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EVIDENCE FOR ICE PARTICLE MULTIPLICATION FROM IN-SITU MEASUREMENTS

by

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1. INTRODUCTION

Environment Canada has made in-situ measurements in cold stratiform cloud layers for many years. Evidence has accumulated showing that ice multiplication, or a secondary ice formation process, probably occurs quite often in clouds containing both liquid water and ice, and those containing only ice. Based on statistical summaries of large data sets where many processes might be operating, ice particle concentrations appear to be much higher than can be explained by conventional ice nucleus measurements, and the ice particle number concentration appear to be the same over a wide temperature range (Korolev et al., 2000; Gultepe et al. 2001; Field et al, 2005). Ice particle size spectra consistently show larger numbers of particles at smaller sizes, suggesting that small particles are being produced continuously (Boudala et al., 2002). There are measurement issues associated with ice particles shattering off probe parts and these artifacts being counted (Korolev and Isaac, 2005; Isaac et al., 2006). However, the bulk of the evidence points to the fact that ice multiplication must be occurring in the atmosphere over a wide temperature range, or if secondary ice formation is highly temperature dependent, there must be an efficient redistribution system.

Several secondary ice or ice multiplication processes have been suggested. Rangno and Hobbs (2001) have separated them into four mechanisms:

a) Ice splintering during riming (Hallett and Mossop, 1974) which occurs at temperatures of -3 to -8 ^{o}C with supercooled large drops (>23 µm) present in concentrations > 1cm^{-3} and impact speeds of 0.2-5 ms^{-1}. Foster and Hallett (1982) indicate that this mechanism also works at lower ambient temperatures at higher liquid water contents, where the ice surface temperature is in the Hallett-Mossop range.
b) Fragmentation of ice particles (Hobbs and Farber, 1972; Vardiman, 1978; Oraltay and Hallett, 1989). This grouping might also include the rime breakup mechanism proposed by Vali (1980).
c) Shattering of isolated drops during freezing (Hobbs and Alkezweeny, 1968; Brownscombe and Thorndike, 1968). However, Johnson and Hallett (1968) indicate that this mechanism is unlikely for drops in free fall in the atmosphere.
d) Nucleation of ice at high water supersaturation adjacent to freezing supercooled drops (Dye and Hobbs, 1966; Gagin and Nozyce, 1984).

It is beyond this paper to do a through review of all the evidence or the papers on ice multiplication. This issue will be discussed primarily with the measurements that have been made during Environment Canada field work.

2. OBSERVATIONS/INSTRUMENTATION

In-situ observations from the Canadian Freezing Drizzle Experiment (CFDE) I and III, the Alliance Icing Research Study (AIRS) I and the FIRE Arctic

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Cloud Experiment (Isaac et al., 2001; Gultepe and Isaac, 2002) were used in the analysis. Details on the instruments mounted on the National Reseach Council Conavair-580 can also be found in the same papers

It is well known that there are many issues associated with measurements of small ice particles. The SPEC Cloud Particle Imager (see Lawson et al., 2001) has been quite useful, especially for imaging ice particles, and some new instruments are under development (e.g. Hirst et al., 2001; Baumgardner et al.,2005; Lawson, R.P. et al., 2006). However, the measurement of small ice particles remains a problem and a complete discussion of the issues is beyond this paper.

Isaac et al. (2006) using a high speed video camera were able to show tens to hundreds of small ice particles being generated after impacts with the side of the Nevzorov total water content probe (see Fig. 1). Korolev and Isaac (2005) showed, small ice particles can be generated off the tips of conventional PMS type imaging probes, resulting in false counts (Fig. 2). This illustrates that erroneous enhancements in small partlcle measurements may be introduced into data sets by instrument artifacts, although some correction methods have been described (e.g. Field et al., 2006).

In the presence of irregular ice crystals, *Gardiner and Hallett* (1985) reported that the PMS FSSP measured ice concentrations 2-3 orders of magnitude higher than those derived from a replicator. However, a study by *Arnott et al.* (2000) suggests that the replicator under estimates concentrations of particle (D<50 μm). Therefore, the FSSP may not over estimate particle concentrations by 2-3 orders of magnitude as suggested by *Gardiner and Hallett* (1985). More recent measurements by Field et al. (2003) suggest that the FSSP may only be overestimating the concentration of small ice crystals on average by a factor of two.

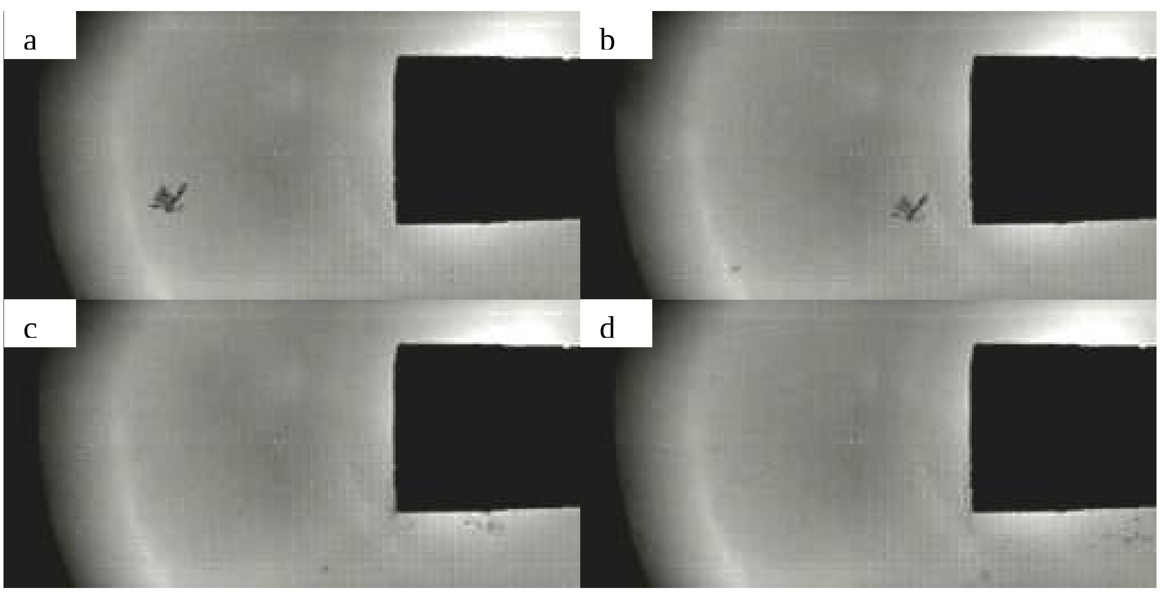

Fig. 1: A sequence (a to d) of pictures from Run 6 on 18 December. Shows an ice particle (1.5 mm in size) impacting with the edge of the cone of the TWC sensor and shattering into many pieces (from Isaac et al., 2005).

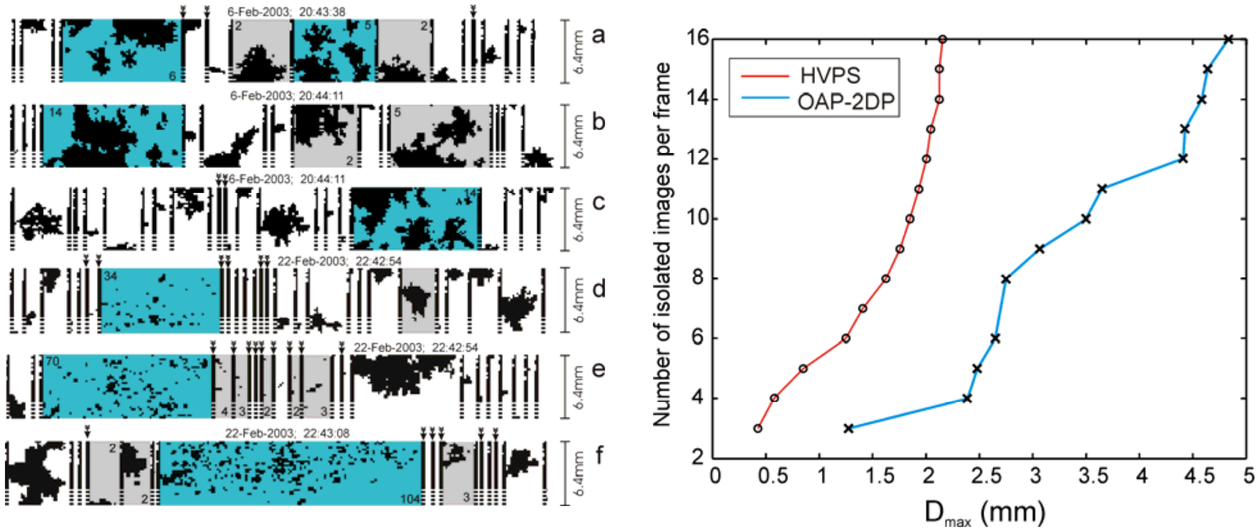

Fig. 2: Left panel shows ice particles shattering off the tips of the PMS 2D-P while the right panel shows the number of images created per frame as a function of particle size (see Korolev and Isaac, 2005).

3. NUMBER CONCENTRATION OF ICE PARTICLES

The number concentration of ice particles appears to be independent of temperature (Korolev et al., 2000; Gultepe et al. 2001; Boudala and Isaac, 2006; Field et al., 2005; Gayet et al., 2006). Gultepe et al. (2001) show this to be true for particles as measured by the PMS 2D-C and FSSP probes. There is a tendency for large particles (>1mm) to be more numerous at warm temperatures, which is consistent with particles growing and aggregating as they fall to warmer temperatures (Gultepe et al., 2001).

Isaac et al. (2004) show the ice particle number concentration for ice particles larger than 100 µm to be independent of temperature. The histogram for each temperature interval also shows a similar spectral width (see Fig. 3), in agreement with the results of Gultepe et al. (2001). Field et al. (2005) also came to the same conclusion for measurements made in the U.K. showing the probability distributions of ice particle concentrations (D>100µm) to be similar for the temperature range 0 to -60 ^oC, with an increase in concentration of a factor ~10 from the coldest to the warmest temperature ranges. Within the temperature range of Fig. 3, Field et al. (2005) did not show much variation. If ice multiplication was occurring in these clouds, and was temperature dependent, as suggested by the Hallett and Mossop (1974) mechanism, then one would expect to see a large peak, especially in the extreme end of the distribution, in the temperature range -3 to -8 ^oC. That does not appear in the probability distributions of number concentration (Fig. 3).

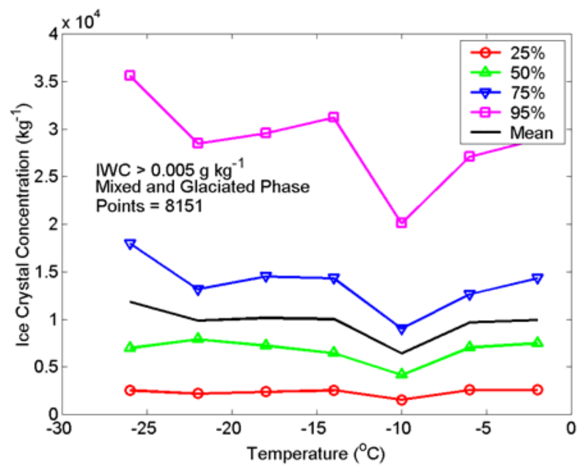

Fig. 3: Ice crystals concentration for sizes > 100 µm in stratiform clouds as discussed in Isaac et al. (2004). The field projects used were the Canadian Freezing Drizzle Experiment (CFDE) I and III, the Alliance Icing Research Study (AIRS) I and the FIRE Arctic Cloud Experiment (Isaac et al., 2001; Gultepe and Isaac, 2002). The concentrations are given in number per kg of dry air and represent 8151 averages of 3km intervals. The analysis was done following the method of Cober et al. (2001).

The average number concentration of ice particles and the width of the number concentration distribution also appears to be independent of geographic location. Gultepe et al. (2001) demonstrated this conclusion using measurements in the Arctic, off the east coast of Canada, and in the mid southern portion of Canada. One might expect differences in aerosol properties in these locations with a resulting difference in ice concentration produced by primary nucleation mechanisms.

4. PARTICLE SHAPE AND ICE PARTICLE SPECTRA

There have been many papers written on ice particle shape recently including ones which summarize measurements made by Environment Canada (Korolev et al., 1999, 2000). Of particular interest for this paper is the observation of Korolev and Isaac (2003) that small ice particles tend to be round, with roundness (R) being defined by the equation

$$R = 4S_{meas}/\pi D^2_{max}$$

where S_{meas} is the measured projection area of the particle image and D_{max} is the maximum dimension of the image (Fig. 4). This suggests that small ice particles are formed from frozen drops. From observations in the laboratory, Korolev et al. (2004) concluded small frozen drops can retain their spheroidal shapes for periods of minutes to tens of minutes under conditions close to saturation over water. This helps explain the observation of many spherical ice particles in natural clouds.

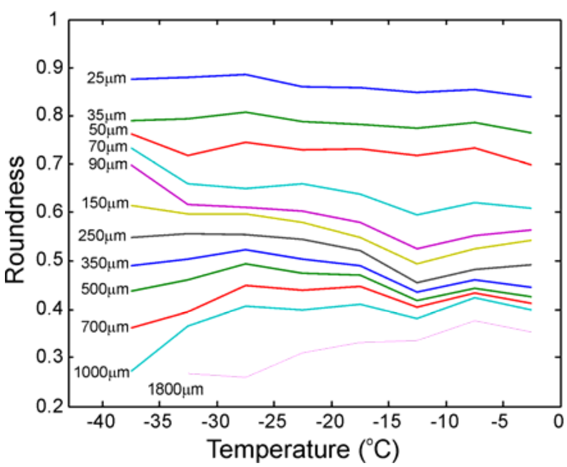

Fig. 4: The roundness of ice particles as a function of particle size and temperature (from Korolev and Isaac, 2003).

It is reasonably well known that ice particle spectra tend to follow an exponential type distribution, generalized gamma distribution, or a power-law relationship (e.g. Mitchell, 1991; Heymsfield et al., 2002; Field et al., 2005) with higher concentration of ice particles as the particle size gets smaller. Fig. 5, as revised from Isaac et al. (2002), shows composite spectra from the CFDE III and AIRS I projects (Isaac et al., 2001) which tend to illustrate that higher concentration of ice particles are observed as the particle size gets smaller.

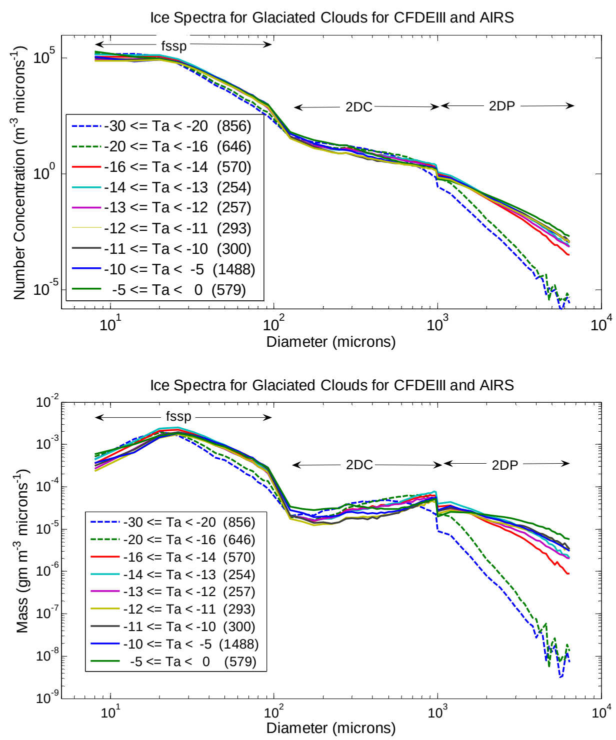

Fig. 5: Composite number concentration and mass spectra for the CFDE III and AIRS projects (Isaac et al., 2001) for glaciated clouds only (modified from Isaac et al., 2002). The mass was calculated using the equivalent melted diameter versus area relationships for aggregates and dendrites as proposed by Cunningham (1978). The spectra were determined using measurements from the PMS FSSP 2DC and 2DP (Knollenberg, 1981) as marked on the diagrams. The FSSP probe is only shown for illustrative purposes because it is designed for liquid drops and will not accurately size small ice particles. The diameter along the x-axis is a circular area diameter determined using a centre-in technique assuming circular geometry.

Although the ability of the FSSP probe to size small ice particles is suspect, even at particle sizes less than 10 µm, the concentration is increasing. Certainly at sizes less than 100 µm, where the confidence level for measurements is higher, the concentration appears to be increasing towards smaller crystal diameters. Note that the composite mass spectrum also tends to show that a great deal of the mass is in small particle sizes. The distributions show a shift to larger sizes at warmer temperatures (Fig. 5) but there are still more ice particles at smaller sizes, even as this shift takes place.

The preponderance of small ice particles suggests a continuous formation mechanism probably near the measurement level. It is unlikely the small particles are coming from above at colder temperatures, because their terminal velocities are small and they would quickly grow out of the small size range. If small particles are being generated by sublimation of larger particles, you would not get such high concentrations. Small particles could be created by sublimation and breakup of larger ice particles, but this is a secondary or ice multiplication formation mechanism. More work is needed to understand the ice particle distribution shape but it does hold clues to ice formation mechanisms.

5. CASE STUDIES

Some interesting cases of what appeared to be ice multiplication occurred during the CFDE III project (Isaac et al., 2001). These measurements were made during the severe Ice Storm that occurred during 1998 over Eastern Canada and the U.S. While doing spiral ascents and descents with missed approaches into Mirabel, Quebec, on both 8 and 9 January 1998, the NRC instrumented Convair-580 aircraft went through freezing precipitation layers which were rapidly glaciating over distance scales of 100s of meters. Figs. 6 and 7 show the profiles over the airport and some PMS OAP 2DC imagery. The above freezing layer is marked in gray, while the bottom layer with the ice crystals being present is marked in a coloured band. Fig. 8 shows the particle spectra in the all liquid portion and the mixed or glaciated portion of the profile. Several profiles were made through these transition zones and the results were repeatable. At the time of the profiles, the surface observer was reporting freezing rain, ice pellets and fog. It is clear that the supercooled liquid is being rapidly converted to ice particles, and the resulting ice particles show a relatively broad size distribution with both small and large particles present. Some of the ice particles are probably frozen drops as the surface observer was reporting ice pellets. Because of the presence of needles or columns and the relatively warm temperatures on 9 January, it was speculated that the Hallett and Mossop (1974) ice multiplication mechanism might be being observed. This possibility is quite real because of the presence of ice pellets, and drops greater than 20 µm in concentrations greater than 2 cm^{-3}. However, the mechanism proposed by Rangno and Hobbs (2001) is also possible whereby "the formation of ice concentrations greatly in excess of ice nuclei in slightly supercooled stratiform clouds in the Arctic appear to be ice splinter production where a few frozen drizzle drops begin to rime; the fragmentation of drizzle-size drops as they freeze; and the fragmentation of existing delicate ice crystals."

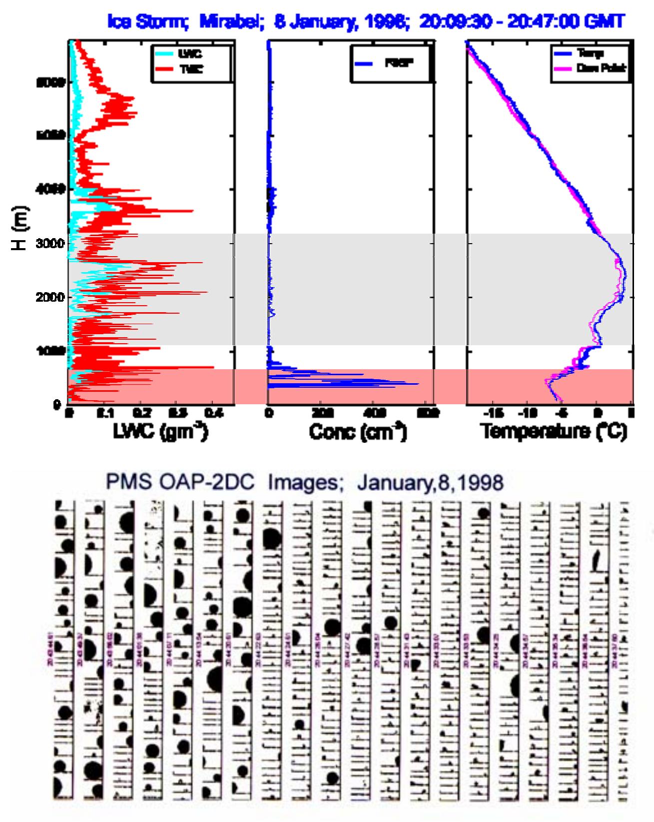

Fig. 6: The top panel shows the profile as measured during the descent over Mirabel, Quebec on 8 January during CFDE III. The bottom panel shows the PMS 2D-C imagery during the sharp transition from the liquid to ice zone which occurred near -3 °C, or near the top of the coloured band near ground level in the top panel.

Another possibility is the interaction of the surface with the air aloft. Freezing precipitation was causing a layer of ice on the trees and other objects at the surface. High surface winds (near 30 km/hr) were blowing large drops around which could have frozen on protruding objects and the resulting freezing, possible shattering and ejection of splinters might be enough to explain the observations when the resulting particles are mixed within the boundary layer. Such a mechanism could also explain why freezing precipitation is rarely observed at temperatures colder than -5°C, with 85% of freezing precipitation occurring between 0-10°C (Cortinas et al., 2004).

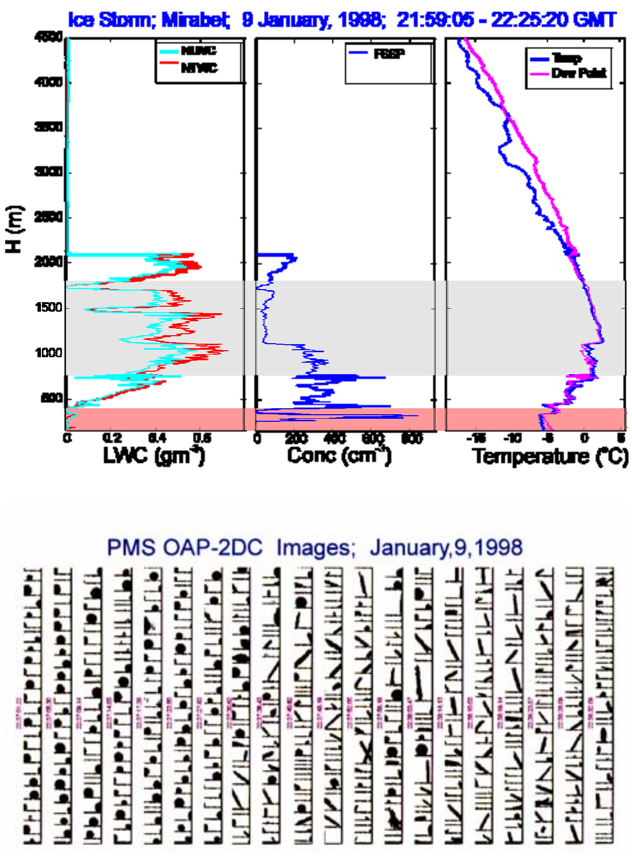

Fig. 7: The top panel shows the profile as measured during the descent over Mirabel, Quebec on 9 January during CFDE III. The bottom panel shows the PMS 2D-C imagery during the sharp transition from the liquid to ice zone which occurred at -4 °C, or near the top of the coloured band near ground level in the top panel.

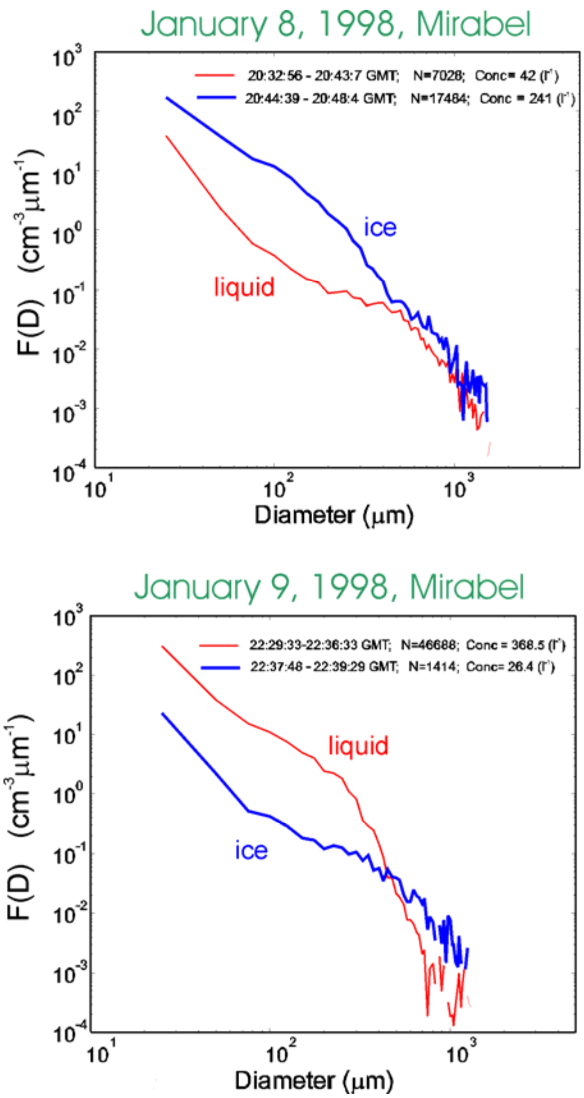

Fig. 8: Spectra as measured with the PMS probes during the profiles described in Figs. 6 and 7. The red curves represent the liquid spectra measured at the higher altitudes and the blue curves represent the ice spectra measured at lower altitudes.

6. DISCUSSION AND CONCLUSIONS

Although there are many uncertainties associated with counting and sizing small ice particles, those uncertainties are not enough to discount the conclusions or points listed below.

- Based on statistical summaries of large data sets, the number concentration of ice particles appears to be independent of temperature. As Gultepe et al. (2001) have shown, the concentration is similar in many different geographic locations.
- The relatively constant number concentration distribution with temperature suggests that the dominant ice multiplication mechanism is not temperature dependent. If it was temperature dependent, then one would need a fast mixing process to spread around locally produced ice particles as has been observed in hurricanes (Black and Hallett, 1986).
- The characteristic ice particle size distribution spectra, showing many ice crystals at small sizes, suggests a continuous formation mechanism. It is very rare to see a spectrum of ice particles with no small ice particle present.
- Small ice particles appear circular in 2D images. This suggests that most of these small ice particles are probably frozen drops (see Korolev and Isaac, 2003).
- Liquid clouds can be converted into glaciated clouds over short distance scales as the case studies of January 1998 have shown.

The observations can be explained by an ice multiplication mechanism(s) or a time dependent ice nucleation mechanism. It is well known that some nucleation mechanisms such as contact freezing are time dependent (e.g. Isaac and Douglas, 1972). The preponderance of spherical appearing small ice particles does support the idea that small drops are constantly being nucleated. Since the concentration of ice particles appears to be independent of geographic location where the aerosol characteristics would be different, this tends to discount the continuous heterogeneous nucleation mechanism. The "sudden" appearance of ice crystals in some instances also does not support the continuous nucleation mechanism.

The dominance of spherical ice particles at small sizes (Fig. 4) suggests that any generalized ice multiplication theory must consider the possibility of cloud droplets being nucleated and frozen as proposed by Dye and Hobbs (1966) and Gagin and Nozyce (1984).

It would be valuable to compare measurements of ice concentration made in different parts of the world by various groups. Many groups use the same instrumentation, so this should be possible. However, it is difficult to do now with any precision because differences in software used in the analysis can be significant. Korolev et al. (2000) and Field et al. (2005) used the same software and equipment, and looked at mid-latitude frontal cloud types. The Field et al. ice particle concentrations (D>100µm) appear to be only 2-3 times higher than Korolev et al. (D>125µm), showing measurements in the UK are similar to those in Canada. It is interesting to note that the ice particle concentrations (D>100µm) of Gayet et al. (2006) as measured in cirrus clouds in the southern hemisphere (-25 to -60 ^{o}C) are very similar to those reported by Korolev et al.

The observations cannot be explained using the ice nucleation parameterization proposed by Meyers et al. (1992), which is widely used in the numerical modeling community. Gultepe et al. (2001) compared several ice nucleation parameterizations with the observed ice particle measurements and could not find any good agreement. So ice multiplication or secondary ice formation mechanisms should be considered in numerical models. However, Boudala and Isaac (2006) suggest a new way of parameterizing ice processes in numerical models without explicitly modeling either primary or secondary ice initiation. This method has the advantage of simulating the observations for ice mass, ice deposition and riming rate, and particle terminal velocity.

It is clear that the observations cannot be easily explained by any known ice multiplication theory. More research needs to be performed in order to solve this problem. The uncertainty in the measurements also needs to be minimized with improved instrumentation.

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