# P5.3 Observation of the Microstructure of Mixed Phase Clouds.

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## 1. INTRODUCTION

Early observations conducted with the help of airborne impactors and replicators (e.g. Zak 1937; Peppler 1940; Weickmann 1945; Borovikov et al, 1963). have shown that liquid and ice particles may coexist in natural cold clouds down to -40°C. The proportion between ice and liquid phase or the phase composition of clouds is an important parameter in the microphysics of clouds. The phase composition affects the rate of precipitation formation and the life cycle of clouds in general. Knowledge of the mixed phase in clouds is important for radar, lidar, satellite retrievals, radiation transfer calculations, GCM and climate modeling, wave propagation etc. The mixed phase is hypothesized to be one of the major causes for cloud electrification.

This work presents a study of the microstructure of mixed phase clouds based on aircraft *in situ* measurements. The statistics of cloud particle sizes, concentration, total and liquid water content versus the mixed phase composition for the temperatures from 0°C to -35°C are presented.

## 2. INSTRUMENTATION

The analysis of the mixed phase composition of clouds in relation to this work has been conducted with the help of the Nevzorov LWC/TWC probe. The Nevzorov probe is a constant temperature hot wire instrument consisting of two sensors: for measurement of liquid water content (LWC) and for total (ice+liquid) water content (TWC). The threshold sensitivity to water and ice was estimated as 0.003 - 0.005g $m^{-3}$. Questions related to the measurement accuracy of the Nevzorov probe were discussed in detail in Korolev et al. (1998). The phase discriminating capability of the Nevzorov probe was tested in the National Research Council (NRC) wind tunnel and natural clouds (Korolev et al. 1998; Korolev et al. 2002).

The other cloud microphysical instrumentation relevant to this study include: two Rosemount temperature probes and a reverse flow temperature probe; a Cambridge dewpoint hygrometer EG&G; PMS FSSP-100 (Knollenberg, 1981), which measured droplet size distributions in size range 5 - 95 $\mu$m, respectively; two PMS King probes (King et al. 1978); a Rosemount Icing Detector; a PMS OAP-2DC (25 - 800 $\mu$m); and a PMS OAP-2DP (200 - 6400 $\mu$m) (Knollenberg, 1981). These instruments were installed on the National Research Council (NRC) Convair-580.

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## 3. DATA PROCESSING

The IWC and LWC were calculated from a system of equations described in Korolev et al. (1998)

$$W_{ice} = \frac{\varepsilon_{lL} W_{TWC} - \varepsilon_{lT} W_{LWC}}{\varepsilon_{lL}\varepsilon_{iT} k - \varepsilon_{lT}\beta} \quad (1)$$

$$W_{liq} = \frac{\varepsilon_{iT} k}{\varepsilon_{iT}\varepsilon_{lL} k - \varepsilon_{lT}\beta} W_{LWC} - \frac{\beta}{\varepsilon_{iT}\varepsilon_{lL} k - \varepsilon_{lT}\beta} W_{TWC} \quad (2)$$

where $W_{TWC}$ and $W_{LWC}$ are the uncorrected total and liquid water contents measured directly by the Nevzorov LWC and TWC sensors, respectively; $\varepsilon_{lT},\varepsilon_{iT}$ are the integrated collection efficiencies for liquid droplets and ice particles respectively for the TWC sensor;. $\varepsilon_{lL}$ is the integrated collection efficiency for liquid droplets for the LWC sensor; $\beta$ is a coefficient accounting for the residual effect of ice particles on the LWC sensor; $k = L_i^*/L_l^*$ is a correction coefficient accounting for the difference between expended energy for water ($L_l^*$) and ice particle ($L_i^*$) evaporation. The coefficient $k$ was assumed to be approximately equal to $k = L_i^*/L_l^* \cong 1.12$, and the collection efficiencies $\varepsilon_{lT},\varepsilon_{iT}$, $\varepsilon_{lL}$, for both LWC and TWC sensors were assumed to be equal to unity (Korolev et al. 2002).

The residual effect of ice on the LWC sensor is due to the small amount of heat removed from the LWC sensor during collision with ice particles. The residual effect depends on size, shape and bulk density of ice particle, air speed, air temperature, and the temperature of the sensor.

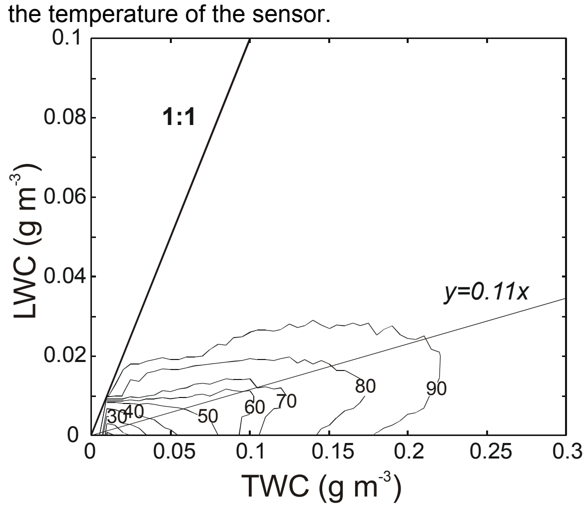


**Figure 1.** Scatter diagram of LWC versus TWC measured by the Nevzorov probe in glaciated clouds. Contour lines indicate isopleths of probability for finding LWC and TWC inside the contour.

The residual ice effect on the LWC sensor for the data set of this paper was estimated from Fig. 1, which shows a scatterplot of $W_{LWC}$ versus $W_{TWC}$ in glaciated clouds. The clouds were identified as glaciated if $W_{TWC}$ exceeded a threshold value of $W_{thresh}$ =0.005g/m$^3$, and the ramp voltage ($V_{RICE}$) of the Rosemount Ice Detector (RICE) probe was not increasing, i.e.

$$W_{TWC}>W_{thresh} \quad (3)$$

$$dV_{RICE}/dt\leq 0. \quad (4)$$

Mazin et al. (2001) theoretically estimated the threshold sensitivity of the RICE probe as 0.006 g m$^{-3}$ at 100m s$^{-1}$. The threshold sensitivity for the RICE probe deduced from *in-situ* measurements was estimated as 0.01 g m$^{-3}$ for a 30 second averaging time interval (Cober et al. 2001). The best-fit linear regression forced through the origin for the scatter diagram in Fig. 1 gives $W_{LWC}$=0.11$W_{TWC}$. and it results in the coefficient $\beta$=0.11

It should be emphasised that the coefficient $\beta$=0.11 applies to air speeds typical for the Convair 580, i.e. $U$~100m/s. The residual effect increases with aircraft velocity and it may reach up to 50% of the indicated IWC at $U$ > 200m s$^{-1}$ (Strapp et al. 1999).

## 4. DESCRIPTION OF THE DATA SET

The data on mixed phase were collected during five field campaigns: the Beaufort Arctic Storm Experiment (BASE) in September-October 1994, the First Canadian Freezing Drizzle Experiment (CFDE I) in March 1995 (Isaac et al. 2001changed font to 9), the Third Canadian Freezing Drizzle Experiment (CFDE III) December 1997-February 1998 (Isaac et al. 2001 changed font to 9), FIRE.ACE in April 1998, and the Alliance Icing Research Study (AIRS) December 1999-February 2000 (Isaac et al. 2001changed font to 9). Usually the duration of the NRC Convair-580 flights was between 2 and 5 hours.

The bulk of data were collected in stratiform clouds (*St, Sc, Ns, As, Ac*), usually associated with frontal systems. During the BASE and FIRE.ACE projects, a number of flights sampled cirrus clouds. The scale of spatial averaging is about 100m (1s). The total flight length in cloud with $W_{TWC}$>0.01 g m$^{-3}$ was 44013 km. The temperature and altitude of measurements ranged from 0 to -35$^o$C and from 0 to 6 km, respectively. The frequency of occurrence of different parameters associated with the analysis of the phase composition of clouds was calculated for seven 5-degree temperature intervals in the range 0$^o$C to -35$^o$C.

## 5. DEFINITION OF MIXED PHASE CLOUDS

Within the cloud physics community there is no clear definition of “liquid”, “mixed” and “ice” phase cloud. For example, should a cloud be defined as a “mixed”, if it has one ice particle per 10$^1$ or 10$^{100}$ droplets? Should a cloud be considered as “glaciated” if it contains one liquid droplet per 10$^{100}$ ice particles, or is it still “mixed”?

It is suggested here to characterize the phase composition of clouds with the help of a so-called phase composition coefficient, $\mu_n$ (Korolev 1998)

$$\mu_n = \frac{\sum_j \alpha_{ice\ j} N_{ice\ j} D^n_{ice\ j}}{\sum_j \alpha_{ice\ j} N_{ice\ j} D^n_{ice\ j} + \sum_i \alpha_{liq\ j} N_{liquid\ i} D^n_{liquid\ i}} = \quad (5)$$

$$= \frac{\alpha_{ice} N_{ice} \overline{D^n_{ice}}}{\alpha_{ice} N_{ice} \overline{D^n_{ice}} + \alpha_{liq} N_{liquid} \overline{D^n_{liquid}}}$$

Here $N_{liquid}$ and $N_{ice}$ are the number concentration of liquid droplets and ice particles, respectively; $D_{liquid}$ and $D_{ice}$ are the droplet diameter, characteristic size of ice particles, respectively; $n$=0,1,2,3… is the moment of the phase composition coefficient; $\alpha_{ice}$, $\alpha_{liq}$ are the coefficients equal to unity, or extinction efficiency, or bulk density, etc, depending on the moment $n$. The advantage of $\mu_n$ is that it changes in a limited interval, i.e. from $\mu_n$=0, when the cloud is all liquid, to $\mu_n$=1, when the cloud is completely glaciated.

The moment $n$ of the phase composition coefficient $\mu_n$ should depend on the problem being solved. Thus, $\mu_6$ should probably be used in radar studies, while radiative transfer models and lidar sensing should use $\mu_2$, etc.

Since we are considering cloud water content, the mixed phase will be characterized by the phase composition coefficient of third moment, i.e.

$$\mu_3 = \frac{\rho_{ice} N_{ice} \overline{D^3_{ice}}}{\rho_{ice} N_{ice} \overline{D^3_{ice}} + \rho_{liq} N_{liquid} \overline{D^3_{liquid}}} \cong \frac{W_i}{W_i + W_l} \quad (6)$$

The phase composition coefficient used in this study is similar to ‘fractional ice content’ in the work of Tremblay et al. (1996). Some authors are using ‘liquid water fraction’ to describe mixed phase composition of clouds (e.g. Moss and Johnson 1994). The liquid water fraction is related to $\mu_3$ as 1-$\mu_3$.

Due to instrumental limitations $\mu_3$ cannot be resolved with an accuracy better than 10%. Therefore, clouds with $\mu_3$<0.1 will be defined as “liquid”; clouds with 0.1≤$\mu_3$≤0.9 as “mixed”; and clouds having $\mu_3$>0.9 as “glaciated” or “ice” clouds. It should be emphasized that these definitions of “liquid” and “ice” clouds are motivated by limited instrument resolution, rather than by physical concepts. Therefore, the clouds defined as “liquid” may still contain a small fraction of ice, whereas “ice” clouds may contain some liquid droplets.

## 6. EXPERIMENTAL RESULTS

### *6.1 Mixed phase composition versus temperature*

Figure 2 a,b shows the frequency and cumulative distributions of $\mu_3$ in seven 5-degree temperature intervals in the range -35$^o$C<$T$<0$^o$C. The curves in Fig. 2a show explicit minima in the range 0.1<$\mu_3$<0.9 for all temperature intervals. At the same time, there are two explicit maxima: one for $\mu_3$<0.1 (liquid clouds) and another one for $\mu_3$>0.9 (ice clouds). The frequency of liquid clouds ($\mu_3$<0.1) decreases with the decrease in temperature from about 50% at -5°C<$T$<0°C down to 5% for -35°C<$T$<-30°C. However, the occurrence of glaciated clouds increases with decreasing temperature from 20% at -5°C<$T$<0°C up to 50% for -35°C<$T$<-30°C. The frequency of occurrence of the mixed phase clouds stays approximately constant in the range $0.2<\mu_3<0.5$. An increase of the threshold, $W_{thresh}$, results in a decrease of the fraction of glaciated clouds and an increase of the fraction of liquid zones, whereas the shape of the density distributions of $\mu_3$ stays about the same as in Fig. 2 a.

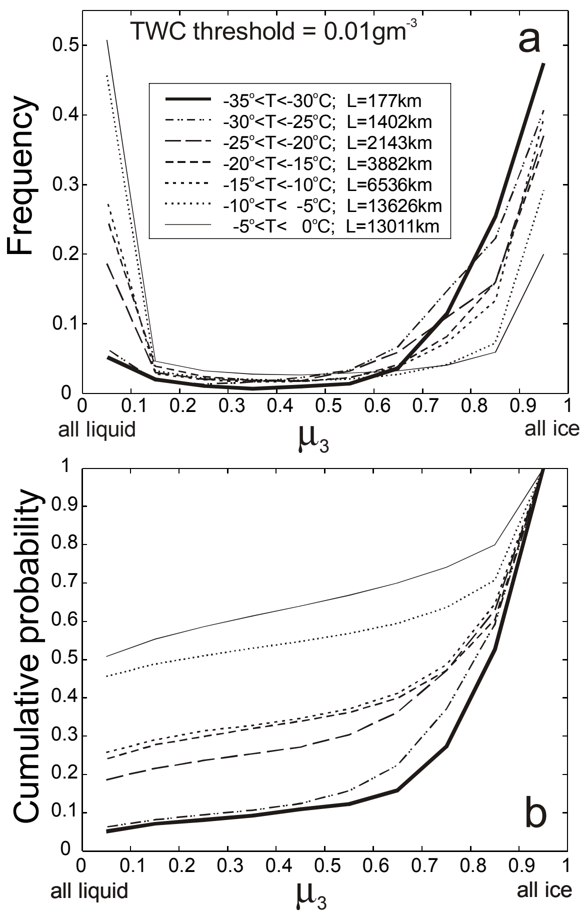


**Figure 2.** Density (a) and cumulative (b) probability distributions of mixed phase composition coefficient □3 for different temperature intervals. $\mu_3$=0 in liquid clouds; $\mu_3$=1 in ice clouds.

### *6.2 Relation between mixed phase composition and cloud water content*

Figure 3 shows the dependence of IWC, LWC and TWC versus temperature for the whole range of $\mu_3$, i.e in all cloud regardless of their phase composition. As seen from Fig. 3 IWC, LWC and TWC are monotonically decreasing from 0.04 to 0.02g m$^{-3}$, from 0.1 to 0.01g m$^{-3}$, and from 0.14 to 0.03g m$^{-3}$, respectively, when the temperature decreases from 0°C to -35°C. It is worth noting that IWC does not change as much as LWC and stays nearly constant.

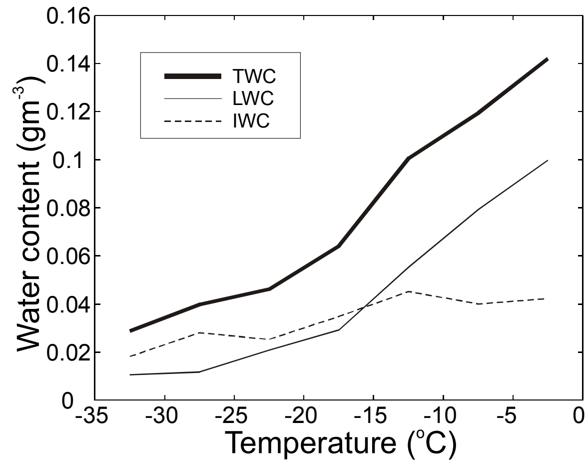


**Figure 3.** Dependence of IWC, LWC and TWC versus temperature averaged over all clouds.

### *6.3 Relation between mixed phase composition and cloud particle concentration*

Recent studies have suggested that FSSP measurements could be used for estimation of cloud particle number concentration in mixed and ice clouds (Gayet et al. 1996; Arnott et al. 2000; Ivanova et al., 2001). These findings create the basis in this study for the use of FSSP measurements of cloud particle concentration in mixed phase and ice clouds. In-situ measurements conducted in ice clouds using replicators and optical size spectrometers have showed that the concentration of ice particles having $d<50\mu$m is usually 1-3 orders of magnitude more as than that for $d>50\mu$m (Heymsfield and Platt 1984; Ivanova et al. 2001). Therefore under these conditions, the FSSP measured concentration are assumed as an estimate of particle concentration. The errors of concentration related to undercounting of particles outside of the FSSP range would not exceed, on average, a few percent.

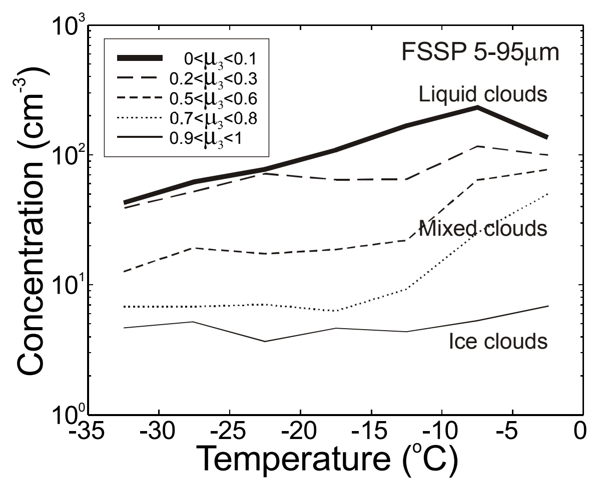


**Figure 4.** Average concentration of cloud particles versus temperature for different $\mu_3$. The concentration was measured by FSSP in the size range 5-95$\mu$m.

Figure 4 shows the dependence of the cloud particle concentration versus temperature for different ranges of $\mu_3$. The most interesting phenomenon is that the particle concentration stays approximately constant in ice clouds ($\mu_3$>0.9). The particle concentration varies somewhat between 4cm$^{-3}$ and 6cm$^{-3}$. The concentration measured in glaciated clouds may be interpreted as an estimate of the concentration of ice particles.

The independence of the ice particle concentration on temperature is an interesting observation requiring further explanation. This observation is in agreement with Gultepe et al. (2001).

### *6.4 Characteristic size of particles in ice and liquid clouds*

Measurements of particle number concentration and their mass enable an estimate of mean volume diameter of cloud particles as:

$$\overline{D}_3 = \left(\frac{6W}{\pi\rho N}\right)^{1/3} \quad (7)$$

Here $W$ is cloud water content measured by the Nevzorov probe; $N$ is the concentration measured by FSSP; and $\rho$ is the density of cloud particles.

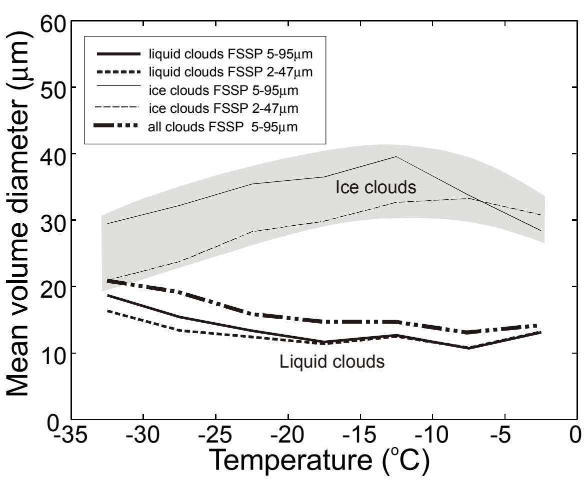


**Figure 5.** Average mean volume diameter of cloud particles versus temperature for liquid, ice, and all clouds derived from the Nevzorov LWC/TWC and FSSP concentration measurements

Figure 5 shows the dependence of $\overline{D}_3$ on temperature for ice ($\mu_3$>0.9) and liquid clouds ($\mu_3$<0.1). The mean volume diameter in liquid clouds slightly increases with a decrease of temperature from about 12$\mu$m to 18$\mu$m. In ice clouds $\overline{D}_3$ varies from approximately 20$\mu$m to 35$\mu$m. As seen from Fig. 11, $\overline{D}_3$ has a maximum at temperatures -15°C<$T$<-10°C, which is consistent with maximum growth rate of ice. Mean volume diameters calculated for all clouds is rather close to $\overline{D}_3$ in liquid clouds and it increases from about 15$\mu$m to 20$\mu$m with a decrease of temperature (Fig. 5).

The density of ice particles in the atmosphere varies from about 100 kg/m$^3$ to 900 kg/m$^3$ and it depends on particle size and its habit (e.g. Heymsfield 1972; Ryan et al. 1976). The density of ice particles increases with decreasing particle size approaching 900 kg/m$^3$ for small ice particles. The mean volume size of ice particles, as shown in Fig. 5, was calculated with an ice density of $\rho$=800 kg/m$^3$. Such an assumption may be justified by the small characteristic sizes of ice particles found in this study. It is worth noting, that since $\rho$ in Eq. 7 is raised to -1/3 power, a decrease of the density of ice by a factor two would result in only a 20% increase of $\overline{D}_3$. Therefore, the expected uncertainty due to $\rho$ would not significantly affect the values of $\overline{D}_3$ shown in Fig. 5.

The mean volume diameter of ice particles, as shown in Fig. 5, was found to be rather small. Evidence regarding the characteristic size of ice particles comes from measurements of the effective diameter of cloud particles. It can be shown that, for any size distribution, $D_{eff} \geq \overline{D}_3 \geq \overline{D}_2 \geq \overline{D}$. Based on a large data set of aircraft data, it was shown that $D_{eff}$ deduced from in-situ measurements of TWC and extinction coefficient at -50°C<$T$<-30°C where all cloud particles are presumably ice, was found to be about 14$\mu$m (Korolev et al. 2001). This is even less than $\overline{D}_3$ shown in Fig. 5. Comparison of $\overline{D}_3$ and $D_{eff}$ suggests that $\overline{D}_3$ should be even less than that shown in Fig. 5.

### *6.5 Correlation between LWC and IWC*

Analysis of in-situ measurements show no correlation between IWC and LWC in mixed phase clouds. An example of simultaneous measurements of LWC and IWC is shown in Fig. 6 a. The scatterplot of LWC and IWC in Fig 6 b indicates an absence of any correlation between these parameters. The correlation coefficient between LWC and IWC for this case was found to be K=0.13. In general, the correlation coefficient between LWC and IWC changes somewhat from 0 to 0.3.

Since the IWC in mixed clouds grows at the expense of LWC, one would expect a high correlation coefficient between these two parameters. However, the low correlation may be because the rate of growth of IWC is governed by the supersaturation over ice. The supersaturation in mixed phase clouds stays constant and nearly equal to the saturation over water before complete evaporation of liquid droplets (Korolev and Isaac 2002) and therefore it is independent of the current value of LWC. The spatial correlation may also be reduced by the fact that glaciation in different cloud regions may start at different times.

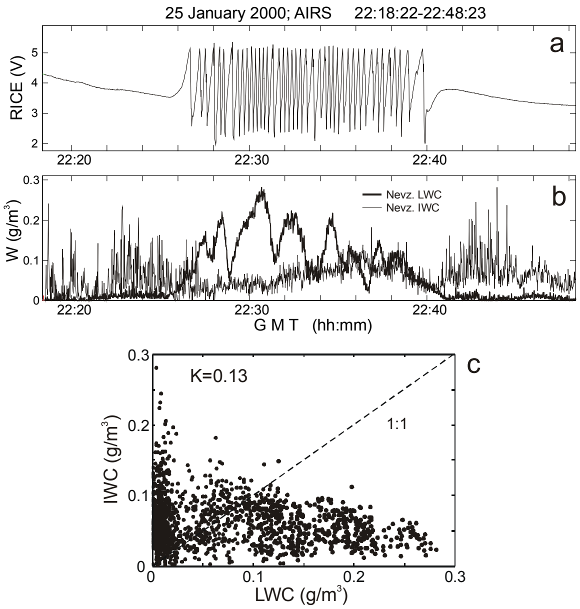


**Figure 6.** Example of low correlation between LWC and IWC. RICE signal (a), LWC and IWC measured by Nevzorov LWC/TWC probe (b), scatterplot of LWC and IWC (c). The correlation coefficient between LWC and IWC for the indicated period of time is 0.13.

## 7. COMPARISON WITH PREVIOUS STUDIES OF MIXED PHASE CLOUDS

Borovikov et al. (1963) based on nearly 9000 aircraft impactor samples derived the frequency of occurrence of liquid mixed and ice clouds versus different temperature intervals (Fig. 7). In their study, an impactor sample was considered to be liquid if it did not contain ice particles, and it was considered as ice if there were no spherical particles that could be identified as droplets. The rest of the samples were assumed to be mixed phase. Such an approach implies that Borovikov et al. (1963) defined the mixed phase composition based on number concentration, which is the zero moment of the phase composition coefficient $\mu_0$ (Section 5).

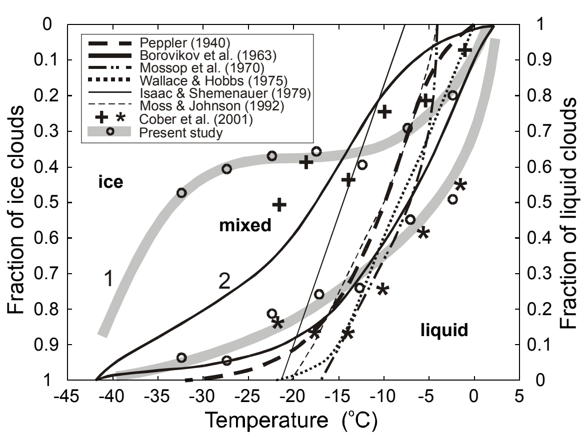


Figure 7. Comparison of fraction of ice, mixed and liquid clouds from the present and previous studies. Lines 1, 2 and * marks refer to left y-axis, all the rest curves refer to the right y-axis

Figure 7 also shows a comparison of the fraction of ice, mixed and liquid clouds obtained in a number of other different studies. It should be noted that the studies of Mossop, et al. (1970), Isaac and Schemenauer (1979), and Wallace and Hobbs (1975) refer to a boundary separating convective clouds with and without ice particles as detected by aircraft instruments. Peppler (1940) based his conclusion on impactor samplings similar to those used Borovikov et al. (1963). Moss and Johnson (1992) estimated mixed phase from OAP-2DC and Johnson-Williams liquid water probe measurements. Fig. 7 gives some idea about how different definitions and instruments can produce alternate conclusions about the mixed phase composition in different type of clouds.

Mazin et al. (1992) and Nevzorov (2000) use a set of instruments including several particle spectrometers, the Nevzorov LWC/TWC probe and the cloud extinction meter, concluded that only a small fraction of clouds are purely liquid and purely glaciated. The majority of clouds were found to have mixed phase. One of the reasons for such a large discrepancy with the findings of the present study is related to a difference in definition of ice and liquid clouds. Another reason may be related to an underestimate of the residual effect of ice on the LWC sensor ($\beta$=0.03) used in their works.

A recent study of phase composition of clouds was conducted by Cober et al. (2001) using the same data sets from the CFDE I and CFDE III projects as used in the present work. The differences can be explained by some differences in the data processing and that the fact that the current study presents a wider range of data encompassing some additional geographical areas.

## 8. DISCUSSION

### *8.1 Relation between glaciation and residence time of cloud particles*

The experimental results presented in Fig. 2 show explicit minima of $\mu_3$ in all temperature intervals. These minima are a result of the instability of mixed phase clouds and the fact that the lifetime of such clouds is limited. At the same time, the maxima at $\mu_3$ =1 indicate that the clouds transit through the mixed phase stage and convert into ice. This leads to an important conclusion that the glaciation time ($\tau_{gl}$) is usually less than the characteristic residence time of liquid particles in clouds ($\tau_{res}$), i.e.

$$\tau_g << \tau_{res}. \quad (8)$$

If the above inequality was reversed, liquid clouds would predominate and the maximum at $\mu_3$ =1 for ice clouds would not exist. The residence time of cloud particles cannot exceed the lifetime of the cloud. However, the residence time of the particles may be significantly less than that of the cloud.

### *8.2 Mechanisms of ice nucleation in clouds*

The concept of increasing activity of ice forming nuclei with decreasing temperature has been demonstrated in numerous laboratory experiments (e.g. Roberts and Hallett 1968) and is consistent with the observed increase of ice concentration in a near adiabatic updraft as measured by aircraft penetration at different levels or by following the updraft in an ascending glider (Dye et al. 1986). Data presented in Fig. 4 indicate that the average concentration of particles in ice clouds is a weak function of the ambient temperature. Moreover, there is a slight trend towards a decrease in the ice particle concentration at colder temperatures. This result conflicts with the parameterizations of ice forming nuclei (IFN) proposed by Fletcher (1962) and Meyers et al. (1992). Fletcher (1962) predicted an increase of ice particle concentration of about nine orders of magnitude when a drop in temperature from 0$^o$C to -35$^o$C. The study of Meyers et al. (1992) proposed an increase of ice concentration of approximately 100 times for the same temperature interval. At the same time, the parameterization of Meyers et al. (1992) gives a concentration of ice particles of $10^2 l^{-1}$ at -35$^o$C, which is more than ten times less than the result obtained in this study. Such a difference cannot be explained by the errors of measurements of particle concentration in the present work. Gultepe et al. (2001) showed a similar contradiction between observations and parameterizations.

The data presented herein are collected in different scenarios, since flight tracks are quite unrelated to the cloud updraft/downdraft structure and must include ice produced under situations other than nucleation. Rime splintering (Hallett and Mossop 1974) occurs over a narrow range of temperature (-3 to -8$^o$C) However, Fig. 4 do not show any significant increase in the concentration of ice above -10$^o$C in glaciated clouds. Therefore it can be concluded that, *on average,* ice multiplication does not affect the concentration of ice particles.

It could be argued that the high concentration of ice at the warmer temperatures may result from ice particles falling from the layers above having colder temperatures and subsequently containing higher concentration of ice particles. However, this does not explain the high concentration of small ice particles in shallow frontal cloud systems with cloud top temperature between -20$^o$C to -15$^o$C. It is also not clear how small ice particles having small terminal fall velocity can fall a few kilometers after having been formed at –35$^o$C without a large change in their size.

One can hypothesize about the existence of a "universal" mechanism of ice nucleation in stratiform clouds, which dominates over the direct formation of ice on ice nuclei (deposition nucleation) and ice multiplication. One of the possible candidates for this role is the formation of ice particles through freezing of liquid droplets. Studies of nucleation in natural clouds have found that the freezing of liquid droplets appear to be the dominant mechanisms in many clouds (Cooper and Vali 1981; Hobbs and Rangno 1985). Following this hypothesis, the formation of ice occurs in two stage. At the first stage the vapour pressure exceeds the saturation pressure over water and liquid droplets get activated. At the second stage, the freezing of droplets occur through activation of contact or immersion ice nuclei.

Ice nucleation during evaporation may provide an attractive explanation of the "universal" mechanism of ice enhancement. This mechanism appears to be fairly general, since evaporation of droplets usually occurs in greatest numbers in downdrafts inside clouds and at the cloud interfaces. Rosinski and Morgan (1991) suggested that evaporating cloud droplets might provide an additional source of ice nuclei.

The concept of ice nucleation associated with solute nucleation following droplet evaporation is suggested by Hallett, Queen and Teets (in preparation). Laboratory experiments have demonstrated that ice and solute nucleation may occur under different situations as the temperature falls and solute concentration increases by evaporation. For NaCl the solubility falls only slightly with decreaseing temperature, whereas ammonium sulfate falls more rapidly. In the former case nucleation is best achieved by evaporation; the latter by cooling. The ice-solution-solute phase diagram shows an unstable region for solution supercooling with respect to ice and a region supersaturated with respect to the solute (Hallett 1968; Queen and Hallett 1990) The solute/solution lines and the ice/solution lines intersect and below and either side of this intersection point *either* phase may nucleate leading to crystal growth driving the phase to the respective equilibrium line. Once this happens the other phase may nucleate; if a eutectic is possible the remaining solute crystallizes. In either case ice will be formed. Thus evaporating solutions lead to ice nucleation in the range of temperature -20$^o$C to -30$^o$C, depending on the solute.

## 9. CONCLUSIONS

The following results were obtained from this study:

(a) The frequency of occurrence of the phase composition coefficient, $\mu_3$=IWC/TWC, was obtained for seven 5-degree temperature intervals for the temperature range from 0$^o$C to -35$^o$C. The frequency of occurrence of $\mu_3$ was found to have explicit minima in the range 0.1<$\mu_3$<0.9 and two maxima for $\mu_3$<0.1 (liquid clouds) and $\mu_3$>0.9 (ice clouds) in all temperature intervals.

(b) The observed frequency of occurrence of $\mu_3$ suggests the typical glaciation time of clouds is much less as compared to the residence time of cloud particles, i.e. $\tau_g << \tau_{resid}$.

(c) The IWC averaged over all clouds was found to be almost constant with temperature (Fig. 3). TWC and LWC are decreasing with the decrease of temperature.

(d) The average concentration of cloud particles measured by two FSSPs in glaciated clouds was found to be approximately constant at -35$^o$C<$T$<0$^o$C and it changes in the range from 4 cm$^{-3}$ to 6 cm$^{-3}$. The observation of a constant concentration of ice particles over all cloud temperatures may be interpreted as an indication that there exists a universal mechanism of ice formation in tropospheric clouds. No simple explanation at this time is possible for the phenomenon and it requires further studies.

(e) The average concentration of particles measured by FSSP in “liquid” clouds was found to decrease with a decrease of temperature. It changes from approximately 200 $cm^{-3}$ at -10°C to 30$cm^{-3}$ at -35°C.

(f) The average $D_3$ in glaciated cloud varied between 20 and 35$\mu$m having a maximum around -15°C and then decreasing towards cold temperatures.

(g) No spatial correlation between LWC and IWC was found in stratiform clouds. The correlation coefficient between LWC and IWC on average varies from 0 to 0.3.

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## REFERENCES

Arnott, W.P., Mitchell, D.L. Shmitt, C. Kingsmill, D. Ivanova, D. and Poellot, M. 2000: Analysis of the FSSP performance for measurement of small crystal spectra in cirrus clouds. *Proceedings of 13th Int. Conf. On Clouds and Precipitation*, Reno, 14-18 August, 2000, 191-193

Borovikov, A.M., Gaivoronskii, I.I., Zak, E.G. Kostarev, V.V. Mazin, I.P. Minervin, V.E. Khrgian, A.Kh. and Shmeter, S.M. 1963: *Cloud physics*. Israel Program of scientific translations, Jerusalem, 393

Cober, S.G., G.A. Isaac, and A.V. Korolev, 2001: Assessing the Rosemount icing detector with in-situ measurements. *J. Atmos. Oceanic Technol*., **18**, 515-528.

Cober, S.G., Isaac, G.A. Korolev, A.V. and Strapp, J.W. 2001: Assessing cloud-phase conditions. *J. Appl. Meteor*., **40**, 1967-1983

Cooper W.A. and Vali, G. 1981: The origin of ice in mountain cap clouds. *J. Atmos. Sci*., **38**, 1244-1259.

Dye, J.E., Jones, J,J., Winn, W.P., Cerni, T.A., Gardiner, B., Lamb, D., Pitter, R L., Hallett, J., and Saunders, C. 1986: Early electrification and precipitation development in a small, isolated Montana Cumulonimbus. *J. Geoph. Res*., **91**, 1221-1247

Fletcher, N.H. 1962 *Physics of Rain Clouds*. Cambridge University Press, Cambridge

Gayet, J.F., Febvre, G. and Larsen, H. 1996: The reliability of the PMS FSSP in the presence of small ice crystals. *J. Atmos. Ocean Techn*., **13**, 1300-1310

Gultepe, I., Isaac, G.A. and Cober, S.G. 2001: Ice crystal number concentration versus temperature. *Int. J. Climatol*., **21**, 1281-1302

Hallett, J. 1968: Nucleation and growth of ice crystals in water and biological systems, *in Low temperature biology of foodstuffs*, Ed. J. Hawthorne, Pergamon Press, 23 -52

Hallett, J. 1979: Measurement of size, concentration, and structure of atmospheric ice particles by the airborne continuous particle replicator. AFGL-TR-76-0149 Airforce Geophysics Laboratory, Hanscom, AFB MA 01731

Hallett, J. and Mossop, S. C. 1974: Production of secondary particles during the riming process, *Nature,* **249**, 26 –28

Heymsfield, A. 1972: Ice crystal terminal velocities. *J. Atmos. Sci*., **29**, 1348-1357

Heymsfield A.J. and Platt, C.M.R. 1984: A parameterization of the particle size spectrum of ice clouds in terms of the ambient temperature and the ice water content. *J. Atmos. Sci*., **41**, 846-855

Hobbs, P.V. and Rangno A.L. 1985 Ice Particle Concentrations in Clouds. *J. Atmos. Sci*., **42**, 2523–2549.

Isaac, G.A., and Schemenauer, R.S. 1979: Large Particles in Supercooled Regions of Northern Canadian Cumulus Clouds. *J. Appl. Meteor*., **18**, 1056–1065

Isaac, G.A., S.G. Cober, J.W. Strapp, A.V. Korolev, A. Tremblay, and D.L. Marcotte, 2001: Recent Canadian research on aircraft in-flight icing. *Canadian Aeronautics and Space Journal*, **47**, 213-221.

Ivanova, D., Mitchell, D.L. Arnott, W.P. Poellot M. 2001: A GCM parameterization for bimodal size spectra and ice mass removal rates in mid-latitude cirrus clouds. *Atmos. Res.,* accepted for publication

King, W. D., Parkin D. A. and Handsworth, R. J. 1978: A hot-wire water device having fully calculable response characteristics. *J. Appl. Meteor*., **17,** 1809-1813

Knollenberg, R. G. 1981: Techniques for probing cloud microstructure. In: *Clouds, their formation, Optical properties, and Effects*, P. V Hobbs, A. Deepak, Eds. Academic Press, 495 pp.

Korolev, A.V. 1998: About a definition of liquid, mixed and ice clouds. *Proc. Of FAA Workshop on Mixed-Phase and Glaciated Icing Conditions*, Dec.2-3 1998, FAA, Atlantic City, NJ, 325-326.

Korolev, A. V., Strapp, J. W. Isaac, G. A. and Nevzorov, A. N. 1998: The Nevzorov airborne hot-wire LWC-TWC probe: principle of operation and performance characteristics. *J. Atmos. Oceanic Technol.* **15**, 1495-1510.

Korolev, A.V., Isaac, G.A. Cober, S., Strapp, J.W. and Hallett, J. 2002: Observation of the Microstructure of Mixed Phase Clouds. *submitted for publication*

Korolev, A.V. and Isaac, G.A. 2002: Phase transformation in mixed phase clouds. *submitted for publication*

Korolev, A., Isaac G.A., and Hallett, J. 2000: Ice particle habits in stratiform clouds. *Q. J. Roy. Meteor. Soc.*, **128**, 2873-2902.

Korolev, A., Isaac, G.A., Mazin I.P. and Barker, H. 2001: Microphysical properties of continental clouds from in-situ measurements. *Q. J. Roy. Meteor. Soc.*, **127**, 2117-2151

Mazin, I. P., A. V. Korolev, A. Heymsfield, G. A. Isaac, and S. G. Cober, 2001: Thermodynamics of icing cylinder for measurements of liquid water content in supercooled clouds. *J. Atmos. Oceanic Technol.,* **18**, 543-558.

Mazin, I.P., Nevzorov, A.N., Shugaev, V.P. and Korolev, A.V., 1992: Phase structure of stratiform clouds. . *Proceedings of 11th Int. Conf. On Clouds and Precipitation*, Montreal, 17-21 August, 1992, 332-336.

Meyers, M.P., DeMott, P.J. and Cotton, W.R. 1992: New Primary Ice-Nucleation Parameterizations in an Explicit Cloud Model. *J. Appl. Meteorol.* **31**, 708–721

Moss, S.J., and Johnson, D.W. 1994: Aircraft measurements to validate and improve numerical model parameterization of ice to water ratios in clouds. *Atmos. Res.*, **34**, 1-25.

Mossop, S.C., Ono, A. and Wishart, E.R. 1970: Ice particle in maritime near Tasmania. *Q. J. Roy. Meteorol. Soc*., **25**, 889-508

Nevzorov, A. N. 1983: Aircraft cloud water content meter. *Transactions of Central Aerological Observatory (Trudi TsAO)*, **147**, 19-26 (in Russian).

Nevzorov, A.N. 2000 Cloud phase composition and phase evolution as deduced from experimental evidence and physico-chemical concepts. *Proceedings of 13th Int. Conf. On Clouds and Precipitation*, Reno, 14-18 August, 2000, 728-731

Peppler, W. 1940: Unterkühtle Wasserwolken und Eiswlken. *Foschg. U. Erfahrung*, Reichsamt f. Wetterdiennst. B., No 1

Queen B. and J Hallett, J. 1990 Crystallization of highly supersaturated solutions. *Proc. AMS Conf. Cloud Physics*, San Fancisco CA, 97 - 100

Roberts P. and Hallett J.1968: A laboratory study of the ice nucleating properties of some mineral, particulates. QJRMS vol 94 25 -34

Rosinski, J., and Morgan, G. 1991: Cloud condensation nuclei as a source of ice forming nuclei in clouds. *J. Aerosol Sci*., **122**, 123-133

Ryan, B.F., Wishart, E.R. and Shaw, D.E. 1976: The growth rates and densities of ice crystals between -3$^o$C and -21$^o$C. *J. Atmos. Sci.,* **33** 53-70.

Strapp, J.W., Chow, P. Moltby, M. Beezer, A.D. Korolev, A.V. Stronberg, I. and Hallett, J. 1999: Cloud microphysical measurements in thunderstorm outflow regions during Allied/BAE 1997 flight trials. *AIAA 37th Aerospace Sci. Meeting and Exhibit*, Reno, Nevada, 11-14 January 1999, AIAA 99-0498, pp10.

Tremblay, A., Glazer, A. and Benoit, W.Yu,R. 1996: A mixed-phase cloud scheme based on a single prognostic equation. *Tellus*., **48**, 483-500.

Wallace J.M. and Hobbs, P.V. 1975: *Atmospheric Science: An introductory survey*. Academic Press, New York

Weickmann H. 1945: Formen und Bildung atmosphärischer Eiskristalle. *Beitr . z. Phys. d. fr. Atmos.*, **28**,

Zak, E. G. 1937: A characterization of frontal clouds using aircraft soundings. *Soviet Meteorology and Hydrology*, **8**