LABORATORY SIMULATION OF HYDROMETEOR STRUCTURE RESULTING FROM CRYSTAL GROWTH FROM THE VAPOR ON PREVIOUSLY FROZEN SUPERCOOLED DROPLETS.

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1. INTRODUCTION

Under conditions typical for the troposphere, water vapor crystallizes with a sixfold-symmetry and forms a hexagonal solid crystals with two basal planes and six prism planes. Laboratory experiments revealed that the rate of propagation of the basal faces relative to that of prism faces varies with temperature and supersaturation in a characteristic manner (e.g. Nakaya 1954; Kobayashi 1957; Hallett and Mason 1958; and others). Observations summarized by Magono and Lee (1966) showed a variety of hexagonal structures in ice particles grown in natural clouds. The diffusional growth of ice crystals can be treated in the same manner as for drops by making an analogy between the governing equation and boundary conditions for electrostatic and diffusional problems. The diffusional growth equation can formally be applied for ice spheres. This approach was used in a number of numerical models for estimations of the rate of glaciation of clouds. However, the spherical growth of ice particles has been considered as a hypothetical possibility, which does not exist in nature.

The present paper describes laboratory experiments on diffusional growth of water vapor on ice spheres.

2. EXPERIMENTAL INSTALLATION

The thermal diffusion chamber used in this study has been developed in the Ice Research Laboratory of the Desert Research Institute (DRI). A schematic of the thermal diffusion chamber is shown in Fig. 1.

video window	top plate	nent sing crank thermocouple
camera	ice T_t gla gla $T_b < T_t$ l fila	ss ment
microscope	bottom plate	chamber wall

Figure 1. Schematic of the static diffusion chamber used in the study.

The chamber consists of two stainless steel plates separated by a short, thick, acrylic cylinder. The top plate has a stainless steel wick, which can be

Corresponding author : Alexei Korolev e-mail: Alexei.Korolev@ec.gc.ca saturated with water, and the bottom plate is covered by a 3mm thick layer of water. The temperatures of the two plates are independently controlled by refrigeration units, and once the liquid layers are frozen, a difference in temperature between the two creates an environment supersaturated with respect to ice. Two double paned windows set in recessed ports in the chamber wall allow illumination and observation. A small flow of dry air between the panes keeps them ice free so that crystal growth can be recorded with a time lapse camera and microscope system.

To study frozen droplet growth, ice crystals were initially nucleated (see Bailey and Hallett, this conference) and grown on 50µm glass filaments suspended in the center of the chamber at the temperature to be studied and with the plate temperatures set to give highly saturated conditions. Crystals were rapidly grown to various sizes and then were completely melted with a stream of dry nitrogen introduced via a small stainless steel tube from a port in the side of the chamber after the ice supersaturation had been reduced to zero or a nominally low value by making the two plates isothermal or nearly so. This allowed well dispersed drops to be formed of a predetermined size by evaporating most of the smaller drops left adhering to the filament. Once the desired droplet sizes were obtained, they were nucleated by briefly introducing a copper rod into the chamber that had been cooled with liquid nitrogen, freezing the drops by touching the glass filament or by contact nucleation due to small crystals nucleated by the cold rod in a slightly supersaturated chamber environment. Once crystals were nucleated, the temperatures of the two plates were changed to create a desired supersaturation and the growth was recorded. The glass filament could be rotated so that frozen droplets could be viewed from any angle as growth proceeded. The study was carried out at a laboratory air pressure of 850 hPa.

3. RESULTS

The experiments on ice growth were conducted for the temperatures from -6° C to -14° C and supersaturation over ice from 2% to 15%. The sequences of successive images of growing ice particles are shown in Figs. 2-6.

3.1 Case S_i=2%, T= -14°C

Figures 2a,b,c show a sequence of images of ice particles growing on a thread at supersaturation over ice S_i =2% and temperature T= -14°C. To analyze the growth rate of ice particles, the contour of the ice particle image at the moment of complete freezing was superimposed with those for the later moments of time (Fig. 2d,e). The characteristic size of the frozen droplets in the direction perpendicular to the thread at the moment of nucleation was about D_{min} =125µm. In one hour the size the ice particles increased up to D_{min} =165µm. Figures 2d,e show that within one hour the frozen droplets were growing approximately uniformly layer by layer.

3.2 Case $S_i = 6\%$, $T = -14^{\circ}C$

Figures 3a,b,c show images of ice particles growing at temperature T= -14°C and supersaturation over ice *S*_i=6%. This corresponds to approximately half water saturation at this temperature. The initial size of the top drop at the moment of freezing was D_{min} =220µm (Fig. 3a). Fig. 3 shows a spherical growth of ice spheres as in the previous case but at higher supersaturation. However, after about half an hour some ice crystals start to grow out of the surface of the top ice particle as seen in Figs. 3b and c.

3.3 Case $S_i = 6\%$, $T = -6^{\circ}C$

The growth of frozen drops was conducted at $T = -6^{\circ}C$ at an ice supersaturation near water saturation, which corresponds to approximately $S_i = 6\%$. Figure 4 shows the sequence of ice particle growth during 15 minutes after the moment of drop freezing. Under these conditions as clearly seen from Figs. 4b,c the layer growth of the frozen drops is concurrent with the growth of a number of hollow columns extending from the surfaces of the frozen droplets.

3.4 Case S_i=14%, T=-12°C

In this experiment the droplet of approximately 800µm in diameter was placed at the end of the thread. The droplet was frozen at low supersaturation and then the supersaturation started increasing (Fig.

5a,b). At approximately 12:23 (Fig. 5c), the ice supersaturation reached near water saturation, i.e. about 14% supersaturation over ice. After that moment, the humidity stayed approximately constant and the ice particles were grown under stable conditions. While the ice supersaturation increased towards water saturation, quasi-spherical growth of the frozen droplet was observed (Figs. 5f,g), and ice spears started to grow out of the frozen drop. During approximately 15 minutes from 12:23 to 12:38 at near water saturation, the frozen drop was completely covered by ice particles growing in radial directions, which made the spherical shape of the "source" ice particle indistinguishable.

4. **DISCUSSION**

4.1 Comparison of spherical ice growth with Maxwellian growth

The rate of mass growth of ice particles in the thermal diffusion chamber was estimated from images. The mass of the particles was calculated with the assumption that drops were prolate spheroids having an ice density of 900 kg/m³. Accuracy of the measurement of particle sizes from the images was about 15%. The estimated mass of the growth particles was compared with that calculated from the equation of diffusion ice growth

$$\frac{dm}{dt} = 4\pi CBS_i \tag{1}$$

Here S_i is the supersaturation over ice; *B* is a known coefficient dependent on pressure and temperature (Pruppacher and Klett, 1998); *C* is the capacitance, which is a function of particle geometry. For a prolate spheroid with semi-major and minor axes *a* and *b*

$$C = \frac{A}{\ln((a+A)/b)}; \quad A = \sqrt{a^2 + b^2}$$
 (2)

Figure 6 shows comparisons of experimental results with the theoretical calculations. As seen from Fig. 6, ice particle mass estimated for the case of spherical growth agrees reasonably well with that observed during growth in the thermal diffusion chamber.



Figure 2. (a-c) Sequence of images of frozen drops growing in thermal diffusion chamber at $T=-14^{\circ}C$ and $S_{i}=2\%$; (d) superposition of contours of images (a) and (b); (e) contours of images (a) and (c).



Figure 3. (a-c) Same as in Fig. 2. $T = -14^{\circ}C$ and $S_i = 6\%$.



Figure 4. (a-c) Same as in Fig. 2. $T = -6^{\circ}C$ and $S_i = 6^{\circ}$.



Figure 5. (a-c) Same as in Fig. 2. (a-b) $T = -12^{\circ}C$ and $S_{i0} < S_i < S_{w0}$; (c-e) $T = -12^{\circ}C$ and $S_i = 14\%$. (f) superposition of contours of images (a) and (b); (g) contours of images (a) and (c); (h) contours of images (a) and (d).



Figure 6. Comparison of the mass growth calculated from Eq.(1) in assumption of spherical growth and derived from experimental data.

4.2 Observation of frozen drops in natural clouds

Figure 7 shows NRC Convair 580 sounding data from a deep *As-Ns* cloud system in an approaching front over the northern part of Lake Ontario conducted during Canadian Freezing Drizzle Experiment 3 (CFDE3) (Isaac et al. 2001). The diagrams presented in Fig. 7 demonstrate vertical profiles of total (ice+liquid) and liquid water content measured by the Nevzorov probe, concentration of ice particles measured by FSSP and static temperature. From the analysis of the Nevzorov probe data (Fig. 7a) it can be concluded that the cloud near the top from 6100m to 5600 was liquid. The lower layer from 5600m to 4400m was classified as mixed phase. The layer between 4400m and 2900m was glaciated.



Figure 7. Vertical profiles of (a) total and liquid water content measured by the Nevzorov probe; (b) concentration of particles measured by two FSSPs in the ranges $3-47\mu m$ and $5-95\mu m$; (c) air temperature.

Figure 8 shows images of cloud particles measured by Cloud Particle Imager (CPI) (Lawson et al. 2001) at two different levels indicated by the arrows (1 and 2) on the right of Fig. 7. Fig. 8.1 shows images of drizzle size drops $100-200\mu m$ in diameter

generated by the top cloud layer. The terminal fall velocity of such drops at this altitude is about 0.4-1m/s. Fig. 8.2 shows the images of cloud particles taken approximately 1300m below compare to that in Fig. 8.1. At least eight particles in Fig. 8.2 may be identified as frozen drizzle. Some of the frozen drops are stuck to bigger ice particles. It is Interesting to note that the frozen drops in Fig. 8.2 kept spherical or quasi-spherical shapes and none of them had developed any significant ice crystal structures on their surface. The air in the mixed cloud layer due to the presence of liquid droplets was close to saturation over water. It was not clear how long these drops stayed frozen, however, the maximum time can be estimated as 15-20 minutes, the time required for them to fall out of the top liquid layer and reach the level associated with Fig. 8.2.

Laboratory experiments discussed above show that frozen drops during some period of time may grow as spheres. The characteristic time of deviation of the shape of particle depends on supersaturation and temperature.



Figure 8. Images of cloud particles measured by CPI at two different levels indicated by arrows on the right in Fig. 7

Analysis of cases 3.3 and 3.4 gives a characteristic time, when at near water saturation for a frozen drop some hundreds of micrometers in diameter to become completely hidden by crystals growing from the vapor overt its surface. At lower ice supersaturation, the frozen droplet may keep a spherical or quasi-spherical shape during a longer period of time. In this regard it would be relevant to mention the work of Juisto and Weikcmann (1973) discussing nucleation of ice particles on frozen droplets.

It is assumed that this time also depends on particle size. Large frozen drops may keep their spherical shape for a longer period of time.

5. CONCLUSIONS

Within the contest of this study, the following results were obtained:

- (a) Spherical growth of frozen droplets with diameter larger than 100μm was observed in the thermal diffusion chamber.
- (b) The characteristic time of spherical growth depends on temperature and supersaturation. At low supersaturation (S_i< than half of saturation over water) the frozen droplet may keep a quasispherical shape during tens of minutes. At higher supersaturation individual ice crystals start to grow in different directions out of the frozen drop eventually completely changing their shape.
- (c) It is not uncommon to observe in clouds frozen drops in the form of almost smooth spheres. This suggests that the droplets were either recently frozen or had been growing at low supersaturation.

Ice nucleation through freezing cloud droplets plays an important role in the formation of ice in tropospheric clouds. The above results contributeto a better understanding of the evolution of ice particles during their diffusional growth.

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