ICE PARTICLE GROWTH UNDER CONDITIONS OF THE UPPER TROPOSPHERE

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ABSTRACT: Atmospheric conditions for growth of ice crystals (temperature and ice supersaturation) are often not well constrained and it is necessary to simulate such conditions in the laboratory to investigate such growth under well controlled conditions over many hours. The growth of ice crystals from the vapour in both prism and basal planes was observed at temperatures of -60 °C and -70 °C under ice supersaturation up to 100% (200% relative humidity) at air pressures derived from the standard atmosphere in a static diffusion chamber. Crystals grew outward from a vertical glass filament, thickening in the basal plane by addition of macroscopic layers greater than 2 µm, leading to growth in the prism plane by passing of successive layers conveniently viewed by time lapse video.

Keywords: Ice crystal growth; layer growth; laboratory ice study; NOx

1. INTRODUCTION

The purpose of this study is to show that crystals may form through the deposition of water vapour in layers on one crystal surface, as one part of the process of forming upper troposphere ice crystals. An ice crystal macroscopic layer is defined as a layer of new growth on an ice crystal, occurring on one facet of the ice crystal. To be observed in the laboratory, layers must have sufficient thickness (in this case 2 µm) to be visible in the microscope images. Layer growth is calculated as the thickness of each new macroscopic layer, times the rate at which new macroscopic layers form (Nelson and Knight 1998). After a layer initially forms, the subsequent changes in thickness of the new layer are at least five times less than the overall thickness of the crystal.

The currently accepted mechanism for the formation of macroscopic layers is step bunching. Microscopic layers (defined by Markov (2003) as tens of molecules thick) on a crystal surface do not necessarily grow at the same rate. This occurs due to competition between layers that start close to each other (Pruppacher and Klett 1997). As faster propagating layers catch up to slower propagating layers, the result is a step sufficiently thick to be visible through a microscope.

To avoid confusion, the term "macroscopic layer" is used in this paper, following Markov's (2003) "macrostep" which was a minimum of hundreds of molecules thick. This is the first study to examine macroscopic layers growing in the prism plane in tropical cirrus conditions. Layered growth on the prism plane initiates by growth exclusively in the a-axis through the addition of new layers. Each layer propagates in the c-axis direction through additional vapour deposition that occurs preferentially on the layer edge, with no further growth in the rest of the crystal; the crystal only gets thicker at a point by passing layers.

2. EXPERIMENT

In this experiment ice crystals were grown in a static diffusion chamber similar to Bailey and Hallett (2002, 2004) consisting of two stainless steel plates separated by a short acrylic annular ring (Figures 1, 2). The top and bottom plates were ice-coated stainless steel, with copper coils bolted to the outside surfaces of the plates, with cooling fluid from refrigerators to chill the plates to desired temperatures, and independent temperature control for each plate to within 0.3º C. The plates were separated at the outer edge by a hollow acrylic cylinder that allowed a width to height ratio of 10:1, to eliminate thermal and vapor convection effects from the edges of the chamber.

Air was filtered before it was brought into the chamber to remove ambient aerosols in laboratory air. The substrate for growing ice crystals was a glass thread drawn to a thickness of 50 to 70 micrometers from soda-lime glass rods and suspended from the top plate in the middle of the chamber. Soda-lime was chosen due to its chemical similarity to kaolinite and other common mineral dust nucleating aerosols in the atmosphere. Cracks in the thread serve as preferential sizes for water vapor deposition. The top plate was always set to a higher temperature than the bottom plate to maintain convective stability. Supersaturation was determined as a function of chamber height from the temperature profile of the chamber, with the assumption that the layer of air just touching the top and bottom plates was at ice saturation, equivalent to zero ice supersaturation. This method is valid for temperatures between 0 and -100º C, and is further outlined in Bailey and Hallett (2002).

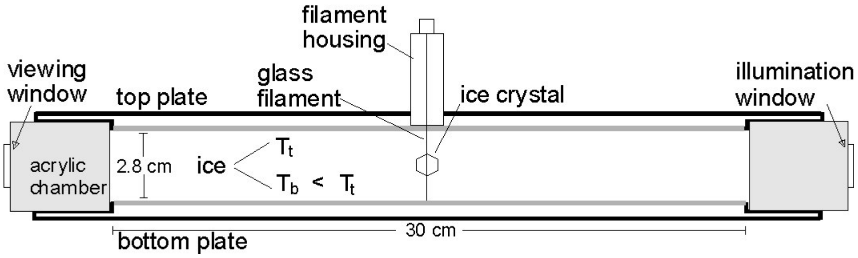


Figure 1: Diffusion chamber schematic. T_t indicates the temperature of the top plate, and T_b indicates the temperature of the bottom plate. The space between the acrylic walls and the plates is air-filled at a pressure equivalent to the standard atmosphere.

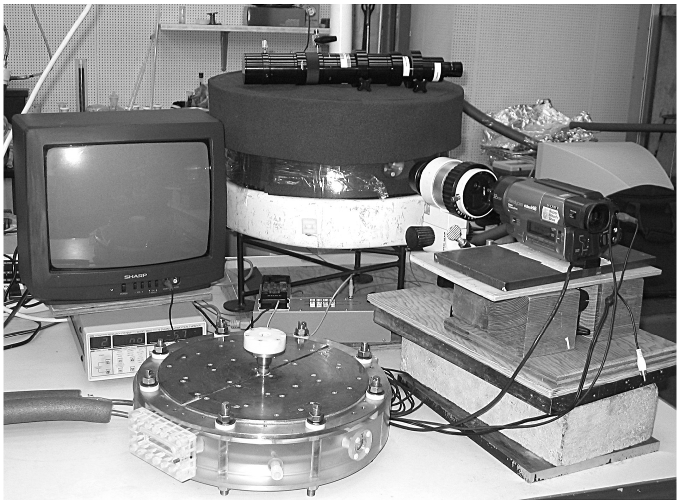


Figure 2: Diffusion chamber with stainless steel plates. The actual chamber is in the foreground, while a similar chamber constructed of aluminum is shown with insulation in the background to show how the chamber is used operationally. The microscope and camera are on the right hand side, with a monitor for real-time viewing of microscope images.

The primary benefit of using a laboratory experiment to study ice crystal growth is the greater control of temperature and supersaturation as compared with studies in the real atmosphere. The experimental setup allowed a range of supersaturations inside the chamber between just saturation with respect to ice and 1.01, with temperatures between 0 and -70° C. The uncertainty in temperature for each plate was 0.3º C. The value of ice supersaturation was taken to be that at the vertical center of the chamber, and varied by approximately 1%. (Ice supersaturation is not linear with height in the chamber.) Ice crystals observed were within 1 mm of the vertical center of the chamber, leading to an error in supersaturation no more than 1%. Chamber air pressures were consistent with the standard atmosphere, with 550 mb at -20° Celsius decreasing to 150 mb at -70° C. For this study chamber temperatures of -60 and -70° C were used, in order to best simulate conditions in the tropical tropopause within the limits of this apparatus. While shadowing was an effect for crystals that grew close to other crystals, crystals formed early on during the growth run were able to grow sufficiently far from the thread as to grow free of vapor competition from neighboring crystals, as judged by uniformity of layering velocity. The higher heat conductivity of the glass thread as compared to air leads to accelerated growth, which partially offsets shadowing. Images of ice crystals grown in the chamber were obtained using a video camera attached to a microscope to achieve time-lapse photography.

3. RESULTS AND DISCUSSION

56 crystals were grown at temperatures of -60° C (49 crystals) and -70° C (7 crystals), and ice supersaturations from 12% to 101%. Each crystal discussed below was observed by video with derived measurements of length (c-axis), width (a-axis), and the presence of layers. Layered growth was observed in 6 crystals, all at a temperature of -60° C and ice supersaturation of at least 25% (Table 1). The majority of crystals observed, including all crystals at -70° C, did not have visible macroscopic layers.

S_i	25%	50%	101%	Total
# of crystals	17	24	8	49
# with macroscopic layers	3	2	1	6

Table 1: Summary of ice crystals observed at -60° C and S_i greater than or equal to 25%. Out of 49 crystals observed, 6 had macroscopic layers.

Figure 3 shows the progression of macroscopic step growth in an ice crystal grown at -60° C and 101% ice supersaturation. The high supersaturation present was favorable for layered growth to proceed, though growth was shadowed on the right hand side of the crystal. The added crystal layers emerge from the bottom, with the top layer being the original ice crystal. It is important to note that the second new layer originates from the left hand side of the crystal, not from the filament. Therefore, it is established that layered growth is able to occur apart from the presence of a nucleating surface such as a glass filament. The crystal thickened by passage of layers only; furthermore, the newest layer also lengthened the fastest of all layers. This occurred due to the newest layer having the least competition for water vapour of all crystal surfaces.

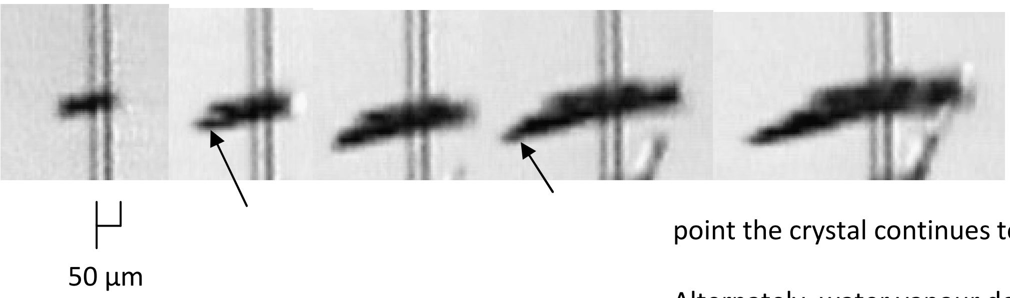
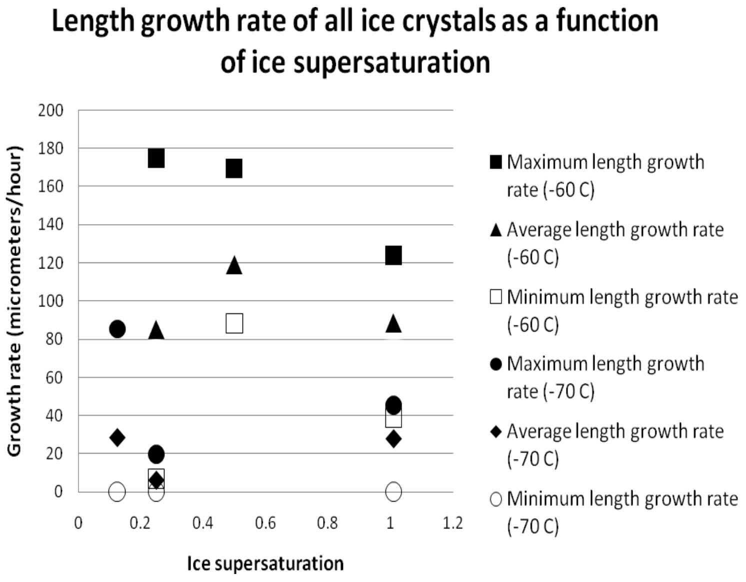


Figure 3: An ice crystal grown at -60±1 °C and 101±1% ice supersaturation. This and other crystals were examined for rates of lengthening, thickening, and the presence or absence of layered growth. Growth rates were measured on the ends of crystals furthest away from the filament, so they would not be influenced by neighboring crystals. In this image each new step is indicated by an arrow.

The growth of ice crystals via layered growth divulges important information about the crystal itself. Assuming the distribution of water vapour in the surrounding air is roughly uniform, water vapour molecules are traveling via surface migration across non-growing regions of the ice crystals to the growing regions. The water vapour molecules then deposit when they reach either a layer on the ice crystal surface or other localized nucleation sites, such as an exposed region near the edge of the original crystal or a layer edge, at which point the crystal continues to grow. Alternately, water vapour deposition may occur at a local maximum in ice supersaturation (Hallett 1987). This site then becomes the point of nucleation for the formation of the new layer at the crystal edge. When ice crystal surfaces are not in the vicinity of local maxima of ice supersaturation and/or do not contain nucleation sites, layered growth would not be expected to occur, as with the majority of crystals in this study.

Figure 4 shows the minimum, average, and maximum rates of growth for crystals observed in this study. The large rate of thickening of one crystal at -60° C and 25% ice supersaturation was caused by the glass filament; this effect was not present in any other crystals observed, and was only present for one hour in this crystal. In all sets of conditions crystals were observed on average to lengthen faster than they thickened. The lack of an overall increase in growth rate with increasing ice supersaturation can be explained by first assuming that layers begin to advance in the c-axis direction (lengthening) through deposition of water vapour molecules at the base of the layer, as this is the most energetically favorable location for water vapour molecules to attach to the ice crystal. Further growth away from the crystal in the a-axis direction (thickening) requires sufficient supersaturation for the process to be energetically favorable. Based on these results the necessary ice supersaturation is greater than 100%. Since crystals such as those in Figure 3 do not grow except through the addition of layers, the result is that an increase in ice supersaturation does not necessarily lead to an increase in the growth rate of ice crystals.



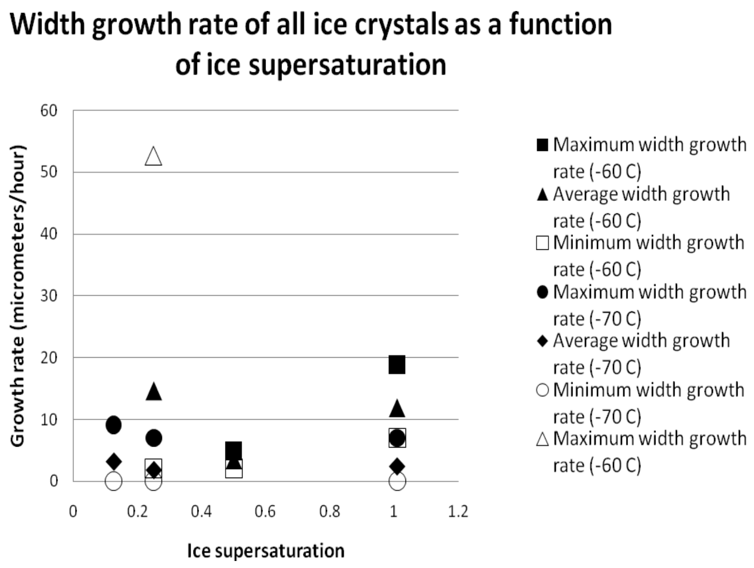


Figure 4: Rate of growth in length (top graph) and width (bottom graph) of all ice crystals observed, as a function of temperature and ice supersaturation. Comparison of the two graphs shows that crystals overall lengthened faster than they thickened. The outlier on the bottom graph for maximum width growth at 0.25 ice supersaturation and -60 °C (open triangle) was due to a single instance of enhanced thickening due to the glass thread, illustrating the difference between growth solely caused by the environment and growth caused by the filament. Crystals that have growth solely influenced by the filament have been ignored for the purpose of this study; the outlier is shown as an example of where it was not ignored.

4. ICE CRYSTALS GROWN IN THE PRESENCE OF NO_x

A similar study was undertaken to examine for the effects of NO_x on ice crystal growth. This study focused on ice crystal habit rather than growth rates, though crystals were qualitatively examined for observable changes in growth rate. A chamber with similar characteristics, including cooling coils and a glass thread, was used to grow ice. Images of ice crystals were obtained using a video camera, as before.

The main difference in this study was the temperature, ice supersaturation, and chamber pressure present. The first test was undertaken with a top plate temperature of -

12° C, and a bottom plate temperature of -27° C, for an ice crystal growth temperature of -19.5° C. The initial chamber pressure was 650 mb. NO_x was added to the chamber after an ice crystal had been growing for one hour (Figure 5), raising the pressure to 700 mb. The result of this test was a change in ice crystal habit due to thickening of crystals as a result of adding a small amount of NO_x to the chamber and observing the crystal for an additional hour.

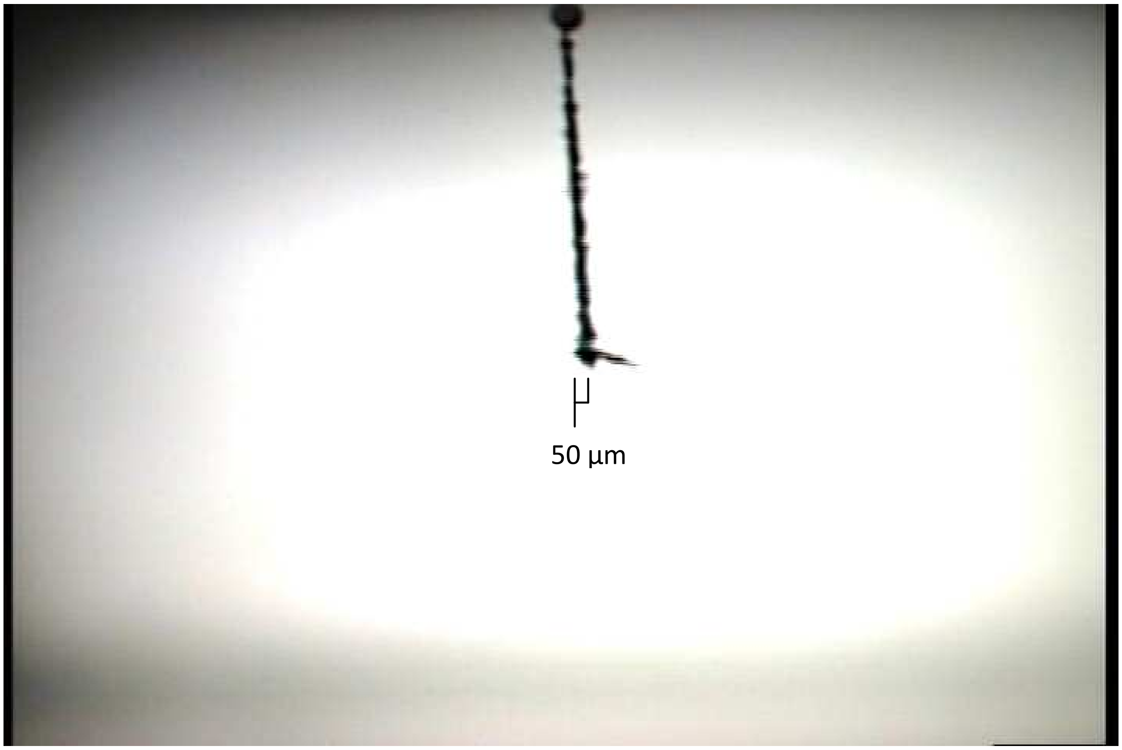


Figure 5: Ice crystal growth after one hour. In this test, NO_x levels were equal to the background concentration of 0.003 ppm, +/- 0.001 ppm.

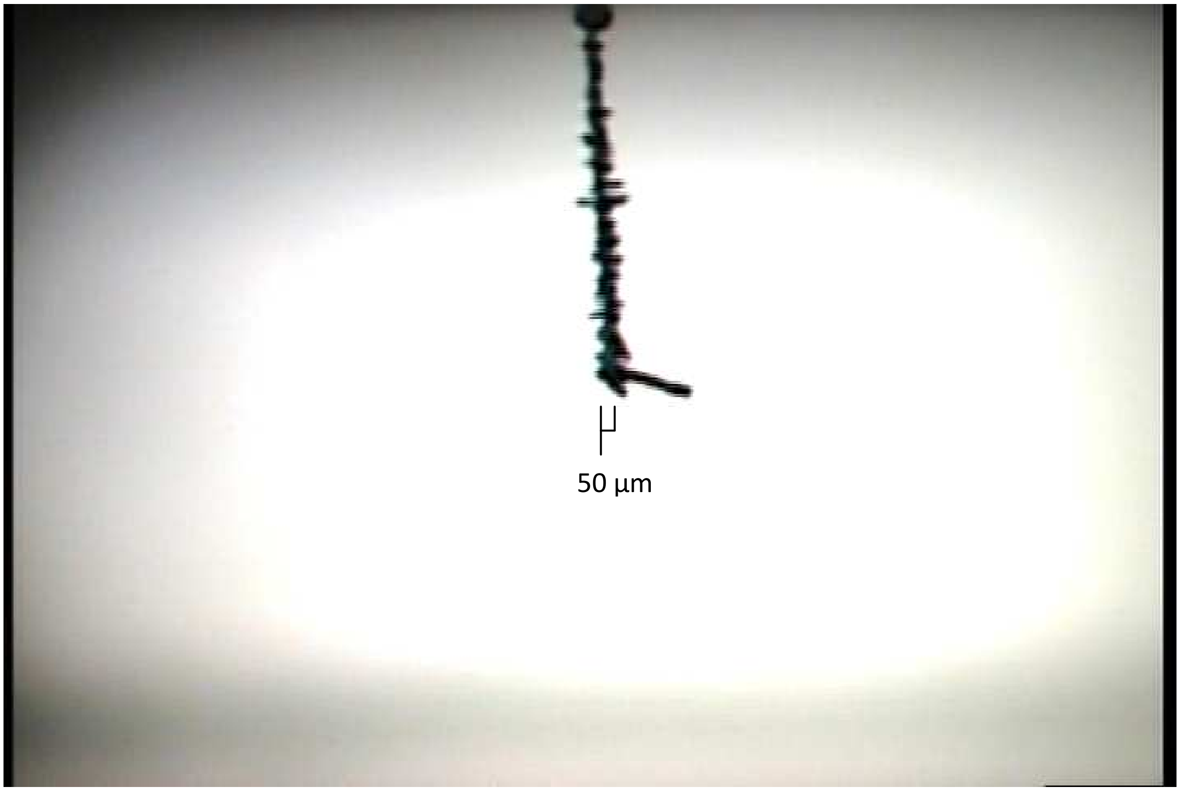


Figure 6: One hour after the picture in Figure 5 was taken. After taking the picture in Figure 5, nitrogen gas with 5.17 ppm NO was added to the chamber to bring the pressure from 650 to 700 mb. Therefore, the concentration of NO in the chamber was 0.37 ppm. After adding NO, some crystals (note the one on the bottom of the glass thread) grew thicker at a rate faster than they grew longer.

A second test was undertaken by evacuating the chamber, then filling it with 5.17 ppm NO in nitrogen, and then growing ice crystals under the same temperature, pressure, and ice supersaturation conditions (Figure 7). In this case, there was no observable difference in habit as compared to growing ice in background NO concentrations.

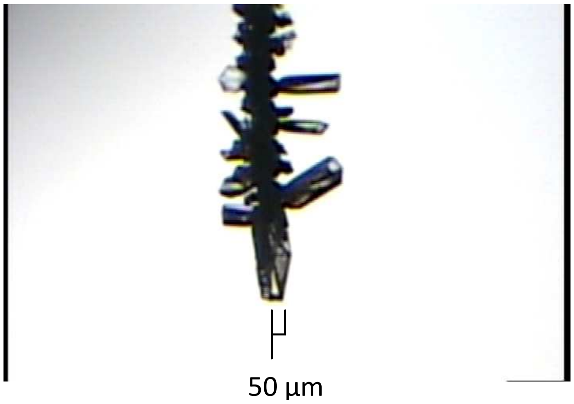


Figure 7: Ice crystals grown in air with 5.17 ppm NO. Crystals grew in habits expected if the chamber air had background concentrations of NO_x. Therefore, it is assumed that the presence of NO_x prior to the start of crystal growth does not have an effect on ice crystal habit.

The first test was then repeated using the same temperature and ice supersaturation, but a starting pressure of 627 mb and an ending pressure of 727 mb when NO was added. In this case the crystals not only grew thicker, but overall faster than in background NO concentration air. This indicates a significant effect, as the increase in chamber pressure would otherwise decrease the vapor diffusivity and thus decrease the growth rate of ice crystals. Figure 8 shows the ice crystals after growing in air with background NO concentrations for one hour. Figure 9 shows the same ice crystals after NO was added and the crystals were allowed to grow for an additional 20 minutes.

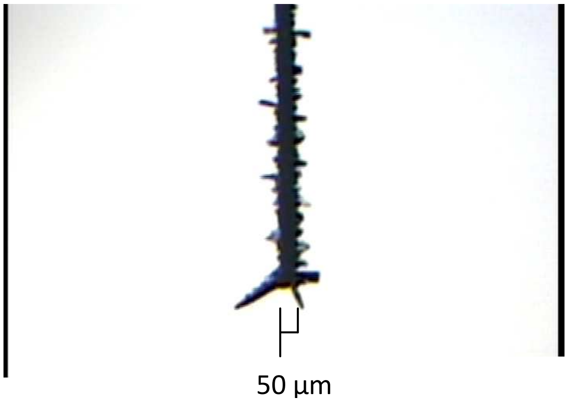


Figure 8: Ice crystal growth over the course of one hour, before adding NO. This is assumed to be normal growth for this temperature, pressure, and ice supersaturation. Ice crystals were not present before the start of this time period.

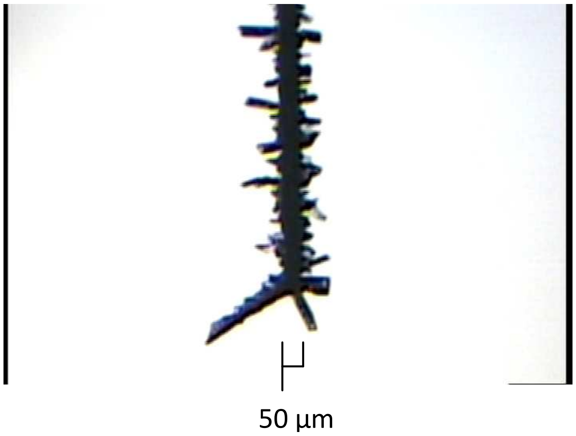


Figure 9: Ice crystal growth after adding NO and allowing the crystals to grow for 20 minutes. The concentration of NO is 0.71 ppm. Note that some crystals have doubled in size, indicating a threefold increase in growth rate (since the time interval is three times less). Also note the thickening of many crystals, especially the one on the lower right of the thread.

5. CONCLUSIONS

The laboratory study described above allows constraining of temperature, air pressure and ice supersaturation environment for the growth of ice crystals. Six out of 49 ice crystals above 12% ice supersaturation grew through the additional of layers on the prism plane. This agrees with the results of Anderson and Hallett (1979) and Hallett et al (2002), which indicates that non-thickening crystals will begin to thicken only when the ice supersaturation is increased above a critical value. Lan et al (2006) also predicted 20-30% ice supersaturation as the minimum needed for steps to form via surface diffusion.

In future studies additional time and special resolution will be of interest, allowing for measurement of the rate of deposition of molecular layers. Adding measurements at -70 °C as well as -80 °C will provide useful data; Bailey and Hallett (2004) and Heymsfield (2007) found that crystals of similar habit and mass are longer and thinner with decreasing temperature. Additionally, measurements at an ice supersaturation of 15% and 20% will be of interest, to further define the minimum ice supersaturation needed for macroscopic layers to form. Subsequent measurements above the minimum ice supersaturation will help to bound the maximum attainable thickness for macroscopic layers at a given temperature and ice supersaturation.

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