ORIGIN AND PECULIARITIES OF COLUMNAR TYPE CRYSTALS IN THE ATMOSPHERE

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ABSTRACT

After a brief introduction to the history of ice crystal observation in the atmosphere, several theories and hypotheses of the origin and evolution of columnar and bullet type crystals are discussed. The main conclusions of theoretical studies are confronted with the ice crystal observations made in the laboratory and in the free atmosphere as well. The cavities in the central part and at the basal faces of columnar type crystals often reveal the principal mechanism of the single column formation and also the potential effect of droplet freezing after coagulation.

INTRODUCTION

Countless varieties and precision of shape have attracted man to observe ice crystals and to relate their shapes to the state of the atmosphere before Christian era (Aristoteles, 384-322 B.C.; Chinese observations in the second century B.C.). Also the first sketches by Olaus Magnus, archbishop of Uppsala in 1555, and studies by R. Descartes (1637) and Hooke (1665) reveal their admiration for perfect shaped ice crystals. Outstanding amongst the observations of ice crystals in the 16th and 17th centuries stands the work of the astronomers at the court of Emperor Rudolf II at Prague. In an atmosphere of great discoveries and ideas which shaped the principles of modern science,Johann Kepler wrote in Latin a treatise on the hexagonal structure of snow (Strena Seu De Nieve Sexangula, Francofurti ad Moenum, apud Godefridum Tampach, Anno 1611). Observations of individual ice crystals led him to the formulation of the law of preservation of constant angles in a crystal, later to become one of the basic axioms of crystalography. The first attempts to classify ice crystals by Scoresby in his "An Account of the Arctic Regions" (Edinburgh, 1820), by Sekka and Zusetsu (Japan, 1833), and later by Hellmann in the monograph "Schneekrystalle" (Munchenberger, Berlin, 1893), reveal that the authors were very aware that besides star and plate type crystals there is a large population of columnar crystals found under specific atmospheric conditions.

This notion was stressed by admirable and enthusiastic work by A. Dobrowolski, who, as a member of the expedition which came to Antarctica by "Belgica" in 1887-1889, made hundreds of descriptions and drawings of individual ice crystals in polar regions. Several of his articles connect the existence of optical phenomena (halo) observed in nature with the columnar type crystals settling in the atmosphere (e.g., Dobrowolski, 1903). The importance of columnar ice crystals and the higher frequency of their occurrence at temperatures below -20°C was stressed by Westman

1903), who made valuable observations of ice crystals at Spitzbergen. Later Heim (1914) published statistics of ice crystal forms at low temperatures which cover mostly the crystal shapes in "diamond dust" and speculated on the relationship between the crystal shape and environmental temperature. He found that at -12°C, columns represented the minority of the total crystal population, at -23°C columns started to prevail, and at -27°C the vast majority of ice crystals were short columns. These observations on the ground and explanation of optical phenomena at higher altitude were thoroughly discussed by J.M. Pernter, and F.M. Exner in their "Meteorologische Optik" (Meterological Optics - W. Braunmuller, Wien, 1922). Since this time the ice crystals in free atmosphere have been assigned an important role in the so-called indirect aerology (see e.g., Weickmann, 1949).

However, turning the pages of the beautiful book "Snow Crystals" by W.A. Bentley, and W.J. Humphreys (McGraw-Hill Book Co., 1931 and Dover, 1962) where several decades of hard work and fine artistic taste culminated in a unique document inspiring both scientist and artist, one has the impression that columnar type ice crystals are not sufficiently represented. One might experience a similar impression looking at the illustrations in the old but important article by Shedd (1919) on "The Evolution of the Snow Crystal".

There are many books on cloud physics (e.g., Mason, 1971; Pruppacher and Klett, 1978) and monographs on ice crystal formation (Nakaya, 1954; Zamorskii, 1955; Klinov, 1960; and Hobbs, 1974) where the reader will find also excellent illustrations and references on many more recent investigations not mentioned in this paper.

The main aim of this short contribution is to point out several features of columnar type crystals observed in nature and in the laboratory, and in this way compensate to some extent for the prevailing attention paid to the morphology of star and plate type crystals.

SHAPE OF COLUMNAR CRYSTALS

Columns are ice crystals in which the ratio of the main to the side axis is equal to or larger than 1 and smaller than 7. Indeed, there is no sharp distinction between a thick plate with an axis ratio 0.8 and a column, and there is often a continuous transition from columns to needles beyond the ratio 7. The main axes of compact individual columnar crystals rarely surpass 1.0 mm and their fall velocities in the atmosphere usually amount to several tens of cm sec^{-1}.

The third class of the Classification of Ice Crystals accepted by the International Commission on Snow and Ice in 1949 includes columns with hollow space at the base, bullet type columns and aggregates of crystals (i.e., rossettes) besides individual compact columns. Many of the ice columns found in nature are rimed, and many columns bear plates at both ends. All these varieties of columnar type crystals are stressed and their origin explained in the more sophisticated ice crystal classification by Nakaya (1954). Nakaya based his genetical classification on many laboratory experiments during which columns originated mostly at low supersaturation relative to ice between -5°C and -13°C and again between -17°C and -21°C. At very high supersaturation (>10%) and low temperatures were found long hollow columns. This scheme was slightly modified by Shaw and Mason and by Kobayashi amongst others (Mason, 1971) (Fig. 1). The spectacular cyclic change in crystal habit (plate-column-dendrite-plate-column) around the mean temperature of dendrite occurrence (-15°C) can be explained by a cyclic change of crystal growth along the direction $[10\bar{1}0]$ and [0001].

Despite the considerable efforts of many investigators, it is unlikely that a complex model encompassing all processes participating in the ice crystal origin and crystal form transformation will be established very soon. There is still a considerable gap between the models based on the formation of a surface embryo and the advance of mono-molecular step as a random walk problem and the macroscopic diffusional model. In general, the evolution of a single unrimed columnar crystal from its basic form will be governed by several processes such as diffusional deposition of water vapor molecules on the crystal surface, and interaction between the mass transfer and hydrodynamic (thermodynamic) field around the falling crystal. A diffusional model enables one to calculate the increase of ice crystal mass Δm in the time Δt according to the formula

$$\frac{\Delta m}{\Delta t} = -4\pi CD(\rho_o - \rho_\infty) \qquad (1)$$

where C is the "capacity" related to the crystal shape; D is the coefficient of diffusion of water vapor molecules and ρ_o, ρ_∞ are the densities of water vapor above the crystal surface and far from it. Equation (1) describes the growth rate of an ice crystal if its "capacity" is known. Mason (1971) analyzed the values of individual terms of Eq. (1) and was able to show that the fastest crystal growth happens around -14° C. However, this approach and its extension into the "electrostatic analogy", elaborated in more detail by Marshall and Langleben (1954) and by Podzimek (1966), could not explain the various habits and crystal forms as a function of temperature observed in nature.

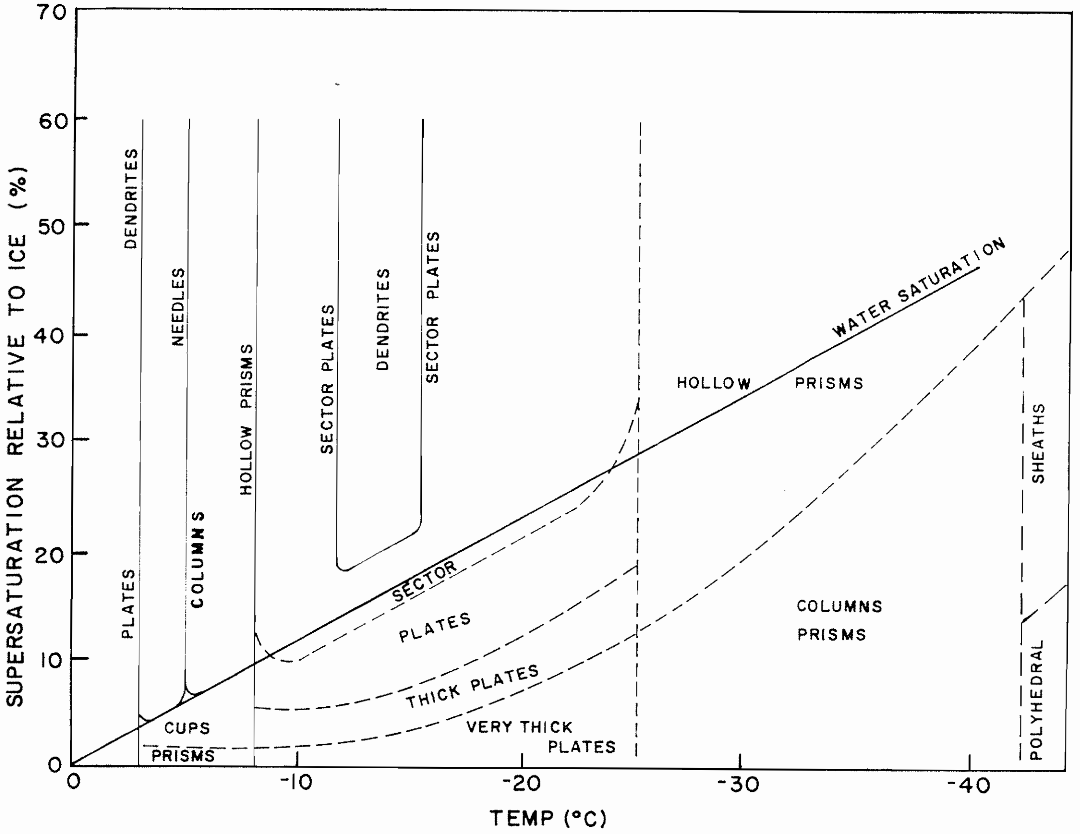

Fig. 1 Modified Nakaya's diagram of the variation of ice crystal habit with changing temperature-supersaturation.

Experimental studies performed during the last three decades have proved, in particular, the importance of surface diffusion of water vapor molecules in forming different habits of crystals (Bryant et al., 1959; Kobayashi, 1966). The molecules impacting the crystal are supposed to migrate for a certain time before being built into the crystal lattice or rebounded into the environment. Therefore, the process of crystal growth can be considered as a formation of surface embryo which grows by attachment of the migrating molecules. In this way steps originate on the crystal surface.

Several authors have successfully studied the motion of steps on the basal face of ice and deduced a simple formula for the velocity of the step growth (Hallett, 1961; Mason et al., 1963), namely

$$V = \frac{A[2x_s + h]}{\rho_i h} , \qquad (2)$$

where A is the net flux of impacting molecules which remain on the unit area of the crystal surface: x_s is the mean migration path (distance) of the molecules; h is the height of the step and ρ_i is the density of ice. One must emphasize that step height means an observed macrostep with a height of tens to hundreds of Å[10^{-8} cm]. Unfortunately, a relationship between the macrostep and microstep (often corresponding in theoretical calculation to the monomolecular layer which properties such as A or ρ_i are unknown) is not yet well established. Disregarding this fact, however, Mason and his fellow workers and later Kobayashi (1966) succeeded to find a dependence of the velocity of the step growth (which is for known A, ρ_i and h proportional to x_s) on the temperature. This discovery explains to a large extent the ice crystal habit evolution on a basal face, B (Fig. 2).

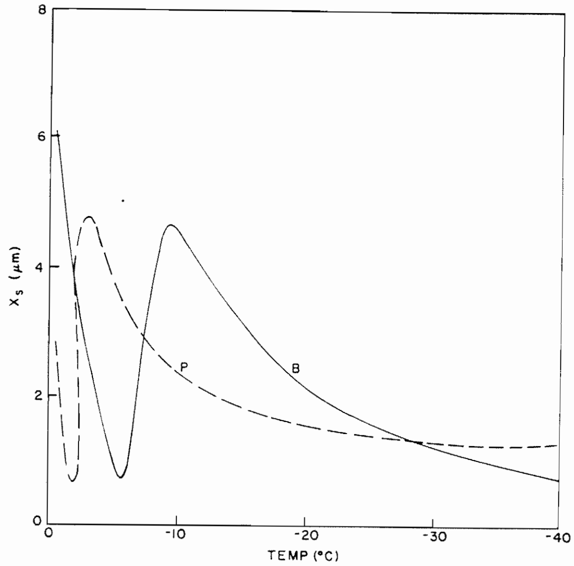

Fig. 2 Variation of the molecule diffusion path, x_s, with temperature, T[°C], on an ice basal face [0001], B, and on a prismatic face, P.

To the author's knowledge, no satisfactory theoretical explanation of the different migrational velocities of molecules on the prismatic and basal face of an ice crystal has yet been found, and very few experimental studies of the growth of prismatic face have been performed (e.g., Lamb and Scott, 1972). In order to overcome the existing standstill in the pursuit of a complex explanation of the ice crystal habit growth, Mason (1971) suggested a superposition of two x_s-curves, one for the basal (full line in

Fig. 2) and the other for the prismatic face (dashed line). This speculative approach to the solution of the cardinal problem of ice crystal growth is supported by past experiments which showed a dramatic change of crystal habits and well-defined critical temperatures for different habits in the temperature range - 2°C to -10°C and the successive transition of plates into columns below -22°C (Fig. 1).

The other factor influencing the shape evolution of an ice crystal is due to the interaction between both of the previous processes and the hydrodynamic flow bringing more water vapor molecules to the tips and edges of a crystal than to its plane faces. This process, governed mainly by the different gradient of water vapor density in the airflow deflected by an obstacle, was considered by Marshall and Langleben (1954) and expressed in the form of a "ventilation" factor. Its value deduced from dimensionless parameters (such as Reynolds and Best number) corrects the rate of growth of a quiescent ice crystal described by Eq. (1). (Pruppacher and Klett, 1978; p. 452). Because the vast majority of the crystals found in nature fall at Re < 50, one can assume the horizontal position of the axis of an unrimed column (Fig. 3) as the most probable and suitable for model flow calculation. For solid columns a simple formula for the drag coefficient was deduced (Podzimek, 1969)

$$C_D = A \sim Re^B \text{ ,} \qquad (3)$$

where the coefficients had different values for different ratio of the main to side axis (e.g., A = 5.5 and B = -0.306 for 1:1; A = 3.95 and B = -0.213 for 2:1; A = 3.17 and B = -0.306 for 4:1). However, the hollow space at the end of columns, the bullet type ends, plates at the ends of a column and riming cause many of the crystals observed in nature to tumble. This instability of motion is enhanced by the horizontal component of the wind and air turbulence.

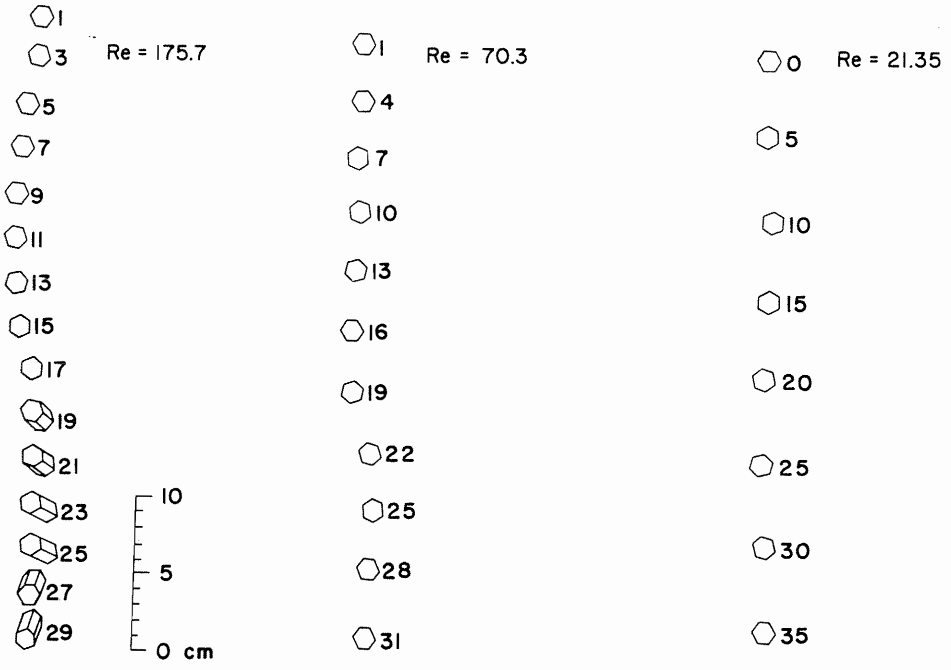

Fig. 3 Model of a columnar (prismatic) crystal with cavities and axes ratio of 2:1 falling in a liquid at different Reynolds numbers, Re. Speed: 24 frames per second.

PECULIARITIES OF HOLLOW COLUMNAR CRYSTALS

During several years of observation of ice crystals generated in a low pressure chamber and in nature the author was attracted by the formation of hollow spaces in columnar crystals. He was mainly interested in how the geometry of the hollow space reflected the physical state of the environment and how the potential irregularities of the shape reveal, for instance, the influence of crystal motion or the aggregation with other crystals.

The stylized drawing in Fig. 4 was produced from a photograph of an ice crystal taken when the crystal settled onto a microscopic slide covered by formvar (1% solution in ethylene dichloride by weight) in a 2 m^3 expansion chamber. The expansion started at a temperature of -24.6°C, ended at -29.1°C and the whole process lasted for four minutes. After 190 sec, during an expansion rate corresponding to an updraft velocity of 2 m sec^{-1}, white fog was observed, which started to convert to ice fog after 20 sec. At the time when the larger crystals settled through the ice cloud a sudden expansion corresponding to an updraft of 15 m sec^{-1} was applied for 15 sec. The shapes of the symmetrical cavities at the ends of the ice column truly reflect the process described above. The embryonal crystal, probably resulting from an impaction of two supercooled droplets was marked in the center of the column (A). If one makes a plausible assumption that the molecules are distributed equally from the place of impaction towards the prismatic and the basal faces, one can conclude that the velocity of an advancing macrostep on the prismatic face will be much larger than that on the basal face (2.8:1) close to the inner narrow cavity. The ratio of the velocities along the largest crystal cavity is 3.3:1 and this corresponds to the increased updraft of 15 m sec^{-1}. The spectacular cavities almost parallel to the crystal shape can be explained by the steadily increasing supersaturation to more than 25 percent during the expansion.

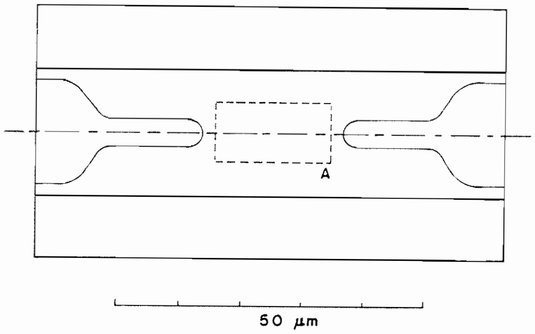

Fig. 4 Drawing of an ice crystal with cavities generated in an expansion chamber.

Fig. 5 Column (prism) 250 μm long with cavities on its faces.

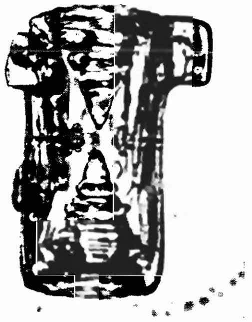

Fig. 6 Column 210 μm long with frozen drops on prismatic wall.

A different situation occurs in nature, where large scale humidity and temperature fluctuations often shape the cavities in a very bizarre way. Figure 5 represents a columnar crystal collected at high relative humidity in the neighborhood of geysers and hot pools in Yellowstone Park at a temperature of -30°C. One clearly sees the initial part of the cavity with a wider cone of basal cavity and the cavity on the prismatic face too. The ribs across the cone reveal inhomogeneities in the concentration of water vapor in the environment. The relative humidity close to the hot pools was very high, and the variety of origin and crystal path explain the features of several example crystals in Figs. 6, 7, and 8. The crystals were sampled in the morning hours on 6 January 1970 at the same place as the crystal in Fig. 5. They reveal their dwelling in a high supersaturation field (in Figs. 6, 7, and 8 crystals with large cavities) and also their stay in a normal atmosphere at low temperature (compact columns in Figs. 7 and 8). The cavities and shapes of aggregated crystals are different from what can be considered as a proof of the high sensitivity of the crystals to the field inhomogeneity. Many of the several thousands observed crystals have a separation line in the middle of the column, indicating the possible origin from frozen droplets. This finding is supported also by the prevailing ratio 2:1 of the c and a axis among the freshly formed ice crystals around the hot pools. For a temperature range - 29°C and -30°C the mean ice crystal concentration was for the axis ratio 1:1 around 55 in 1 liter of air, for 2:1 more than 618 and for 3:1 around 13 in 1 liter of air. In the temperature range -21°C to -25°C the ice crystal concentration corresponding to the same three axis ratios were: 22, 121, and 10 in 1 liter of air. The mean length of columnar type crystals at the lower temperatures was 65 *µ*m and about 120 *µ*m at higher temperatures.

Figures 6, 7, and 8 show columns with frozen drops on their surface which started to develop into crystals with the main axis parallel to that of the collector. It is interesting to compare this case with that of frozen drops deposited on the basal plane of a plate at the end of a short column (Fig. 9). One sees clearly that many of the droplets freeze and start to assume a hexagonal shape. This reveals interesting features of drops frozen on different faces of an ice crystal. The attached crystals from frozen drops assume the crystalographic structure of the substrate.

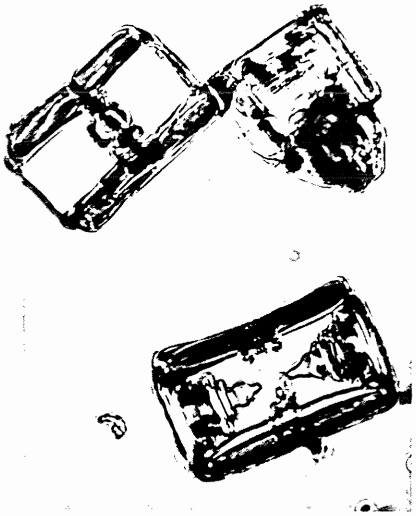

Fig. 7 Column with cavities and column 145 µm long with separation line between its two parts.

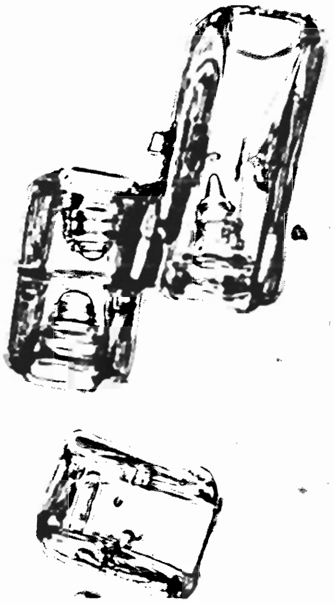

Fig. 8 Prisms with distorted cavities and one compact column 125 µm long.

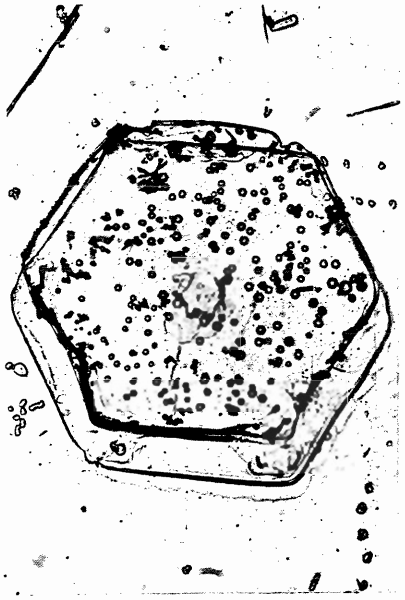

Fig. 9 Short column with two large plates 300 µm in size and frozen droplets.

A symmetry of the cavities is supposed to exist in the case of individual columnar crystals falling with their main axis in horizontal position (Fig. 5), whereas, the irregular and aggregated crystals (Figs. 7 and 8) have different cavities at both ends, revealing the interaction with the hydrodynamic field and the mass transfer between aggregated crystals. This simple picture has to be corrected, however, because a detailed analysis of several well replicated ice columns led to a surprising conclusion that the symmetry of cavities does not exist in reality. The two halves of crystal cross sections in Fig. 10 do not match in detail, although the mean shape is alike. That poses a new question of how far the main processes contributing to crystal growth do interact at the edges and inside of the cavities. In general, we see on all well-developed single columnar type crystals with cavities that the cavities on prismatic faces are much more alike than those on two basal faces. This fact is clearly related to the more steady ventilation of the central part of the long column and variable ventilation of the basal faces of a crystal which usually tumbles.

The other element disturbing the regular formation of cavities in columnar type crystals is the proximity of other crystals or the effect of droplets frozen on the crystal surface. Usually, the cavity tends to bend towards the larger ice mass and is deformed in accordance with the model of surface diffusion (ice sintering). However, the calculated values of linear step growth using an equation similar to Eq. (2) or assuming a more complex model of the linear growth rate due to a screw dislocation (Scott and Lamb, 1978) reveal still a considerable discrepancy between the values calculated for the growth of a prismatic and basal face and the observed growth rate at a specific temperature.

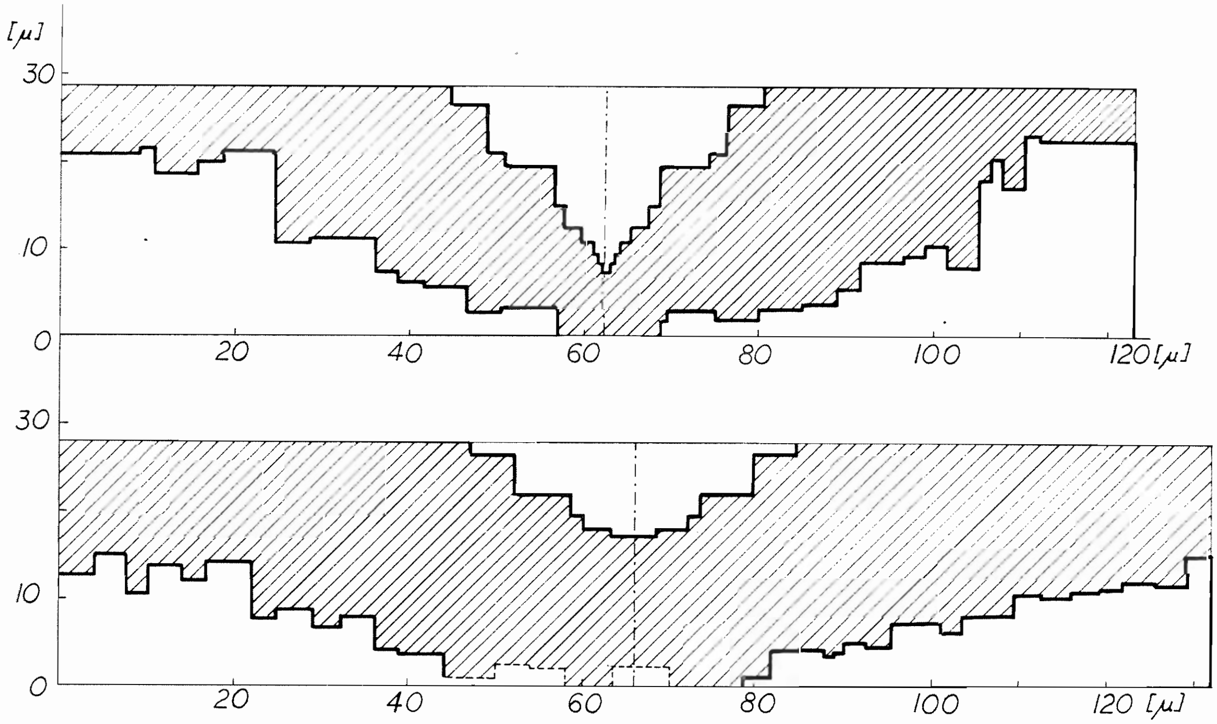

Fig. 10 Cross sections of two different columnar half-crystals collected at a Yellowstone Park hot pool at an air temperature close to -30°C.

CONCLUSION

In spite of some progress in explaining the ice crystal habit at different temperatures, it is not possible at present to provide an accurate calculation of the growth rate of a falling ice crystal at different temperature and supersaturation in the atmosphere. The satisfactory explanation of the many processes affecting the growth rate of falling crystals is impeded by our limited knowledge of the basic parameters used in our models such as the sticking (deposition) coefficient and the mean migration distance of water molecules at the crystal surface at different temperature. The cardinal question of the physical (perhaps anomalous) behavior of the surface layer and the related water phase transformation and step growth remains still unanswered.

In this situation the study of physical and morphological properties of columnar type crystals seems to be a unique tool for establishing a more sophisticated theory of crystal growth in the atmosphere and for explaining the results of laboratory experiments. More attention will be certainly paid to the growth of prismatic face and to the basic processes dominating its interaction with basal face for stationary and free falling columns.

These studies will be of utmost importance for the investigation of ice crystal evolution in cirrus clouds and ice fogs. Other practical applications are related to the study of the hollow spaces in column type crystals for "indirect aerology" and for the explanation of the ice crystal aggregation in the presence of freezing water droplets.

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