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Snow Crystals and Aerosols*

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In our former experiments the shape of snow crystals is found to be determined by the temperature T_a and supersaturation s . Consequently each of the various types of snow crystals corresponds to a domain in T_a - s diagram, but this diagram was not final. In T_a - s diagram all crystals belonging to needle and cup types and most of the dendritic crystals are formed above water saturation. This condition must be in cloud condition, and it was verified by experiment that the air is full of minute droplets about 1μ in diameter. When these minute droplets are brought in contact with the surface of snow crystal, they do not freeze in a droplet form, but spread over the ice surface. The crystal thus produced shows the same appearance as formed by sublimation.

The electron microscope study of snow crystal nuclei has been carried out by KUMAI. He found, besides the center nucleus which exists at the center of crystal in most cases, numerous aerosol particles at any portion of various snow crystals. The concentration of these aerosol particles was measured. If we assume that each of these aerosols is a condensation nucleus of the droplet of the order of 1μ in diameter, most portion of water substance in the snow crystal is explained as supplied by attaching of these minute droplets to the crystal. Recently M. I. T. people found by using an infrared transmissometer that such minute droplets are abundantly observed in the atmosphere at the top of Mt. Washington, N. H.

From these considerations it was expected that the amount of aerosols might be the third controlling factor for the shape of snow crystal. In order to check this point, the artificial snow experiment was carried out by using the aerosol free air. The aerosols were removed by filtering with cotton filter and then passing the air through a thermal precipitator. Two expansion chambers were set in the inlet and the outlet of the air to and from the snow-making apparatus. When no or very few fog particles are observed with the expansion ratio 1.23, the air is considered "aerosol free" in this experiment. Results of the experiments showed that the shape of snow crystal is quite different from the case when the ordinary air is used, other conditions having been kept the same. For example, a needle or an elongated sheath was obtained in the formerly dendritic condition, a solid column was produced in the plate condition, etc. It was confirmed that the presence of aerosols affects the crystal shape remarkably, and any theory on the formation of various shapes of snow crystals must consider this point seriously.

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§ 1. Introduction.

The external conditions controlling the shape of snow crystals was studied in the Low Temperature Laboratory of Hokkaido University during 1938 through 1945. Various types of snow crystals were made artificially by changing the temperature and the supersaturation. It was found that the shape of snow crystals is determined by the temperature where the crystal is made, T_a , and the supersaturation s . In other words, each point in the T_a - s diagram corresponds to a certain shape of crystal. Classifying the type of crystals into dendritic, plate, columnar, needle-like etc., each type occupies a certain domain in the T_a - s diagram. These results together with over one thousand microphotographs of natural snow crystals were published in one volume "Snow Crystals, natural and artificial" from Harvard University Press in 1954¹⁾.

The T_a - s diagram reproduced in that book is not final, but must be considered as showing the results obtained up to 1945. Some of the domains are not clearly distinguished. For example, several marks representing plate crystals are scattered in the domain of dendritic type, and the ordinary plate and the thick plate cannot be separated as different domains, and occasionally a column is produced under the supposed condition of plate formation. This suggests that T_a - s diagram is a first approximation and there may be another unknown factor, besides the temperature and supersaturation, for controlling the shape of crystal.

After "Snow Crystals" was published, many experiments were carried out in the laboratory of Hokkaido University and we came to the conclusion that the aerosol particles in the atmosphere will be the third factor which controls the shape of snow crystal, specially the dendritic form is closely connected to the presence of aerosols in the air. Recently we carried out the artificial snow experiment by using the air from which almost all aerosol particles are removed. The result showed that the shape of crystals is quite different from the case when the ordinary air is used. For example, needle-like crystals are obtained at the temperature of dendritic condition. Although this paper deals with the results of the preliminary experiments, it is confirmed that the amount of aerosol particles in the air is the third factor for controlling the

shape of snow crystals.

§ 2. T_a - s Diagram and the Supersaturation.

The results of experiments obtained up to 1945 are summarized in T_a - s diagram²⁾ reproduced in Fig. 1. The shape of crystals is grouped into eight kinds, and each of them is represented with different symbols. W is a line giving the saturated vapor pressure with respect to supercooled water.

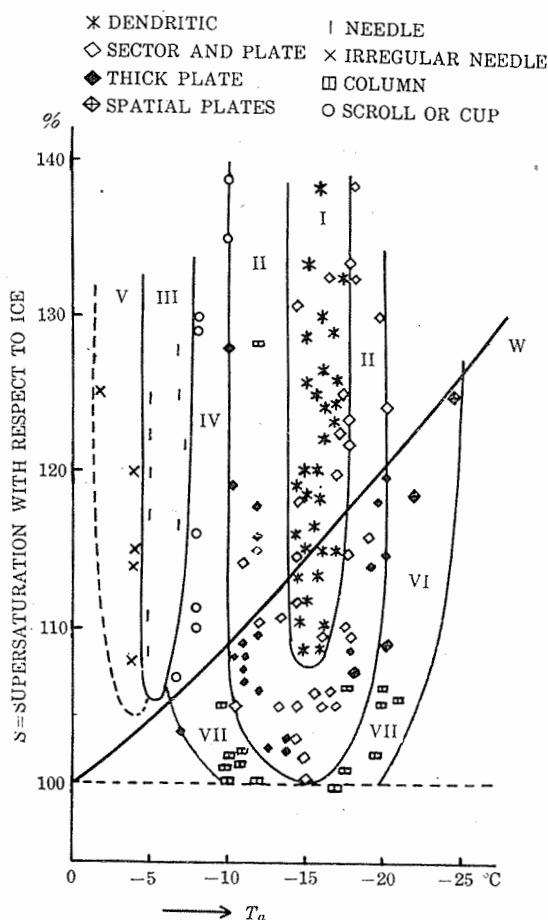


Fig. 1. T_a - s diagram, showing the conditions of formation of various shapes of snow crystals.

One point to be noticed in this diagram is that all crystals belonging to irregular needle, needle and cup types are produced in the region above water saturation and most of the dendritic crystals are also formed above water saturation. In the region above W we must expect the presence of minute droplets suspended in the air saturated with water vapor. The effect of curvature of the droplet is sometimes over-estimated, but $(p-p_0)/p_0$ is not so large for the ordinary *small* droplets, being only 0.011% for $r=10\mu$, 0.11% for $r=1\mu$ and 0.215% even for the very small fog particle of $r=0.5\mu^{3)}$. In this discussion, therefore, we put aside the effect of curvature for the moment.

The term *supersaturation* is always used in the text-book of meteorology, but the definition is sometimes ambiguous. The supersaturation above water saturation must mean the excess of water vapor above the saturated value in its literal sense. It is, however, very unconceivable that more water vapor than the water saturation exists in a purely gaseous state in the natural atmosphere. As pointed out by BENNETT⁴⁾ more than twenty years ago, "the evidence is merely negative as to whether supersaturation does or does not exist, and positive evidence is urgently required." This question seems to be still left unanswered.

In our snow making apparatus, it was found that a large number of minute droplets of diameter about 1μ or less is contained in the upward current of air. We assume that above water saturation the supersaturation means the amount of water in the droplet state plus the saturated vapor with respect to water at that temperature. In this meaning the degree of supersaturation is the total water content in unit volume of the atmosphere. s is measured by taking a known volume of air from the neighborhood of the spot of crystal formation, and weighing the amount of water absorbed by P_2O_5 .

Thus,

$$s = \frac{w + \rho'}{\rho_0} ,$$

where w is the liquid water content per unit volume, ρ' is the vapor density at saturation over supercooled water, and ρ_0 is the vapor density over ice. This technique was developed by HANAJIMA⁵⁾.

The region above W in Fig. 1 is considered to be in the condition of cloud formation. The liquid-water content is calculated from the equation,

$$w = s\rho_0 - \rho'$$

and the curves of equal water content are shown in Fig. 2.

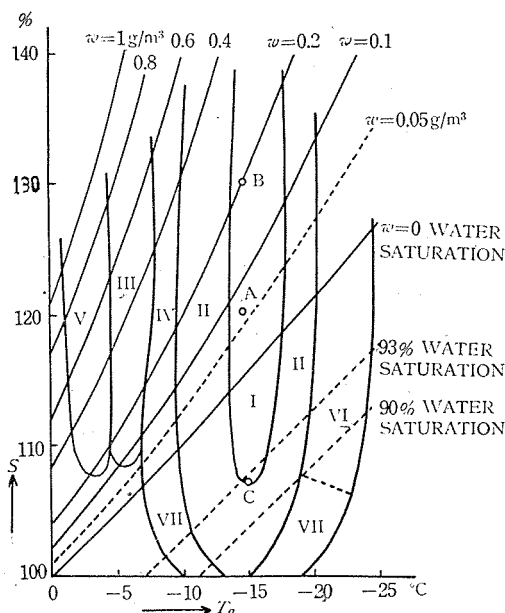


Fig. 2. T_a - s diagram, showing the equal liquid content of water droplets⁶⁾.

At the point A in the figure, that is, in the most favorable condition for dendritic development, the water content is 0.06 g/m^3 . As the probable diameter of minute droplets is about 1μ , the number of droplets per cubic centimeter n is calculated from the equation

$$w = \frac{4\pi}{3} n \rho r^3 = 0.06,$$

$$n = 1.2 \times 10^5 / \text{cm}^3.$$

This value is of the same order of magnitude as the concentration of aerosols in city air. At the point B in Fig. 2, the water content

is about 0.2 g/m^3 , and the mean diameter of the droplets comes out as 1.5μ , assuming the same value of n .

From the considerations above mentioned it is probable that the presence of water droplets contributes to the formation of snow crystals and consequently aerosol particles in the atmosphere may play some role in the growth of snow crystals, because the formation of water droplets is effected by the presence of aerosols.

There is one problem left untouched in this discussion, that is the problem whether fog particles exist or not in the region below the line $w=0$. If the phenomenon takes place in a nearly equilibrium state, no such droplet can exist below water saturation. It is, however, well known that in the growing cloud or fog many results of measuring the humidity often show the values less than 100 percent. It is not rare that the humidity in a fog comes down to 90 percent or less. The lower limit of humidity in dendritic condition is 93 percent, as seen in Fig. 2, point C. If the humidity is 90 percent in this case, the difference of 3 percent will mean that 1×10^5 droplets of 1μ in diameter must exist in 1 cm^3 of the air. In our artificial snow experiment, it is not confirmed yet whether minute droplets exist in the condition below $w=0$, but we cannot go into a hasty conclusion that such droplets never exist below $w=0$.

§ 3. Electron-microscope Study of Snow Crystal Nuclei.

Since 1948 Mr. M. KUMAI of our Laboratory has engaged in the electron-microscope study of snow crystal nuclei, and a part of the results was published in 1951⁷⁾. He found that a relatively large solid nucleus was found almost always at the center of a snow crystal. This "center nucleus" was fairly large and was of the order of one micron or a few microns. Besides this center nucleus, he found numerous "condensation nuclei" of much smaller size, the mean diameter having been measured as about 0.1μ . An electron photomicrogram of any portion of various snow crystals was found to be full of these condensation nuclei. This kind of nucleus is called in this paper the aerosol. The mean concentration of these condensation nuclei per unit area of crystal is roughly 100 per square micron for thicker crystals, and 10 per

square micron for thinner ones. If we assume that each of these condensation nuclei was the nucleus of the water droplet 1μ in diameter, most part of water substance in the snow crystal can be explained as supplied by the effusion of these droplets.

Usually a snow crystal is made of transparent ice, and this crystal of ice is considered as the result of condensation of water vapor by sublimation; that is, the direct condensation of water vapor on ice without passing the liquid state. It is, however, often observed that many crystals of snow are attached with numerous frozen droplets. They are called the rimed crystals. When supercooled fog particles suspended in air are brought in contact with a snow crystal, the droplets are got frozen in a very short time and gives rise to this rimed crystal. The diameter of these frozen droplets was measured with respect to natural rimed crystals, and it was found that most of them are between 20μ and 40μ , the most probable size being 30μ . During the course of our former artificial snow experiment it was found that the larger droplets of 20μ or 30μ in diameter are frozen to the snow crystal in a droplet form but the minute droplets of a few microns in diameter do not freeze in a droplet form but spread over the ice surface when they are brought in contact with ice. The minute droplets, therefore, do not give rise to a rimed crystal, but a transparent ice crystal which is the same in appearance as the crystal made by the sublimation. In other words, these minute droplets behave, as a result, like water vapor in the process of condensation.

This phenomenon was first observed by YOSIDA⁸⁾ with respect to hoar crystals developed on glass surfaces. In most cases, the glass surface is covered with numerous supercooled droplets in the initial state. When the hoar crystal develops through the field of scattered droplets and a tip of the crystal approaches them, most of the droplets vanish or diminish their size markedly by evaporation caused by the vapor-pressure difference of ice and water. Often, however, some remaining minute droplets are caught by the streamer of hoar crystals. The droplet in this case does not freeze to the crystal in the form of a droplet, but appears to spread on the crystal surface. This process is beautifully demonstrated in the microphotographs reproduced in his paper.

This phenomenon may be explained by the sudden evaporation

of a droplet, as pointed out by aufm KAMPE, WEICKMANN and KEDESZ⁹⁾. They suggest that a droplet of diameter $1\ \mu$ can evaporate at -10°C within 0.7 sec at water saturation. This is one way of explaining this phenomenon, but there is another interpretation. That is the supposed surface diffusion of H_2O molecules on the surface of ice crystals. The surface nature of ice crystals has been an old problem since the days of FARADAY and TYNDALL, and the existence of a "liquid" water film on the ice surface was suggested. Recently a simple experiment showing the existence of a liquid-like film on the surface of ice was carried out in our Laboratory¹⁰⁾. Two small ice balls were suspended by very thin filaments. One of the filaments could be moved with a screw motion so that the top of the filament was displaced horizontally. Two balls are separated at a certain inclination of the filament, giving the normal adhesive force. Sometimes, however, it was observed that the ice spheres showed a rotation before the separation. The rotation took place more frequently in the range of temperature near the freezing point, but it was once observed at -7°C . This rotation phenomenon is explained by assuming the existence of a liquid-like film on the surface of ice.

Assuming this liquid-like film on the ice surface, the spreading of minute water droplets on the surface can be explained without difficulty. Due to the viscosity of the supercooled water droplet and the "viscosity" of the "liquid" film on the ice surface, it will take some time for the spreading of the droplet on the surface. If the droplet is large, $20\ \mu$ or $30\ \mu$, this time will be fairly long and the droplet will freeze before it is completely spread over the ice surface. In this case the rimed crystal is formed.

The condensation of minute droplets to snow crystal can thus produce the ordinary snow crystal which has hitherto been considered as the product of condensation of water vapor by sublimation. Next problem is that whether an ample amount of such minute droplets do exist or not in the cloud of snow conditions. The former results were negative, but it is considered that this negative result might be chiefly due to the difficulty in catching these minute droplets, because most of the observations were done by collecting cloud particles in an oil film. Recently an infrared transmissometer was designed in M.I.T. and JOHNSON, ELDRIDGE

and TERRELL¹¹⁾ report the results of the observations at the top of Mt. Washington N. H. The method consists of the observation of changes in intensity with wave length when infrared radiation is transmitted through a cloud. Size distribution and liquid water content were measured for 42 clouds, and majority of cloud droplets were found to be of two microns diameter or less. The number of droplets $1\ \mu$ diameter was more than $20,000/\text{cm}^3$ in 20 cases, more than $30,000/\text{cm}^3$ in 14 cases, the maximum having been $68,323/\text{cm}^3$. We can expect, therefore, that such minute droplets may exist abundantly in the cloud of snow conditions.

§ 4. Preliminary Experiment on the Effect of Removal of Aerosols on the Shape of Snow Crystals.

From the considerations above described, the author is led to consider that the aerosol particles may play some role in controlling the shape of snow crystals. The characteristics in the pattern of snow crystals, for example, the dendritic branching and the fine striae in the sector and etc. are probably due to the fluctuation in conditions, but at the same time it is expected that these characteristics are closely connected with some crystal defects produced in the course of crystallization. If the crystal is made up chiefly by attaching of minute droplets to the crystal, the aerosol particles which are the nuclei of those droplets will give rise to the crystal defects and act as a controlling factor for determining the crystal shape.

In order to check this point, the artificial snow experiment was started by using aerosol free air. The experiment was carried out in the Low Temperature Laboratory of the Transportation Technical Research Institute at Mitaka, Tokyo, which belongs to Ministry of Transportation. The experiments were done chiefly by Dr. HANAJIMA with the collaboration of Dr. OGUCHI, Mr. KUMAI and Mr. MUGURUMA. The details will be published later, but the result of the preliminary experiment is reported in this paper.

a) Removal of aerosols from atmospheric air.

Aerosols were removed by filtering the air with cotton filters 2 meters in total length, and then passing a thermal precipitater. The function of the thermal precipitater was examined by using

an expansion chamber.

Very few fog particles were observed with the expansion ratios up to 1.23. When the ratio is increased to 1.24, several numbers of particles made the appearance. It seems to exist a discontinuity in the action of the expansion ratio, and the fog particles suddenly increases in number when the ratio exceeds 1.25. Above 1.25, very tiny fog particles appear mixed with those of the ordinary size. After the expansion above 1.25 is once performed, many fine fog particles appear with the smaller ratios than 1.23, if the same air is used. This after effect will be due to the formation of new nuclei produced by the large rate of expansion. This point is of great interest for the study of the nature of various kinds of aerosols.

b) *Validity of the former diagram of condition of formation of snow crystals.*

The former diagram of condition of formation of snow crystals shows that the crystal shape is determined by the temperature of air where the crystal is made, T_a , and the temperature of the water in the reservoir below, T_w . The latter is a measure of supersaturation. The former is a more important factor than the latter. The old T_a - T_w diagram is reproduced in Fig. 3 for the convenience sake. T_a - T_w diagram is of a transit nature, and it must be converted into T_a - s diagram, by measuring s as the function of T_w and T_a . In the present state of experiment s is not yet measured. Therefore, all discussions in this section will be referred to T_a - T_w diagram. No essential difference exists between T_a - T_w diagram and T_a - s diagram for the qualitative discussion of the crystal shape.

Snow making experiments with ordinary air were carried out by using the new apparatus which was designed to study the effect of removing aerosols on the shape of snow crystals. The result was the same as the former case, and the T_a - T_w diagram obtained in our former experiments was verified to be applicable in this series of experiments.

According to the former diagram, dendritic crystals are obtained with T_a between -14°C and -17°C and relatively high values of T_w . One example is shown in Photo. 1, Pl. I, which is

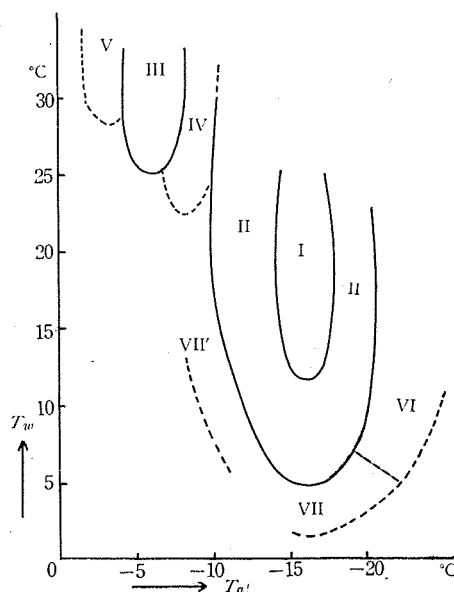


Fig. 3. T_a - T_w diagram, showing the condition of formation of various types of snow crystals.

I dendritic, II plate & sector, III needle, IV scroll or cup, V irregular needle, VI spatial assemblage of plates, VII column, VII' column.

made with $T_a = -15^\circ\text{C}$ and $T_w = +19^\circ\text{C}$. In the former experiments needle crystals were found to be produced with T_a between -4°C and -7°C and the higher values of T_w . This point is also verified with this new apparatus. Photo. 2, Pl. I is a crystal of dendritic type with many needles grown perpendicularly to the basal plane. First the dendritic crystal was made and then T_a and T_w were changed to -6.5°C and $+31^\circ\text{C}$ respectively, which was the favorable condition for needle growth. This is a good example showing that needle crystals are formed with T_a about -6°C . All these results are concordant with the results of our former artificial snow experiments.

c) *Results obtained with "half clean" air.*

Similar experiments were repeated by using "half clean" air, which means most of the dust and aerosol particles are removed from the atmospheric air. By using the cotton filter only or

the thermal precipitater being not in its full efficiency, most of aerosols are removed but still a considerable amount is detected by the expansion chamber. This state of air is called "half clean" in this report for the descriptive purpose. It was found by experiment that the shape of snow crystal is, under the same condition of T_a and T_w , quite different from that produced in the ordinary air. Photo. 3, Pl. I is made with $T_a -14^\circ\text{C}$ and $T_w +20^\circ\text{C}$ by using "half clean" air. The thermal condition is almost the same as the case of Photo. 1, Pl. I, but the crystal developed into a plate type instead of a dendritic one. Comparing Photo. 1, Pl. I and Photo. 3, Pl. I, it is remarkable that the shape of snow crystal is influenced by the presence of aerosol particles. Sometimes crystals of apparently dendritic form are produced in the half clean air, as shown in Photo. 4, Pl. I. This crystal, however, is different in the inner structure from the proper dendritic snow crystal. The dendritic appearance is the result of assemblage of small plates.

d) *Results obtained with "aerosol free" air.*

The "aerosol free" air means the state of air in which no or very few number of fog particles are observed with the expansion ratios up to 1.23. The experiments carried out by using this "aerosol free" air gave a more remarkable result. Two expansion chambers were provided in the inlet and outlet of the snow making apparatus respectively, so that the air is checked before being sent into the apparatus and also after it came out from the apparatus. The snow making experiment was carried out by streaming the aerosol free air slowly through the apparatus. Artificial snow experiment was started after it had been confirmed that no or very few fog particles were produced both in the expansion chamber at the inlet and in that at the outlet.

In this case the crystal always developed into a sheath type or a needle, inspite of the condition of dendritic formation. The sheath type crystal is slender in thickness and elongated in length, giving the form like a needle. One example of the sheath-like crystal is shown in Photo. 5, Pl. II, and the needle in Photo. 6, Pl. II. T_a and T_w for the former are -14°C and $+19.5^\circ\text{C}$ respectively, and -16°C and $+16^\circ\text{C}$ for the latter, both cases being in the con-

dition of dendritic development, if the ordinary air is used. From the results described above, it may be concluded that the dendritic development of snow crystal is much affected by the presence of aerosol particles in the air.

e) Another experiments by using "aerosol free" air.

In the former experiments using ordinary atmospheric air, the dendritic development transformed into a plate type when the supersaturation became less than a certain value, the temperature being kept in the condition favorable for dendritic formation. In the corresponding case, a solid column is produced, when aerosol free air is used. Photo. 7, Pl. II is an example of this solid hexagonal column, which is made with $T_a -15^{\circ}\text{C}$ and $T_w +8^{\circ}\text{C}$, which is the condition for plate formation in the case of ordinary air.

With the supersaturation suitable for dendritic development, the crystal grows into a plate type in the ordinary air, when the temperature deviates from the range between -14°C and -17°C , which is the condition for dendritic formation. In the corresponding case of aerosol free air, the crystal develops into a sheath in the form of a thick column, Photo. 8, Pl. II. This crystal is made with $T_a -11^{\circ}\text{C}$ and $T_w +16^{\circ}\text{C}$:

f) Summary of the preliminary experiment.

All the results above described show that snow crystals have a tendency to develop in the direction of the principal axis, when aerosol particles are removed from the air. At least it is made clear that the shape of snow crystal is determined not only by the temperature and supersaturation but the presence of aerosols plays an important role in determining the shape. Any theory about the formation of various shapes of snow crystals must take this fact into considerations. Details of the experiments and the whole results obtained will be published as a separate paper in a near future.

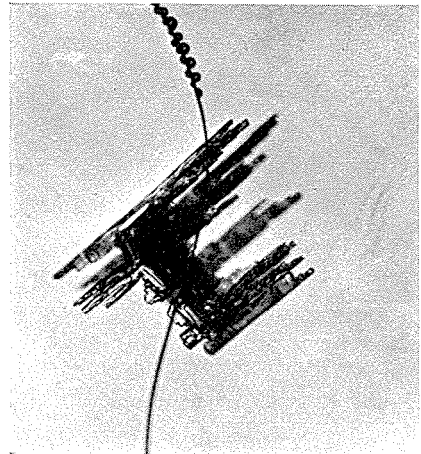
In conclusion the author expresses his sincere sense of gratitude to the authorities of Transportation Technical Research Institute for the use of the Low Temperature Laboratory.

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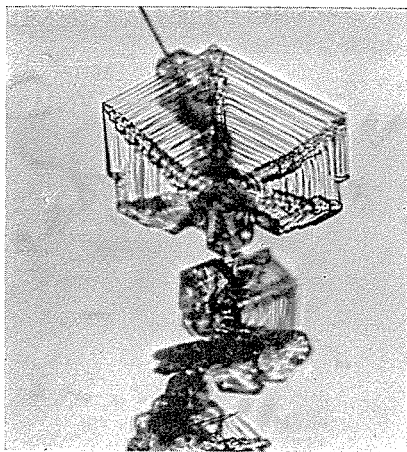
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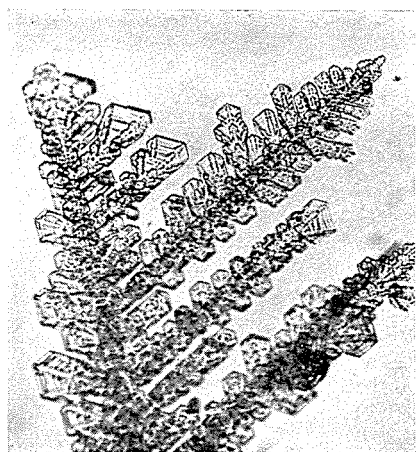
1. Snow crystals made in ordinary air. Dendritic crystal; No. 34, $T_a -15^\circ\text{C}$, $T_w +19^\circ\text{C}$. $\times 15$.



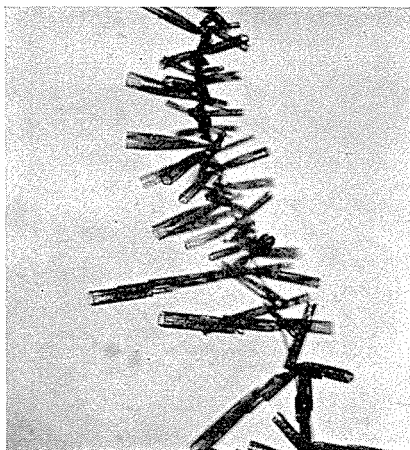
2. Snow crystals made in ordinary air. Dendritic crystal with many needles attached; No. 26, $T_a -6.5^\circ\text{C}$, $T_w +31^\circ\text{C}$. $\times 25$.



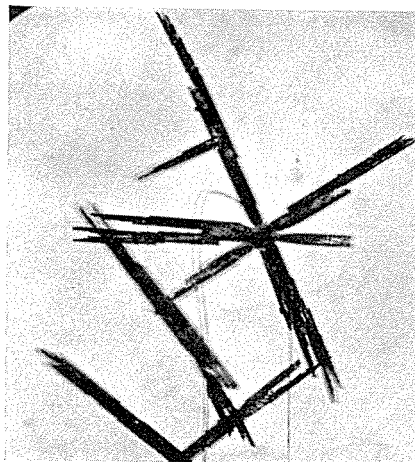
3. Snow crystals made in "half clean" air. Plate crystal produced in the dendritic condition; No. 49, $T_a -14^\circ\text{C}$, $T_w +20^\circ\text{C}$. $\times 43$.



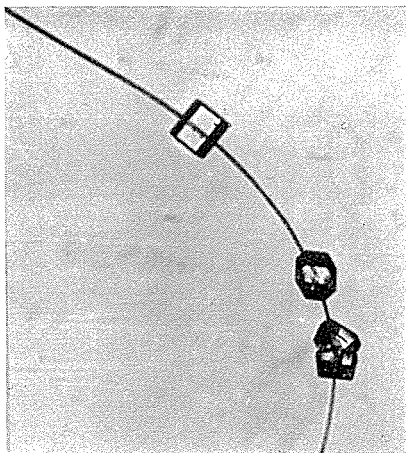
4. Snow crystals made in "half clean" air. Pseudo-dendritic crystal made of an assemblage of small slates; No. 8, $T_a -16.5^\circ\text{C}$, $T_w +16.5^\circ\text{C}$. $\times 43$.



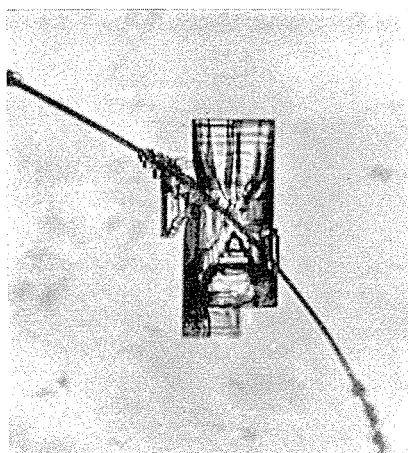
5. Snow crystals made in "aerosol free" air. Sheath type crystal produced in the dendritic condition; No. 36, $T_a -14^{\circ}\text{C}$, $T_w +19.5^{\circ}\text{C}$. $\times 15$.



6. Snow crystals made in "aerosol free" air. Needle crystal made in the dendritic condition; No. 35, $T_a -16^{\circ}\text{C}$, $T_w +16^{\circ}\text{C}$. $\times 15$.



7. A solid column made in aerosol free air under the plate condition; No. 42, $T_a -15^{\circ}\text{C}$, $T_w +8^{\circ}\text{C}$. $\times 43$.



8. Sheath-like crystal made in aerosol free air under the plate condition; No. 39, $T_a -11^{\circ}\text{C}$, $T_w +16^{\circ}\text{C}$. $\times 43$.