SNOW CRYSTALS AND THE IDENTIFICATION OF THE NUCLEI IN THE NORTHERN UNITED STATES OF AMERICA

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ABSTRACT

Snow crystals are born at high altitudes and grow into various forms while falling through the atmosphere. In this research work, the relations observed among snow crystals were crystal form, nucleus, and temperature and humidity of the mother cloud based on radiosonde sounding data. Almost all crystal forms which are shown in "Snow crystals" by Nakaya (1954) were observed in a winter season at Houghton (Keweenaw Field Station), Michigan which is situated on a small peninsula along the southern shore of Lake Superior. The factors that influence snow-crystal form are mainly the air temperature and the humidity at which the crystal grows. Previously there have been no data for the formation of pyramid-shaped snow crystals in natural conditions. However, in the observations of Houghton, it was found that the pyramid shaped crystals were formed in the clouds at temperatures between $-6C$ and $-10C$. This coincides with the condition of growth of artificial snow crystals of the cup and scroll type. Needle crystals are made in the temperatures between $-4C$ and $-6.5C$; pyramids, bullets, and columns, between $-6C$ and $-10C$; hexagonal plates between $-8C$ and $-12.6C$; and dendritic forms between $-14C$ and $-16C$. These observations agree well with the Nakaya-Hanajima diagram obtained from measurements made in convection snow-making apparatus and with the Mason-Hallet diagram obtained with a diffusion chamber.

Three hundred snow crystals were collected; successful electron micrographs were obtained of the center nucleus of 271 of these. The nucleus of snow crystals can be classified as clay-mineral particles, hygroscopic particles, combustion products, microorganism and unknown (unidentified) materials. Clay-mineral nuclei accounted for 87 per cent, hygroscopic nuclei 1 per cent, combustion products 2 per cent, unknown material 9 per cent, and no nuclei 1 per cent of the sample.

A relation was found between the sizes of the snow-crystal nuclei and the snow-crystal forms. The size of the maximum frequency of needle-crystal nuclei is $3.5\ \mu$, and that of the hexagonal-crystal nuclei is $1\ \mu$. In other words, the sizes of maximum frequency of the nuclei of snow crystals which are formed at warmer temperatures are larger than those at colder temperatures. In this investigation, no relationship between crystal forms and the substances of the nuclei was found.

1. Introduction

The nucleation of a snow crystal is one of the main problems of cloud physics. In the field of physical meteorology, the electron microscope is used for the studies of nuclei of snow crystals, fog, and cloud droplets, of the surface nature and the growth of snow crystals, and of atmospheric aerosols. Snow crystals are forms of solid precipitation which are observed on the earth's surface. The initial stage of a snow crystal can be seen in the cirrus cloud. This small crystal is called an ice crystal. In the process of formation of a snow crystal in the free atmosphere, the ice crystal is first formed. The factors, determined by Nakaya and Hanajima (1951, 1958) that influence snow-crystal form, are mainly the air temperature and humidity at which the crystal grows.

Volmer and Weber (1926) have developed the theory of homogeneous nucleation in which the new phase appears without any foreign substance. Nucleation of ice crystals occurs more commonly on foreign nuclei in the atmosphere. The nuclei of ice and snow crystals have been studied for many years. The presence of sublimation nuclei was suggested by analogy with condensation nuclei by Wegener (1911). It was speculated that natural sublimation nuclei were substances having a crystalline structure similar to that of ice. In this line, Vonnegut (1947) found that silver-iodide crystals were effective as ice nuclei at temperatures lower than $-5C$. Kumai (1951) and Isono (1959) studied the nuclei of snow crystals by the use of an electron microscope and identified the nucleus substances by the electron diffraction method. Many of them were found to be minute crystalline substances insoluble in water, a kind of clay mineral on which ice may form by oriented overgrowth. In general, the formation of an ice crystal may be (1) by the spontaneous transformation of a supercooled water droplet below the threshold temperature for nucleation, (2) by the direct condensation of water vapor on a solid nucleus, or (3) as the result of natural and artificial seeding. After nucleation, ice crystals grow in an atmosphere of water vapor supersaturated with respect to ice. The snow crystals and the identifications of nuclei described in this paper resulted from collections made at Houghton, Michigan.

2. Observations of snow-crystal forms and the temperatures of the mother cloud

During the past thirty years, the dependence of crystal shape on temperature was studied by many physicists and meteorologists. Experimental work by Nakaya and Hanajima (1951; 1954) have shown the relations between the shape, the temperature and the total water content with the use of a convection cold chamber. Aufm Kampe *et al.*, (1951) showed the dependency of crystal shape on temperature in a cold-box experiment. Gold and Power (1954) observed snow-crystal forms on the ground and then compared the forms with the meteorological conditions as measured by a radiosonde. Magono and colleagues (1959) studied the forms of snow crystals and their growth rates at five stations on a mountain slope. These results agree fairly well with Nakaya's diagram.

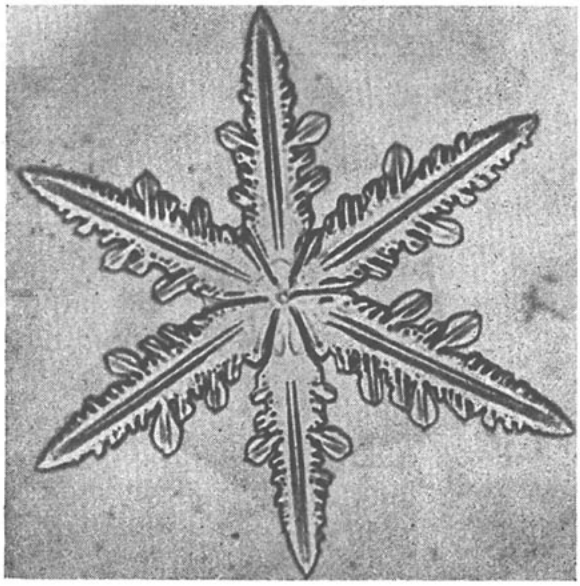

FIG. 1. Dendritic crystal (×60).

Hallet and Mason (1958) showed the relations between crystal forms and conditions of temperature and humidity with the aid of a diffusion cold chamber. In their diagram, plates and dendrites should form at temperatures between 0C and −2C. Nakaya, Hanajima, and Muguruma (1958) also formed plates and dendrites in the same temperature region by the use of dust-free air in a convection cold chamber. Therefore, the temperature dependency of snow-crystal forms is rather well agreed upon among the experiments of Nakaya, Mason and Kobayashi (1958).

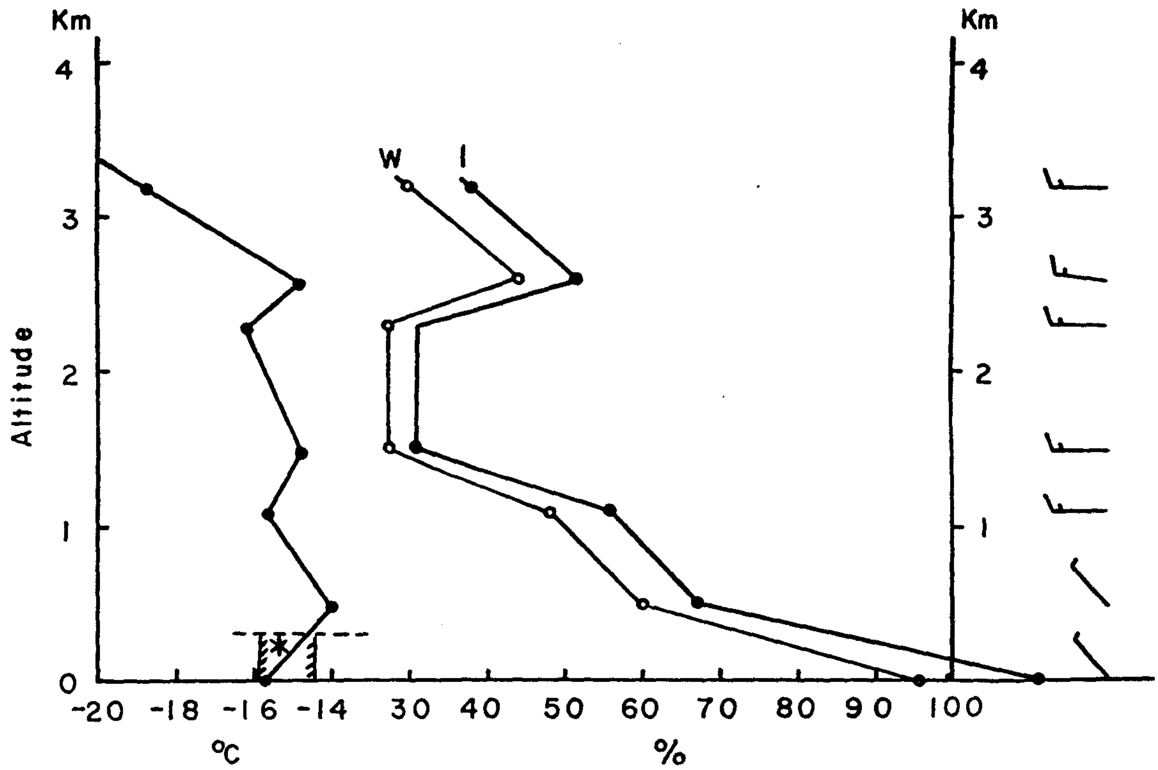

FIG. 2. Temperature and humidity conditions observed during collection of dendritic snow crystals. Humidity (w: water saturation; I: ice saturation), 0938 EST 26 January 1959. Estimated cloud top indicated by dashed line; wind speed full barb equal to 10 m per sec.

However, the dependency for crystal forms of the humidity between water saturation and ice saturation is still unresolved.

In the present paper, the observations of snow-crystal forms and the temperature of the mother cloud were made during four weeks in midwinter at the Keweenaw Field Station in Houghton, Michigan. There were seventeen days of snowfall in this period. Almost all forms of snow crystals were collected, and 685 micrographs and 30 sheets of snow-crystal replicas were collected. From these, a relationship between radiosonde sounding data and the falling crystal forms was obtained.

Previously, no data had been reported for the formation of pyramid-shaped snow crystals. However, in this observation, it was found that pyramid crystals formed in the cloud at the temperature between −6C and −10C. Artificial snow crystals of cup and scroll shape form at temperatures between −6C and −10C. Meteorological conditions of the formation of dendritic, hexagonal, needle, pyramid and bullet crystals agree with the diagrams of Nakaya-Hanajima and Hallet-Mason.

The variety of snow-crystal forms in Upper Michigan paralleled that of the observation in Hokkaido by Nakaya (1954). In a month's observation at Houghton, Michigan, almost all crystal forms were observed. They were needle (simple and combination), column (simple and combination), capped column, pyramid, bullet, hexagonal, dendritic crystals (three-, four-, six-, and twelve-branch type, malformed and spatial type), irregular particles and graupel.

On any given day, several forms of snow crsytals might be observed. However, usually one or two forms of snow crystals would predominate during the one hour or more that radiosonde soundings were conducted. From such data it is possible to get the relationship between the falling crystal forms and the meteorological conditions of the mother clouds as revealed by the radiosonde sounding data. Observations of the mother clouds were convenient because the cloud bases were low—that is, less than 500 m. However, snow crystals observed on the ground at the radiosonde station were formed in the windward cloud. The temperature of the mother cloud of the crystals was measured leeward of the station. It is possible to establish a relationship between snow-crystal form and the temperature of the mother cloud, provided the falling snow is of one form and the same meteorological condition continues 30 minutes or more.

A. *Dendritic crystals.* On 26 January 1959, as shown in fig. 1, a heavy snowfall of dendritic crystals occurred from early morning until 1000 EST. Since no other shapes of snow crystals fell, it is very easy to determine the correlation between the forms and the temperature. The air temperature was measured by the use of a radiosonde released at 0938 EST 26 January. The data are shown in fig. 2. The air temperature from the ground to the altitude of 2,800 m was between −14C and −16C. It is presumed that the dendritic crystals grew in the lower cloud near the ground while the radiosonde sounding was conducted. Therefore, the dendritic crystals grew in the cloud at temperatures between −14C and −16C. This verifies that the conditions of the natural snow-crystal growth agree with that of artificial dendritic crystals. In general, the humidity in the mother cloud, as measured by the radiosonde, was below water saturation, and it was rather lower than the estimated value for the experiments in the cold chamber growth of snow crystals. From the radiosonde data, the expected cloud height was 300 m; the mean wind speed in the cloud was 5 m per sec. The falling rate of dendritic plane crystal is 30 cm per sec; therefore, it is estimated that the crystal would have taken about 17 min to fall 300 m in height. During this time, they would move 5 km horizontally.

B. *Hexagonal crystals.* On 28 January 1959, hexagonal crystals fell from 0830 to 1130 EST. The altitude of the cloud base was 500 m. The air temperature and the humidity were measured by a radiosonde, released at 1038 EST. From the humidity data, the top of the mother cloud of hexagonal snow crystals is estimated to be 1300 m. The temperature of the mother cloud of the hexagonal snow crystals from 500 m to 1300 m in altitude was between −10C and −12.6C.

C. *Needle crystals.* On 29 January 1959, a heavy shower of needle crystals occurred from 0900 until 1000 EST. The smallest crystal observed was 0.8 mm, and the largest one was 5 mm in length. The altitude of the cloud base was 200 m. The radiosonde was released at 0938 EST. From the radiosonde data, the cloud top was estimated to be at 2,500 m. The air temperatures in the cloud between 200 m and 2,500 m were between −4C and −6C. From the radiosonde data and the observation of the crystal form at the ground, it is concluded that these needle crystals were growing in the cloud at temperatures between −4C and −6C. This agrees with the growing condition of artificial needle crystals.

D. *Pyramid, bullet, and column crystals.* Pyramid crystals are a rare form of snow first reported by Scoresly in 1832. Only two pyramid crystals were taken in micrograph in three winters of observation by Nakaya in Hokkaido. After that, during approximately ten winters, the writer and others continued to take micrographs of snow crystals but did not find them. On 26 February 1959, many pyramid crystals were observed from 0800 to 1000 EST. Fig. 3a and

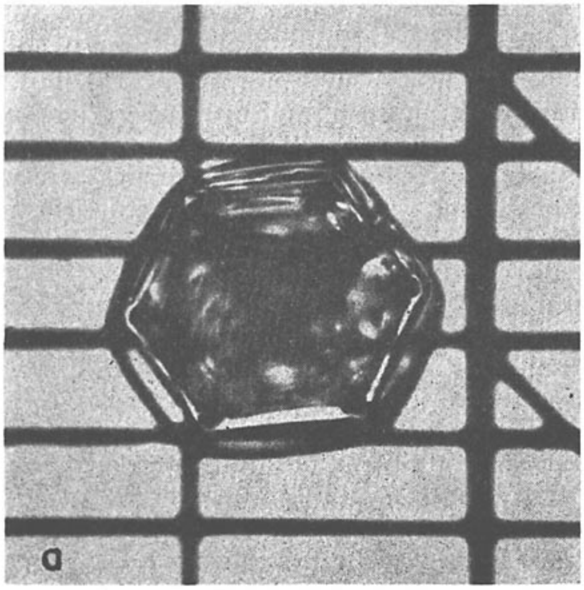

FIG. 3a. Top view of pyramid crystal (×100).

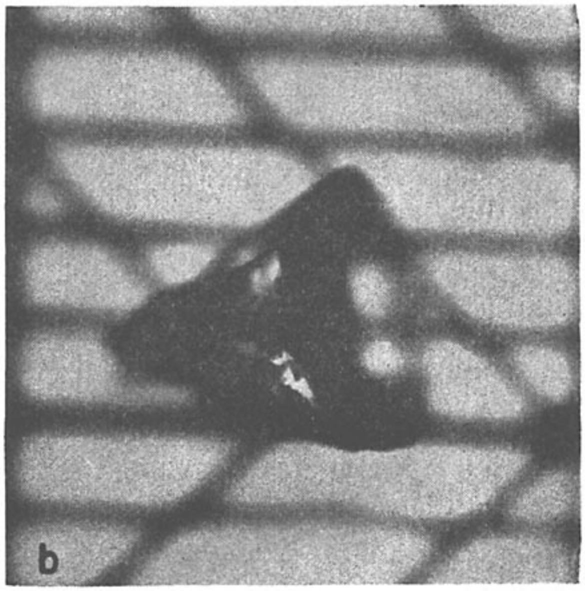

FIG. 3b. Side view of the same crystal (×100).

3b show an example of such a pyramid crystal. Fig. 3a shows the top view; the base plane is hexagonal as seen in this micrograph. Fig. 3b shows the side view of the same crystal; the bottom is an empty hole as seen in this micrograph. The size is rather small, the diameter of the bottom is 0.45 mm, and the height is 0.44 mm. It is thought that these pyramid crystals were born at a low altitude. A radiosonde was released at 0840 EST; the data are shown in fig. 4. A great number of pyramid and bullet crystals were observed, and at the same time hexagonal crystals in small numbers were falling. At that time,

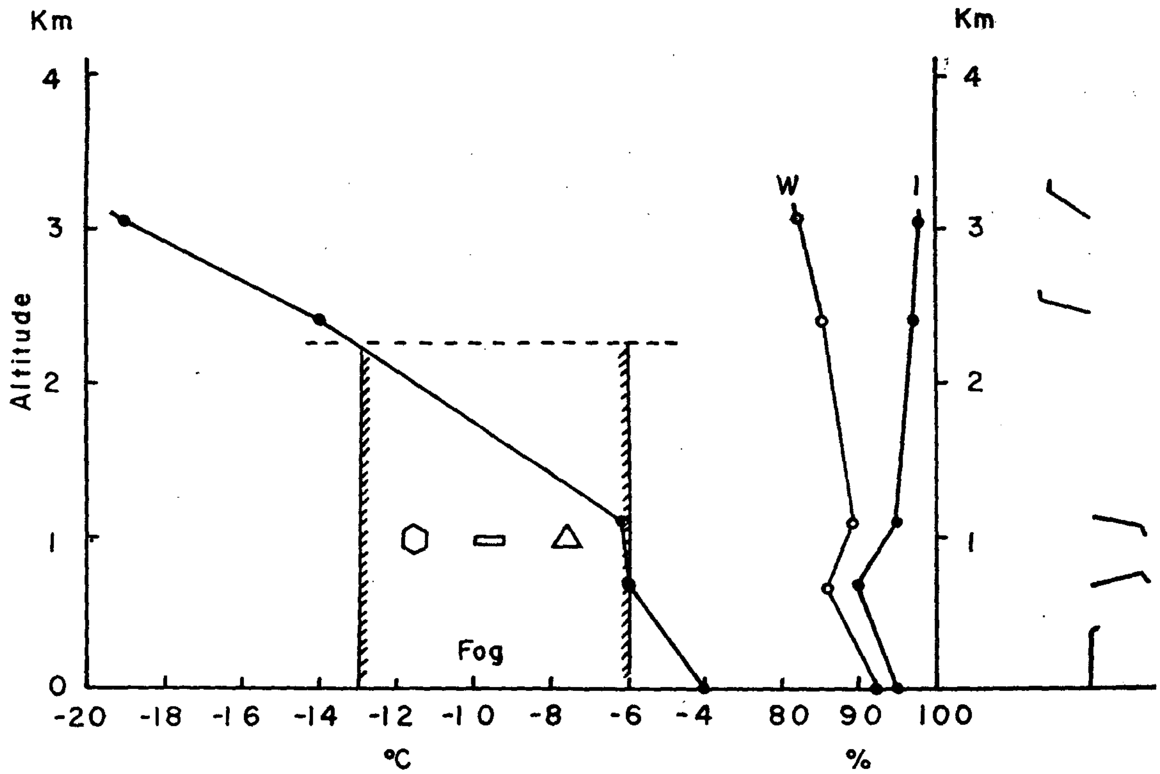

FIG. 4. Temperature and humidity conditions observed during collection of pyramid, bullet and hexagonal crystals, 0840 EST 26 February 1959.

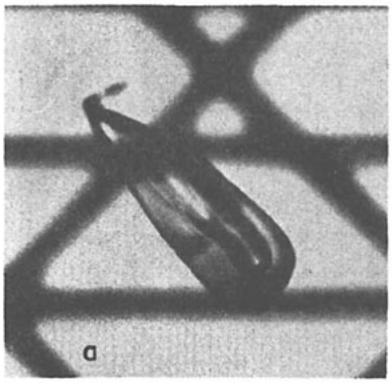

FIG. 5a. Sublimation process of a snow crystal (×150).

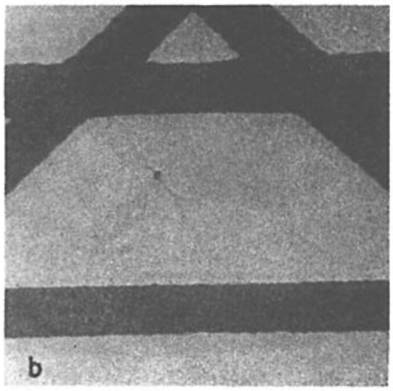

FIG. 5b. Sublimation process of a snow crystal (×150).

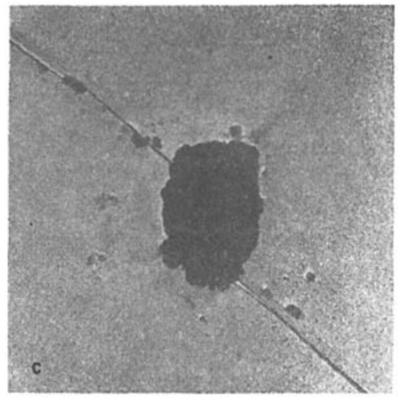

FIG. 5c. Sublimation process of a snow crystal (×150).

the ground was covered with fog. These crystals formed in the cloud at the temperatures between −6C and −13C. However, hexagonal crystals were observed at temperatures between −10C and −12.6C. Therefore, it appears as though the pyramid, bullet, and column crystals were formed in the temperature range −6C to −10C. The temperature region of pyramid (cup, scroll), bullet, column, and hexagonal crystals coincides with that of the diagrams of Nakaya and of Mason.

E. *The correlation between the shape of natural snow crystals and Nakaya's diagram.* An attempt was made to obtain correlations between the shape of the crystals and the temperature of the mother cloud by the use of snow crystal replicas. This is very difficult because, in general, two or three types of crystals fall to the ground at the same time. However, on several occasions at Houghton, Michigan in the winter of 1959, one or two types of snow crystals were observed falling during one or more hours concurrent with a radiosonde release. For these periods, it is very easy to get definite correlations. Table 1 covers all observations for the crystals which formed at temperatures above −17C.

TABLE 1. The comparison of the correlation between growing temperature and forms of natural and artificial snow crystals.

Shape of snow	Observation at Houghton	Artificial snow (Nakaya's diagram)
Dendritic	−14C−−16C	−14C−−17C
Hexagonal	−8C−−12.6C	−8C−−20C
Pyramid, bullet (scroll or cup)	−6C−−10C	−6C−−10C
Column	−6C−−15C	−10C−−20C
Needle	−4C− −6.5C	−5C− −6C

3. Methods of making specimens of snow-crystal nuclei

The difficulty of this experiment lies in making certain that the image obtained in the electron micrographs is that of the nucleus of snow. For this purpose, great care must be taken in locating the center of a snow crystal in the field of the mesh. Also, as there are many aerosols in the free atmosphere, it is desirable to choose a spot where the natural dust is at a minimum.

At Houghton, Michigan the aerosols were at a minimum in midwinter. The specimens of snow-crystal nuclei for the electron microscope were prepared in an igloo—*i.e.*, a snow cavern made for this purpose. The temperature in the igloo was between −5C and −15C, and snow crystals could be handled at will, without any fear of melting. A long and slender sliver of wood resembling a match stick was made and broken into two pieces. The whisks of the broken end were convenient to pick up snow crystals without melting because of the thermal insulation of the stick.

When the central part of a large snow crystal of the hexagonal-plate variety is examined under an ordinary microscope, it usually appears as a tiny ice crystal of either a stellar or hexagonal form about 30 μ in diameter. In the case of stellar crystals, the central part is a circle about 20 μ in diameter. These central patterns are called the central portion of the snow crystal in this paper. Since the diameter of the central portion is between 20 and 30 μ, it can be arranged in a void of the mesh. In the case of snowfalls not accompanied by strong winds, snow crystals fell into the igloo through a ceiling opening. They were collected on a clean glass plate. The snow crystals were then transferred to a mesh covered with collodion film. It is important to observe the mode of sublimation process of snow crystals to see whether the

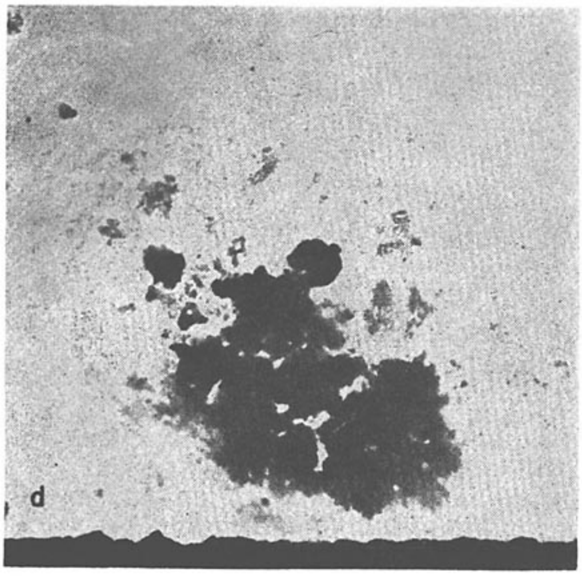

FIG. 5d. The nucleus (illite mineral) of the snow crystal in fig. 5a (×4000).

supposed nucleus can be expected to remain on the collodion film without displacement. Successive stages of sublimation of specimen crystals were taken through a microscope with a magnification of about sixty. A snow crystal begins to evaporate by sublimation from the tips of the crystal; the thicker, central portion of the crystal remains to the last stage without marked displacement, as shown in figs. 5a, 5b, and 5c. Inside the igloo, it took about 10 min for a small crystal to completely sublime.

In preparing the crystals for study, they were mounted on collodion film kept in a desiccator in the igloo. In this condition, the snow sublimed leaving the nucleus on the collodion film. All experience shows that the nucleus remains on the collodion film of the mesh. The region at which the central portion vanished in the last stage of sublimation (which is recorded by an optical photomicrograph) can be observed through an electron microscope, and then the center nucleus pattern can be taken by means of an electron micrograph as shown in fig. 5d.

4. Method of identification of snow-crystal nuclei

It has been found that a relatively large, solid nucleus almost always exists at the central portion of the crystal. Innumerable very small nuclei are observed in the remainder of the crystal (Kumai, 1951; Isono, 1959). The former is called "center nucleus" and the latter is called "condensation nuclei" in this paper. The composition of the center nuclei has been identified through analysis of electron diffraction patterns by comparing their shapes with electron micrographs of known substances (Kumai, 1957; Isono, 1959) and by observing specimen changes due to electron bombardment in the microscope (Burton and Ellis, 1947; Yamamoto and Ohtake, 1953).

The nucleus of natural snow crystal is an aerosol in the atmosphere. Aerosols consist of many kinds of substances from the various sources—*i.e.*, the ground, the ocean, air pollution as a result of human activities, and materials from outer space.[1] They are insoluble crystalline substances on which ice may form by oriented overgrowth.

Analysis of these nuclei showed that most of them were clay-like minerals. Most clay minerals are hydrous aluminum silicates, but they vary considerably both in physical properties and chemical composition. The most common clay minerals belong to the kaolin, montmorillonite and illite group. All are finely crystalline or metacolloidal and are likely to occur in flakelike or dense aggregates of various types as shown in table 2. Electron micrographs of these minerals have been taken by Davis and colleagues (1950). The composition of the clay minerals are often related to their origin. In general, alkalic feldspars and micas tend to alter to kaolin. Ferromagnesium mineral, calcic feldspars and volcanic glasses tend to alter to members of the montmorillonite group. These clay minerals are affected by climatic conditions, such as the length of time during which weathering conditions prevail. The identification of these minerals is possible from the analysis of the electron-diffraction patterns and the morphology of these minerals.

In this study, the specimens of snow-crystal nuclei were shadowed by chromium vapor in a vacuum vessel. The indexing of rings of Debye-Sherrer patterns from the chromium were used as interplanar distances. Eighty per cent of the snow-crystal nuclei from 271 specimens showed electron-diffraction patterns. Electron-diffraction-ring patterns indicated that many of them were aggregated from minute crystalline particles.

It was found necessary to take two or more selected electron-diffraction patterns of the nucleus with different exposure time for each specimen. Some examples of the identification of the snow-crystal nuclei are discussed below.

A. *Illite*. Illite is a widely distributed clay-mineral constituent of argillaceous sediments apparently related to the mica group. Fig. 5a shows a hexagonal snow crystal which nucleated on illite. After sublimation, a solid particle remained at the position where

[1] Interest in the possibility that nuclei may be extra-terrestrial stems from the Bowen hypothesis (1953) that precipitation may be influenced by meteor showers. Schaefer (1955) and Mason and Maybank (1958) have studied this as yet unsolved problem.

TABLE 2. Characteristic shapes of minerals.

Kinds		Characteristic shape
Kaolin group	Kaolinite	Fine-grained, euhedral, occasionally elongated, and six-sided crystals.
	Halloysite	Partial splitting and unrolling of cylindrical forms, or typical elongated striated structures.
	Nacrite	Irregularly shaped and angular flakes.
Montmorillonite group	Montmorillonite	Irregular flake-like particles with no definite crystal outline.
	Nontronite	Lath-like particles with rather well-defined outlines.
Illite group	Illite	Irregular flaky particles.
Related minerals	Pyrophylite	Distorted and curved crystal flakes. The dark wavy lines are frequently observed in small micaceous plates.
	Serpentine	Characteristic short and stubby rods with centered tubes.
	Alpha Sepiolite	Long and short bundle of fibers. A long fiber shows flexible habit. Shorter ones result from breakage.
	Attapulgite	Typical rods of fibers which occasionally occur in bundles.

the crystal center vanished by sublimation as shown in fig. 5d. Many small particles were found around the center nucleus. The particles had been in the branch of the snow crystal. The particle 8 μ in diameter shown in fig. 5d is the nucleus of this snow crystal. The electron-diffraction pattern of this nucleus is shown in fig. 5e. The interplanar distances of the pattern are tabulated in table 3 with that of

TABLE 3. Interplanar distances of illite.

Illite		Snow-crystal nucleus	
d	I/I_1	d_{obs}	Intensity
4.91	0.5	4.96	M
4.39	0.5	4.30	M
3.64	0.2	3.63	W
3.40	0.1	3.38	VW
3.29	1.0	3.26	S
3.20	0.1	3.15	VW
2.93	0.1	2.95	VW
2.83	0.4	2.82	M
2.55	1.0	2.55	S
2.47	0.2	2.45	VW
2.36	0.2	2.36	VW
2.22	0.2	2.25	VW
2.00	0.5	2.01	M
1.65	0.4	1.64	W
1.50	0.8	1.51	W
1.35	0.2	1.38	VW
1.25	0.2	1.24	VW

d: d spacing of an illite mineral.
I/I_1: Relative intensity of x-ray diffraction patterns for the illite mineral.
d_{obs}: Observed d spacing of the snow-crystal nucleus.
Intensity: S strong, M medium, W weak, V very.

the illite mineral. Their interplanar distances d (d spacing) agree well with each other, and the shape of this irregular flaky particle is similar to the illite mineral as shown in "Electron Micrographs of Reference Clay Minerals." Therefore, the nucleus can be identified as an illite mineral.

B. *Kaolin*. The kaolin group is characterized by a two-layer type of crystal lattice including kaolinite, anauxite, dickite, nacrite and halloysite. The chemical formula of kaolinite is recognized as $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$, and its shape is hexagonal. Fig. 6a shows the center nucleus of a dendritic snow crystal. The nucleus is a thin hexagonal crystal whose largest extent is 4 μ. The shape is similar to kaolin. Many minute substances are seen around the nucleus; some of them were found to be soluble in water. The electron-diffraction pattern of the nucleus in fig. 6b shows a hexagonal cross grating which is expected to be obtained when the direction of the incident beam is normal to the base plane of kaolin, a clay mineral. The interplanar distances are 2.59 Å for d_{200} and

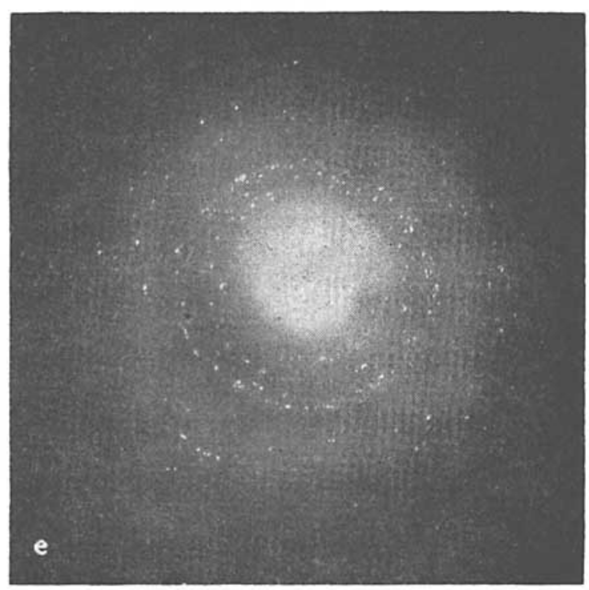

FIG. 5e. Electron diffraction pattern of the nucleus in fig. 5d.

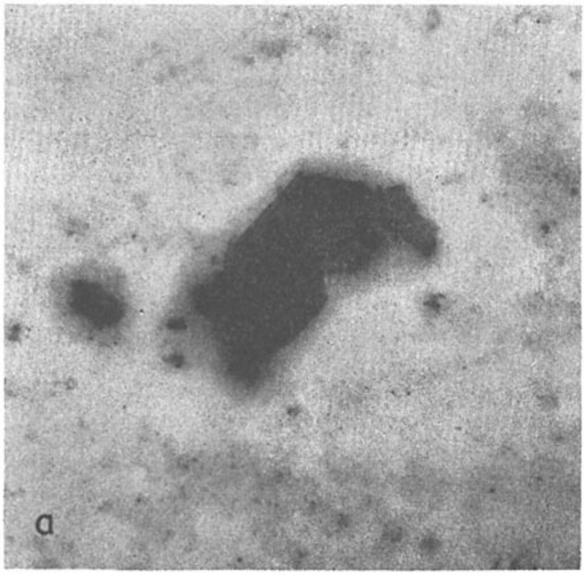

FIG. 6a. A nucleus (kaolin mineral) of a dendritic snow crystal (×8000).

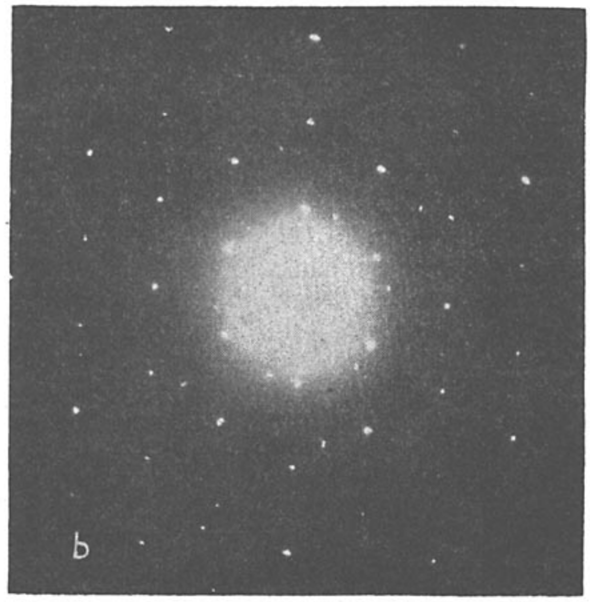

FIG. 6b. The electron diffraction pattern of the nucleus in fig. 6a.

4.49 Å for d_{020}. The length of the unit cell along the a-axis of the crystal is 5.18 Å. The cell dimensions of kaolinite are 2.56 ± 0.012 Å for d_{200} and 4.435 ± 0.06 Å for d_{020} and the length of the unit cell along the a-axis is 5.12 ± 0.12 Å (Pinsker, 1953). The nuclei lattice values coincide with these within the experimental errors. Thus, the nucleus of this snow crystal can be identified as a kaolin mineral.

A crystal which formed on a nucleus of aggregated kaolinite is shown in fig. 7. A snow crystal of bullet form on the mesh is shown in fig. 7a. After the sublimation, a particle remained at the position where the centers of the crystal vanished by sublimation as seen in fig. 7b. Many small particles were found around the nucleus. They had been contained in the other parts of the crystal. The nucleus consists of coagulated small particles of hexagonal shape as shown in fig. 7c. The electron-diffraction pattern of this nucleus was compared with that of kaolinite. The interplanar distances and the shape of the nucleus coincide with the kaolinite mineral; therefore, the nucleus can be identified as a kaolin mineral.

C. *Montmorillonite.* Montmorillonite exists as an essential mineral in soil or in rock bentonite. Nontronite, beidellite, hectorite and saponite are related structurally to montmorillonite and belong to the montmorillonite group. Fig. 8 shows a montmorillonite 2.5 μ in diameter, from a columnar snow crystal. This nucleus showed a Debye-Scherrer pattern, and the interplanar distances were compared to those of montmorillonite. The nucleus consists of thin irregular plates. The interplanar distances and the shape of the nucleus resemble montmorillonite in the common clay mineral.

D. *Attapulgite.* Attapulgite is a hydrous magnesium aluminum silicate in the palygorskite group. The

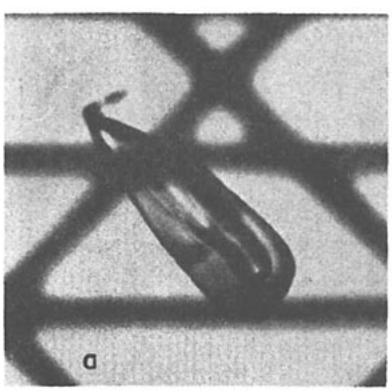

FIG. 7a. A bullet-shaped snow crystal (×160).

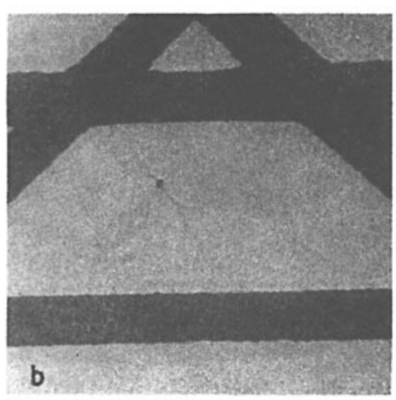

FIG. 7b. The nucleus of the snow crystal in fig. 7a.

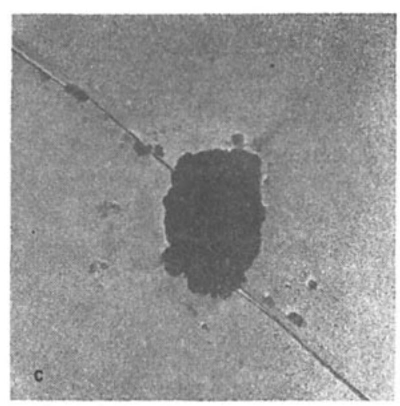

FIG. 7c. The nucleus (kaoline mineral) of the snow crystal in fig. 7a. Cr. shadowing (×5000).

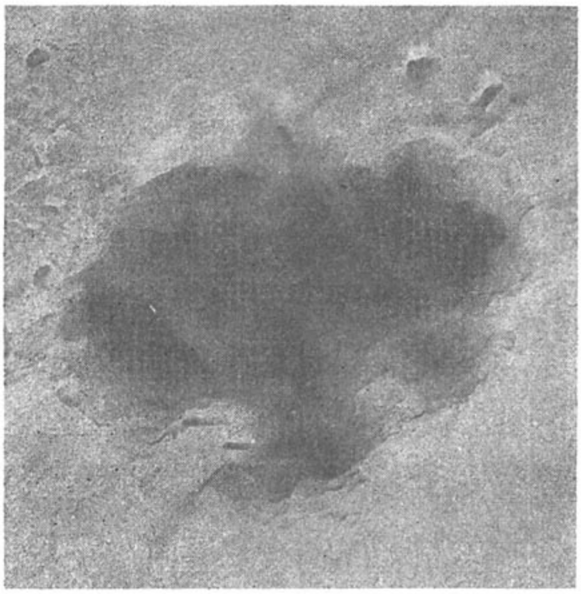

FIG. 8. A nucleus (montmorillonite mineral) of a columnar snow crystal. Cr. shadowing (×25,600).

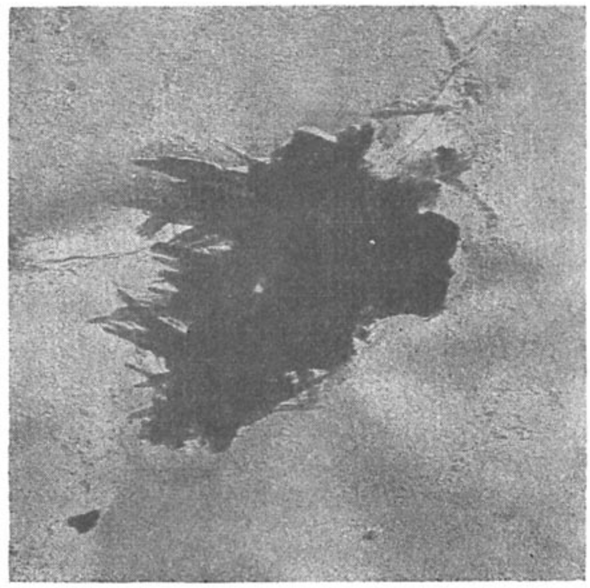

FIG. 9. A nucleus (attapulgite mineral) of a dendritic snow crystal. Cr. shadowing (×8500).

electron-diffraction patterns of attapulgite and montmorillonite are similar. However, the shape of these two minerals as seen in electron micrographs is different. Attapulgite has a rod-like fiber which is different from the flaky structure of montmorillonite. The composition of the ideal cell of attapulgite is $(OH_2)_4(OH)_2Mg_5Si_8O_{20}\cdot 4H_2O$, and there are two molecules present. The dimensions of the cell are $a_0 = 12.9$ Å, and $b_0 = 18$ Å.

Fig. 9 shows a nucleus of a dendritic snow crystal. The nucleus is a bundle of typical rods of fibers, and it shows an electron-diffraction pattern. The d spacing of the nucleus coincides with attapulgite. Attapulgite is a common clay mineral which is of wide distribution. The nucleus can be identified as attapulgite.

E. *Potassium chloride.* The nuclei of two snow crystals among 271 specimens showed a hygroscopic pattern, and the electron-diffraction pattern coincided with the d spacing of the potassium chloride as shown in table 4.

F. *Unknown material.* Some nuclei were not identifiable in this research work. One example is shown in fig. 10. This is a nucleus of a pyramidal crystal. Because of its elliptic shadow, the shape of the nucleus is known to be a sphere. Four other nuclei of spherical shape were found in hexagonal and pyramidal snow crystals. These spherical nuclei show electron-diffraction patterns. The d spacings of these nuclei are similar to Cr_2O_3, Fe_2O_3, Al_2O_3, but the indexing of diffraction patterns is not sufficient to determine the substance. The minerals such as Cr_2O_3, Fe_2O_3, and Al_2O_3 are components of both meteorite and terrestrial minerals. The shape of the nucleus is a sphere, and it is presumed to be a fused particle. It

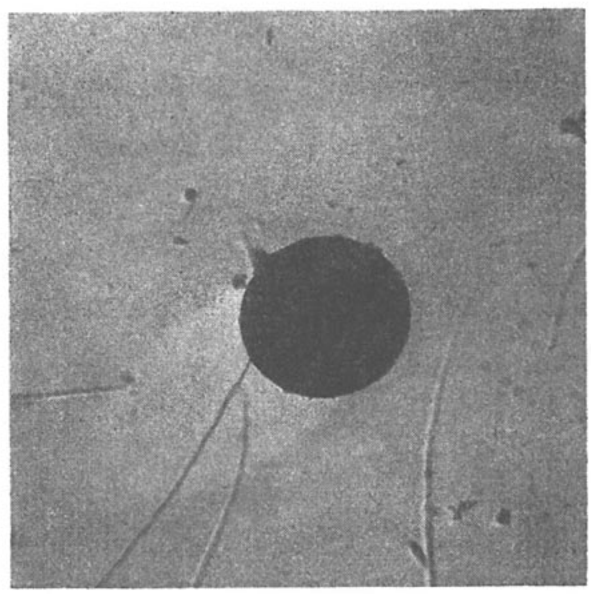

FIG. 10. A spherical nucleus (unidentified mineral) of a pyramidal snow crystal. Cr. shadowing (×8500).

TABLE 4. Interplanar distances of KCl and a snow-crystal nucleus.

KCl		Snow-crystal nucleus	
d	I/I_1	d_{obs}	Intensity
3.13	1.00	3.05	S
2.21	0.61	2.15	M
1.81	0.14	1.89	W
1.57	0.06	1.57	VW
1.401	0.12	1.35	W
1.280	0.06	—	—
1.108	0.02	—	—
1.047	0.02	—	—
0.991	0.02	1.00	VW

TABLE 5. The identification of the substances of snow-crystal nuclei in each snowing day.

Substances	Jan. 21	Jan. 24	Jan. 26	Feb. 16	Feb. 17	Feb. 23	Feb. 26	Feb. 27	Total	%	
Clay Minerals:											
Kaolin group	1	4	0	10	13	50	21	39	138	51	
Montmorillonite group	3	7	0	1	1	11	7	6	36	14	
Illite group	0	3	2	2	0	4	13	9	33	12	87
Attapulgite or Sepiolite	0	1	0	2	0	4	4	1	12	4	
Related mineral	1	1	1	3	1	2	3	4	16	6	
Other substances:											
Potassium chloride	0	0	0	2	0	0	0	0	2	1	
Carbon particle	0	0	0	0	1	3	1	1	6	2	13
Unknown material	1	0	0	5	0	6	4	9	25	9	
Not observed	0	0	0	0	1	2	0	0	3	1	
Total	6	16	3	25	17	82	53	69	271	100	100

TABLE 6. Snow-crystal forms and the nuclei substances.

Substances	Hexagonal plate	Dendritic crystal	Column	Pyramid & bullet	Needle	Total
Clay Minerals:						
Kaolin minerals	76	35	6	13	8	138
Montmorillonite minerals	24	3	3	5	1	36
Illite minerals	16	6	6	5	0	33
Attapulgite or Sepiolite	6	3	2	1	0	12
Related minerals	8	4	3	0	1	16
Other Substances:						
Potassium chloride	0	2	0	0	0	2
Carbon particles	4	1	0	1	0	6
Unknown material	13	8	1	2	1	25
Not observed	3	0	0	0	0	3
Total	150	62	21	27	11	271

is known that fly ash from industrial chimneys have spherical shapes and that some of their substances are metallic components. These spherical nuclei are classified as unknown material in this paper.

5. The center nucleus substance of snow crystal

As described above, a solid particle was found at the center of all but one per cent of the snow crystals examined. This solid particle is thought to have served as the nucleus of the snow crystal. The results of the identification of the substances of the nuclei are summarized in table 5. It can be seen that most of the snow-crystal nuclei were mineral particles which can be identified as one of the aluminum-silicate clay minerals. In addition, some rock and metal minerals such as petarite or pyrite were found; these minerals are classified as related minerals in this table. Almost all kinds of minerals (within this group) were found as snow-crystal nuclei in each of the sample days as shown in table 5. Aluminum-silicate minerals are one of the most abundant substances on the earth's surface. Clay minerals are produced by the weathering of igneous rock. In this table, kaolin minerals include kaolinite, nacrite and halloysite. The montmorillonite group consists of montmorillonite and nontronite. Illite, attapulgite, sepiolite, *etc.* are also common, wide-spread minerals.

According to the cold-box experiments of Mason and Maybank (1958), kaolinite induces ice-crystal formation initially at temperatures below −9C and, after pre-activation, at −4C. Montmorillonite (bentnite) is initially inactive as a nucleating agent above −25C, but after pre-activation it becomes active at −10C. As shown in table 5, mineral particles accounted for 87 per cent of the nuclei; among these, the kaolin group accounted for 51 per cent, montmorillonite group 14 per cent, attapulgite or sepiolite 12 per cent, and related minerals 6 per cent. Carbon-particle nuclei accounted for 2 per cent of the nuclei, potassium chloride 1 per cent, unknown material 9 per cent, and no nuclei 1 per cent. It is concluded that clay-mineral particles are the main source of efficient atmospheric ice nuclei.

The relation between snow-crystal forms and the nuclei substances is shown in table 6. The crystal forms of the specimens are hexagonal, dendritic, column, pyramid, bullets and needle. As seen in table 6, no correlation between the shapes of snow crystals and the composition of their nuclei could be found.

Mason and Maybank (1958) found that in their cold-box experiment there was no difference in the threshold temperatures of kaolinite in the two size ranges 0.2 to 0.5 μ and 5 to 10 μ diameter. If there is a size dependence, it has a relatively small effect.

A relation was found between the size of the snow-crystal nuclei and the snow-crystal forms in this research work. The relationships between snow-crystal forms, maximum frequency in size of snow-crystal nuclei, and their growing temperature by radiosonde sounding are tabulated in table 7. Snow-crystal forms were hexagonal, dendritic, column, pyramid, bullet and needle. The nuclei diameters of maximum frequency are for hexagonal crystals, 1 μ; dendritic, 2.5 μ; column, 3 μ; pyramid, bullet, and needle, 3.5 μ. These relations show a tendency for snow crystals forming at warmer temperatures, such as needle crystals, to have larger nuclei than those formed at colder temperatures, such as hexagonal or dendritic crystals. All nuclei were found to be between 0.2 μ and 8 μ in the largest extension.

TABLE 7. Snow-crystal form, maximum frequency in size of snow-crystal nuclei and the growing temperature by radiosonde sounding.

Form	Max. freq. in size of nuclei: Diameter μ	Frequency %	Temperature by radiosonde
Hexagonal plate	1.0	38	−8−−21
Dendritic crystal	2.5	23	−14−−18
Column	3.0	19	−16−−15
Pyramid & Bullet	3.5	21	−6−−10
Needle	3.5	36	−4− −6.5

Table 8 shows the comparison of snow-crystal nuclei observed in Hokkaido and Honshu, Japan, and Houghton, Michigan. The differences may be due to the meteorological conditions or the locations where the specimens have been collected. The composition of snow-crystal nuclei in Honshu is similar to that in Houghton; mineral particles such as hydrous aluminum silicate accounted for 57 per cent of the nuclei in Hokkaido and 87 per cent in Houghton. There is a smaller percentage of hygroscopic nuclei in Houghton than in Hokkaido. All hygroscopic nuclei in Houghton were potassium chloride. However, in Hokkaido most of them were sea-salt particles; the remainder were potassium chloride. This is because Houghton is located far from the ocean and Honshu is a large island, but Hokkaido is a relatively small island. The percentage of combustion products or carbon particles is smaller for Houghton than for Hokkaido. Micro-organisms (that is, bacteria) were not found in Houghton. In Hokkaido, five per cent of the center nuclei were unobservable; in Houghton, one per cent were unobservable. These facts show that the nuclei in the formation of snow crystals are mainly mineral particles.

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TABLE 8. The comparison of snow-crystal nuclei as observed at Hokkaido and Honshu, Japan and Houghton, Michigan, U. S. A.

Estimated substances	Hokkaido, Japan 1948–1956 Number	%	Honshu, Japan 1955–1958 Isono (1959) Number	%	Houghton, Michigan U.S.A. 1959 Number	%
Mineral particle	176	57	46	88	235	87
Hygroscopic particle	57	19	0	0	2	1
Combustion product	26	8	2	4	6	2
Micro-organism	3	1	0	0	0	0
Unknown material	30	10	4	8	25	9
Not observed	15	5	0	0	3	1
Total	307	100	52	100	271	100

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