THE FORMATION OF ICE CRYSTALS IN THE LABORATORY AND THE ATMOSPHERE[1]

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This paper summarizes some of the author's laboratory and field activities in the realm of experimental meteorology.

Some properties of natural snow particles in the free atmosphere and their electrical characteristics observed in snowstorms are given. A method for classifying snow particles is listed and the typical varieties illustrated by photomicrographs.

Methods of preparing plastic replicas of snow crystals and of snow on the ground are described. Techniques for detecting sublimation nuclei in the free atmosphere are given, involving the use of a cold chamber and supercooled films supported on rings or on thin plastic films.

A description is given of the technique used in simulating natural clouds in the laboratory, and the basic procedures which should be followed are listed.

A brief description of the rôle that certain types of dust particles play in serving as sublimation nuclei is mentioned and the results observed with a considerable number of typical samples and their effectiveness at various temperatures.

The effect on the crystal habit of ice produced by the blocking action of adsorbed surface-active polar molecules is shown.

The latter part of the paper describes briefly some of the aspects of the cloud modification studies initiated by the author in 1946 and subsequently carried out as Project CIRRUS, a joint Army–Navy–Air Force–General Electrical research project concerned with the physical nature of precipitation as snow and rain. A few of the observed results are mentioned, as well as the relationships which are believed to exist in unstable clouds.

During the past seven years, various phases of snow studies have been carried out by the writer and several of his associates of the General Electric Research Laboratory at Schenectady, New York. The earlier work was conducted in a snow laboratory on a slope of the Mohawk Valley in eastern New York. Subsequent studies starting in 1943 and continuing at the present time were also conducted at the Research Laboratory at Schenectady, New York, and the Mt. Washington Observatory in the state of New Hampshire.

The work has included such studies as the electrical properties of snow, the physical effects causing precipitation static on aircraft radio, various techniques for making snow crystal replicas, studies of the changes in snow on the ground, the physical processes related to the deposition of ice on aircraft surfaces, a

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study of supercooled water droplets, both in bulk phase and as an aerosol, methods for detecting sublimation nuclei in the free atmosphere, and finally, various phases of cloud physics, particularly "seeding" techniques which may be used to "trigger off" unstable cloud systems.

Neither space nor time permits more than brief descriptions of the various subjects which have been or are being studied at the present time. References are made, however, to papers which are available in the literature or as reports issued by our laboratory which may be used to obtain more detailed information on the various subjects discussed.

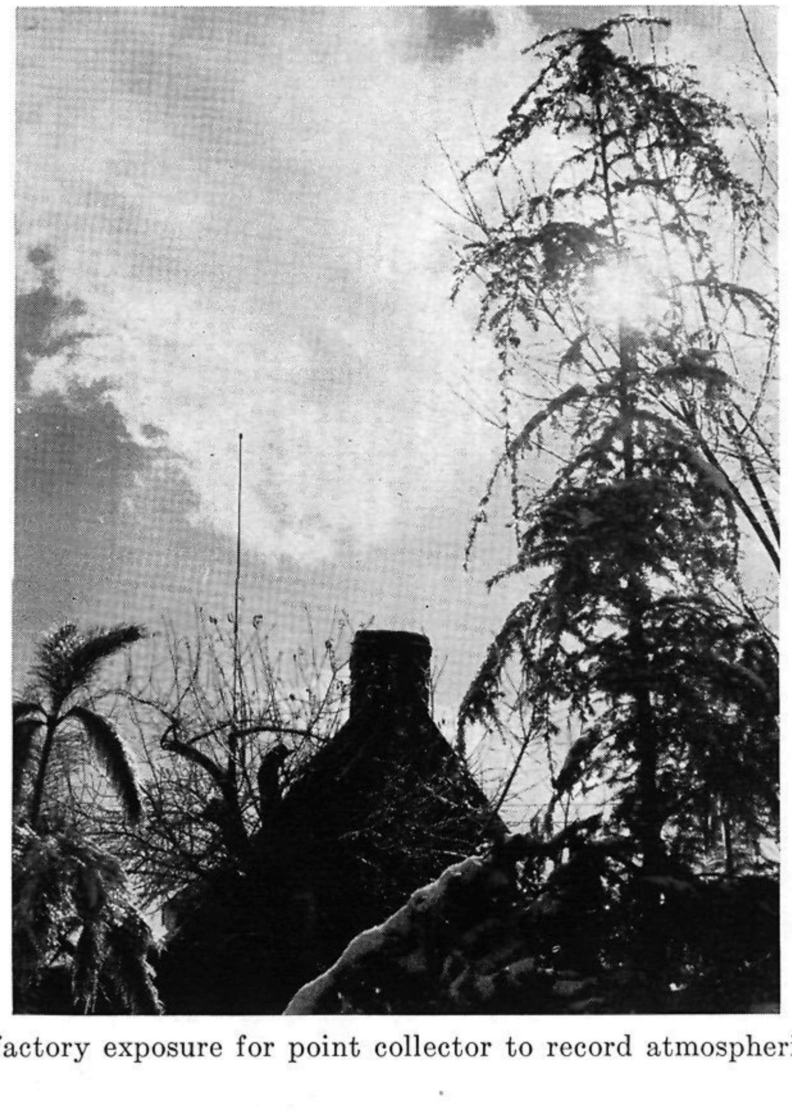


FIG. 1. Satisfactory exposure for point collector to record atmospheric electricity

THE ELECTRICAL PROPERTIES OF SNOW

Early in 1943 several of us in the Research Laboratory were asked to make a study of some of the causes of precipitation static. This is a trouble experienced when aircraft fly through various types of precipitation. Under such conditions, and particularly when the precipitation is in the form of snow, much static noise develops on the aircraft radio. Under severe conditions, the radio becomes useless because of the high noise level, and at times St. Elmo's fire becomes prevalent on the wing and propeller tips and other parts of the airplane.

To understand certain phases of the phenomenon better, the writer made a study of the electrical characteristics of snow particles and snowstorms. The general results of this research have been published (13).

In brief, the studies indicated that one of the main causes of static noise is the charging effect produced when snow particles hit a plane and are shattered, the fragments rubbing and bouncing across the metal surface thus serving as units of a frictional electrical generating system. The studies showed that a single snow crystal may be broken into thousands of fragments. As the plane becomes charged, it finally reaches an unstable condition at which time an electrical discharge occurs which may assume the form of St. Elmo's fire, as previously mentioned. These electrical discharges account for the radio noise.

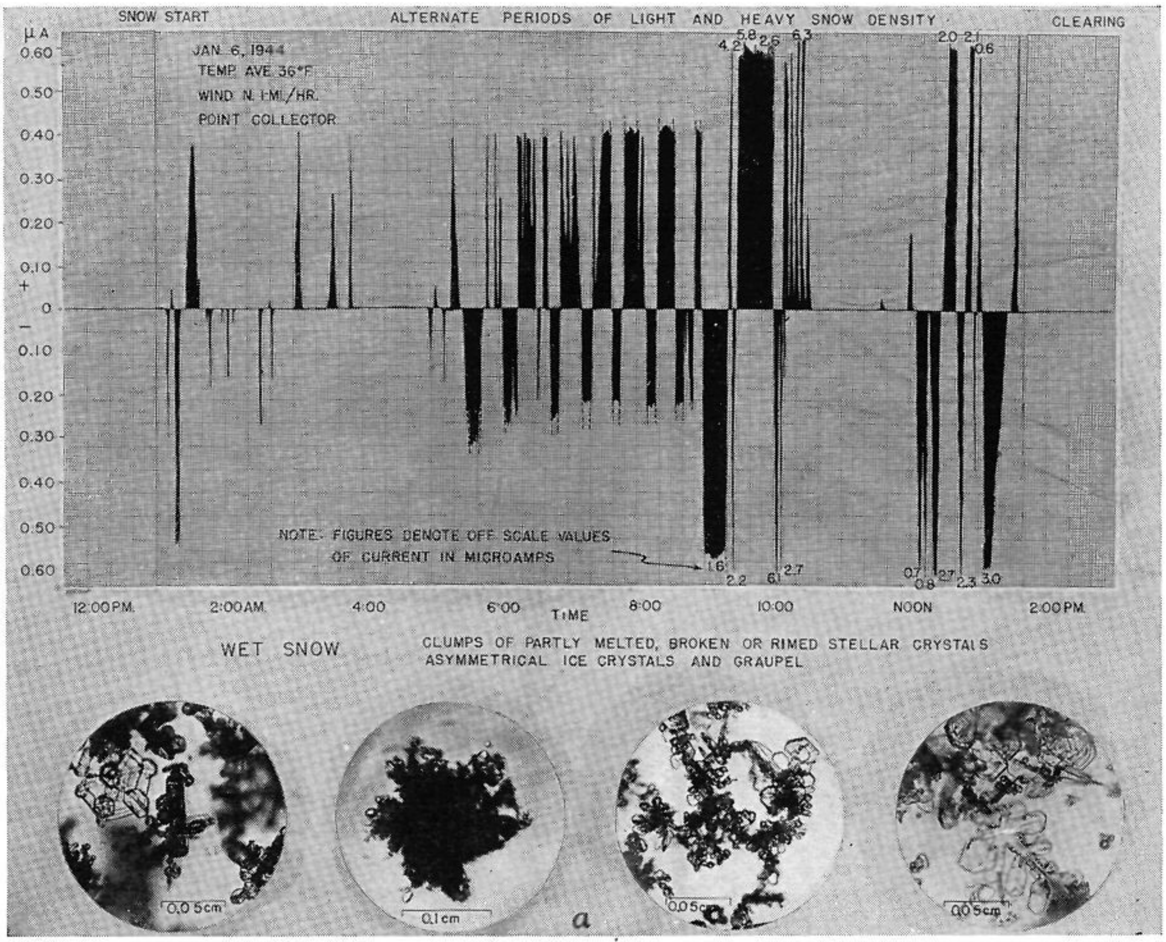
TABLE 1

Comparison of general properties of positive current and cross current storms

GENERAL PROPERTIES	TYPE OF STORM	
	Positive current	Cross current
Air temperature	0–25°F.	25–35°F.
Precipitation forms	3, 4, 5, 7, 8	1, 2, 6, 8, 9
General nature of snowfall	Dry	Wettish
Clumping tendency	Rare	Common
Frozen water content	0.05–0.2 g./m.^{3}	0.05–1 g./m.^{3}
Rate of snowfall	0.1–1 cm./hr.	0.1–3 cm./hr.
Density on ground	0.05–0.15	0.05–0.75
Weight of crystals	4–100 micrograms	4–1000 micrograms
Falling velocity	50 cm. sec.$^{-1}$	20–250 cm. sec.$^{-1}$
Range of falling velocity	Small	Large
Rime deposition	Rare	Common
Cloud structure	Non-convective	Convective
Air-to-ground current	Positive	Positive and negative
Observed atmospheric electricity, C_p collector	Less than 0.25 μa.	-7.3 to $+9.7$ μa.
Frequency of electrical activity	Sporadic and short	Nearly continuous
Relationship of electrical activity to specific clouds	Not obvious	Often quite definite
Rain forms	Light	Thunderstorms

Among the interesting effects observed during the study of the electrical properties of snowstorms was that two types of snowstorms could be identified in terms of their electrical properties. These were termed cross current and positive current storms, the evidence indicating that the former were storms having clouds with a considerable amount of convective activity, while the latter storms apparently consist of relatively stable clouds.

The electrical activity during storms is easily measured by mounting a well-insulated collector such as a needle in a well-exposed position, as shown in figure 1. If the needle is connected by shielded cable to a recording microammeter and then to the ground, records such as shown in figure 2 are obtained in practically all snowstorms. After studying many storms, Table 1 was developed



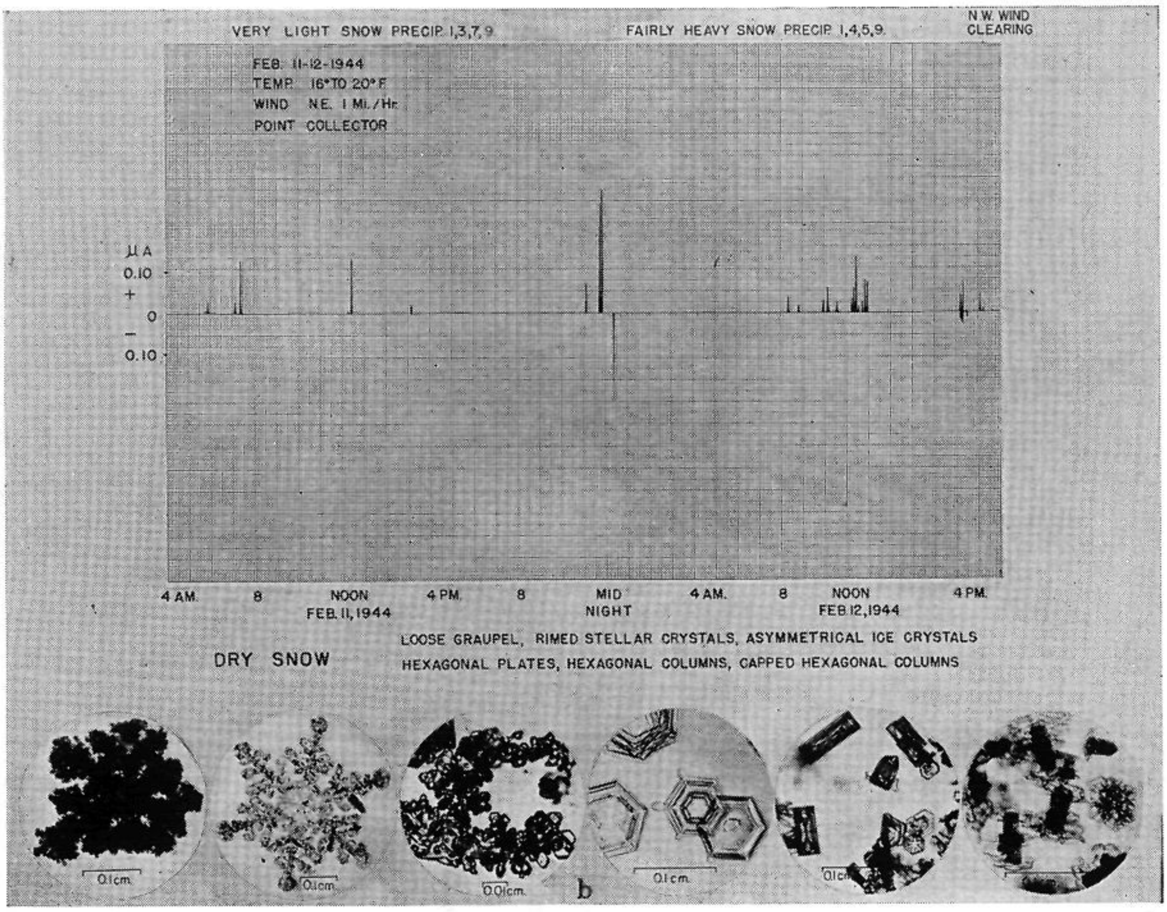


FIG. 2. Typical atmospheric electricity from positive current and cross current snowstorms.

to show the general characteristics of the storms producing these two interesting types of atmospheric electricity.

THE FORMATION OF ICE ON AIRCRAFT

One of the interesting developments following our study of precipitation static at the Mt. Washington Observatory was a shift of our interest from that subject to one of even more importance—the icing of aircraft.

Since Mt. Washington is noted for having "the worst weather in the world," it was found that the summit was an ideal spot to study some of the basic problems related to the icing of surfaces from supercooled clouds. Studies at the Observatory have been carried out each season since 1943 in an effort to under-

FIG. 3. View of Mt. Washington Observatory and supercooled clouds

stand better some of the fundamental phenomena. Much of the results of this work has been published in reports (11), most of which are available from either the U. S. Air Forces or our Laboratory.

It was the writer's visits to Mt. Washington and many observations made on the arduous climbs and descents of the mountain during icing storms and snowstorms that eventually led to the studies resulting in cloud modification techniques. A view of the Observatory after a mild icing storm is shown in figure 3. The clouds above and below the summit are all in a supercooled condition. Under somewhat similar cloud conditions but later in the season, figure 4 shows a relatively thin orographic cloud system which shed snow continuously for several days. A consideration of these observations and many others laid the groundwork for subsequent discoveries.

THE CLASSIFICATION OF SNOW PARTICLES

Any fundamental work with snowstorms and snow crystals immediately points out the necessity of having a simple classification chart to record the crystal forms occurring at a specific time from a storm.

Other investigators in this field (1, 2, 5, 8) have suggested classification methods, but a survey indicated that they were either more complicated than necessary or else failed to include one or more important types. The classification chart shown in figure 5 was prepared by the writer in 1943 for a group of co-

FIG. 4. View of snow clouds from summit of Mt. Washington, New Hampshire

operative observers who assisted him in making a study of the distribution of crystal forms which occur during widespread snowstorms in the northeastern part of the United States. This has been revised several times as the result of experience in the field, but in its present form has now been successfully used for more than three years.

In order to describe the crystal forms observed at a particular time in a snowfall, the code number related to the crystal type is preceded by the letter P. If the crystal is coated with cloud particles so that it has a frosted appearance, the letter R following the code number denotes that condition. Thus the designation P 1 R, 6, 7 indicates that at the observation time stellar crystals coated with rime, ice needles, and asymmetrical crystals were falling from the sky.

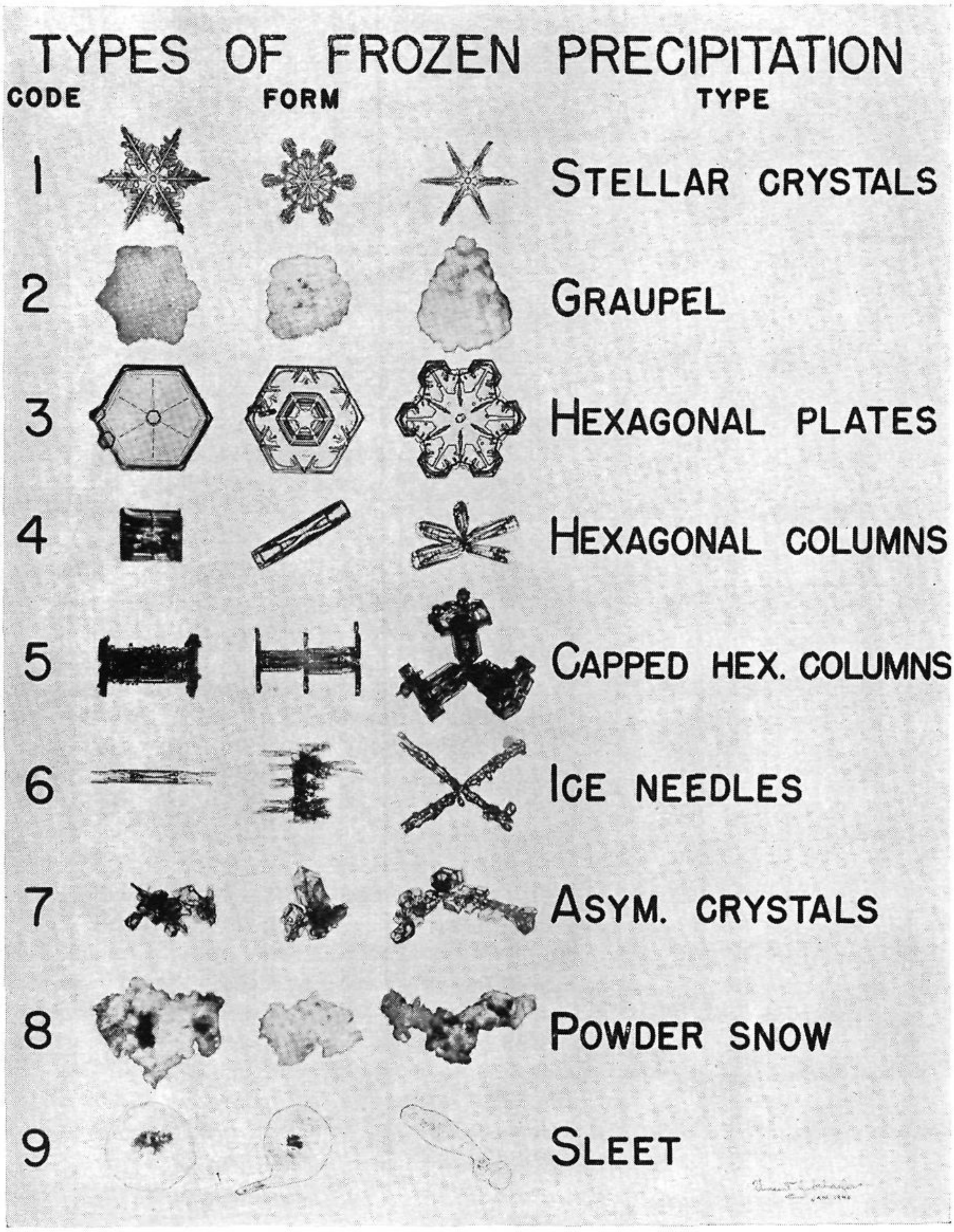


FIG. 5. Classification chart of various forms of frozen precipitation

Greater detail may be included by affixing a subscript number indicating by tenths the relative quantities of each type at the observation time. The photomicrographs used to illustrate the chart show some of the variations which occur with each general type of snow particle. Table 2 indicates the general features and relative sizes of the nine different forms of precipitation. Figure 6 shows the range in size which is commonly observed with a typical crystal form—the hexagonal plate. The larger one comes from relatively low level clouds con-

TABLE 2

General features and relative sizes of frozen precipitation

1. Stellar crystals	Six-rayed starlike forms occurring as single crystals or in clumps; crystals vary in a wide range of form and size, often comprising cottony flakes sometimes 2 in. in diameter; size $\frac{1}{32}$–$\frac{1}{2}$ in.; clumps common
2. Graupel	Snow crystals which are covered with frozen cloud particles forming a rime deposit; this results in pellets of soft snow, which range from hexagonal to rounded kneaded forms; size $\frac{1}{16}$–$\frac{1}{4}$ in.; rarely clumped
3. Hexagonal plates	Thin, hexagonal, solid, or semisolid plates often containing internal structure due to air inclusions; the smallest form of this crystal and the stellar form is known as "diamond dust"; size $\frac{1}{1000}$–$\frac{3}{16}$ in.; rarely clumped
4. Hexagonal columns	Generally flat-ended, sometimes with one end pointed, hexagonal prisms mostly of transparent ice, many with air inclusions; often a group of columns grow from a common source radiating in several directions; the former type produces 22° and 46° halos around the sun; size $\frac{1}{64}$–$\frac{1}{8}$ in.; sometimes clumped
5. Capped hexagonal columns	Similar to hexagonal columns with the exception that both ends terminate as expanded hexagonal plates; a hexagonal plate sometimes is positioned midway between the two plates; many queer forms occur; size $\frac{1}{64}$–$\frac{3}{16}$ in.; sometimes clumped
6. Ice needles	Long slender shafts with hexagonal cross-section often terminating with sharp irregular points on periphery; needles occasionally grow in masses in random directions; size $\frac{1}{64}$–$\frac{3}{8}$ in.; often clumped
7. Asymmetrical crystals	Angular crystals without a symmetrical outline occurring as simple or compound crystals; sometimes crystalline rays originate from a common center but grow in many directions; size $\frac{1}{64}$–$\frac{1}{8}$ in.; often clumped
8. Powder snow	Dry bits of snow without angular form, often of irregular shape; size $\frac{1}{64}$–$\frac{1}{8}$ in.; rarely clumped
9. Sleet	Pellets of translucent to transparent ice often bearing bumps or other protuberances; they are frozen rain drops but are not always round; size $\frac{1}{32}$–$\frac{1}{4}$ in.; very rarely clumped

taining moderate quantities of moisture. Such clouds are generally below the 10,000 ft. level. The crystal shown in the middle of the figure is common to cirrus type clouds and may fall from an altitude as great as 40,000 ft. The smaller crystal is the type known as diamond dust, which commonly forms when the air is supersaturated with respect to ice. Such conditions generally happen with strong temperature inversions accompanied by cloudless skies, the crystals coming from a region less than 1000 ft. above the ground.

COLD CHAMBER STUDIES

The basic experiment (12, 16) in which a supercooled cloud of liquid water droplets may be converted to a mass of snow crystals is a spectacular one which may be easily reproduced with very simple apparatus.

The essential articles for the experiment are an open-topped, well-insulated chamber having a volume of about 1/20 cu.m., e.g., 48 cm. long, 24 cm. wide, 48 cm. deep, which may be cooled to a temperature of −10° to −20°C. The

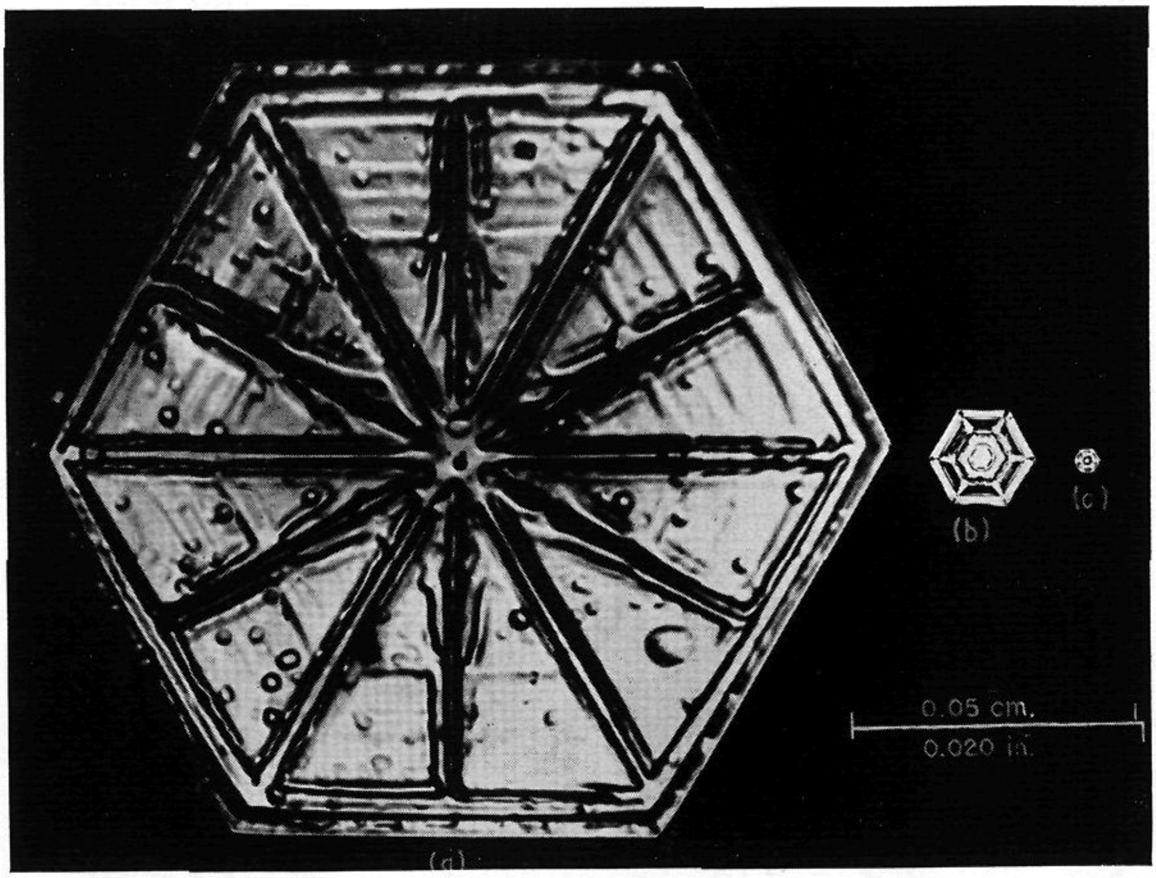


FIG. 6. Relative sizes of hexagonal plate type of snow crystals

walls and floor of the chamber should be painted dull black or lined with black velvet, cloth, or cardboard.

Illumination is simple but important. For best results, the light should produce a brilliant beam and is most satisfactory when mounted above the chamber and directed into it at an angle of 45° from the horizontal.

A supercooled cloud is produced by placing a moist cloth in the cold air of the chamber. The moisture condenses into small water droplets, generally in the size range of 10–20 microns diameter. Another simple method for forming the cloud is merely to breathe into the cold air. Several breaths will produce a cloud having a liquid water content equivalent to 1 g./cu.m. A chamber with a cloud in it is shown in figure 7.

Under ordinary conditions, few crystals will be seen in the chamber. A single

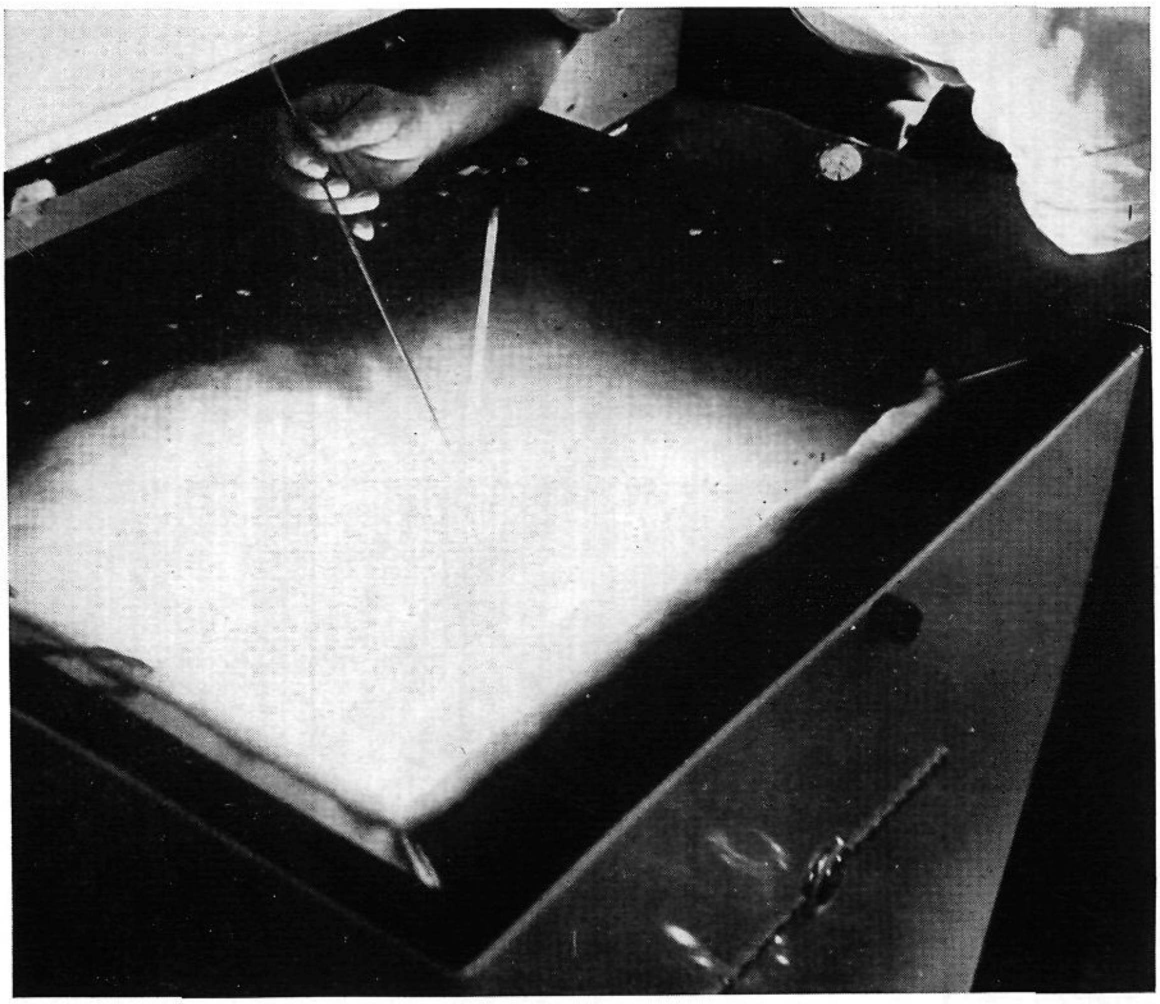


Fig. 7. View of cold chamber containing supercooled cloud

Fig. 8. Trail of ice crystals produced in cold chamber by seeding with needle cooled below −39°C.

crystal may easily be seen among the water droplets, since it grows rapidly, scintillating in the light. They are most easily seen when viewed at right angles to the axis of the beam, since much of the light from them is specular reflection. This is particularly true of hexagonal plates, since they become oriented with their flat surfaces horizontal.

It can easily be shown that most of the condensed cloud formed in the chamber is supercooled. If a fine wire is rotated at 180 R.P.M. in the chamber, it becomes coated with ice. If left alone, the cloud slowly disappears by condensing onto the cold walls of the chamber in the form of frost.

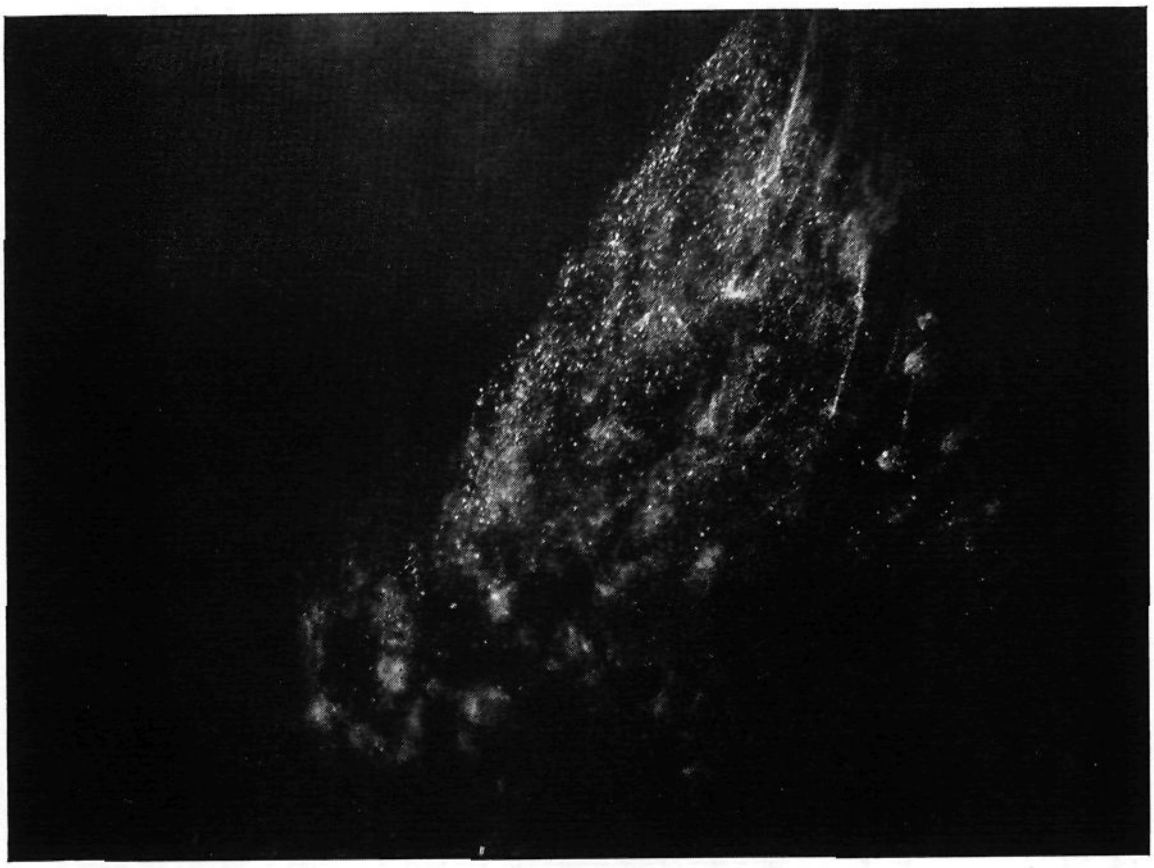


FIG. 9. Twinkling crystals

Within a minute after the supercooled cloud has been formed, the crystals which develop from sublimation nuclei present in the air grow and fall to the floor of the chamber. The supercooled cloud may be suddenly converted to snow crystals by briefly introducing an object colder than −39°C. This may be a needle cooled in liquid air or a tiny particle of dry ice. A single passage of such a cold object produces an effect (figure 8) which quickly changes to a mass of twinkling crystals like those in figure 9. These crystals range in size from 10 to 200 microns. By the time they reach this latter size, the falling velocity is of the order of 1 cm. $sec.^{-1}$, so that it is difficult to keep the particles from "snowing out." Typical crystals produced in this manner are shown in figure 10.

One of the most convenient ways to convert supercooled clouds to ice crystals is by the use of dry ice. Dry ice is easily produced by permitting liquid carbon dioxide to expand into the air. A very convenient source of dry ice for experimental studies is the small 10-g. shells, such as used in soda fountains and in various home appliances. A simple valve for controlling the opening of such containers is available as part of a small model-airplane carbon dioxide engine. By valving the container held in an inverted position above the cold chamber, tiny bits of dry ice will be seen to fall through the cloud, leaving behind tiny

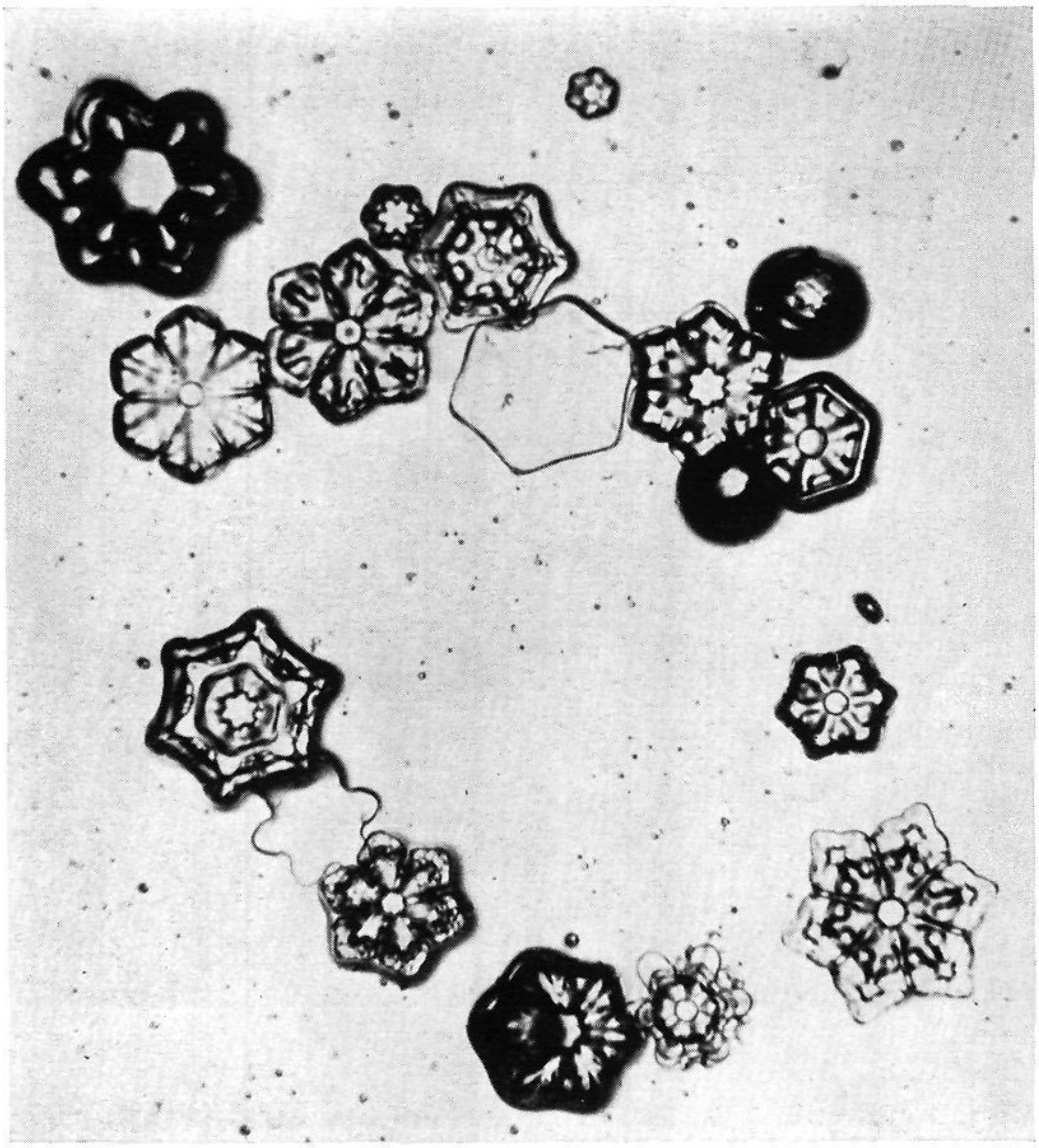


FIG. 10. Typical photomicrograph of snow crystals produced in cold chamber

white "contrails." These rapidly grow until the cloud is completely replaced by a mass of twinkling ice crystals.

Several very interesting phenomena may be seen in this experiment. The cold chamber, when operated with the lid removed, has a temperature gradient with the air warmest at top, colder below. This temperature inversion produces a very stable air condition in the chamber, which resists convection and turbulence. Thus when seeded with dry ice or a cold needle, the heat released by the crystallization process is not sufficient to overcome the inversion stability. If the region of ice crystals is observed carefully, a dark area quickly develops at the interface between them and the supercooled cloud. This is caused by the fact that the ice crystals grow at the expense of the water droplets which evaporate, the ice crystals growing from the condensation of gaseous water vapor, since the vapor pressure of ice is below that of water at all temperatures below 0°C. The dark area often grows to a width of 1.5–2 cm. in a few seconds, showing in a striking manner the phenomenon of diffusion. Another interesting and unusual effect is observed whenever the concentration of ice crystals in the chamber exceeds about 500 per cubic centimeter. In addition to the glint from the crystals as they tumble about in the chamber, a background twinkling is observable. This is due to Brownian motion and is an elegant way to see this interesting phenomenon.

Many other experiments could be described which may be carried out in the cold chamber. These must be deferred at this time, but it should be emphasized that the cold chamber as described is so simple to use that it should become a familiar research tool and demonstration unit for those working in meteorology and other related physical studies.

THE PREPARATION OF SNOW CRYSTAL REPLICAS

Excellent replicas of snow crystals may be prepared by a very simple method which has been described elsewhere (9, 10).

Briefly, it consists in using a dilute solution of a resin in a solvent which does not dissolve ice. An excellent solution consists of 1 per cent polyvinylformal dissolved in ethylene dichloride. When cooled below 0°C., it may be used to produce exact replicas of snow crystals by letting natural crystals fall on a collecting sheet of black velvet cloth or paper and then placing them in a small drop of solution previously placed on a piece of cold glass or slightly absorbent black cardboard. A variation of the technique is to coat a cold glass plate or black cardboard sheet with a thin film of the replica solution, which is then exposed to the falling snow crystals for a specific time period ranging from 5 to 60 sec., depending on the density of the snowfall. This produces a permanent record of the snowfall at that particular time in terms of snow crystal type, concentration of crystals, and other factors of importance to obtaining a better understanding of the physical features of such storms.

The technique is so easy that relatively untrained observers can obtain excellent results by observing a few basic rules. The method also lends itself admirably to sampling from planes, at remote stations, and by automatic exposure instruments. The samples thus obtained may then be studied at leisure in a warm laboratory. A photomicrograph of a replica is shown in figure 11.

THE PREPARATION OF REPLICAS OF SNOW ON THE GROUND

In addition to the various replica techniques previously described, there is another which may be used to advantage in preparing replicas of snow on the ground. For this purpose, a more concentrated solution of polyvinylformal should be used, having a concentration of between 3 and 5 per cent.

To illustrate one of the possibilities, replicas were prepared of a cross section of snow on the ground, as shown in figure 12. To prepare these records, a 5-cm. strip of 3 per cent solution was spread lengthwise on a piece of window glass 24 in. long. A vertical face was cut into a fresh blanket of snow and the coated glass pressed firmly against it, withdrawn, and then placed in a well-ventilated but sheltered location to permit the solvent to evaporate. Until all traces of the solvent have evaporated, the replica must be kept at a temperature colder than 0°C. The best replicas are obtained when the snow particles are permitted to sublime. If it is desirable to hasten the process, however, the sample may be warmed as soon as the solvent is gone, which may take from 20 to 60 min.

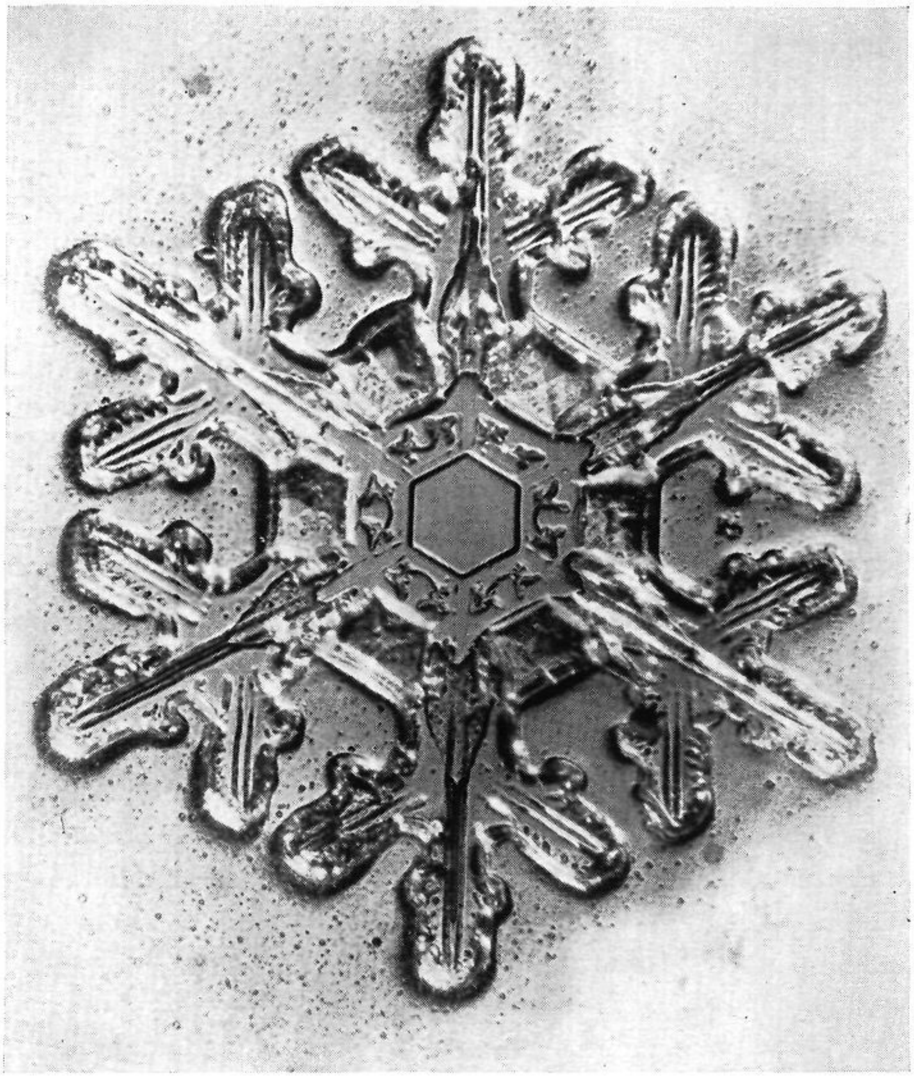


FIG. 11. Photomicrographs made by transmitted light of snow crystal replicas

depending on the ventilation and the temperature of the replica sample. After obtaining a cross-section replica as described, subsequent samples may be obtained by successive applications of the solution side by side until the surface area of the backing is exhausted.

The sample shown in the photograph illustrates a vertical shrinkage of the snow layer during a period of 5 successive days during which time the air temperature remained continuously below 0°C. Although the shrinkage amounted to more than 50 per cent in snow thickness, the density increased from less than 0.10 to more than 0.20, so that the content of liquid water remained nearly the same. Examination of the structure of the snow particles on the fifth day showed that, although most of the original snow consisted of perfect crystals of stellar, hexagonal plate, and column types, a major change occurred during the 5-day period, producing granular snow approaching the rounded firn forms.

Subsequently, as shown by the photograph, several additional light snowfalls occurred which were also recorded by the replica technique.

It is hoped that eventually a snow replica solution may be developed employing the solventless type of plastic in which several monomers are catalyzed by an oxidizing agent to form a true three-dimensional structure. Until such a material becomes available which employs materials which do not modify ice, satisfactory replicas may be prepared as described, particularly when good ventilation is provided.

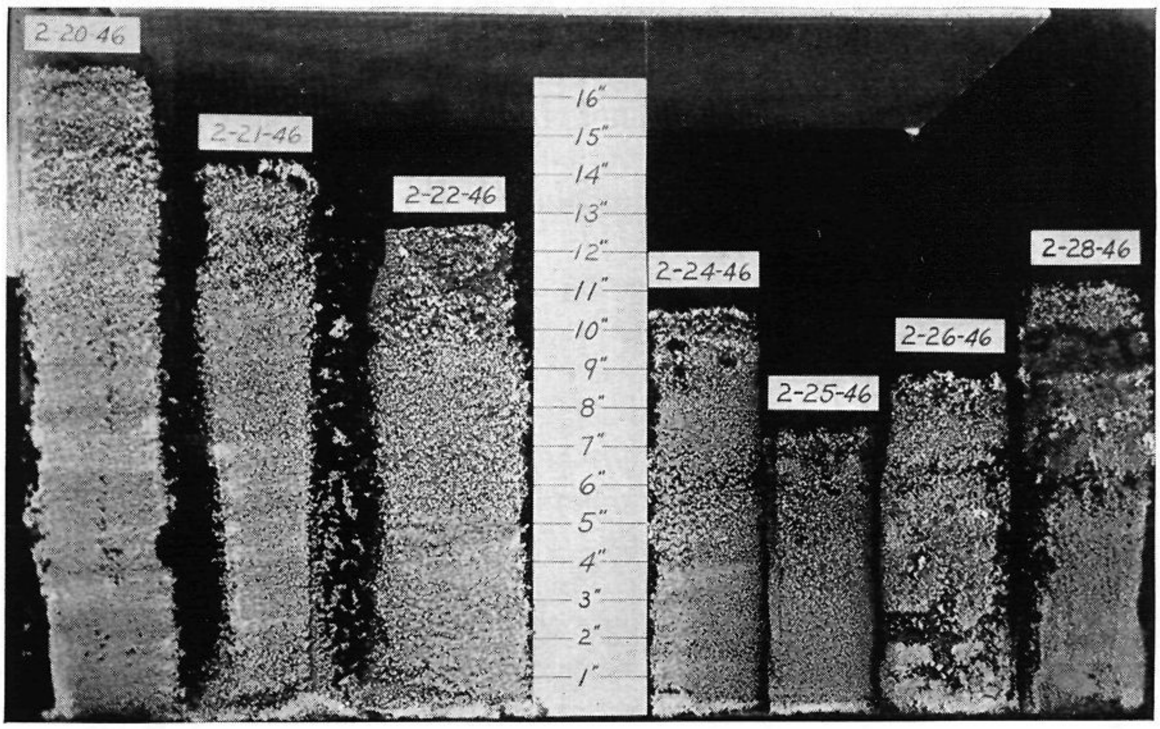


FIG. 12. Replica of cross section of snow deposit on ground

THE DETECTION OF ICE NUCLEI IN THE FREE AIR

The random occurrence of supercooled clouds in the free atmosphere is a well-known fact. The reason for this seemingly haphazard phenomenon is probably closely related to the concentration of sublimation nuclei in the particular air mass when clouds occur.

A better knowledge of sublimation nuclei is needed, particularly of particles other than ice which lead to the formation of ice crystals in air supersaturated with respect to ice. Unfortunately, such particles are generally of microscopic or submicroscopic size and, in addition, include only a small fraction of the dust in a given sample of air.

It is important to differentiate between *condensation* and *sublimation* nuclei. The former are always much more common than the latter. As will be shown in a subsequent section, a third type of particle, the *freezing* nucleus, is also of importance in this connection.

Three methods have been devised by the writer for detecting sublimation nuclei in the clear air of the atmosphere (17).

The most satisfactory of these methods is to use the cold chamber of the type previously described. A sample of the surrounding air is introduced into the chamber by simply removing the air within it by employing a suction fan. Unless the air which is introduced has a very low humidity or was nearly as cold as that in the chamber, condensation will occur. By illuminating this newly formed cloud with a strong beam of light, every active sublimation nucleus which was present will appear as a twinkling snow crystal, easily discernible among the surrounding supercooled cloud droplets. The temperature of the air in the cold chamber will have a direct relationship to the number of particles likely to be observed.

Observations made during 1947 and 1948 on the summit of Mt. Washington (6288 ft.), New Hampshire, and in the Mohawk Valley show the concentration of sublimation nuclei at a cold chamber temperature of -20°C. to vary from none observable to values up to 10 per cubic centimeter. The type of crystal which forms under such conditions probably bears a definite relationship to the substance serving as the active nucleus. These crystals may be examined by direct examination under a microscope kept in the chamber, by preparing plastic replicas, or by permitting the crystals to seed a thin supercooled water film.

Certain "soap" bubble solutions serve admirably for this latter purpose. Most solutions work except those containing large amounts of glycerol or similar freezing-point depressants. A film is formed on a ring 5–10 cm. in diameter and then held in the cold air of the chamber. The film supercools in about a second. If visible crystals are in the air, the number of separate crystals which develop and grow in the film appears to be in direct proportion to that concentration. At least four types of crystals grow in the film. A six-membered stellar form appears when the crystals are hexagonal plates. A symmetrical cross or x-shaped crystal appears when hexagonal columns occur. An irregular-shaped crystal predominates when asymmetric crystals are present. It should be remembered that there may also be crystals developing from freezing nuclei within the solution. Whenever this occurs, the crystals tend to have frostlike forms with a tendency to be curved. There is a time lag also before such crystals appear, as compared to the effects seen when sublimation nuclei or ice crystals are present.

A variation of this method is to blow bubbles in the free air when its temperature is below freezing. When this is done, crystals will sometimes develop in the bubble. Occasionally a single crystal will grow completely around a bubble 10 in. in diameter. Figure 13 illustrates an effect commonly seen. Tables 3, 4, and 5 show typical data indicating the variations which have been observed during the past winter.

Permanent replicas of such effects may be prepared in the following manner: On a frame of convenient size, such as 9 x 12 cm., a thin film of cellulose acetate is stretched and cemented. This film is then coated with a thin layer of 3–5 per cent of polyvinyl alcohol dissolved in water. This solution readily supercools and when seeded with sublimation nuclei or ice crystals will produce effects

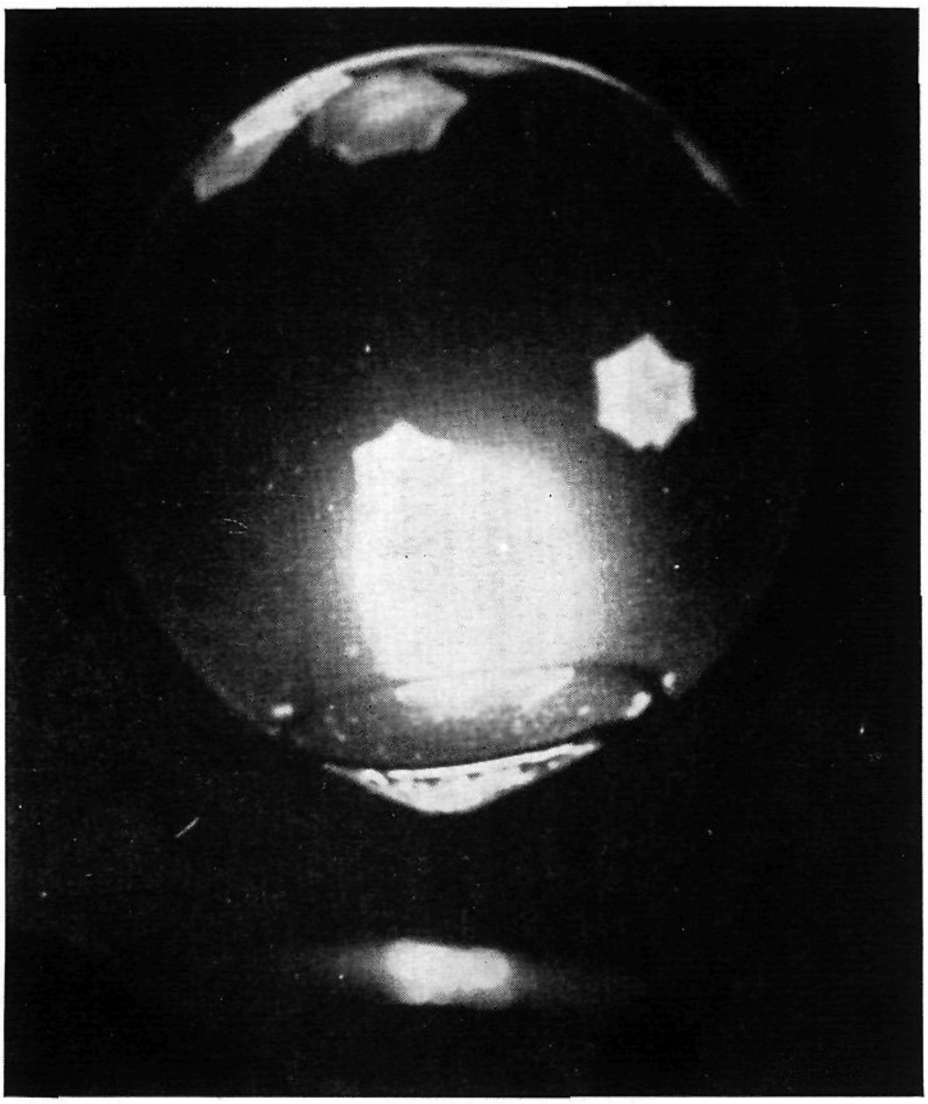


FIG. 13. Supercooled bubble in which ice crystals are growing

TABLE 3

Extremes in number of sublimation nuclei as a function of air temperature

DATE	TIME	TEMPERATURE	NUMBER OF NUCLEI	TYPE	COMMENTS
		°C.			
February 11, 1948	0800	−18	>50	Hexagonal	Alto stratus clouds
February 24, 1948	0700	−18	20–30	x	Supercooled fog in lowlands
December 25, 1948	0730	−14	30–40	Hexagonal	Clear
January 23, 1948	0800	−14	10–15	Hexagonal	Stratus overcast
January 22, 1948	0815	−10	10–20	Hexagonal, x	Stratus overcast
February 13, 1948	0745	−10	3–5	Hexagonal, x	Cirrus haze
January 25, 1948	1200	−7.8	20–30	Hexagonal, x	"Frosty" air
February 19, 1948	0730	−7.8	1–5	x	Cirrus haze
February 15, 1948	0800	−6.6	5–10	Hexagonal, x	0.8 strato cumulus
March 1, 1948	2200	−6.6	None		Cirrus haze
February 24, 1948	2300	−6.1	None		Clear, 22° halo
March 15, 1948	0645	−6.1	None		Cirrus haze

identical with those already described for the soap film. Figure 14 shows two samples of the growth of ice crystals in such films. If, after the crystallization is

TABLE 4

Variation in number of ice nuclei under nearly isothermal conditions

DATE	TIME	TEMPERATURE	NUMBER OF NUCLEI	TYPE	COMMENTS
		°C.			
February 11, 1948	1300	−9.5	20–30	Hexagonal, x	Alto cumulus
January 22, 1948	0815	−10	10–20	Hexagonal, x	Thin cumulus humilus
January 23, 1948	0745	−9	10–20	Hexagonal	Cumulus humilus
February 23, 1948	0800	−9.5	5–10	Hexagonal, x	Scattered fracto cumulus, hazy
February 13, 1948	0745	−10	3–5	Hexagonal, x	Cirrus haze
March 11, 1948	0645	−9.5	2–5	Hexagonal	0.8 strato cumulus sun pillar
February 22, 1948	2200	−9.5	1–3	Hexagonal, x	Clear
March 6, 1948	2130	−9	1–2	Hexagonal, asymmetrical	Cirrus haze
March 13, 1948	2200	−9	1–2	x	Thin haze
February 20, 1948	0745	−9.5	0.1–1.0	Hexagonal	Clear

TABLE 5

The relationship of concentration of sublimation nuclei to the temperature of the free atmosphere

Cold chamber method—Mt. Washington Observatory

FREE AIR TEMPERATURE	NUMBER OF OBSERVATIONS SHOWING VARIOUS CONCENTRATIONS			
	ca. 1×10^6/m.^{3}	ca. 1×10^3/m.^{3}	$< 1 \times 10^2$/m.^{3}	None
°C.				
−30	4	5	1	1
−26	3	3	1	4
−22	5	4	0	1
−18	6	4	0	1
−14	3	5	0	3
−10	5	4	0	3
−6	1	2	5	4
−2	0	1	3	7
+2	0	3	3	6
+6	2	0	6	3
+10	0	2	3	6
+14	0	1	9	1

completed, the film is kept below freezing until the ice disappears by sublimation, a permanent replica remains. This was formed by the polyvinyl alcohol being precipitated at the interfaces between the ice crystals.

THE EFFECTIVENESS OF CERTAIN DUSTS AS SUBLIMATION NUCLEI

It is obvious that there are physical processes in the natural atmosphere which lead to the formation of snow at temperatures only a few degrees below 0°C. In the past, it has been suggested (3) that graphite and quartz dust carried into

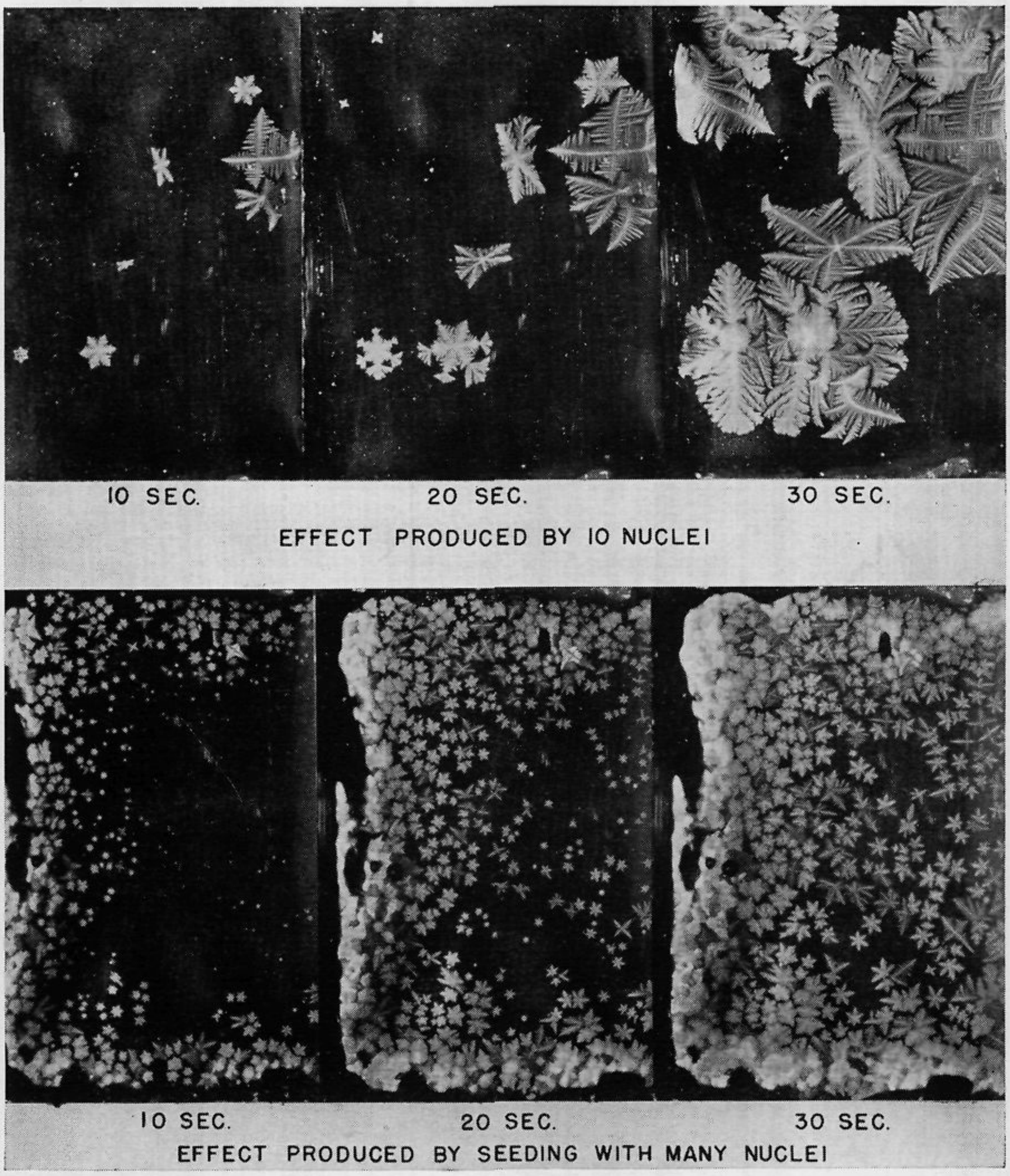


FIG. 14. Stages in growth of ice crystals in supercooled 3 per cent solution of polyvinyl alcohol.

the atmosphere were the active sublimation nuclei for ice crystals. The hexagonal structure of these materials in their crystalline form was thought to be of importance in their activity. So far as I am aware, no information has been published of any verification of these ideas by laboratory experiments or observations of nuclei of actual crystals.

It was found (19) by Dr. Vonnegut of our laboratory that small particles of silver iodide were quite effective as ice nuclei. This material has a rather high temperature coefficient, the warmest temperature at which any particles are active being about $-5°C.$ with a much greater activity at $-10°C.$

During the past year, I have found a number of naturally occurring dusts which serve as effective sublimation nuclei. Among these are volcanic dusts, silicates, and clays. In most instances, these materials have been selected from extensive natural deposits in arid or semiarid regions noted for their duststorm activities. Each material has its own temperature range of activity for producing effective nuclei. For example, a sample of loess from an ancient lake bed near Richland, Washington, when formed as an airborne dust shows the

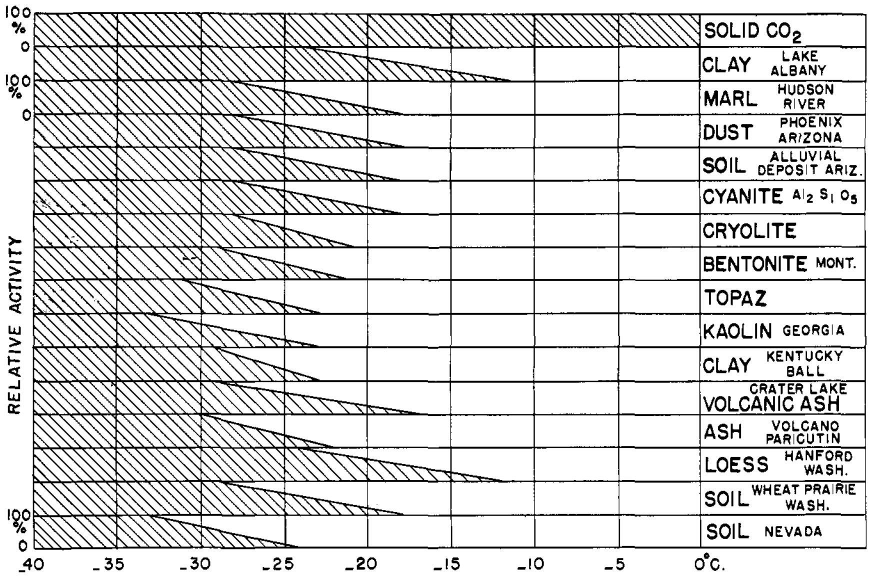


FIG. 15. The temperature–activity relationships of various kinds of foreign particle ice nuclei.

presence of a small percentage of particles that form snow crystals in saturated air at a temperature of $-12°C.$ At $-24°C.$, all of the particles are "active" nuclei. Another dustlike soil from a desert in Nevada does not show any activity until the temperature reaches $-24°C.$ and does not reach 100 per cent activity until the temperature is $-33°C.$ The temperature–activity relationships of a number of such natural dusts are shown in figure 15 and several typical snow crystals grown on such foreign nuclei are shown in figure 16.

It is believed that the mechanism responsible for the activity of such particles is the fact that the surface of the particle is conducive to the formation of frost. As soon as a frost deposit occurs on the surface of the particle, growth continues as though the particle were made up only of ice. Dust particles larger than 1 micron generally produce asymmetrical crystals with irregular platelike growths, although smaller particles often lead to the development of very symmetrical ice crystals.

Since these dusts are active in the temperature range where snow is commonly observed, it is quite likely that they are responsible for the initiation of many snowstorms. It is unlikely, however, that such materials would be so concentrated that they could provide the tremendous quantities of nuclei necessary to keep a snowstorm going. It has been my observation that the majority of snow particles from storms coming from relatively low clouds are asymmetrical crystals which seem to be growing on fragments of other crystals. This strongly suggests that snowstorms might often be initiated by foreign

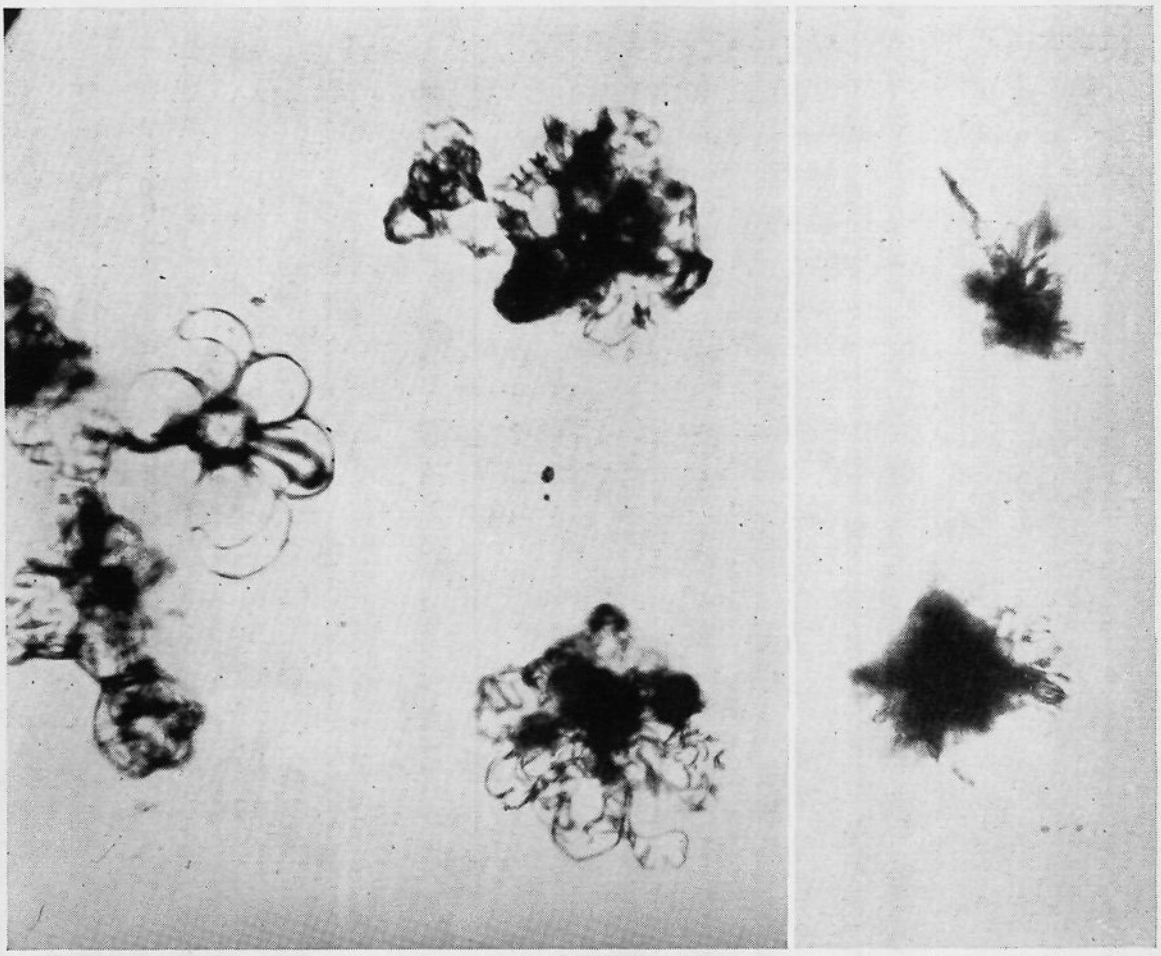


FIG. 16. Typical snow crystals formed on foreign particle nuclei

particle sublimation nuclei in the early stage of development of the storm. These particles grow big enough and have a sufficient concentration so that for a short time they collide with one another. The resulting fracture from collision and abrasion and the electrical effects which result would then produce large numbers of tiny ice fragments, each of which would serve as an effective nucleus up to the freezing temperature. This mechanism is thus a chain reaction of a type somewhat comparable to that described by Langmuir (6) for the development of precipitation in warm clouds.

THE MODIFICATION OF SNOW CRYSTAL FORMS BY VAPORS

The variations in snow crystal forms shown in figure 5 are generally believed to be due to variations in temperature and moisture supply. It has been shown by Nakaya (7) that most of these variations in crystal form may be duplicated in a laboratory cold chamber by carefully controlling the temperature and moisture supply as a crystal grows on a supporting fiber.

Work in our laboratory (20) shows that modifications to the crystal form may also be produced by introducing certain organic vapors into the chamber in which the crystals grow. Unlike Nakaya's experimental method in which single crystals are grown, our experiments generally produce a minimum of 10^7 crystals.

A typical experiment employs air having a mean temperature of -15°C. Moisture is added to the air by the evaporation of water from a porous porcelain

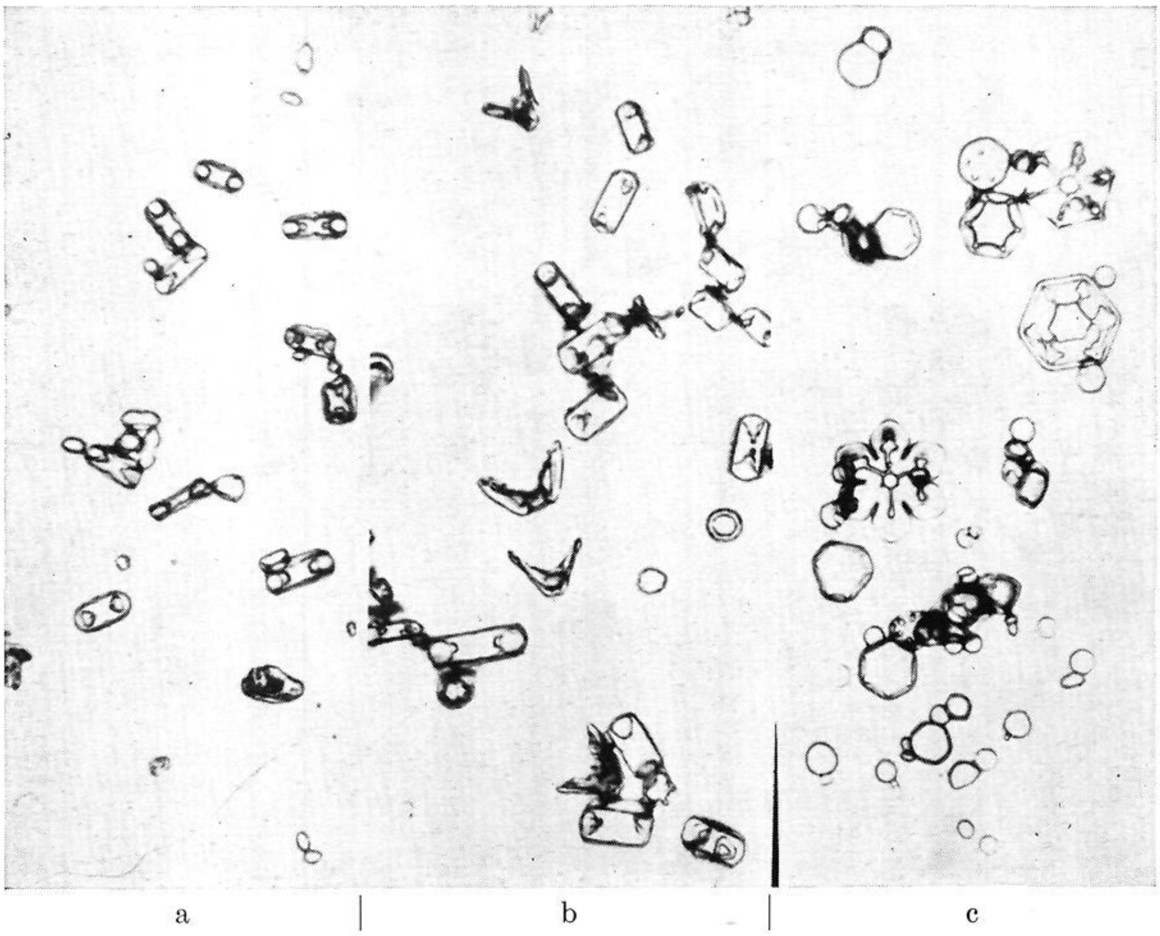
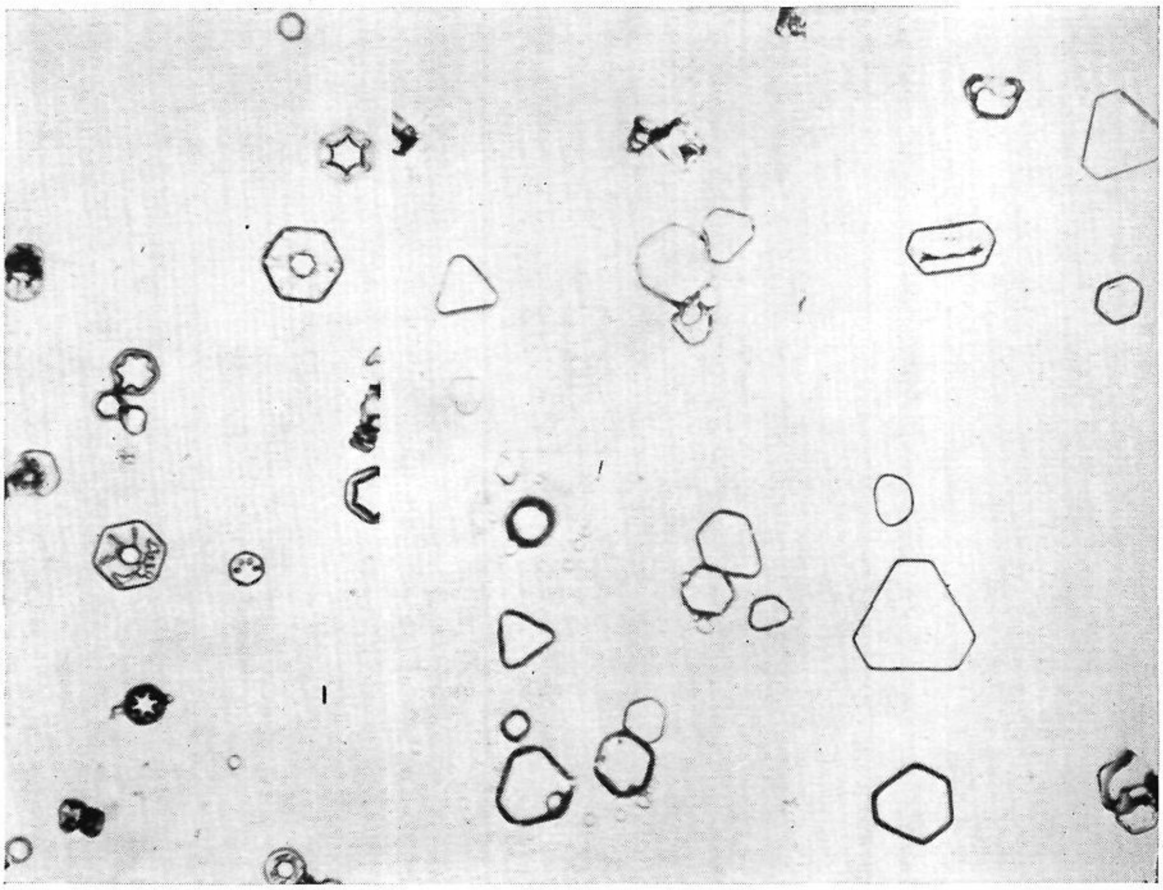


FIG. 17. The effect of acetone vapor on the crystal habit of ice. (a) Effect of 1 molecule of acetone with 10 molecules of water; (b) effect of 1 molecule of acetone with 100 molecules of water; (c) effect of 1 molecule of acetone with 1000 molecules of water.

container or by use of a nebulizer (a small aspirator constructed within an outer glass container which directs an atomized water stream against the surface of the outer wall, the finer particles and accompanying saturated air emerging as a stream of water condensate from a tube on the top of the nebulizer).

Under normal conditions when the supercooled cloud is "seeded" with dry ice or when adiabatic expansion cools a small sample of the air below -39°C., the ice crystals which form in ordinary laboratory air are simple hexagonal plates. When, however, a small amount of certain organic chemicals is introduced into the air, either by vaporization or by use of the nebulizer, the crystals which form are modified. For example, as shown in figure 17, acetone in concentrations as low as one molecule of acetone to 100 molecules of water causes all of the crystals to become either hexagonal columns or a peculiar combination of plate and column. On the other hand, the introduction of the vapor of nitric acid or nitrous oxide (figure 18) tends to emphasize the trigonal symmetry of the water molecule so that the majority of the crystals are triangular with some of them being almost perfect isosceles triangles with sharp corners bearing no suggestion of the hexagonal pattern so common to crystals formed in "normal" air.

The use of acetic acid (figure 19) encourages the development of large stellar-like crystals containing intricate structural designs. Larger molecules of the



a | b | c

FIG. 18. The effect of nitrous oxide and nitric acid vapor on the crystal habit of ice. (a) Crystals formed in ordinary air; (b) effect of trace of nitric acid vapor in air; (c) effect of nitrous oxide smoke in air.

fatty acid series having an appreciable vapor pressure, such as valeric and caprylic acids, produce the columnar form of prism similar to that observed with acetone.

The aldehydes also modify the ice crystal structure, formaldehyde causing rather short columns while heptaldehyde produces a peculiar crystal somewhat akin to the capped hexagonal column.

Other chemical types which produce definite modifications are dioctyl sodium sulfosuccinate (OT 100), heptyl and octyl alcohols (methyl and ethyl are not effective), certain of the silicones, notably dimethyldichlorosilane, pentamethylaminomethyldisiloxane, and the ketones.

A preliminary consideration of the results shows that the most effective chemicals for producing modification are those which have strong polar properties. These are generally surface-active substances and presumably produce the modification by becoming preferentially adsorbed on certain faces of the crystals. This restricts growth in some directions, allowing more growth on others.

The study up to the present time indicates that the modification of the crystal is a continuing phenomenon and not one which is directly controlled by the crystal habit of the nucleus. By producing a batch of crystals by the adiabatic expansion of air containing acetone vapor and then permitting them to grow in saturated air under normal conditions, a typical crop of hexagonal plates was produced.

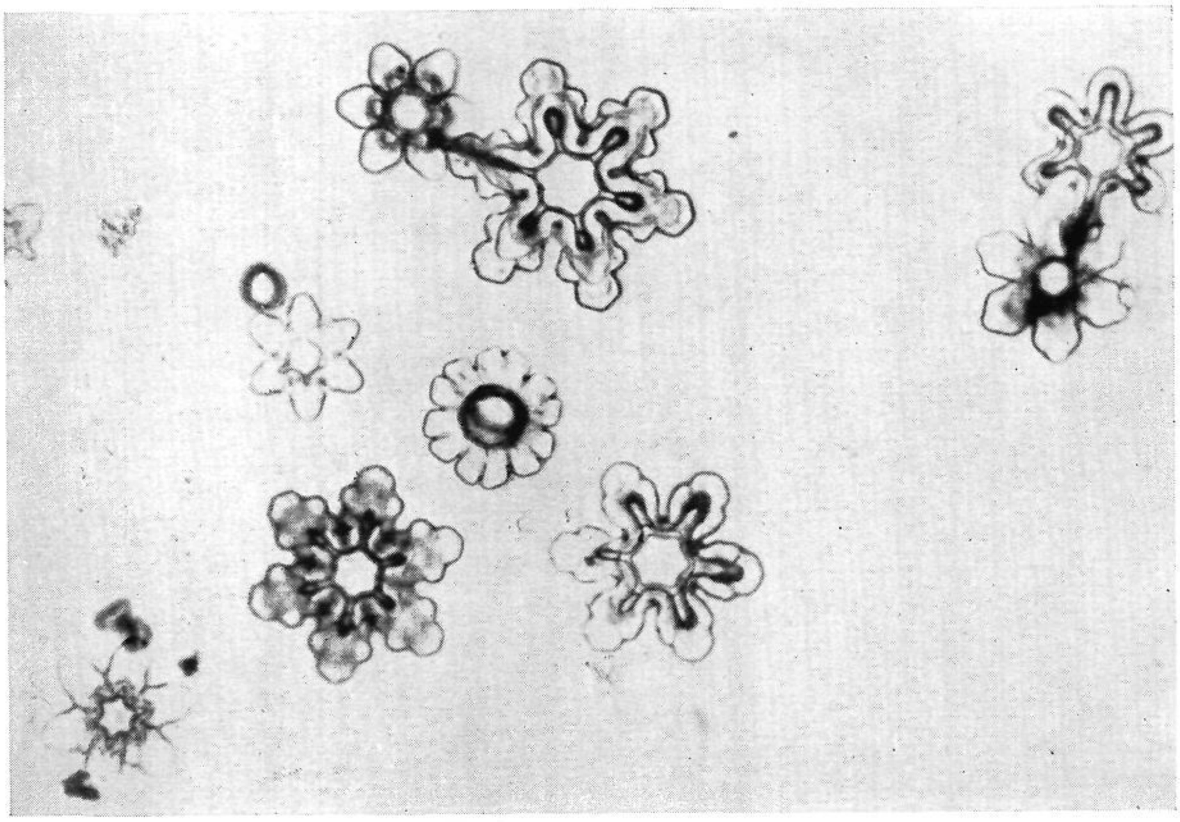


FIG. 19. The effect of acetic acid vapor on the crystal habit of ice

This experiment was made by placing a tiny wad of filter paper soaked in acetone into a "pop" chamber having a capacity of 0.025 cc. Saturated air at 25°C. was introduced into this chamber under a pressure of 30 lb. per square inch. By the sudden opening of the chamber, using a high-speed release system, the air expanded and cooled below the critical temperature of −39°C., causing the spontaneous formation of ice crystals in great numbers. These grew at the expense of a supercooled cloud in the cold chamber. Instead of forming hexagonal columns, as would be the case if the original crystal habit were dominant, the crystals which formed were of the "normal" variety of hexagonal platelets.

Work in this field is continuing, since many fundamental phenomena in the field of crystallography may be studied with great ease with the techniques which have been developed.

NOMENCLATURE IN EXPERIMENTAL METEOROLOGY

As the new science of experimental meteorology develops, especially as applied to cloud seeding and modification, certain experimental procedures are necessarily so often described that it becomes worthwhile to introduce new terms to simplify description and reduce the chance of ambiguity.

Since clouds occur in air which may be warmer or colder than 0°C., it was decided by our group to follow a suggestion of Langmuir's and adopt the following terminology.

The term a *cold cloud* shall be used to describe a cloud of water droplets which is supercooled and all of the cloud is in air which is colder than 0°C.

A *warm cloud* consists of a cloud of water droplets existing in air of which all parts are warmer than 0°C.

A *cool cloud* shall be the term used to describe a cloud which combines the properties of both *warm* and *cold* clouds; that is, the cloud exists in both above and below freezing air, with the freezing level extending through some part of it.

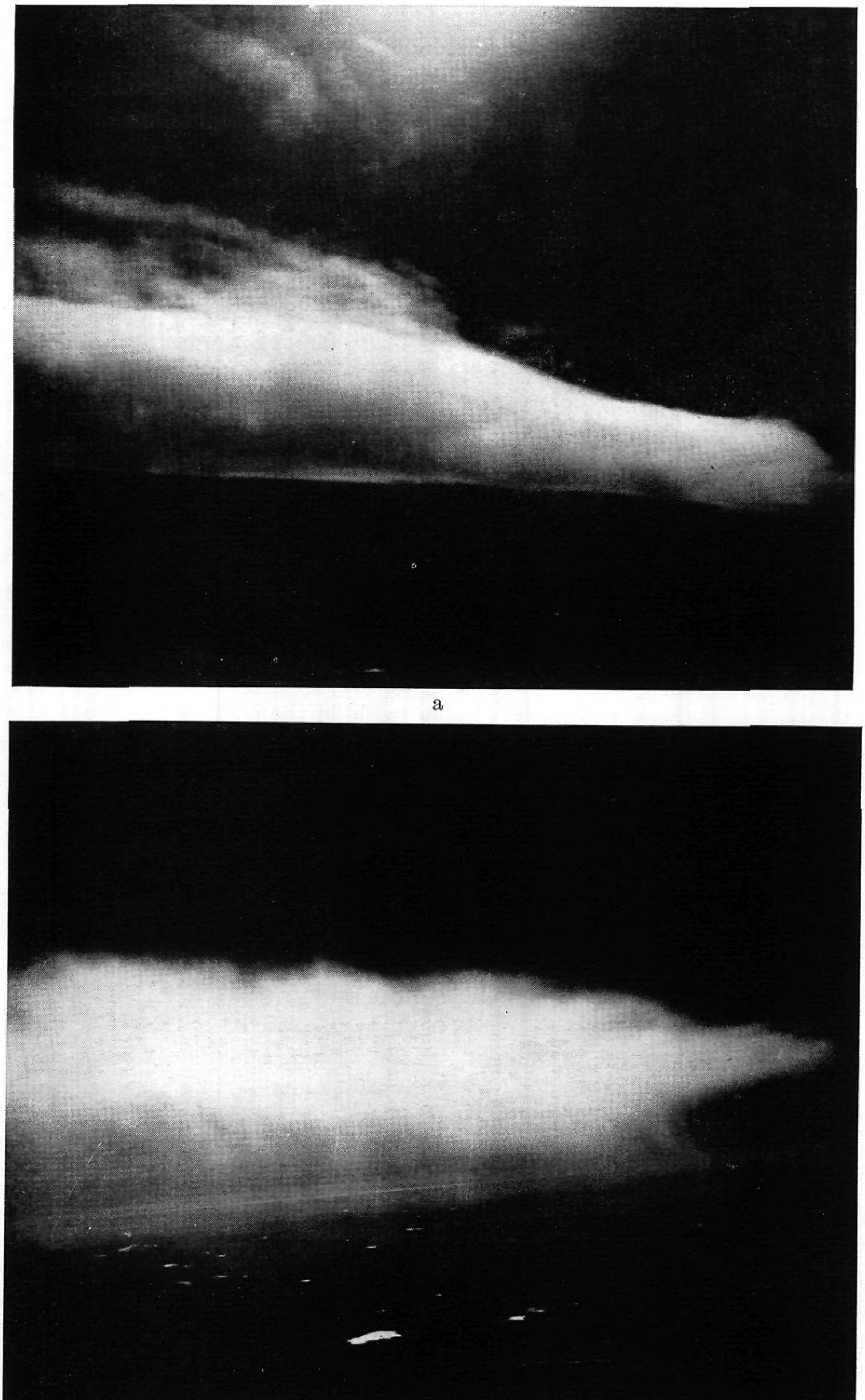
Other terms are being devised to describe some of the observable features peculiar to cloud modification studies. These will be described in greater detail in a later paper.

THE PRODUCTION OF SNOW IN SUPERCOOLED CLOUDS

As a result of the experimental studies made in the cold chamber during the summer of 1946 (12, 16), which has already been mentioned, the first experiment in the free atmosphere carried out by the writer took place on November 13, 1946. On that day a 4-mile supercooled alto stratus cloud having a temperature of -18.5°C. and containing no trace of ice crystals was "seeded" with 6 lb. of dry ice by flying through it in a small Fairchild monoplane at an altitude of 14,000 ft. Half of the fragments were put out from a mechanical dispenser operating on the floor of the plane; the rest were scattered by merely letting the slip stream suck the fragments from cardboard containers held out of the open window. Figure 20 shows the cloud before and after seeding.

Since that time many experiments have been carried out by the writer, by members of Project Cirrus—a joint Army–Navy–Air Forces–General Electric weather research program—, and by many other investigators. Some of these results may be studied by referring to detailed reports and papers (4, 14, 15, 18). Positive results are always observed when supercooled stratiform clouds are seeded with dry ice at the rate of about 1 lb. per mile. The magnitude of the effect is dependent on the stability of the air, the thickness of the cloud, and whether or not the air flow into the region is of a convergent nature. If convergence is present, it is quite possible that the "triggering effect" produced by seeding with dry ice would result in a self-perpetuating storm spreading out from the area originally modified. Similarly, cumulus clouds may be profoundly modified with dry ice whenever the cloud contains supercooled water droplets.

Laboratory experiments indicate that at least 10^{16} ice nuclei may be produced from 1 g. of dry ice when it is introduced into a supercooled cloud. If all of these particles could be supplied with moisture and allowed to grow to a small



a

b

Fig. 20. View of first experiment in natural clouds above Mt. Greylock, Massachusetts (a) before seeding; (b) 5 min. after seeding with dry ice.

snow crystal, some 300,000 tons of snow would result. It is, of course, impossible to achieve such results under natural conditions, since there is not enough moisture available in the area that can be affected. The important fact to recognize in this respect is that by using dry ice it is quite feasible to produce nuclei on the tremendous scale that is characteristic of natural weather phenomena. For example, the number of snow crystals required to produce 1 ft. of snow over an area of 10,000 square miles is approximately 1×10^{18}. That number of ice nuclei could easily be produced with 2 lb. of dry ice. Since it would not be feasible to spread 2 lb. of dry ice over such a large area, it would, however, be practical to use 200 lb. at the rate of $\frac{1}{2}$ lb. per mile and exert a large influence over an area of 10,000 square miles.

Under actual seeding operations, it has been found that about 1 lb. of granulated dry ice will convert 5 cubic miles of supercooled stratus clouds to snow in about 30 min. when the dry ice is introduced from a 2-in. pipe in a plane flying at the top of a cloud having a vertical thickness of a mile. Dry ice pellets in the size range 1–10 mm. in diameter are used so that the crystals are produced within a minute or so along a plane extending from top to bottom of the cloud. The heat generated in the crystallization process ranges from 0.5 to 1°C. in such a cloud, causing turbulence which spreads the crystals upward and outward. Thus it is a common observation to see the affected area expand as shown in figure 21 at a rate of nearly 5 miles an hour. Figure 22 is a photograph of such an area.

So many crystals are produced when dry ice is dispensed at the rate of 1 lb. per mile that the affected region often shows a persistent, stable snow crystal cloud remaining in the modified region after all of the adjoining clouds have disappeared by precipitation. Such an effect is recognized as "overseeding" but is only experienced with very stable clouds, thin clouds, or under conditions free of convergence.

Such effects would not be expected with cumulus clouds unless they were dissipating or when large quantities of dry ice are used.

Some evidence has been seen that extensive cumulus activity may be generated in thick and unstable stratiform cloud systems by certain seeding techniques. Studies are now under way by our group to explore these possibilities.

SOME POSSIBILITIES AND LIMITATIONS IN MODIFYING CLOUDS

There are, of course, definite limitations to the effects which may be expected when supercooled cloud systems are artificially modified with a material such as dry ice.

If any appreciable amounts of precipitation are to result from such activities, the atmospheric conditions must be such that it is likely that snow or rain would occur from natural processes. The method does, however, present the definite possibility of "triggering off" a storm sooner than it would develop if the natural sequence prevailed.

It must be remembered that there is actually only a relatively small amount of condensed water in a cloud. Thus, if all of the moisture in an ordinary stratus cloud having a vertical thickness of a mile were to be precipitated, only about 2 mm. of water would result. Towering cumulus of the congestus type may contain from 1 to 3 cm. of precipitable water, depending on vertical height and liquid water content.

While it is not possible to produce ice crystals in clouds using the dry ice method unless they are *cold* clouds, with temperatures colder than 0°C., it is obvious that if a *cool* cloud were modified in the supercooled region, the resulting

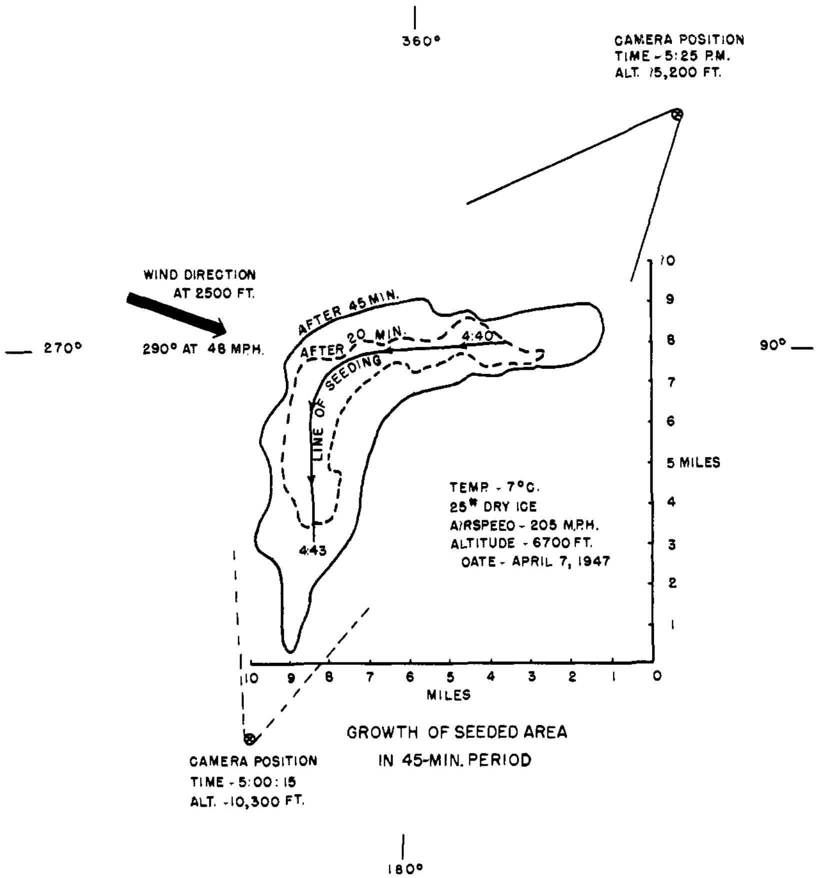


FIG. 21. Rate of growth of area in stratus cloud seeded by Project Cirrus

snow crystals would melt and become rain at a short distance below the freezing level, thus providing water droplets which would lead to the formation of rain. If Langmuir's chain reaction theory is correct, this effect would quickly lead to a rapid development of heavy rain in large cumulus clouds.

The tremendous quantities of ice crystals which may be produced with dry ice in cold clouds raises the distinct probability that, if correctly introduced, it should be possible to eliminate dangerous hailstorms. With a large concentration of sublimation nuclei present in the region where hail might form, the competition between particles for the available moisture would be so keen that no particles could grow large. It would, indeed, be foolish to think that such results could be obtained immediately. Much careful quantitative research experiments must be conducted to hope for eventual success in this field. In a similar manner, violent thunderstorms may eventually be subdued by preventing the development of any appreciable thickness of supercooled cloud in the cold portion of the cool cumulus congestus cloud.

In fact, any type of storm system which contains appreciable amounts of supercooled clouds may be profoundly modified by intelligent seeding with dry ice.

FIG. 22. Photograph of modified stratus cloud seeded at rate of 1 lb. per mile. The modified area which appears dark is 15 miles long and 2 miles wide.

Supercooled ground fogs may also be modified by following a proper seeding procedure. There is a likelihood of "overseeding" with ground fog which not only reduces visibility but also makes the fog much more persistent. Proper amounts used will, however, clear the fog quite rapidly.

There is also a real possibility that the aircraft icing problem may be affected to a considerable degree by modifying supercooled clouds in the vicinity of airports and along heavily travelled air lanes. There is also a distinct possibility that the local area ahead of a flying aircraft may be modified with projectiles carrying and dispensing seeding media, such as dry ice from liquid carbon dioxide. In this way, it may be possible that the icing hazard could be reduced or eliminated as the plane reached the treated area.

One of the immediate results which is proving to be of much value in the physical studies of clouds is the fact that the dry ice seeding process permits the marking of clouds in a unique way. By photographing the subsequent appearance of the modified cloud, many interesting features may be measured in a quantitative manner by special photographic techniques and procedures under development by our Project Cirrus operation.

The science of experimental meteorology is just beginning. The next few years will show rapid advances in this fascinating field. The results will surely aid meteorologists and others in reaching a better understanding and appreciation of weather phenomena.

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