Ice Nucleation in Clouds by Liquefied Propane Spray

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ABSTRACT

The inducement of cloud glaciation by cooling with evaporating droplets of liquefied propane was investigated in laboratory experiments and in field tests. The efficiency of ice crystal production was shown to be near 10^{12} crystals per gram of propane for temperatures colder than -2C; 10^{10} crystals were produced at supercoolings of few tenths of a degree. Observed variations of the efficiency with changes in cloud liquid water content and in wind velocity indicate that the rate of vapor supply to the cooled plume is the limiting factor in ice crystal production. Ice crystal habits and growth rates were also examined and inferences could be drawn regarding the nucleation mechanisms of the ice crystals in the cooled zone. The results indicate that liquefied propane is an effective, easy-to-use, and safe nucleating agent. The experiments helped delineate the factors which are to be considered in designing practical applications of the technique.

1. Introduction

The first instances of artificial production of ice crystals by Langmuir and Schaefer (Suits, 1962; Schaefer, 1946) were achieved by the introduction of objects at very low temperatures into moderately supercooled clouds. This discovery has led to the first documented modifications of natural cloud systems by the dropping of small pellets of dry ice into the clouds. Another method of producing very low temperatures in a cloud, to thereby create large numbers of ice crystals, was given by Serpolay (1954); this method rests on the large self-cooling that accompanies the rapid evaporation of highly volatile liquid droplets.

In the 25 years since the original discovery of nucleation by cooling, numerous field experiments have been carried out and several laboratory measurements were made to determine the effectiveness of the different coolants. Dry ice has been used the most extensively because of convenience in dispersing it into a cloud. The original experiments of Langmuir (1948) were followed by large numbers of attempts, notable being those by aufm Kampe *et al.* (1957) and Weickmann (1957). The quantitative aspects of ice crystal production by dry ice were examined by Eadie and Mee (1963), Chao (1964) and more recently by Fukuta *et al.* (1971) who found the efficiency of ice crystal production to be very near 10^{12} ice crystals per gram of CO_2 over a wide range of temperatures. Theoretical estimates of the expected ice crystal production by Langmuir (1948) and Gaivoronskii *et al.* (1968) are much higher, generally in the range of 10^{14} to 10^{17} per gram of dry ice. Reasons for the discrepancy between predicted and observed efficiencies were discussed by Fukuta (1965b). Recent field applications of dry ice seeding are those reported by Appleman (1968a,b), Kornienko *et al.* (1968) and Bigg and Meade (1971a,b).

The use of sprays of volatile liquids has not been as extensive as that of dry ice primarily because of problems created by the necessity of releasing the spray directly into the cloud to be modified. The dispersion of supercooled ground fogs has nonetheless been attempted by such methods as reported by Serpolay (1965), Hicks (1967) and Gerdel (1968). Some of the factors governing the effectiveness of evaporating spray droplets was discussed by Fukuta (1965a,b) who also explored practical means of utilizing these sprays for cloud seeding. Fukuta *et al.* (1970) reported the ice crystal production efficiencies of nine liquids at -10C. The highest numbers of crystals (10^{11}–10^{12} crystals per gram of liquid) were found for propane and for chlorodifluoromethane (Freon 22); substances having higher boiling points showed lower ice crystal production efficiencies. Andro and Serpolay (1970) have reported the results of laboratory determinations of the efficiencies of propane and carbon dioxide sprays. Over the temperature range -5 to -15C, they found propane to produce 5×10^9 crystals per gram of propane and CO_2 to be half as effective.

The high nucleating efficiency of propane, its ready availability, and ease of handling prompted a further investigation into its performance. The experiments to be described here were undertaken to determine the ice crystal production efficiency of propane spray under various conditions. Laboratory tests were first performed to investigate the nucleation efficiency of propane over a large range of temperatures with other variables held constant. Following this, a set of field tests were performed in which a quantitative evaluation of the seeding efficiency was obtained under natural conditions. The results of these tests, in good agreement with one another, have shown propane to produce nearly 10^{12} crystals per gram of propane, and that cloud seeding by this technique is both simple and safe. In many situations seeding with propane spray may offer advantages over nucleation by silver iodide smoke particles for induction of the formation of the ice phase.

2. Laboratory measurements

a. Test facilities

This phase of the project was conducted at the U. S. Army Cold Regions Research and Engineering Laboratory. A walk-in type refrigerated room 3.85 m long, 2.30 m wide and 3.63 m high (31.64 m^3 effective volume) was used as a cloud chamber. The inside walls and ceiling were painted black to improve the visual contrast. A low-pressure steam outlet was installed to provide the moisture needed for fog generation.

Cooling was accomplished by circulating a cold brine (-56C) through cooling cores located near the ceiling of the chamber. Thermostatically controlled blower and brine valves kept the chamber at any desired temperature down to a minimum of about -40C.

b. Instrumentation

Fog density in the cloud chamber was measured by a transmissometer having a path length of 33 cm. The tests were performed in dense fogs which had a visual range of about 1 m. Fog density was controlled by admitting more or less steam into the cloud chamber.

The primary temperature measurement was obtained from a fast-response thermocouple installed at a height of 185 cm above the floor. The output of this thermocouple was continuously recorded. Indications of vertical temperature gradients were obtained from six thermocouples mounted 38 cm away from the center of one wall at heights of 60, 125, 188, 246, 309 and 334 cm above the floor. The outputs of these sensors were also recorded. In addition, frequent checks of temperature were made with a direct-reading thermistor system and a liquid-in-glass thermometer.

c. Propane dispenser

The dispenser used for the liquefied propane consisted essentially of a glass tube with a small toggle valve at its bottom. The glass tube was filled from an inverted propane cylinder after which the valve separating the tube from the cylinder was closed. The propane was released by depressing the toggle valve for an instant. The amount of propane dispensed could be determined from the change in the height of the liquid column by taking into account the density of propane at the ambient temperature. Generally, $\sim$0.1 gm of propane was used for each test.

d. Ice crystal collection and evaluation

Ice crystal concentrations in the cold room were determined by replication of ice crystals from a known volume of air. This was achieved by placing a microscope slide, previously coated with Formvar and allowed to dry, on a horizontal surface and then covering it with a cylinder of 16.3 cm height closed at its upper end. Crystals were thus deposited onto the slide from an air column of 16.3 cm in height. The slide was withdrawn from the cylinder after 3 min and exposed in a closed container to chloroform vapor for approximately 20 sec. The replicas so obtained were then examined under a phase-contrast optical microscope.

e. Procedures

The typical sequence of operations for a test was as follows:

1) The cloud chamber was cooled to several degrees below the desired temperature.

2) Steam was introduced into the room until the desired temperature was reached.

3) The fog was allowed to decay naturally until a meteorological range of approximately 1 m was indicated on the transmissometer.

4) Propane was released and the air fanned with a piece of cardboard to distribute the ice embryos throughout the cloud chamber.

5) When all the fog droplets had disappeared and only ice crystals were visible, the microscope slide was exposed.

With this procedure it was found to be somewhat difficult to simultaneously control the temperature and the desired fog density. This was due to temperature changes that accompanied the introduction of additional steam into the cold room. Even so, it was found possible to achieve near constancy of the temperature and to determine this temperature to better than 0.5C accuracy.

f. Results

The concentration of ice crystals in the cold room was determined from the crystal replicas and was assumed to be constant throughout the cold room. The efficiency of crystal production was then derived by calculating the total numbers of crystals produced and taking into account the amount of propane used in the particular test.

The numbers of crystals produced per gram of propane at the various test temperatures are shown in Fig. 1. It is seen that the production rate is fairly constant near 4×10^{11} for temperatures colder than

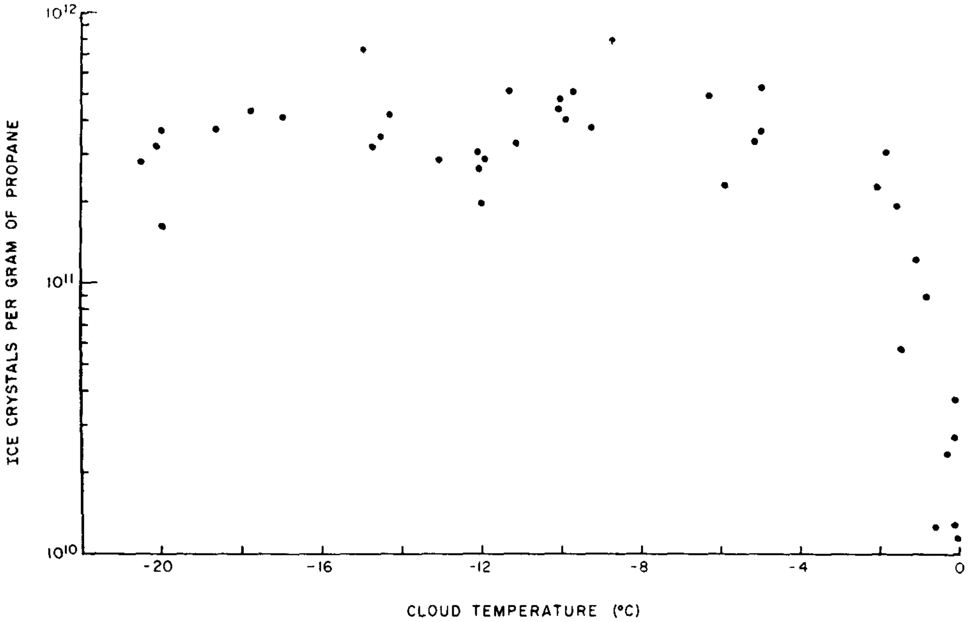

FIG. 1. Ice crystal production efficiency of liquefied propane in cloud chamber tests.

−2C. Crystal production drops off quite abruptly at temperatures warmer than −2C with approximately 10^{10} gm^{-1} produced within a few tenths of a degree of 0C. At the warmer temperatures some liquid fog persisted in the cold room in addition to the crystals.

It should be noted that the temperatures quoted above and used in Fig. 1 are those of the environment in which the ice embryos grew; the temperatures at which nucleation occurred in the plume of evaporating propane droplets were very much colder.

3. Field experiments

a. Site of field experiments

Field testing of the nucleating efficiency of liquefied propane was carried out at the Elk Mountain Laboratory of the University of Wyoming. This Laboratory is located near the peak of Elk Mountain at an elevation of 3350 m MSL. From the direction of the prevailing winds (west) Elk Mountain has a sharply rising slope with the Laboratory located 200 m beyond (to the east) and 50 m below the crest of the mountain. Orographically induced clouds frequently form around the peak of the mountain with the base of the cloud typically 600 m below the level of the Laboratory.

The propane seeding experiments were carried out by releasing propane at the crest of the mountain and by monitoring the seeding effects at the Laboratory. The position of the seeding generator was selected in accordance with prevailing winds so that the plume would be carried directly toward the Laboratory. In the direct vicinity of the release points the terrain was in all cases free of obstructions and was snow covered. Wind at the release point was typically strong and gusty; at the Laboratory the winds were considerably lighter. Because of the irregular terrain, the airflow over the crest and in the area around the Laboratory is quite turbulent resulting in rapid broadening of the seeded plumes. This was occasionally made evident by the release of colored smoke plumes prior to seeding.

Temperature and wind conditions both at the crest of the mountain and at the Laboratory were routinely measured during the experiments. The size distribution of cloud droplets was determined in a few cases using a portable impactor device in which the droplets impinge on a soot-coated slide at a known air velocity. These measurements revealed the drop spectra to have peaks at around 10 μm diameter with the droplet concentrations typically being 200–400 cm^{-3}.

b. Techniques

Difficulties are usually encountered in field evaluations of cloud seeding systems due to the need to define the volume of air affected by the seeding agent and the distribution of products within the seeded plume. In the experiments with propane these problems were circumvented by the use of a tracer material which was integrally mixed with the liquid propane. Particles of doped ZnS were mixed into the propane and released together with it, so that the relative release rates of seeding agent and tracer were predetermined by the relative amounts of the two materials used in the generator.

The experiments thus consisted of releases of propane and tracer, with a mass output ratio of R, and of determinations of the time-averaged concentrations of

TABLE 1. Summary of data for field experiments. See text for explanation of symbols.

Test no.†	Date	Release time	Temperature (°C)	Wind (m sec^{-1})	LWC (gm m^{-3})	R	Time of tracer sample	K (m^{-3})	C (m^{-3})	N (crystals gm^{-1})
1	Mar 1/72	1000–1004	−16.3	5	0.1	1.02×10^{-2}	1000–1007	230	7.0×10^{5}	1.5×10^{11}
2	Mar 1/72	1346–1351	−14.7	5	0.1	1.02×10^{-2}	1347–1354	290	1.4×10^{6}	2.4×10^{11}
3	Mar 2/72	0927–0932	−9.6	15G25	0.3	1.02×10^{-2}	0928–0934	190	1.7×10^{6}	4.5×10^{11}
4	Mar 2/72	1324–1335	−8.1	20G30	0.5	1.02×10^{-2}	1324–1347	130*	5.0×10^{6}	1.9×10^{12}
5	Mar 2/72	1336–1347	−8.1	20G30	0.5	1.02×10^{-2}			2.0×10^{7}	5.3×10^{12}
6	Mar 2/72	1602–1607	−7.0	20G35	0.6	1.02×10^{-2}	1602–1607	190*	1.2×10^{7}	4.3×10^{12}**
7	Mar 2/72	1617–1621	−7.0	20G35	0.6	1.02×10^{-2}	1617–1621		2.3×10^{6}	
8	Mar 3/72	0828–0835	−6.8	15G30	0.4	1.02×10^{-2}	0829–0837	17	$<10^{4}$	$<3\times10^{10}$
9	Mar 3/72	0900–0907	−7.3	15G30	0.4	1.02×10^{-2}	0900–0907	220	$<10^{4}$	$<2\times10^{9}$
10	Mar 20/72	2023–2030	−6.2	10	0.1	2.70×10^{-2}	2025–2030	400	1.7×10^{6}	5.7×10^{11}

† The same release point was used for all tests, except test 7.
* Adjusted in proportion to release rate.
** Assuming same plume diffusion as test 5 and adjusted for release rate.

ice crystals, C, and of tracer particles, K, at some point in the plume. The rate of ice crystal production per unit mass of propane was computed from

$$N = nR\frac{C}{K}, \tag{1}$$

where n denotes the number of particles per unit mass of tracer (i.e., the inverse of the average mass of the tracer particles). This equation applies regardless of the location of the observation point relative to the seeded plume provided that the diffusion of the tracer particles and of the generated ice crystals is similar. This assumption of plume similarity is believed to be reasonably correct for situations where ice crystals remain small, so that no differential sorting due to fallout occurs. The median crystal sizes in these experiments were around 50 μm corresponding to fall velocities of about 10 cm sec^{-1}. The travel time of the plume from the release point to the observation point was about 1 min so that the separation between the center of mass of the ice crystal plume and the tracer plume was less than 10 m at the point of observation; this is considered to be insignificant considering that the plume was quite turbulent with its depth estimated to be roughly 100 m.

Ice crystal concentrations were again determined by replication. Microscope slides coated with 1% solutions of Formvar in dichloroethane were waved through the cloud by hand. The velocity of motion of the slide was approximately 1 m sec^{-1} and the plane of motion was roughly at right angles to the wind direction. From five to ten back-and-forth sweeps were taken to minimize the velocity variation due to wind. The collection efficiencies for ice crystals between 20 and 80 μm in size vary quite rapidly with velocity in the neighborhood of a few meters per second according to Ranz and Wong (1952), and since the velocity of the slides was not accurately controlled no attempts were made to correct the observed ice crystal distributions for collection efficiency. This leads to an underestimation of the seeding efficiency by perhaps as much as a factor of 2.

The concentrations of ZnS tracer particles at the observation point were determined by capturing tracer particles on a filter from a known volume of air and determining the numbers of particles on the filter by counting under a microscope with UV illumination. Ideally, the duration of filter sampling in each test would have matched the time of passage of the seeded pulse at the observation point. Shorter sampling times would reduce the statistical validity of the particle counts obtained and longer times would lead to underestimates of the actual concentration due to dilution by particle-free air. In practice, the filter samples were taken simultaneously with the release of propane or in some cases the sampling time was shifted by 1–2 min with respect to the release times. Due to the uncertainties involved in selecting the appropriate sampling period the particle concentrations are considered to be accurate only within ±20%. As Eq. (1) indicates, this error is transferred directly to the calculated seeding efficiency.

The liquid propane was dispensed from standard 20-lb containers. These were fitted with a short piece of pipe and a simple nozzle having a 1.5-mm orifice. Inside the containers the propane outlet was fitted with a short pipe so that material would not be taken from the very bottom of the container when it is operated in an inverted position. The tanks were loaded with somewhat more than 7 kg of propane to which from 1–3%, by weight, of ZnS particles were added. It was established in preliminary experiments that the ZnS is practically insoluble in propane and that the particle size distribution of the ZnS was not affected. However, since the particles settled rapidly in the propane, continuous mixing during discharge was necessary. This was accomplished by mounting the propane tanks in "tumbling racks" so that the tanks could be rocked back and forth prior to and during the release of the C_3H_8-ZnS mixture. Several steel balls were placed inside the containers to increase the agitation. The ZnS used was a commercial product; the powder was used as received. Particle size analyses of both the powder and of the

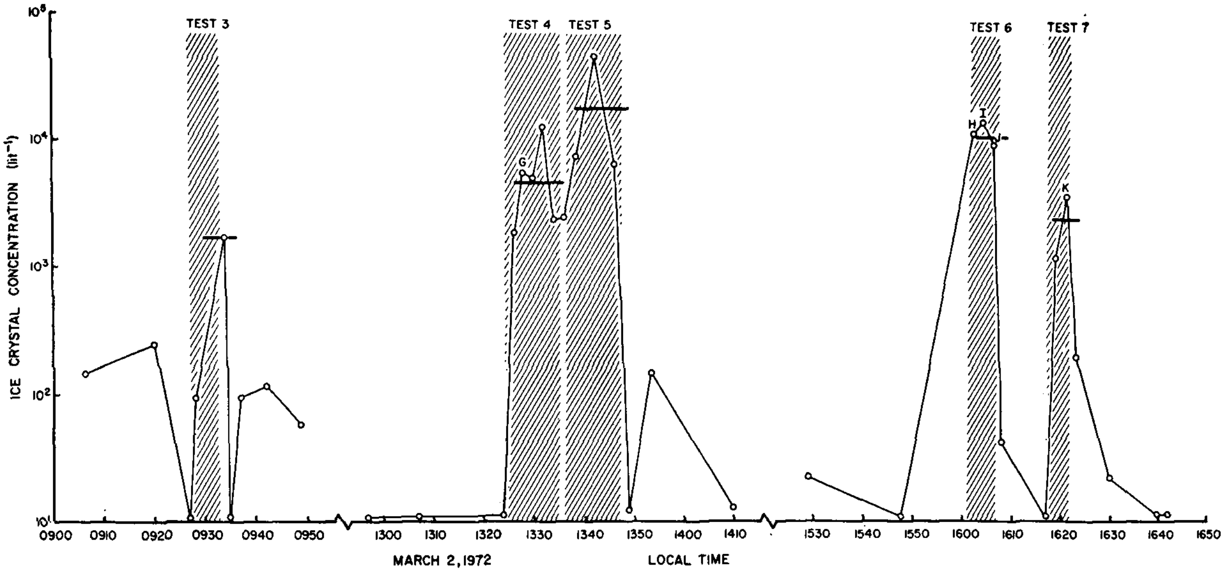

Fig. 2. Variation of ice crystal concentration with time during Tests 3 to 7. The horizontal heavy lines indicate the average values for given tests.

aerosol produced in the propane spray yielded $n = 5\times10^9 \pm 20\%$ particles per gram of ZnS.

The propane releases were made by fully opening the valve on the tanks and by allowing the liquid to escape through the nozzle under the internal pressure of the propane. Since the vapor pressure of propane is a function of temperature the release rates varied from case to case. The actual release rates were determined by noting the time necessary to empty the entire contents of the tanks. Typically, this time was 10 min which corresponds to release rates in the vicinity of 700 gm min^{-1}. (In one instance an anomalously slow discharge rate was observed for unknown reasons.) It may be noted that the actual release rate was not of critical importance, since, as shown by (1), only the propane-to-tracer ratio enters into the calculations and this ratio was determined by the relative amounts of material loaded into the tanks.

c. *Results*

A total of 10 separate tests was performed over four days. Temperatures and other environmental conditions varied over considerable ranges during the tests and the effect of these variables can be detected in the results. Table 1 lists the relevant data.

The measured ice crystal concentrations at the observation point are shown for several tests in Fig. 2. Each point in this figure was derived from one crystal replica slide on which 10–20 randomly selected areas of 1.7 mm^2 each were examined to obtain the numbers of ice crystals replicated. For the highest concentrations this corresponds to several thousand crystals being counted; the lowest concentrations correspond from 1 to 10 counted crystals. As shown by Fig. 2, the propane releases have produced easily recognizable peaks in ice crystal concentration, well above the background. The ice crystal concentrations given in Table 1 are averages computed from the measurements obtained during particular tests with a 1-min time lag allowed for the travel of ice crystals from the generator to the observation point. The averaging of ice crystal concentrations over several minutes was necessary, not only for smoothing the data but because of the need to get compatible data with the tracer particle concentrations which were determined with a long-term collection to which averaging is inherent. As the typical data shown in Fig. 2 reveal, although there was appreciable scatter in the individual measurements, the averages are still quite meaningful and the differences between tests appear significant. The use of time averages for the concentrations of ice crystals and tracer particles does not mean that the plume is assumed to be in a steady state; short-term variations do not invalidate the data as long as the fluctuations in C and K are correlated. Of some concern was the sufficiency of the sampling frequency for ice crystals. While a detailed analysis of this problem is not possible, consistency in the data for cases where sampling frequency was high indicates that the problem is not serious.

The efficiency of ice crystal production deduced by (1) is plotted in Fig. 3 as a function of environmental temperature, each data point being designated by the appropriate test number. Tests 8 and 9 failed to produce detectable ice crystal concentrations at the observation point; the points in Fig. 3 for these tests designate the detection limits only. No definite explanation has been found for the apparent failures in these two tests. The other seven measurement points in Fig. 3 indicate

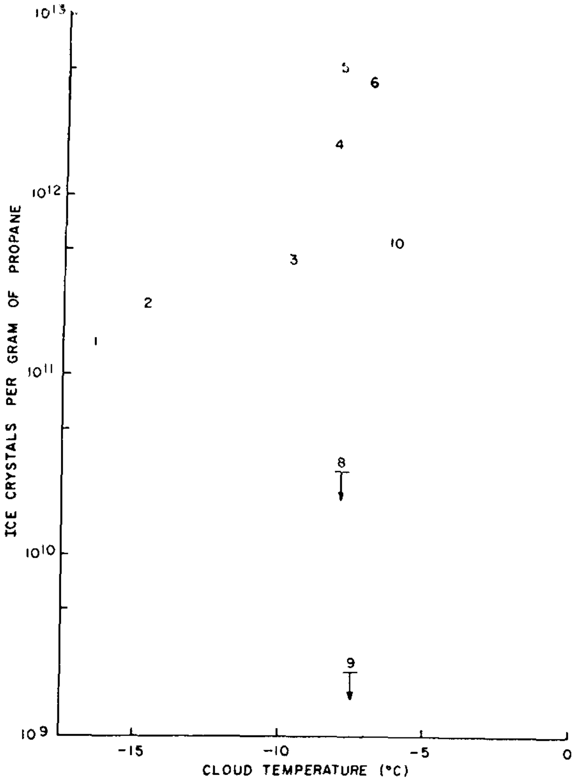

FIG. 3. Ice crystal production efficiency of propane in field tests. Numerals designating points in graph refer to test numbers. For Tests 8 and 9 only the detection limits are indicated.

efficiencies in the vicinity of 10^{12} crystals per gram of propane over the temperature range −15 to −6C. There is an apparent increase in efficiency with warming temperature but this is believed to be caused in reality by changes in the liquid water content of the cloud and in wind velocities.

The liquid water content of the cloud was deduced from the observed thickness of the cloud layer below the Laboratory (assuming adiabatic liquid water content) and from the visibility at the Laboratory. The estimated values are given in Table 1 and the observed ice crystal production efficiencies are related to liquid water content in Fig. 4. Data points in this figure are designated by the test numbers and the wind velocity at the release point is indicated adjacent to the test numbers. As can be seen, higher liquid water contents happened to exist when wind velocities were also higher so that it is not possible to separate the effects of these two variables. Both the higher liquid water contents and the higher wind velocities can be expected to lead to greater ice crystal production since increases in these variables result in greater rates of vapor supply to the cooled plume; insufficient supply of vapor in the plume limits the nucleation of new crystals because of competition between crystal embryos. This tendency is borne out by the findings presented in Fig. 4.

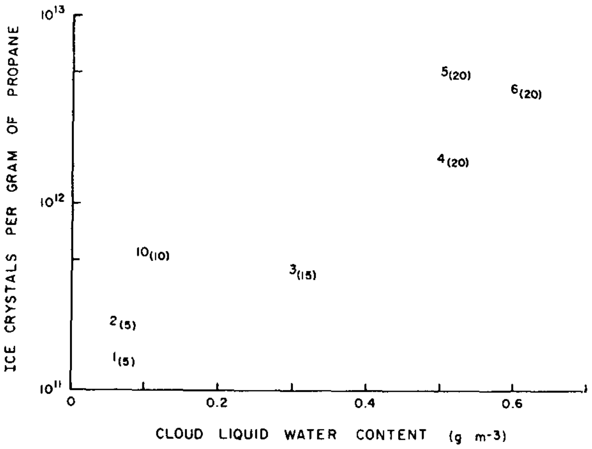

FIG. 4. Variation of the production efficiency of propane with cloud liquid water content. Each point in the graph is designated by the test number with the wind velocity (m sec^{-1}) given in parentheses.

4. Ice crystal morphology

The morphology of the ice crystals produced by propane seeding was studied in order to gather indications regarding the growth environments of the crystals. Fig. 5 shows the variations with temperature in the proportions of different crystal types which were obtained in the laboratory experiments. Here the crystals developed in an environment saturated with respect to water, as some fog persisted in the chamber throughout the growth periods of the crystals. Nonetheless, local regions of lower vapor pressure may have existed transiently.

Fig. 6 illustrates the proportions of various crystal shapes which were observed in the field experiments at different temperatures and Fig. 7 shows typical examples of these crystals. Comparison of Fig. 5 with Fig. 6 indicates that particular crystal types occurred over similar temperature regimes in both the laboratory and in the field tests but the agreement is not complete. Particularly conspicuous is the absence of stellar

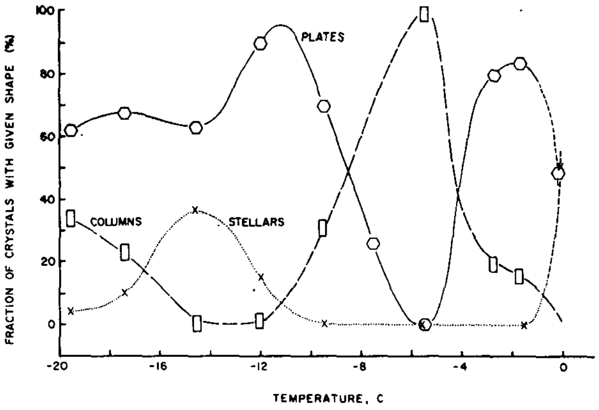

FIG. 5. Proportions of various crystal types produced by propane seeding at different test temperatures in cloud chamber.

crystals in the field experiments at −15C. Also, columnar crystals were found in the laboratory test between −8 and −12C whereas none were observed in the field tests for that temperature regime.

It is interesting to compare the crystal habits observed in the field tests with the results of investigations on the conditions governing the formation of crystal habits. For example, the crystals observed at −16.3C (Test 1) were mostly hollow plates (Fig. 7a) whereas dendritic crystals were observed at this temperature by Mason (1971) and Magono and Lee (1966) in conditions somewhat above water saturation. On the other hand, Rottner (1971) found hollow plates growing in laboratory experiments at −16C and at $S_w=0.94$ ($S_w=1$ being water saturation), and Auer (1970) also detected crystals similar to those in Fig. 7a close to the point of generation of crystals by dry ice in a natural cloud. Some columns were also present in Test 1; Rottner (1971) found that columns grow at −16C if $S_w=0.87$ which further agrees with the indication given by Magono and Lee (1966). Thus, it appears from the observed crystal types that the saturation ratio in the seeded plume was depressed below water saturation between the release and observation points. This inference is supported by the lack of cloud droplets on the corresponding replicas (Fig. 7a) and is made plausible by the low value of the liquid water content of the cloud (<1 gm m^{-3}).

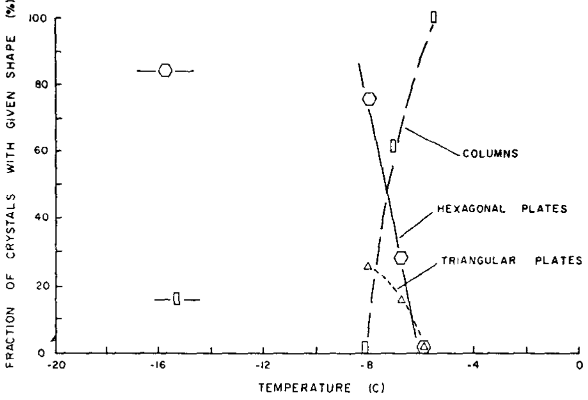

Fig. 6. Proportions of various crystal types produced by propane seeding at different test temperatures in field tests.

In contrast with the above situation, the presence of

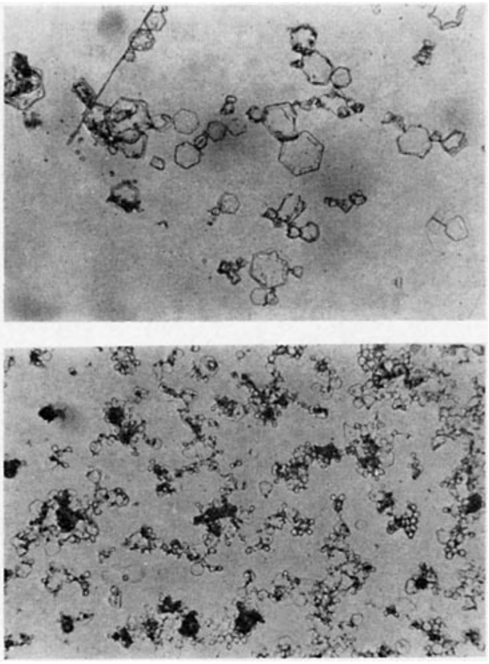
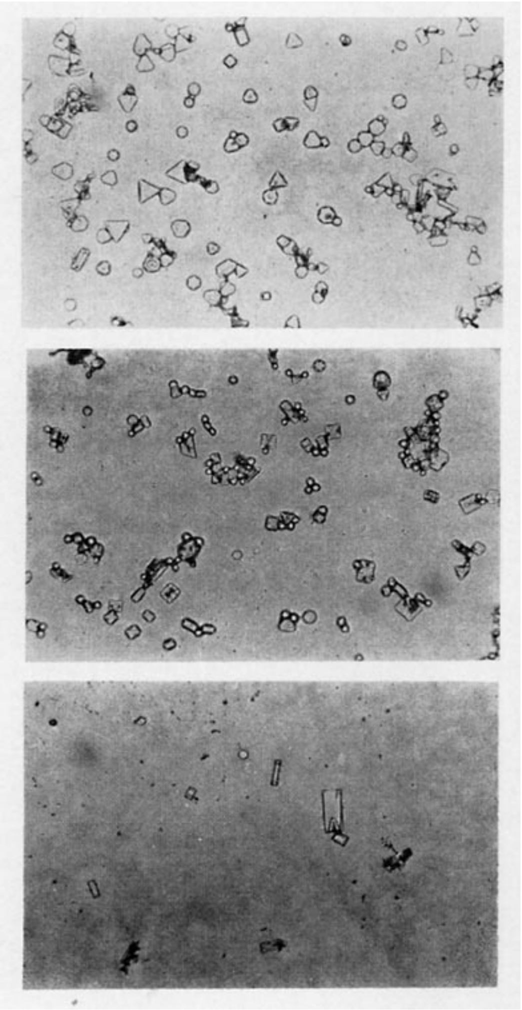

Fig. 7. Examples of crystals replicated in field tests. From top to bottom: (a) Test 1, −16.3C; (b) Test 4, −8.1C; (c) Test 5, −8.1C; (d) Test 6, −7.0C; (e) Test 10, −6.2C. Viewed areas are 0.88 mm in width.

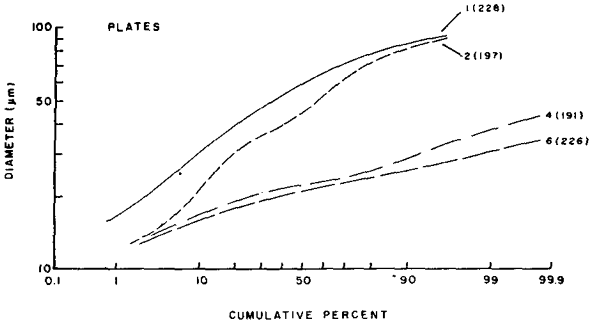

FIG. 8. Size distributions of plate crystals in different tests. Each curve is labeled by the test number and the number in parentheses indicates the number of crystals used for evaluation.

numerous droplets on the replicas from Tests 4, 5 and 6 indicates that these crystals grew in saturated conditions with respect to water (see Figs. 7b and d). The types of crystals observed here correlate well with the observation of Mason and of Magono and Lee for saturation. The cloud liquid water contents for these cases were considerably higher than for Test 1 (cf. Table 1). Only one of the replicas, Fig. 7c, from Tests 4–6 showed an absence of cloud droplets, this being the one taken at 1343 in the highest concentration of ice crystals observed throughout the experiment (cf. Fig. 4). The indication is that even with a liquid water content of about 0.5 gm m^{-3} there were pockets within the seeded plume where cloud droplets had completely evaporated.

It can thus be concluded that under the conditions of the field tests and with the release rates of propane that were used, the vapor pressure in the seeded plume was below water saturation in clouds of 0.1–0.2 gm m^{-3} water content and in portions of plumes in clouds of about 0.5 gm m^{-3} water content. This is yet another manifestation of the dependence of seeding effect on the vapor supply. In fact, the observations make it most plausible that in the direct vicinity of the propane spray nozzle the saturation ratio did not exceed unity with respect to ice except at the point of nucleation. It is even possible that in some interior pockets of air the vapor pressure fell below ice saturation leading to evaporation of some crystals; this can arise as a pocket of air which was cooled by the evaporating propane begins to warm but surrounding crystals stop the flow of water vapor toward it. (In any event, evaporation could have occurred only in very limited regions at the start of the plume as no evidence of sublimed crystals was found on the replicas.)

5. Ice crystal growth rates

For practical applications of propane seeding, it is of importance to determine the rate at which the produced ice crystals develop. Such information is necessary for proper targeting of seeding effects.

Measurements of the crystal dimensions were taken on the replicas and, thus, the data given here represent the growth of the crystals over the travel between the release and observation points. The mean time of travel of the plume can be estimated to be 40 sec for the lowest and 10 sec for the higher wind velocities experienced. The size distributions of the replicated crystals are shown in Figs. 8 and 9. A comparison of these data with Table 1 shows that generally larger crystals were observed for experiments with lower wind velocities, as is to be expected. The variations in the median crystal sizes from test to test are between factors of 2 and 2.5 which are smaller than the ratio of the mean travel times. This is reasonable since the rate of development of the linear dimensions of crystals is not directly proportional to growth time. Noteworthy is the fact that the crystal sizes showed considerable dispersion. The 10- and 90-percentile values for the distributions had ratios as high as 3.5 for the plates in Tests 1 and 2; the lowest ratio was 1.5 for the plates in Tests 4 and 6. The greater dispersion for Tests 1 and 2 is probably related to the longer travel times in those tests. The dispersion as a whole is a result of inhomogeneities of vapor pressure within the plume and the turbulent motions of the plume leading to variations in travel times. The magnitude of the dispersion was somewhat surprising in view of the short travel distance between release and observation points.

From the above data and the material presented in Section 3, the role of wind in determining the seeding effect can be seen to be twofold: it affects the vapor supply to the cooled region and it influences crystal sizes at some given distance away from the seeding point. These two effects can nearly balance out, as a calculation of the total flux of ice crystal mass can show. The distribution of crystal masses for Test 10 is shown in Fig. 10 indicating that the median crystal mass was 1.5×10^{-8} gm. Crystal masses were readily calculated from the observed diameters and lengths of the column crystals. Since no direct measurement of the thickness was available for plate crystals, their masses were calculated from the mass-diameter relations of Auer and Veal (1970) and the well-known relation between mass median diameter and median diameter for log-normal size distributions (Herdan, 1960). The mass calculations show that the total flux of ice which passed

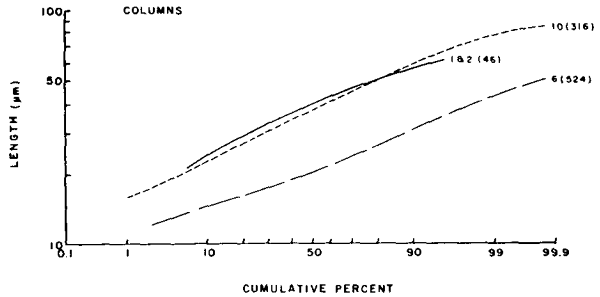

FIG. 9. As in Fig. 8 except for columnar crystals.

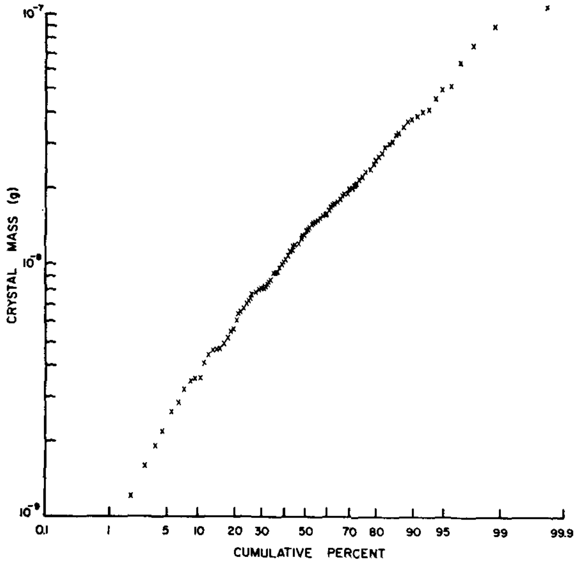

Fig. 10. Cumulative size distribution of crystal masses for columnar crystals produced in Test 10, based on measurements of 316 crystals.

the observation point varied between 1.5×10^3 and 9×10^3 grams of ice per gram of propane released. The lowest value was obtained for Test 1 while the highest one was for Test 6. It appears that the high liquid water content in Test 6, as compared to Test 1, contributed substantially to the mass of ice formed; on the whole, however, the relative constancy of the mass values showed the compensating effects of higher numbers of crystals nucleated and smaller sizes achieved for greater wind velocities.

The growth times of the observed crystals can also be estimated from growth rate calculations by Koenig (1971), Jayaweera (1971) and Hindman and Johnson (1972). It was found, assuming water saturated conditions, that the calculated crystal growth times are less than 1 min for the median crystal sizes in agreement with the estimated travel times. The agreement with the experimental results of Fukuta (1969) is also good. Detailed calculations of the crystal growth times were not performed since, for the small crystal sizes here dealt with, the calculations are very sensitive to the assumed initial size of the crystal and require knowledge of the ambient vapor pressure.

6. Summary

Laboratory tests at various temperatures but with constant cloud densities have yielded ice crystal production efficiencies between 2×10^{11} and 6×10^{11} crystals per gram of propane. Above -2C the efficiency decreases rapidly but is still 10^{10} gm^{-1} within a few tenths of a degree of 0C.

The results of the field tests showed more variability, the nucleating efficiency varying between 10^{11} and 5×10^{12} gm^{-1} in apparent response to the combined effects of cloud density and wind velocity. Since these factors influence the rate of vapor supply of the seeded plume, this rate appears to be a limiting factor for the numbers of crystals being produced. On the whole, the agreement between laboratory and field tests is quite satisfactory so that the extension of the field results, on the basis of the laboratory tests, to temperatures not covered in the field tests appears to be justified. Thus, it can be stated that the liquefied propane will produce 10^{12} crystals per gram of propane under average conditions and at temperatures colder than -2C. Particular circumstances may lead to approximately tenfold increases or decreases in efficiency. This result is in good agreement with the findings of Fukuta *et al.* (1970) but is at variance, for unknown reasons, with the results of Andro and Serpolay (1970).

The influence of the discharge rate of propane on the efficiency has not been determined in the present tests. It can be anticipated that the optimum discharge rate and spray pattern will be that which best compromises the increased cooling effect accompanying high propane droplet concentrations with the decreased vapor supply of a small plume.

The shapes of ice crystals produced and their growth rates indicated that the vapor pressure within the plume was frequently depressed below water saturation. These observations further confirm the critical role of vapor supply in determining the seeding effect in terms of the numbers of crystals produced and the sizes reached by the crystals over a given path length.

The experiments also provided some information on the nucleation mechanisms involved. The process of nucleation is of great theoretical interest but it need not be considered in detail here. Ultimately the nucleation and growth processes will have to be examined so that the dependence of the production rate on environmental conditions may be elucidated; the use of the propane system may then be accompanied by quantitative predictions of the outcome.

The experiments described in this paper have again confirmed the possible utility of liquefied propane as a cloud seeding agent. Its advantages over other materials are seen in that: 1) it is effective at near-zero temperatures; 2) it is readily available, inexpensive, easily stored and transported; and 3) it is not considered to be detrimental to the environment since it diffuses readily and does not react harmfully with other components of the atmosphere (Carroll, 1970).

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