Observations of Ice Crystal Nucleation by Droplet Freezing in Natural Clouds

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1. Introduction

During the autumn of 1967, a close scrutiny of the ice crystal climatology within the orographic cap cloud was initiated in an attempt to isolate some features of the natural precipitation process. The isolated cap cloud was selected for study because it is perhaps the simplest of the orographic clouds and, therefore, presents an opportunity for investigating some of the important nucleation processes.

Early studies of ice crystal shapes by Nakaya *et al.* (1936) and Nakaya (1954) described observations of dendritic crystals of double forms, often exhibiting an asymmetric structure. Weickmann (1947) confirmed and expanded the 1936 observations of Nakaya *et al.* by noting that the components of a single asymmetric crystal indeed belong to one crystal but were growing in different planes. Furthermore, Weickmann (1947) and Weickmann *et al.* (1970) noted that in each case these "double crystals" possessed a circle in their center resembling a cloud droplet; he therefore premised that small droplets, upon freezing, could become tiny monocrystalline prisms whose two base planes then grow into a hexagonal or dendritic crystal form.

In the same paper Weickmann *et al.* reported on cloud chamber nucleation activities and again found ice crystals exhibiting this characteristic asymmetric or double feature, suggesting that the silver iodide nucleant used in these studies was presumably generating these ice crystals by the droplet freezing mechanism.

Expanding on the implications of these findings, we observed the depositional and droplet freezing modes of nucleation *within* orographic cap clouds during natural and seeded conditions. This paper will discuss the results of these observations and suggest possible implications concerning the various modes (depositional or droplet freezing) of nucleation.

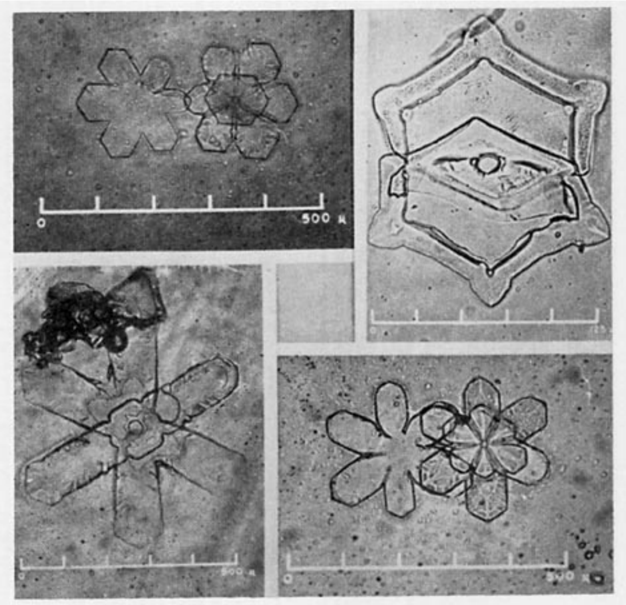

FIG. 1. Examples of representative ice crystal replicas with and without droplet centers and accompanying asymmetric or double structures.

2. Procedures

Ice crystals growing within the cap cloud were captured and replicated on Formvar-coated slides at very slow impact speeds. The replicas were then inspected under a microscope to determine the crystal type (Magono and Lee, 1966), to measure the dimensions of the double crystal asymmetries, if any, and to check for the presence of the circular (droplet) center. Magnifications of 100–400× were required to insure sufficient resolutions since the circular center cannot be precisely determined with lower magnifications.

The crystal types studied were limited to hexagonal plates, thick plates, stellars and dendrites since the structural patterns of interest are most easily ascertained from such crystal types. Therefore, the findings in this note are applicable only over the temperature range corresponding to the temperature habitat of these crystals; namely, −8 to −25C as found in this study and also suggested by Magono and Lee.

3. Results

Fig. 1 illustrates some representative crystal replicas with circular (droplet) centers and corresponding double or asymmetric structures found in the untreated orographic cap cloud; note how clearly the crystals with circular (droplet) centers can be delineated from their neighbors without such a structure. The circular (droplet) center is always accompanied by the double or asymmetric structure for plate family crystals.

Assuming an ice crystal cross-sectional structure similar to that shown in Fig. 2a, the relationship shown in Fig. 2b was generated from observations of the circle (droplet) diameter d within the observed ice crystals and the separation h (including crystal thicknesses) between the double or asymmetrical crystal faces. The proximity of the plotted data points to the $d=h$ line in Fig. 2b suggests a nearly spherical shape of the center of the crystal.

Fig. 3 is a comparison between the average observed cloud droplet spectrum for Elk Mountain cap clouds and the observed circle (droplet) center spectrum from double or asymmetric ice crystals found within the same clouds. It can be seen from Fig. 3 that the population of droplets serving as crystal centers (mean diameter 11.7 μ) lies within the observed spectrum of cloud droplet diameters (mean 8.6 μ).

An inspection, then, of Figs. 1–3 indicates that the circular centers of the double or asymmetric ice crystals possess spherical shape and arise from the population of cloud droplets commonly observed within the cap cloud. These observations, when combined with evidence for single crystalline structure of frozen cloud droplets (Weickmann *et al.*), clearly imply that ice crystals having a center circle and possessing double or asymmetric structures have their genesis from frozen cloud droplets.

Since identification of ice crystals originating from frozen cloud droplets is now possible, it was decided to investigate the number of these ice crystals occurring in natural clouds. This enumeration consisted of determining the total concentrations per unit volume of *cloud*

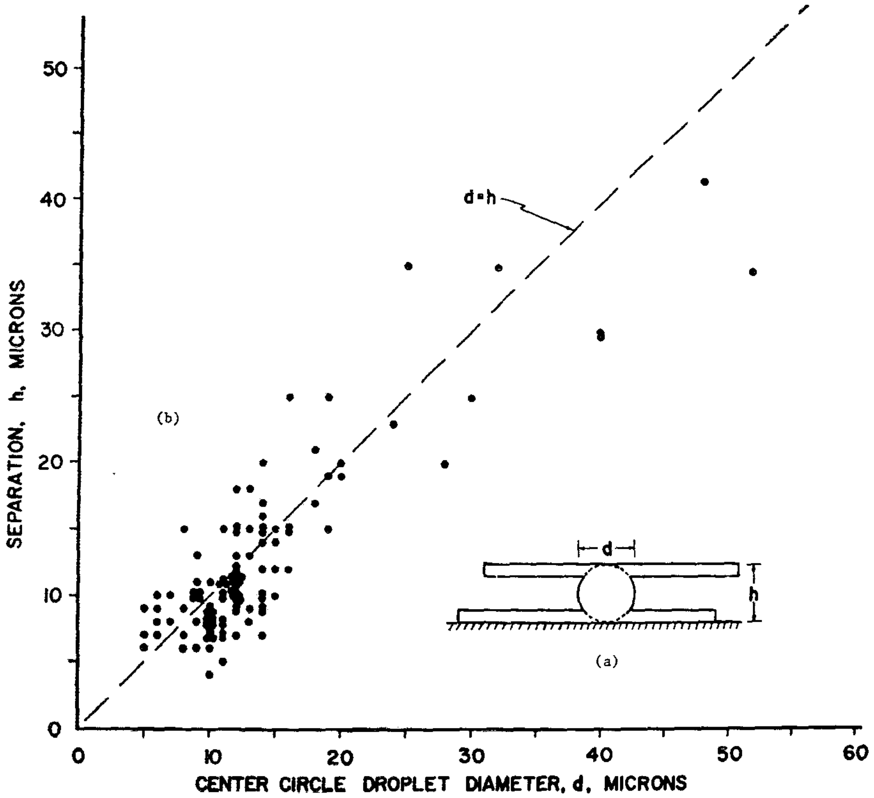

FIG. 2. Assumed cross-sectional structure of a double or asymmetrical crystal, (a), and the relationship between the circle (droplet) diameter *d* and the separation *h* (including crystal thickness) between the double or asymmetrical crystal faces, (b).

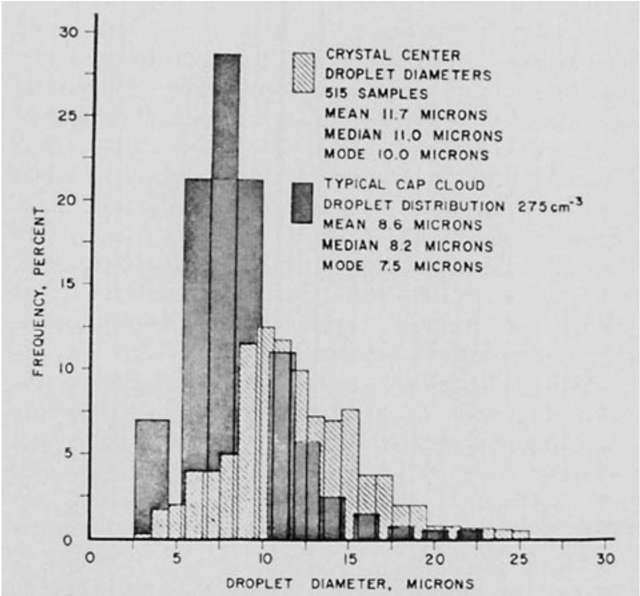

FIG. 3. Comparison between the average observed cloud droplet spectrum for Elk Mountain cap clouds and the observed circle (droplet) center spectrum from double or asymmetric ice crystals found within the same clouds.

air and the fraction of that total possessing frozen droplet centers.

Fig. 4 shows the relationship between the observed cap cloud limit temperatures and the corresponding total ice crystal concentration, "frozen droplet" crystal percentage, and "frozen droplet" crystal concentration. From the 48 samples presented in Fig. 4, it can be seen that the frozen droplet crystal percentage reaches a maximum average value near 45% in the temperature range −13 to −16C. Thus, in this temperature range it would appear that the ice nucleation mechanisms (deposition or droplet freezing) may occur with equal ease. For both warmer and colder temperature regimes, the contribution of frozen droplet crystals appears to be ~25% of the total ice crystal population. The data shown in Fig. 4 also suggest some degree of temperature dependence for all three parameters between −8 and −25C.

Conditions of ice supersaturation but water subsaturation are commonly observed atop Elk Mountain, particularly during periods of time prior to formation of the cap cloud. Carefully calibrated sling psychrometers were used to measure the degree of ice supersaturation and water subsaturation; while such a technique is inadequate for accurate assessment of the degree of ice supersaturation, it is sufficiently accurate to determine if ice supersaturation exists. During the periods of ice supersaturation and water subsaturation, small concentrations of ice crystals would be expected to form naturally on deposition nuclei; and due to the absence of condensed water droplets (neglecting those very few hygroscopic nuclei possibly present), ice crystals would not be expected to exhibit any double crystal or asymmetric structure with circular centers. Fig. 5 illustrates a representative sample of an ice crystal replica acquired during ice supersaturation but water subsaturation conditions which exhibits an absence of frozen cloud droplet centers and double or asymmetric structures. Frozen cloud droplet centers were never found in ice crystals replicated during water subsaturated conditions. Hence, ice crystals found in ice supersaturated but water subsaturated conditions are presumably the result of growth by deposition.

Similar results were also to be expected during periods of seeding with dry ice, since the deposition nuclei are activated by the strong cooling and resultant high supersaturation induced by the dry ice. Indeed, ice crystals formed in such a manner did not possess frozen droplet centers and/or asymmetric structures as shown in Fig. 5.

Based on this evidence, it may be suggested that the respective concentrations of the frozen droplet crystals

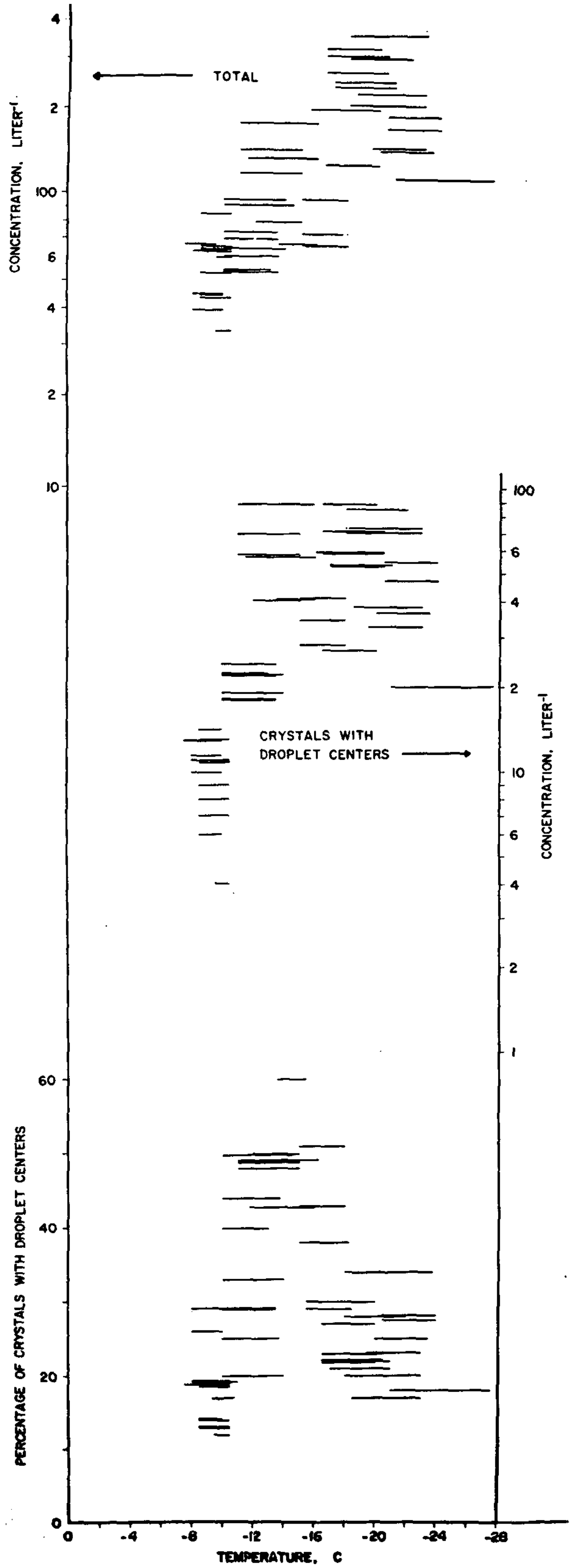

FIG. 4. The relationship between the observed cap cloud limit temperature and the corresponding total ice crystal concentration, percentage of "frozen droplet" crystals, and the "frozen droplet" crystal concentration (48 samples).

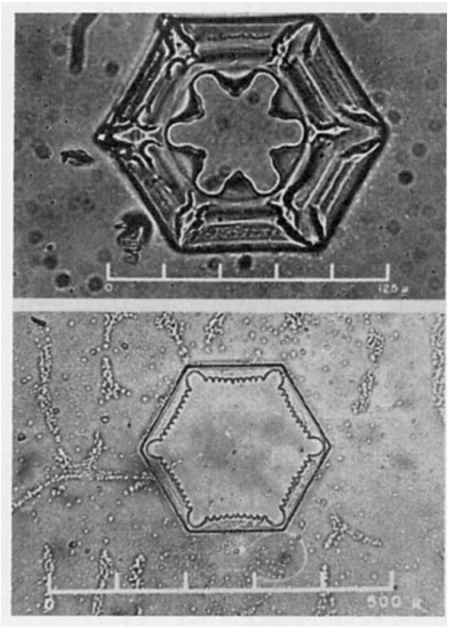

FIG. 5. Hexagonal plate crystals without both a droplet center and double or asymmetric structure observed (top) within an ice supersaturated but water subsaturated environment atop Elk Mountain (−16C, 115% with respect to ice), and (bottom) during dry ice seeding within the cap cloud at −13C.

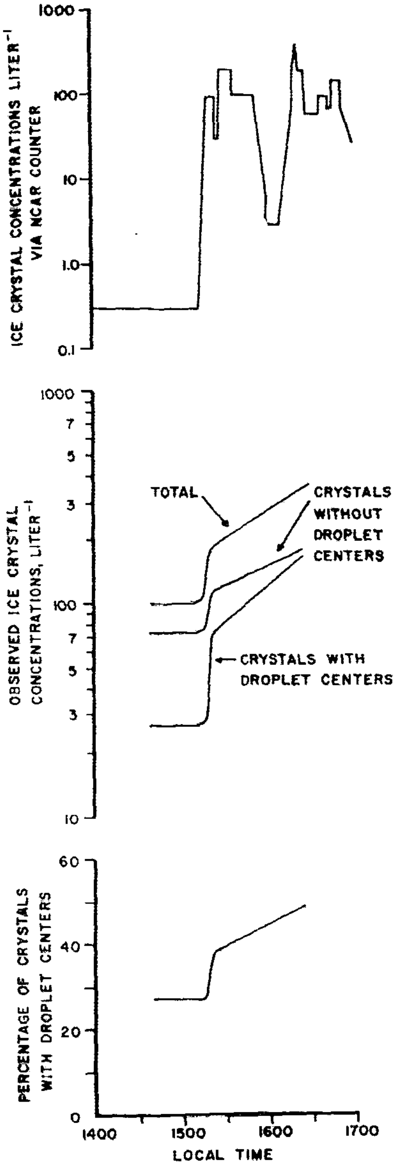

Fig. 6. Time section of the NCAR acoustical counter output and observed ice crystal data during a cap cloud seeding experiment with silver iodide on 18 December 1968.

and of the balance of the ice crystal population may be considered as representative estimates of the freezing and deposition nuclei concentrations, respectively, disregarding any ice multiplication processes.

4. Implication

To this point the discussion has focused on ice formation by droplet freezing in an untreated cap cloud. In addition to the above observations, ice crystal replica signatures were also obtained during cap cloud seeding with an acetone generator burning a 2.5% solution of AgI prepared with NaI. These replicas were examined for the purpose of detecting any possible changes in the percentage of frozen droplet crystals during seeding intervals as implied from the cloud chamber nucleation studies of Weickmann *et al.*

Fig. 6 shows a time cross section of ice crystal (hexagonal and thick plates) data and ice nucleus concentrations detected by an NCAR acoustical counter for a period of time prior to and during a cloud seeding experiment on 18 December 1968 at the Elk Mountain facility. Throughout the seeding experiment, the height of the cloud base (2900 m MSL), the cloud base temperature (−17C), and the Observatory (3300 m MSL) temperature (−20C) remained steady. The seeding agent was injected into the cloud from below cloud base; at all times during the experiment, water saturated conditions were maintained through the treated cloud depth.

During background conditions prior to 1515 MST, the percentage of frozen droplet crystals lay near 27%, a value to be expected from other independent data shown in Fig. 4. Following the arrival of the AgI (detected by the NCAR acoustical counter at the Observatory at 1515 MST), there was a corresponding increase in the total ice crystal concentrations, especially in the percentage and concentration of the frozen droplet crystals.

If the respective concentrations of the frozen droplet crystals and the balance of the ice crystal population may be considered to be representative estimates of the droplet freezing and deposition nuclei concentrations, as

Table 1. A summary of observed ice crystal data obtained prior to and during a cap cloud seeding experiment on 18 December 1968.

Time (MST)	Total ice crystal concentration (liter⁻¹)	Frozen droplet crystals (percent)	Frozen droplet crystal concentration, i.e., freezing nucleus concentration (liter⁻¹)	Balance of ice crystals, i.e., depositional nuclei (liter⁻¹)
Prior to 1515	100	27	43{ 27	38{ 73
1520	181	38	70 }141	111 }106
1625	347	49	168	179

premised earlier, then the contributions from these respective nuclei to the increase nucleation activity during the cap cloud seeding can be assessed from Fig. 6 or Table 1.

During the first several minutes following the detection of AgI the *absolute* increases in the droplet freezing and deposition nuclei concentrations were nearly equal; however, in the *relative* sense, the droplet freezing nucleus concentration increased nearly $2\frac{1}{2}$ times, while the deposition nucleus concentration increased by a factor of 1.5. After more than 1 hr of seeding, the droplet freezing and deposition nucleation modes appeared responsible for nearly equal numbers of crystals, but the increases in the concentration of the respective nuclei (droplet freezing vs deposition) over background concentrations seemed to slightly favor the freezing nucleus. It may thus be concluded from this experiment that the droplet freezing and deposition nucleation activities of AgI produced from a generator burning an acetone solution of silver iodide and sodium iodide yielded equal numbers of observed ice crystals in orographic cap clouds for the case of cloud activating temperatures between -17 and -20C.

Examples of ice crystals generated by cloud droplet freezing during other silver iodide seeding sessions are shown in Fig. 7. The two photographs illustrate the crystal structures found during cap cloud treatment (-12 to -14C) with 15% $AgIO_3$ pyrotechnics (supplied by the Naval Weapons Center, China Lake, Calif.) and with an acetone generator burning a 2.5% solution of AgI prepared with NH_4I. It is again emphasized that cloud droplet freezing does produce ice crystals in untreated clouds; on the other hand, silver iodide, produced by various methods, increases the number of ice crystals possessing cloud droplet embryos above background levels.

Further research concerning the relevance of the findings in this paper and those, for example, of Edwards and Evans (1968) concerning the role of silver iodide in the drop freezing process in natural clouds, are forthcoming.

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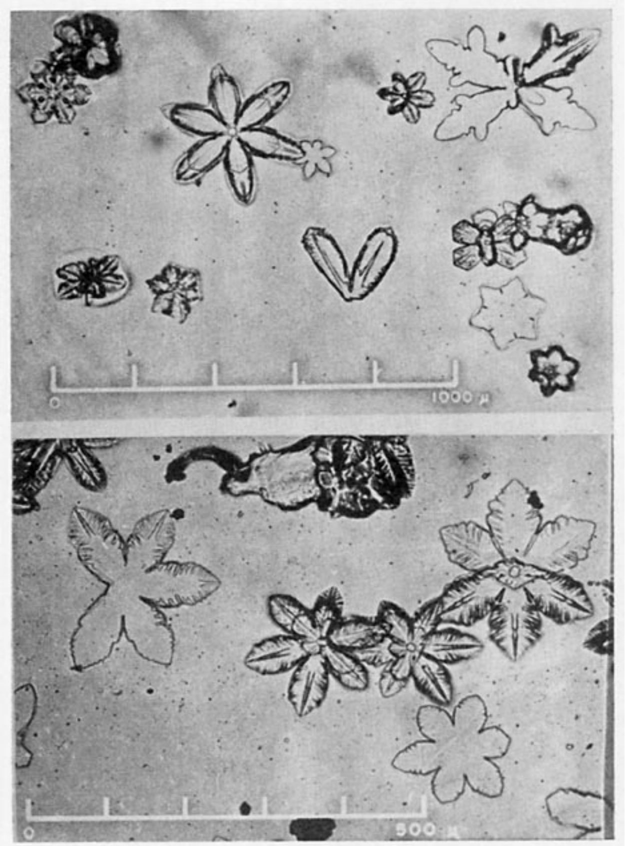

FIG. 7. Representative collection of plate family crystals observed (top) within the cap cloud (-12 to -15C) during silver iodide seeding with 15% $AgIO_3$ pyrotechnics, 10 March 1970 at 1335 MST, and (bottom) seeding with an acetone generator burning a 2.5% solution of silver iodide prepared with ammonium iodide, 10 March 1970 at 1510 MST.

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