Inferences about Ice Nucleation from Ice Crystal Observations

AUGUST H. AUER, JR.

Dept. of Atmospheric Resources, University of Wyoming, Laramie 82070

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ABSTRACT

The total concentration of ice crystals found in natural cap clouds appears to result from the nucleating behavior of at least two types of ice nuclei, i.e., freezing and deposition. The respective contributions of these two modes of nucleation have been partitioned by the use of techniques in which the identification of ice crystals resulting from frozen droplets can be made. Furthermore, the identification of deposition ice crystals at water saturation, when compared with similar data obtained during water subsaturated but ice supersaturated conditions, has yielded an insight as to the dependence of deposition ice crystal concentration (deposition nuclei) upon both temperature and ice supersaturation.

1. Introduction

In the past, both laboratory and field experiments have devoted considerable effort to the study of natural ice nuclei, those minute airborne particles which are believed to play a vital role in starting the growth of ice crystals in clouds of water droplets that have risen high enough to be cooled well below 0C. Only recently have attempts been made to examine ice crystals under known conditions with natural clouds. Since it is generally accepted that the concentration of ice crystals vitally affects the formation and growth of precipitation in many cloud systems, measurements of ice crystal concentrations can thus provide important basic knowledge concerning weather modification attempts which depend, in principle, upon introducing ice crystals into supercooled clouds in which, supposedly, no crystals have yet formed. Concentrations (interpreted as deposition[1] nuclei) effective at various temperatures and unspecified ice supersaturations have been used extensively as a primary index for estimates of ice crystal concentrations forming at corresponding temperatures.

However, the importance of cloud droplet freezing (i.e., freezing nuclei[2]) as a method of generating ice in natural clouds is gaining in popularity and the appropriate conditions for its occurrence are being defined. Recently, Auer (1971) has shown that frozen cloud droplets of a monocrystalline structure appeared to act as the embryos for planar ice crystal formation in natural untreated cap clouds. Continued literature research in this area has shown that Auer's extended observational evidence was actually a revival of thinking first premised by Bentley (1924) in which he said:

> "That many of the crystals seize upon and crystallize around frozen cloud droplets seems very probable, as the nuclei of at least one-half the (dendritic) crystals are tiny circular figures, looking much like an encased cloud droplet. Further support is given this theory by the fact that they correspond in size, also, with the cloud droplets (Figs. 10, 24, 25 and 28)."

It is interesting to note that Bentley's "one-half" corresponds strikingly close to the 45% estimate by Auer (Fig. 4) of the number of dendritic crystals forming as a result of frozen droplets! Nakaya *et al.* (1936), Nakaya (1954) and Weickmann (1947) came to the same qualitative conclusion concerning the possible role of frozen (monocrystalline) cloud droplets as embryos for planar ice crystals.

Many observations of planar ice crystals with frozen droplet (monocrystalline) centers appear to exist, but the significance of these observations never appeared to be emphasized. For example, the reader is referred to aufm Kampe *et al.* (1951, Figs. 8, 9, 10); Okita (1954, Photographs 3, 9, 14); Schaefer (1962, Fig. 2); Schaefer (1968); Khrgian (1963, Fig. 46c); Bashkirova and Pershina (1964, Fig. 2); Magono and Lee (1966, Figs. 38, 48, 104); Mossop *et al.* (1967, Figs. 1a–b); Ono (1969, Figs. 7a, 12); and Ono (1970, Figs. 14b, 15c–d) for examples of ice crystals similar to those cited in the unabridged work of Auer (1970).

Fig. 1 illustrates a representative pair of ice crystal replicas, with and without the frozen cloud droplet center and accompanying double crystalline structure (Nakaya, 1954; Weickmann, 1947; Auer, 1971). Fig. 2

[1] Deposition nucleus (sublimation nucleus): any particle upon which an ice crystal may grow by the transition of water to the solid from the vapor phase.

[2] Freezing nucleus: any particle which, when in contact with or present within a mass of supercooled water, will initiate growth of an ice crystal about itself.

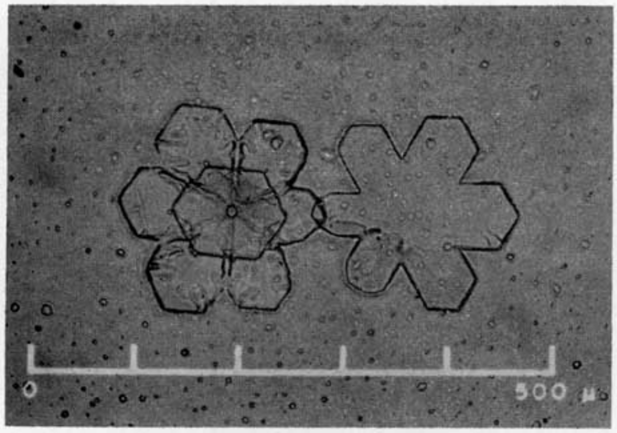

FIG. 1. Comparison of broad-branched crystals with and without a frozen droplet center and a double crystal structure.

shows a side view of a dendritic crystal possessing a droplet center and double crystalline structure. In this case, the frozen droplet diameter is near 25 μ, approximately twice the average value of 11 μ cited by Auer (1971), but still within the droplet population of the cap cloud. The larger droplet center made it possible to obtain a better photograph. Notice also that the crystal in Fig. 2 exhibits a threshold degree of riming of cloud droplets whose diameters are very nearly the same as that of the droplet center, suggesting that all droplets came from the same cloud droplet population (Bentley, 1924; Weickmann *et al.*, 1970; Auer, 1971).

If the concentrations of crystals with frozen droplet centers and those of the balance of the ice crystal population may be considered as representative estimates of the freezing and deposition nucleus concentrations, respectively, disregarding any fragmentation nucleus by ice multiplication processes, then it may appear that sufficient qualitative information is at our disposal to suggest some estimates of the natural ice forming power in the atmosphere.

2. Procedures

During the past several years, a climatology of the ice crystal concentrations occurring within the orographic cap cloud over Elk Mountain, Wyoming, has

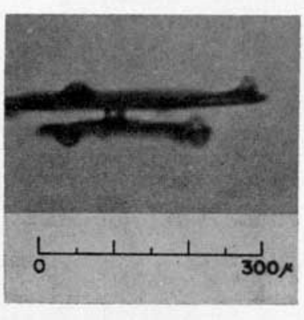

FIG. 2. Side view of an ice crystal showing the frozen droplet center and double crystal structure.

been obtained. These ice crystal concentrations were determined using hand-held Formvar-coated slides and sweeping through a small, known volume (typically 10 liters) of cloud at heights of 2–3 m above the surface. Estimates of collection efficiency for this method of ice crystal sampling were obtained utilizing the inertial collection theory of Ranz and Wong (1952). For the typical sampling situation (-10C, 700 mb, 5 m sec^{-1} impaction velocity), the collection efficiencies were estimated at 0.5 and >0.9 for 25 and 75 μ diameter crystals, respectively. Thus, the impactor sampling procedure appeared applicable (within the limits of the Ranz and Wong theory); and since fewer than 10% of the ice crystals observed during typical Elk Mountain cap clouds are <25 μ diameter, no collection efficiency corrections were used. Normally, the sampling was performed over a time interval of $\sim$300 sec downwind from the leading formation edge of the cap cloud. The effect of blowing snow on the measured ice crystal concentrations within the immediate environment of the Elk Mountain Observatory is considered to be negligible for the following reasons: strong, gusty winds in this location are infrequent; if gusty winds are present, the ice crystal sampling is limited to periods of intervening lulls; and the irregular or partially sublimed structure of ice crystals due to blowing snow can be easily distinguished from the regularly shaped and well-structured ice crystals occurring in the cap cloud. Schmidt[3] has found during blowing snow conditions that the contribution of ice particles at 1 m heights in 8 m sec^{-1} winds is less than 1 liter^{-1}.

Typically, a small vertical temperature gradient of 3–6C exists through isolated cap clouds; the methods of determining the cap cloud temperature limits and their reliability are described in Auer *et al.* (1969).

It appears that the possible sources of observational error (e.g. impaction speed, sampling volume, temperature, crystal size) in this sampling procedure could account for differences of a few tens of a percent, but certainly not an order of magnitude discrepancy in ice crystal concentrations. It should be emphasized that the ice crystal observations collected for this report are from a quasi-steady-state cloud system, and are not the result of an aged, decaying or seeded cloud system.

3. Results

Observations of ice crystal concentrations averaged over 1C intervals of cloud temperature are presented in Fig. 3 together with the 70 and 95% confidence limits obtained by application of appropriate statistical estimation theory for normal and/or skewed distributions. Three specific observations of ice crystal concentration at temperatures <-28C are also individually plotted.

[3] Schmidt, R. A., 1971: Personal communication relating to blowing and drifting snow studies. Rocky Mountain Forest and Range Experiment Station, U. S. Forest Service, Fort Collins, Colo.

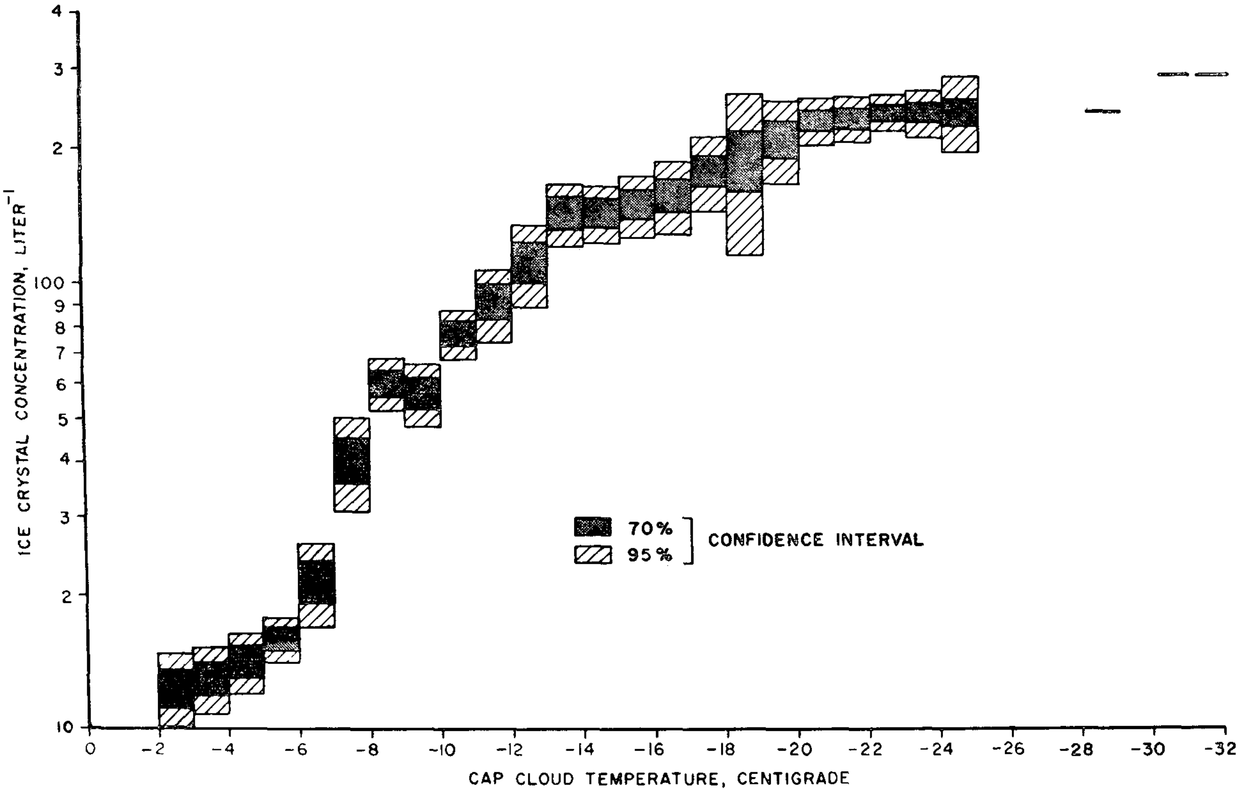

FIG. 3. Observed relationship between ice crystal concentration (70–95% confidence limits) and cap cloud temperature, including three specific observations at temperatures between −28 and −32C.

Ice crystals are found in all cap clouds whose cloud top temperatures[4] were colder than −2C. The concentrations of ice crystals observed within the Elk Mountain cap cloud are in encouraging agreement with other recent observations taken in orographic or stratiform clouds (Koenig, 1967; Mossop *et al.*, 1967; Mossop, 1968; Hobbs, 1969; Hobbs and Ryan, 1969; Mossop and Ono, 1969; McTaggart-Cowan *et al.*, 1970). The minor discrepancies that do exist between observations at Elk Mountain and those cited above appear to stem from the possibility of inaccurate determination of the temperature gradient through the orographic or stratiform clouds.

Table 1 utilizes the results shown in Fig. 3 along with the concentrations of ice crystals (with or without frozen droplet centers) as found by Auer (1971, Fig. 4)

[4] The reader is reminded at this point that while cloud top temperature could be as low as −2C, the temperature at the replicating site must be at least −1C to insure Formvar replication.

TABLE 1. A summary of ice crystal concentration data observed within the Elk Mountain cap cloud (50 clouds).

Cap cloud temperature (°C)	Total ice crystal concentration, (liter⁻¹)	Percentage of frozen droplet crystals	Concentration of ice crystals from freezing nuclei (liter⁻¹)	Concentration of ice crystals from deposition nuclei (liter⁻¹)
− 9 to −10	56	19	11	45
−10 to −11	77	26	20	57
−11 to −12	90	32	29	61
−12 to −13	111	36	40	71
−13 to −14	140	41	58	82
−14 to −15	142	44	62	80
−15 to −16	150	48	72	78
−16 to −17	157	39	61	96
−17 to −18	180	30	54	126
−18 to −19	188	27	51	137
−19 to −20	210	24	51	159
−20 to −21	231	23	53	178
−21 to −22	233	23	54	179

TABLE 2. A summary of ice crystal concentrations observed during 12 individual temperature and ice supersaturation conditions on Elk Mountain.

Observed temperature (°C)	Saturation with respect to ice (SS_i) (percent)	E (percent)	Deposition nucleus concentration (liter^{-1})
−2.5	101.5	61	1
−4.0	103	69	2
−7.6	105	65	5
−10.6	106	58	7
−11.1	103	26	1
−13.2	108	60	7
−14.0	107	47	4
−14.3	109	54	5
−14.5	107	48	7
−15.3	110	60	13
−16.2	112	72	17
−20.6	115	69	47

and from more recently acquired data. Again, the reader is reminded that the concentrations of the frozen droplet crystals and of the balance of the ice crystal population are considered as representative estimates of the freezing and deposition nucleus concentrations, respectively.

It is interesting to note from Table 1 or Fig. 3 that the concentration of deposition nuclei appears to increase with decreasing temperature to about −20C, thereafter remaining nearly constant; the freezing nucleus concentration remains nearly constant between 50–60 liter^{-1} for temperatures < -13C.

Conditions of ice supersaturation (but water subsaturation) are occasionally observed atop Elk Mountain, particularly during periods prior to the formation of the cap cloud. Carefully calibrated psychrometers were used to measure the degree of ice supersaturation and/or water subsaturation; such a technique is adequate for assessing the degree of ice supersaturation to $\pm 3\%$ (−20C) and $\pm 1\%$ (−2C). Auer (1971) suggested that all crystals observed during conditions of ice supersaturation (but water subsaturation) form on deposition nuclei (neglecting those very few hygroscopic nuclei possibly present) and grow by diffusion. Table 2 summarizes the observations of temperature, ice supersaturation and, hence, deposition ice crystal concentrations from 12 individual events. In addition, Table 2 contains a parameter E defined by

$$E = \left[\frac{e_x - e_i}{e_w - e_i} \right] \times 100,$$

where e_x, e_w and e_i are the ambient, water saturation,

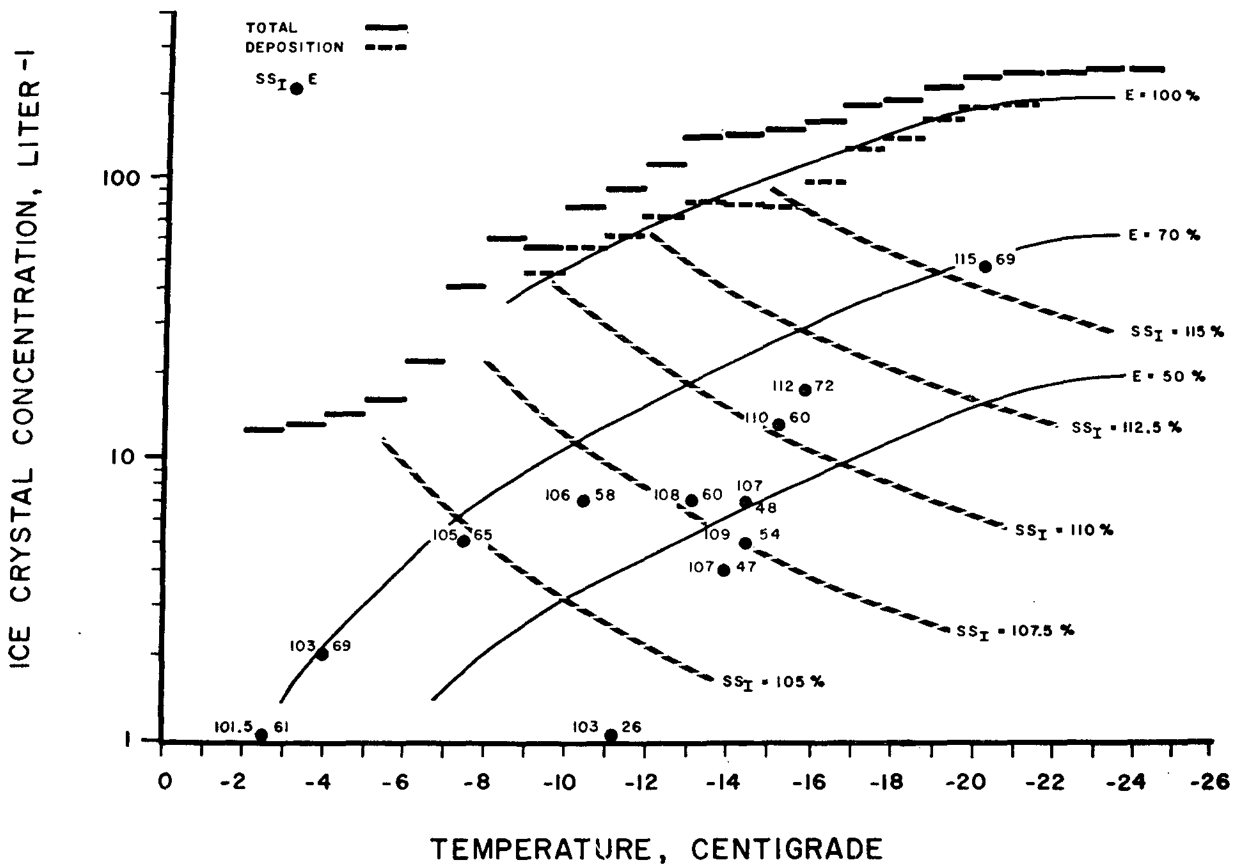

FIG. 4. Relationship between the observed concentration of deposition ice crystals (activated at or below water saturation) as a function of cap cloud temperature.

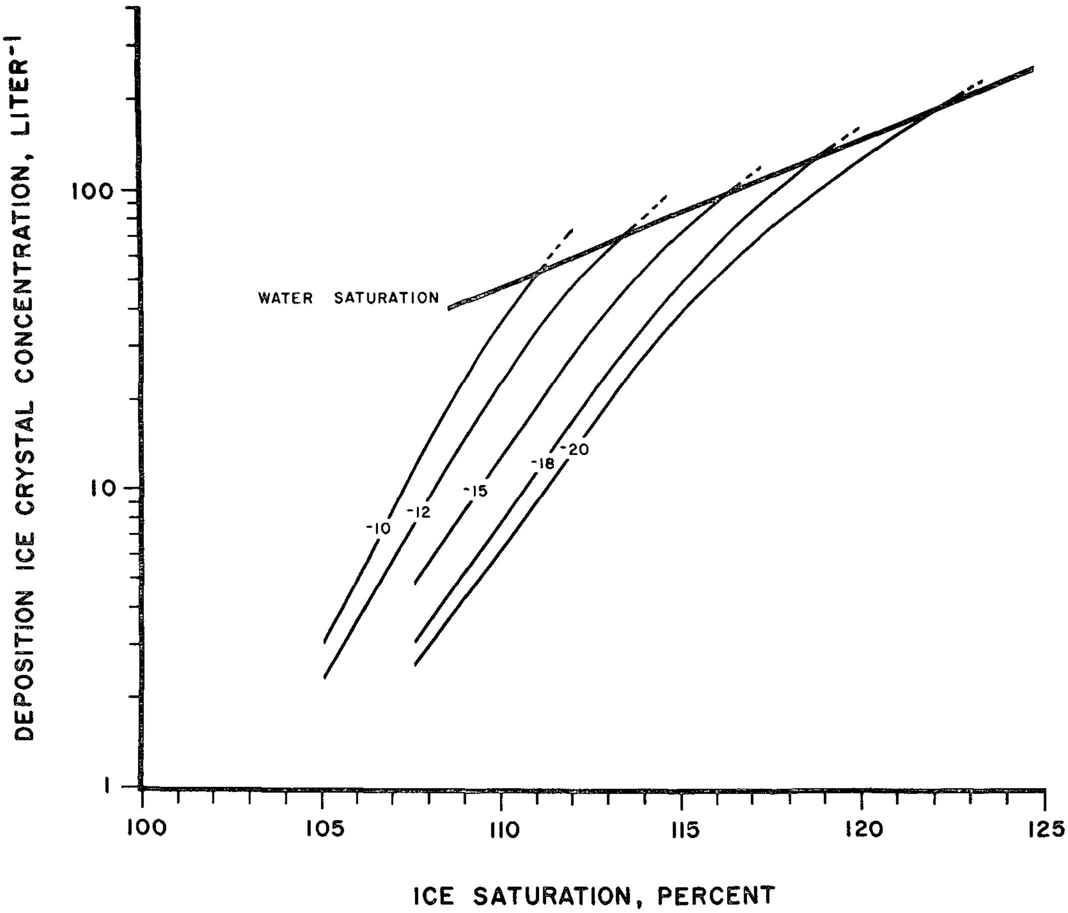

FIG. 5. Relationship between observed concentration of deposition ice crystals as a function of observed ice saturation and temperature.

and ice saturation vapor pressures, respectively. Thus, the parameter E is the percentage or degree of water saturation attained above ice saturations (SS_i); e.g., for $SS_i=100\%$, $E=0$; for $SS_i=121\%$ (−20C), $E=100\%$.

Fig. 4 is derived from the data presented in Tables 1 and 2 and illustrates the temperature dependence of the total ice crystal concentration, the deposition ice crystal concentration at water saturation conditions, and the observed deposition ice crystal concentration at various ice supersaturation (but water subsaturation) conditions. In addition, values of E are listed at each data entry for ice supersaturation. Isopleths of ice supersaturation and the parameter E were analyzed by eye and appear as dashed lines.

While the concentration of deposition ice crystals (active at water saturation) shown in Fig. 4 may be considered representative of some sort of average deposition nucleus concentration for the approximate 150 samples, it is not certain how representative the limited number of observations of deposition nuclei (active at water subsaturation) is. The analysis in Fig. 4 is based on the assumption that the deposition nuclei active at water subsaturation are from the same population as those deposition nuclei active at water saturation.

Fig. 5 presents data identical to those contained in Fig. 4 and is intended to assist the reader in interpreting the dependence of the activated deposition nucleus concentration upon ice saturation. In Fig. 5 the deposition ice crystal concentration (i.e., deposition nuclei) is plotted as a function of ice saturation for temperatures of −10, −12, −15, −18 and −20C.

An inspection of either Fig. 4 or 5 illustrates the dependence of the deposition ice crystal concentration upon the degree of ice supersaturation. For example, at −16C with $SS_i=108\%$, a concentration of 5 liter^{-1} is found, while at the warmer temperature of −12C with $SS_i=112\%$ (still water subsaturated), the concentration is 50 liter^{-1}. Again, if the temperature remains −16C but the SS_i is increased to 114%, the concentration of deposition ice crystals is increased to 50 liter^{-1}. It is evident, therefore, that the active number of deposition nuclei is dependent on the supercooling as well as the degree of ice supersaturation.

4. Implications

The physical basis for most weather modification operations in the past has been the hope that seeding would produce colloidal instability in clouds either prematurely or with greater efficiency than in nature. Most cloud seeding activities presume that a portion of the treated cloud is supercooled and that nature is not producing sufficient ice nucleating embryos at the temperature of the cloud; thus, treatment with chemical agents should change a proportion of the supercooled liquid cloud to ice. Since the evolution of the precipitation particles does take place within the cloud, it seems apparent that the measurements of ice crystal concentrations *within* the cloud are more germane to the subject of precipitation production and ice-water budgets.

It is now apparent that the total concentration of ice crystals found in natural cap clouds results from the nucleating behavior of at least two types of ice nuclei, i.e., freezing and deposition. Since observed ice crystal concentrations far exceed measured ice nuclei concentrations within natural untreated clouds (Mossop *et al.*, 1967; Mossop, 1968; Auer *et al.*, 1969; Burrows and Robertson, 1969; Hobbs, 1969), it does not seem feasible in the future to use deposition ice nucleus concentrations as the primary index for evaluating the potential artificial treatment. Concentrations of deposition ice crystals at water saturation, when compared with similar concentration data obtained during water subsaturated but ice supersaturated conditions, have yielded an insight as to the dependence of deposition ice crystal concentration (deposition nuclei) upon both temperature and ice supersaturation. Furthermore, the concentrations of ice crystals from freezing nuclei (Table 1) in themselves exceed by 1–2 orders of magnitude the generally accepted ice nucleus concentrations of 0.1–1 liter^{-1} at temperatures between -15 and -20C, respectively.

The observational evidence of this influence of temperature and ice supersaturation upon the nucleation activity of deposition nuclei may have bearing on the interpretations given to ice nucleus measurements by various cloud chamber techniques. It appears imperative that the degree of ice supersaturation, as well as temperature, within such chambers should be closely monitored in order to avoid questionable interpretations concerning "ice nucleus concentrations."

Ludlam (1955), Bergeron (1959), Schaefer (1967) and Grant (1968) concluded that the practicability of a seeding operation hinges on the necessity of providing an ice crystal concentration in the treated cloud on the order of 10–200 liter^{-1} at -20C. Some revision of these authors' estimates of the optimum ice crystal concentration may be in order in light of the data presented herein. During observations of such ice crystal concentrations within the untreated cap cloud, copious amounts of riming occur due to an abundance of supercooled water droplets impinging on exposed surfaces.

The observed ice crystal concentrations reported in this paper and by other authors (already cited) should not be interpreted as an indication of colloidal stability within clouds, making them insensitive to artificial treatment. For example, Auer and Veal (1970) found that, in spite of such "high" background values of ice crystal concentrations, the ice contents of the untreated cap clouds were such that cloud seeding experiments resulted in distinguishable (factors of 5–10) increases in the ice content while still maintaining a supercooled liquid water cloud (i.e., water saturation).

Further research concerning the relevance of the findings in this paper to the modes of ice nucleation in clouds by silver iodide is forthcoming.

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