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APPLICATION OF CONTACT-FREEZING NUCLEI IN CONVECTIVE CLOUD SEEDING

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Abstract. Continuing ice crystal generation and larger and longer seeded area development as well as the tendency to form desirable rimed crystals and graupel, previously reported with silver iodide (AgI) seeded convective clouds were examined in comparison with dry ice seeding cases. The slow contact-freezing nucleation by AgI particles was identified as the main reason for this. It was pointed out that the contact-freezing mechanism avoids over-generation of ice crystals by deactivating when cloud droplets are exhausted. In order for the mechanism to occur effectively in clouds, use of a nucleant whose preceding fast ice nucleation mechanisms (deposition nucleation in particular) are not highly active is advantageous to avoid early nuclei loss. From their relatively low deposition nucleation activity and slower contact-freezing rate observed in the laboratory, the application of organic ice nucleants to convective cloud seeding was suggested, and based on the Brownian coagulation formula, the desirable nucleus size range was estimated for this process.

1. INTRODUCTION

In ice phase weather modification, it has been recognized that an understanding of the working physical processes is necessary in order to achieve the desired effect. The understanding of physical processes is therefore a test of the seeding hypothesis which has to be formulated at the beginning of a field program, and also provides clues to improve the hypothesis later. However, a seeding hypothesis consists of a series of physical processes and varies depending on how it is formulated and what physical processes it includes. In other words, the proper choice of physical processes as well as their ingenious use in the hypothesis must precede the seeding study. Thus, a proper knowledge of such physical processes is essential for the formulation of the hypothesis. In this paper, we shall discuss an unusual behavior of organic ice nucleus particles as a possible physical basis for a new seeding hypothesis formulation.

2. DIFFERENCE OF CLOUD REACTIONS SEEDED BY DRY ICE AND AgI

Between the two most commonly used seeding agents, i.e., silver iodide (AgI) and dry ice, there exists a well-known ice nucleation mode difference. The former nucleates ice by the so-called heterogeneous mechanisms which may be further classified into three mechanisms, i.e., deposition, condensation-freezing (including immersion-freezing), and contact-freezing nucleations (Schaller and Fukuta, 1979). These mechanisms depend mainly on the temperature and supersaturation of the environment, cloud droplet size and number concentration (at least for the contact-freezing mechanism), the size and physico-chemical characteristics of nucleus particles, and the time. Thus, the ice nucleation mechanisms of AgI are indeed complex. On the other hand, dry ice nucleates ice crystals by strong cooling, in which it is believed that the formation of water droplets is followed by a homogeneous freezing of them. These processes proceed quickly regardless of the temperature of supercooled clouds. However, as far as the behaviors of nucleated ice crystals are concerned, there is no evidence that they are noticeably different for AgI and dry ice.

When these two ice nucleants were tested in the field, there seemed to be a substantial difference in temporal variation between ice crystal number concentration and the shapes of the ice particles formed. Schaefer (1965) observed in his Yellowstone Park winter field experiments that ice crystals from AgI particles tended to last much longer than those from dry ice. When the droppable AgI flare is compared with dry ice pellets in supercooled cumuliform clouds, ice crystals resulting from dry ice seeding showed a shorter duration due to fall out. Also, they were not accompanied by newer and smaller ice crystals, indicating practically instantaneous initial ice nucleation. However, in the plume seeded with AgI flares, ice crystals lasted much longer and the seeded areas greatly increased in comparison to those seeded with dry ice. Large ice crystals were always accompanied by smaller and newer ones. Dry ice seeded ice crystals frequently grew into aggregates, while AgI flare seeded crystals tended to become rimed crystals and graupel (Strap et al., 1979; Marwitz and Stewart, 1981; English and Marwitz, 1981). Similar behavior was detected in the plume seeded with an AgI-NH_4I-acetone burner in supercooled stratus clouds (Fukuta et al., 1984). In vapor-activated metaldehyde (MA) seeding which was anticipated to trigger instantaneous ice nucleation under penetration seeding in stratus clouds, the ice crystals detected were unaccompanied by smaller and newer ice crystals as much as in the case of dry ice seeding. AgI flares are known to produce at least partially smaller AgI particles that can be active for the contact-freezing mechanism (Sax et al., 1979).

It appears that the difference in ice crystal characteristics found between AgI and dry ice seeding is due to the nucleation mode differ-

ence rather than the characteristic difference of ice crystals shortly after ice nucleation. Thus, although the mechanisms involved in ice nucleation by AgI particles are complex, what contributes to the difference between the ice particles grown as well as their temporal behavior, with AgI seeding and those of dry ice seeding must be the slow acting mechanism of ice nucleation by AgI. Except for zones near the nucleation threshold, the deposition and condensation-freezing nucleations are so fast that they can be considered instantaneous relative to the time scale involved in cloud seeding. This leaves us with the contact-freezing mechanism for the slow activation process of AgI seeding. However, a large portion of AgI smoke particles can activate instantaneously when a suitable condition occurs, due to its relatively hydrophilic nature (low contact angle with water). There are other heterogeneous ice nucleants which are more hydrophobic, such as organics, and they can be engineered to act as contact-freezing nuclei in a much higher proportion. For this reason, we shall re-examine their behavior below.

3. ORGANIC ICE NUCLEANTS AS CONTACT-FREEZING NUCLEI

3.1 Laboratory Behaviors

During our past studies of organic ice nucleants, we noticed a puzzling behavior difference between AgI and organics. In the test using a laboratory mixing cloud chamber where cloud duration is rather limited, even if the particle sizes were about the same, AgI particles always nucleated in a much higher proportion compared with organic particles. It seems now that the reason has finally become clear. It is partly because organic nucleants are more hydrophobic. Table 1 gives the contact angles of water on nucleus compound surfaces (Fukuta, 1975b).

Table 1
Contact angle of water on ice nucleus compounds
(Fukuta, 1975b)

Ice Nucleus Compound	Contact Angle (Degrees)
Metaldehyde	66
1,5-Dihydroxynaphthalene	50
Phloroglucinol	26
Lead Iodide	78
Silver Iodide	17

1,5-Dihydroxynaphthalene (DN) for example, has a contact angle of 50 degrees, compared with an AgI contact angle of 17 degrees. Contact-freezing nucleation is known to be the most effective mechanism of ice nucleation as far as the temperature threshold is concerned (Fukuta, 1975a). However, it is not only the contact-freezing nucleation that controls the efficiency of slow but long lasting nucleation. Since there are other ice nucleation mechanisms that act more quickly than the contact-freezing mechanism, their activities govern the remaining fraction of the nucleus particles available for the contact-freezing mechanism. If they were highly active, a large fraction of the nucleus particle population would be lost in the early stages of seeding. Therefore, those fast mechanisms should be examined first. Figure 1 shows an example of fast condensation-freezing nucleation followed by the slow but long lasting contact-freezing nucleation of MA (Schaller and Fukuta, 1979).

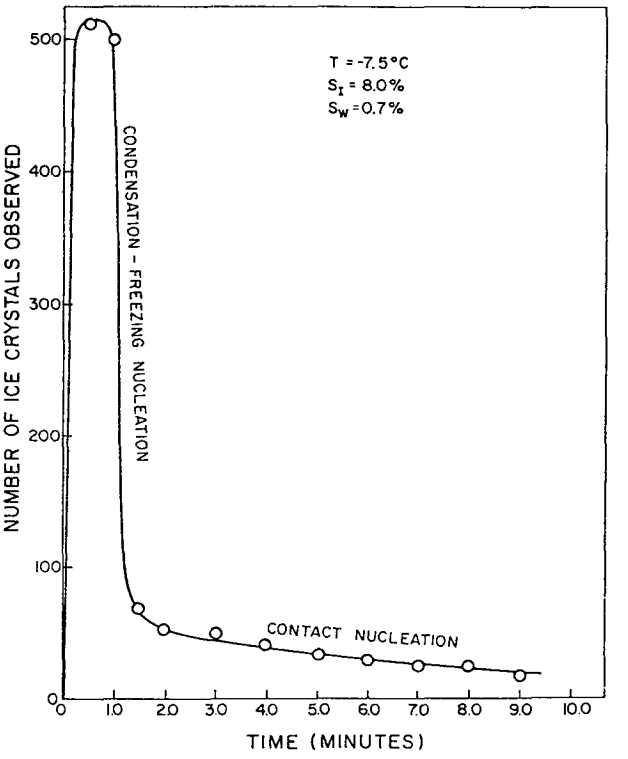

Fig. 1. Condensation- and contact-freezing nucleations for metadehyde smoke particles (Schaller and Fukuta, 1979).

The supersaturation thresholds of ice nucleants appear to approximately parallel their hydrophobicity (See Fig. 2). AgI showing the lowest contact angle of water, exhibits the lowest supersaturat-

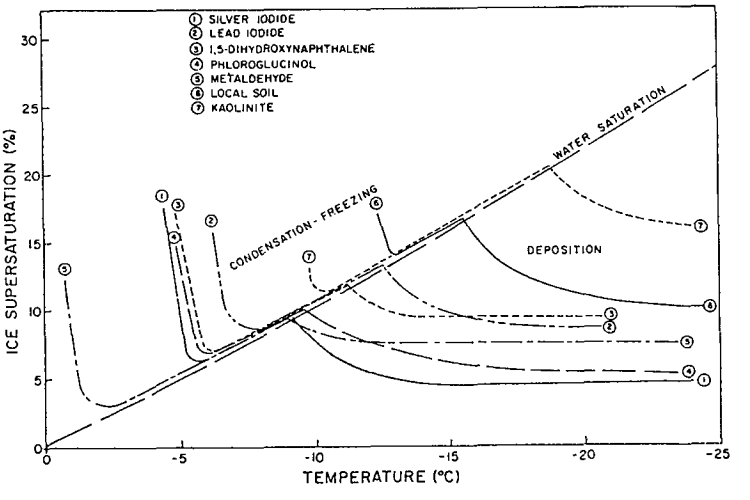

Fig. 2. Condensation-freezing and deposition nucleation behaviors of seven selected ice nucleants. The threshold curves represent 1.3% of nucleation in smoke particles in 1 min. (Schaller and Fukuta, 1979).

ion for a given level of the deposition nucleation threshold. Organics require higher ice supersaturation for nucleation at that level. This implies that organics are not likely to appreciably lose ice nucleus particles at the early stage. Nucleation of a small fraction of nucleus particles is commonly detected with organics in the laboratory mixing chamber where fast mechanisms, deposition nucleation in particular, dominate. This is evident from the measurements carried out in the isothermal chamber of Colorado State University (C.S.U.) where the fog supply was kept up for a long time (See Fig. 3). The DN particles are compared

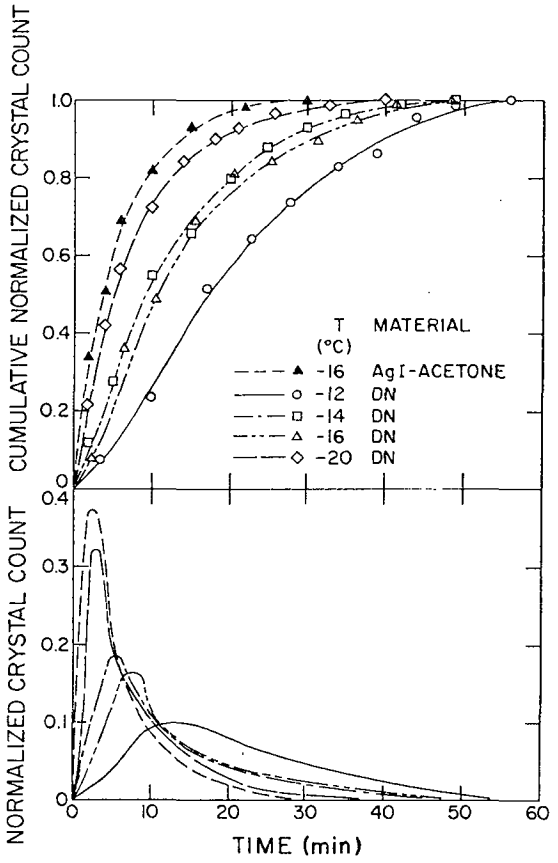

Fig. 3. Ice crystal count in the Colorado State University isothermal chamber for 1,5-dihydroxynaphthalene smoke particles (Fukuta et al., 1977).

with AgI particles generated by an acetone burner. Data taken at -16°C clearly show that DN is slower than AgI over a period of 50 min.

The organic ice nucleants are known to evaporate and decay. The lifetime of organic nucleants (estimated based on a mixing chamber measurement for nucleus particles being kept under elevated temperatures) did not match the trend of evaporation decay (Vasquez and Fukuta, 1984). Instead, they decayed far faster than the predicted evaporation loss. This, and the fast ice nucleation mechanisms detectable in the laboratory mixing chamber, led us to believe that the decay measured in their study was concerned only with fast ice nucleation processes and not contact-freezing. The slow contact-freezing nucleation is difficult to estimate in the laboratory due to the fact that the chamber condition has to be sustained for tens of minutes or even hours. Since the contact-freezing requires collision between the supercooled cloud droplets and ice nucleus particles, it should be of interest to estimate the collision process, at least by Brownian diffusion.

3.2 Factors Affecting Contact-Freezing Nucleation in Clouds

Contact-freezing nucleation is a multi-step process requiring a nucleus particle to collide with a supercooled droplet at the nucleation temperature and an active mechanism of the freezing nucleation upon collision. The latter depends on the physico-chemical characteristics of the nucleus particles and is essentially given at the moment of nucleus compound selection under the environmental temperature. However, the former depends on the physical parameters of both the nucleus particles and the clouds droplets and it is to some extent possible to estimate these. In a cloud system where the slow contact-freezing mechanism plays a significant role in precipitation development, the cloud air is nearly saturated which prevents the diffusiophoresis and thermophoresis from effectively working around a cloud droplet. Thus, we can ignore the phoretic processes and estimate the collision frequency of nucleus particles with cloud droplets and other nucleus particles by Brownian diffusion.

Using subscripts a and d for an aerosol (nucleus) particle and cloud droplet, the rate of aerosol particle number concentration change by collision with cloud droplets may be described as

$$-\frac{dn_a}{dt} = 2\pi(D_a + D_d)(r_a + r_d)\, n_a\, n_d \quad , \qquad (1)$$

where D is the diffusion constant given by

$$D = kT\left[1 + \frac{A\ell}{r}\right] / 6\pi\eta r \quad , \qquad (2)$$

with k the Boltzmann constant, A a constant near unity, ℓ the mean free path length of air, r the radius, η the dynamic viscosity of air, and n the number concentration.

Since $D_a \gg D_d$ and $r_d \gg r_a$, Eq. (1) becomes

$$-\frac{dn_a}{dt} = 2\pi\, D_a\, r_d\, n_d\, n_a \quad , \qquad (3)$$

which, after integration and omitting the subscript a for aerosols, yields

$$\frac{n}{n_0} = e^{-K_d t} \quad , \qquad (4)$$

where

$$K_d = 2\pi\, D_a\, r_d\, n_d \quad , \qquad (5)$$

and n_0 is the initial number concentration of aerosol particles.

For simultaneous particle loss by Brownian collision with droplets and coagulation among aerosol particles,

$$-\frac{dn_a}{dt} = K_c n_a^2 + K_d n_a \quad , \tag{6}$$

where K_c is the coagulation constant obtainable from Eqs. (1) and (2) by setting d = a, or

$$K_c = \frac{4kT\,(r + A\ell)}{3\eta r} \quad . \tag{7}$$

After integration, Eq. (6) becomes

$$\frac{n}{n_0} = \frac{K_d e^{-K_d t}}{K_d + n_0 K_c (1 - e^{-K_d t})} \quad . \tag{8}$$

Figure 4 shows the computed, normalized decay of

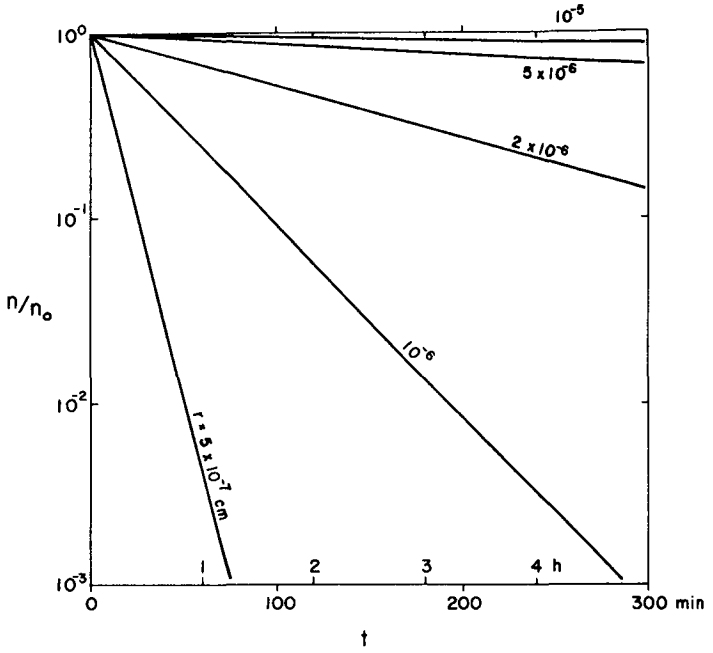

Fig. 4. The normalized decay of the number concentration of aerosol particles by Brownian collision with cloud droplets, n/n_0 with time t in an idealized continental cumulus cloud. The cloud condition: $r_d = 10^{-3}$ cm and $n_d = 5 \cdot 10^2$ cm^{-3}. For notations, see the text.

the number concentration of aerosol particles by Brownian collision with cloud droplets n/n_0 with time t, in an idealized continental cumulus cloud with $r_d = 10^{-3}$ cm and $n_d = 5\text{x}10^2$ cm^{-3}. For all computations, the temperature and pressure of the cloud were taken to be 263.2°K and 1000 mb. $r = r_a$ is the aerosol particle radius. Although $r = 5\text{x}10^{-7}$ cm is somewhat beyond the limit of the coagulation theory, the number concentration decreases very rapidly; within 50 min., 99% will collide. The $r = 10^{-6}$ cm case may be more suitable for obtaining a long lasting and larger seeding effect. It takes about 3 hours to reach the 1% level or the 99% collision. During a 3 hour period, the collision rate seems to remain sufficiently high to achieve a desirable seeding effect. For cases with $r > 5\text{x}10^{-6}$ cm, the collision rate may be too low.

Figure 5 shows the case for an idealized maritime cumulus with $r_d = 2.5\text{x}10^{-3}$ cm and $n_d = 50$ cm^{-3}.

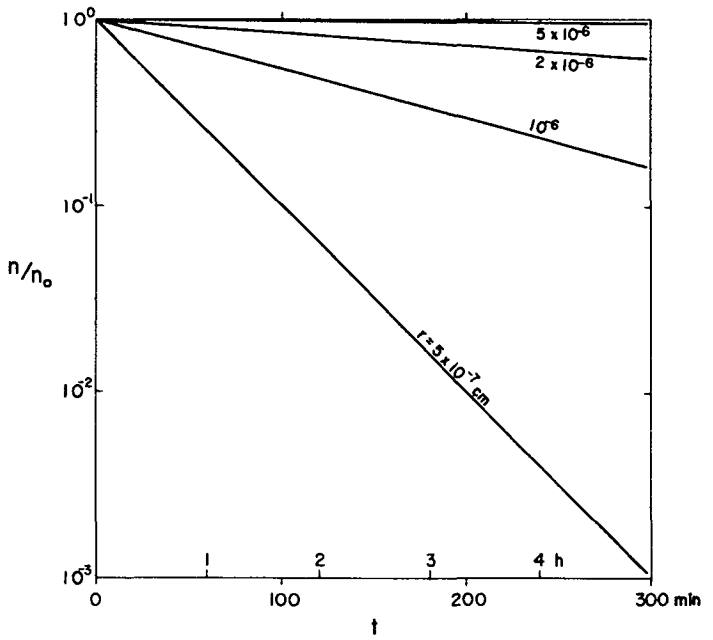

Fig. 5. Same as Fig. 4 except in an idealized maritime cumulus. The cloud condition: $r_d = 2.5 \cdot 10^{-3}$ cm, $n_d = 50$ cm^{-3}.

Generally, the collision rate is lower mostly due to the more dominant effect of a reduced n_d value compared to the effect of an increased r_d value. In this cloud, aerosol particles with r_d 2x10^{-6} probably produce ice crystals too slowly.

Figure 6 presents a comparison between the

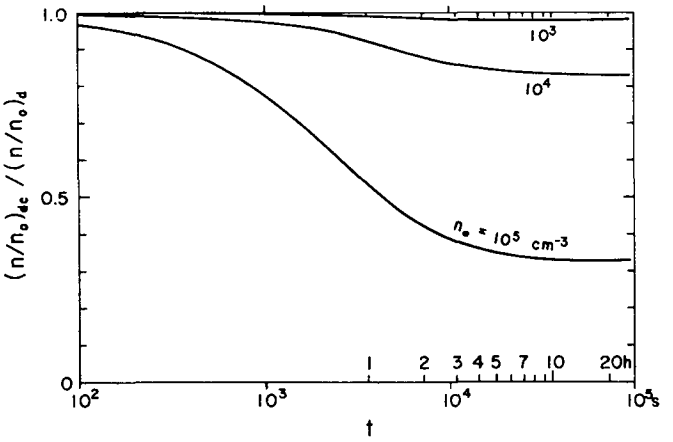

Fig. 6. The ratio between the normalized decay of the number concentration of aerosol particles by simultaneous mutual coagulation and collision with cloud droplets, and that by collision with cloud droplets alone plotted with time t. The cloud condition: $r_d = 10^{-3}$ cm and $n_d = 2 \cdot 10^2$ cm^{-3}.

case for collision loss of nucleus particles with cloud droplets alone and that in addition to mutual coagulation loss, based on Eqs. (4) and (8) with different n_0 under $r_d = 10^{-3}$ cm and $n_d = 2\text{x}10^2$ cm^{-3}. As can be seen from the figure, the mutual coagulation loss of nucleus particles is not high unless $n_0 > 10^4$ cm^{-3}, which may be avoided rather easily by dilution of the nucleus particles

with turbulent diffusion.

4. SLOW CONTACT-FREEZING NUCLEI IN CLOUD SEEDING

As we have seen above from studies in cloud seeding, as well as in the laboratory, there exists a difference between AgI and dry ice in microphysical characteristics of the resultant ice particles. The advantageous features of AgI seem to stem from the slow acting contact-freezing mechanism of the particles.

The slow contact-freezing process automatically avoids over-generation of ice crystals by stopping the mechanism when the supercooled droplets are exhausted. The preserved nuclei will be activated when the seeded plume diffuses into a new supercooled cloud volume and new cloud droplets appear, thereby inducing a long lasting effect in a large cloud volume and allowing desirable forms of ice particles such as rimed crystals and graupel to grow. Although there are other factors that control the form of the ice particles (e.g., Fukuta et al., 1984), a proper number concentration regulated by the contact-freezing mode is a requirement even for these particles. Because of the slower contact-freezing process, assisted by a smaller fractional loss from condensation-freezing and deposition nucleations compared with AgI, organic nuclei show promise in this regard, provided that the particle size is engineered properly in the range $r = 1 \sim 5 \times 10^{-6}$ cm. The supersonic expansion method of nucleus generation (Fukuta and Paik, 1976) can certainly perform such a function, but even the simpler evaporation-condensation method of the powder injection generator (Fukuta et al., 1977) should provide a sufficient number of particles in this range. For calibration of the contact-freezing nucleation, caution must be exercised to sustain supercooled clouds for a sufficient period of time without transitional supersaturation development.

A new or improved seeding hypothesis based on the use of such nucleants appears possible and its specific test in the field should be of interest to ice phase weather modification.

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5. REFERENCES

English, M. and J.D. Marwitz, 1981: A comparison of AgI and CO_2 seeding effects in Alberta cumulus clouds. J. Appl. Meteor., 20, 483-495.

Fukuta, N., 1975a: A study of the mechanism of contact ice nucleation. J. Atmos. Sci., 32, 1597-1603.

-----, 1975b: Comments on "A Possible Mechanism for Contact Nucleation." J. Atmos. Sci., 32, 2371-2373.

-----, and Y. Paik, 1976: A supersonic expansion method of ice nuclei generation for weather modification. J. Appl. Meteor., 15, 996-1003.

-----, M.N. Plooster, J.A. Armstrong, A. Gorove, J.A. Butz, R.C. Schaller and L. Glen, 1977: The development of organic ice nuclei generators for weather modification. Final Rept. to N.S.F. under Grant No. ENV-73-02910, June 1977.

-----, H.-S. Chang, J.L. Sutherland and D.A. Griffith, 1984: Comparative airborne tests of vapor activated metaldehyde and silver iodide particles in supercooled stratus clouds. Extended Abstract, 9th Conf. on Weather Mod., May 21-23, 1984, Park City, Utah, 6-7 (Boston, MA, Amer. Meteor. Soc.).

-----, N.-H. Gong and A.-S. Wang, 1984: A microphysical origin of graupel and hail. Proc. 9th Internat. Cloud Physics Conf., Tallinn, U.S.S.R., August 21-28, 1984, 257-260.

Marwitz, J.D. and R.E. Stewart, 1981: Some seeding signatures in Sierra storms. J. Appl. Meteor., 20, 1129-1144.

Sax, R.I., D.M. Garvey and F.P. Parungo, 1979: Characteristics of AgI pyrotechnic nucleant used in NOAA's Florida Area Cumulus Experiment. J. Appl. Meteor., 18, 195-202.

Schaefer, V.J., 1965: Studies of cirrus-type clouds at ground level. Proc. Internat. Conf. on Cloud Phys., May 24 - June 1, 1965, Tokyo and Sapporo, 414-418.

Schaller, R.C. and N. Fukuta, 1979: Ice nucleation by aerosol particles: Experimental studies using a wedge-shaped ice thermal diffusion chamber. J. Atmos. Sci., 36, 1788-1802.

Strapp, J.W., H.G. Leighton and G.A. Isaac, 1979: A comparison of model calculation of ice crystal growth with observations following silver iodide seeding. Atmos. Ocean, 3, 234-252.

Vasquez, H.R. and N. Fukuta, 1984: Evaporation decay of organic ice nucleus particles. J. Weather Mod., 16, 9-18.