Reply

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17 March 1991

1. Introduction

Aside from some questions concerning terminology, and the presentation of some evidence concerning the activity of AgI–AgCl as a condensation freezing nucleus, the main point of the comment by DeMott (1991), hereafter referred to as D, centers on the validity of our conclusion that contact nucleation alone cannot account for the ice-crystal concentrations we observed to result from seeding in an experiment conducted on 22 December 1986. The observations from this experiment were presented in Deshler and Reynolds (1990), hereafter referred to as DR, and have recently been questioned by Finnegan and Pitter (1991). In our reply to Finnegan and Pitter (Deshler and Reynolds 1991) we defended our interpretation that the increased ice-crystal concentrations (ICC) observed at the seedline resulted from seeding and also presented some additional justification for our opinion that contact ice nucleation could not account for the ICC observed. DeMott continues to question this last conclusion and points out several aspects of the ice nuclei (IN) described in DR, which should have been mentioned in characterizing the IN employed in the seeding experiment on 22 December 1986, and also in a case on 18 December 1986 presented by Deshler et al. (1990). With a proper accounting of these characteristics D indicates that observations reported by DR may agree with the laboratory results. Here we focus on repeating the calculations used by DR to predict the ICC expected from aircraft seeding using a 3% by weight solution of AgI, NH_4I, and NH_4ClO_4 at a 30 mol % level with respect to AgI. As pointed out by DeMott et al. (1983) and Finnegan and Pitter (1991), the active ice nucleus is AgI–AgCl.

These new calculations with a more careful accounting for the characteristics of the IN used does not change the conclusions of DR; that is, contact nucleation alone cannot explain the ICC observed to result from seeding with AgI–AgCl. To justify this conclusion and to clear up the confusion that has resulted from our previous discussions, we will present these calculations in some detail. Unfortunately, part of the confusion resulted from some order of magnitude calculations in DR, which were presented solely to indicate upper limits to the ICC and were not carefully separated from the more exact calculations based on scavenging rates for contact nuclei. We will refrain from speculating on what may be the active nucleation mechanism in these cases. First, however, we would like to defend our use of the word persistence.

Both D and Finnegan and Pitter (1991) have objected to our use of the word persistence. At the risk of being didactic, the definition given by Finnegan and Pitter was found in only one of three dictionaries checked and then was the third of three possible definitions. The first was the act of continuing firmly or steadily when faced with difficulty. This was the intended meaning as used by DR, and we feel it describes the phenomenon observed. In fact, to our knowledge, this case represents the longest period of time in which seeding material and its effects have been tracked with a research aircraft. In our view the seeding material and its effects continued as an entity, in spite of the dispersive nature of airflow in a cloud over a mountain barrier, to the downwind boundary of the cloud. We realize this is a much different phenomenon than is being described by Bigg and Turton (1988) and Mather et al. (1990) and that the time scales described by DR are much shorter.

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2. Calculation of ice-crystal concentrations expected from contact nucleation

In DR we did not intend to imply that we are questioning the conclusions of DeMott et al. (1983) for the laboratory situation. On the contrary, we accepted those conclusions and attempted to apply those laboratory results to a field situation. The fact that the seeding material tested in the laboratory was not reproduced faithfully will afflict many field experiments that are not designed specifically to test a laboratory result, and the Sierra Cooperative Pilot Project is a case in point. Still that should not prevent a comparison from being made, with appropriate allowance for the differences between laboratory and field environments. That brings up the two points raised by D, which were overlooked by DR (aside from the chemical terminology) in characterizing the IN used. First, DR used a 3% by weight solution of AgI, NH_4I, and NH_4ClO_4 at a 30 mol % level with respect to AgI instead of the 2% by weight solution tested by DeMott et al. Second, DR failed to account for the differences in draft past the IN generator between the natural draft used by DeMott et al. and the forced draft which an aircraft IN generator at flight speed experiences.

Both of these differences will affect the number and size of the IN produced, although by far the largest difference will result from the differences in draft past the IN generator. The differences in the efficiency of ice nucleation between a 2% and 3% by weight solution of AgI, NH_4I are much smaller than other uncertainties in the calculation to follow. Considering Fig. 9 in DeMott et al. (1983), it is clear that a 3% solution will increase the nucleation efficiency of the IN only a few percent due to the production of slightly larger particles. The difference between the 2% and 6% by weight solutions shown is 16% versus 24% at −10°C. The effect of differences in draft causes much larger changes. Again based on Fig. 9, the effect of the maximum draft was to create smaller and thus less efficient IN. The nucleation efficiency of a 2% solution decreases from approximately 16% to 2.4% at −10°C, when comparing natural draft to maximum draft. However, even though particle size and thus nucleation efficiencies, are reduced by a factor of 6.8, the number of particles created is greater and their diffusion (and thus coagulation with cloud droplets) more rapid. DeMott et al. estimate that the number of particles created per gram of AgI–AgCl increases by a factor of 7 when comparing maximum and natural draft, almost exactly canceling the decrease due to the lower nucleating efficiency. Thus, due to the offsetting factors of a 6.8-fold decrease in nucleating efficiency, but a 7-fold increase in the number of particles, the IN production rates of 10^{13} g^{-1} at −6°C and 6×10^{14} g^{-1} at −10°C used by DR appear correct. But this assumes the particle number concentration generated in the laboratory can be used in the field. In fact this is probably not the case.

The mechanics of seeding generators have been treated in many articles and reviewed by Dennis (1980), who points out that because of Brownian coagulation the number of particles produced by a seeding generator is limited to about 10^{14} m^{-3} of effluent. Thus, the number of particles produced by an aircraft generator is controlled primarily by the amount of air or exhaust gas traveling through the region where the particles form from the vapor. For the aircraft IN generators used for the cases described by DR and by Deshler et al. (1990) the exhaust gas flow rate is not well characterized. The generator consists of a tapered cylinder with an opening 25.4 cm in diameter on the upstream side and an opening 15.2 cm in diameter at the exhaust. However, the center 22.9 cm of the upstream opening is filled with the solution tank, limiting the opening on the upstream side to a 2.5-cm-wide ring with an interior diameter of 22.9 cm. With this opening and the seeding aircraft speed of 80 m s^{-1} the flow rate past the aerosol generator is 0.8 m^3 s^{-1} assuming no back pressure resulting from constriction of the flow as the cylinder tapers. Considering just the exhaust diameter of 15.2 cm the flow rate would be 1.5 m^3 s^{-1}. Thus, based on these characteristics a reasonable estimate of the exhaust gas flow rate appears to be 1 m^3 s^{-1}. In fact this is probably an overestimate, since around the combustion chamber there is an air baffle that will further constrict the flow.

With these considerations and the assumption of contact nucleation we now proceed directly with a calculation of the ICC to be expected from seeding. From Slinn (1971) the number of particles scavenged at time t, N_s, from an initial concentration of N_0, is

$$N_s(t) = N_0(1 - e^{-Kt}),$$

where $K = 4\pi DRN$; D is the particle diffusion coefficient, R the radius of the cloud drops, and N the concentration of cloud droplets. Assuming the aerosol to be approximately monodispersed with a 0.03-μm diameter (DeMott et al. 1983) gives us $D = 5 \times 10^{-5}$ cm^2 s^{-1}. From the observations of DR we have $R = 5$ μm and $N = 40$ cm^{-3}, the maximum cloud-droplet concentration observed by DR. With these values $K = 1.3 \times 10^{-5}$ s^{-1}, and since $t < 10^4$ s we may approximate the number of particles scavenged by $N_s = N_0Kt$.

With some assumptions as to the dispersal rate of the seeded volume the concentration of scavenged particles for any time after seeding may now be calculated. The volume of air affected at time t by 1 s of aircraft seeding is $\pi H_d V_d t^2 L/4$, where H_d is the horizontal dispersion, V_d the vertical dispersion, and L the distance traveled by the seeder aircraft in 1 s. We use $H_d = 1.0$ m s^{-1}, $V_d = 0.1$ m s^{-1}, and $L = 80$ m. Thus, in the dispersing seedline the concentration of particles scavenged by water drops is $CN_s = 4N_0K/\pi H_d V_d tL$, where N_0 is the number of particles created in 1 s by the seeder aircraft. Using the results presented by Dennis (1980)

TABLE 1. Ice-crystal concentrations expected at −10°C from seeding with AgI–AgCl, assuming contact nucleation, compared with observations.

	Calculations				Observations		
Cloud-drop concentration	10 cm^{-3}		40 cm^{-3}			Seedline	Near seedline
Time (min)	CN_s* (l^{-1})	ICC (l^{-1})	CN_s (l^{-1})	ICC (l^{-1})	Temperature (°C)	ICC (l^{-1})	ICC (l^{-1})
10	83	2.0	333	8.0	−6	45	55
70	12	0.2	48	1.1	−10	40	15
90	9	0.2	37	0.8	−12	20	<1

* CN_s = concentration of scavenged particles.

and our estimate of 1 m^3 s^{-1} for the IN generator effluent flow rate gives us $N_0 \approx 10^{14}$. To obtain the number of ice crystals to be expected from seeding we now need only to multiply CN_s by the nucleation efficiency of the AgI–AgCl particles, which depends on both temperature and the draft. From Fig. 9 of DeMott et al. (1983) at maximum draft the ice nucleation efficiencies are 5.2% at −12°C, 2.4% at −10°C, and 0.02% at −6°C.

The results of these calculations for a temperature of −10°C are presented in Table 1 for two cloud-droplet concentrations, 10 cm^{-3}, the average observed by DR, and 40 cm^{-3}, the maximum observed. The ICC observed on 22 December 1986, both at and near the seedline, are also shown in Table 1. After 70 min, for a cloud-droplet concentration of 40 cm^{-3}, the expected ICC is 1 l^{-1} at −10°C, whereas the observations of DR show an increase from 15 to 40 l^{-1} at the seedline, suggesting that seeding created an additional 25 l^{-1} of ice crystals. This is clearly an increase over what is predicted based on contact nucleation, and perhaps a conservative one. This calculation has assumed that all ice crystals nucleated within the dispersing volume are still contained therein when in fact many of the ice crystals that nucleated early would have grown and fallen below the seeded volume by the time of the measurements at 70 min. These calculated concentrations are lower than reported by DR, since in the earlier calculations the limiting aspects of Brownian coagulation in the aerosol plume were not considered, allowing DR to assume an initial aerosol concentration a factor 10 higher than was used here. Thus, D's suspicion that DR overestimated the available IN is correct, but note that with the more realistic estimate of available IN, contact nucleation is less able to explain the observations. It is true that the concentration at the center of the seeded plume would be higher, but it is difficult to argue that the research aircraft penetrated the center on each of its 5 penetrations after 60 min. The vertical standard deviation for plume spread considering a Gaussian plume is only between 300 and 400 m for 60–90 min downwind (Pasquill 1961; Gifford 1961).

Thus, again we come to the conclusion that limiting the nucleation mechanism to contact nucleation cannot explain the results we see in the field using AgI–AgCl as the nucleating agent. The uncertainties in this calculation center primarily around N_0 and the nucleation efficiency, but note that to produce the ICC observed would require that our estimate of the product of these two numbers is low by a factor of 25 for a cloud-droplet concentration of 40 cm^{-3}, and by a factor of 100 for a droplet concentration of 10 cm^{-3}. If we use the laboratory measurements of DeMott et al. (1983) for maximum draft the aerosol production rate is estimated to be 2×10^{16} g^{-1}. This is in direct agreement with Fig. 5.3 in Dennis (1980) for the number of particles expected per kilogram of AgI for a lognormally distributed aerosol of 0.03-μm geometric mean diameter and geometric standard deviation between 1.5 and 2. With this estimate N_0, the number of particles released per second, becomes approximately 10^{15} and the ICC calculated in Table 1 would be increased by a factor 10. This is still lower than the observations by a factor of 3 to 10, and such a high initial concentration would suffer rapid depletion due to Brownian coagulation immediately after creation.

The conclusion reached here is even more apparent in a case study on 18 December 1986 (Deshler et al. 1990), where evidence is presented that suggests that seeding with AgI–AgCl produced, within 20 to 30 min, ICC of 100 l^{-1} at −6°C. Note that in the cloud on 18 December 1986 the cloud droplet concentration was 100 cm^{-3} at the seedline. This has the effect of increasing the ICC expected from seeding by a factor of 10 over the results presented in Table 1 for a cloud-droplet concentration of 10 cm^{-3}.

In conclusion we feel it imperative to point out that if laboratory results are to be applied to a field situation, care must be taken to ensure that the differences between the laboratory and the field situation are taken into account. To the extent possible we feel this has now been done for the case of 22 December 1986. Clearly more field studies are necessary to characterize the time dependent nature of ice-crystal production due to seeding with artificial ice nuclei, and to compare this production with laboratory measurements. These studies are important to help determine the best seeding materials and rates for future operational seeding programs.

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