Comments on "The Persistence of Seeding Effects in a Winter Orographic Cloud Seeded with Silver Iodide Burned in Acetone"

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1. General comments

The article by Deshler and Reynolds (1990), hereafter referred to as DR, contains errors and misconceptions concerning the chemistry, generation, characterization, and functioning mechanisms of silver iodide-containing ice nucleus aerosols. These errors may be due in part either to the authors not understanding the processes of ice nucleation and ice crystal formation, which are critical aspects of cold cloud modification, or to their careless use of jargon. In this comment, only the errors in DR relevant to ice formation are discussed.

First, use of the term "persistence of seeding effects" in the title is confusing. Persistence is the continuation of an effect *after its cause is removed.* The functioning of the ice nuclei (IN) in supercooled liquid-water clouds, producing ice crystals several tens of minutes after their generation, is regulated by collision rates of nuclei with cloud droplets and is not a matter of persistence. In the reported experiment, if IN function by collision with cloud drops, ice crystals will be produced *over a period of time* according to the equation:

$$d[\mathrm{IC}]/dt = K(T)[\mathrm{IN}][D], \qquad (1)$$

where [IC], [IN], and $[D]$ are the number concentrations of ice crystals, ice formation nuclei, and droplets; and K is the kinetic rate, which is dependent on the temperature (T).

Second, *silver iodide* is not "burned in acetone," as stated in the title. A *solution* is burned to yield the nucleating aerosol. When an acetone–water solution of AgI NH_4I plus 30 mole percent NH_4ClO_4 is burned, the resulting aerosol is a mixed chloroiodide of silver, $AgCl_{0.3}I_{0.7}$ (hereafter abbreviated AgClI). This aerosol is the nucleating agent, not the mixture of chemicals used to make the solution (as the text states several times). The AgI NH_4 INH_4ClO_4 mixture does not survive the combustion process.

Third, it is incorrect to ascribe the effectivity of AgClI to its AgI content alone. Vonnegut and Chessin (1971) studied the improved effectivity of $AgBr_{0.3}I_{0.7}$ (henceforth AgBrI) over that of AgI in freezing bulk water and concluded that the improvement was due to a better structure match of the new solid solution nucleant with ice, but they also pointed out that the improvement could have been due to other surface effects produced by the bromide substitution for iodide in the aerosol. This may also be the case for the AgClI aerosol used by DR. Vonnegut and Chessin noted that x-ray diffraction patterns of AgBrI lacked patterns characteristic of pure AgI or AgBr.

By way of clarification, although DR stated that the acetone–water solution of AgI NH_4I NH_4ClO_4 was *burned* in a pressurized stainless steel container (a Carley-type generator), it should be noted that the solution was *stored* in the container, but it was burned at atmospheric pressure in the generator's combustion chamber.

Finally, pyrotechnics used for cloud seeding in the United States do not contain silver *iodide*, as DR stated. They contain silver *iodate* ($AgIO_3$), which is the pyrotechnic oxidizer and source of the AgI aerosol. The IN composition can be modified to AgClI by inclusion of chlorine compounds in the pyrotechnics (Sax et al. 1979). Droppable pyrotechnics are droppable nucleus aerosol generators, not droppable seeding agents similar to dry ice pellets.

2. Mechanism of ice formation

The article contains several assumptions in its argument, which concludes that contact freezing is not the primary ice formation mechanism. Even so, their observational data taken during the IN plume encounters at 5 and 12 min after treatment (DR, Fig. 2) do not indicate differences in the ice particle concentrations within or outside the plume. The article states that 40 ice particles per liter might be attributed to the seeding agent at 10 min, but the data reveal no such signals in the fluctuating ice crystal concentrations. Rather, the laboratory result of DeMott et al. (1983), which demonstrated that contact freezing is the ice

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crystal formation mechanism for silver iodide nucleus aerosol, and which DR estimate would yield about 1 ice crystal per liter at 10 min, is consistent with the observational results presented in the article.

In the instrumentation section, DR focused on a comparison between the anticipated results of two mechanisms of ice formation. The article compared the way IN would function by contact freezing (DeMott et al. 1983) with the way the same IN would function by forced condensation-freezing (Finnegan and Pitter 1987). Deshler and Reynolds estimated that nuclei functioning by a contact freezing mechanism would initially yield about 1 ice crystal per liter per 10 min.

Finnegan and Pitter (1987) found that high concentrations of very small ice crystals were visible within 30 s downwind of a ground-based generator, indicating almost immediate ice formation. They suggested that the moisture in the exhaust plume condensing on IN was responsible for the high initial rate of ice formation. No quantitative measurements were taken in the field study to determine the yields, but Feng and Finnegan (1989), studying a hygroscopic nucleating agent (AgI · AgCl–4NaCl aerosol), found that the yields at −6°C were comparable whether at water saturation or at water supersaturation. Meanwhile the time for 90% of the IN to function in the Colorado State University cloud chamber decreased from 15 min at water saturation to less than 3 min at water supersaturation. Ice crystals take time to grow and fall out before they are counted. Therefore, ice formation occurred almost instantaneously in the water supersaturation case.

The small ice crystals DR observed after 90 min (10–70 l^{-1}), approximately coincident with the IN plume, may have formed from any of several mechanisms. Unfortunately, the field experiment did not yield sufficient data to determine which mechanisms operated. The IN plume was penetrated too infrequently for *rates* of ice formation within the plume to be measured. and the data do not allow reconstruction of the cloud conditions within the plume (e.g., temperature, drop concentration, and vapor density) that govern the rates and mechanisms of ice crystal formation. The small ice crystal sizes DR observed after 90 min suggest that the ice crystals had been formed within perhaps the 5–15 min prior to the time they were sampled. The ice crystals may have formed on natural IN or they may have formed by ice crystal multiplication (Hallett and Mossop 1974; Pitter and Finnegan 1990). If sufficiently strong updrafts were present to form local regions of transient supersaturation, they may have formed by condensation-freezing of generator-produced IN or they may have formed by contact freezing of generator-produced IN.

3. Summary

The nucleation and ice crystal formation terminology in DR is incorrectly articulated. Scientific concepts and results need to be presented in a manner that can be understood. The errors in DR can lead to misconceptions of critical concepts involved in cold cloud modification and hinder future research.

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