A Quest for Effective Hygroscopic Cloud Seeding

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ABSTRACT

It is shown here that hygroscopic seeding requires two orders of magnitude more hygroscopic agent than can be delivered by flare technology for producing raindrop embryos in concentrations to detect by cloud physics aircraft the microphysical signature of rain initiation. An alternative method of finely milled salt powder is shown to be capable of achieving this goal. During field experiments the use of a sulfur hexafluoride (SF₆) gas tracer to identify the exact seeded cloud volume and to quantify dilution of the seeding agent showed that the seeding agent dilutes to the order of 10⁻¹⁰ of its released concentration in updrafts at a height of ≥1 km above cloud base. This means that the theoretically expected changes in the cloud drop size distribution (DSD) would not be detectable with a cloud droplet spectrometer in a measurement volume collected during the several seconds that the seeded volume is traversed by an aircraft. The actual measurements failed to identify a clear microphysical seeding signature from the burning of hygroscopic flares within the seeded convective clouds. This uncertainty with respect to hygroscopic flare–seeding experiments prompted an experimental and theoretical search for optimal hygroscopic seeding materials. This search culminated in the production of a salt powder having 2–5-μm-diameter particle sizes that are optimal according to model simulations, and can be distributed from a crop duster aircraft. Such particles act as giant cloud condensation nuclei (GCCN). Any potential broadening of the DSD at cloud base by the competition effect (i.e., when the seeded aerosols compete with the natural ambient aerosols for water vapor) occurs when the seeding agent has not been substantially diluted, and hence affects only a very small cloud volume that dilutes quickly. Therefore, the main expected effect of the GCCN is probably to serve as raindrop embryos. The salt powder–seeding method is more productive by two orders of magnitude than the hygroscopic flares in producing GCCN that can initiate rain in clouds with naturally suppressed warm rain processes, because of a combination of change in the particle size distribution and the greater seeding rate that is practical with the powder. Experimental seeding of salt powder in conjunction with the simultaneous release of an SF₆ gas tracer produced strong seeding signatures, indicating that the methodology works as hypothesized. The efficacy of the accelerated warm rain processes in altering rainfall amounts may vary under different conditions, and requires additional research that involves both observations and simulations.

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1. Introduction

This study examines the spreading and dilution of seeded hygroscopic aerosols from two perspectives—their impacts on cloud drop size distribution (DSD) and how that might affect the precipitation forming processes. Before describing the experiments, we briefly state the physical background. Hygroscopic seeding for rain enhancement in convective clouds is aimed at accelerating autoconversion (i.e., the conversion of cloud water to precipitation). This was reviewed extensively by Bruintjes (1999) and Silverman (2003). Three main conceptual models have guided the hygroscopic-seeding experiments.

1) The rain embryo particles: seeding with ultra-giant cloud condensation nuclei (UGCCN; size >10 μm), which serve as embryos for raindrops. This has been done by dumping milled salts from aircraft into the clouds. Because of their large particle sizes, hundreds of kilograms–tons of salts had to be used to have a detectable signal on the rainfall (Braham et al. 1957; Biswas and Dennis 1971; Silverman and Sukarnjanaset 2000).

2) The competition effect: seeding with large CCN (LCCN; diameter near 1 μm) for greater competition for the vapor, decreasing peak super saturation at cloud base, and hence reducing cloud drop number concentrations and broadening the DSD. This causes larger drops that coalesce faster into raindrops (Cooper et al. 1997).

3) The tail effect: seeding with giant CCN (GCCN; between 1 and 10 μm) adds drops to the large tail end of the cloud drop size distribution, and hence accelerates the further widening of the DSD and leads to the formation of raindrops (Segal et al. 2004). Model simulations (Segal et al. 2007) show that seeding with large and giant CCN accelerates the autoconversion, mainly by the tail effect with very little contribution from the competition effect.

Accelerating the autoconversion can produce rain showers from clouds that are too shallow to precipitate naturally, as has been demonstrated by Biswas and Dennis (1971). However, too fast acceleration of autoconversion in clouds with warm bases induces early warm rain without release of the latent heat of freezing, which leads to a greater reduction of rainfall later in the life cycle of the cloud [see Rosenfeld et al. (2008a) and references therein].

2. Considerations for effectiveness of hygroscopic seeding

Hygroscopic seeding of shallow clouds has less potential to add significant amounts of water than deeper clouds because of the smaller fraction of vapor that condenses in these clouds. Therefore, the added water in very shallow clouds can be rather small even if the seeding is very successful in terms of percentage increase. For example, the average rain volume from the experimental units in the warm cloud hygroscopic-seeding experiment in Thailand was $10^5$ m$^3$ (Silverman and Sukarnjanaset 2000), whereas the rain volume within the identically defined experimental units in the cold cloud glaciogenic-seeding experiment in the same area was on the average about $6 \times 10^7$ m$^3$ (Woodley et al. 2003). This rain produced by cumulonimbus convection is a factor of 600 more than by warm rain clouds. Therefore, from a practical standpoint, hygroscopic seeding should address the deep convective clouds that can reach cumulonimbus stature.

This means that the impacts of hygroscopic seeding on the mixed-phase precipitation forming processes must be addressed.

To avoid the possible negative effects of early warm rain (Rosenfeld et al. 2008a) due to hygroscopic seeding of convective clouds that reach the freezing level, hygroscopic seeding of such clouds should not be done in a way that would cause such early warm rainfall. Because seeding with UGCCN creates raindrops already very low in the cloud (Johnson 1982), using UGCCN is probably not a good idea in deep convective clouds. Seeding with LCCN and GCCN appears more appropriate, because it would increase the general population of the cloud drops and widen the DSD that reach the supercooled levels without premature warm rain.

Hygroscopic-seeding experiments have been done in deep continental convective clouds that extend well above the 0°C isotherm level in South Africa (Mather et al. 1997) and Mexico (World Meteorological Organization 2000). Therefore, the hygroscopic-seeding effects on the mixed phase have to be considered. It has been postulated that enhancement of coalescence in supercooled clouds can enhance also the ice precipitation processes (Braham 1964). Ice is produced faster in clouds with larger drops (Hobbs and Rangno 1985). Larger cloud drops are rimed more effectively on ice crystals and graupel, thereby accelerating the growth of these hydrometeors and expediting the conversion from cloud water to ice. The enhanced rate of freezing of clouds with larger cloud- and raindrops was postulated to produce dynamic invigoration of the updraft, prolong the precipitation, and enhance the volumetric rain production of the cloud (Rosenfeld and Woodley 1993). Although the ultimate goal is to enhance rainfall in deep convective mixed-phase clouds, the immediate impact of the seeding must be upon the warm rain process, and thus the present study seeks to understand first the impacts of hygroscopic seeding on the initiation of warm rain before progressing to the more complex mixed-phase clouds.
3. Testing the hygroscopic flares

The apparent success of the cloud-base hygroscopic flare-seeding program was described by Mather and Terblanche (1994), Mather et al. (1997), Bruiniets et al. (1993), Cooper et al. (1997), and Bruiniets (1999). The publications and personal communications prior to 1995 provided the incentive to investigate that methodology in field experiments in west Texas for potential application there.

The broadening of the DSD has been presented as a key link in the conceptual model guiding the hygroscopic flare-seeding experiments. Therefore, the effort of the authors in the Texas Experiment in Augmenting Rainfall through Cloud Seeding (TEXARC) program of 1995 in west Texas concentrated on verifying this link in the conceptual chain. The approach was to release a sulfur hexafluoride (SF$_6$) tracer gas from the seeder aircraft flying at cloud base with the simultaneous burning of “South African” hygroscopic flares, so that the seeded cloud volume could be identified unambiguously. A second monitoring aircraft, instrumented with an array of Particle Measuring Systems (PMS) cloud physics probes and an SF$_6$ detector, was flown in coordination with the seeder aircraft. The monitoring aircraft climbed into the cloud in the region where the plume from the flares was entering cloud base. The seeded cloud volume was detected by the presence of SF$_6$, and the cloud microstructure within the plume was compared to other cloud volumes in which SF$_6$ was not detected.

This method is based on the assumption that the seeding material and nucleated large cloud drops are moving together with the SF$_6$. The terminal fall velocity of a large cloud drop of 40 $\mu$m that might have been induced by the seeding material is 6 cm s$^{-1}$. In a modest updraft of only 2 m s$^{-1}$, for every kilometer that the air ascends, the cloud drop will fall only 30 m relative to that air. With turbulent velocities at least two orders of magnitude greater than the terminal fall velocity, the differences between the trajectories of the SF$_6$ gas, flare particles, and induced large cloud drops can be neglected.

On 20 August 1995, the microphysical impact of the South African hygroscopic flares was tested. The seeder aircraft burned two flares simultaneously while circling under the base of a small convective cloud cluster. The burning time of the flares was 5 min. Cloud base was at 2.5 km at a temperature of 15°C. The seeded cloud cluster was not vigorous and developed only to a height of about 5 km before dissipating. The seeding was accompanied by a release of SF$_6$ gas. This was the only available experimental case where all materials were available and all systems worked properly.

The Forward Scattering Spectrometer Probe (FSSP) on the cloud physics aircraft was used to measure the cloud DSD within and outside the SF$_6$ plume marking the seeded cloud volumes. The SF$_6$ was found in most of the volume of the seeded clouds, in concentrations varying between 10 and 400 parts per trillion (ppt), as shown in Fig. 1. Assuming that the hygroscopic flare particles moved with the SF$_6$ gas, the seeded volumes are clearly identified above the background by the SF$_6$ gas. The cloud-pass integrated drop size distribution is also presented in Fig. 1. The passes with the greatest SF$_6$ concentrations are presented at the various heights, but there is no indication there for the DSD in the seeded clouds to be wider than the unseeded cloud volumes. The cloud drop number concentrations were not smaller in the seeded clouds either. This absence of observed widening of DSD stood in direct contradiction to the results reported by Mather et al. (1997) that TEXARC had tried to replicate. It also differs from the results presented by Ghate et al. (2007) for the seeding of marine stratoscumulus. The differences in the results of those studies versus the results presented here are discussed further in section 8.

One cannot completely exclude that there is actually an effect that went undetected. Only one flight is available for examining the seeding signature from hygroscopic flare so that there is no guarantee that the cloud volume examined was seeded with a high concentration of seeding particles and experienced less dilution in the updraft core, where we expect to see the most significant seeding signature in the DSD. In fact, SF$_6$ concentrations for the hygroscopic flare–seeding case range from 10 to 400 ppt while those for the salt powder–seeding cases to be discussed later range from 50 to 4800 ppt. Additional such experiments are needed to reach more conclusive results. However, if the effect dominated the cloud properties, it is difficult to imagine how it went undetected when tagging the seeded cloud volume with SF$_6$.

4. Some simple estimates of nucleant concentrations and activity

Knowing the rate of seeding, the rate of SF$_6$ dispersion, and the concentration of SF$_6$ detected in cloud, it is possible to make some calculations of the dilution of the seeding material. The small convective cloud on 20 August 1995 was seeded with a total of five 1-kg South African flares during 15 min. The seeding was accompanied by the release of SF$_6$ gas as a tracer at a rate of about 0.7 L s$^{-1}$. Based on the measurements made by the cloud physics aircraft, the following results and conclusions were obtained:

- Detection of SF$_6$ at a concentration of 100 ppt in the cloud means a dilution rate of the seeding material by
about the same factor (i.e., of $10^{-10}$). This means that 1 L of dispersed SF$_6$ gas spread to a volume of 0.01 km$^3$. With five 1-kg flares, each producing $6.4 \times 10^{15}$ particles throughout their burn (integrated from the particle size distribution shown in Fig. 2) during a total seeding time of 15 min (about $10^3$ s), the average seeding rate is $6.4 \times 10^{15} \times 5/900 = 3.5 \times 10^{13}$ such particles per second. In practice, two flares were burned
simultaneously for the first 10 min, and one for the last 5 min or 300 s. Two simultaneous flares translate to \(6.4 \times 10^{15} \times 2/300 = 4.3 \times 10^{13}\) particles per second.

- With a release rate of 0.7 L s\(^{-1}\) for the tracer gas, this means there are \(4.3 \times 10^{13}/0.7 \approx 6 \times 10^{13}\) particles dispersed with each 1 L of gas.
- After dilution by a factor of \(10^{-10}\), the seeding material would expand to a volume of \(0.7/10^{-10} = 7 \times 10^6\) L, or about 0.007 km\(^3\) s\(^{-1}\) of air if diluted homogeneously to 100 ppt. With two flares burning simultaneously, 1 kg of flare material is consumed every 150 s. Therefore, the total volume that 1-kg flare would dilute into is 0.007 km\(^3\) s\(^{-1}\) \(\times\) 150 s \(\approx\) 1 km\(^3\). The seeding started 500 s prior to the monitoring pass and ended a short time before the pass, at 1 km below the monitoring level. This is a comparable dispersion rate to that observed in cumulus clouds over the high plains, as calculated by tracking the concentrations of ice crystals produced by seeding with dry ice. The time to fill an updraft with a 1-km diameter was calculated to be 260 s (Weil et al. 1993).
- With a gas dilution factor of \(10^{-10}\), the concentration of seeded particles would be about \(6 \times 10^{13} \times 10^{-10} = 6000\) L\(^{-1}\) or 6 CCN cm\(^{-3}\). With natural CCN in the hundreds per centimeter cubed, no significant effect on the main part of the DSD can be expected because of such a low concentration of artificial CCN. With 0.05% of the particles \(\geq 2\) \(\mu\)m, only 0.0005 \(\times\) 6 = 0.003 cm\(^{-3}\) or 3 L\(^{-1}\) and can potentially create large cloud drops. This concentration is nearly a factor of 6 smaller that the detection limit of the FSSP of one count per second, which, for a measurement volume of 60 cm\(^3\), is 1000/60 = 17 L\(^{-1}\). This concentration is not detectable using the FSSP for integration times \(<\)6 s.
- As shown earlier, observations of the DSD with the FSSP instrument did not show any clear difference between seeded and nonseeded cloud volumes, in accordance with these simple calculations.

The calculations of Cooper et al. (1997) of the seeding effectiveness of the flare used a concentration of CCN \(> 1\) \(\mu\)m of 1 cm\(^{-3}\). The differences in the flare particle size distributions in Fig. 3b of Cooper et al. (1997) and Fig. 3b here are because Cooper et al. (1997) approximated the distributions to the sum of three lognormal distributions, whereas here we used the exact measured distribution shown in Fig. 1 of Cooper et al. (1997). The total particles per 1-kg flare were \(8 \times 10^{14}\) when integrated on the lognormally parameterized distribution, and \(6.4 \times 10^{15}\) when the exact distribution was integrated. Hence, the approximation caused an undercount by a factor of 8 in the concentrations in the simulations using this parameterization, mainly the small particles. According to the distribution in Fig. 3b, 0.5% of the particles in the South African flare exceed 1 \(\mu\)m. For comparison, diluting this flare particle size distribution to the observed factor of \(10^{-10}\) produces 0.005 \(\times\) 6 = 0.03 cm\(^{-3}\) of CCN \(> 1\) \(\mu\)m. The respective concentrations that Cooper et al. (1997) used are 1 cm\(^{-3}\) of flare particles \(> 1\) \(\mu\)m. Hence, the simulation used a flare aerosol concentration that is 33 times that actually measured in cloud. The calculations of Cooper et al. (1997) have indicated that the fastest enhancement of coalescence should occur because of the competition effect for seeding with 1-\(\mu\)m CCN particles in concentrations of 50–200 cm\(^{-3}\). Such a hypothetical 1-kg flare, assuming a salt density of 2 g cm\(^{-3}\), produces 10\(^{15}\) particles of 1-\(\mu\)m diameter. When dispersed homogeneously to 100 cm\(^{-3}\) it fills a volume of 0.01 km\(^3\). This means that a competition effect, if at all, would occur only in a very small volume at the base of the cloud. The observed concentration of \(< 1\) cm\(^{-3}\) particles \(\geq 1\) \(\mu\)m is not likely to cause much of a competition effect. Therefore, it seems unlikely that the common practice of seeding with these flares is greatly affecting the rainfall in accordance with the conceptual model of the CCN competition effect that has guided these experiments. However, the competition effect is relevant mainly to cloud base, where most cloud drops are nucleated. The competition effect in a small cloud volume on the order of 0.01 km\(^3\) can still occur, but can it lead to a growth of raindrops before getting diluted with the rest of the cloud? The observations of the SF\(_6\) concentrations indicate that the dilution occurs very quickly. This limits the growth...
the parameterized particle distribution of the South African flares (Cooper et al. 1997), with concentrations of 150–1500 cm$^{-3}$. Taking into account the factor of 8 between the approximated and actual particle numbers per flare, the concentration that was used in the simulations of Yin et al. (2000) was from 150/(6/8) = 200 to 1500/(6/8) = 2000 times greater than the concentrations of 6 cm$^{-3}$ actually found in the experimental measurements reported here. The simulated seeding effect was nearly doubled when increasing the concentrations from 150 to 1500 cm$^{-3}$. This means that a concentration that is lower by two orders of magnitude would have a very small effect in this kind of simulation.

It is interesting to note that the greatest indicated rain enhancement in the simulations of Yin et al. (2000) was obtained when seeding above cloud base, where the competition effect cannot occur, leaving only the GCCN effect as a cause for the added rainfall. This result was obtained despite a modest decrease in cloud-base drop concentrations when simulated seeding occurred below cloud base.

Furthermore, the more recent simulations of Segal et al. (2007) indicate that the previous studies have overestimated the efficiency of the competition effect. This is further supported observationally by the lack of indicated increase in the cloud drop effective radius in the seeded cloud volumes shown in Fig. 1 and later in this study. The competition effect, by reducing cloud drop number concentrations, should have increased the drop size in the main part of the distribution along with the drops in the tail, and hence increased the cloud drop effective radius.

The remaining possibility of the hygroscopic flares influencing the precipitation would seem to be mostly by their activity as GCCN. About 0.05% of the particles produced by the flare are GCCN (i.e., >2 μm). According to the SF$_6$-based dilution observations, this would give about 0.0005 × 6000 = 3 GCCN particles per liter. Such an effect should be manifested in the large-size tail of the DSD that is below the detection limit of the FSSP. Furthermore, GCCN were calculated to be >5 μm for serving as efficient embryos of raindrops (Ivanova et al. 1977). The concentration of such CCN is about 10$^{-5}$ of the particles produced by the flare, or about $10^{-5} \times 6000 = 0.06$ L$^{-1}$ or 6 × 10$^{-5}$ cm$^{-3}$. According to Fig. 6 of Segal et al. (2007), a minimum concentration of 0.025 cm$^{-3}$ such particles is required for a noticeable increase in the warm rain production in a rising cloud parcel under typical conditions in Texas. Hence, the observed concentration was a factor of 400 smaller than necessary for producing warm rain in the ascending cloud parcel according to the calculation of Segal et al. (2007). The significance of these results with respect to past hygroscopic-seeding experiments is discussed in section 8.

of the raindrops that would be potentially released from this volume to the rest of the cloud.

Other simulations of hygroscopic seeding that resulted in an indicated large increase in rainfall used a much greater mass of hygroscopic material than is technically possible with flare technology. Reisin et al. (1996) simulated 450 kg of salt with particle diameters of 12 μm for nearly double the total rainfall. Yin et al. (2000) used

![Diagram](http://journals.ametsoc.org/doi/abs/10.1175/2010JAMC2307.1)
5. Developing an optimal hygroscopic-seeding material

The considerations in the previous section indicate that the reported rainfall enhancements by hygroscopic flares have been produced by the large end tail of the CCN distribution from the flares. Rain enhancement by GCCN is a possibility. However, the probable numbers of GCCN produced by the flares are still far less than optimal. Simulations of the effect of particle size on the warm rain forming processes, using an explicit microphysics cloud parcel model with 2000 size bins, revealed that the sub-micron particles actually suppress the warm rain forming processes (Segal et al. 2004). Larger hygroscopic particles produce raindrops faster, but fewer of these larger particles are available for a given amount of seeding material that can be dispersed into the cloud. This would produce a smaller number of raindrops, as simulated by Segal et al. (2004), which is an undesirable result. This means that an optimum particle size must exist for a maximum rate of conversion of cloud drops into warm rain. This optimal size was found to have a flat maximum diameter between 2 and 5 \( \mu \)m of NaCl particles. The effectiveness of the flare particles falls sharply out of these bounds (Segal et al. 2004). This optimal size range is marked by the two vertical bars in Fig. 3. According to Fig. 3, only 7% of the mass of the South African flare produces hygroscopic particles of the optimal size. Indeed, the simulations of this flare were found to be enhancing warm rain much less effectively than the same mass produced by the same size range (Segal et al. 2004). The next obvious step was a search to produce such a seeding agent. Figure 4 shows the particle size resulting from fine spraying of concentrated brine from the evaporation ponds of the Dead Sea. The spray was made by an agricultural sprayer aircraft operating at a pressure of 80 bars using the finest available nozzles. The drops were measured with a Cloud, Aerosol and Precipitation Spectrometer (CAPS) probe being flown on an aircraft behind the sprayer in a very dry atmosphere where hygroscopic growth of the particles was not possible. Almost all (99%) of the spray mass was above the upper bound of the optimal size of 5 \( \mu \)m. The size of 2–5 \( \mu \)m proved to be too small for spray technology, so again the search continued for another method. The solution was found by producing fine NaCl powders of the specified size, with additives that prevent coagulation of the fine salt grains. Figure 5a shows the particle size distribution as measured in the factory. The material is dispersed from an agricultural crop duster, shown in Fig. 6. Most (58%) of the mass is within the optimal size range of 2–5 \( \mu \)m, and none is in the detrimental size range of <1 \( \mu \)m. Field measurements of the particle size that actually dispersed from the aircraft showed enlargement of the median volume particle size from the factory size of 4–5.5 \( \mu \)m in the field (see Fig. 5b). These measurements were made with an optical aerosol counter on the ground under a low-level pass of the seeder aircraft. The number of giant CCN > 2 \( \mu \)m produced by a given amount of the salt power is greater by a factor of about 3 than the number produced by the same mass of South African hygroscopic flare, mainly because of smaller concentrations of particles >5 \( \mu \)m. That ratio increases to 20 for particles >5 \( \mu \)m. A powder-seeding rate of 10 kg min\(^{-1}\) is 20 times the burning rate of two simultaneously burning hygroscopic 1-kg flares, which is the common practice. This sends to the air 3 \( \times \) 20 = 60 times greater number concentrations of GCCN that can enlarge the tail of the DSD and enhance the coalescence rate, or serve directly as raindrop embryos. The respective factor for particles >5 \( \mu \)m is 20 \( \times \) 20 = 400. This concentration is double the threshold for a noticeable enhancement of the rain in the parcel model of Segal et al. (2007).

6. Design of the field experiment to test the salt powder

The salt powder seeding was first tested in northern Israel during the late winter of 2004. It did widen the DSD above cloud base, but it was added to an already existing tail of up to 35-\( \mu \)m drops. The natural tail was likely caused by sea-spray aerosols that were carried from the
Mediterranean Sea just 10–30 km to the west of the measured clouds. These mixed results demonstrated the need to repeat the salt powder–seeding experiment with three important changes: 1) performing the experiment in a region far from the sea, where sea salt cannot be a major confounding factor, 2) identifying the seeded cloud volume unambiguously with an SF$_6$ gas tracer, and 3) development of a real-time display that shows the flight scientist and the pilots of the monitoring cloud physics aircraft the location of the seeder aircraft, its back trajectory, and its seeding coordinates drifting with the wind.

Thus, the experiment was planned for the semiarid region of west Texas. The necessary seeder tracking system was developed and installed in the Seeding Operations and Atmospheric Research (SOAR) aircraft (Rosenfeld et al. 2008b). The SF$_6$ monitoring instrument was installed in the SOAR cloud physics aircraft. The salt powder was loaded on an agricultural aircraft for cloud seeding through a spreading device similar to that used in Israel. The SF$_6$ gas also was installed on this aircraft. The experimental seeding flights took place in the early summer of 2005. There were seven salt experiments with monitoring by the cloud physics aircraft. During these experiments the total expenditures of sized salt and SF$_6$ gas were 556.4 and 59.2 kg, respectively. The SF$_6$ and salt powder were dispersed simultaneously. This means a ratio of about 1 kg of SF$_6$ marking each 10 kg of salt powder. The SF$_6$ gas was detected by the SF$_6$ detector on the cloud physics aircraft during portions of four out of the seven
experiments and an apparent microphysical seeding signature was noted in the strongest updraft cases, especially on 25 and 31 May 2005.

The number concentrations of salt powder supermicron particles after a dilution to 100 ppt are calculated based on the practice of releasing 10 kg of salt powder marked by 1 kg of SF$_6$. This allows us to evaluate the expected impact of the seeding agent on the measured cloud volume. For example, if the seeding is changing the DSD toward larger drops, the expected change cannot involve a larger number of drops than the number of hygroscopic particles can produce under the best assumptions.

Based on the particle size distribution shown in Fig. 5, this seeding rate after dilution of $10^{-10}$ amounts to 11 particles L$^{-1}$. With 80% of the powder particles >2 $\mu$m (90% by mass), 9 particles L$^{-1}$ of that size can be expected. At a cloud droplet probe (CDP) sampling volume of about 20 cm$^3$ s$^{-1}$ (assuming an airspeed of 80 m s$^{-1}$), there is a probability of detecting about $\frac{1}{6}$ particle s$^{-1}$. This is below the detection limit of the CDP. The sample volume of the cloud imaging probe (CIP) for small raindrops of 0.3–0.4-mm diameter is about 8 L s$^{-1}$ (assuming an airspeed of 80 m s$^{-1}$). This factor of 400 greater sample volume of the CIP with respect to the CDP allows the detection of the impact of 60 salt particles >2 $\mu$m s$^{-1}$, or about 1 salt particle >8 $\mu$m s$^{-1}$. This means that drizzle drops that may develop because of the hygroscopic seeding can be detected by the CIP at a dilution factor of 10$^{-10}$, which is indicated by an SF$_6$ concentration of 100 ppt. These calculations show that the hypothesized impact of the salt seeding should be detectable by the CIP at an SF$_6$ dilution factor of $10^{-10}$, whereas the hygroscopic flares produce concentrations that are below the detection limit.

The SF$_6$ detector was less sensitive and stable than that used in the 1995 TEXARC experiment. Its measurements were in units of millivolts. Each millivolt corresponded to about 12 ppt of SF$_6$, which was found only in small portions of the cloud passes at heights of about 1 km above cloud base. The SF$_6$ spread into much of the cloud volume at 3 km above base about 20–30 min after seeding.

The tracking system was used for directing and coordinating the measuring and seeding airplanes. The GPS coordinates of the seeder were transmitted to the measuring aircraft, and plotted on its computer screen (the yellow track in Fig. 7). The track of the measuring aircraft was plotted on the same screen (the green track in Fig. 7). The coordinates of the tracks of both airplanes were corrected for the wind drift, based on the inferred wind from the air data probe on the monitoring aircraft. An inset graph of the measured liquid water content (LWC) and SF$_6$ was displayed (bottom right of Fig. 7). When the SF$_6$ level exceeded a threshold that was set by the flight scientist (shown as the horizontal white line on the SF$_6$ inset plot), the green line track of the measuring aircraft was highlighted with red. The background of the line was colored with blue, where the width was proportional to the logarithm of the LWC to display the location of the cloud water with respect to the flight track. This system provided the flight scientist with the ability to find the seeding plume in the cloud and then navigate back into it while maneuvering in cloud, as shown in Fig. 7.

7. Results of the field testing of the salt powder

Examples of the salt powder–seeding signatures and their evolution are shown in Figs. 8–11. Figure 8 shows a pass 1 km above cloud base into two adjacent cloud segments with identical LWC. It is the last cloud (right-most cloud) in the LWC inset plot of Fig. 7a. Segment a of that cloud was seeded and segment b shown in Fig. 8 was not seeded, as tagged by the SF$_6$ readings (Fig. 8). The nonseeded cloud had no large droplets >31 $\mu$m (Fig. 9). The seeded cloud (Fig. 10) was identified by an SF$_6$ level of 70 mV above the baseline, which translates to about 840 ppt. This relatively high concentration means that the seeding signature of a possible GCCN effect should be observable by both CDP and CIP instruments. This was manifested in Fig. 10 as a tail of larger cloud and small drizzle drops. About one large drop per second is evident in the tail of the DSD. If this drop concentration is induced by seeding, the particle size that caused it can be estimated. Taking into account the dilution of $8.4 \times 10^{-10}$, which can be approximated to $10^{-9}$, 10 kg of salt powder dispersed into about 10$^9$ L of cloud volume. In such dilution there is about one salt particle >3 $\mu$m in the CDP-measured volume of 20 cm$^{-3}$. This is roughly the same concentration of cloud drops >30 $\mu$m in the seeded cloud segment. The aircraft returned to the seeded cloud volume in the next pass 2 min later (see Fig. 11) and found already that the tail of the large cloud drops was reduced, and instead drizzle and small raindrops at smaller concentrations appeared. The continued dispersion of the SF$_6$ in the cloud was documented for the subsequent 20 min (see Figs. 7b, 7c). The gas filled initially only small portions of the seeded clouds, in agreement with the observations of Stith et al. (1986), who concluded that the seeding agent spreads mainly as filaments in the cloud. The gas-tagged portions of the cloud contained what in many cases appeared to be anomalously large cloud droplets and some raindrops relative to nearby regions where the gas was not found. However, because of cloud maturation and the spreading of the SF$_6$ it was no longer possible to ascribe that rain unambiguously to the seeding.
This case (the second case on 31 May 2005) was the best of the four cases in which SF$_6$ plumes were identified in the seeded clouds. The monitoring showed that it was well seeded, and it did not reach the vigor that precluded repeated penetrations in the experimental units on 25 May and for the first case on 31 May. The case of 30 May 2005 is shown in Fig. 12. This figure shows a conspicuous increase of the maximum cloud drop size within the seeded cloud volume, as identified by the sharp increase in the SF$_6$ concentration. However, no discernible change is evident in the cloud drop effective radius. The same is indicated in the case shown in Fig. 8. This type of seeding signature means that the seeded particles acted to create larger drops by extending the tail of the distribution, but did not greatly affect its main body. This is consistent with the hygroscopic particles acting as GCCN, but not producing a discernible competition effect.

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The other two cases of the TEXARC experiment had similar microphysical seeding signatures. The correspondence between the calculated hygroscopic particles after
dilution and the added large cloud droplets supports the suggestion that the salt particles act mainly by being embryos for raindrops, and not appreciably by the competition effect.

8. Significance of the results with respect to past flare-seeding experiments

The repeated observed rate of dilution at 1 km above cloud base by a factor of about $10^{-10}$ can all by itself be used for making some useful calculations, which show that the concentrations of flare particles are too small for producing significant rain enhancement in cloud simulations. These estimates must be considered with respect to the reported enhanced precipitation in the hygroscopic-seeding experiments of deep continental convective clouds in South Africa (Mather et al. 1997) and Mexico (World Meteorological Organization 2000).

Suggestions that the rain enhancement effect of hygroscopic flares is produced by a broadening of the cloud-base spectrum due to the competition effect (Mather et al. 1997; Cooper et al. 1997) are consistent with the observations by Mather et al. (1997) of the broadening of the cloud-base DSD in the area where the seeding material was observed to be entering the cloud base, and where the seeding material is still highly concentrated in a very small cloud volume. Mather et al. (1997) reported that they visually tracked the plume from two simultaneously burning flares on the seeder aircraft into the cloud, and documented the effect 200 m above cloud base. The authors of the present study could no longer detect the seeding signature after dilution that would fill about 1 km$^3$ at 1 km above cloud base, despite the positive identification by the SF$_6$ tracer. Any competition effect that might have occurred there on broadening the DSD was diluted below the detection limit. The small extent and short lifetime of the cloud volume that received the concentrated smoke from the flare and possibly broadened its

![Fig. 8. A seeded cloud pass in Texas on 31 May 2005, 1000 m above cloud base and 500 s after the start of seeding. The LWC is marked by the blue line (10 gm$^{-3}$). The CDP-measured largest cloud drop (µm) is shown by the green line. The cloud droplet effective radius is shown by the purple line. The SF$_6$ readings in millivolts are shown by the red line. The peak translates to a concentration of about 840 ppt. The seeded volume is identified by the high SF$_6$ readings, and seen by the larger maximal size of cloud droplets. Two LWC comparable seeded (segment a) and not-seeded (segment b) cloud segments are marked by the respective time bars. The DSDs for these cloud segments are show in Figs. 9 and 10.](image)

![Fig. 9. DSD for the nonseeded volume (segment b in Fig. 8) in the cloud pass shown in Fig. 8. No droplets >31 µm were observed. Each line represents DSD during 1 s of flight path, at the time and altitude (m) shown in the legend as hhmmss.aaaa. (top) The CDP measurements. (bottom) The combined CDP and CIP distributions. Note the logarithmic scales for the drop concentrations per drop size.](image)
DSD probably do not allow the development of raindrops before it gets diluted. While the competition effect vanishes with the dilution, the resultant hydrometeors formed by the GCCN just get diluted while continuing to grow.

Given the above considerations, we try to understand how the reported positive results of the seeding experiments in South Africa and Mexico could be possibly related to the seeding. If the statistical results are not just by chance, the remaining alternative is that the GCCN impacts clouds that extend well above the 0°C isotherm level. In such clouds GCCN can create supercooled raindrops that freeze to graupel and hailstones, which fall and melt into large raindrops. With the most favorable assumption, flare seeding with 6 m$^{-3}$ GCCN $>5 \mu$m that would grow into raindrops of 5-mm size, there would be as many such drops per meter cubed. The radar reflectivity factor $Z$ (mm$^6$ m$^{-3}$) is defined as $Z = \Sigma N_i D_i^6$, where $N_i$ is the drop concentrations (m$^{-3}$) and $D_i$ is the drop diameter (mm) for the $i$th drop within one cubic meter. According to the definition of $Z$ and terminal fall speed of the raindrops, this would create a reflectivity of 49.7 dBZ, and a rain rate of 13 mm h$^{-1}$, originating from 1 km$^3$ of seeded cloud volume per 1-kg flare mass. With the operationally used $Z$-$R$ relation of $Z = 300R^{1.4}$ for the U.S. Weather Radar Network, where $R$ is rain intensity (mm h$^{-1}$), this would result in a radar-inferred $R$ of 60 mm h$^{-1}$. Furthermore, when only a few GCCN create isolated raindrops, they do not collide and break up until reaching 8 mm (Beard et al. 1986), producing exceedingly high reflectivities with little rain intensity.

These calculations probably provide the upper limit of the effects. In reality, the efficiency of growth of all GCCN $>5 \mu$m into large raindrops is $<<1$, because only a fraction of the flare material gets ingested into the updraft and experiences these highly idealized conditions.
The flare must be burned continuously for 4–5 min after ignition, while the aircraft can stay in the updraft only a fraction of that time.

If indeed the seeding effects are dominated by the GCCN rain embryo effect, hygroscopic seeding could produce overestimated radar-indicated rain rates. This is in agreement with the overestimate of the radar-indicated seeding effect on rainfall by as much as a factor of 3, as simulated by Levin et al. (1999) and Yin et al. (1999). This is also consistent with the observations of Woodley and Rosenfeld (1999), who reported that clouds that were seeded with hygroscopic flares had the greatest radar reflectivity with respect to nearby comparable clouds, but with a much less visibly dense rain shaft. The possibility of radar rainfall overestimates of the seeded clouds was not investigated for the South Africa and Mexico experiments, but it does not mean that such effects did not exist there. Recently, flare seeding in marine stratocumulus (Sc) was reported to broaden the DSD (Ghate et al. 2007). The seeding was done by six flares burning simultaneously while flying within the Sc, midway between cloud base and cloud top. Because the competition effect occurs at the cloud-base drop nucleation level, a competition effect could not have been possible in this experiment, and all the observed effects are due to the GCCN effect. The observed concentrations of aerosols ascribed to the flares about 10 min after seeding were 1000–2000 cm$^{-3}$. This is larger by a factor of 250 than the calculated concentrations in the present study. When taking into account the triple rate of release, the dilution is about 80 times less in the stable marine Sc off the coast of California than in the highly turbulent convective clouds over Texas. This difference in dispersion rate is supported by the observations that ship tracks in marine Sc off the coast of California on average spread at a rate of 1.4 km h$^{-1}$ (Durkee et al. 2000), or 0.39 m s$^{-1}$, as compared with a dispersion rate of 2 m s$^{-1}$ in the high plains convective clouds (Weil et al. 1993). The volume dispersion is approximately the cube of the linear rate. Hence, the ratio of the volume dispersion rate is given by $(2/0.39)^3 = 134$. Given the crude calculations here, this is in general agreement with the ratio of 80 calculated above.

Assuming that the flare used by Ghate et al. (2007) had the same particle size distribution as the South African flare, this 250-fold greater concentration of the flare material would mean a concentration of GCCN $> 2 \mu m$ of 750 L$^{-1}$, or 0.75 cm$^{-3}$. This is already well within the detection limit of the cloud drop spectrometer. This is in agreement with the conclusion of Ghate et al. (2007) that the GCCN dominated the cloud response. This underlines the necessity of increasing the hygroscopic seeding rate by a factor of several hundred for obtaining a significant microphysical response in the cloud.

In summary for this section, given all these considerations, it is difficult to establish the physical basis for the radar-reported rain enhancement due to cloud seeding with hygroscopic flares (Mather et al. 1997; Bruintjes et al. 2001). Our measurements and calculations suggest the possibility that the indicated enhancement was caused by GCCN that produced small concentrations of large hydrometeors,

FIG. 12. As in Fig. 8, but for 31 Jul 2005, 600 m above cloud base. The seeding started at 2048:45 UTC and continued through the time of monitoring. Note the large increase in the maximum cloud droplet size coinciding with the SF$_6$ peak, which is not reflected in the cloud drop effective radius. This indicates that seeding affected cloud DSD by creating just a few large drops without changing the main body of the distribution much, hence not producing a discernible competition effect.
which had a large impact on the radar reflectivity that was manifested in a much smaller increase in rain rates than indicated when applying a fixed Z–R relation.

These considerations prompted an experimental and theoretical search for optimal hygroscopic-seeding materials. It is now possible to produce inexpensively a salt powder agent, having a 2–5-μm diameter particle size, that is optimal according to model simulations, and to distribute the agent using crop duster aircraft. It appears that the main mechanism for expected rain enhancement is the GCCN serving as drizzle and subsequently as raindrop embryos, and not the competition effect. This seeding method is more effective by two orders of magnitude than the hygroscopic flares in producing GCCN that are responsible for initiating rain in clouds with naturally suppressed warm rain processes. This takes into account both the differences in the particle size distributions and the 20 times greater seeding rate of the salt powder with respect to the commonly practiced seeding rate with hygroscopic flares. Experimental seeding, aided by an SF6 gas tracer, found strong microphysical seeding signatures indicating that the methodology works as hypothesized. The significance of the accelerated warm rain processes in terms of changing rainfall amounts may vary in different conditions, and require additional research that involves both observations and simulations.

10. Concluding remarks

The use of increasingly sophisticated numerical models and instrumentation has made it obvious that hygroscopic seeding for precipitation enhancement is far more complex than might have been envisioned originally. Models indicate that the seeding outcome for individual clouds and for groups of convective clouds is strongly dependent on the sizes and amounts of the dispersed nucleant and on the time the seeding action is taken. Initiating precipitation too early in the convective cycle can sometimes result in less precipitation than if no seeding were undertaken at all (Rosenfeld et al. 2008a). With such complexity the day is coming when seeding will be guided in real time by a combination of cloud physics measurements and validated numerical models. Demonstrating such model capabilities should be a focus of weather modification research programs.

This paper has called into question current conventional wisdom with respect to hygroscopic-seeding experiments. Nucleant sizes and amounts are major considerations. According to model simulations, the CCN aerosols can be so small (<0.5-μm diameter) that they suppress precipitation or they can be too large (>5-μm diameter), resulting in early precipitation that truncates the convective cycle. The results of recent (since 1990) hygroscopic-seeding experiments would appear to still be open to interpretation. Although they produced apparent increases in radar-estimated rainfall, the attendant observations are not consistent with the “competition effect” conceptual model. The results presented here suggest that the tail of the drop size distribution is the key to the apparent seeding effect. If this is the case, it creates problems for the past radar evaluation of the hygroscopic-seeding experiments. Not only do the usual uncertainties apply with respect to radar estimation of rainfall, the problem is exacerbated by the likelihood that hygroscopic seeding alters the drop size distribution of the seeded clouds, producing overestimates of the rainfall and, therefore, the effect of seeding. Although this casts serious doubt on the reported rainfall increases, there is other evidence for the effects of seeding. The reported increased longevity of the seeded clouds is the most intriguing apparent effect that has not likely been compromised by the increased raindrop sizes in the seeded clouds. How flare seeding might cause such an effect remains an open question.

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