

#### 4.4 THE SEARCH FOR THE OPTIMAL SIZE OF HYGROSCOPIC SEEDING PARTICLES

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##### 1. ABSTRACT

It is well known that large concentrations of small aerosols, such as produced by smoke and air pollution, can suppress coalescence and precipitation. Our observations have shown that when giant salt CCN (e.g., sea spray) restore the precipitation from such polluted clouds (Rosenfeld et al., 2002).

Aircraft and satellite microphysical observations in Israel and adjacent areas show that hygroscopic seeding of clouds occurs naturally by sea spray, but the effect diminishes with distance inland.

Cloud simulations and observations show that hygroscopic seeding is most effective with 2-4  $\mu\text{m}$  diameter particles. This is larger than can be produced currently with hygroscopic flares, but smaller than can be produced with other methods, such as spray seeding. The calculations also show that the mass that needs to be dispersed for such "optimal" seeding is larger than can be delivered by current flare technology, but starts to be feasible with spray seeding, and most practical with powder seeding. The results from a couple of salt spray experiments in Israel showed early formation of large raindrops and somewhat wider cloud drop size distribution in the seeded clouds, in accordance with the theoretical expectations. The small concentrations of the seeding-induced raindrops suggest that the concentrations of seeded particles were too small. We realized that the spray system produced particles that are much larger than the optimal size, increasing the number concentrations can be achieved by getting closer to the optimal size for hygroscopic seeding, using smaller particles in the form of salt powder.

Field tests in Israel have specified the size distribution of the manufactured powder, which was found to be in the desired range specified by the models.

Preliminary results with powder salt seeding in Israel and more recently in Texas have shown that the seeded clouds were affected by the salt powder, producing high drizzle concentration and rain drops in the seeded cloud.

##### 2. BACKGROUND

Artificial hygroscopic seeding has been done in a wide range of particle sizes, but the concentrations were constrained by the amount

of material that could be carried by the aircraft and by its rate of dispersion into the clouds (Silverman and Sukarnjanaset, 2000). Hygroscopic flares appeared to be an attractive option (Mather et al., 1997, Bruintjes, 1999), but the problem with such technique is that it produce mostly sub-micron large CCN (Cooper et al., 1997). Their calculations showed that the sub-micron particles of the South African hygroscopic flare did not contribute to the enhanced coalescence. If this is true, only about 30% of the flare mass was in particles that were sufficiently large for enhancing the coalescence and precipitation.

Further calculations (Segal et al., 2004) showed that a wide range of optimal diameter exists between 2 and 5  $\mu\text{m}$ . Because dispersion of such particles at large seeding rates is not yet technologically feasible with flares, the next best thing that we could think of was to develop a new seeding method that imitates what nature does with sea spray (Rosenfeld et al., 2002).

##### 3. SPRAY SEEDING

An agricultural spray aircraft was fitted with high-pressure pump and nozzles, designed for high capacity spraying of small drops. Concentrated Dead Sea brine, containing 500 g salt  $\text{l}^{-1}$ , was sprayed at a rate of 12 liters per minute. The size distribution of the spray was measured with the Israeli research aircraft flying in the plume of the seeder aircraft at a distance covered by less than 2 seconds of flight time. In view of the short distance and dry atmosphere (~60% relative humidity), the sprayed particles had not changed much between dispersion and measurement.

The spray seeding was tested in Israeli clouds on several case studies. Here we describe one of the cases. On 25 February 2002 experimental spray seeding took place in northern Israel over the western lower Galilee. The seeder circled just below cloud base at 2500 feet and a cloud physics aircraft penetrating the seeded and adjacent similar unseeded clouds at various levels above cloud base. The distance in time and space from the spray seeding coordinates was used to identify the potentially seeded cloud volumes. The cloud physics aircraft was equipped with a CAS cloud droplet and aerosol spectrometer (0.3-50  $\mu\text{m}$ ), a precipitation particle imager OAP-2DC (25-800  $\mu\text{m}$ ), and KING and DMT hot wire liquid water content probes. In addition, temperature, pressure and GPS navigation parameters were recorded. Cloud tops reached 9000 feet, at  $-3.5^{\circ}\text{C}$ . Very light rain showers

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occurred from these clouds from all-warm-rain processes. Mean cloud pass droplet concentrations  $> 2 \mu\text{m}$  were about  $450 \text{ cm}^{-3}$  for both unseeded and seeded clouds. The proximity to the sea (10 to 40 km) likely added a background of natural sea spray particles at unknown concentration. The surface wind over sea was about 15 knots. From visual inspection, white caps did not occur at the sea surface of the coastal waters. The near cloud-base spectra of the seeded and unseeded clouds were similar, but they deviated with altitude such that the DSD of the seeded clouds widened somewhat faster than the unseeded clouds (see Figure 1). Raindrops of 0.4 to 0.8 mm diameter appeared in the seeded clouds at 7900 feet, starting at 10 minutes after initial seeding. After additional 7 minutes the seeder aircraft reported the first raindrops falling through cloud base. These raindrops became “large” (by the appearance of the impacts on the seeded windshield of the seeder aircraft) after additional 4 minutes. The seeding was terminated after 25 minutes because the cloud drifted out of the area where air traffic control allowed flight. The unseeded clouds developed small raindrops of about 200 micron just at their tops near 9000 feet.

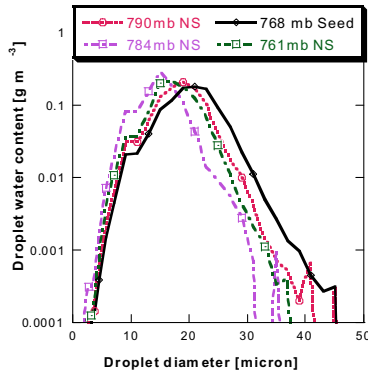


Figure 1: Normalized drop size distributions from full cloud passes of spray-seeded (black bold line) and not seeded (colored broken lines), done at height of 7700 feet (5200 feet above base), on 25 February 2002 in north Israel.

#### 4. MODEL SIMULATION

The NaCl equivalent dry particle size distribution of the spray is given in Figure 2, along with the particle size distribution of the SA hygroscopic flares and the background concentrations that were used by Cooper et al. (1997) in their model simulations. The simulated cloud had a base height of 700 m, a temperature of  $21^\circ\text{C}$ , and a cloud base updraft of  $2 \text{ m s}^{-1}$ . The updraft accelerated to about  $10 \text{ ms}^{-1}$  near the 5 km level and there leveled off. The model-simulated rainfall flux (with respect

to the rising air) and the drop effective radii are presented in Figures 3 and 4, respectively.

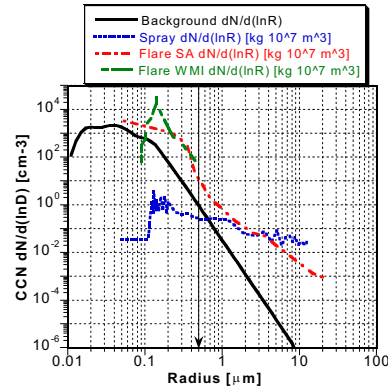


Figure 2: Size distribution of CCN in dry NaCl equivalent, for a spray of Dead Sea brine (blue), SA hygroscopic flares (red), a hypothetical flare that produces only sub-micron particles (green), and the background CCN (black). The concentrations are calculated for 1 kg of dry material diluted in  $10^7 \text{ m}^3$  of air.

The calculation assumed that the dry mass of 1-kg seeded particles was diluted in  $10^6$ ,  $10^7$  or  $10^8 \text{ m}^3$  of air well below cloud base. The results show the following:

Background aerosols started to develop precipitation ( $R > 2 \text{ mm/hr}$ ) at a height of 5.5 km. Seeding with the South African flares at a dilution of 1-kg in  $10^6 \text{ m}^3$  lowered the rain onset slightly from 5.5 to 5.3 km. Increasing the concentration of the flare effluent to 1-kg in  $10^7 \text{ m}^3$  had a much larger effect and lowered the rainfall onset to 4700 m. Further increase of the concentration to 1-kg in  $10^8 \text{ m}^3$  did not result in additional enhancement of the precipitation. This means that the concentration of 1-kg in  $10^7 \text{ m}^3$  is already near the optimum.

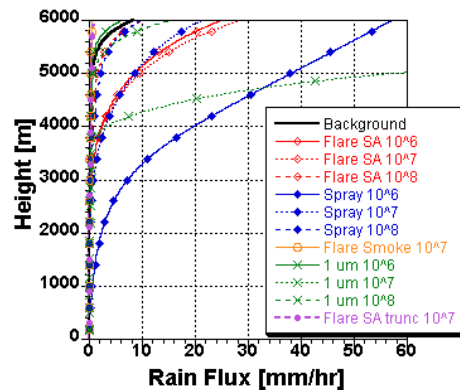


Figure 3: Simulated rain flux for clouds containing the background CCN as assumed for the simulations by Cooper and Bruintjes

(1997) and in addition, the dry NaCl equivalent of spray of Dead Sea brine, South African hygroscopic flares, the SA flare with particle truncated at diameters  $> 1\text{-}\mu\text{m}$ , and a hypothetical flare that produces only 1-micron particles.

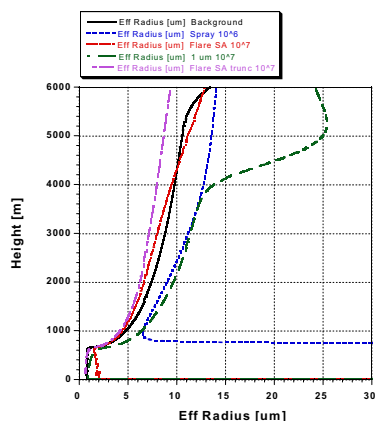


Figure 4: Simulated effective radius of cloud droplets, for clouds containing the background CCN (black bold) as assumed for the simulations by Cooper et al. (1997) and in addition, the dry NaCl equivalent of a spray of Dead Sea brine (blue), SA hygroscopic flares (red), and the South African flare with particles truncated at diameters  $> 1\text{-}\mu\text{m}$  (green) and a hypothetical flare that produces only 1-micron particles (purple).

Truncating all the particles with diameters  $> 1\text{-}\mu\text{m}$  at the “optimal” concentration of  $1\text{-kg in } 10^7 \text{ m}^3$  resulted in a strong suppression of the precipitation compared to the background. Its effect was similar to the hypothetical smoke flare. According to Figure 4 the truncated distribution created a cloud with reduced droplet sizes with respect to the background. Even the “optimal” concentration of the untruncated flare reduced the droplet size in the lower part of the cloud. This suggests that the smaller flare particles still nucleated and that they actually caused the cloud to produce more droplets at its base. The raindrop-embryo effect, produced by the super-micron flare particles, was mainly responsible for the rain enhancement, and it worked against the suppressing effect of the smaller particles. If sub-micron particles do not contribute to the apparent seeding effect, it is worthwhile to see what would be the result for a flare with a narrow distribution near  $1\text{-}\mu\text{m}$ , which Cooper et al., (1997) have shown would produce optimal results. Here again, at a dilution of  $1\text{-kg in } 10^8 \text{ m}^3$  only a slightly positive effect was noted, but it was dramatically increased when the concentration increased to  $1\text{-kg in } 10^7 \text{ m}^3$ . According to Figure 3, the rainfall enhancement occurred due to the size increase of the cloud-base droplets, which coalesced into drizzle and then continued their growth to raindrops at a height of 3.9 km. Increasing the concentration

further, resulted in a return to near background precipitation, probably due to excessive concentrations of  $1\text{-}\mu\text{m}$  CCN, which already constituted the bulk of the cloud droplets.

Increasing the spray concentration to  $1\text{-kg in } 10^6 \text{ m}^3$  to simulate the real situation enhanced the rain flux further, even beyond the composition of the “optimal flare” of  $1\text{-}\mu\text{m}$  particles, at least initially in the clouds. The rain started forming at a height of  $< 2 \text{ km}$ , but its development with height was slower than for the “optimal flare” of  $1\text{-}\mu\text{m}$  particles. The large advantage of the  $1\text{-}\mu\text{m}$  “optimal” device is that it converts most cloud water quickly to drizzle and rainfall between 4 and 5 km, leading to very efficient precipitation in clouds that exceed this depth, whereas the spray seeding would leave most of the water still in the form of cloud droplets. The large number of drizzle particles can also recalculate into the cloud more easily and spread the seeding effect to cloud portions that are not seeded directly. However, dispersion of  $1\text{-}\mu\text{m}$  salt particles in sufficient quantities for cloud seeding is still an unresolved technological challenge. Furthermore,  $1 \text{ }\mu\text{m}$  is still smaller than optimal, as shown next.

## 5. SALT POWDER SEEDING

Segal et al. (2004) tested a large combination of sizes and concentrations for the maritime, Mediterranean and Texas clouds using the parcel model to obtain an answer to this question. They showed that a wide optimal size-range occurs between 2 and 5 microns diameter. This optimal size enhances rainfall even in maritime clouds in contrast to the action of even the best of the hygroscopic flares. The required mass was only  $1 \text{ kg to } 10^8 \text{ to } 10^9 \text{ m}^3$ . No over-seeding is possible.

### 5.1. Salt Powder Field Experiment

The next challenge was the production of such particles. This cant be done currently with flare and spray technologies. We found the solution with a world-patented method for processing salt (NaCl) powder to these sizes, with anti-clumping additives. An example of such material is shown if Figures 5-7.

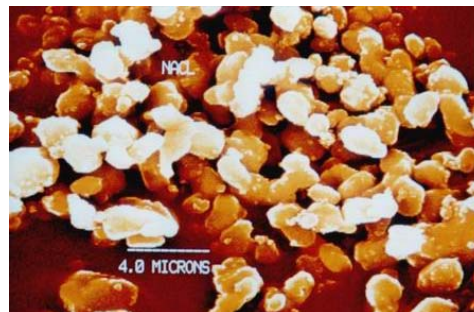


Figure 5: Scanning electron microscope image of the salt powder.

Hundreds kg of salt were milled to specification of diameter between 2 and 5  $\mu\text{m}$  and field tested by dispersing from an agricultural duster aircraft. The experiment took place on 24 December 2003 in Megido airstrip at northern Israel, at 60% relative humidity. The deliquescence point of NaCl is 75%. The airplane made low dusting passes over the measuring instruments (see example of the dispersal powder in Figure 7), which consisted of a Welas optical aerosol spectrometer (see Figure 6) and filters. The dispersion rate was about  $7.5 \text{ kg km}^{-1}$ . If this material were to disperse uniformly in  $1 \text{ km}^3$  of cloudy air, the salt concentration would be nearly optimal according to the model calculations discussed earlier. The dispersal rate can be controlled by the pilot to be smaller or higher, as desired.



Figure 7: The aircraft pass over the airport measurement site on the 24.12.2003. Note the plume of salt powder behind the aircraft.

The results seemed very encouraging so the next step was to test the powder seeding in the Israeli clouds and monitoring the effects with the cloud physics aircraft.

## 5.2 In-Situ Salt Powder Seeding in Israel

The powder seeding was tested in Israeli clouds on couple of case studies. On 23 April 2004 experimental of powder seeding took place in northern Israel over Karmiel in the lower Galilee (32.92N/35.30E). The seeder circled just below cloud base at  $\sim 4500$  feet and a cloud physics aircraft penetrating the seeded and adjacent similar unseeded clouds at various levels above cloud base. The distance in time and space from the spray seeding coordinates was used to identify the potentially seeded cloud volumes. Cloud tops reached 9000 feet, the clouds were shallow and no rain was observed. The surface wind over sea was about 10-15 knots and from visual inspection, only little white caps occurred at the sea surface of the coastal waters. Figure 8 represent the DSD as measured by the CAS, we can see that in the seeded clouds we get a tail of large droplets comparing to the unseeded clouds for approximately the same height and amount of LWC. Figure 9 represent the Effective radius ( $R_{\text{eff}}$ ) of cloud droplets as measured by the CAS comparing to the LWC at height of 8000 feet, looking at the plot, one can see that in the seeded clouds the effective radius is larger than in the unseeded clouds for the same LWC, that means that the seeding affected the cloud by enlarging the cloud droplets, the same results were repeated for various of heights above cloud base

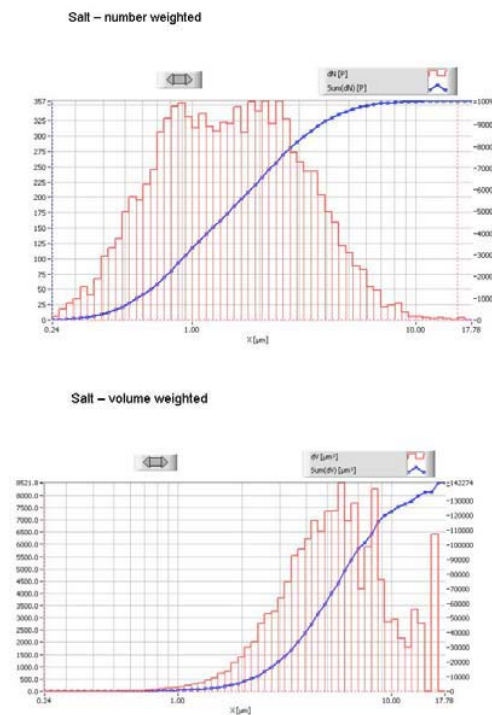


Figure 6: Particle size distribution of the salt powder after it was dispersed from the aircraft. Background concentrations are negligible. The abscissa is the particle diameter, and the left ordinate is the concentration density by numbers on the top panel [ $dN/d\ln(D)$ ], and by volume on the bottom panel [ $dV/d\ln(D)$ ]. The right ordinate provides the negative of the cumulative distribution of the particles.

### 5.3 In-Situ Salt Powder Seeding in Texas

The Powder Salt seeding was tested again on the 4 September 2004 In Texas. The aircraft involved were the research aircraft (Cheyenne turboprop) and salt seeder. The aircrafts departed toward a cumulus cloud field in the NW part of Texas (33.56N/102.99W) close to New Mexico border.

We were using a very low seeding rate (about 3 kg/minute) in clouds that were capped at 17000 feet at the  $-7^{\circ}\text{C}$  isotherm and base at about 11000 feet (2 km deep only) and of continental microphysical nature without warm rain. The clouds that capped at the  $-7^{\circ}\text{C}$  were good for testing hygroscopic seeding because they had no natural rain coming from above, and the expectancy was that any rain would be a result of the seeding.

Faint echoes (<20 dBZ) existed in the seeded cloud before seeding started when its top exceeded 20000 feet, but the echoes faded even more with the collapse of the tops to 17000 feet. After seeding echoes intensity reached again 20 to 30 dBZ. Unseeded clouds with tops at 17000 feet did not echo at all.

Seeded cloud started producing small raindrops 16 minutes after first seeding, visible on the cloud physics aircraft windshield. After additional 10 minutes light rain was reported also by the cloud base aircraft. No raindrops were encountered in unseeded clouds.

Figure 10 summaries the seeding effect, using the data that obtained from the CDP (Cloud Droplet Probe) and CIP (Cloud Imaging Probe), and so are the concentrations. The different in CDP effective diameter accruing at heights 4500-5600 m, but looks less dominant comparing to the effect we got from the CIP.

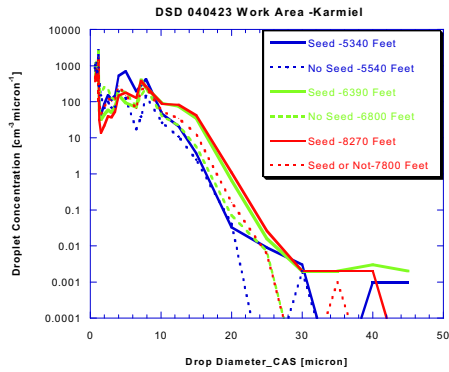


Figure 8: The Drop Size Distributions as measured by the CAS at different heights of cloud passes in seeded clouds (bold lines) and unseeded clouds (broken lines), on 23 April 2004 in north Israel-Karmiel (32.92N/35.30E). The cloud base was 4500 Feet. Note that the seeded clouds have a larger tail of the distribution.

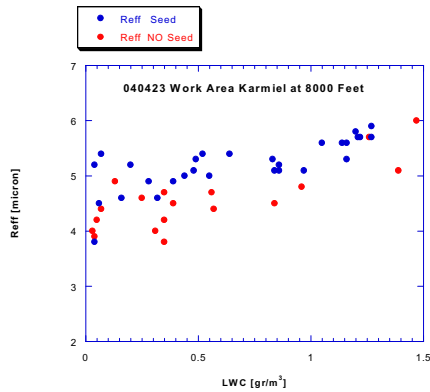


Figure 9- The Effective radius (Reff) of cloud droplets as measured by the CAS vs. the LWC in seeded clouds (blue dots) and unseeded clouds (red dots) over Karmiel (32.92N/35.30E) at height of 8000 Feet on the 23 April 2004.

The large apparent positive impact of the salt powder seeding on the DSD prompted us to continue the experiment, while the experiments with the salt seeding in Israel were conducted only on shallow clouds that were too close to the Mediterranean sea (~40 Km) and could be affected from sea spray in conditions of strong winds. We wanted to continue testing the salt powder effect on deeper clouds in a place that is not affected from marine source.

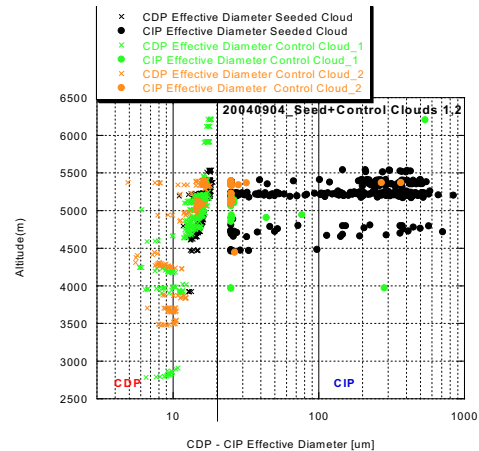


Figure 10- The Effective Diameter (Deff) of cloud droplets as measured by the CDP and CIP vs. the Altitude in seeded cloud (black) and unseeded control clouds (green and orange colors) over NW Texas (33.56N/102.99W) on the 4 September 2004.

The salt powder seeding appears to have the intended effect on the cloud droplets as seen by the sizes and the concentration of the CIP images in the seeded cloud comparing to the control clouds that were analyzed for the same amounts of LWC at the same heights above cloud base.

The seeding rate, as we found, was very small. The material was visible coming out faintly, and the consumption was about 3 kg/minute. Spraying the salt powder in higher rate should affect the clouds even more.

## 6. SUMMARY

We have shown that hygroscopic flares produce mostly sub-micron large CCN particles that are under the optimal size. Dispersion of such particles near-optimal size of 1- $\mu$ m seeding rates is not yet technologically feasible with flares, so the next step was to develop a new seeding method that imitates what nature does with sea spray.

Both observations and simulations showed that the spray seeding produced particles that were much larger than the optimal size. This led us to continue the search for the optimal size. Interactive modeling and observations have been used to identify an optimum method of hygroscopic seeding and a new patented method has been used to process the salt to the sizes (diameters of 2-5 micron) indicated as optimum by the use of a 2000 bin cloud parcel model.

Field tests in Israel have specified the size distribution of the manufactured powder, which was found to be in the desired range specified by the models.

In Situ measurements in the Israeli clouds using an agriculture seeding aircraft which dispersed the salt under the cloud base were very encouraging and prompt us to continue the experiment in Texas in a place which is far from marine source to avoid chance of sea salt aerosols in the air.

The recent results from Texas case study were promising. The DSD spectra in the seeded cloud were wider and the cloud produced raindrops comparing to the control clouds.

The detection of SF<sub>6</sub> gas by the research aircraft and the radar tracking is very important and will allow much greater confidence to determine if the observed changes in the clouds occurred due to the seeding material. Hopefully next summer will continue the experiment using SF<sub>6</sub> detectors.

## 7. ACKNOWLEDGMENTS

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