

Some Size Distribution Measurements of AgI Nuclei with an Aerosol Spectrometer

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ABSTRACT

The Goetz Aerosol Spectrometer, generally considered to possess only a fair ability in resolving size distributions of polydispersed aerosols, operates properly following a modification to the geometry of the entrance to the instrument's deposition channels. Its accuracy is demonstrated with an electron microscopic evaluation of a collecting surface deposit of a thermally produced polydispersed AgI aerosol with particle sizes ranging from 60 to 1000Å in diameter.

Thus calibrated, the instrument was utilized to investigate the activity of the same aerosol as freezing nuclei. The AgI particles on the hydrophobic chrome-plated collecting foil were nucleated by sorption at water saturation for temperatures of -15 and -20°C . The results appear to reflect the influence of the Kelvin effect since the activity decreased at a faster rate than predicted by the "surface area rule" and since it showed a sharp cutoff corresponding to Fletcher's theoretical size temperature predictions for ideal sublimation nuclei.

Also, field measurements were conducted on 12,000-ft Chalk Mountain (Climax, Colo.) for the purpose of measuring the sizes of active AgI-NaI nuclei emanating from acetone ground generators located at least 6 mi upwind. The size distribution of the nuclei on seeding days proved similar to what might be expected from this generator type. On non-seeding days, the number of active nuclei decreased sharply while the peak of the size distributions shifted to larger sizes.

1. Introduction

It is important for AgI particle nucleation theory, generator design and efficiency, and aging effects to determine the sizes of AgI particles which are active as ice nuclei. The technique used to establish the active sizes of particles from generators suitable for weather modification has been to size all the particles with an electron microscope (e.g., Mossop and Tuck-Lee, 1968) and then to apply the theoretical activity curves of Fletcher (1958, 1968) which predict the minimum particle size for which 100% nucleation efficiency exists for a given temperature. This method appears to be no longer applicable since experiments by Katz (1962), Edwards *et al.* (1962), and Mossop and Jayaweera (1969) have shown much lower efficiencies. It is necessary to determine the sizes of only those particles which have the ability to form ice crystals. Very little work has been done along these lines due to a lack of suitable experimental techniques. The usual approach has been to analyze the AgI content of individual ice crystals (e.g., de Pena, 1964; Mossop and Jayaweera, 1969). However, due to the involved nature of this technique, it is difficult to attain the statistical accuracy required by particle-size histograms.

Recently a method was suggested by Weickmann⁴ and developed by Allee *et al.* (1968, 1969) which promises to meet the need for a practical ice nuclei sizing technique. Essentially, the Goetz Aerosol Spectrometer, a high-speed centrifuge developed by Goetz *et al.* (1960), is utilized to deposit the AgI particles along a polished chrome foil according to their Stokes diameters. The foil is removed from the spectrometer, cooled to the desired subfreezing temperature, and then humidified by positioning an ice surface several millimeters above it. Since the temperature of the ice is controllable, the vapor pressure over the sample can be adjusted. By keeping the vapor pressure equal to or less than water saturation, an insignificant amount of water is condensed on the foil which is hydrophobic if carefully cleaned before use. In this controlled environment, the active AgI nucleates by sublimation and grows into visible ice crystals. [The effect of a clean metal substrate on the activation of the AgI particles is apparently insignificant as concluded by Edwards and Evans (1968) in similar experiments.] To find the diameter of the active ice nuclei, only the location of the corresponding ice crystals need be determined since the coordinates of the foil are related to particle size by Stokes law after calibration of the spectrometer.

The spectrometer was used to size active AgI particles originating from two different sources. The first source consisted of aerosol produced by heating a small

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quantity of AgI-amine complex to about 1000C in a stream of nitrogen. Since the complex decomposes at a lower temperature, the AgI evaporates last and, therefore, should condense into pure AgI particles. These particles were used to investigate the activity of the AgI particles as a function of size at two temperatures. Since the peak in the size distribution of the particles would probably be of the order of several hundred angstroms, it was expected that this experiment would demonstrate the influence of the Kelvin curvature effect and test Fletcher's theoretical predictions for heterogeneous nucleation of AgI ice nuclei.

The second source consisted of acetone AgI-NaI ground generators located in the vicinity of Chalk Mountain, Colorado. Many previous measurements of ice nuclei concentrations conducted on top of Chalk Mountain for the cloud-seeding project of Colorado State University (CSU) have shown that periods of seeding correlated strongly with an increased count of ice nuclei (Reinking and Grant, 1968). Although the increased counts leave little doubt as to the origin of the ice nuclei, their positive identification had yet to be made. Ten spectrometer measurements on Chalk Mountain over a period of two weeks were compared to discover any unique characteristics in the size distributions of seven non-seeding and two seeding days.

Before discussing the measurements in detail, it is first necessary to place the utilization of the spectrometer on a firmer basis since the scientific community currently questions the ability of the instrument to size atmospheric particulates accurately (e.g., Green and Lane, 1966).

2. Performance of the Goetz Aerosol Spectrometer

The current reputation of the spectrometer is largely a result of a theoretical study of the aerosol dynamics inside the centrifuge channels (Stöber and Zessack, 1964) and of the subsequent attempts to verify the theoretical predictions (Stöber and Zessack, 1964; Raabe, 1966; Baust, 1967). Theory and experiment did not agree except in demonstrating that the spectrometer did not operate in the manner of a horizontal elutriator (Walton, 1954), as was specified by its developers. Over its intended range of operation, the spectrometer must form homogeneous deposits with sharp cutoffs along its collecting foil for monodispersed particles.

a. For 0.1 to 2.0 μm particles

In recent experiments, Gerber (1970) investigated the behavior of the spectrometer for particle sizes between 0.1 and 2.0 μm . He found that for specific operating conditions of the stock instrument (flow rate and centrifuge rotor rpm), the desired homogeneous deposits and cutoffs existed. However, these conditions only corresponded to particle sizes $\gtrsim 1 \mu\text{m}$. When the

spectrometer was modified by removing the baffle from the instrument's inlet, operating conditions were found for which proper performance existed over the entire investigated size range.

The inconsistencies of the earlier work were explained. The key oversimplification in the theoretical work was the assumption of a steady Hagen-Poiseuille type velocity profile in the flow along the entire length of the instrument's channels. Experiment showed a non-parabolic profile at the channel entrances, resulting in a strong influence on the particle trajectories in the channels due to the modifying velocity profile in the laminar flow. The previous experimental work agreed closely with that of Gerber. Nevertheless, the limited choice of operating conditions of the former investigations and a lack of emphasis that only a small portion of the operating range of the spectrometer had been investigated, led to the unfavorable verdict of the instrument.

b. For 200 to 1500 Å AgI particles

Usually the majority of the particles from AgI generators are smaller than 0.1 μm . Therefore, it is also necessary to demonstrate that the spectrometer functions properly for these sizes. Goetz *et al.* (1960) suggested that their instrument could capture and size particles as small as 250 Å. However, this was only an estimate determined by extrapolating calibration data for particles $> 0.1 \mu\text{m}$. No experimental work has been done to test the spectrometer for sizes much under 0.1 μm .

It does, however, appear entirely feasible that the spectrometer could collect particles as small as 100 Å. By operating the instrument at high rotor speeds and lowering the flow rate so as to lengthen the time that the particles are exposed to the centrifugal field, Stokes law predicts that 100 Å particles can be captured.

1) EFFECT OF BROWNIAN DIFFUSION

Whether the spectrometer can still accurately size these small particles depends not only on the instrument's proper operation as a horizontal elutriator but also on the limiting effects of Brownian diffusion of the particles. Since the horizontal elutriator is limited to accurately sizing particles $> 0.5 \mu\text{m}$ due to the diffusion effect, it is an appropriate question to ask how this limit applies to the spectrometer, which can collect much smaller particles.

In both instruments the particles are introduced across the entire width of the channel entrances. As the dispersion medium carries the particles down the channels of rectangular cross section, the gravitational field in the elutriator and the centrifugal field in the spectrometer (a maximum of 30,000 times greater) causes the particles to fall out against one of the walls. Therefore, along the channels and opposite the deposit wall there will be an ever-increasing volume from which the

particles have already fallen. For each differential size group in a polydispersed aerosol this boundary between the particles and the dispersion medium must remain clearly defined. Brownian diffusion blurs the boundary, and the sharp deposit cutoff necessary for sizing accuracy no longer exists.

In an analogous problem Fuchs (1964) gives the distribution of particle concentration, $n(x,t)$, across a completely permeable wall. Under the boundary conditions of $n(x,0)=n_0$ for $x>0$, and $n(x,0)=0$ for $x<0$, which means that initially no particles are beyond the wall, the concentration perpendicular to the wall after the particles have been permitted to diffuse for a time t is given by

$$n(x,t) = \frac{n_0}{2[1 + \operatorname{erf}x/(4Dt)^{1/2}]}, \quad (1)$$

where D is the coefficient of diffusion.

When the diffusion in the two instruments is small, Eq. (1) gives the spread that the otherwise sharp deposit cutoffs would have. This, in turn, is equivalent to the broadening of peaks in the size distribution corresponding to monodispersed particles.

Due to the higher acceleration that the particles experience in the spectrometer, their fall times are much less than in the elutriator. As a result, the diffusion effect should become important only in the spectrometer for particle sizes significantly smaller than $0.5 \mu\text{m}$. For 100\AA particles, one standard deviation of the broadening is equivalent to an acceptable 0.17 cm on the spectrometer deposit surface or less than 5\AA upon conversion to particle size.

2) CALIBRATION OF THE SPECTROMETER

The calibration aerosol consisted of the previously mentioned AgI particles which were thermally generated by vaporizing a small quantity of AgI-amine complex in a nitrogen atmosphere. The generator was developed by Katz and Davis of Colorado State University for the purpose of establishing an easily reproducible source of pure AgI particles. It consisted of an open glass tube $\sim 60 \text{ cm}$ long and 5 cm inside diameter. An ordinary 12 V automobile lighter was situated in the center of the tube and a small petri dish, the diameter of the lighter's heating element, was placed on the lighter. In these experiments $\sim 0.25 \text{ cm}^3$ of AgI-isopropyl amine (0.2% AgI by weight) was placed in the dish. Nitrogen at $5 \text{ liters min}^{-1}$ was blown through the tube while the AgI evaporated at a temperature of $\sim 1000\text{C}$.

Eight times the output of the generator was passed into an open 10-liter flask from which the spectrometer drew its sample at a measured $0.49 \text{ liter min}^{-1}$. The spectrometer was operated at $18,000 \text{ rpm}$ and with exit jets 16 mils in diameter. These operating conditions were chosen since previous calibration work (Gerber,

1970) had shown that the same conditions resulted in the proper deposits for 0.234 and $0.109 \mu\text{m}$ monodispersed latex particles. The lengths of the deposits on the 40 cm long collecting surface were 7.5 and 11.2 cm , respectively, and as a result, suggested that the same operating conditions would form proper deposits of smaller particles over the rest of the collecting surface.

Since the majority of the AgI particles was expected to be smaller than $0.1 \mu\text{m}$, it was necessary to evaluate their sizes on the spectrometer foil with an electron microscope. Eight carbon film grids, A, B, C, D, F, G, H and J were cemented along the foil surface at distances of $7.7, 9.8, 12.5, 15.5, 18.5, 22.6, 29.0$ and 37.0 cm . After the AgI particles were collected, the grids were removed, shadowed with a few angstroms of platinum, and then analyzed with a Siemens Elmicope I. Photographs were taken of each grid at a magnification of 10^5 . The projected diameter of about 3500 particles was measured for the seven size distributions shown in Fig. 1 (grid G was accidentally destroyed during handling).

Fig. 1 clearly demonstrates that there is a different maximum in the size collected by each of the grids. For the grids further along the collecting surface, the size

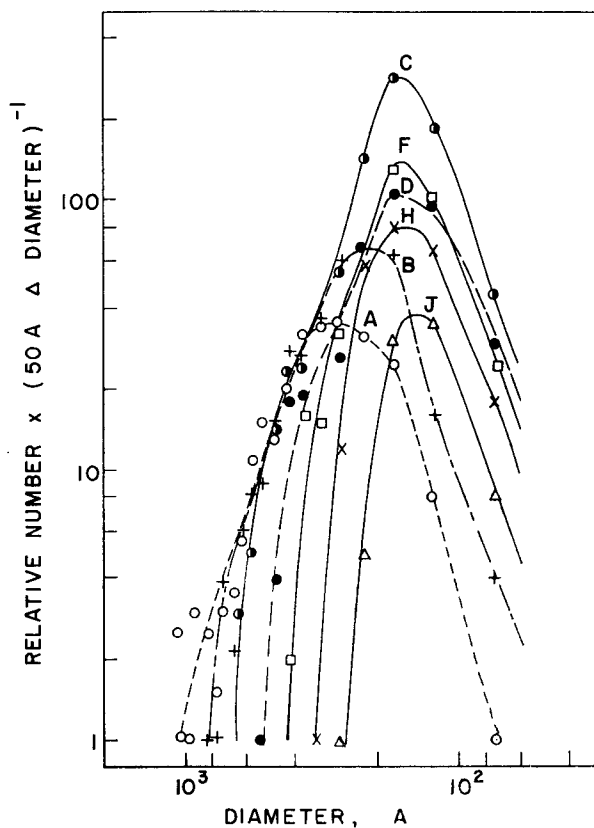


FIG. 1. Size distributions of the projected diameters of AgI particles (Katz-Davis generator) deposited on electron microscope grids (A-J) cemented along the spectrometer collecting surface at progressively longer distances from the instrument's inlet.

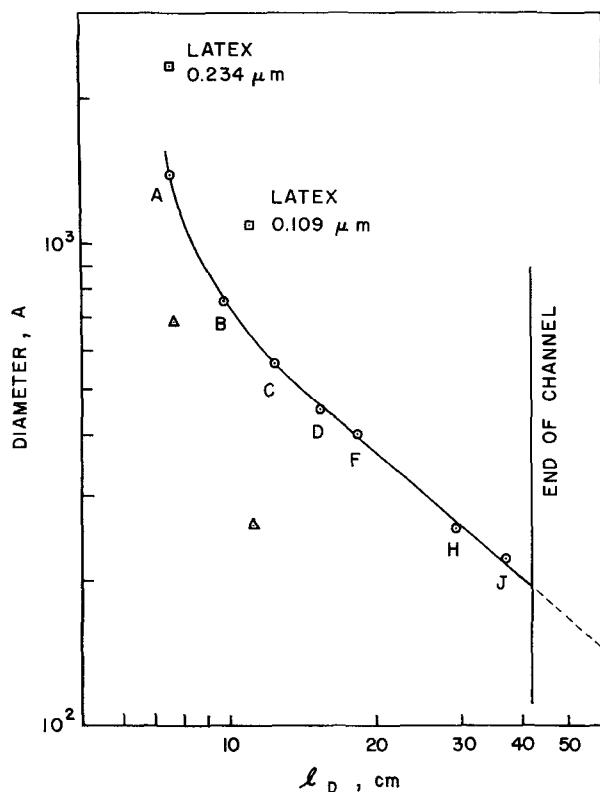


Fig. 2. The maximum deposit length l_D of AgI particles (Katz-Davis generator) as a function of their projected diameters, circles. The latex particle calibration points, squares, are utilized to find the Stokes diameter of spherical AgI particles, triangles, as well as the effective density, 2.06 gm cm^{-3} , and shape factor, 2.17, of the generated AgI particles (see text).

cutoff exists for progressively smaller particles. This pattern is consistent with the proper operation of the spectrometer, where the larger particles are deposited nearer the beginning of the collecting surface, as predicted by Stokes law. The cutoffs indicate that all particles larger than the cutoff size for each grid must have been deposited on the collecting surface before that grid was reached. In other words, the location of each grid gives the maximum distance l_D along the surface that particles of the cutoff size will penetrate. The relationship between l_D and projected particle size is given in Fig. 2. This calibration curve can now be utilized to predict the deposit lengths of particles of similar AgI aerosol without having to again size individual particles.

Fig. 2 shows that 200\AA is the minimum size of AgI particles that can still be collected 100% under the specified operating conditions. The 60\AA particles are only partially captured by the spectrometer. However, by slowing the flow rate sufficiently, probably all those particles could also be captured.

The second requirement for proper operation was that the deposits of monodispersed particles must be homogeneous up to their respective cutoffs. This signifies

that the curves in Fig. 1 should have been identical for all particle sizes except in the cutoff regions. Fig. 1 shows that all curves are not alike; instead, except for A and B, they are similar in shape and displaced vertically from each other.

A probable explanation of the shift in the curves can be found in the effect that the grids have on the flow near the collecting surface. Since the copper grids were 14\AA thick and 25 mm wide, and the cement utilized to attach them to the surface was probably thicker in spots, there could well have been a small perturbation in the flow near each grid. In the vicinity of the perturbation the deposit concentration would be affected. This is borne out by past experience which has shown that large dust particles on the surface affect the concentration of smaller particles in the immediate area. If the lengths of the perturbations due to the grids was small in comparison to the l_D of the particles, the effect on the deposit concentration in the vicinity of the grids would have been independent of the sizes of the particles, and curves similar to the ones in Fig. 1 would have resulted. Also, it appears highly unlikely that the constant percentage difference between the curves in Fig. 1 would exist if the deposits of each size group of AgI particles were anything but homogeneous.

The smaller particles could not be resolved on grids A and B since the surface was covered with many larger soot-like particles. Insulation on one wire leading to the lighter was scorched during the generator operation and was the likely source of these particles.

3) PARTICLE SHAPE

The electron microphotographs showed the dispersed phase of the AgI aerosol to consist of single particles with both cubic and hexagonal crystal structure. Columns appeared to be predominant, although some plates were also seen. It is unlikely that the shapes of the particles varied extensively since otherwise the cutoffs in Fig. 1 would have been much broader.

The effective density and the shape factor of the AgI particles were found from Fig. 2. According to Stokes law the density times the diameter squared of any particles must be constant if they are deposited on the same location of the collecting surface. Hence for latex particles ($\rho_1 = 1.0 \text{ gm cm}^{-3}$) of size D_1 and spherical AgI particles ($\rho_2 = 5.8 \text{ gm cm}^{-3}$) of size D_2 situated on the same deposit location, the relationship

$$\rho_1/\rho_2 = (D_2^2/D_1^2)[(1+2Al/D_2)(1+2Al/D_1)] \quad (2)$$

must hold, where the quantity in brackets is the Cunningham correction.

With Eq. (2) and the two latex particle calibration points in Fig. 2, it is possible to compute D_2 . The resulting values (Fig. 2, triangles) are less than the experimental AgI sizes D_3 measured for the same l_D . This confirms that AgI particles are not spherical but of an

irregular shape which gives the particles a smaller settling velocity.

Eq. (2) is again applied to find the effective density ρ_3 of the AgI particles (ρ_3 is the density of spherical particles with the same Stokes velocity as the AgI particles). After replacing ρ_2 and D_2 in Eq. (2) with ρ_3 and D_3 , inserting the values for ρ_1 , $D_1=0.234 \mu\text{m}$, and $D_2=0.109 \mu\text{m}$, Eq. (2) is twice evaluated to yield $\rho_3=2.12 \text{ gm cm}^{-3}$ and $\rho_3=2.00 \text{ gm cm}^{-3}$. The values for ρ_3 appear to be large enough to rule out the possibility that each AgI particle is a conglomeration of much smaller particles, as has been sometimes postulated. If the value of ρ_3 is assumed to be about the same value for the smaller particle sizes in Fig. 2, then Eq. (2) can be used to find the calibration curve for AgI particles of any other known effective density.

The shape factor γ is defined as the projected diameter divided by the Stokes diameter (Green and Lane, 1966). For the AgI particles $\gamma=D_3/D_2$ and has the average value of 2.17. In comparison, anthracite dust has a shape factor of 2.05 (Hamilton, 1954), while the more nearly spherical softer coal particles have $\gamma=1.34$ (Watson and Cruise, 1954).

3. Size distribution of active AgI particles

a. Laboratory aerosol

The AgI aerosol was again provided by the Katz-Davis generator. Since a much lower concentration of particles was desired on the spectrometer foil, the output of the generator was diluted in the 1500-liter CSU isothermal cloud chamber. The generator and the spectrometer were operated in exactly the same manner as before. Hence, it was felt that the AgI particle size distribution on the foil would be closely reproduced from the calibration test.

For each of three separate tests the spectrometer drew an ~ 1 -sec sample out of the chamber after 1.5 min had elapsed since initial introduction of the particles. After the deposit for the first test was cooled to -20°C , an ice surface at -18°C was placed several millimeters above the foil for 60 sec. The ice surface simulated vapor pressure saturation with respect to water since the saturation vapor pressure of ice at -18°C is 1.24 mb, while for water it is a nearly identical 1.25 mb. The second and third tests were each exposed to water saturation at -15°C by using an ice surface at -13.5°C for 60 sec. Immediately upon removal of the ice surface the foil was photographed. The ice crystals grown on the active ice nuclei were made visible by illuminating the foil at a shallow angle.

Fig. 3 shows the count of ice crystals per unit area on the foil as a function of position on the deposit for each of the three tests. The size of the active nuclei corresponding to the ice crystals is found from Fig. 2 and is also given as an abscissa in Fig. 3. The data is relative since it was not possible to accurately determine the

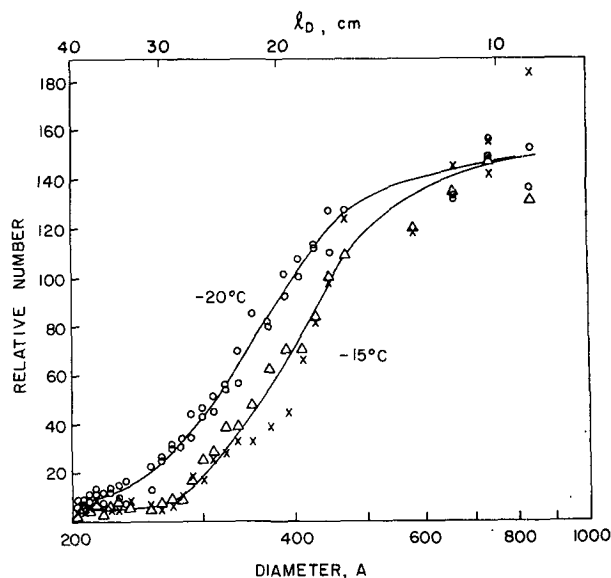


FIG. 3. The relative number of ice crystals grown on AgI particles (Katz-Davis generator) per unit area on the spectrometer collecting surface as a function of l_D , the projected diameter of the AgI particles, and the temperature. The curve for -15°C represents two independent tests.

sampling time and thus the total concentration of the particles for each test. When the total number of crystals counted for the first -15°C test was normalized to the total of the second -15°C test, the good agreement shown in Fig. 3 was found between the two tests. This strengthens the assumption that the generator produces the same AgI particle sizes each time.

The curves in Fig. 3 are related to the integrated size distribution of the nuclei since the spectrometer forms homogeneous deposits for each narrow-size interval. Therefore, to obtain the size distribution, the curves must be differentiated and multiplied by the area deposition function which gives the area on the foil covered by particles of each size interval (Goetz and Kallai, 1962). The final size distribution of the active AgI particles is given in Fig. 4 along with the distribution of all the AgI particles determined by electron microscopy in the last section. Also shown is the distribution of that portion of all the particles which would be active according to the "surface area rule" (Katz, 1962; Edwards *et al.*, 1962), which states that the probability for a nuclei to have an active nucleation site is proportional to its surface area. The values of the distributions are relative. Since the data are plotted on semi-log paper, the shapes of the curves are absolute although their proper positions with respect to the ordinate are unknown.

The outstanding difference between the curves in Fig. 4 is the lack of activity for small particles in the measured distributions of active particles. For the larger particles it is difficult to say how well the measured distributions follow the "surface area rule" since the data in Fig. 3 show large statistical fluctuations

above 500Å. However, below 400Å for the -15°C curve and below 320Å for the -20°C curve, the measured activities initiate a sharp decrease. Within a 30% reduction in each of these particle sizes, a 95% decrease takes place in the activity of the AgI particles.

It is customary to compare the activity of ice nuclei to the classical theory of Fletcher (1958), who determined the particle size and temperature-dependent nucleation threshold for nucleation by freezing and sublimation. At a temperature of -15 and -20°C , Fletcher's computations show a particle size threshold of 85 and 20Å, respectively, for freezing and 500 and 300Å for sublimation. Comparison of these values with the data in Fig. 4 shows reasonable agreement if the AgI particles used in this experiment are assumed to nucleate ice by sublimation. Earlier work by Edwards and Evans (1960) confirms this assumption since in their experiments AgI particles with a maximum diameter of 200Å failed to nucleate ice in a water vapor environment unless quickly included into droplets. The slight shift in the activity threshold in Fig. 4 from Fletcher's values might indicate that the AgI particles behaved more nearly like ideal sublimation nuclei. Also, the activity did not demonstrate the sharp cutoff specified by the theory. Perhaps both deviations are related to the uncertain relationship between the projected diameters in Fig. 4 to the same diameters of the spherical particles in the theory. However, the basic agreement between experiment and Fletcher's theory

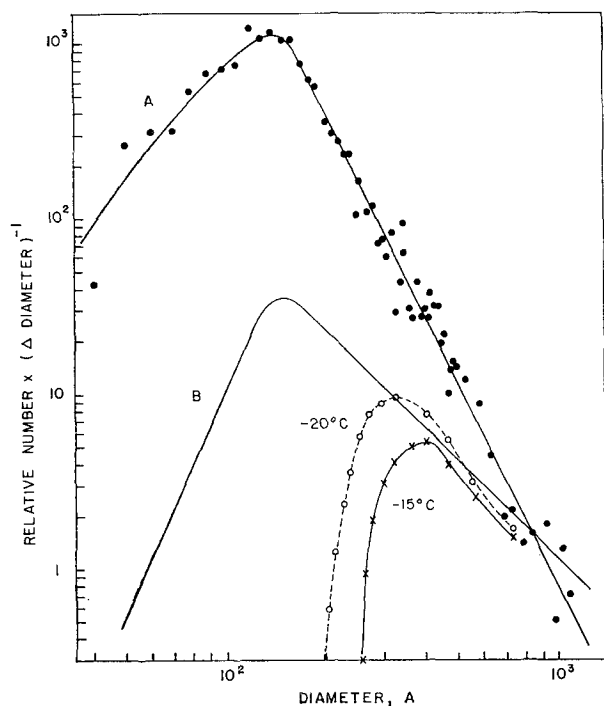


FIG. 4. The size distributions of active AgI particles (-15 and -20°C), of the AgI particles sized by electron microscopy (A), and of the active AgI particles according to the "surface area rule" (B). The Katz-Davis generator was utilized.

TABLE 1. Spectrometer measurements of ice nuclei at 12,000 ft on Chalk Mountain, Climax, Colo.

Test	Date (September 1969)	Time (MST)	rpm (10^3)	Jet diameter (mils)	Number of nuclei (liter^{-1})	Peak in dN/dD (μm)
a	4	1348-1727	18	30	0.25	>0.7
b	5*	1018-1403	18	30	>50	<0.2
c ₁	10	0825-0923 0956-1028	18	30	0.1	>0.7
c ₂	10	1226-1302 1310-1558	18	30	0.15	>0.7
d ₁	11*	1403-1700	18	16	30.4	0.1
d ₂	11*	1858-1957	18	16	550	0.12
e	12	0945-1250	18	30	1.7	0.22
f	16	1315-1435	18	30	2.6	0.35
g	17	0924-1425	18	30	1.9	>0.7
h	18	1108-1508	18	30	1.0	0.5

* After 12 hr of AgI seeding from upwind ground generator.

suggests the appropriateness of most assumptions in his derivation. Therefore, contrary to some of the other currently popular nucleation theories [e.g., the water cluster theory of Zettlemoyer *et al.* (1961) and the two-dimensional crystallization theory of Edwards and Evans (1968)], it is necessary to conclude that ice embryos in the shape of spherical caps form on the active sites of AgI when nucleation occurs by sublimation.

b. Aerosol generated with an acetone burner in the field

Over a two-week period, ten measurements of the atmospheric ice nuclei size distribution were analyzed with the spectrometer on the 12,000-ft Chalk Mountain in Colorado. This site is situated in the remote, mountainous, central portion of the state and is the seeding target area for the CSU cloud-seeding project (Grant and Mielke, 1967). Six modified Skyfire needle-type ground generators (Grant and Steele, 1966) are located in the two quadrants west of Chalk Mountain, at distances ranging from 13 to 59 km. On days when the randomized seeding experiment stipulated seeding, at least two of the generators were operated. Of the eight days that the spectrometer was used, two were seeding occasions. The operating mode of the spectrometer was either 18,000 rpm with a 16-mil jet, described in the preceding sections, or 18,000 rpm and a 30-mil jet, which accurately sizes 0.1 to 0.75 μm particles (Gerber, 1970). After the samples were collected, the foils were exposed to water saturation at -20°C .

The results are summarized in Table 1. On each of the two seeding days, the concentration of ice nuclei was much higher than the background concentration which averaged slightly more than 1 liter^{-1} for seven measurements. These results are in good agreement with the findings of Reinking and Grant (1968), which correspond to a much larger volume of data collected on Chalk Mountain. The peak in the particle size distributions was noticeably shifted between seeding and non-seeding conditions. On the seeding days the peak was located at $\sim 0.1 \mu\text{m}$. Since for these measurements the

spectrometer deposit was evaluated for calibration curves specified for spherical particles with a density of 1.0 gm cm^{-3} , this peak shifts to a Stokes diameter of $\sim 0.07 \mu\text{m}$, if it is assumed that the effective density of the particles actually is 2.0 gm cm^{-3} [Eq. (2)]. The latter size agrees with the mean projected diameter found by Mossop and Tuck-Lee (1968) for AgI-NaI particles generated by similar means and evaluated by electron microscopy. On the non-seeding days the peak shifted to the larger values given in Table 1 for particles with $\rho = 1.0 \text{ gm cm}^{-3}$. These findings clearly demonstrate that the AgI-NaI particles actually advected to the summit of Chalk Mountain from the ground based-generators, that little if any coagulation occurred in the aerosol during its trajectory, and that the measurement of particle size is a good indicator of the presence of the seeding agent.

Fig. 5 shows the ice nuclei size distributions for the second seeding day and for three non-seeding days when enough nuclei were deposited on the foil to make the evaluation statistically feasible. The distributions on the seeding day are typically narrow while the non-seeding curves are much broader and also extend to smaller sizes than are usually attributed to background ice nuclei. Although the curves must end abruptly according to the chosen operating mode of the spectrometer, it is still possible to extend their approximate position beyond the measured values. Accordingly, since the spectrometer also collects 100% of the particles which are larger than the sizes that can be accurately repro-

duced, the total count of ice crystals on the foil should not be more than the integrated size distribution unless additional peaks exist for those larger sizes. For all five curves in Fig. 5, the two totals were within 30%. This argument cannot be applied to the very small particles which are only partially captured by the spectrometer. However, since the integrated distributions showed low values for the five measurements at maximum l_D , all the curves in Fig. 5 will continue to decrease in value for smaller sizes.

An unexpected consequence of the size measurements was an observation made on the nature of the background nuclei. After the moisture from the ice crystals on the 18 and 19 September 1969 foils had evaporated, the locations of the ice nuclei were checked with a light microscope. By humidifying the foil by breathing upon it, at least one of the particles at each site was seen to be strongly hygroscopic. When reactivation of the nuclei was attempted by exposing the foil to the same subfreezing environment as before, most nuclei were inactive. Soulage (1957) came to similar conclusions as to the nature of the ice nuclei in his examination of the residues of evaporated snowflakes. However, when next the foil was heated to temperatures $> 250\text{--}300\text{F}$, more than ten times the original number of nuclei surprisingly became active after the identical exposure in the cold box. The activation and deactivation could be cycled if exposures in the cold box after heating followed exposures after an excessive use of moisture. An explanation has not been found for this phenomena; but perhaps it explains the persistently lower ice nuclei counts which the acoustical counter (Langer, 1965) gave in relationship to the spectrometer and the Bigg-Warner expansion chamber (Warner, 1957), which were all operated simultaneously. Since the sample aerosol is strongly humidified before its passage into the acoustical chamber's cloud chamber, the potential ice nuclei might be deactivated.

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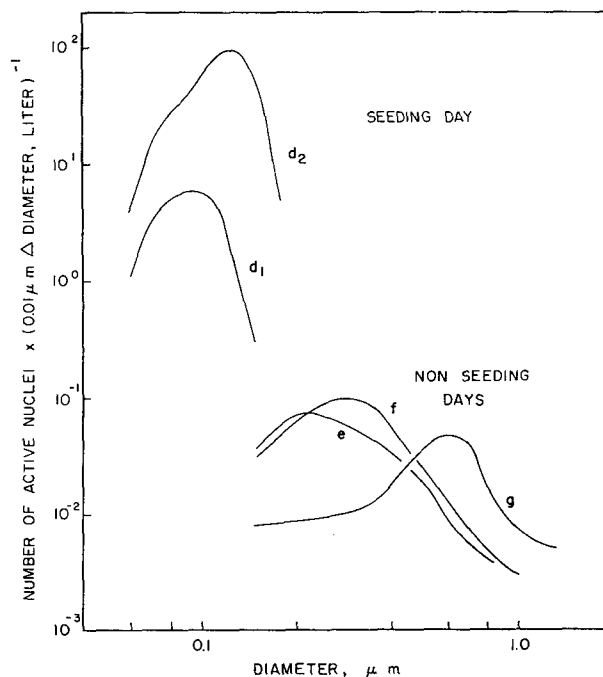


FIG. 5. The size distributions of ice nuclei measured on Chalk Mountain with the spectrometer on a seeding day (tests d_1 , d_2 , Table 1) and on nonseeding days (tests e , f , g , Table 1).

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