

AgI-NaI Aerosols as Ice Nuclei

S. C. MOSSOP AND K. O. L. F. JAYAWEERA

Radiophysics Laboratory, CSIRO, Sydney, Australia

(Manuscript received 10 September 1968, in revised form 19 November 1968)

ABSTRACT

The ice-nucleating properties of cloud-seeding aerosols produced by burning a solution of AgI and NaI in acetone have been studied in a laboratory cloud chamber. The ice-forming ability of these mixed aerosols is not influenced as markedly by the supersaturation in the test cloud as is the case with "pure" AgI aerosols. This is ascribed to the hygroscopic nature of the particles which promotes drop formation at low supersaturations and allows the AgI to nucleate by a freezing mechanism. The relationship between nucleus size and nucleation temperature has been investigated and is compared with theory. Fletcher's theory, as recently modified to take account of the soluble component in mixed AgI aerosols, does not explain the rapid decrease in nucleation temperature with decreasing particle size and the wide size range of nuclei active at a particular temperature. A theory which assumes that nucleating sites of widely differing efficiencies are randomly distributed over the surface of the nucleating material is favored.

1. Introduction

Despite the search for other ice-nucleating substances, silver iodide remains the most commonly used material for "seeding" supercooled clouds. When used by itself as an aerosol of "pure" silver iodide it suffers from an important drawback. As shown by the experiments of Edwards and Evans (1960, 1968), a particle of AgI is much less effective in nucleating ice by the "sublimation" mechanism (when suspended in the interdroplet space in a cloud) than when actually enclosed in a water drop and able to act as a freezing nucleus. Because of the comparatively hydrophobic nature of AgI, it has been argued (Fletcher, 1959a; Edwards and Evans, 1960) that particles will be unable to grow into droplets at the small supersaturations found in natural clouds. Being generally of a size unsuitable for capture by impaction or by Brownian diffusion, they can therefore nucleate ice only by the comparatively inefficient "sublimation" process. Most laboratory tests of AgI as ice nuclei have been done in cloud chambers in which large supersaturations were produced, so that nucleation may have taken place by the condensation-freezing process. It is therefore doubtful whether the results can be applied to natural clouds.

It has generally been assumed that these strictures would not apply to the aerosols commonly used in seeding experiments. These smokes are produced by burning an acetone solution of AgI with an alkali halide added to enable the AgI to dissolve. The particles are therefore expected to be hygroscopic, so there would be no difficulty in forming water drops around the AgI particles, thus enabling them to nucleate by the efficient freezing process. These assumptions will now be examined.

2. Composition and hygroscopic nature of AgI/alkali-halide aerosols

Our discussion will be concerned with the aerosols produced by burning a solution of AgI and NaI in acetone, as used for cloud seeding in Australia (Smith *et al.*, 1966).

The composition of these particles was investigated by Mossop and Tuck-Lee (1968), who found that they are a mixture of beta-AgI and NaI. Mason and Hallett (1956), on the other hand, could find no trace of beta-AgI and ascribed their electron diffraction patterns to the cubic form or to sodium iodide. However, their results applied to AgI-NaI vaporized from a hot wire where temperature conditions may have been quite different to those in a seeding generator.

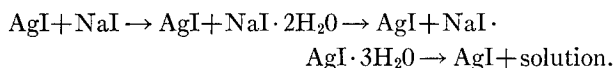
Mossop and Tuck-Lee also found that all particles of the mixed aerosol were hygroscopic down to the size limit of the method used (0.05 μ diameter). Further confirmation of the ability of these mixed aerosol particles to assist droplet formation in clouds comes from work reported by Mossop (1968), in which the ability of the particles to grow into droplets was studied and compared with that of "pure" AgI particles. As illustrated in Fig. 1, the mixed particles take up water far more readily. At a supersaturation of 1%, half of them grow into water drops whereas only 4% do so in the case of AgI particles.

The mixed particles will start to take up water at humidities well below saturation. Tompkins *et al.* (1963) have shown that AgI and NaI in contact take up water at a relative humidity (unstated) below 55%. According to St. Amand *et al.*¹, complex hydrates will then form.

¹ St. Amand, P., W. Finnegan, L. Burkardt and F. K. Odenrantz, 1968: Effects of the type of nucleant on modification of clouds for the stimulation of rainfall. Paper presented at First Natl. Conf. on Weather Modification, Albany.

In the work that follows we shall be discussing aerosols produced by burning an acetone solution of AgI and NaI, the latter constituting one-quarter of the weight of the AgI. We have made an electron microscope comparison of the size distribution of the mixed particles with that of the AgI constituent alone (after dissolving away the NaI). This shows that the proportion of AgI to NaI in the aerosol particles does not differ markedly from that in the original solution.

Let us consider the changes that occur when these mixed particles take up water in a humid atmosphere. The phase diagram of St. Amand *et al.* shows the following successive transitions:



The complex salt starts to dissolve when about 7% by weight of water has been taken up. The solubility tables of Linke (1966) indicate that the proportion of the AgI which dissolves reaches a maximum of 21% when the particle has taken up 10% by weight of water. As more water is taken up, AgI will start to precipitate. By the time the solution droplet reaches equilibrium in a water-saturated environment, only 18% of the AgI will be in solution, if we consider a particle containing an amount of AgI equivalent to a sphere of diameter 0.01 μ ; even less AgI will be in solution for larger particles.

There is therefore no danger with our aerosols that the presence of the NaI will cause the AgI particles to go completely into solution when first introduced into a cloud and in this way prevent them nucleating ice on reaching low temperatures. This can only happen when the proportion of sodium iodide to silver iodide is much greater. The solubility tables of Linke (1966) show that a mole ratio of NaI to AgI of 1.9 (i.e., a weight ratio of 1.2) is necessary for all the AgI to dissolve.

Even though it escapes going into solution in the early stages of drop formation, a small AgI particle may dissolve completely later, should the drop in which it resides grow to large size. This can happen despite the low solubility of AgI (2.6×10^{-6} gm liter⁻¹). We must therefore examine the possibility of small AgI particles being found within large water drops. Considering a cloud with a high supersaturation, say 1%, we see from Fig. 1 that only 50% of our mixed particles are likely to grow into cloud droplets. These will, in general, be the largest 50% of the particles. A particle of the median size has an AgI component about 0.08 μ in diameter, which will need a 100 μ drop to dissolve it completely. In view of the long growth time of such drops it is doubtful whether a significant proportion of AgI-NaI particles dissolve before reaching the supercooled regions of our clouds, provided they are introduced into a region of updraft.

It has been suggested by Russian workers [as reported

by Battan (1965)] that AgI is less effective as ice nuclei when introduced into cloud base at temperatures above 0C than when injected directly at the -5C level. The reason for this reported inactivation is not clear. The arguments given above indicate that in the case of our AgI-NaI aerosols, inactivation due to the AgI being completely dissolved is unlikely. This does not rule out the possibility that active ice nucleating sites may be dissolved away when a droplet forms on an AgI-NaI particle. Obviously this important point merits further investigation.

3. Comparison of AgI-NaI and AgI aerosols as ice nuclei

The behavior of AgI-NaI particles and "pure" AgI particles as ice nuclei has been studied in a mixing-type cloud chamber.

a. Production of AgI-NaI and AgI aerosols

An airborne-type generator (Smith *et al.*, 1966) was operated on the ground, using a blower to simulate the air flow that would be present in aircraft operation. A solution of AgI and NaI in acetone was burned at the rate of 2.5 ml sec⁻¹ (approximately 0.15 gm sec⁻¹ of AgI). The products were fed into a wind tunnel and three successive dilutions of the wind-tunnel effluent were then made in plastic bags. These were 500-liter bags made of plastic sheeting metallized on the inside and containing clean, dry nitrogen as carrier gas. The concentration of aerosol particles in the final bag was about 1 cm⁻³. Theoretical and practical tests showed that particle losses in the dilution and storage system could be ignored.

Measurements of the concentration and size distribution of the aerosol particles were made immediately after production are described by Mossop and Tuck-Lee (1968). They found that the particle size distribution follows a log-probability law with median diameter 0.085 μ and standard deviation factor 1.47.

The AgI aerosol was produced by heating a few milligrams of the salt in a silica tube in a stream of nitrogen. The aerosol was stored and diluted as described above. Some particles when examined in the electron microscope were found to be aggregates of several lumps, indicating that the smoke in the silica tube had been insufficiently rapidly diluted. Particle diameter was taken as that of the largest constituent lump. The size distribution found in this way could not be represented by a single log-probability relationship. Diameters ranged from 0.03-0.4 μ with a median value of 0.09 μ . Even measured in the conservative way described above, the sizes of these AgI particles were, on average, somewhat larger than those of the AgI-NaI particles, and the number per gram of AgI was correspondingly less.

This aerosol was also used to determine the AgI condensation nucleus spectrum shown in Fig. 1.

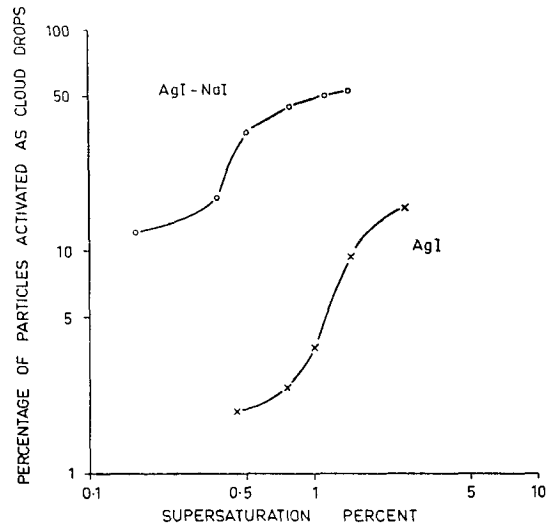


FIG. 1. Percentage of aerosol particles which grow into water droplets at various supersaturations. Upper curve: AgI-NaI aerosol, experiment of 27 July 1966. Lower curve: aerosol produced by heating AgI in silica tube, experiment of 20 May 1966.

b. Measurements of ice nucleating ability

The proportion of particles acting as ice nuclei at various temperatures was measured by injecting into a cloud chamber a volume of aerosol, either 0.1 or 1 liter, taken by metal syringe from whichever of the last two stages of dilution was the more suitable at the particular cloud chamber temperature.

A 10-liter mixing-type cloud chamber was used. Ice crystals were detected by means of a tray of super-cooled sucrose solution on the bottom of the chamber. After injection of the test aerosol a cloud was made by introducing a semi-permeable cylinder (sausage skin) containing warm water, for 2 min. The humidifier was moved round the chamber in a circular path during this time. It was then removed, and the ice crystals which fell into the sucrose tray were counted at the end of a further 2 min. To avoid frost formation the walls were coated with a 50% glycerol-water mixture. The temperature of the cloud chamber was taken as that of its coldest part, i.e., the surface of the sucrose solution. Because of the temperature gradient from top to bottom of the chamber it is possible that the mean temperature to which nuclei are subjected is up to 1.5C warmer than this value. This possible error is not important when we are comparing sets of observations made with the same cloud chamber.

Ice nucleus spectra are plotted in Fig. 2a for AgI-NaI aerosols using humidifier temperatures of 35 and 45C. Even allowing for the scatter of the observations, there appears to be a difference between the two spectra at the warm temperature end. However, the behavior of these aerosols contrasts markedly with that of "pure" AgI smokes, which show a much greater influence of humidifier temperature on ice nucleus spectrum (Fig. 2b).

The method of cloud production used here has the disadvantage that large supersaturations are undoubtedly produced in the immediate vicinity of the warm water source. The use of two source temperatures enables one to explore the effect of changing this local supersaturation. The results for AgI are consistent with the comparatively hydrophobic nature of the

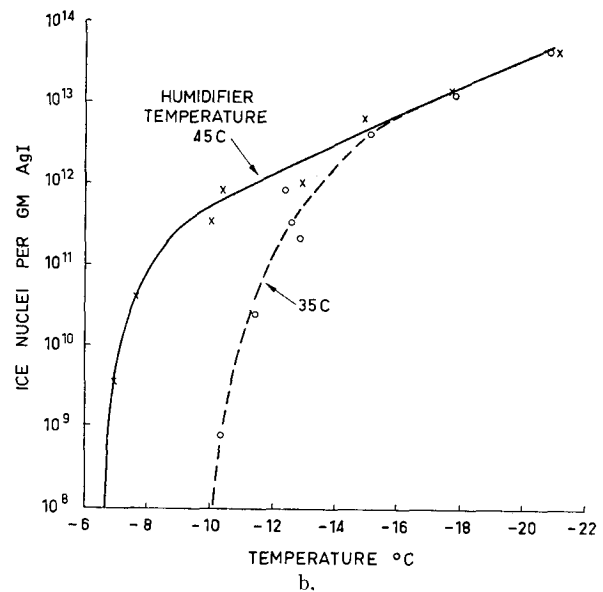
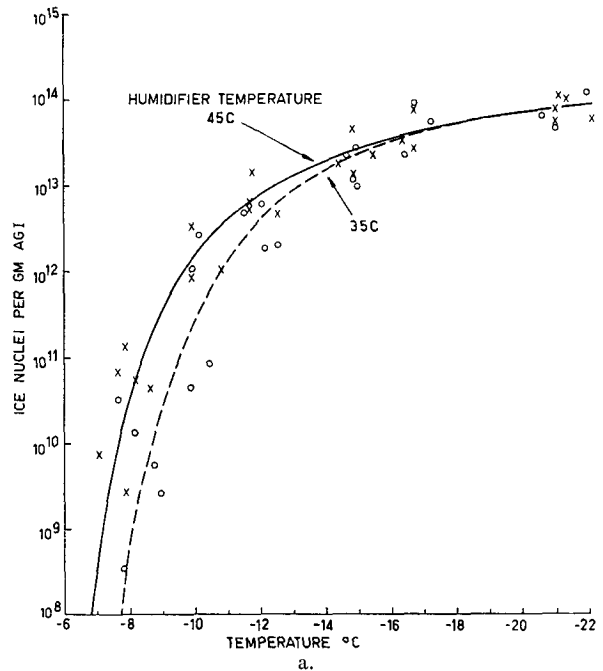


FIG. 2. Ice nucleus spectra for AgI-NaI aerosol, a., and "pure" AgI aerosol, b., in 10-liter mixing cloud chamber, expressed as number of ice nuclei per unit weight of AgI. Humidifier at 45C, solid line (points shown by crosses); humidifier at 35C, broken line (points shown by circles). Data for a. were derived from experiments of 10 April and 16, 18 and 23 May 1967, and for b. from experiment 20 May 1966. The behavior of the AgI aerosol, b., as a condensation nucleus is shown in Fig. 1.

particles as shown by Fig. 1 and with the idea that higher supersaturations increase the number of AgI particles that can nucleate droplet condensation and then act as freezing nuclei. With the mixed aerosols the effect of humidifier temperature is much less pronounced. Therefore, the results using a source temperature of 35C can be applied to natural clouds without fear of large errors. Rather, the main uncertainty arises in the measurement of the effective temperature in the cloud chamber, where, as we have seen above, a possible error of 1.5C exists.

4. Comparison between measured AgI-NaI ice nucleus spectrum and theoretical predictions

The only comprehensive theoretical treatment of nucleation of water by AgI is that of Fletcher (1959a, 1962). Because of the hydrophobic nature of AgI, he held that it must produce ice crystals direct from the vapor phase rather than by a condensation-freezing process. He gives curves relating nucleation temperature to AgI particle size for this "sublimation" process. These are based upon a threshold nucleation temperature of -4°C for large AgI particles (100μ radius). In view of the evidence (Bryant *et al.*, 1960; Edwards and Evans, 1960) that this threshold applies only to the freezing process, and that nucleation by sublimation is of negligible importance until a temperature of -12°C is reached, it is apparent that these sublimation curves are incorrect.

In dealing with aerosols produced by cloud-seeding burners, Fletcher (1959b) made the working assumption

that they behave in all respects essentially like pure AgI smokes, and nucleate ice by the sublimation process. If we consider the hygroscopic nature of the mixed aerosol particles, it seems that this assumption is unjustified.

Fletcher (1968) has now amended his treatment of nucleation by mixed smokes to take account of their hygroscopic nature. He treats the case of particles produced by burning a solution of AgI and KI in acetone. He shows that when such a particle enters a cloud and takes up water, the fraction of AgI which dissolves is negligible. The AgI nucleates the drop by the freezing process. He introduces into his equation relating nucleation temperature to particle size another term to express the lowering of the freezing point of the drop by the KI when the drop is in equilibrium with an environment at water saturation. In this way he develops new curves for the nucleation temperature of supercooled water by these mixed particles as a function of the radius of the AgI component. His curve for an AgI-KI complex is shown in Fig. 3 (curve B) along with the curve for pure AgI (curve A).

We have now applied Fletcher's new treatment to the AgI-NaI aerosols used in our cloud-seeding work. The NaI in the smoke particles is assumed to have one-quarter the mass of the AgI component, as in the original acetone solution. In these calculations, a value of 20 ergs cm^{-2} was used for the surface free energy of the water-ice interface, following Fletcher (1962). The relationship between nucleation temperature and the size of the AgI freezing nucleus is shown in Fig. 3

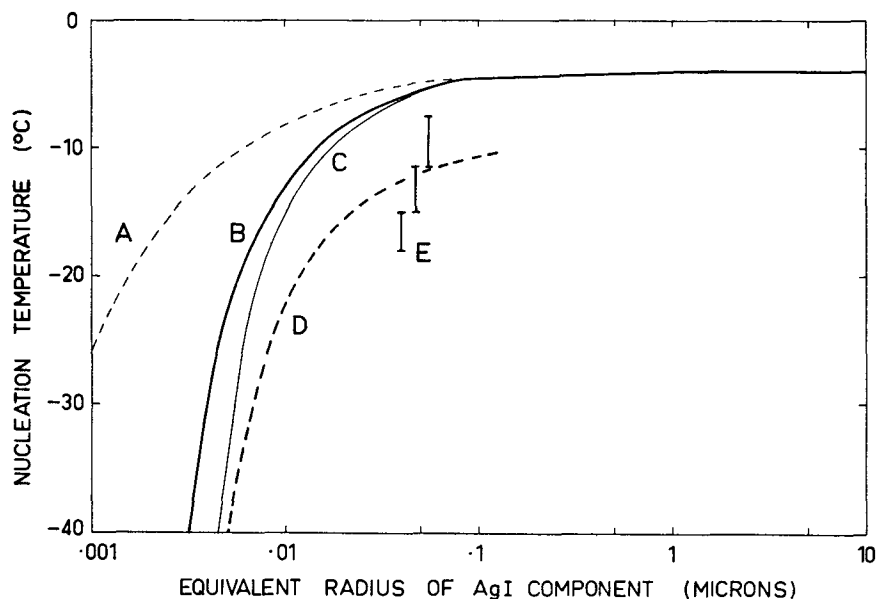


FIG. 3. Relationship between size of AgI nucleus and nucleation temperature: A, AgI as freezing nucleus (Fletcher, 1968); B, freezing promoted by AgI-KI (Fletcher, 1968); C, curve for mixed aerosol of AgI, NaI in ratio 4:1 by weight, calculated after Fletcher (1968) assuming 1μ radius particle acts as a freezing nucleus at -4°C ; D, as C, but assuming that 0.125μ radius particle causes freezing of water at -10.4°C ; and E, measured median radius of central nuclei of ice crystals nucleated by AgI-NaI aerosols in temperature ranges indicated by vertical bars.

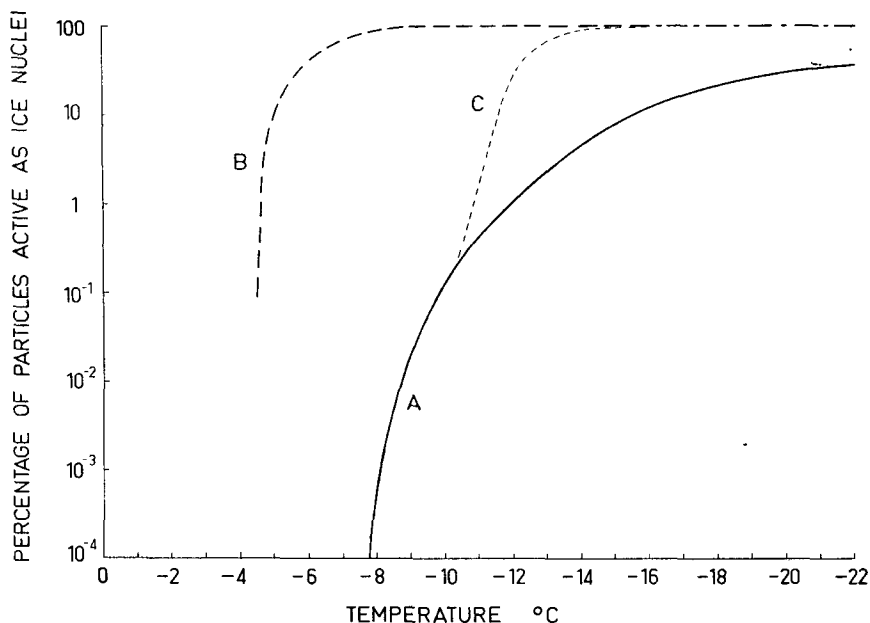


FIG. 4. Freezing nucleus spectra for AgI-NaI aerosols: A, as measured in 10-liter mixing chamber using 35C humidifier; B, as calculated following Fletcher (1968) assuming nucleation by a 1μ radius particle at -4C ; C, as calculated following Fletcher (1968) assuming nucleation by a 0.125μ radius particle at -10.4C .

(curve C). No correction has been made for the small amount of AgI which goes into solution.

By combining the theoretical relationship between size and nucleation temperature with the size distribution of our AgI-NaI smoke particles, we obtain a freezing nucleus spectrum (Fig. 4, curve B) which can be compared with that actually measured (Fig. 4, curve A). It is apparent that the theoretically derived curve overestimates the activity of our aerosol by at least three orders of magnitude for temperatures $> -10\text{C}$. Even if we add the full 1.5C possible correction to our cloud-chamber temperatures, there is still a wide difference between the two curves.

One possible explanation of the discrepancy is that our AgI-NaI aerosol is intrinsically poorer as an ice nucleator than the AgI for which a threshold of -4C has been found. The assumption that a particle of 1μ radius acts as a freezing nucleus at -4C leads to a value of 0.85 for Fletcher's parameter M . If instead, we use our own observation that the largest 0.2% of our AgI-NaI particles have diameters $\geq 0.25\mu$, and assume that these particles constitute the most effective 0.2% of all nuclei and therefore cause freezing at -10.4C and over, we find that $M=0.5$. The relationship between AgI particle size and nucleation temperature calculated on this basis is shown in Fig. 3 (curve D). This approach allows a test of Fletcher's theory that does not involve the uncertainty in cloud-chamber temperature measurement.

From this new curve and the known size distribution we have recalculated the freezing nucleus spectrum, as shown in Fig. 4 (curve C). Comparing this with the

actual measured freezing nucleus spectrum, we find, of course, an enforced agreement at the temperature -10.4C . At lower temperatures the two curves deviate considerably, the measured nucleating efficiency being much lower than the theory would indicate. This lack of agreement brings seriously into question the validity of Fletcher's treatment.

5. Direct investigation of the relationship between size and nucleation temperature

Besides following Fletcher's treatment to deduce theoretically the relationship between particle size and nucleation temperature for AgI-NaI aerosols, we have also investigated this relationship in the laboratory. In this work we followed the method used by de Pena (1964), in which ice crystals formed in a cloud chamber at a given temperature are allowed to fall upon Formvar-coated electron microscope grids. The Formvar is then exposed to chloroform vapor so that a replica of each ice crystal is made. This is subsequently examined in an electron microscope and the diameter of the AgI particle at the center of the ice crystal is measured. De Pena worked with particles produced by spraying an acetone solution of AgI-NaI.

We have carried out experiments on similar lines using the 10-liter cloud chamber with 35C humidifier in the manner described above. Crystals nucleated by AgI-NaI aerosol particles were replicated at chamber temperatures of -11.5 , -15 , and -18C . The crystals were about 15μ in diameter, mostly hexagonal, with a few short columns at the highest temperature. In all

cases there was an electron-dense particle at or very near to the geometrical center, and this was assumed to be the ice nucleus. In a number of cases there were additional electron-dense particles more remote from the center. The size distributions of the central particles are shown in Table 1. By plotting these data on log-probability paper, we obtained smoothed size distributions. Then by making use of Fig. 4 (curve A), we find that nuclei active at -11.5C constitute 10% of all those active at -15C , and thus by subtraction of the appropriate numbers in each size interval, we can find the size distribution of nuclei active in the temperature interval -11.5 to -15C , and similarly for the range -15 to -18C . In this way the size distributions shown in Fig. 5 were obtained. The nuclei active at -11.5C may be assigned to the interval -7.5 to -11.5C since no nuclei active at warmer temperatures have been found. These data are summarized in Table 2 and are plotted in Fig. 3 (data points E).

We see from Fig. 5 that nuclei active in the three temperature ranges span virtually the entire size range in each case. As others have remarked before (Edwards *et al.*, 1962), particles of the same size show a wide range in nucleating ability. This is at variance with the basic assumption of Fletcher's theoretical treatment that there is a unique relationship between nucleus size and nucleation temperature.

Furthermore, it is apparent from Fig. 3 that the nucleating ability of our aerosol particles decreases much more rapidly with size than is indicated by the Fletcher theory. This was already implicit in the comparison of curves in Fig. 4. Over the size range examined in the present work we would expect the nucleation temperature to be depressed about 1C , a negligible effect compared with the observed depression of about 7C for a decrease in median particle size of about 30%.

Fletcher's treatment of heterogeneous nucleation, as illustrated by the curve for AgI as freezing nucleus in Fig. 3 (curve A), is to consider how a particle which possesses a nucleating site intrinsically active at -4C becomes increasingly inefficient as the curvature of the

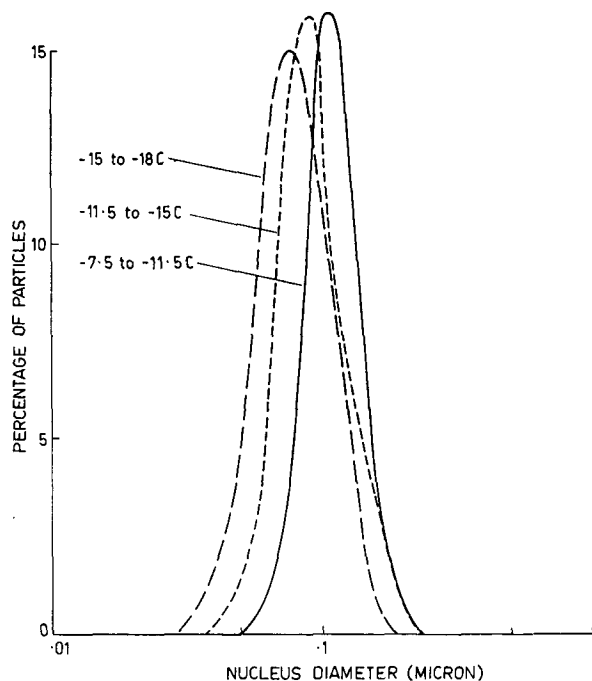


FIG. 5. Size distribution of central nuclei found in ice crystal replicas nucleated by AgI-NaI aerosols in various temperature ranges.

particle increases. Objections have been raised that this purely geometrical approach ignores the basic physical nature of the nucleation process, dependent as it is upon the occurrence of nucleating sites of widely differing efficiencies upon the surface of the nucleating material. Edwards *et al.* (1962) and Katz (1962) explain the effect of particle size on nucleation temperature by postulating that nucleation sites are randomly distributed over the surface of the nucleating material, the probability of finding a site on a given particle being a function of the area of the particle only. It follows that, though large particles have a higher probability of possessing a nucleating site, there is no certainty that an individual large particle will possess a nucleating site.

Since this hypothesis qualitatively explains the present results it is of interest to test it quantitatively. In Fig. 5 we show the size range of nuclei active over the temperature range -7.5 to -11.5C . Knowing the size distribution in our original smoke we can determine the

TABLE 1. Size distribution of AgI-NaI particles active at various temperatures.

Diameter range ($10^{-2} \mu$)	Percentage of particles at temperature of		
	-11.5C	-15C	-18C
3-4	0	0	1
5-6	0	6.1	12
7-8	11	24.8	28
9-10	20	26.1	26
11-12	23	15.4	18
13-14	26	12.7	8
15-16	13	8	4
17-18	4	4.3	2
19-20	1.4	1.8	1
21-22	1.4	0.8	0
Total number	70	130	100

TABLE 2. Size of AgI particles active as ice nuclei in given temperature intervals.

Temperature interval ($^{\circ}\text{C}$)	Median diameter of nucleus (μ)	Minimum diameter of nucleus (μ)
-7.5 to -11.5	0.115	0.05
-11.5 to -15	0.095	0.04
-15 to -18	0.08	0.028

ratio

$$R_D = \frac{N_D}{P_D},$$

where N_D is the percentage of particles of diameter D found among the nuclei active at -11.5°C and P_D is the percentage of particles of the same size present in the original smoke. This ratio is then a measure of the probability of a particle of the given size possessing an active nucleating site, and should be proportional to the particle surface area if the theory is correct.

In Fig. 6 we have plotted R_D against particle surface area assuming that the particles are spheres. The data are grouped in 0.03μ diameter intervals and ascribed to the mean diameter in each interval.

It will be seen from Fig. 6 that the probability of a particle possessing an active nucleating site is roughly proportional to surface area for the smaller particle sizes. Above a particle diameter of 0.18μ , however, this linear relationship no longer holds good. Whether this is a real effect indicating that the largest particles have a lesser probability of possessing active sites is doubtful, in view of the small number of particles in the original data (only 2 out of 70 active at -11.5°C were of diameter ≥ 0.18).

A more searching test of this hypothesis was provided by Edwards *et al.* (1962), who showed that the probability of possessing a nucleation site increased linearly with particle area for monodisperse AgI particles ranging from 0.17 – 3.5μ in diameter.

6. Conclusions

In the above work we have attempted a comprehensive study of the properties and behavior of the AgI-

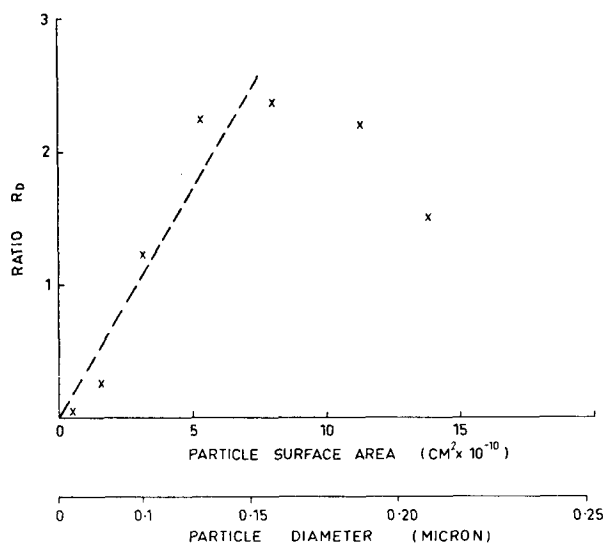


FIG. 6. The relationship of the ratio R_D , which is a measure of the probability of a particle containing an ice nucleus active at a given temperature (-11.5°C), to the surface area of the particle.

NaI cloud-seeding aerosols used in Australia and some other countries.

Individual particles are found to be of mixed composition and are hygroscopic in nature. When introduced into a cloud they will take up water in sufficient quantity to cause all the NaI and a minor fraction of the AgI to dissolve. If the temperature is low enough the AgI will cause the drop to freeze, the dissolved salts depressing the freezing point by an amount that will depend on their concentration at the time of freezing. To this extent the NaI decreases the nucleating activity of the AgI. On the other hand its original action as condensation nucleus plays an essential part in assisting the AgI to nucleate by the freezing process rather than the much less efficient sublimation mechanism.

Because of their ability to take up water at and below saturation these aerosols are not very sensitive to the supersaturation under which their ice-nucleating ability is tested.

The behavior of "pure" AgI aerosols is quite different. In order to nucleate ice by the condensation-freezing process they require higher supersaturations, because of their much more hydrophobic nature. Cloud-chamber supersaturations greater than those found in natural clouds will therefore exaggerate their ice-forming ability. Such supersaturations are common in experimental chambers that produce cloud by using i) rapid, large expansions, ii) high-temperature sources of water vapor, and iii) sprayed water drops at a temperature higher than that of the chamber. They can also be produced by injecting the test aerosol along with a warm moist carrier gas into the cold chamber.

In the case of new methods of dispersing AgI, for example by means of rockets and explosives, there is very little evidence on the composition or hygroscopic nature of the particles produced. Tests of their nucleus output per gram should therefore be made under conditions that avoid supersaturations of more than 1–2%.

The ice nucleus output of our cloud-seeding generators has been compared with Fletcher's theory as modified by him to take account of the freezing-point depression by the soluble component of the aerosol particles. For the measured particle size distribution, Fletcher's theory predicts a higher output than that observed, by at least three orders of magnitude at temperatures $> -10^\circ\text{C}$. If the parameter M in the nucleation equation is altered to give agreement with the observations at about -10°C , then the shapes of the theoretical and experimental ice nucleus spectra are quite different at lower temperatures.

Direct measurement of the size of nuclei active at known temperatures support this evidence that the nucleating ability of AgI-NaI falls off much more rapidly with decreasing particle size than indicated by Fletcher's treatment. It is also found that the sizes of particles active at any given temperature span almost the entire size range present in the aerosol, so that a

unique size for each nucleation temperature cannot be assumed.

In views of these shortcomings of Fletcher's theory our data was used to test an alternative hypothesis that nucleation sites are randomly scattered on the surface of the nucleating material. The probability that any particle will possess a nucleating site active at a particular temperature will then be proportional to its surface area. Some support for this hypothesis is provided by our data, taking into account the narrow size range and comparatively small number of particles in the sample.

Acknowledgments. We acknowledge with thanks the assistance of Dr. A. Ono and Messrs. N. S. C. Thorndike and K. Davidson and the late K. J. Heffernan in the experimental work described above. Dr. S. Twomey supervised the cloud nucleus measurements. We are indebted to the Sydney University Electron Microscope Unit, and to C. Shorey in particular, for the electron microscopy.

REFERENCES

- Battan, L. J., 1965: A view of cloud physics and weather modification in the Soviet Union. *Bull. Amer. Meteor. Soc.*, **46**, 309-316.
- Bryant, G. W., J. Hallett and B. J. Mason, 1960: The epitaxial growth of ice on single-crystalline substrates. *J. Phys. Chem. Solids*, **12**, 189-195.
- de Pena, R. G., 1964: Étude des noyaux glaçogènes artificiels au microscope électronique par la méthode des répliques. *J. Rech. Atmos.*, **1**, 122-131.
- Edwards, G. R., and L. F. Evans, 1960: Ice nucleation by silver iodide I. *J. Meteor.*, **17**, 627-634.
- , and —, 1968: Ice nucleation by silver iodide, III. *J. Atmos. Sci.*, **25**, 249-256.
- , — and V. K. La Mer, 1962: Ice nucleation by monodisperse silver iodide particles. *J. Colloid. Sci.*, **17**, 749-758.
- Fletcher, N. H., 1959a: On ice-crystal production by aerosol particles. *J. Meteor.*, **16**, 173-180.
- , 1959b: The optimum performance of silver iodide smoke generators. *J. Meteor.*, **16**, 385-387.
- , 1962: *The Physics of Rainclouds*. London, Cambridge University Press, 386 pp.
- , 1968: Ice nucleation behavior of silver iodide smokes containing a soluble component. *J. Atmos. Sci.*, **25**, 1058-1060.
- Katz, U., 1962: Wolkenkammeruntersuchungen der Eiskeimbildungsaktivität einiger ausgewählter Stoffe. *Z. Angew. Math. Phys.*, **13**, 333-358.
- Linke, W. F., 1966: *Solubilities of Inorganic and Metal-organic Compounds*. Washington, Amer. Chem. Soc., Vol. I, 94-95.
- Mason, B. J., and J. Hallett, 1956: Artificial ice-forming nuclei. *Nature*, **177**, 681-683.
- Mossop, S. C., 1968: Silver iodide as nucleus for water condensation and crystallization. *J. Rech. Atmos.*, **3**, 185-190.
- , and C. Tuck-Lee, 1968: The composition and size distribution of aerosols produced by burning solutions of AgI and NaI in acetone. *J. Appl. Meteor.*, **7**, 234-240.
- Smith, E. J., J. A. Warburton, K. J. Heffernan and W. J. Thompson, 1966: Performance measurements of silver-iodide smoke generators on aircraft. *J. Appl. Meteor.*, **5**, 292-295.
- Tompkins, L. M., D. A. Muus and T. Pearson, 1963: Water adsorption in the system AgI-KI-H₂O. *J. Geophys. Res.*, **68**, 3537-3539.