Field Observations of the Persistence of AgI-NH_4I-Acetone Ice Nuclei in Daylight

ARLIN B. SUPER AND JACK T. MCPARTLAND

Division of Atmospheric Water Resources Management, Bureau of Reclamation, Miles City, Mont. 59301

JAMES A. HEIMBACH, JR.

Department of Earth Sciences, Montana State University, Bozeman 59715

(Manuscript received 6 June 1975)

ABSTRACT

A field method of estimating the persistence of a commonly used silver iodide seeding agent is described. The method involved measurement of the AgI plume structure at two downwind distances from the ground generator(s). Distances between the nine available pairs of downwind measurement planes ranged from approximately 10 to 100 km. An NCAR acoustical ice nucleus counter in a light twin aircraft was used to sample the AgI plumes. A series of passes was made through the entire vertical and horizontal extent of the plume at each downwind distance. These measurements, together with pilot balloon observations, permitted calculation of the flux of AgI through each vertical cross-sectional plane. The difference in flux measurements yielded an estimate of the persistence of the seeding agent over the period of transport between the two vertical planes.

This method was applied at three separate locations, during different seasons, and with various degrees of cloudiness. Resulting estimates of deactivation rates of the ice nucleating ability of AgI ranged from no loss to 70% loss per hour. The implications for possible cloud seeding effects beyond the intended target area are discussed.

1. Introduction

It has been generally accepted that the ice nucleating ability of silver iodide (AgI) smoke decreases rapidly during exposure to sunlight. Several laboratory investigations have shown this to be the case. These include Inn (1951), Reynolds *et al.* (1951), Vonnegut and Neubauer (1951), Birstein (1952), Reynolds *et al.* (1952), Mason and Hallett (1956), Bryant and Mason (1960), Rowland (1964), and St. Louis and Steele (1968). In addition, Smith and Heffernan (1954, 1956) and Smith *et al.* (1955) utilized airborne measurements to investigate the persistence of smoke produced by both a hydrogen- and a kerosene-burning silver iodide generator. The rate of loss of ice nucleating ability, usually attributed to photolytic deactivation, differed widely among the studies cited, ranging from a factor of two to several orders of magnitude per hour. However, several investigations indicated about one to two orders of magnitude decay per hour.

The silver iodide used in these studies was generally either in the form of relatively large silver iodide crystals, or, more commonly, was produced by burning AgI-NaI-acetone solutions. The latter, which results in a complex different than pure silver iodide, has been widely used in cloud seeding operations and experiments during the 1950's and 1960's.

The field observations to be discussed allowed estimates of persistence of smoke from an AgI-NH_4I-acetone solution which should be in the form of a relatively pure AgI particulate (St. -Amand *et al.*, 1971).

2. Instrumentation and experimental approach

The experiments to be described used an ice nucleating agent produced by spraying a 3% (by weight) AgI, 1% NH_4I, 3% H_2O, 93% acetone solution into a propane flame. Modified Skyfire-type generators were used which are described by Super *et al.* (1972). The rate of AgI consumption was about 30 g h^{-1}. Calibration at the Colorado State University (CSU) test facility indicated that this type of generator and solution produces 6×10^{14} ice nuclei per gram AgI at natural tunnel draft, effective at -20°C (CSU, 1972). Output at maximum tunnel flow (10 m s^{-1} across the burner head) increased to 2×10^{16} nuclei g^{-1}. Data are not available at intermediate speeds. For the burn rate of 30 g h^{-1} used in the experiments to be discussed, the corresponding output range is 0.5 to 17 $\times10^{13}$ nuclei s^{-1}.

An NCAR acoustical ice nucleus counter was used for all AgI concentration measurements. This unit, considerably modified from its commercially manu-

factured configuration, is similar to that described by Langer (1973) except that it has a smaller cloud chamber. Langer reported excellent agreement between an airborne-type counter like that used here and the isothermal cloud chamber at CSU. Comparisons were made using silver iodide from the same type of generator and solution used in this study. Super (1974) found very good agreement between estimated generator output based on the CSU calibration, and downwind ice nucleus flux estimated from measurements made with the NCAR counter utilized in this study.

The method used to estimate the persistence of the silver iodide's ice nucleating ability in the atmosphere relied on estimates of the flux of ice nuclei through two parallel vertical planes, aligned approximately perpendicular to the mean horizontal component of the wind, at different downwind distances. The distances between measurement planes varied from day to day, but corresponded to AgI transport periods greater than $\frac{1}{2}$ h. The observed differences between fluxes yielded estimates of AgI persistence.

At each downwind distance, a series of horizontal passes was made through the AgI plume, following a line approximately across the mean wind direction. The horizontal passes would usually be made at several different elevations starting from one to a few hundred meters above ground level and extending to above the AgI plume. The vertical interval used varied depending on vertical mixing, downwind distance, and flight time remaining. Figs. 1 and 2 illustrate the sampling procedure for two downwind distances on a particular day.

Ice nucleus measurements were extracted from the counter chart recorder at time intervals corresponding to 1 km travel distance or less. The indicated ice nucleus concentration data were multiplied by a factor of 10 to compensate for instrument chamber losses (Langer, 1973). Each data point was assumed to represent the concentration of silver iodide in a rectangular volume, centered on the point. The volume had the crosswind dimension of the distance between data points which was a function of aircraft speed v, was 1 m in downwind extent, and had a vertical dimension determined in the following manner. The ice nucleus data from a particular measurement level were assumed to represent the vertical interval defined on the top by the mean elevation of that level and the next highest measurement level, and on the bottom by the mean of the given level and the next

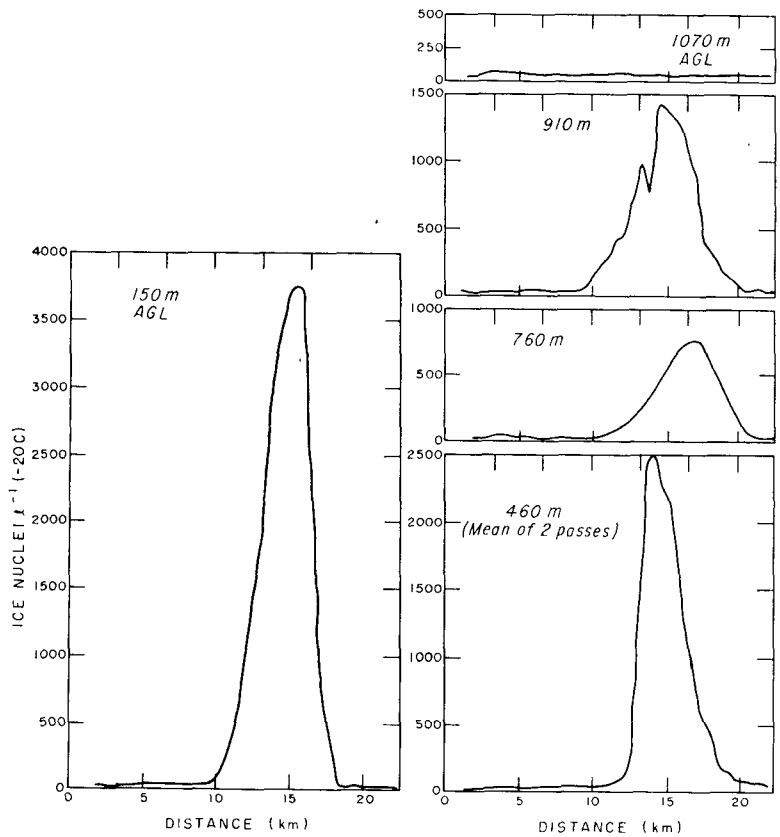

FIG. 1. AgI plume measurements 5 km downwind of generator at Rapelje, Mont., 7 August 1972.

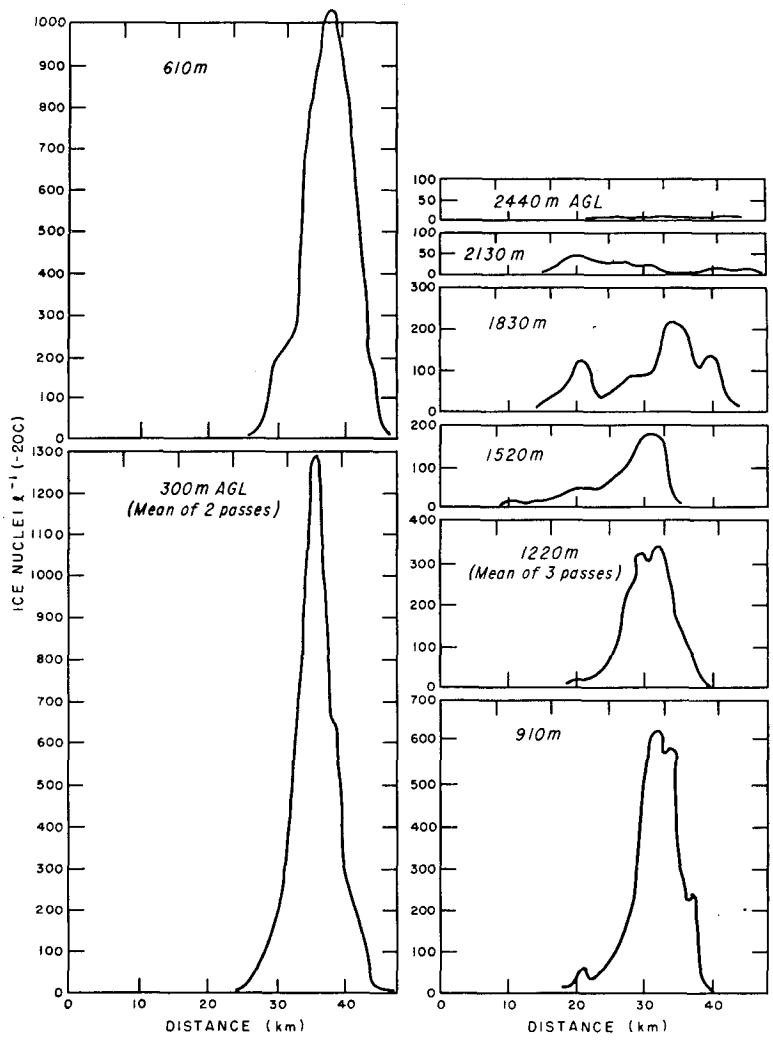

FIG. 2. As in Fig. 1 except for 37 km downwind.

lowest level. The lowest measurement pass was assumed to represent the vertical increment from average ground elevation below the flight path to one-half the vertical distance from the lowest to the next lowest pass. The top of the plume was either actually measured (e.g., by spiral flight) or was assumed to extend 150 m above the elevation of the highest pass. In general, ice nucleus concentrations on the highest passes were close to background levels (<2 nuclei ℓ^{-1}). Thus, any error in the vertical extent to which the measurement was applied would not significantly alter the calculated ice nucleus flux.

The ice nucleus concentration for each rectangular volume was multiplied by the volume, yielding the number of nuclei within it. The total number of nuclei was summed for all similar volumes along each flight elevation, and in several cases, two passes at the same level were averaged to estimate the flux for that layer. This quantity was multiplied by the wind speed (m s^{-1}) at that level. These values were summed for all measurement levels to yield the flux of nuclei s^{-1} passing through the entire vertical cross-section plane. In some cases, the cross-section plane significantly deviated from the crosswind direction for one or more levels and a cosine adjustment was made to estimate the flux along the direction of plume transport.

The formula used to estimate the AgI flux through the layer Δz for one pass was then

$$\text{Flux} = uvT\Delta z|\cos(\theta)|\sum_{i=1}^{N} C_i/N. \qquad (1)$$

This represents an average over N samples of nucleus concentration C_i, taken over the time period T. The flow of material through the sum of the basic volumes is expressed by the wind speed u, and the angle of

the pass to the crosswind plane, θ, is compensated for by the cosine term.

Eq. (1) assumes a negligible contribution by diffusion to the downwind flux where the total downwind flux is

$$\text{Flux}=uC-\left(K_x\frac{\partial C}{\partial x}\right), \tag{2}$$

where u is the mean wind speed and x the downwind coordinate. If the downwind flux due to diffusion, $-K_x(\partial C/\partial x)$, compares in magnitude to the advective flux uC, errors in AgI persistence estimates would result from comparing the flux from (1) at two downwind distances. To show the insignificance of the diffusion term, the following conservative scales are assigned to the parameters involved:

$$\left.\begin{aligned} &u\sim 1\text{ m s}^{-1},\quad x\sim 1\text{ km},\quad C=\text{unity},\\ &\frac{\partial C}{\partial x}\sim\text{one order of magnitude per km }(-0.0009\text{ m}^{-1})\end{aligned}\right\}.$$

A practical estimate of the eddy diffusivity K_x can be found applying the analytical solution to one-dimensional Fickian diffusion (Haltiner and Martin, 1957). This solution is equivalent to the Gaussian diffusion model if

$$\sigma_x=(2K_xx/u)^{\frac{1}{2}}, \tag{3}$$

where σ_x is the downwind Gaussian diffusion coefficient. Assuming isotropic turbulence in the horizontal plane and negligible vertical wind shear allows σ_x to be equivalent to the crosswind Gaussian coefficient σ_y (Roberts *et al.*, 1970). Taking the extreme case of Pasquill stability class A, $\sigma_x\approx\sigma_y=220$ m at 1 km (Turner, 1969). Solving for K_x from (3) gives $K_x=24$ m^2 s^{-1}.

With these values, advective flux has a relative value of 1, and flux due to diffusion contributes 0.02 or almost two orders of magnitude less than that from advection under this extreme example. It is therefore thought justifiable to neglect transport from downwind diffusion in favor of advection.

In all cases, winds aloft were measured by either single or dual theodolite tracking of pilot balloons launched near the generator site. Pibals were tracked at approximately hourly intervals and average speeds were calculated for the period of AgI plume measurement.

Silver iodide persistence estimates were derived from measurements made in three different areas during different seasons. The first measurements were made in south-central Montana near Rapelje during July and August, 1972. A detailed discussion of this work and ice nucleus concentration measurements at each level sampled have been given by Super and McPartland (1973). Either one or five generators were operated at ground level. Pertinent information is summarized in Table 1.

Two estimates of AgI persistence were possible from the tracing of ground-released silver iodide over the

TABLE 1. Summary of ice nucleus flux estimates and associated information.

Date	Location	Number of measurement levels	Approximate height of plume top above generator (m)	Mean wind speed at generator level (m s^{-1})	Cross-section distance downwind of generator (km)	Exposure time between cross-sections (h)	Ice nucleus flux* per generator (10^{13} s^{-1})	Apparent loss per hour (%)	Sky condition
7/24/72	Rapelje	3	1200	3	15	1.8	1.4	~50%	clear
		4	1400		44		0.3		
7/28/72	Rapelje	2	2700	3.5	15	3.6	4.4	~50%	clear
		3	2800		97		0.6		
8/7/72	Rapelje	5	1000	6.5	5	0.8	7.8	none	clear
		8	2300		37		11.0		
2/27/73	Bridger Range	4	900	2	5	0.5	0.5	none	broken
		5	900		16		0.6		
2/6/73	Bridger Range	4	1200	2	5		1.0		scattered to broken
		6	1200		18	0.9	0.9	~10%	
		5	1200		39	1.4	0.8	~10%	
5/3/74	Colstrip	5	1500	>7	2	1.0	8.2	~70%	scattered cirrus
		5	1700	(fan)	18		2.3		
5/22/74	Colstrip	3	700	>7	6		2.3**	none	
		1	3000	(fan)	105	2.4	11.9	~50%	scattered cirrus
		1	3000		190	2.1	3.2		

* Effective at −20°C.
** Generator output reduced—see text.

Bridger Range of southwestern Montana during February and March, 1973. This work is described in detail by Super *et al.* (1974). Resulting data pertinent to silver iodide persistence is given in Table 1.

Silver iodide was released from the top of a 90 m tower at Colstrip in southeastern Montana during April and May, 1974. In this case, the AgI was used as the tracing material for an atmospheric dispersion study concerned with the air pollution potential of large coal-fired plants in semi-rugged terrain (see Heimbach *et al.*, 1975). The AgI generator was similar to those previously used except that a large fan was directed upward at the flame chamber in an attempt to both increase and stabilize the output.

The flux estimates of 22 May 1974, given in Table 1, require elaboration. The generator was hoisted to tower top at 1315 MDT and was operating properly with an AgI consumption of 30 g h^{-1} at that time. However, when the generator was taken down at 1830 MDT, it was discovered that the AgI line was partially plugged. This malfunctioning likely accounted for the relatively low ice nucleus flux 6 km downwind which was sampled from about 1525 to 1550 MDT.

A pair of sampling passes was then made 105 km downwind of the source, followed by another pair 190 km downwind. The latter passes were made between 1730 and 1750 MDT. Aircraft fuel limitations prevented further sampling. Consequently, the fluxes estimated 105 and 190 km downwind were based on sampling at only one level. However, based on previous sampling on similar days (see Super and McPartland, 1973), it appeared reasonable to assume a homogeneous concentration of AgI throughout the vertical extent of the mixing layer. The top of the mixing layer was estimated to be 4000 m MSL from the 1800 MDT rawinsonde at Rapid City, located approximately 145 km from the most downwind passes.

Winds aloft were relatively steady in speed and direction throughout the sampling period. The average speed for the layer was approximately 12 m s^{-1}. At this speed, AgI produced just after the generator was lifted to tower top would have just reached the sampling line by 1740 MDT, the mean sampling time for the two measurement passes 190 km downwind. Consequently, it is likely that quasi-steady-state conditions were not yet established that far downwind, and the estimated flux may be too low. If so, the loss of ice nucleating ability is overestimated in Table 1.

Examination of Table 1 reveals apparent losses of AgI, effective as ice nuclei at $-20°C$, ranging from as high as 70% per hour to none (apparent increase in ice nucleating ability in three cases—see column 8, Table 1). Much, if not all, of the variability in these apparent losses or gains can probably be attributed to the method of flux estimation. It is difficult to determine the accuracy of any given flux estimate as it depends upon the accuracy of the wind speed (pibal) estimates sometimes extrapolated over long distances, the number of measurement levels, ice nucleus counter reliability, and other factors. Also, the generator output varies with wind speed in only a partially known manner.

Perhaps the best estimate of the accuracy of the method is given by comparison among the flux estimates themselves. The sixteen flux estimates presented in Table 1 range from 0.3 to 11.9×10^{13} nuclei s^{-1}, with ten of the estimates between 0.3 and 2.4×10^{13} nuclei s^{-1}. In addition, a total of ten flux estimates were made during winter over the Bridger Range (Super *et al.*, 1974) when wind speeds at the AgI generator were light (<2 m s^{-1}) so AgI output should have varied little. Flux estimates ranged only from 0.5 to 1.0×10^{13} ice nuclei s^{-1} in these cases. Thus, the method is sufficiently accurate to detect any losses in ice nucleation ability as rapid as the one to two orders of magnitude per hour cited in several previous investigations.

Most of the estimates of Table 1 indicate that the AgI lost its ice nucleating ability by no more than a factor of two per hour, and possibly not at all. In fact, at least two of the nine flux comparisons suggested downwind increases in ice nuclei. This finding has importance in the consideration of extra area cloud seeding effects. It would appear that the type of AgI used can persist for long periods in the atmosphere. Thus, if not carried to ground in an intended target area, the AgI might modify clouds located hundreds of kilometers further downwind.

Another consequence of the persistence of this type of AgI is that it can be used as a tracing material over considerable distance. It is noteworthy that plume measurements made 190 km downwind of the generator on 22 May 1974 were approximately 30 times greater than background concentrations. Because of the small size (~0.03 μm), a given mass of AgI can produce a million times as many particles as a tracing material of equivalent density and 3 μm size. It should be possible to trace this type of AgI hundreds of kilometers with a source strength of several hundred grams per hour.

It would be useful to conduct similar experiments using AgI-NaI-acetone as the tracing agent. The rapid photolytic decay rates suggested in earlier, largely laboratory studies should be verified under actual field conditions. If the ice nucleating ability of this type of AgI diminishes rapidly under sunlight, it may be preferable to use it instead of AgI-NH_4I-acetone in situations where downwind contamination is undesirable.

The heights to which the AgI mixed at each downwind distance are also given in Table 1. Dry adiabatic lapse rates were found throughout the layer of mixing during the flights near Rapelje and Colstrip, with the

exception that superadiabatic conditions prevailed near the surface. Lapse rates ranged from moist to dry adiabatic over the Bridger Range. The substantial vertical mixing during summer flights, together with the persistence of the AgI, suggests that ground-based seeding of convective clouds may be feasible under some conditions. Further consideration of this possibility is given by Super and McPartland (1973).

3. Summary

The experimental data obtained under field conditions indicate that the cloud seeding material produced from the AgI-NH_4I-acetone solution does not rapidly lose its ice nucleating ability during daylight. While previous work with AgI-NaI-acetone or large AgI crystals generally showed decay rates of one to two orders of magnitude per hour, usually attributed to photolytic effects, the present work indicates that decay is no more than a factor of two per hour and may even be non-existent. This finding has interesting implications for cloud seeding effects outside the intended target area. It also suggests that the AgI-NH_4I-acetone seeding agent can be used as an inexpensive, mesoscale tracing material.

Acknowledgments. This research was supported by the Bureau of Reclamation under Contract 14-06-D-6798 and by the Montana Power Company.

Grateful acknowledgment is made of the substantial assistance and cooperation of Gerhard Langer of NCAR, and Dave Roger and Jim Stradley of the Gallatin Flying Service, Belgrade, Mont.

REFERENCES

Birstein, S. J. 1952: The effect of relative humidity on the nucleating properties of photolyzed silver iodide. *Bull. Amer. Meteor. Soc.*, **33**, 431–434.

Bryant, G. W., and B. J. Mason, 1960: Photolytic de-activation of silver iodide as an ice-forming nucleus. *Quart. J. Roy. Meteor. Soc.*, **86**, 345–357.

CSU, 1972: Montana State Skyfire ground generator, effectiveness test with AgI, NH_4I solution. Report no. 7204C, Cloud Simulation and Aerosol Lab., Colorado State University, 19 pp.

Haltiner, G. J., and F. L. Martin, 1957: *Dynamical and Physical Meteorology*. McGraw–Hill, 470 pp.

Heimbach, J. A., Jr., A. B. Super and J. T. McPartland, 1975: Dispersion from an elevated source over Colstrip, Montana. Paper No. 75-26.6, *Proceeds. 68th Annual Convention Air Pollution Control Assoc.*, Boston, Mass., 15–20, June 1975.

Inn, E. C. Y., 1951: Photolytic inactivation of ice-forming silver iodide nuclei. *Bull. Amer. Meteor. Soc.*, **32**, 132–35.

Langer, G., 1973: Evaluation of NCAR ice nucleus counter, Part I: Basic operation. *J. Appl. Meteor.*, **12**, 1000–1011.

Mason, B. J., and J. Hallett, 1956: Artificial ice-forming nuclei. *Nature*, **177**, 681–683.

Reynolds, S. E., W. Hume, B. Vonnegut and V. J. Schaefer, 1951: Effect of sunlight on the action of silver iodide particles as sublimation nuclei. *Bull. Amer. Meteor. Soc.*, **32**, 47.

——, —— and M. McWhirter, 1952: Effects of sunlight and ammonia on the action of silver iodide particles. *Bull. Amer. Meteor. Soc.*, **33**, 26–31.

Roberts, J. J., E. S. Croke and A. S. Kennedy, 1970: An urban atmospheric dispersion model. *Proc. Symp. Multiple-Source Urban Diffusion Models*, Research Triangle Park, N.C., 6-1 to 6-72.

Rowland, S. C., 1964: Photolytic activation of silver iodide in the nucleation of ice. *J. Atmos. Sci.*, **21**, 698–700.

Smith, E. J., and K. J. Heffernan, 1954: Airborne measurements of natural and artificial freezing nuclei. *Quart. J. Roy. Meteor. Soc.*, **80**, 182–197.

——, and ——, 1956: The decay of the ice-nucleating properties of silver iodide released from a mountain top. *Quart. J. Roy. Meteor. Soc.*, **82**, 301–309.

——, —— and B. K. Seely, 1955: The decay of ice-nucleating properties of silver iodide in the atmosphere. *J. Meteor.*, **12**, 379–385.

St. -Amand, P., W. Finnegan and L. Burkhardt, 1971: Understanding of the use of simple and complex ice nuclei generated from pyrotechnics and acetone burners. *J. Wea. Mod.*, **3**, 31–48.

St. Louis, P. T., and R. L. Steele, 1968: Certain environmental effects on silver iodide ice nuclei. *Proc. Intern. Conf. Cloud Physics*, Toronto, 26–30 August, 178–182.

Super, A. B., 1974: Silver iodide plume characteristics over the Bridger Mountain Range, Montana. *J. Appl. Meteor.*, **13**, 62–70.

——, and J. T. McPartland, 1973: Feasibility of seeding cumulus clouds with ground-released silver iodide. Tech. Rept. 73-1, Bureau of Reclamation Contract 14-06-D-6798, Montana State University, 95 pp.

——, C. A. Grainger, J. T. McPartland, V. L. Mitchell and R. H. Yaw, 1972: Atmospheric Water Resources Management Program, Final Report—Part I. Bureau of Reclamation Contract 14-06-D-6798, Montana State University, 388 pp. (NTIS PB218705).

——, J. A. Heimbach, J. T. McPartland and V. L. Mitchell, 1974: Atmospheric Water Resources Management Program, Final Report—Part II. Bureau of Reclamation Contract No. 14-06-D-6798, Montana State University, Bozeman, 191 pp. (NTIS PB234012).

Turner, D. B., 1969: *Workbook of Atmospheric Diffusion Estimates*. Nat. Air Pollution Control Agency, Cinncinnati, 84 pp.

Vonnegut, B., and R. Neubauer, 1951: Recent experiments on the effect of ultraviolet light on silver iodide nuclei. *Bull. Amer. Meteor. Soc.*, **32**, 356.