An Interpretation of the Mechanisms of Ice-Crystal Formation Operative in the Lake Almanor Cloud-Seeding Program

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ABSTRACT

In a 1984–85 winter cloud-seeding program at Lake Almanor, California, indium sesquioxide (In_2O_3) aerosol particle generators were collocated with silver iodide (AgI) aerosol particle generators as a source of inert tracer aerosols. The In_2O_3 aerosol served as an indicator of the amount of AgI aerosol scavenged. Based on the aerosol emission rates, if AgI aerosol was only captured by scavenging processes, and played no part in forming ice crystals and snowfall, the silver to indium ratio (Ag:In) in the analyzed snow would be 0.8.

Analysis of snow samples from the target area produced frequent Ag:In ratio values in excess of 1.1. In the snowfall at the closest sampling sites to the aerosol generators, the high ratios of Ag:In cannot be explained by the contact-freezing ice formation mechanism. A mechanism with a much faster rate than possible by contact freezing is necessary to produce the high Ag:In ratios that were observed. Part of the AgI seeding aerosol functioned rapidly to produce ice crystals by a forced condensation-freezing mechanism immediately after generation, and those ice crystals contributed to the snowfall at those sites closest to the generator.

1. Introduction

Evaluation of precipitation enhancement in winter orographic cloud-seeding programs is an important but difficult task in the practice of weather modification. The natural variability in precipitating weather systems and incomplete knowledge of the processes that occur in the atmosphere contribute to this difficulty in evaluating the effects of a snowpack augmentation program.

Methods of evaluating precipitation enhancement in winter orographic cloud-seeding programs based upon the analysis of physical data are under development in the Atmospheric Sciences Center of the Desert Research Institute (for example, Warburton et al. 1985). This paper introduces a method of evaluating hypotheses concerning the *rates and mechanisms* of ice-crystal formation that occurred during a winter cloud-seeding program and the action of the treatment-induced ice-crystal formation process in the development of snowfall. The approach holds promise in evaluating precipitation enhancement results, and in guiding the planning and conduct of future cloud-seeding programs.

2. The field program

The Lake Almanor field program is a long-term, nonrandomized, operational snowpack augmentation program conducted by Pacific Gas and Electric Company in the Lake Almanor watershed of the Sierra Nevada of northeastern California. Figures 1–3 depict the topographic map of the target area. The maps show, for snow sampling conducted at three times, the locations of aerosol generator pairs that operated during storms prior to snow collection and sampling sites where both silver and indium were detected significantly above background.

Silver iodide (AgI) aerosols were generated by combustion of acetone solutions of AgI and ammonium iodide (NH_4I) in auxiliary propane flames from ground-based generators located at high elevations on mountain peaks to the south and west of the target area.

To differentiate between silver content in snow from ice formation events and from nonfunctioning, scavenged AgI aerosol in snowfall, the concurrent release of indium sesquioxide (In_2O_3) aerosol with the AgI aerosol was initiated in the winter of 1983 (Warburton et al. 1985). Indium sesquioxide is water insoluble, and is known not to be an ice-nucleating material. Background indium concentrations in snow are low, about 1 part per trillion (ppt). Indium sesquioxide aerosol was generated by combustion of aqueous alcoholic solutions of indium nitrate [$In(NO_3)_3$] in an auxiliary propane flame using an aerosol generator that is similar to that used for generation of the AgI aerosol. The indium sesquioxide aerosol generators were located 10–20 m from the silver iodide aerosol generators. The aerosol plumes from the two collocated aerosol generators followed nearly identical trajectories. The col-

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located generators were operated to emit 18.75 g h^{-1} of AgI and 12.5 g h^{-1} of In_2O_3, respectively. Proximate coagulation of AgI aerosol particles with In_2O_3 aerosol particles was assumed to be negligible.

If the AgI and In_2O_3 aerosols were incorporated into precipitation by identical mechanisms (namely, aerosol scavenging), then the value of Ag:In in the snow would have been close to 0.83, provided that the precipitation at the site incorporated approximately equal portions of both plumes. Thus, the amount of indium in the snow was a measure of the amount of AgI aerosol in the precipitation, which did not act in ice-crystal formation, but was scavenged.

Transmission electron microscope studies of the two aerosols showed that the particle size distributions were similar for the two aerosols, with mean diameters about 0.06 μm and standard deviations of about 0.02 μm. Atomic absorption spectroscopy, sensitive to 1 ppt, was used for silver and indium analysis. Silver and indium concentrations above 4 ppt were accepted as being significantly above background.

Snowpack sampling was conducted on 20 December 1984, 8 March 1985, and 30 March 1985. Generator operation times pertinent to these periods are presented in Table 1. Atmospheric conditions during the storms preceding the snowpack sampling are represented by the 700- and 850-mb temperatures and winds taken from soundings at Sheridan, California, located near Sacramento, and are summarized in Table 2 (Huggins 1992, personal communication; Hemmer et al. 1985). Generally, low-level winds in the Almanor basin are channeled by topographic features, and temperatures in the study area are usually several degrees colder than at Sheridan.

Snow samples were collected by R. Stone using a careful handling procedure to minimize the potential for contamination. Indium and silver concentrations in the snow samples were also determined by R. Stone by using the techniques described in Warburton et al. (1985), with accuracies to 1 part per trillion (1 part in 10^{12}). Samples were divided into 2-cm depth segments for analysis of silver and indium. Those sections in the snow core at a snow sampling site that contained both Ag and In in concentrations of 4 ppt or greater, called the "qualifying sections," were used in the analysis, and the average silver and indium concentrations and Ag:In ratio for those sections were determined by weighting the individual measured concentrations by the appropriate snow mass. The results are presented in Table 3. During other periods of the storms, the plumes from the aerosol generators missed the sampling site, so those periods are not used in the analysis.

This treatment obtains snow-mass-averaged concentrations of Ag and In during those times when the aerosol plumes from the generator sites contributed to the snow sampling site and removes from analysis those times when the aerosol generators did not contribute.

TABLE 1. Generator operating periods.

Snowstorms in the period 10–16 December 1984	
On	Off
1800 UTC 10 December	10 December 2015 UTC
0001 UTC 11 December	11 December 1100 UTC
0900 UTC 12 December	12 December 2000 UTC
1200 UTC 15 December	15 December 2040 UTC
0200 UTC 16 December	16 December 1300 UTC

Snowstorms in the period 4–7 March 1985		
On	Off	
0400 UTC 4 March	2120 UTC 4 March	except 1500–1600
0200 UTC 5 March	1300 UTC 5 March	UTC

Snowstorms in the period 24–28 March 1985		
On	Off	
0200 UTC 24 March	1150 UTC 24 March	
1600 UTC 24 March	1500 UTC 25 March	except 0300–0400 UTC
0400 UTC 26 March	2000 UTC 26 March	except 1500–1600 UTC
0000 UTC 27 March	2025 UTC 27 March	except 1200–1300 UTC
0000 UTC 28 March	1200 UTC 28 March	

The storms that occurred prior to 20 December 1984 were the most northwesterly of the three cases studied. The Ag:In results obtained from the snow samples are depicted in Fig. 1. Although the 850-mb temperatures were often above 0°C during the rawinsonde soundings at Sheridan, the surface isotherms between Sheridan and Almanor suggest that the aerosol generators located above 6000-ft (1800 m) elevation were operating at temperatures colder than −6°C.

The soundings and analyses available for storms sampled on 8 March 1985 also suggest that the aerosol generators were operated at temperatures colder than −6°C. These storms were associated with low wind speeds, generally from the southwest. The mass-averaged Ag:In ratios for snow samples that contained qualifying sections values are plotted in Fig. 2.

The samples collected on 30 March 1985 included snow from storms that occurred from 24 March to 28 March. Although the thermal, moisture, and kinematic profiles of these storms varied significantly during the period, the winds were predominantly from the southwest and the cloud seeding generators were operated at temperatures colder than −6°C at least part of the time during the series of storms. The mass-averaged Ag:In ratios for snow samples that contained qualifying sections are plotted in Fig. 3.

3. Results

Figure 1 presents the distribution of snow-mass-averaged Ag:In ratios in qualifying sections of the snow

TABLE 2. Atmospheric conditions during storm periods—Sheridan radiosonde conditions.

Date	Time (UTC)	700 mb			850 mb		
		T (°C)	T_d (°C)	Wind (deg/m s^{-1})	T (°C)	T_d (°C)	Wind (deg/m s^{-1})
10 December	1900	−7	−16	215/20	+3	+2	175/20
10 December	2300*	−7	−17	330/10	+2	−1	205/5
11 December	2200*	−2	−9	320/45	+3	+2	330/10
12 December	2000*	−6	M	330/60	+1	−8	330/10
13 December	1900*	−8	M	010/60	−1	−13	025/25
4 March	2100*	−13	−27	280/15	−2	−5	220/5
5 March	0000*	−13	−17	250/15	−2	−4	180/15
5 March	0300	−12	−12	200/10	−4	−4	200/15
24 March	1500*	−4	−4	230/30	+3	+3	220/25
24 March	2100	−5	M	220/35	+1	+1	220/10
25 March	0000	−7	M	200/30	+1	+1	220/10
25 March	0300*	−7	−23	240/20	−1	−8	220/10
26 March	1500*	−7	−7	260/40	−3	−7	180/25
26 March	1800	−6	−6	250/45	−5	−5	170/40
26 March	2100*	−6	−6	250/50	−1	−1	190/50
27 March	0000	−5	−5	250/50	+2	+1	210/55
27 March	0400	−7	−7	250/50	0	0	260/30
27 March	1500	−13	−13	230/35	−2	−2	220/25
27 March	1800	−12	−12	250/35	−2	−6	220/20
27 March	2100*	−12	−13	240/30	−1	−1	210/30
28 March	0000	−12	−12	250/30	−1	−1	220/35
28 March	0300	−11	−20	240/45	−2	−2	240/25

* Sounding taken during storm, but at time when aerosol generators were not on.

cores sampled on 20 December 1984. During this period, northwesterly winds prevailed. Most of the region shows Ag:In ratios well in excess of 0.8, with only one site reporting a lower concentration ratio. The highest value, 13.2, was located at the sampling site farthest to the west.

Figure 2 displays the distribution of snow-mass-averaged Ag:In ratios in qualifying sections of the snow cores sampled on 8 March 1985. At two sites, both Ag and In were not detected above background in any sections. Southwesterly winds prevailed during these storms. Two of the three most western snow sampling sites yielded Ag:In ratios in excess of 14. The Ag:In ratios exceed 0.8 in all but the two easternmost snow sampling sites.

Figure 3 displays the distribution of snow-mass-averaged Ag:In ratios in qualifying sections of the snow cores sampled on 30 March 1985. At three sites, both Ag and In were not detected above 4 ppt in any sections. Southwesterly winds prevailed during these storms. The sites with Ag:In ratios in excess of 10 were located in the western group and at the eastern edge of the target area. Ag:In ratios exceeded 0.8 at all the sites that had qualifying snow sections.

Sounding data at Sheridan, California, near Sacramento, must be applied to the Almanor region with care. Sheridan is located in the broad, low-elevation, central California Valley about 140 km south of Almanor. Almanor is located in a mountain basin at 4500-ft (1400 m) elevation. At 700 mb and above, the Sheridan winds are representative of those at Almanor when consideration is given to the synoptic curvature associated with low pressure systems, and 700-mb temperatures at Sheridan are warmer than those at the same level over Almanor. The 850-mb winds at Sheridan do not reflect the lower-level winds in the Almanor basin. During westerly synoptic-flow conditions, the lower level winds are often channeled through the basin, splitting north and south at Dyer Mountain, just east of Lake Almanor. Under southwesterly synoptic-flow conditions, the low-level flow often exhibits significant southerly or westerly components as a result of local terrain effects.

4. Discussion

a. Mechanisms of ice-crystal formation

The snow-mass-averaged indium concentration in the qualifying sections is a measure of the amount of aerosol scavenging that occurred during those times. Since the AgI and In_2O_3 aerosol particles have similar distributions, the probability of an indium particle being scavenged and arriving in the snowfall at a site approximately equals the probability of a silver particle being scavenged and arriving in the snowfall at the site during the same time interval. If scavenging were the only removal process for AgI or In_2O_3 aerosols, then the Ag:In ratio would be 0.8, based on the aerosol emission rates by the generators.

Hydrophobic silver iodide aerosols have been found in laboratory cloud chamber studies to function predominantly by contact freezing at water saturation and

temperatures above −20°C (DeMott et al. 1983). The rate of ice-crystal formation depended on the product of the nucleus particle concentration and the droplet concentration in the cloud. In the cloud chamber, contact freezing of 7-μm-diameter droplets resulted in 90% of the aerosol particles functioning within about 20 min at −10°C, for a droplet concentration of 4300 cm^{-3}. In the Sierra Nevada, droplet concentrations of 20–75 cm^{-3} with mean diameters of 10–15 μm are typical in winter orographic clouds (Marwitz and Stewart 1982), indicating that the rate of contact freezing in Sierra storms should be about two orders of magnitude slower than in the cloud chamber experiments.

When AgI aerosols act to increase the snowfall at a site, then the Ag:In ratios in the qualifying sections at the site will be greater than 0.8. The effect of contact freezing on the Ag:In ratio may be investigated by assuming that all supercooled drops in the cloud will eventually contain either no AgI or In_2O_3 aerosols or, at most, one such aerosol particle. No multiple scavenging events are considered, since this is a second-order effect. The rates of scavenging of aerosol particles are based on their relative airborne concentrations at any point, which in turn are set by their generation rates. Natural and artificially nucleated ice crystals collect supercooled droplets by riming and fall to the ground as snow. At −6° to −8°C, fewer than 1% of the generated AgI aerosols are effective ice nuclei by contact freezing (DeMott et al. 1983), so the rimed drops (those that freeze only by collection on the ice crystal) are expected to contain an Ag:In ratio of 0.8. Similarly, ice-crystal scavenging of aerosols contributes Ag:In at a ratio of 0.8. Contact freezing, contributing silver but no indium, will slightly increase the Ag:In ratio in the qualifying snow sections.

Studies at Colorado State University on the rates and mechanisms of ice-crystal formation by artificial ice nuclei demonstrated a very rapid ice-crystal formation mechanism termed *forced condensation freezing,* which requires transient supersaturation with respect to liquid water and temperatures colder than −6°C. The forced condensation-freezing mechanism caused aerosols to function almost instantaneously, and at −7° to −10°C the experiments produced higher yields by forced condensation freezing resulting from transient water supersaturation than by contact freezing at water saturation over several tens of minutes (Blumenstein et al. 1987; Feng and Finnegan 1989).

In a field study, Finnegan and Pitter (1988) reported rapid ice-crystal formation in the immediate vicinity of the exhaust plume of a ground-based generator operated at −8°C in a supercooled fog. Rapid ice-crystal formation resulted from AgI aerosol particles functioning at water supersaturation, produced when water vapor formed by combustion of acetone and propane cooled during mixing of the plume with the ambient air. High concentrations of very small ice crystals were observed within 30 s downwind of the aerosol generator.

TABLE 3. Snow-mass-weighted concentrations of silver and indium in snow in qualifying sections, and Ag:In ratios.

Site	Ag concentration (ppt)	In concentration (ppt)	Ag:In
	20 December 1984 snow sampling		
A	116.0	8.8	13.2
B	20.2	7.4	2.7
C	25.1	13.7	1.8
D	11.3	18.7	0.6
E	15.4	12.2	1.3
F	88.4	12.8	6.9
G	49.0	11.2	4.4
H	58.1	10.7	5.4
	8 March 1985 snow sampling		
A	115.0	6.7	17.2
B	33.1	14.8	2.2
C	206.8	14.6	14.2
D	19.5	9.7	2.0
E	6.3	12.9	0.5
F	4.8	9.1	0.5
	30 March 1985 snow sampling		
A	40.1	10.1	4.0
B	102.2	8.6	11.9
E	31.2	9.6	3.3
F	10.4	9.4	1.1
H	116.8	10.9	10.7

In the Lake Almanor program, forced condensation freezing must have occurred at the aerosol generator sites, since it occurs only at temperatures below −6°C during transient supersaturation with respect to water, due to the cogenerated water by combustion of propane and acetone by the generator. At −6° to −8°C, less than 1% of the generated AgI aerosols function by forced condensation freezing (Feng and Finnegan 1989), leaving the remainder to interact with the cloud in another manner. In the snow fallout from this rapid ice-crystal formation, which is expected to occur close to the generator sites, there will be minimal time for scavenging of the generated aerosols and the Ag:In ratio can far exceed the value of 0.8. If the snow close to the generator site resulted only from natural ice-crystal formation processes, but scavenged the aerosol plumes, it would have an Ag:In ratio of 0.8. The possibility of snowfall interacting with only one of the two plumes is small, since the plumes disperse laterally. Also, since each 2-cm section contains snowfall for 30 min to several hours, the possibility of continued interaction with only one of the two plumes becomes very small.

A depression of the Ag:In ratio is possible if a site is located at sufficient distance from the generators that a considerable fraction of the AgI aerosol has already contributed to the snowfall by some mechanism other than contact freezing. Under such conditions, the con-

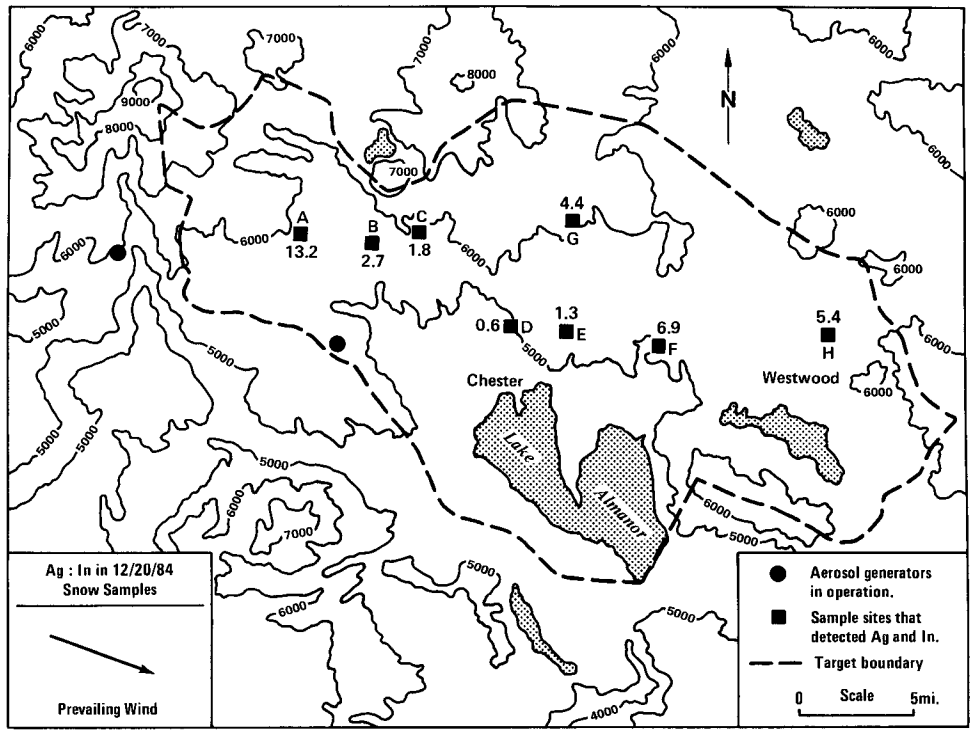

FIG. 1. Map of the Lake Almanor area showing 1000-ft terrain contours, the Lake Almanor study area, locations of operating aerosol generators and snow sampling sites. Results of Ag:In ratio in snow samples taken on 20 December 1984.

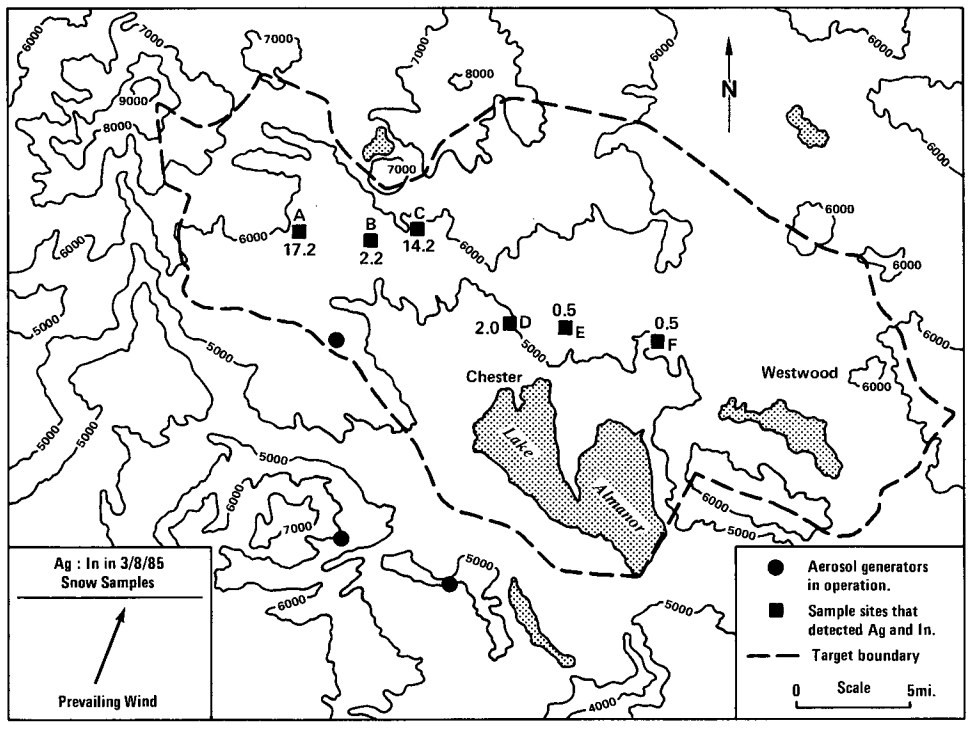

FIG. 2. Same as Fig. 1. Results of Ag:In ratio in snow samples taken 8 March 1985.

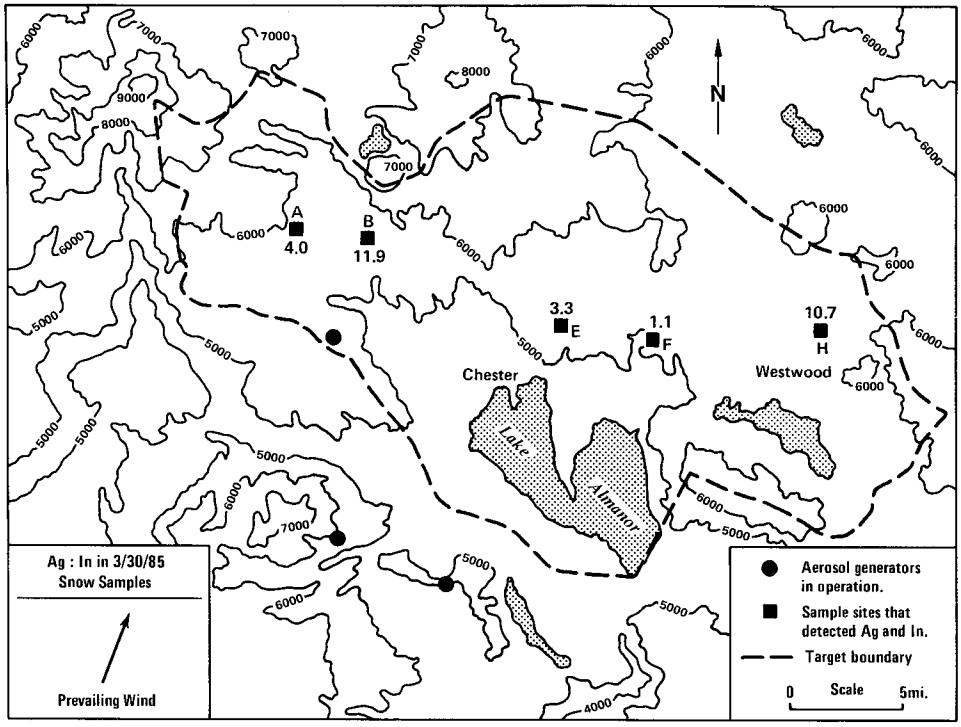

FIG. 3. Same as Fig. 1. Results of Ag:In ratio in snow samples taken 30 March 1985.

tinued scavenging of AgI and In_2O_3 contributes Ag and In to cloud water and ice crystals at less than the initial 0.8 mass ratio, so under some circumstances it is conceivable that even the enhancement of AgI by ice-crystal nucleation may not raise the Ag:In ratio to 0.8.

Recent dynamic cloud chamber work by DeMott and colleagues (DeMott et al. 1992) found that a small fraction of the ice crystals formed from hydrophobic AgI aerosols during adiabatic expansion may have done so before the liquid water cloud was formed. This mechanism was called deposition freezing, which is defined as any mechanism that operates below water saturation. It may have been condensation freezing, due to inadvertent hygroscopicity of the mixed ice forming aerosol particles, or a forced condensation-freezing mechanism induced by supersaturation developed during expansion prior to cloud formation. The uncertainty of this mechanism and its rate precluded further analysis in the present study.

Pitter and Pruppacher (1972) found that immersion freezing is effective only at much lower temperatures than contact freezing, so the immersion freezing mechanism is not evaluated in this study.

b. *High Ag:In ratios*

The high incidence of Ag:In ratios significantly in excess of 0.8 in the snow at various snow sampling sites, shown in Figs. 1–3, indicates that silver iodide aerosols contributed to the snowfall formation by functioning as ice nuclei. The frequent observations with ratios significantly greater than 0.8 suggest that the AgI aerosol did not function by contact freezing, but rather could have functioned by forced condensation freezing. Finally, the high Ag:In ratios in the snow at the sampling sites closest to the generators suggest that a significant amount of ice-crystal formation occurred close to the generators. These results are consistent with the hypothesis that forced condensation freezing occurred on generated aerosol particles.

The observations of high silver concentrations and water mass in the present study are similar to those obtained in the Bridger Range, Montana (Heimbach and Super 1988). In their 1986–87 Snow Pack Augmentation Program, a statistically significant 40% increase in snowpack was claimed within 4 km downwind of the AgI aerosol generators.

Thus, the observational data support the hypothesis that forced condensation freezing was an important mechanism by which generated aerosols function to contribute to snowfall close to the generators in the Lake Almanor program.

c. *Results of low Ag:In ratios*

Silver to indium ratios in snow close to 0.8 (between 0.7 and 1.0) were not found at any site sampled. These results support the hypothesis that a measurable fraction of the AgI aerosol acted to form ice crystals that contributed to the snowfall in the target region. The

study was not conducted in such a manner as to determine what that fraction was.

Cases where the Ag:In ratios were 0.6 or lower were found only three times in the three sampling periods. More data are required to determine the patterns of low Ag:In ratios. As noted above, a low Ag:In ratio may indicate that the AgI aerosol functioned as ice nuclei and contributed to snowfall between the generator and the site, thereby lowering the airborne Ag:In ratio. Subsequent scavenging in the AgI-depleted air, scavenging that contributes chemicals to the snowfall at the snow sampling site, would tend to yield Ag:In ratios of less than 0.8.

5. Summary and conclusions

Snow samples from the Lake Almanor basin were examined for silver and indium concentrations as part of a program to test the effectiveness of AgI aerosols as artificial ice nuclei for increasing precipitation in winter orographic storms. A non-ice-nucleating aerosol, In_2O_3, was used to determine, by analogy, the rate of passive incorporation of AgI into the precipitation.

An Ag:In ratio of 0.8 is expected in snow samples if both aerosols are scavenged and the AgI aerosol does not function to form ice crystals. If AgI functions by a contact-freezing mechanism, the Ag:In ratio is expected to be only slightly higher than 0.8.

The analyses frequently found Ag:In ratios in collected snow samples significantly in excess of 0.8, and as high as 17.2. These observations are consistent with the hypothesis that these hydrophobic AgI aerosols, which would be expected, from laboratory cloud chamber experiments, to function by contact freezing, function by a forced condensation-freezing mechanism in the supersaturated generator plume, almost immediately upon generation. The rapid forced condensation-freezing ice-crystal formation mechanism then leads to increased precipitation close to the generator sites.

Additional information might be gained on the mechanisms of ice-crystal formation by employing a hygroscopic nucleus aerosol of the type AgI · NaCl. Such an aerosol is not expected to affect the results at sites closest to the generators, since forced condensation freezing occurs rapidly and with nearly identical efficiency for hydrophobic AgI aerosols and for hygroscopic AgI · NaCl aerosols, as shown by Blumenstein et al. (1987) and Feng and Finnegan (1989). If the ice-crystal formation mechanism responsible for the downwind peak is freezing of drops with immersed nuclei, then the change in aerosol chemistry would not be expected to change the location of the downwind peak. If the mechanism is condensation freezing, then the use of hygroscopic nuclei is expected to shift the downwind peak closer to the generators.

Summarizing,

(a) Data support the hypothesis that a portion of the AgI ice nucleus aerosol functioned almost immediately on exit from the aerosol generators, by forced condensation freezing in the transient supersaturated plume, and contributed to the snowfall downwind of the generators.

(b) Data support the hypothesis that AgI contribution to snowfall was apparent at all the snow-sampling sites where silver and indium concentrations were found.

(c) Even though the generated AgI aerosol was hydrophobic, data indicated that contact freezing was not the major AgI ice-crystal formation mechanism contributing to the snowfall at the sampling sites in the target area.

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