THE ASSESSMENT OF SNOWPACK ENHANCEMENT BY SILVER IODIDE CLOUD-SEEDING USING THE PHYSICS AND CHEMISTRY OF THE SNOWFALL

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Abstract. A new experimental technique is being developed for assessing whether observed changes in the numbers of ice crystals and the quantities of water arriving at the ground at a specific site and in a given time period, are related to the seeding of clouds passing over that site. The method, based upon a "source-receptor" concept, involves simultaneous releases from the same "source" locations of ice-nucleating silver iodide complexes and non ice-nucleating indium sesquioxide aerosols into orographic winter storm clouds in the Sierra Nevada. These two aerosols have similar particle size distributions. The concentrations of silver and indium in the snow samples collected during the seeding periods at the "receptor" sites are measured, together with the numbers of ice crystals per unit water mass in those snow samples. These measurements are used to determine if any observed changes in the ice crystal deposition are attributable to the seeding process. They are also used for estimating whether there have been any associated changes in the quantities of water which have been precipitated to the ground.

Preliminary results from the first field trials of the technique are given. The mean minimum enhancement (E_{min}) in the number of ice crystals deposited during one 8 h seeded storm period at one sampling site in the 1992-93 winter season was 16%. The results suggest that there may have been an associated minimum precipitation enhancement (P_{min}) around 3% of the natural precipitation. During a second 5 h seeded storm period in the same winter season, the mean minimum enhancement (E_{min}) in the number of ice crystals deposited at that same sampling site was 91%. Results suggest that there may have been an associated minimum precipitation enhancement (P_{min}) for this 5 h period around 7% of the natural precipitation. The data from these two seeded storms also showed that the targeting of the aerosols to the sampling site was good; approximately 85% of the snow samples collected contained silver above the background concentration. The proportion of those same samples which contained indium above its background was much lower (average 35%), indicating that more AgI was being incorporated into the precipitation than In_2O_3. In one of the case studies, it was found that the mean ice crystal mass was higher in the snow samples containing silver above background, than in the snow samples where the silver concentration was not above the background of 2.0×10^{-12} g.ml^{-1}. This is consistent with a process involving the production of new ice embryos by the seeding agent followed by aggregation of the ice crystals into larger ice particles. No such differences were found in the second seeded snow storm study. These investigations are continuing in the central Sierra Nevada region of the U.S.A.

1. INTRODUCTION

The concentration of chemical impurities in precipitation usually decreases with increase in precipitation rate or amount, Junge (1963). However, as shown by Warburton et al.(1986), snowfall from Sierra Nevada storms which had been seeded with silver iodide complexes, often had silver concentrations which increased with precipitation rate and amount. If the aerosols are coming from continuous sources and are being removed entirely by scavenging processes, we might expect to observe chemical concentrations which are directly proportional to precipitation amount. In fact Warburton et al. (1986) showed that this was true for a non-ice-nucleating aerosol of indium sesquioxide. This was not the case for the ice-nucleating aerosol of silver iodide mixtures released at the same times and locations as the indium aerosol. Its departure from the norm was interpreted to be due to some of the silver iodide material being removed by mechanisms other than scavenging. Ice-nucleation followed by ice-crystal aggregation was proposed as a mechanism for producing the observed results. On a more negative note, it has been shown by Warburton et al.(1979) and Super and Huggins (1992), that only 15% to 30% of snow which fell in western U.S. target areas during seeding experiments, contained "seeding silver". If the snow falling in seeded areas not containing "seeding silver" is natural, then the majority of the snow treated in randomized seeding projects as "seeded" would, in fact be "unseeded". This must make it more difficult to detect seeding effects using precipitation statistics.

2. BASIS FOR CHEMICAL AND PHYSICAL ASSESSMENT

In an attempt to improve the "signal to noise ratio" in winter orographic seeding experiments conducted over large target areas, aerosol tracer and chemical techniques have been developed to determine the

possibility of using only the ice-phase precipitation samples containing the seeding chemicals in the target area as "seeded", and those which do not, as "unseeded". Aerosols from AgI.NH_4I and of In_2O_3 are being used for the studies. The silver and indium in the snowmelt are measured by flameless atomic absorption. The background concentrations of silver (B_{Ag}) and indium (B_{In}) in this region are 2.0(σ = 1.0) x 10^{-12} g.ml^{-1} and 1.5 (σ =1.0) x 10^{-12} g.ml^{-1} of snowmelt respectively, (see Warburton et al. 1995a).

3. EXPERIMENTS USING TWO SUBMICRON-SIZED AEROSOLS SIMULTANEOUSLY

3.1 Equipment Details

Initial observations using the two aerosols were made in the Lake Almanor watershed (40 km east-west and 32 km north-south) in the Sierra Nevada of northern California in the winter of 1984-85. This program has been described in detail by Warburton et al. (1995 a,b). All seeding and tracer releases in these experiments were from five pairs of ground-based generators. One generator of a pair used the AgI.NH_4I mixture, the other $In(NO_3)_3$. The generators of each pair were about 50 m apart to minimize coagulation of the two aerosols produced near their sources where the concentrations in the air can be quite high. The aerosols containing silver were produced by burning solutions of AgI.NH_4I in acetone. According to Davis et al. (1975), the active nucleant produced from this mixture is termed phase B_n, which may activate ice growth at temperatures as warm as -1°C. Hence the use of "AgI" in this paper recognizes that the ice nucleant is very probably B_n which is a hydrated double salt. The In_2O_3 aerosols were produced by burning solutions of $In(NO_3)_3$ in ethyl alcohol. No ice crystals were produced when 20 separate samples of In_2O_3 aerosol were placed in a 6.7 m^3 cloud chamber at -20°C. Neither aerosol is hygroscopic.

The snowfall which occurred during the seeding periods in this project, was sampled in 2 cm depth increments immediately following each of the several snowfall events. This was done at the same eight sites in the target area throughout the winter season by a vertical profiling (coring) method described by Warburton et al. (1995a). A 2 cm increment is now referred to as a "slab" of snow. It is important to point out that the observed indium and silver contents of the snowfall depend upon the integrated effects of the aerosols' capture throughout their size spectra. Each 2 cm "slab" of snow represents time periods on the order of one half hour to three hours or more, depending on precipitation rate. It is not possible in this type of study to distinguish between differential rates of removal of aerosols as functions of particle size, nor to differentiate amongst removal by the several scavenging mechanisms,all of which are common to both the ice-nucleating and non ice-nucleating aerosol particles with one exception. B_n can be removed by ice-nucleation but the In_2O_3 cannot. Figure 1 shows the locations of the aerosol generators and sampling sites.

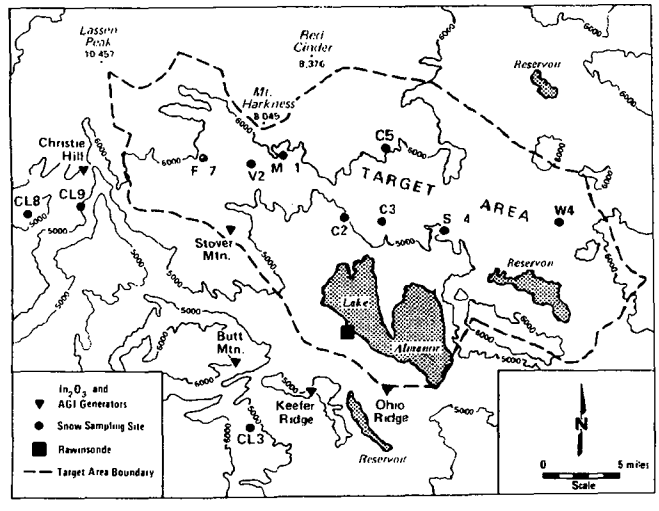

Figure 1. Lake Almanor catchment area showing locations of aerosol generators and snow coring sites used in the 1984-85 expereiments.

Results from Lake Almanor Studies: The particle size distributions of the B_n phase silver iodide and In_2O_3 aerosols used at Lake Almanor were measured by electron microscope analysis of eight samples of each of the aerosols which were collected 10 m from the generator sites on scanning electron microscope grids. The sampling was conducted in non-precipitation conditions at the sites where the generators were permanently located. No attempt was made during these measurement periods to determine particle concentrations in the air nor to determine seeding rates. Analysis of the aerosols sampled, resulted in mean particle diameters of AgI and In_2O_3 of 0.060 μm and 0.065 μm respectively. Both aerosols had the same mass median diameter of 0.066 μm. The geometric standard deviations were 0.021 μm and 0.016 μm for the AgI and In_2O_3 respectively.

Table 1 shows the seeding rates used for both aerosols in this 1984-85 program. These are the mean rates of consumption of the seeding solutions by the aerosol generators as determined throughout the winter season by Pacific Gas and Electric Company (PG&E) professional staff. For the seeding rates shown, the ratio of Ag/In "expected" in the snow, if scavenging alone

were responsible, was calculated to be 0.83:1. The data used to determine this ratio are shown in the Table. The actual Ag/In ratios observed in the snow samples collected at the eight Lake Almanor sites following four seeded storm periods are given in Table 2. Since the experimental ranges of variability in the measured concentrations of silver and indium are ±15%, this can result in experimental standard deviations of 0.2 from the "expected" mean Ag/In ratio of 0.83:1. Using this criterion, there is a 95% confidence level that ratios greater than 1.4 provide evidence of removal processes for silver iodide other than scavenging.

The results show that 22 of the 27 snow cores analyzed contained Ag/In ratios significantly greater than 0.83:1. If more AgI is being removed than In_2O_3, ratios greater than 0.83:1 will occur. Values less than 0.83:1 can occur because the original ratio of release rates has not been maintained due to greater removal of the AgI than the In_2O_3. This occurred at site F7 in the second seeded storm period, at site C2 in the first and second storm periods and at sites C3 and S4 in the third storm period. The nearest generator site to F7 is 13 km distant. However, for the second storm period the closest seeding unit was 24 km to the south. This may provide a reason for the low Ag/In ratio on this occasion compared with the other significantly higher values observed at F7 during the other three seeding periods when the generators closer to this sampling site were in operation. Again, in Table 2 we note that during storm period #3, the four most easterly sampling sites had either no silver detection or Ag/In ratios less than 0.83. This seeding was in cold southerly flow conditions and all four sampling sites were 21 to 35 km north to north east of the nearest generator. Eight of the eleven highest Ag/In ratios occurred at sites within 17 km of the nearest operational generator and six of these were within 11 km range, (see Chai et al.1993).

Table 1. "Expected" Ag/In (mass/cm^2) deposition in Lake Almanor snowfall

Quantity	Silver iodide	Indium sesquioxide
Mean particle diameter:	0.060μm (σ=0.021)	0.065μm (σ=0.016)
Mean particle volume:	1.1 x 10^{-16}cm^3	1.4 x 10^{-16}cm^3
Density:	5.67 g.cm^{-3}	7.18 g.cm^{-3}
Molecular weight:	234.8 g/mole	277.6 g/mole
Mean mass of element/particle:	3.0 x 10^{-16}g	8.5 x 10^{-16}g
Ratio: Indium/silver masses per particle: 2.90:1		
Experimental release rates:	AgI 18.75 g/h	In_2O_3 12.5 g/h
Ratio: AgI/In_2O_3 release rates: 1.5:1		
Release rates, particles/hr:	AgI 2.9 x 10^{16}	In_2O_3 1.2 x 10^{16}
Ratio: AgI/In_2O_3 particles release rates: 2.4:1		
"Expected" observed ratio of masses deposited per unit area in precipitation by scavenging alone:	Ag/In: 0.83:1	

Table 2. Silver/Indium (mass/cm^2) ratios observed in snow profiles collected in Lake Almanor Seeding Experiment Target area.

	Snow Sampling Sites Arranged in order from West to East							
Storm Period	F-7	V-2	M-1	C-2	C-3	C-5	S-4	W-4
1	9.6	2.4	1.8	0.6	1.3	4.4	6.9	5.4
2	0.6	1.3	4.3	0.7	1.4	1.2	2.3	2.5
3	17.2	2.2	14.2	2.0	0.5	ND	0.5	ND
4	4.0	11.9	ND	ND	3.3	ND	1.1	10.7

ND = Silver and Indium not detected above threshold values.

4. DISTINGUISHING BETWEEN SEEDED AND NON-SEEDED SNOWFALL

The results presented in Tables 1 and 2 above suggested the feasibility of using the precipitation chemistry to distinguish between seeded and non-seeded snowfall. By combining this with simultaneous measurements of the numbers and masses of the ice crystals in the same snow samples which contained the seeding chemicals, it may become possible to assess whether there have been enhancements or reductions in ice crystal deposition and whether such changes are related to the seeding.

4.1 Assumptions Made in Ice Crystal Enhancement/Reduction Method

The method makes five assumptions:

(I) The two aerosols (AgI and In_2O_3) have similar size distributions and follow the same general paths to the snow-sampling sites.

(ii) The In_2O_3 particles can only be removed by precipitation scavenging mechanisms.

(iii) The AgI particles can be scavenged and also be captured by ice nucleation.

(iv) The scavenging efficiencies by supercooled drops or ice crystals are the same for both aerosols.

(v) For aerosol release rates used, each supercooled drop can catch, at most, one aerosol particle (either AgI or In_2O_3).

4.2 Theory of Method and Properties to be Measured

In order to determine if any ice crystal enhancement or reduction has occurred and whether any changes have been produced the precipitation amounts, it is necessary to measure or calculate the following quantities.

A. Aerosol Generators:
- G_{agI} = generation rate, AgI aerosol
- G_{inO} = generation rate, In_2O_3 aerosol
- R_o = ratio G_{AgI}/G_{InO} at the origin
- M_{ag} = mean mass of silver in each AgI particle
- M_{in} = mean mass of indium in each In_2O_3 particle

(both masses determined from aerosol size spectra)

B. Snow Samples:
- w_t = total water mass in "slab"
- C_{ag} = silver concentration in "slab"
- C_{in} = indium concentration in "slab"
- N_{ag} = number of ice crystals containing silver
- N_{in} = number of ice crystals containing indium

C. Ice Crystals:
- N_{xt} = number of heterogeneously nucleated crystals/gram of water
- N_{ct} = No. of ice crystals in "slab" $= w_t \times N_{xt}$

We now define the minimum ice crystal enhancement as:

$$E_{min} = \frac{\text{No. of AgI nucleated crystals}}{\text{Total crystals - No.of AgI nucleated crystals}}$$

$$= \frac{N_{Ag} - R_o N_{In}}{N_{ct} - (N_{Ag} - R_o N_{In})}$$

$$= \frac{w_t C_{Ag}/M_{Ag} - R_o w_t C_{In}/M_{In}}{w_t N_{xt} - (w_t C_{Ag}/M_{Ag} - R_o w_t C_{In}/M_{In})}$$

$$= \frac{C_{Ag}M_{In} - R_o C_{In} M_{Ag}}{N_{xt} M_{Ag} M_{In} - (C_{Ag}M_{In} - R_o C_{In} M_{Ag})}$$

$$= \frac{1}{F - 1}$$

Where $$F = \frac{N_{xt} M_{Ag} M_{In}}{C_{ag}M_{In} - R_o C_{In} M_{Ag}}$$

$$= \frac{\text{Total No.of crystals in "slab"}}{\text{AgI nucleated crystals in "slab"}}$$

The term E_{min} is used because of the presence of the ratio of release rates of the two aerosols, R_o, in the denominator of the equation. Since we have no way of determining the actual value of this ratio at times subsequent to the aerosols' release, it is assumed in the calculation that this ratio remains constant for the period of time the aerosols are traveling from their original "source" sites to the snowfall "receptor" site. This could be true both aerosols were being scavenged at the same rates. However, if AgI is also being removed by ice-nucleation, as indicated in the Lake Almanor results presented in Table 2, then the actual aerosol ratios would be smaller than R_o, yielding a value of enhancement higher than E_{min}.

The most difficult property to measure in this method is N_{xt}, the number of individual crystals per gram of water in the melted snow samples. This is because of difficulties introduced into the analysis by riming, aggregation and crystal fracturing.

Earlier studies of ice crystal sizes reaching the ground in the central Sierra Nevada had shown (Deshler,1988), that their mean diameters could be surprisingly uniform over periods of hours. The measurements were made with an aspirated 2DC probe providing continuous data on crystal sizes over the range from 50 to 3000 μm. For the particular study reported by Deshler the mean diameter, after correction for aspiration, was around 550 μm ± 15% over a period of one hour of continuous snowfall.

This study, together with the work of Mitchell et al. (1990), provided some confidence to this present program in using short period sampling methods for obtaining the needed ice particle characteristics. It is fully understood that such short term sampling could result in atypical crystal diameters used in the estimation of ice crystal enhancement, but because of the need to determine both ice crystal masses and the principal dimensions of the ice crystal types, it was necessary to design a method which, despite its current shortcomings, would yield both the masses and the mass-dimensional relationships to be used in the E_{min} calculations. Initially, therefore, N_{xt} is being determined from microphotographs of the precipitating ice crystals during the snowfall periods, and of the water drops produced when those same ice crystals are melted. The experimental procedures used for deter-mining N_{xt} have been described in detail by Pitter et al. (1994). Firstly the principal ice crystal habits are identified using the Magono-Lee (1966) method of classification. In cases of small aggregates, the number of individual crystals in each aggregate is counted prior to melting. For those aggregates which are too large to count all of the individual crystals, (e.g., needle haystacks which occur frequently in Sierra Nevada snowfall), the melted drop is used to determine the mass of that aggregate and the ice crystal photograph is used to identify the sizes and

shapes of the individual ice crystal types in the aggregate. The masses of those individual ice crystal types in the aggregate are then determined from the mass-dimensional relationships developed by Mitchell et al. (1990) for Sierra Nevada snowfall, and the total number of crystals needed to account for the aggregate mass is calculated. It has been estimated that the values of N_{xt} being determined by these methods have an observational and measurement error of ±20%.

5. ESTIMATING PRECIPITATION CHANGES WHICH MAY BE RELATED TO SEEDING

From the definition of minimum ice crystal enhancement, we have:

$$E_{min} = \frac{\text{No of AgI nucleated crystals}}{\text{No of natural crystals}} = \frac{\text{No of AgI nucleated crystals}}{(w_t.N_{xt} - \text{No of AgI nucleated crystals})}$$

Now, all "slabs" of snow in a profile or core which has been collected immediately following a seeded storm period, and which do not contain silver above background, form an "unseeded" comparison set which is used for calculating changes in precipitated water at the site, during that period. This is done as follows.

First, we determine for the "n" unseeded slabs (i = 1 to n), $N = \Sigma w_{ti}.N_{xti}$ = Total number of crystals in the unseeded "slabs" and $W = \Sigma w_{ti}$ = Total water in these unseeded "slabs". Hence, W/N = average crystal mass in the unseeded "slabs", $= m_{xt,ns}$.

We then calculate the natural number of crystals in each "slab" <u>containing silver above the background</u> as follows: $N_{natural}$ = Total number of crystals in "slab", minus the number of AgI nucleated crystals, $= w_t.N_{xt} - \{w_t.C_{Ag}/M_{Ag} - R_o.w_t.C_{In}/M_{In}\}$.

Now there are two groups of "slabs" in the seeded category, one where both $[Ag] > B_{Ag}$ and $[In] > B_{In}$; the other where <u>only</u> $[Ag] > B_{Ag}$. We can use the expression above to calculate $N_{natural}$ for both groups, noting that for the second group (without indium), the third term in the equation above becomes zero.

We can then calculate the amount of natural water, $W_{natural}$, which should be in a "seeded slab". $N_{natural} \times m_{xt,ns} = W_{natural}$. $W_{natural}$ is the <u>EXPECTED</u> water content assuming there have been no seeding effects for that "slab".

We then measure the actual amount of water in that same "slab" by weighing it. This quantity we denote as $W_{measured}$. Then for that "slab",

$$W_{measured} - W_{natural} = W_{seeded} = W_s$$

where W_s = water difference relatable to the seeding. It is apparent that this assessment method is designed to test the Null Hypothesis: "That seeding has no effect on the precipitation amount".

Hence, for each "seeded" precipitation "slab" analyzed, if $W_{natural} = W_{measured}$, seeding has had no effect; if $W_{natural} > W_{measured}$, W_s is negative, and precipitation is reduced; if $W_{natural} < W_{measured}$, W_s is positive, and precipitation is increased. For a complete snow profile (core) of "n" slabs, in which some (n_s) contain seeding chemicals, and others do not, we have:

$$\frac{\sum_{1}^{n_s} W_s}{\sum_{1}^{n} W - \sum_{1}^{n_s} W_s} = P$$

where $\sum_{1}^{n} W$ is the total measured water in the complete profile, and P is the water enhancement in the profile.

6. ICE-CRYSTAL ENHANCEMENT EXPERIMENTS 1993

The first two preliminary experiments using these new concepts were conducted in 1993 in an area surrounding Lake Tahoe in the central Sierra Nevada as shown in Figure 2 where the natural background of silver is 2.0×10^{-12} g.ml^{-1} and that of indium is 1.5×10^{-12}g.ml^{-1}. Ice crystal and melted water drop micro-photographs and snowfall samples were taken at site "A"; weather stations were at sites "M" and a dual channel microwave radiometer was located at site "R" to guide seeding operations. Three pairs of generators releasing AgI and In_2O_3 were located at sites "G" west of the Sierra Nevada crestline. The aerosol characteristics for these 1993 experiments were as follows:

$M_{ag} = 1.4 \times 10^{-16}$ g; $M_{In} = 2.0 \times 10^{-16}$ g;
Mean AgI Particle Mass = 3.0×10^{-16} g;
Mean In_2O_3 Particle Mass = 2.0×10^{-16} g;
AgI release rate = 11.6 g.h^{-1}; In_2O_3 release rate = 12.5 g.h^{-1}; $R_o = 0.74$.

<u>Results of February 17-18, 1993 Case</u> On this day all three pairs of aerosol generators were operated. These generator sites, from north to south as shown in Fig. 2, are at elevations of 2400 m, 2300 m and 2100 m. The AgI units were activated at 1930 UT on February 17 and continued until 0100 UT, February 18. They

were restarted at 0430 UT and continued until 1400 UT on February 18. The In_2O_3 units at the same sites, operated continuously from 1600 UT on February 17 until 1400 UT on February 18. Note that in this series of experiments, the release rates of the aerosols are different from those given in Table 1 for Lake Almanor. Using the other aerosol details given in Table 1, this leads to an "expected" Ag/In ratio in the precipitation of 0.52:1 from scavenging alone.

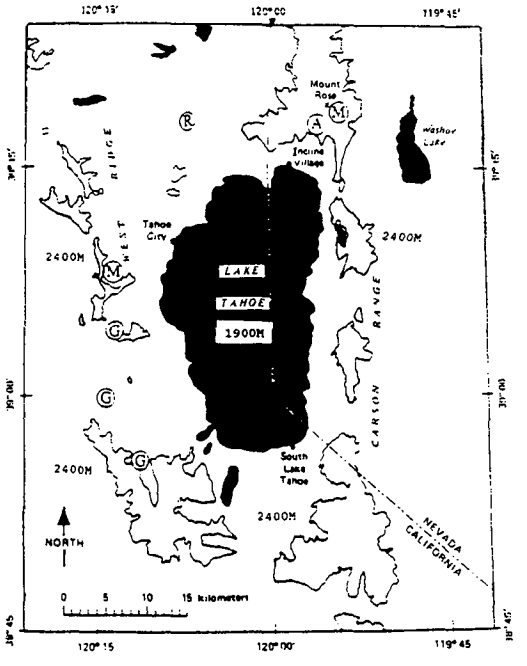

Figure 2. Truckee-Tahoe catchment area showing sites of generator pairs used for producing the AgI and In_2O_3 aerosols, the snow sampling sites and meteorological observing stations.

Falling snow was sampled at site A (elevation 2610m) at 10 to 15 minute intervals from 1930 UT to 0330 UT. The first ice crystal measurements were made at 2000 UT. Weather conditions were monitored continuously at Slide Mountain (elevation 2950m) close to site A (see Fig. 2). Wind directions were from 181° to 210°between 2000 UT and 0430 UT. Temperatures at that weather station ranged from -5.5°C to -6.7°C during the measurement period, being a little warmer toward the end of this time period than at the beginning. This suggests that the temperatures at the aerosol release sites were probably around -1°C. A total of 46 snow samples were collected and analyzed for their silver (Ag) and indium (In) contents, (see Table 3). Each determination consists of 5 separate determinations of 10 aliquots of the snowmelt sample. The values quoted are the means of these 5 determinations.

The reproducibility of these values has been determined from many thousands of such measurements as ±20%. Thirty-four of the samples contained silver in concentrations > (B_{Ag} + 1σ). Eight contained indium > (B_{In} + 1σ). The samples which did not contain Ag were distributed in time throughout the sampling period. All samples which contained Indium occurred during the first three hours of the sampling period.

Table 3. Analysis of snow collected February 17-18, 1993.

Time (UT) Start	End	C_{Ag} (e-12)	C_{In} (e-12)	w_1 g	N_{xt} (e+05)	Principal crystal types
1930	1945	7.3	6.1	111	1.06	small
1945	2000	1.9	6.0	536	1.06	unrimed needles
2000	2015	5.8	7.1	1000	1.61	rime needle aggr.
2015	2030	1.8	5.6	793	2.04	rimed bullets
2030	2040	8.4	3.3	458	1.12	columns rosettes
2040	2050	7.6	5.4	675	0.60	
2050	2100	4.1	7.4	528	1.16	rimed needles
2100	2110	4.2	1.4	659	0.47	columns, hex plates
2110	2120	2.9	1.7	750	0.47	
2120	2130	2.4	1.4	847	0.47	short columns
2130	2140	3.8	1.3	759	0.47	needles
2140	2150	3.7	1.5	730	0.61	dendrites
2150	2200	2.3	0.0	729	0.52	dendrites columns
2200	2210	3.9	1.7	1105	0.15	less riming
2210	2220	5.2	1.6	906	0.61	spatial dendrites
2220	2230	3.8	7.6	888	0.61	
2230	2240	6.4	1.4	548	0.32	rimed hollow colms.
2240	2250	9.0	2.0	852	0.18	needle aggregates
2250	2300	2.9	0.0	916	0.27	rimed needle)
2300	2310	10.6	0.0	962	0.27	haystacks)
2310	2320	11.6	0.0	919	0.27	
2320	2330	3.2	0.0	662	0.41	needle aggregates
2330	2340	3.4	0.0	720	0.41	needles/columns
2340	2350	2.1	0.0	982	0.54	needles, graupel
2350	0000	4.1	0.0	657	0.24	needle haystacks)
0000	0010	3.0	0.0	594	0.24	columns, rimed)
0010	0020	3.4	0.0	735	0.40	No observations
0020	0030	2.8	0.0	826	0.40	rimed haystacks)
0030	0040	3.5	0.0	808	0.25	of needles)
0040	0050	0.6	0.0	915	0.18	rimed haystacks)
0050	0100	4.5	2.1	1026	0.62	of needles)
0100	0110	3.5	0.0	616	0.62	aggeg. short needles
0110	0120	16.6	2.1	1020	0.39	broken dendrites
0120	0130	1.5	0.0	1214	0.44	capped columns
0130	0145	4.3	0.0	1720	0.56	rimed haystacks)
0145	0150	4.9	0.0	448	0.56	of needles)
0150	0200	1.3	1.5	950	0.37	raspberry graupel
0200	0210	4.3	0.0	984	0.79	Aggr. rimed needles
0210	0220	4.3	0.0	907	1.01	Aggr. rimed needles
0220	0230	4.5	0.0	677	0.35	needle haystacks
0230	0240	3.2	0.0	929	0.18	needles plates)
0240	0250	1.2	0.0	896	0.31	graupel)
0250	0300	3.5	0.0	877	0.15	Aggr. rimed needles
0300	0310	3.8	0.0	858	0.15	" " "
0310	0320	3.6	0.0	856	0.15	" " "
0320	0330	4.2	0.0	377	0.15	" " "

After subtracting the backgrounds from each of the measured silver and indium values, it was determined that the observed Ag/In ratios ranged from 0.3:1 to 32:1. There is a 95% confidence level that ratios greater than 1.1 provide evidence of removal processes for AgI other than scavenging.

Thirty-six estimations of the number of ice crystals per gram of melted snow water (N_{xt}), were made during this same time period. The results obtained are shown in Fig. 3. N_{xt} was between 1 and 2 x 10^5 per gram during the first hour of observations and then became relatively steady, somewhat reminiscent of the results of Deshler (1988) mentioned earlier, with values between 2 and 6 x 10^4 per gram for the remainder of the observation period. E_{min} was determined for each of

ICE CRYSTAL MEASUREMENTS
February 17-18, 1993

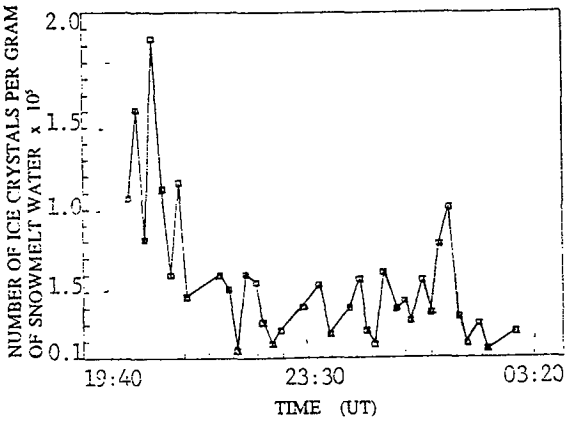

Figure 3. The number of ice crystals per gram of snowmelt (N_{xt}) as a function of time, February 17-18, 1993 at site A in Fig. 2.

the 12 samples which contained both silver and indium above background, (see Table 4). The N_{xt} value used in each calculation of E_{min} was that measured during the time interval of that snow sample collection. The E_{min} values obtained were then weighted by the mass of water in each snow sample. These were summed and the total divided by the total mass of water in all of the snow samples collected during the total 8 h period from 1930 UT, 2/17/93 to 0330 UT, 2/18/93. This weighted mean ice crystal enhancement was calculated to be E_{min} (weighted) = 16%. The six snow samples listed in Table 3 which did not contain silver > 2.0×10^{-12} g.ml^{-1} were used as the unseeded comparison set for calculating the change in precipitated water P at the site during the 8 h period using the following procedure.

First, we determine, for the six unseeded "slabs" (i = 1 to 6) $N = \Sigma w_{ti}.N_{xti}$ = Total number of crystals in the unseeded "slabs". Then calculate: $W = \Sigma w_{ti}$ = Total water in these 6 unseeded "slabs". Hence, W/N = the mean crystal mass of the unseeded "slabs" = $m_{xt,ns}$. Then the natural number of crystals in each "slab" containing silver above the background is calculated as follows: $N_{natural}$ = Total number.of crystals in "slab", minus the number of AgI nucleated crystals $w_t.N_{xt}-(w_t.C_{Ag}/M_{Ag} - R_o.w_t.C_{In}/M_{In})$.

Now there are two groups of "slabs" in the seeded category, one where both Ag and In are above their background concentrations and the other where only Ag is above its background concentration.

For the first group, we can use the expression above to calculate $N_{natural}$, and for the second group we will assume that all of the crystals in the "slab" are natural. This has the effect of yielding a minimum enhancement figure for P which we denote P_{min}.

Table 4. The February 17-18, 1993 case for those snow samples for which E_{min} can be calculated.

Time (UT)	Ag:In ratio	N_{xt} (e+05)	f	E_{min}	w_t(g)
1930-1945	1.15	1.06	4.98	25	111
2000-2015	0.68	1.61	24.18	4	1000
2030-2040	3.56	1.12	2.81	55	458
2040-2050	1.44	0.60	2.29	78	675
2050-2100	0.36	1.16	-16.93	- 6	528
2110-2120	4.50	0.47	8.11	14	750
2200-2210	9.50	0.15	1.15	652	1105
2210-2220	32.00	0.61	2.65	60	906
2220-2230	0.30	0.61	-6.22	- 14	888
2240-2250	14.00	0.18	0.37	-160	852
0050-0100	4.17	0.62	3.88	35	1026
0110-0120	24.33	0.39	0.38	-161	1020

Expected scavenging alone Ag:In ratio = 0.52

Weighted E_{min} (observed) = 16%
Range of actual E_{min} (assuming 20% error in N_{xt}) = 12% to 20%

Mean crystal mass - all snow "slabs" collected not having Ag concentration above background,
= 1.5 e-05 g/crystal

Mean crystal mass - all snow "slabs" collected having Ag concentration above background,
= 2.4 e-05 g/crystal

For the February 17-18 case, the minimum enhancement of the natural (P_{min}) was estimated to be +3%. The combined errors in N_{xt}, C_{Ag}, C_{In} and w_t, suggest that the P_{min} value of +3% has an error of ±60% of that value.

For the "slabs" containing silver above the background, it has been determined that the mean ice crystal mass in those "slabs" was 2.4×10^{-5}g. For those "slabs" where the silver concentration is not above the background, the mean ice crystal mass was calculated to be 1.5×10^{-5}g. This result would be expected if the growth process first proposed by Warburton et al. (1986) is in effect.

This process involves the production of larger numbers of ice particle embryos by the seeding aerosol particles, leading to small ice crystal growth, followed by aggregation of these crystals into larger crystal agglomerates. This process has already been shown to be consistent with other observations of increases in silver concentration with ice particle size and precipitation rate, (see Warburton et al.1995a).

Results of March 24, 1993 Case: All three pairs of generators were used on this day. The AgI units operated from 0600 UT to 1930 UT and the In_2O_3 generators operated from 0330 UT to 1930 UT. The AgI and In_2O_3 release rates were the same as for the

February 17, 1993 case, with a ratio $R_o = 0.74$. Weather conditions were monitored continuously at Slide Mountain Weather Station where wind directions ranged from 206°to 217° and temperatures were between -4.4°C and -5.9°C in these time periods.

Ice crystal sampling and snow sampling occurred at Site A in Fig. 2 at 10 min. intervals. A total of 28 "slabs" of snow were collected between 0745 UT and 1240 UT on this day and analyzed for their silver and indium contents, (see Table 5). Ten of these "slabs" contained both Ag and In above their background values. The Ag/In ratios ranged from 0.2:1 to 4.5:1. There were 16 "slabs" which contained Ag and no In and 2 "slabs" contained In and no Ag. Those without Ag were distributed in time throughout the

Table 5. Analysis of snow collected March 24, 1993.

Time (UT) Start	End	C_{Ag} (e-12)	C_{In} (e-12)	w_t g	N_{xt} (e+05)
0745	0800	2.9	0.0	781	0.68
0800	0810	2.3	0.0	334	0.62
0810	0820	2.5	1.9	544	0.72
0820	0830	2.7	0.0	709	0.67
0830	0840	2.8	0.0	656	1.04
0840	0850	3.5	0.0	694	0.87
0850	0900	2.6	3.0	615	0.23
0900	0910	2.8	2.1	920	0.23
0910	0920	3.5	0.0	735	0.23
0920	0930	3.7	0.0	1078	0.23
0930	0940	5.5	2.5	1148	0.23
0940	0950	3.9	0.0	839	0.23
0950	1000	5.2	2.9	618	0.14
1000	1010	1.9	3.0	404	0.09
1010	1020	2.0	2.6	520	0.16
1020	1030	3.3	0.0	850	0.16
1030	1040	3.3	0.0	1133	0.45
1040	1050	3.7	0.0	1466	0.43
1050	1100	2.2	0.0	888	0.56
1100	1110	2.6	0.0	1193	0.56
1110	1120	2.0	0.0	1235	0.74
1120	1130	2.6	4.0	848	0.58
1130	1140	2.3	2.2	780	1.09
1140	1150	3.5	2.1	506	0.70
1150	1200	2.5	0.0	649	0.67
1200	1210	2.9	1.7	400	0.67
1210	1225	1.7	0.0	583	0.67
1225	1240	3.0	2.0	36	0.67

sampling period, while those with indium tended to be in clusters over 20-30 minute time intervals, a common feature of detection. In this same time period, 26 measurements were made of ice crystal sizes. The numbers of ice crystals per gram of water (N_{xt}) ranged from 0.1 to 1.1 x 10^5 as seen in Fig. 4. For the 10 "slabs" which contained both Ag and In during the 5 h period of observations from 1545 UT to 2040 UT,3/24/93, the weighted mean value of ice crystal enhancement was (E_{min}) weighted = 91%, as shown in Table 6.

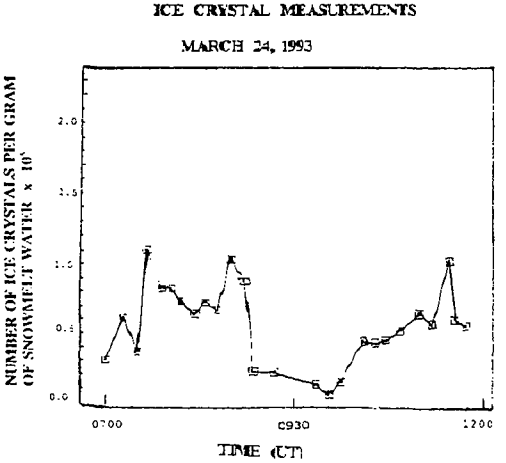

Figure 4. The number of ice crystals per gram of snowmelt (N_{xt}) as a function of time, March 24, 1993 observed at site A in Fig. 2.

Table 6. The March 24, 1993 case for those snow samples for which E_{min} can be calculated.

Time (UT)	Ag:In ratio	N_{xt} (e+05)	f	E_{min} (%)	w_t(g)
0810-0820	1.25	0.72	1.33	3	544
0850-0900	0.40	0.23	0.80	-5	615
0900-0910	1.33	0.23	1.06	18	920
0930-0940	3.50	0.23	1.00	1773	1148
0950-1000	2.29	0.14	0.99	-445	618
1120-1130	0.24	0.58	0.88	-8	848
1130-1140	0.43	1.09	0.00	-1	780
1140-1150	2.50	0.70	1.07	14	506
1200-1210	4.50	0.67	1.10	10	400
1225-1240	2.00	0.67	1.11	9	36

Expected scavenging alone Ag:In ratio = 0.52

Weighted E_{min} (observed) = 91%
Range of E_{min} (assuming 20% error in N_{xt}) = 62% to 135%

Mean crystal mass for all snow "slabs" collected with silver not above the background concentration,
= 1.9 e-05 g/crystal

Mean crystal mass for all snow "slabs" collected with silver above the background concentration,
= 2.0 e-05 g/crystal

There were only two snow "slabs" with silver less than background which could be used as the unseeded comparison set for calculating $m_{xt,ns}$. We then proceeded in the same manner as described for the February 17-18 case, and estimated the minimum precipitation enhancement (P_{min}) to be 7% of the natural precipitation at the site during the 5 h sampling period. Again the combined errors in N_{xt}, C_{Ag}, C_{In} and w_t, suggest that the P_{min} value of +7% has an error of ±60% of that value. The mean crystal mass in the snow "slabs" having silver above the background was 2.0 x 10^{-5}g, while that in the "slabs" not having silver above the background was 1.9 x 10^{-5}g. The difference is not significant.

7. DISCUSSION OF CHARACTERISTICS OF MEASUREMENT SYSTEMS USED

To employ this physical and chemical assessment method, it is necessary that the data be collected

under specific experimental conditions. Firstly, the method of assessment requires a high probability that particle sizes over the entire size spectra of the AgI and In_2O_3 aerosols be represented in the snowfall chemistry. Hence, snow sampling periods on the order of 1/2 h or more should be allowed for the integrated effects of such particle removal to be observed in the precipitation. Secondly, because targeting of the aerosols to the sampling sites in the mountainous terrain where the studies are conducted can be difficult, sufficient aerosol generator sites should be used to increase the probability that some portions of the aerosols will be transported to the chosen "receptor" sites. Thirdly, it is important to know the natural background concentrations of silver and indium in the region where the experiments are being conducted. Finally, in the estimation of N_{xt}, it is essential that the shapes of the water drops formed by melting the sampled ice crystals are well recorded, for example as hemispheres or prolate spheroids. This is to ensure that the mean ice crystal mass, m_{xt}, is not overestimated. Experience so far in this work has shown that the melted water drops from the ice crystals resemble prolate spheroids being about 14% less than the corresponding hemispherical volumes. It is important to point out that the scientific techniques being described in this paper are not intended for general use in operational silver iodide seeding projects. However, it would appear to be suitable for use in small or large scale snowpack augmentation experiments. The assessment method aims to provide a procedure for assessing the results of such experiments using physical and chemical measurements of the precipitation, either as an alternative to, or in support of, the traditionally used statistics of precipitation. The new assessment methodology described here is undergoing early development and some of the difficult measurement methods may contain significant errors at this time. It is hoped that new measurement systems combined with computerized data handling can be developed to overcome these shortcomings.

The requirements of the new method include the deployment of a substantial number of aerosol generator pairs (silver iodide and indium sesquioxide) which can impact larger proportions of a target area than most programs have used in the past. Evidence presented by Warburton et al. (1995b) has shown serious deficiencies in areal coverage of targets with inadequate numbers of generators. The new method also requires sampling of the snowpack using a vertical profiling technique, at a significant number of sites throughout the target area (e.g., one every 50 km^2) and immediately following the storm periods. The profiling method subdivides the snow core into 2 cm depth increments ("slabs") which generally represent 1/2 h to more than 3 h of snowfall depending on precipitation rate. These profiles also provide the basic precipitation data which the project needs for overall evaluation of the seeding experiment. Also, it is essential to collect real time ice crystal observations probably with a combination of particle (PMS) probes and microphotography, at selected sites throughout the experimental storm periods, to obtain truly representative N_{xt} values for use in the assessment process.

8. CONCLUSIONS

This paper describes a new physical and chemical method for estimating whether cloud seeding with silver iodide aerosols has changed ice crystal deposition in a target area, and whether such changes have altered the amounts of ice-phase precipitation reaching the ground. Very preliminary results from two seeded storm periods in 1993 are presented.

Based on a "source-receptor" concept, AgI and In_2O_3 aerosols with very similar particle size distributions are released from separate generator "sources" simultaneously. Measurements are then made of the concentrations of silver and indium in the snowfall at the "receptor" sites and of the numbers of ice crystals per gram of water in the precipitation at those sites. These measurements are then used to calculate the minimum enhanced numbers of ice crystals (E_{min}) produced at each "receptor" site. These values can then be used to estimate if there have been changes in precipitated water (P) at each site.

These new techniques have been used during two seeded winter snow storms in the Sierra Nevada in early 1993. Precipitating ice crystals and bulk snow samples were collected in real time at a high altitude sampling site in the Carson Range, east of the Sierra Nevada crestline, during each of these snowfall events for periods of several hours. Based on the chemical and physical measurements which were made, it was determined that ice crystal enhancements E_{min} of 16% and 91% had occurred at that site during the seeding periods on February 17-18 and March 24, 1993 respectively.

Minimum amounts of natural precipitation enhancement, P_{min}, were then estimated for these two seeded storm periods at that sampling site. The results suggest that the value of P_{min} was +3% ± 60% of that value, over the 8h period for the February 17-18 case. For the March 24 case the results suggest that P_{min} had a value of +7% ± 60% of that value, during the 5 h seeded storm period.

9. ACKNOWLEDGMENTS

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