A Field Technique for Detecting Silver Iodide in Snow

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ABSTRACT

A field technique was developed to detect silver iodide seeding agent in snow samples. The technique consists of collecting snow during a snow storm, forming liquid drops by melting pellets made from the snow, and refreezing the drops. A histogram of frequency of drop freezing plotted against temperature indicates the presence or absence of silver iodide in snow.

1. Introduction

During periods of good weather it is possible to track the dissemination of silver iodide with an instrumented aircraft (Langer *et al.*, 1967). Results of such studies indicate that the seeding agent may be poorly distributed by atmospheric turbulence, but these results are representative only of non-precipitating weather conditions. During storms, silver iodide plumes can best be tracked by analyzing precipitation fallout. For this reason, field methods for on-the-spot detection of the seeding agent in collected snow have been developed.

There are three mechanisms by which silver iodide particles become associated with snow crystals during a seeding operation conducted from the ground.

1) Silver iodide particles may originate ice crystals; in this case, it is generally accepted that each particle will be located in the center of a snowflake.

2) Silver iodide particles may be scavenged during ice crystal growth (phoretic and electrostatic forces) and by snowflakes during their fall through the atmosphere.

3) Silver iodide particles may be deposited on the surface of fallen snow. It is possible to design a series of experiments to differentiate these mechanisms and to determine their contribution to snowfall resulting from the seeding operation. However, this paper describes a field technique for detecting silver iodide in snow regardless of the mechanism by which the silver iodide particles became attached to the snowflakes.

A number of laboratory techniques have been developed to detect silver iodide in precipitation, but true *field* techniques for doing this have not been available. In the Park Range Program[2] neutron activation analysis has been employed. Though this is an extremely accurate and sensitive technique, the time required for analysis is excessive; in the experience gained on the Park Range Program, results have often not been available for months after samples were taken. Its high cost also leaves much to be desired. Innovations have recently been introduced[3] which improve the cost and time factors involved in neutron activation analysis, but measurements still cannot be made in the field. Also, while this method estimates the amount of silver present in the snow sample, it does not give information concerning the source of the silver nor whether it was present as silver iodide.

A technique has been developed (Isono, 1961) for detecting silver in water samples by means of its nucleating property. Dissolved silver iodide is precipitated as a fine colloid by mixing the water sample with a dilute potassium iodide solution, and the sample is then sprayed into a cold chamber where it produces ice crystals according to its silver iodide content. This technique presents many problems, but has been greatly refined by Warburton (1963, 1965) and Warburton and Maher (1965) to yield a very sensitive tool. However, use of this method is time consuming and requires an amount of care that is not feasible in the field.

Koenig (1960) rejected mass techniques and developed a chemical method which allowed determination of the actual position of the silver iodide within the ice crystal.

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[2] Bureau of Reclamation Contract No. 14-06-D-5640.

[3] Warburton, J. A., 1967: The detection of silver in hail, rain and snow by neutron activation analysis. Paper presented at the American Meteorological Society meeting, 20–22 March, Ann Arbor, Mich.

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This would, of course, be the most desirable approach, but difficulties encountered in field application apparently could not be overcome, and the technique was never utilized.

Some variant of the refreezing approach of Isono and Warburton seemed to offer the most promise of a successful field tool. An investigation was undertaken to determine the usability of the refreezing temperature of melted snow pellets as an index of the silver iodide content of snow.

The freezing of drops of rain, melted hail and melted snow "was investigated by Vali and Stansbury (1965). They indicated that, for heterogeneous freezing, the rate of cooling is of secondary importance. They filtered melted snow samples before refreezing. In the present study this was not done as we did not wish to remove any silver iodide with larger particles present in snow.

Emphasis must be placed on the fact that this method is concerned with snow and not rain. Silver iodide is slightly soluble (2.8×10^{-9} gm cm^{-3} at 15C) in water and in the quantities we found in precipitation ($\approx10^{-11}$ gm cm^{-3} background) would be completely dissolved if the sample remained in a liquid state for some period of time. The rate of dissolution of silver iodide in water is very slow, so that by melting the snow sample immediately before analysis, one can be reasonably certain that the silver iodide particles are not dissolved. Fresh silver iodide particles released from a generator are hydrophobic, and it takes a period of time to change the surface to hydrophilic and to transfer the particles into solution.

Measurement of the solubility of silver iodide and its dependence on temperature are not available in the literature and were not determined for this study. However, we have observed under a microscope the behavior of relatively large ($\approx8\,\mu$ diameter) particles of powdered silver iodide when placed on and injected into water droplets during repeated cycles of freezing and melting over several hours. At no time was there any sign of the particles dissolving or even becoming wetted; particles placed on the surface remained on the surface without exception and those placed within the droplet remained on the bottom of the droplet, with, in all cases, no change in appearance. Of course, it is not possible to state firmly that the same behavior will hold for the much smaller particles produced by the various burners in current use. The effect of additives and other combustion products cannot also be overlooked.

As for the dependence of the solubility of silver iodide on temperature, we have observed, during our attempts to adapt the Isono technique for use with the NCAR acoustical counter, that the number of ice crystals which result from a sample depends on the sample's temperature at the moment of spraying. A sample yielding a count rate of 1000 ice crystals min^{-1} (at sample flow of 1 cm^3 min^{-1}) when held at 0C before spraying (chamber temperature, −15C) would yield no counts at all if warmed to 35C. The samples in question were prepared from solution by Nagels' (1958) method and contained 0.01 gm liter^{-1} potassium iodide as specified by Isono (1961). Since the samples were prepared from solution, considerations such as surface wettability were not applicable.

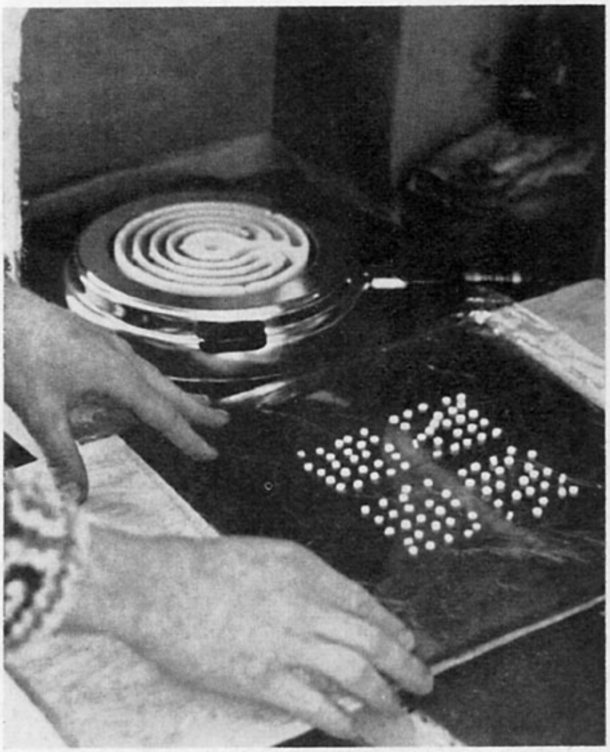

Fig. 1. Pellet-forming equipment.

In the method described here the time of contact between the silver iodide particles and liquid water is kept to the absolute minimum required for melting the snow and placing it in the cold chamber for refreezing. The quantities of silver iodide dealt with in the previous experience were 10^{-11} gm ml^{-1} in unseeded snow and 10^{-11} to 5×10^{-9} gm ml^{-1} in seeded snow as determined by neutron activation analysis.

2. Experimental technique

Drops were formed by pressing snow into a form consisting of a sheet of lucite, 3 mm thick, through which 100 conical holes 5 to 8 mm in diameter have been drilled, and which has been coated with teflon (Fig. 1). A sheet of polarizing plastic film[4] was placed beneath the form so that the snow pellets ware arrayed on the polarizer when the form was lifted away. To avoid the problem of cleaning the polarizing film after each sample, it was covered with a sheet of thin plastic wrap, rubbed on to give good thermal contact. The

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[4] Bausch & Lomb, Inc., Rochester, N. Y.

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same wrap was used as a glove to manipulate the snow sample. To avoid carry-over from sample to sample, the lucite form was rubbed thoroughly each time with the new sample. The plate was also tested several times to see if some of the positions on the plate nucleate preferentially; it was found that freezing was random.

The pellets on the polarizing film were melted (with a hot plate and reflector, or with an infrared flood lamp) until they formed drops, which were about 3 mm in diameter. A maximum time of 2 min was required for melting the drops. The hot plate was tested with the NCAR ice nucleus counter (Langer *et al.*, 1967) and it was found that it did not produce any ice nuclei down to −21C. A thermocouple was taped to the polarizing sheet which was set in a cold box at −27C. An electronic flash unit was situated beneath the sample and another polarizer, at right angles to the first, placed over an opening in the top of the cold box. As the sheet cooled down, it was photographed at pre-chosen temperatures. Trials have shown that, during cooling, the temperature difference between the center and the edge of the sheet did not exceed 0.3C. In each photograph the drops that have frozen show up in high contrast to those remaining unfrozen (Fig. 2).

We have found that the heterogeneous freezing of drops by the procedures described here is dependent on rate of cooling. To eliminate instrumentation for an elaborate constant-cooling rate system in the field, efforts were made to set up reproducible cooling rate conditions. The temperature-time traces recorded in the experiments were uniform. The average cooling rate was 3C min^{-1}; it was faster just after the polarizing plate was transferred into the freezer and slower as the temperature approached −15C. In the neighborhood of −15C there was a slight tendency for the cooling rate to be controlled somewhat by the number of drops freezing. The polarizing sheet is of relatively low mass, and the heat released by the freezing of the drops has a relatively strong effect on the cooling of the sheet. This is unfortunate, but must be lived with if the entire setup is not to become very complex. In using this technique for detecting silver iodide in snow, one should place much more value on the results at warmer temperatures, for example, −10C or −12.5C.

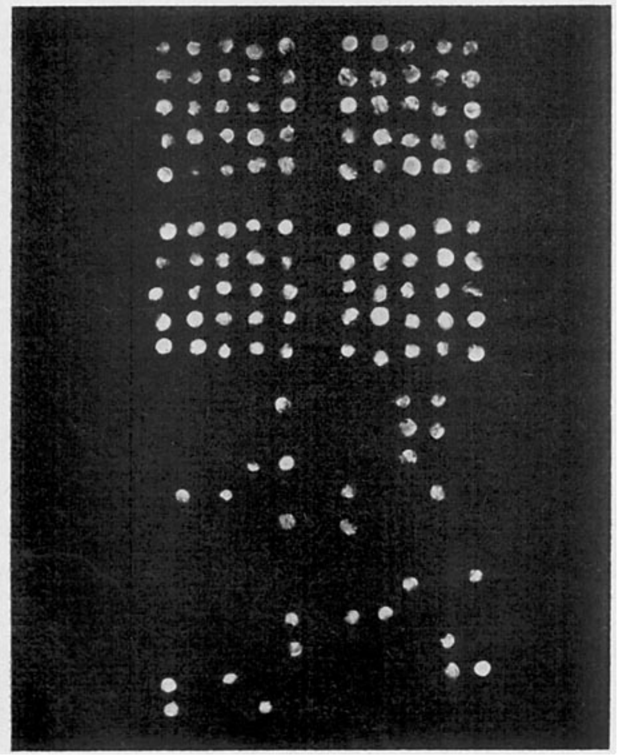

FIG. 2. A sample at various stages of freezing.

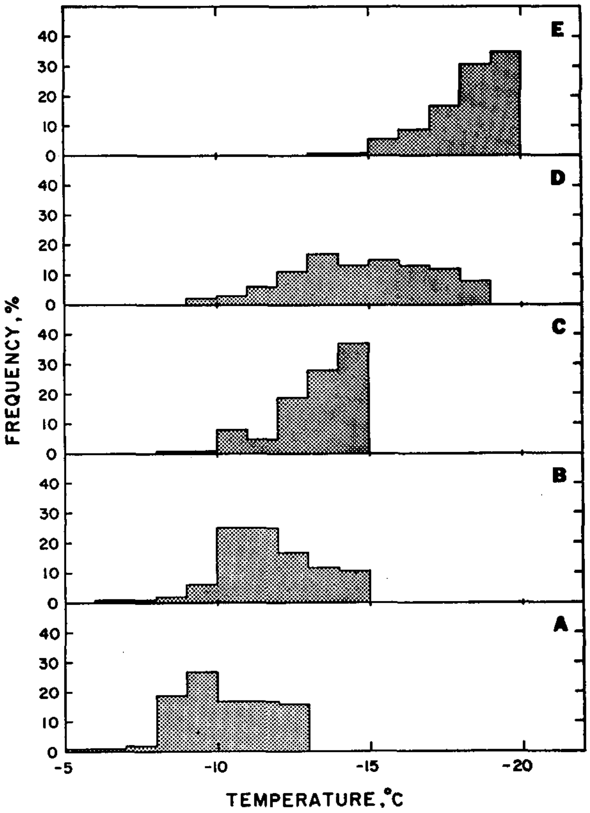

FIG. 3. Freezing spectra of five snow samples: a., 10 m from silver iodide generator; b., 200 m from silver iodide generator; c., 3 km from silver iodide generator; d., 15 km from silver iodide generator; e., uncontaminated snow from unseeded area.

3. Experimental results

Field trials of the experimental equipment were carried out at the Steamboat Springs, Colorado, field facility of the Park Range Atmospheric Resources Program, and in the area of Winter Park, Colo. The results of refreezing melted snow samples from the storm of 31 December 1966 are shown in Fig. 3a to 3d. The storm was seeded with silver iodide and samples

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were collected at different distances from the burner. The histogram in Fig. 3a represents the percentage of drops frozen versus temperature for a sample of snow collected 20 m from the silver iodide burner. The drops had started to freeze at −5C and were completely frozen at −13C. Fig. 3d shows a sample collected 15 km from the burner and not directly downwind from it; it represents clean snow from the seeded area. Figs. 3b and 3c represent intermediate cases, one being 200 m downwind from the burner and the other 3 km away. Fig. 3e represents snow collected during the 30–31 January 1967 storm from the unseeded Winter Park area at an altitude of 3250 m. It is given here for comparison. In addition to differences in the freezing-temperature spectrum, it was noticed that refreezing of seeded snow produced a large number of drops with spicules (up to 25%) as shown in Fig. 4.

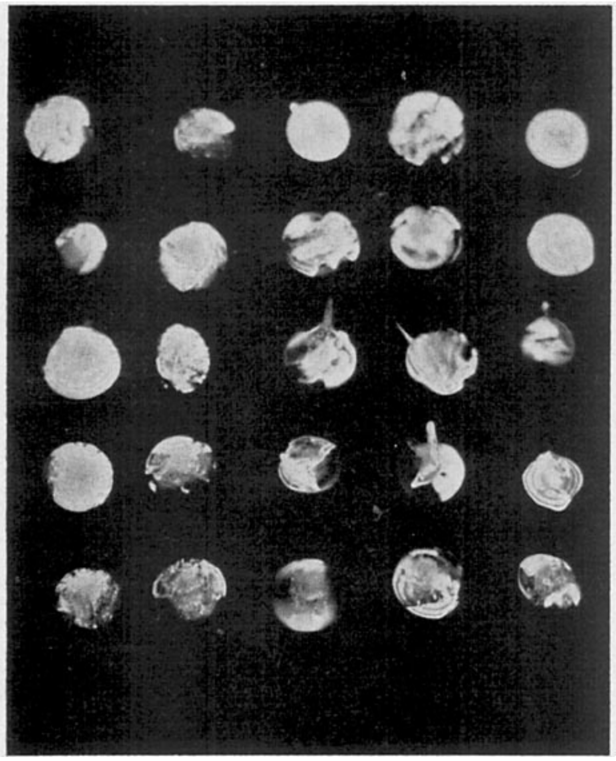

Fig. 4. Drops with spicules.

To represent serial measurements, the percentage of drops frozen at certain temperatures has been plotted against time in Figs. 5 and 6. Fig. 5 is the record of a series taken at the Rabbit Ears Pass highway maintenance camp (2900 m), located about 21 km ESE of the generator site. During this period of silver iodide generation, the wind at 3350 m was from the NW at 46 km hr^{-1}, and the generator, located at Emerald Mountain (2600 m), was being operated pulse fashion, being ignited on the hour and turned off on the half hour.

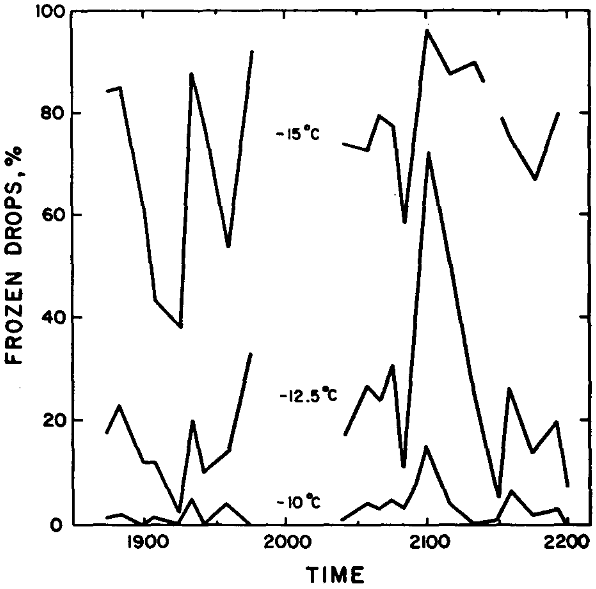

Fig. 5. Per cent of drops frozen for three different temperatures as a function of time (Rabbit Ears Pass, snowstorm of 31 December 1966).

Fig. 6 shows a series taken at Valley View Lodge, on U. S. Highway 40 at 2300 m, 13 km to the SE of the generator. The generator at Emerald Mountain was being operated pulse-wise, on the same schedule as before. The wind at 3350 m was from the NNW at 18 km hr^{-1} during the series; a trough had just passed through at 2000 MST.

Results shown in Figs. 5 and 6 indicate that it would be possible, with a sampling network, to determine the

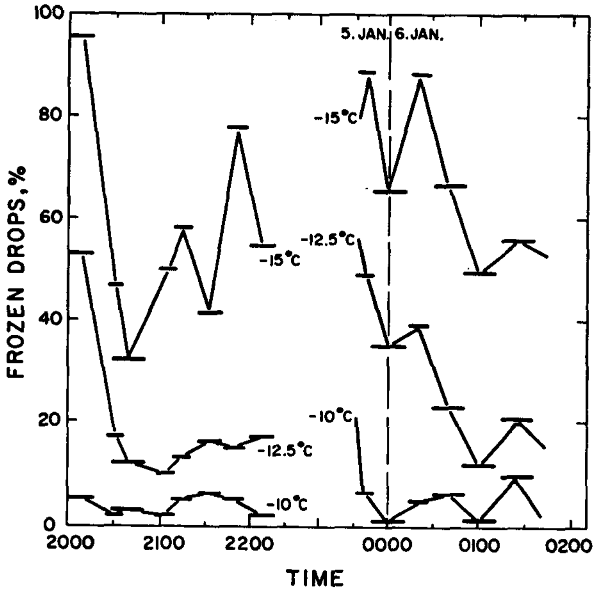

Fig. 6. Per cent of drops frozen for the three different temperatures as a function of time (Valley View Lodge, snowstorm of 5–6 January 1967). Horizontal bars indicate interval over which sample was collected.

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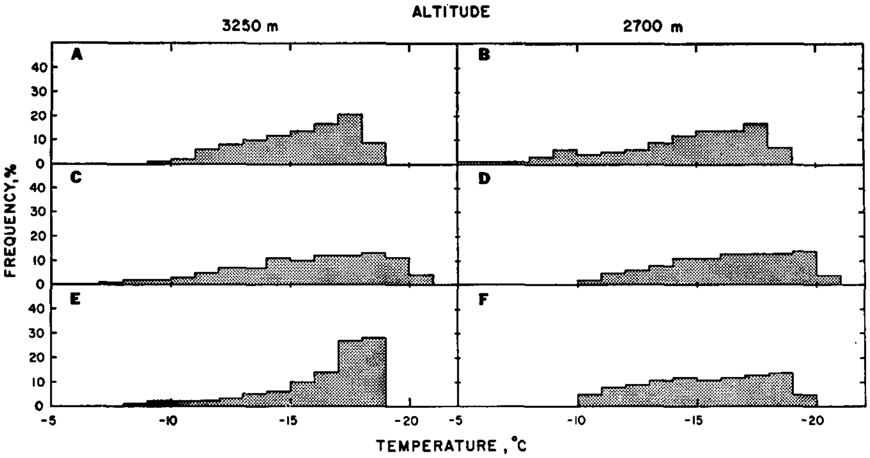

FIG. 7. Freezing spectra of snow collected at two different altitudes as a function of time (Winter Park, Colorado). a, b., 0800–1200; c, d., 1200–1500; e, f., 1500–2000.

coverage with snow associated with silver iodide. Snow collected during a storm and analyzed later requires storage of snow samples. Three experiments were performed during the 31 January 1967 storm to determine conditions under which snow should be sampled and stored. In the first one, snow was collected at an altitude of 2700 m, and was stored at a temperature of −20C. Results were reproducible indicating that storage at low temperatures of sealed samples appears not to affect the temperature spectrum. Prolonged storage of snow samples was not tested. In the second experiment (Fig. 7), snow was sampled in an uncontaminated area at 3250 m altitude and at 2700 m altitude in Winter Park located near U. S. 40. Three snow samples were collected: from 0800 to 1200 MST (Fig. 7a, b), from 1200 to 1500 (Fig. 7c, d) and from 1500 to 2000 (Fig. 7e, f). If the histogram skewed to the right represents uncontaminated snow, snows collected at the lower latitude and at 1200–1500 at the higher altitude contained more solid particles enhancing freezing at higher temperatures than snow collected in the morning and in the evening.

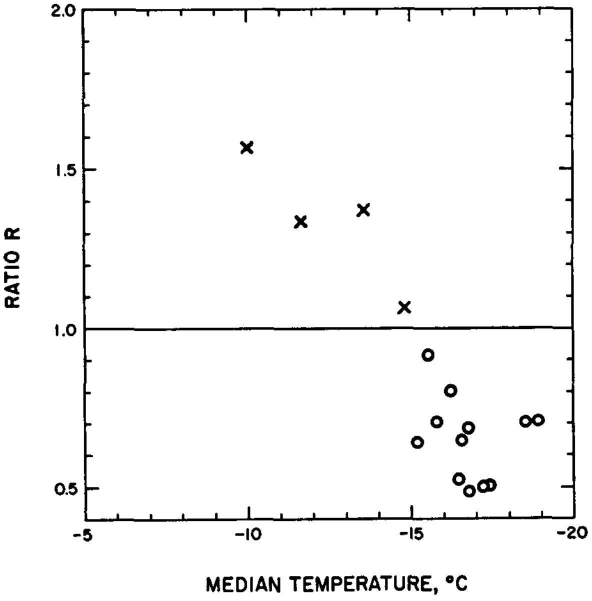

FIG. 8. Ratio R as a function of median temperature of frozen drops.

The third experiment consisted of analyzing snow from different layers, after exposing half of each sample to full sunshine for one day. Differences in the temperature spectrum for different samples of fresh snow exposed to heat from the sun were very erratic. It can be concluded that if it is at all possible, analysis of snow should be performed at the field site.

4. Characteristics of the freezing temperature histograms

The distributions of freezing temperatures for the drops are clearly not normal, and not all alike. Some generalizations can be made, however. The difference between the temperatures of the 25th and 75th percentiles of the cumulative freezing temperature distribution can be used as an index of freezing temperature variation or spread. This spread tends to be small for samples with a high or low median freezing temperature. The spread is larger for intermediate median freezing temperatures, which may indicate interaction of two independent freezing nucleus populations.

An index of the skewness of the distribution can be roughly estimated by the ratio

$$R=\frac{T_{75\%}-T_{50\%}}{T_{50\%}-T_{25\%}},$$

where $T_{a\%}$ is the temperature of the ath percentile of the freezing temperature distribution. This appears to have a strong dependence on the median freezing temperature as shown in Fig. 8. It would appear that R

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TABLE 1. R ratios for different snows.

Snow origin	Location	R	Figure*
Seeded area, Steamboat Springs, Colo., 31 December 1966 storm	20 m from AgI generator	1.56	3a
	200 m from AgI generator	1.33	1b
	3 km from AgI generator	1.37	3c
	15 km from AgI generator	1.06	3d
Unseeded area, Winter Park, Colo., 30–31 January 1967 storm	Altitude 3500 m, clean area	0.70	3e
	Altitude 3250 m, 0800–1200, clean area	0.70	7a
	Altitude 2700 m, 0800–1200, Winter Park	0.64	7b
	Altitude 3250 m, 1200–1500, clean area	0.80	7c
	Altitude 2700 m, 1200–1500, Winter Park	0.82	7d
	Altitude 3250 m, 1500–2000, clean area	0.50	7e
	Altitude 2700 m, 1500–2000, Winter Park	0.91	7f
	Altitude 2700 m, average for storm, Winter Park	0.64	
	Altitude 2700 m, average for storm, Winter Park, exposed to sun	0.70	
Unseeded area, Winter Park, Colo., December 1966 and January 1967 storm	Altitude 2700 m, snow from January storms	0.50	
	Altitude 2700 m, snow from January storms, exposed to sun	0.68	
	Altitude 2700 m, snow from December storms	0.48	
	Altitude 2700 m, snow from December storms, exposed to sun	0.52	

* See corresponding figures in this paper.

is at least as good a parameter for discriminating between samples as the median freezing temperature. It may in the long run be better, because it contains more information about the distribution than the median alone. The R ratios for different snows are given in Table 1.

5. Conclusions

Silver iodide particles present in snow remain in suspension long enough to form active freezing nuclei in the refreezing of freshly melted snow. This makes a refreezing procedure feasible as a field technique for detecting silver iodide in snow samples. The distribution of per cent of drops frozen vs. temperature, and parameters which can be derived from it, vary in a way which can be related to the presence of silver iodide. It is not possible to distinguish in this way the mechanism by which the silver iodide particles become attached to the snow crystals. It is not suggested that this approach to detecting silver iodide can replace the much more exact and sensitive techniques, such as neutron activation analysis, which have been discussed here. The technique developed is, however, one which can be applied directly in the field during seeding operations.

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