

Freezing Nucleus Content of Hail and Rain in Alberta

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ABSTRACT

The concentrations of freezing nuclei in precipitation from different storms have been measured and the variations of nucleus content with space, time, precipitation type and intensity have been examined. It was found that nucleus concentrations are higher in showery rain and in hail than in continuous-type rain. Peaks have been detected in several nucleus spectra and there is some recurrence of such peaks throughout the storms. This type of information may help to improve descriptions of precipitation processes which involve the ice phase.

1. Introduction

In view of the need to establish the nature of the ice-forming processes in clouds and in order to find the sources and to describe the nature of atmospheric ice nuclei, improved ice nucleus measurements continue to be widely sought. The principal techniques for the detection of atmospheric ice nuclei are cloud-chamber activation, filter capture and development, and precipitation analysis. There exist several variants of each of these methods and it is now recognized that even beyond the technical differences, the different methods may be activating and detecting nuclei through fundamentally different (and not always unambiguously defined) processes. Comparisons of techniques are made very difficult by this fact. The observations presented in this paper relate to freezing nucleation by particles suspended in water (rain or melted hail). From the results, estimates can be derived for the contribution made by freezing nuclei to the overall cloud glaciation (Vali, 1968b), but other processes of nucleation will clearly have to be included before a complete picture is obtained.

Drop freezing experiments for the analysis of water have been widely used (Rau, 1944; Vonnegut, 1948; Dorsch and Hacker, 1950; Bigg, 1953; Barklie and Gokhale, 1959; and many others). The results of these experiments were first interpreted on the basis that the freezing of the drops is a stochastic process (Bigg, 1953; Marshall, 1961), but later research on the time-dependence of the freezing showed that the stochastic element is only a small superposition on the basically "singular" character of the process (Vali and Stansbury, 1966). In the singular interpretation each heterogeneous nucleus is taken to become active at its singular or characteristic temperature and the freezing tempera-

ture of each drop in an experiment is determined by its specific nucleus content.

A quantitative description of the drop freezing experiments in terms of differential and cumulative nucleus spectra was given by Vali (1967, 1968b). The differential concentration $k(\theta)$ is derived from

$$k(\theta) = -\frac{1}{VN(\theta)} \frac{dN}{d\theta}, \quad (1)$$

where $N(\theta)$ is the number of drops unfrozen at temperature θ ($^{\circ}\text{C}$) and V the volume of the drops. The differential nucleus spectra represent the specific activities at particular temperatures normalized to 1C intervals. The cumulative concentrations represent the nuclei active at all temperatures above the particular temperature and is obtained by integrating (1).

Analyses of precipitation by the drop freezing technique (Barklie, 1960; Vali and Stansbury, 1966; Vali, 1968a, b) have led to the interesting finding that, in general, the concentrations of nuclei in hail and in showery rain are higher than in steady rain or snow. Extensive sampling of summer storms in Alberta was undertaken in 1968, with the aim of obtaining greater detail on individual storms, so that freezing nucleus content could be related to rainfall rate and other storm parameters. Information about the storms was derived from radar, ground and aircraft observations.

2. Method

a. Sample collection

Sample collections were carried out in the following manner: Polyethelene bags of 25 cm diameter were splayed over and secured to wire hoops and installed on top of poles about 2.5 m over the ground. The bags had welded constrictions, designed to allow rain to

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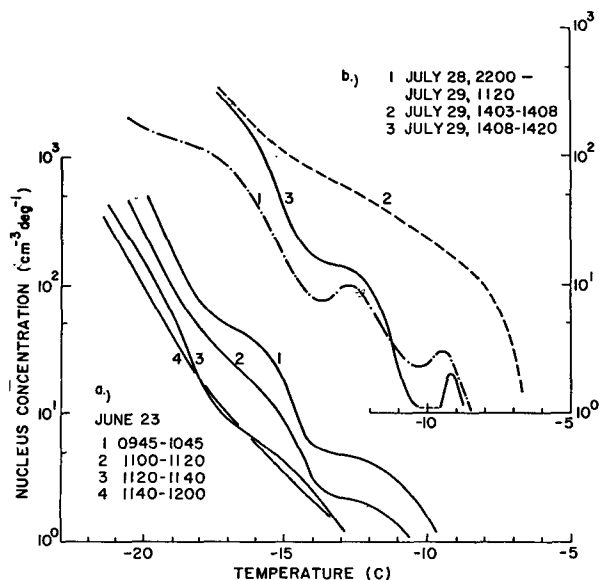


FIG. 1. Differential nucleus spectra for sequences of rain samples on two days.

collect in the bottom section and to retain hail, if any, in the upper section. Some mixing of the hail and rain was certain to occur (soaking of rain into the hailstones and melting of hail into the rain); differences are therefore attenuated. The exposure of collectors was held to a minimum; they were installed just after the onset of rain and taken down when a sufficiently large sample had been obtained or when there was a change in rain intensity. The bags were sealed immediately and frozen over dry ice. Collectors were always set up in pairs to provide a check on consistency, and to allow, in a few cases, other types of analyses to be carried out on duplicate samples.

Most collections were made by mobile units which were directed to the storm areas by radio from the radar control center and/or from the aircraft tracking the storm updrafts. Two units were used primarily for precipitation measurements and collections, three units made collections in addition to other duties, and several more units could be utilized for collections on special occasions.

b. Analysis

The nucleus contents of the samples were determined by drop-freezing experiments. From each sample a 2-cm³ aliquot has been used to produce 200 drops of 0.01 cm³ volume (0.27 cm equivalent spherical diameter). The number of frozen drops were recorded at 0.25C intervals giving 40–60 points for each spectrum. The resultant accuracy of the spectra is such that 90% of repetitions of the same experiment would result in concentrations varying by a factor of 1.5 over most of the spectra, with widening of the error band to approximately factors of 4 at the minimum and maximum con-

centrations. In the figures presented in this paper, peaks at warm temperatures (low concentrations) have occasionally been drawn in when there was an indication for consistency among samples, even though the data defining these peaks may have been within the error band.

3. Results

On 30 days, over a 10-week period, a total of 243 pairs of samples were collected. Little of this was hail, however, due partly to an unusually low incidence of hail in the Alberta Hail Studies project area.

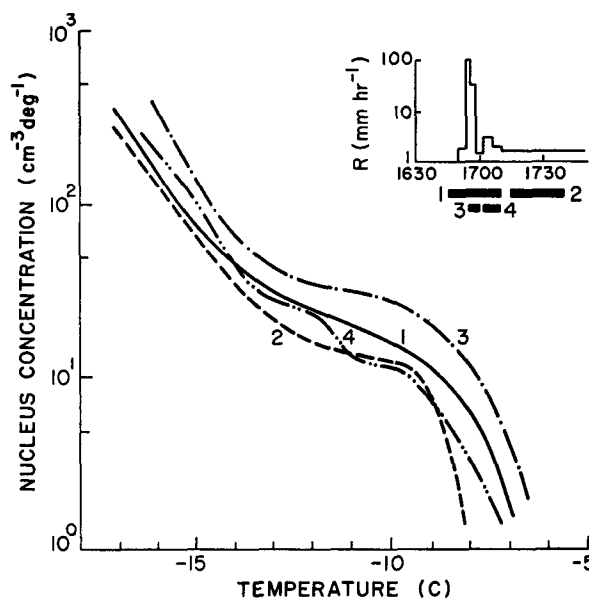


FIG. 2. A sequence of rain samples from a small Cb. Rainfall rates and the durations of samplings are shown by the inset.

The comparisons of sample pairs showed that although there are unaccountable differences in certain cases, the spectra for the pairs agree within a factor of 2 for 90% of the samples. The correlation coefficient for the mean freezing temperatures of drops from sample pairs is 0.87. The spectra given in this paper represent the mean concentrations for sample pairs; no use was made of samples where the differences were held (subjectively) to render an average spectrum meaningless. The only reason for large discrepancies could be contamination during handling.

In the following, results for a few occasions (storms) are presented to illustrate the major factors that are emerging from this work.

a. Variation with rain intensity

1) 23 June 1968 (Fig. 1a). Rain was continuous on this day from early morning, with occasional showers. The first sample was taken from a period of relatively heavy rain, with the rainfall rate varying between 3 and

7 mm hr⁻¹. The intensity of the rain began to diminish afterward, and for samples 2, 3 and 4 the average rates were 2.5, 1.5 and 0.8 mm hr⁻¹, respectively. In another hour or so the rain stopped.

There is an unfortunate ambiguity in the interpretation of these results, in that the sequence of samples with diminishing rainfall rate also forms a succession in time. The decrease in nucleus content could therefore be related to either cause, but the fact that sampling commenced ~5 hr after the onset of rain makes it likely that the variation is due mainly to changes in rainfall rate, and that the gradual cleansing of the air by rain washout contributed little to the observed results. Dingle and Gatz (1966) found the greatest decreases in contaminant concentrations during the initial periods of rain.

2) 29 July 1968 (Fig. 1b). After a major storm of the previous day a collector was left exposed overnight, and a sample of the rain that fell principally in the very early morning was obtained as sample 1. During the day shallow cumuli developed embedded in thin low cloud. At 1402 there was an approximately 2-min burst of small hail accompanied by intense rain. A collector was set up during this downpour, catching a little bit of

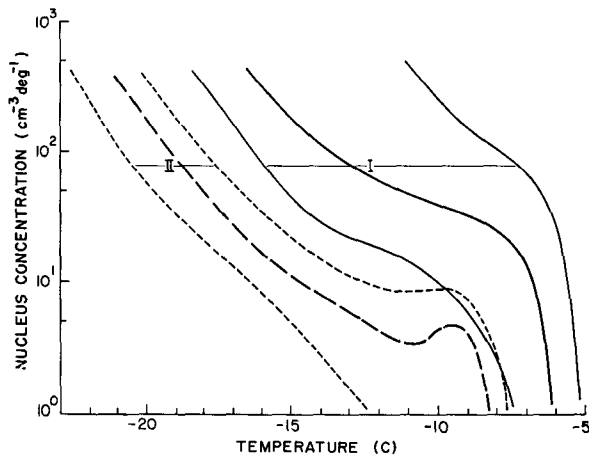


FIG. 3. Ranges of nucleus concentrations in 11 samples from a small area for the first period of high intensity rain (I) and for the following period of lighter rain (II) during the passage of a severe storm. The heavy lines indicate the central tendency in each range.

the hail which melted almost immediately, and collecting rain for 5 min. The average rainfall rate during this time was 45 mm hr⁻¹. This is sample 2. The next 12 min of rain, of average rate of 20 mm hr⁻¹, was collected in sample 3. At 1420 the rain stopped.

The most pronounced feature in the results shown in Fig. 1b is the high concentration of nuclei in the sample from the burst of most intense rain. Both the preceding (although by a long time interval) and the following rain had lower nucleus contents. In these latter two samples there is a very interesting recurrence of peaks at -9.5C and at -13C. (The long collection time for

sample 1 does not seem to have led to unreasonable results, which gives some reassurance on this point.)

3) 3 July 1968 (Fig. 2). This day there was intense convection, with echo tops reaching 12-14 km, but no major storm developed and both hail and rain were minor and sporadic. Samples were obtained from a small isolated Cb, perhaps 5 km in diameter. A distinguishing feature of the cloud was the rather intense electrical activity associated with it. Samples times and rainfall rates are shown by the inset in Fig. 2. The variations in concentration are relatively small among these samples, in spite of the great differences in rainfall rates, but the 5-min samples of very high intensity rain still shows somewhat higher nucleus concentration. All the samples had generally high nucleus contents.

4) 25 July 1968 (Fig. 3). One of the most severe storms of the season occurred on this day. A squall line of about 100 km length and 15 km width, with echo tops at 14 km, travelled over 130 km during its mature stage of 3 hr. It produced copious amounts of small hail during the first 2 hr and rain amounts ≥ 5 cm were recorded over an extensive area. Sampling of this storm was concentrated over a small area, 6.5 km along and 15 km across the line of propagation. From ten mobile and one fixed sampling station, a total of 34 sample pairs were obtained. Two high-speed recording raingages both indicated maximum rainfall rates well exceeding 100 mm hr⁻¹ and average rates of around 50 mm hr⁻¹ for the first one-half hour of rain. This was followed by ~2½ hr of light rain of less than 2 mm hr⁻¹. The last hail from the storm fell just before it reached the sampling area.

Only the most prominent aspect of the results for this day will be described here. Fig. 3 shows the ranges and the "centroids" of the spectra of the 11 first

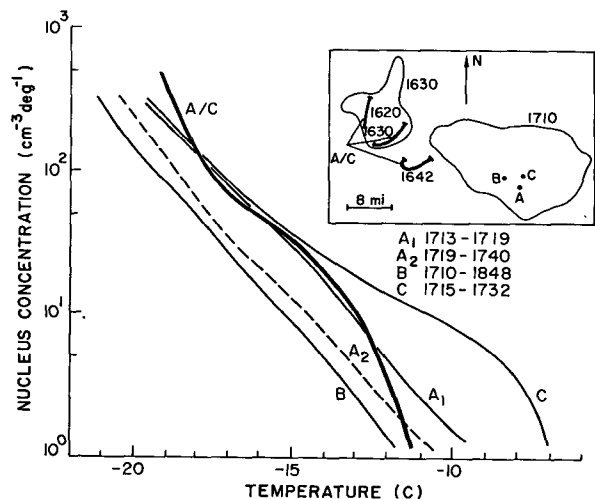


FIG. 4. Rain samples collected just below cloud base from an aircraft (heavy line) and for samples collected on the ground. In the inset, thin irregular lines are radar echo contours at the indicated times. Heavy line segments are the aircraft flight paths during sample collections.

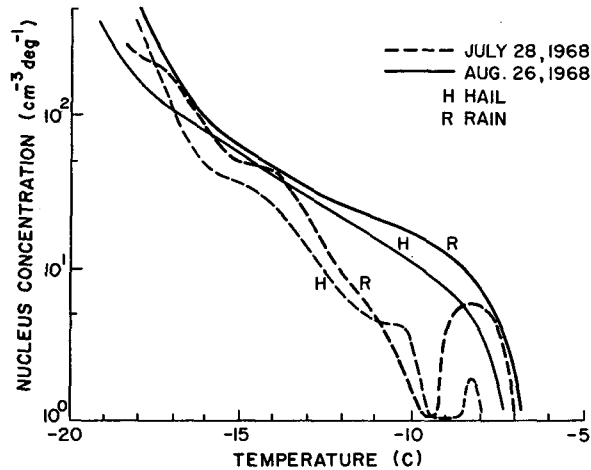


FIG. 5. Nucleus spectra for two pairs of samples in which rain and hail were caught separately from the same periods of precipitation.

samples (I) and of the 11 last samples (II) from the sequences obtained by the stations. The duration of collections varied, so that although all the first samples are of high-intensity rain, some of the last one overlap somewhat into this high-intensity period. Still, the difference is clear: very high concentrations of nuclei, active at temperature near -5°C , in the intense rain, and low concentrations in the light rain.

b. Variation with altitude

1) 1 August 1968 (Fig. 4). Samples of precipitation from a vigorous cumulus were collected by an aircraft about 300 m below cloud base and also by collectors on the ground. Cloud base was at 2.5 km, the temperature at that level being 12°C . The aircraft made three passes through moderate rain and encountered sporadic hail on one pass. At that time the cloud was beginning to dissipate (the echo heights reached their maximum 30 min earlier). Samples on the ground were obtained 40 min later, near the end of the storm's life. There were no reports of hail on the ground anywhere in the area covered by the storm.

The airborne collections were made with small (5 cm diameter) plastic bags which were held on a cylinder attached to a rod. This assembly was held out through the window of the aircraft. The collector was about 15 cm from the fuselage in undisturbed air. The duration of each collection was about 3 min and from 6–9 gm of water per sample were obtained. The nucleus spectra for these three samples were practically identical; all three spectra fall within a factor of 2 of the heavy line shown in Fig. 4.

Four samples of rain were collected at three locations. The nucleus spectra for these are also given in Fig. 4, the inset showing the times and relative locations of the sites of collection. The rate of rainfall for samples A_1 and C is estimated (from the amount of rain collected) to have averaged 8 and 15 mm hr^{-1} , respectively;

for A_2 the average rate was $\sim 2\text{ mm hr}^{-1}$. The duration of rain at B is unknown, and no reasonable estimate of rainfall rate can thus be made.

The agreement between the airborne and the ground-based collections can be said to be good, since the range of variation among samples from different days is much larger than the differences found here. That there are some differences present is not unexpected in view of the spatial and temporal separations of the collection. The somewhat higher nucleus content of sample C appears to be related to the higher rain intensity at that location.

These results are rather reassuring regarding the validity of precipitation collection at ground level for cloud physical interpretation, and show that collection of precipitation and perhaps of cloud from aircraft may provide further important information.

c. Hailstorms

On two occasions both hail and rain were collected in the upper and lower sections, respectively, of the same sampling bag, so that hail and rain from the same time and place and collected in identical fashion could be examined separately.

The rain-hail sample of 28 July 1968 was from a major hailstorm which produced up to 7.5-cm diameter hail. At the collection site the maximum hail size was about 0.6 cm, and the total amount of rain was ~ 15 mm. Since the collectors were left unattended for the 1-hr period of collection, the sequence of rain and hailfall or the amount of hail that fell are not known. At the time the sample bags were taken down there was unmelted hail in the upper section of the bag, but some of the hail had undoubtedly melted and the melt mixed with the rain. As shown in Fig. 5, the nucleus concentrations in the hail and in the rain are not very different. The rain has somewhat higher nucleus concentrations at all temperatures, the difference being greatest at the -8°C peak which shows up very clearly in both the rain and the hail. There is another minor peak in both spectra at -14°C .

The nucleus spectra for the other rain-hail pair, which was collected on 26 August 1968, show the same tendency: relatively little difference between rain and hail, with the rain having a slightly higher nucleus content. This pair of samples originated from a small storm which did, however, produce hail up to 4 cm diameter. The sample contained small hail only, and the details of the precipitation sequence are not known in this case either.

1) 15 July 1968 (Fig. 6). There was widespread and intensive convection on this day. No major hailstorms developed, but numerous small storms occurred over a 9-hr period and most storms produced some grape-to-walnut-size hail. The echo tops reached 10–12 km. Surface winds were generally very light, which meant reduced hazard of contamination of samples.

Samples from a very widespread area were collected. One sample was obtained from a moderate shower. Four essentially simultaneous samples along a 15-km line, 32 km from the first sample, were collected in an area where hail fell; one of these samples had some melted hail with the rain. Forty miles downwind another sample, again with some hail in it, was collected from the same storm. Fifty miles farther in the direction of storm travel, a late evening sample was collected from a dissipating cell. Hailstones of 3 cm diameter from a different storm were picked off the ground 110 km away. The nucleus spectra for two of these stones, after removal of the outer layers, are shown as curves 1A and 1B in Fig. 6. Curves 2 and 3 are for the two rain samples which contained some hail. Four spectra from the other showers are not shown; these all lie between curves 2 and 3. The spectrum for the rain from the dissipating cell is shown as curve 4.

These results indicate again that high nucleus content is associated with strong convection. All these samples have high concentrations of nuclei at temperatures around -8C . The nucleus contents of the hailstones are not unlike that of the rain, except for the much more rapid rise in concentration with decreasing temperatures. The similarity of all these spectra, in spite of the large spatial separation of sampling locations and the 8-hr time spread in collection times, is rather surprising. (The terrain in the area is quite uniform and the storms developed in the same air mass.)

2) 4 August 1968 (Fig. 7). This storm had small scattered hail associated with it, but at some points the amount of hail that fell was considerable. Hail from a deep layer was picked up at A and rain was collected at four other points along the storm track. Up to 25 mm of rain accumulation was measured in this area. At the fringe of this storm, 32 km to the ENE of the region shown on the inset, a sequence of rain samples was obtained. The average rate of rainfall for samples 1 and 2 was 20 mm hr^{-1} ; for samples 3 and 4 the rates were 12 and 6 mm hr^{-1} , respectively.

The spectra for these samples show that the highest nucleus content is found in the hail, that nuclei active at warmer temperatures are present in the rain samples from the hail region, and that the nucleus content decreases with rain intensity. A very striking feature of these results is the clear recurrence of peaks in the spectra at -9C and at -13C (perhaps also at -18C).

4. Summary

The findings regarding the freezing nucleus content of hail and rain in Alberta which emerge from the case studies presented in this paper are as follows:

- 1) There is storm-to-storm variability in the nucleus content of precipitation.
- 2) High nucleus concentrations are found in convective situations.

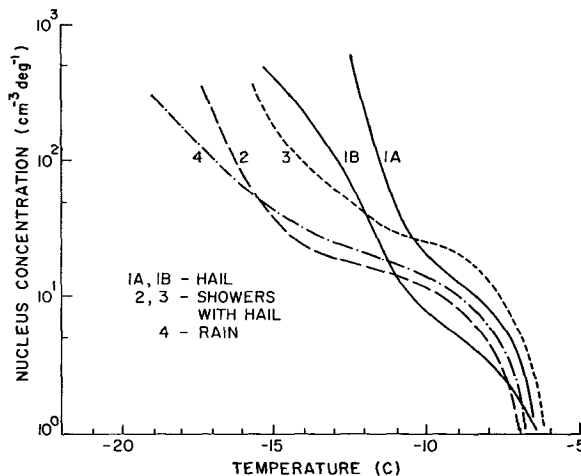


FIG. 6. Nucleus spectra for two large hailstones (1A, 1B), for two showers in which some hail fell (2, 3), and for rain from a dissipating storm (4). Four additional samples of rain from the same storm system had spectra which fell between curves 2 and 3.

- 3) There are often pronounced peaks in the nucleus spectra.
- 4) There are similarities in the nucleus spectra of rain samples from different points in a storm.
- 5) Nucleus content was found to be higher in rain of high intensity than in rain of low intensity from the same storm (one exception to this pattern is known to have occurred).
- 6) The concentrations of nuclei in simultaneous collections of hail and rain at the ground were found to

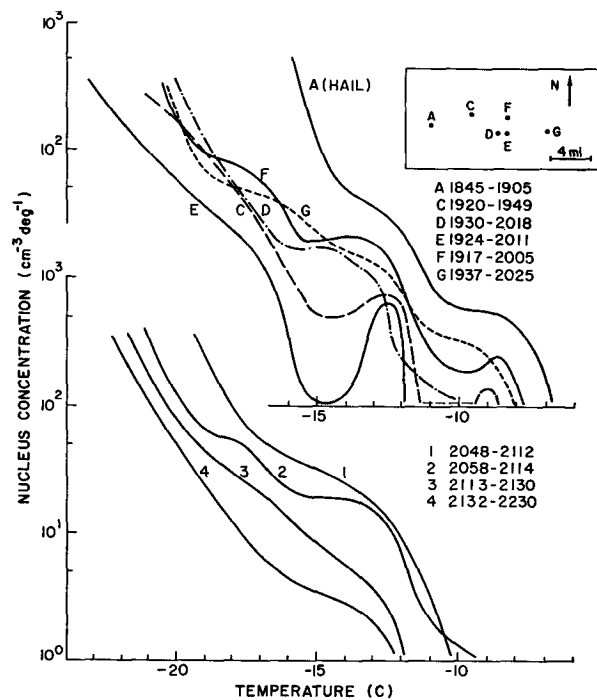


FIG. 7. Nucleus spectra for a sample of hail and five samples of rain from the same storm (upper), and for a sequence of rain samples 20 mi ENE of the region shown (lower).

be nearly the same. (Unavoidable mixing during collection could have masked small differences if they existed.)

The first four points are relevant to considerations of the sources of nuclei, and some general ideas follow from the findings. The high nucleus concentrations that are found in showery rain and hail are easiest to explain by postulating that the nucleus content of the air in which the precipitation developed was high, since differences in scavenging efficiencies between steady and showery types of rainfall would tend to result in lower concentrations for the showery rain (Georgii, 1965). High concentrations of nuclei in air for convective situations in turn may be taken as evidence for low-level origin of the nuclei. The occurrence of peaks in the nucleus spectra and the presence of these in different regions of the storms indicate the existence of specific types and yet widely distributed nuclei.

The direct relation between rain intensity and nucleus concentration is a very important feature and will require a great deal of elaboration. Dingle and Gatz (1969) have explained the direct variation of contaminant concentrations with rain intensity on the basis of storm structure; for freezing nuclei, their active role in the precipitation process may be equally important. More complete study of this relation, along with the patterns of variation for other types of aerosols (not freezing nuclei), may lead to a better description of the role of the nuclei in the precipitation mechanisms.

The fact that nucleus concentrations in hail and in rain were much the same for the cases studied must, if confirmed, be brought into agreement with models of hail growth. A first interpretation of this result would seem to be that there was very little dry growth during which nuclei that had become active at warmer temperatures were rejected.

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