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The Origin of Hailstone Embryos Deduced from Isotope Measurements

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A refined interpretation of the growth history of 30 hailstones is presented. The stones are analysed by the simultaneous determination of D and  $O^{18}$  on the same samples, the application of a new isotopic cloud model and a more accurate determination of the isotope content of vapor at cloud base ( $R_c$ ). Three questions are specifically addressed. 1) Are the frequently observed big-drop hailstones embryos a) merely melted and recirculated graupel, or b) drops grown by the coalescence process? Evidence is provided by the isotope measurements that interpretation b) is more likely. 2) What is the extent of recirculation of hailstones in severe storms? It is shown that by combining isotope, radar and crystallographic measurements, the presence or absence of recirculation can be demonstrated and consistent trajectories and updrafts can be obtained. 3) What are the temperatures of origin of graupel and drop embryos? By comparing the time sequence of these temperatures in hailstones fallen before and after seeding in the same storm, a possible seeding effect is discussed.

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1. INTRODUCTION

One of the fundamental questions in hail formation is the temperature of the origin of hailstone embryos. It is possible to derive these by means of isotopic measurements from hailstones collected on the ground, if, firstly, the relative enrichment of HDO and  $H_2^{18}O$  in the hydrometeors from which hailstones are grown, and its decrease with height can be estimated reliably. Secondly, the isotope content of the vapor feeding the cloud at cloud base ( $R_0$ ) and the degree of its change during the growth time of the hailstones should be known accurately. The 1D-steady state model presented by Jouzel et al. (this conference) gives more realistic profiles of the isotope content of hailstones  $R_H$  than the previously used adiabatic models. In the new model the assumption that the D-content in the hail layers is essentially that of the accreted cloud droplets was dropped and the interactions of 5 water species have been taken into account. Concerning the second problem the isotope content of subcloud vapour was measured during two summer seasons to assess the value of the traditional  $R_0$ -determination and to obtain a measure of the variability of  $R_0$  during a hailday. With these new tools a refined interpretation of the growth history of 33 hailstones fallen on 3 days is presented. The size of the stones, collected from the ground or quenched directly in the chilled hexane of the hail collector, ranged from 11 to 46 mm and the frequency of graupel or frozen drop embryos in all stones collected on those days is given in Table 1.

2. DETERMINATION OF  $R_0$ , THE ISOTOPE-CONTENT OF VAPOR AT CLOUD BASE.

The  $R_0$  value is usually determined from the extreme values of D,  $^{18}O$  measured in hailstones and by attributing those to growth temperatures of -35C and 0C for example. In this way the relationship between the D content of the condensed phase  $R_c$  and  $R_0$  is obtained. Since in the new model the deuterium content of hail  $R_H$  results from the interaction of 5 water species, no analytical relation exists between the isotope content of the hailstone layers  $R_H$  and  $R_0$ . Therefore a trial and error method has been employed: First a reasonable  $R_0$  is estimated using the adiabatic assumption. The model is then run yielding an approximate  $R_H$ -profile. The D-concentration corresponding to a specific extreme temperature or, better, identified from crystallographic analysis (no large crystals at  $T_c < -24C$ , no small crystals at  $T_c > -15C$ ), is compared with the result of this first approximation. New approximations of  $R_0$  are obtained by shifting the theoretical profile to coincide with an increasing number of temperature reference points. The final  $R_0$  value then yields the optimum  $R_H$ -profile as well as the D-profile of the entrained vapor  $R_e$  by using the average decrease  $dR_e/dz = 0.0039$  ppm/m (Ehhalt, 1974). In this method it must be assumed that the isotope content of subcloud vapor is reasonably constant both vertically and horizontally.

\*Crystal size in the growth layers was inferred from off-center thin sections.

	graupel n	frozen drop n	not classified	total number	temperature at CCL (°C)
August 6, 1977	37	49	14	100	6.7
July 14, 1978	38	52	10	100	11.2
August 6, 1978	28	63	9	100	9.3

Table 1: Embryo classification for the 3 storms. Total number of stones and percentage of the embryo types are given. The cloud base (CCL) temperatures are also tabulated.

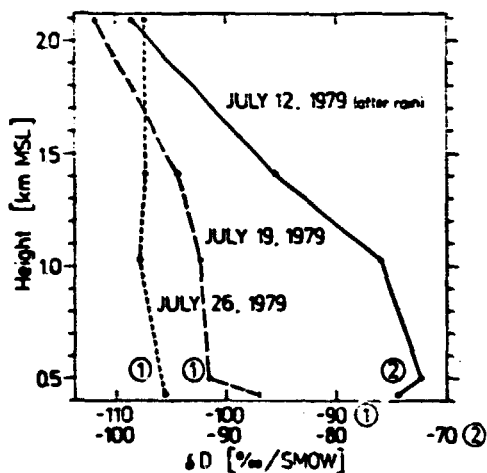


Fig. 1. Three measured deuterium profiles of water vapor in clear air as a function of altitude. Note the different scale on the abscissa for July 12, 1979 which represents a typical gradient after a rain shower.

This has been checked in an experiment with up to 5 stations, sampling simultaneously in and near the experimental area at altitudes between 430 and 2100 m MSL in typical pre-storm conditions. The 3 most important results of a total of 153 vapor measurements on 29 days can be summarized as follows: 1) The D-content of vapor at any one sampling station typically varied by less than 1 ppm within 2-hours (maximum 1.8 ppm at station Neuenkirch probably due to the wet ground). 2) The horizontal variability was surprisingly large, typically 2 ppm and up to 3.5 ppm. This casts doubt on the value of measurements in narrow valleys which might be subject to a very local circulation. The mean relations, obtained on all sampling days are:  
 $\delta D = 7.6 \delta^{18}O + 3.4$  for Emmen (425 m MSL) 1978 and  
 $\delta D = 6.7 \delta^{18}O - 7.0$  for Emmen 1979.  
 Feldmoos 1979 (30 km to the SE, 805 m MSL) yielded  
 $\delta D = 7.1 \delta^{18}O - 2.1$ . The relation for Pilatus (2100 m MSL) resembles most closely the theoretical relation:  $\delta D = 7.0 \delta^{18}O + 9.7$ . 3) The vertical concentration-gradients are small in cloudless situations but up to 3 ppm (for D) after rain (see Fig. 1).

In conclusion, for the  $R_0$ -determination we use the direct vapor measurements of the lowest atmospheric layer, measured in the plain. If none are available, we choose the crystallographic method which gives the smallest intervals for the possible  $R_0$ 's. A comparison of the two independent methods results in almost the same  $R_0$ 's. This gives considerable confidence in the correctness of  $R_0$  and therefore in the model-calculated absolute temperature scales.

## 1. APPLICATIONS TO EMBRYOS

### 3.1 Isotope-content

In Fig. 2 we have plotted the isotope concentrations of the hailstone embryos analysed on August 6, 1977, August 6, 1978 and July 14, 1978 in a  $\delta D - \delta^{18}O$  diagram. The relation  $\delta D = 8.5 \delta^{18}O + \text{const.}$ , valid for equilibrium conditions, is drawn for comparison through the "coldest" ice sample for each day, where equilibrium conditions are most likely to prevail. Any deviation from the equilibrium line is due to kinetic effects from condensation-evaporation processes which are characterized by a slope 3.5 in the above relation (Jouzel and Merlivat, this conference). Fig. 2 shows that the range of  $\delta D$  for the embryos is about 22 ‰ on August 6, 1977 and 1978 and about 32 ‰ on July 14, 1978 which is comparable to the range reported by Roos et al. (1977) for his South African embryos.

Most of the embryos lie close to the equilibrium line and within the measurement uncertainty. If the ice samples from the entire stones are plotted in a similar diagram (not shown), the warmer, clear layers with large crystals tend to show a pronounced evaporation effect; these layers must have been grown in the wet régime. As expected the graupel embryos are closer to the  $D -^{18}O$  equilibrium line than the frozen drop embryos. The latter are generally warmer and show signs of evaporation, especially at the higher isotope contents. This indicates that the drops were evaporating slightly prior to their freezing. This is because they were warmer than the environment, a situation to be

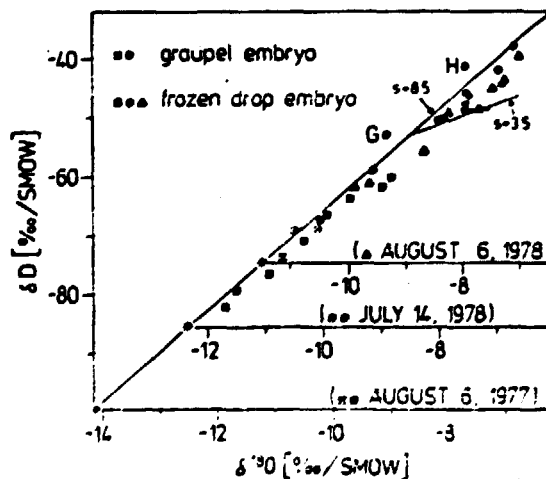


Fig. 2. Plot of the deuterium content vs.  $^{18}O$  - concentration of the 33 analysed hailstone embryos. The equilibrium line with slope 8.5 is plotted through the 3 points (asterisks) representing the coldest ice sample of the day. The non-equilibrium line with slope 3.5 through a selected point is used to correct the measured isotope values for evaporation effects as suggested by Bailey et al. (1969). The error bar indicates the standard deviation of the  $^{18}O$  measurement.

expected with millimeter-sized drops ascending in a moderate updraft and growing by coalescence. On the other hand two frozen drop embryos, designated by letters in Fig. 2, are lying to the left of the equilibrium line for July 14, 1978 which indicates that condensation was occurring on the cold drops prior to their freezing. This and the fact that graupel are generally poorer in isotopes, but drop embryos do not show any substantial evaporation, raises the question whether the drop embryos G and H were merely melted and recirculated graupel, a possibility indicated by workers in Colorado. This problem will be addressed in the next section.

### 3.2 Recirculation

In order to show that it is unlikely that the frozen drop embryos on July 14, 1978 were merely melted graupel, we assume that a graupel grown to a size of 3 mm diameter from an ice crystal nucleated somewhere between the -10 and -15°C-isotherms, would have a D-content between  $R_1$  and  $R_c$  at the -25°C-isotherm (point A in Fig. 3). Suppose now that this graupel falls out of the updraft and is transported below the 0°C-isotherm in weak downdrafts as indicated for instance by the closed loop circulation by Dye et al (1978). The melting time of the graupel to produce a drop of  $r = 1$  mm would be about 100 seconds in a cloud of  $T_c \approx +2^\circ\text{C}$  (Drake and Mason, 1966), whereas the isotopic relaxation time  $t_2$  for the same drop is of the order of 400 s (Stewart, 1975). The situation is schematically depicted in Fig. 3 at the +2°C-isotherm,

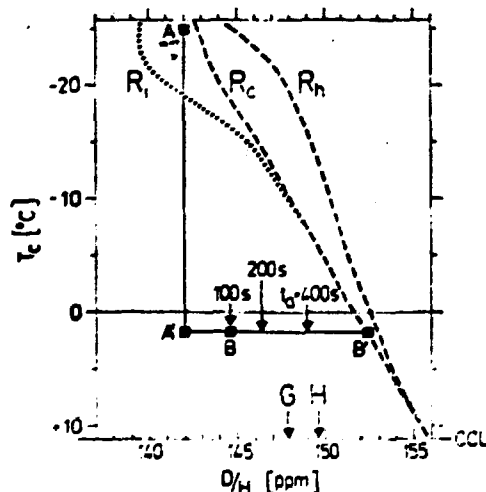


Fig. 3. Deuterium content of cloud water  $R_c$ , cloud ice  $R_1$  and hydrometeor water  $R_h$  as a function of cloud temperature  $T_c$ . The time for a drop of radius  $r = 1$  mm to approach isotopic equilibrium is indicated at the  $+2^\circ\text{C}$ -isotherm. The drop would reach the relaxation time  $t_2$  only after 400 s and would have a D-content of 149.0 ppm. The graupel particle with an assumed diameter of 3 mm starts to melt at A' for 100 s (point B).

where it is shown that the D-content of the drop is always considerably smaller than the one of the water species surrounding it (point B'), unless the drop is kept in balance for an unrealistically long time period. If the drop then started to ascend again in the strong updraft from point B, interacting with cloud droplets only (no breakup), it is unlikely that it would reach D-contents of 147.9 and 149.6 ppm as measured in embryos G and H respectively. It is clear that there are many arbitrary assumptions in this reasoning and each case would have to be calculated separately with varying updraft speeds and LWC's. But the fact that the D-content of all embryos identified as graupel is at least 2 ppm lower than that of the drop embryos and the observation that the two embryos exhibiting condensation have D-contents lying in the range of those for the frozen drop embryos (showing evaporation), leads us to conclude that most embryos were probably grown by coalescence.

### 4. APPLICATION TO TRAJECTORIES

The application of the new isotopic model to the D-profiles of the entire hailstones yields a range of growth temperatures which is much narrower than those obtained earlier with the adiabatic model (Fig. 4). In Fig. 4a the  $\delta D$ -profiles are shown together with the bubble structure of 3 stones of August 6, 1977. From the profile of stone G for example, a rapid upward motion beginning at a radius of  $r = 11$  mm and spanning the whole  $\delta D$ -range of 18 ‰ could be inferred. Incidentally this rapid upward motion is accompanied by the formation of a clear layer, a fact observed in most published isotope analyses of hailstones and already described by Federer et al. (1978). This ascent is seen to be much less dramatic in Fig. 4b where the 3 stones oscillate in a temperature range of merely  $8^\circ\text{C}$  during their entire growth. This means that both embryo and hailstone growth take place in a nearly balanced state and that changes in opacity are not due to large changes in cloud temperature but rather to changes in LWC, ice content and/or drop spectra.

The narrowing of the temperature range of hailstone trajectories due to the use of the more realistic model also changes the earlier calculations of maximum updraft speeds  $u$ . Since the determination of  $u$  is critically dependent on the altitude change of the stone with radius  $dz/dr$ , the earlier excessive speeds of up to 80 m/s would be corrected down to considerably lower values in the present approach.

The amplitudes of the recirculation of large stones which extended over several kilometers in the earlier model are now reduced considerably. Hailstone recirculation is present in only 9 of the 25 stones from which we obtained radial D-profiles. Interestingly, the 3 hailstones with graupel embryos (1 C-type) from the same site on August 6, 1977 show recirculation, whereas the 4 with frozen drop embryos (2A and 2C-type) do not. Crystallographic analysis of the rest of the analyzed ensemble (see Table 1) points in the same

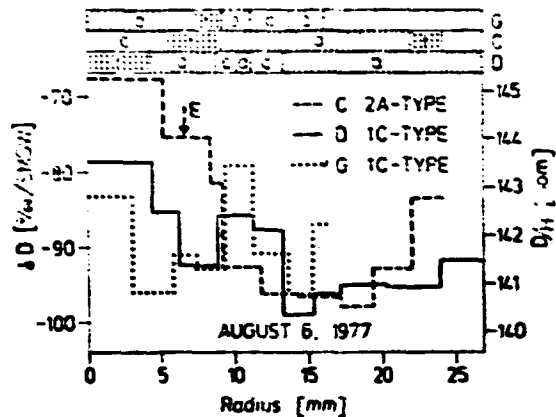


Fig. 4a. Deuterium content  $\delta D$  vs. radius of 3 hailstones from Aug. 6, 1977 with bar indications of bubblyness: opaque, transparent and clear ice.

direction. This is consistent with the graupel originating at a generally colder level than drop embryos and therefore the greater necessity for the graupel to recirculate in order to become large.

#### 5. CONCLUSIONS

The interpretation of the growth history of hailstones and their embryos has been refined with the use of a model which takes into account the interaction of 5 water species, entrainment and fallout. Furthermore the variability of the isotope content of subcloud air ( $R_0$ ) assumed to be constant during the growth time of hailstones, has been investigated by direct measurements.

The following conclusions were obtained from this preliminary data set:

1) The directly determined  $R_0$ -values are very similar to the ones from the independent crystallographic method. This gives considerable confidence in the latter which is used in conjunction with Macklin's composite diagram (no large crystals at  $T_c \leq -24^\circ\text{C}$  and no small crystals at  $T_c \geq -15^\circ\text{C}$ ). Simultaneous  $R_0$ -measurements on the ground show a significant horizontal gradient but a much smaller vertical gradient, both dependent on the local weather condition.

2) The application of the new isotopic model results in a much narrower temperature range for the trajectories indicating that hailstone growth takes place in a nearly balanced state. As the main advantage the new model eliminates some contradictions in the adiabatic model like growth temperatures  $>0^\circ$  and excessive updraft speeds.

3) In most of the hailstone trajectories investigated, no pronounced recirculation of the stones was detected. This is in contrast to results obtained with earlier isotope interpretations which supported the recycling hypothesis.

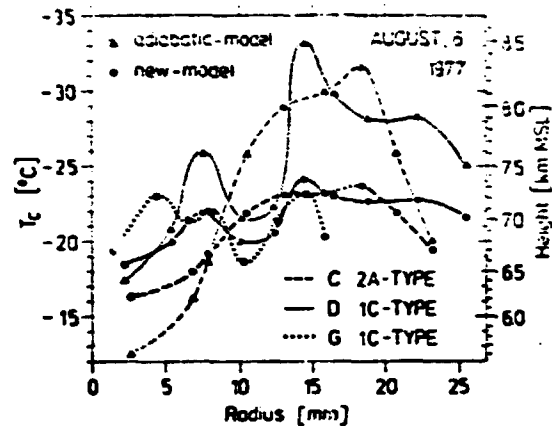


Fig. 4b. Comparison of trajectories of the hailstones in Fig. 4a calculated with the adiabatic (triangles) and the new model (circles).

4) The simultaneous measurement of D and  $^{18}\text{O}$  on the same sample allows to correct for condensation-evaporation effects during hailstone growth. Evaporation effects for D of up to 1 ppm ( $\delta D = 6^\circ/100$ ) are observed in accordance with the laboratory measurements of Bailey et al. (1969).

5) A theoretical comparison of the melting time of a typical graupel with the isotopic relaxation time of the drop produced leads us to conclude that the frozen drops observed were not merely graupel melted below the  $0^\circ\text{C}$ -isotherm and recirculated.

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