Isotopic, Crystal and Air Bubble Structures of Hailstones

William C. Macklin,[1] Charles A. Knight, Howard E. Moore,[2]
Nancy C. Knight and Walter H. Pollock

National Center for Atmospheric Research,[3] Boulder, Colo. 80303

John N. Carras and Suszanne Thwaites

Department of Physics, University of Western Australia, Nedlands

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ABSTRACT

The deuterium, crystal and air bubble structures of 11 large hailstones from three severe storms have been examined. It is emphasized that there are a number of assumptions underlying the interpretation of such data and these are discussed. In seven of the hailstones the ambient temperatures at which they grew were inferred from the crystal size. The deuterium concentrations and ambient temperatures generally show similar variations and the crystal data thereby provide a useful way of placing an absolute temperature scale against the deuterium values. Throughout most of their growth, the hailstones grew in the updraft between about the ambient temperature levels of -17 to $-30°C$. The air bubble analyses showed that the hailstones grew near the wet growth limit or slightly wet and heat balance considerations give values of 2–3 g m^{-3} for the effective liquid water concentrations. On the assumption that the median volume radius of the cloud droplets is 10 μm, the actual liquid water concentrations are then about 4 to 5.5 g m^{-3}.

1. Introduction

The purpose of studying hailstone structure is to obtain the hailstone growth trajectories and the conditions the hailstones experience (updrafts, liquid water concentrations and droplet sizes) during their growth in cumulonimbus clouds. Three methods of hailstone analysis are currently available, i.e., determinations of their isotopic composition and of their crystalline and air bubble structures. It must be emphasized that there are a number of assumptions underlying the interpretation of such data and these are discussed below.

The isotopic method of analysis consists of measuring the ratios of deuterium to hydrogen and of O^{18} to O^{16} as a function of hailstone radius. Isotopic analyses of hailstones have been made by Facy *et al.* (1963), Merlivat *et al.* (1964), Majzoub *et al.* (1968), Macklin *et al.* (1970), Knight *et al.* (1975) and Jouzel *et al.* (1975). The theory of this procedure has been treated in detail in a number of these references and will not be dealt with here. From the present point of view, all that need be noted is that the relative enrichment of the HDO and H_2O^{18} molecular species in the cloud droplets decreases with height [Facy *et al.*, 1963 (Fig. 1); Macklin *et al.*, 1970 (Fig. 9); Knight *et al.*, 1975 (Fig. 2); Jouzel *et al.*, 1975 (Fig. 13)]. However, quantitative calculations of this height variation require knowledge of two factors: 1) the initial value of the isotope ratio in the subcloud layer and its constancy, and 2) the mixing during the rise of the air in the updraft. In the calculations cited it is assumed that the ascent of air is adiabatic, with no mixing. The first factor has been taken into account by some authors by assuming that the most positive isotopic ratios in the hailstones analyzed were produced at a temperature close to 0°C, while the most negative ratios were produced at temperatures about $-35°C$. This gives self-consistent curves for the decrease in isotopic ratio with height (e.g., Merlivat *et al.*, 1964; Macklin *et al.*, 1970; Jouzel *et al.*, 1975). The assumption of constancy of the isotopic ratio of the subcloud vapor over the period of time it takes the hailstones to grow is also necessary for the calculations. If the storm is a traveling one, as are most storms which produce large hail, then it can draw on vapor arising from different sources. Consequently, it is possible that the isotopic ratio of the subcloud vapor fluctuates with time. Strictly, the initial isotopic

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[1] Advanced Study Program Fellow on leave from the Department of Physics, The University of Western Australia.

[2] Department of Physical Sciences, Florida International University, Miami 33199.

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ratios in the subcloud vapor should be sampled and measured but, as yet, this has not been attempted. It should be noted also that, as explained by Bailey *et al.* (1969), there is additional fractionation of the isotopic species during the actual freezing of the droplets on accretion. Bailey *et al.* (1969) have shown that this effect is small and that it is virtually the same for all hailstone layers. Consequently, this only introduces a small systematic difference between the measured isotopic ratios and those calculated from the adiabatic model. This difference can readily be taken into account.

The simplest measure that can be made of the crystalline structure of hailstones is crystal size. This can easily be done from photographs of thin sections of the hailstones taken between crossed polaroids. The crystal size depends on the ambient temperature and decreases as the ambient temperature decreases (Levi and Aufdermaur, 1970; Rye and Macklin, 1975). At ambient temperatures $\lesssim -20°C$, the crystal size is dependent also on the growth temperature of the hailstone which is higher than ambient due to the release of latent heat of the accreted droplets. There are also assumptions underlying this method of analysis:

1) It is assumed that the crystal size data of Levi and Aufdermaur and Rye and Macklin, which were obtained from ice deposits formed on cylinders rotating at rates between 0.5 and 3 Hz, are applicable to hailstone growth. The question of whether the rotation rate affects the crystal size has been investigated by Knight (private communication) who found that the crystal size was unaffected by rotation rates up to 200 Hz. The assumption that the cylinder data are applicable to hailstones is justified by the fact that the crystal size is primarily dependent on the ambient (i.e., droplet) temperature and not on the shape of the object. The physical reason for this is given by Rye and Macklin (1975).

2) It is possible that the crystals have undergone recrystallization, particularly if they have been warmed to 0°C for tens of minutes prior to storage (see, e.g., Knight, 1975). The extent of recrystallization on the structure of hailstones is not at present completely known and it is assumed that, unless obvious, this effect is negligible.

3) The crystal structure will also be affected if the hailstones accrete ice crystals as well as supercooled droplets. Macklin (1961) has shown in icing tunnel experiments that this can occur in the wet growth regime and possibly to some extent in the dry growth regime. Unfortunately there are no aircraft observations of ice crystal concentrations in the hail growth regions of severe storms so that it is not possible to give a quantitative assessment of the magnitude of this effect. However, the agreement between the ambient temperature variations deduced from the crystal size and those deduced from the deuterium data, which are presented below, indicates that any such effect is minimal.

The formation of air bubbles in ice accreted by rotating cylinders has been discussed by Carras and Macklin (1975). These authors showed that the bubble concentration is dependent on the ambient temperature, the deposit temperature and the droplet size. Macklin *et al.* (1976) have shown that there is no difference between the air bubble structures of cylindrical deposits and those of artificial hailstones grown freely suspended in a vertical icing tunnel. Consequently, once the ambient temperature has been determined (either by studies of the isotopic composition or the crystal structure) it is possible to ascertain the hailstone temperature for different droplet sizes. The values of the effective liquid water concentration, EW, experienced by the hailstones can then be calculated from the heat balance equation [see Eq. (2) below].

Macklin *et al.* (1976) used the crystal and air bubble methods of analysis to examine the internal structure of six hailstones having diameters between 4.5 and 6.5 cm. They determined the trajectories from the crystal structure and found that the hailstones remained balanced in the updraft between approximately the -20 and $-25°C$ temperature levels throughout most of their growth. They further showed that the effective liquid water concentrations in which the hailstones grew were between 1 and 3 g m^{-3}. However, their analyses are contingent on the assumptions mentioned above.

In this paper we present analyses of hailstones using all three methods. This serves to remove some of the ambiguities in the isotopic and crystal analyses which arise because of the various assumptions which have to be made.

2. The hailstones and experimental techniques

Eleven hailstones have been analyzed. Six of these fell at Sterling, Colo., on 15 August 1974, five at Yellow Springs, Ohio, on 3 April 1974 and one at Coffeyville, Kans., on 3 September 1970. The hailstones were obtained from interested people who had collected and temporarily stored them in deep-freezers. Prior to analysis the stones were stored in a cold room at a temperature of about $-25°C$. Typical photographs of thin sections of the stones are shown in Fig. 1. The reason for choosing the particular Coffeyville hailstone (CK21) for analysis was its striking layered structure and approximately spherical symmetry, as distinct from most of the Coffeyville hailstones (see Knight *et al.*, 1975).

The isotopic analyses were carried out at the National Center for Atmospheric Research in the manner described by Knight *et al.* (1975). A section about 3 mm thick was cut through growth center of the

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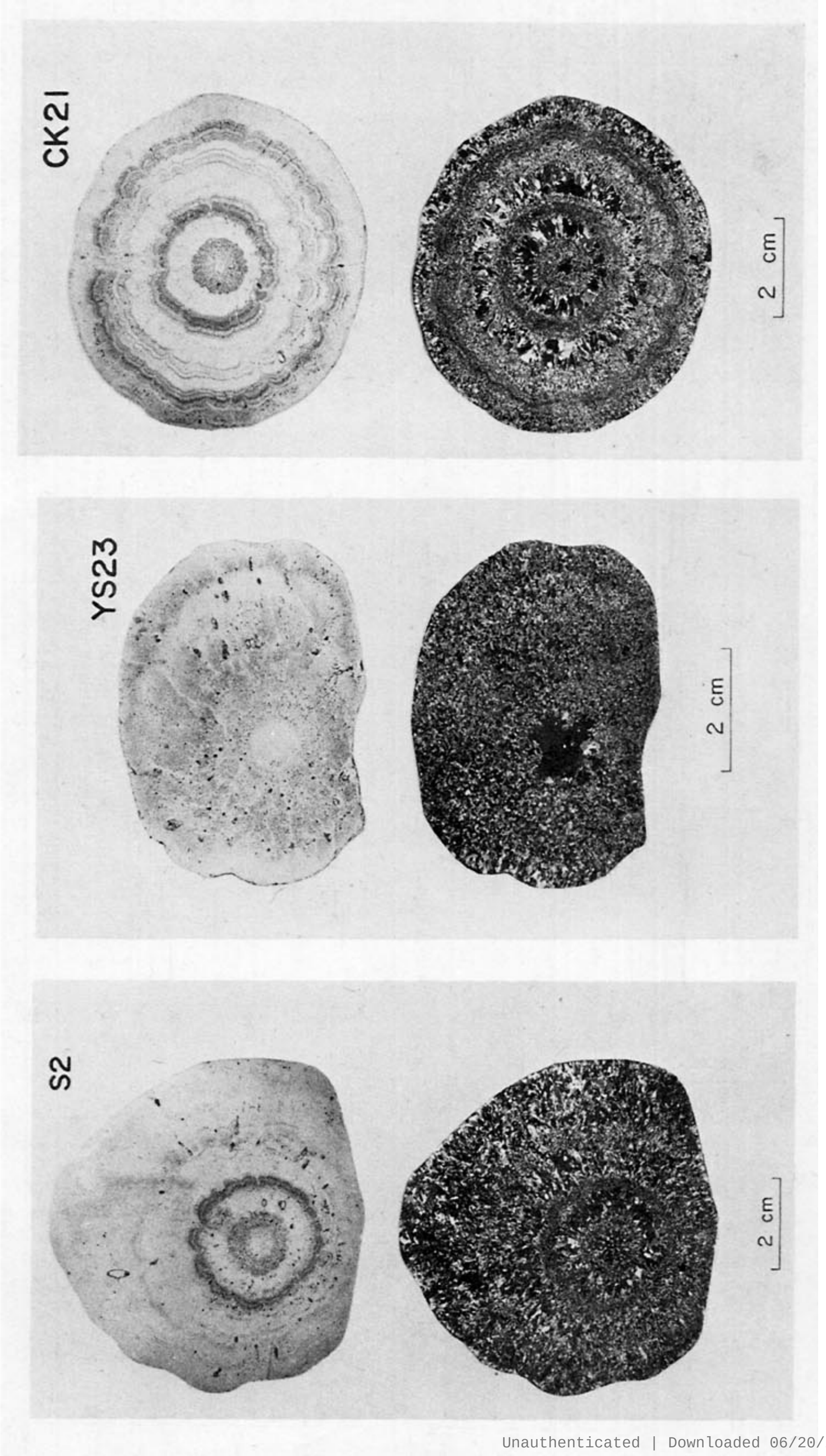

FIG. 1. Thin sections of typical hailstones viewed in transmitted and polarized light.

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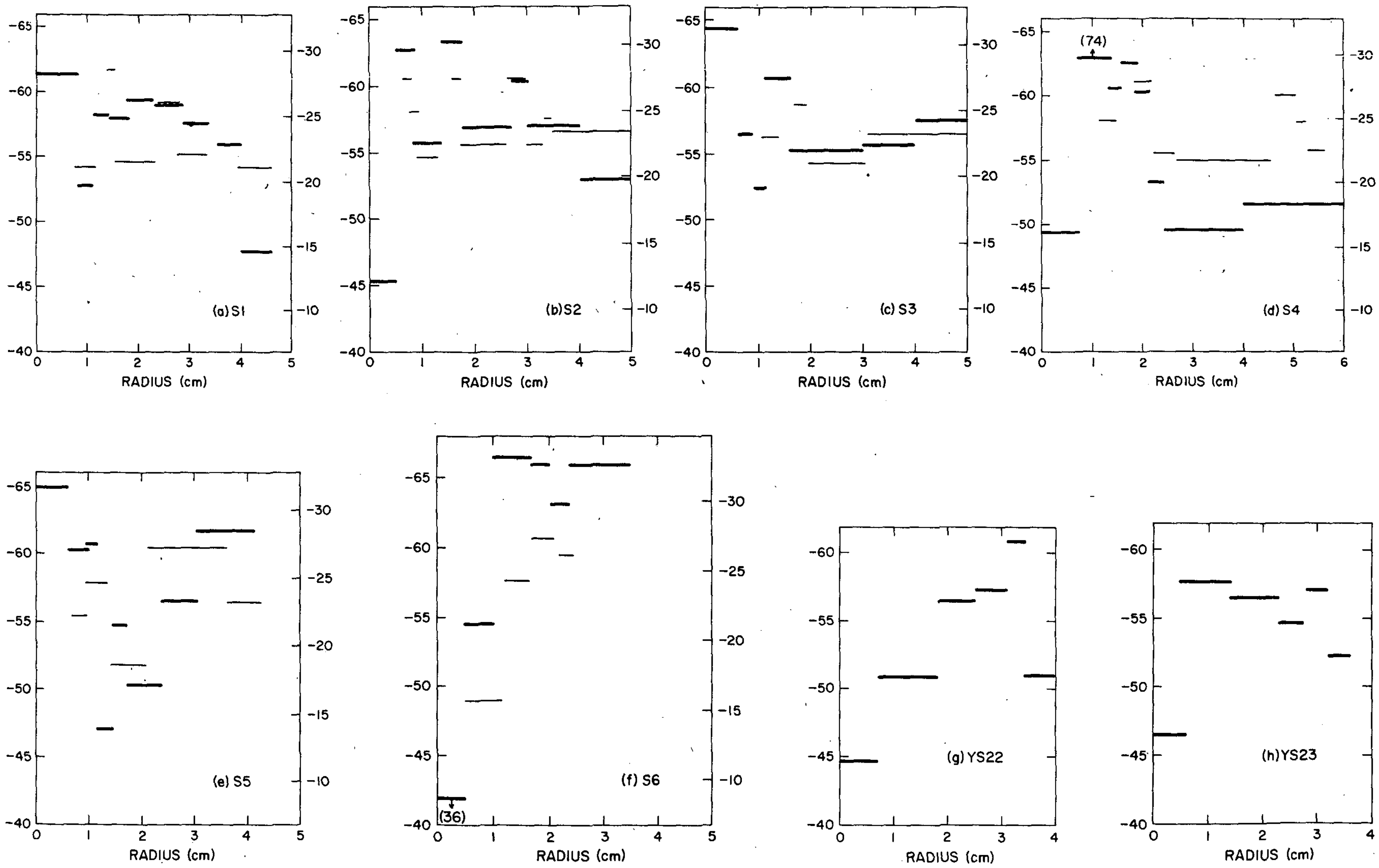

FIG. 2. Values of deuterium concentrations, expressed as δD(‰), as a function of the semi-major axis of the hailstones (heavy lines). Where appropriate the values of the ambient temperature T_a(°C) deduced from the crystal lengths are also shown (light lines). The standard error in the δD values is ±0.5‰ and the error in the ambient temperatures is ±2°C. Hailstones S1, S3 and S5 had bubbly spherical embryos while hailstones YS22 to 25 had frozen drop embryos. The left vertical scale in each part refers to δD, the right to T_a.

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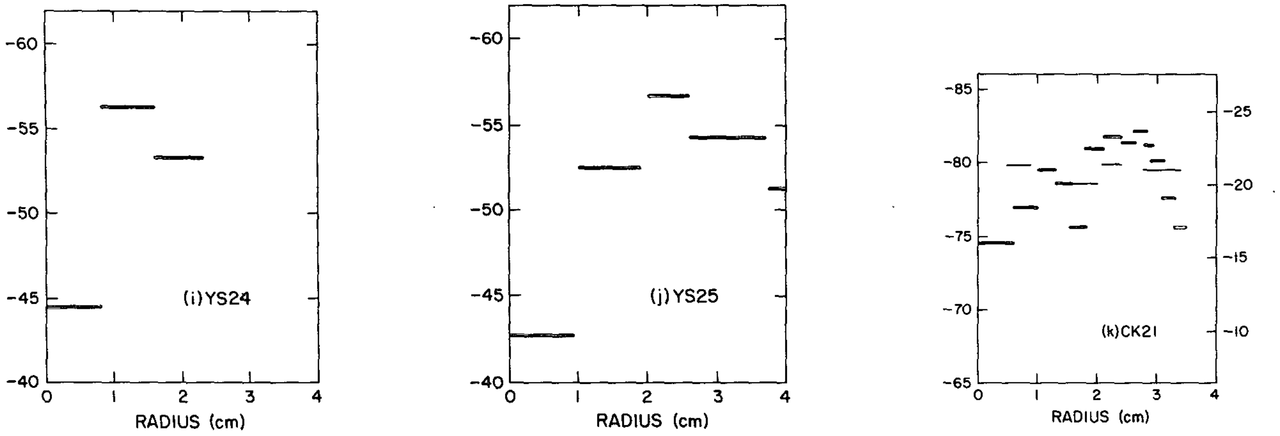

FIG. 2 (*continued*).

stone in such a way that the stone's longest and shortest axes were included in the section. Each growth layer was separated by chipping it off piece by piece with a sharp knife. The chips were then sealed into test tubes. This technique permitted the lobe structure to be followed and provided the minimum size sample (a few cubic millimeters) from each layer necessary for mass-spectrometric analysis. The D/H ratio was then measured by a standard procedure (see, e.g., Friedman, 1953) using an Atlas MS86 mass spectrometer. The deuterium concentration is expressed as a δ value, defined as the relative variation of the D/H ratio R of the sample [SA] from the D/H ratio of standard mean ocean water [SMOW] in parts per mille (Craig, 1961), i.e.,

$$\delta_{\mathrm{SMOW}}=[(R_{\mathrm{SA}}-R_{\mathrm{SMOW}})/R_{\mathrm{SMOW}}]\times 1000. \quad (1)$$

The standard deviation of the δ measurements was ±0.5‰. This is the usual error in mass-spectrometric measurements of this kind (see Knight *et al.*, 1975).

Thin sections of the hailstones, adjacent to those used for the deuterium analyses, were photographed between crossed polaroids to determine the crystal size. The crystals were sized at the Department of Physics, the University of Western Australia. The other halves of the hailstones were air-freighted under dry ice to The University of Western Australia and their air bubble concentrations and size distributions determined in the manner described by Carras and Macklin (1975).

3. Results and discussion

a. *Isotopic composition*

The deuterium data are shown in Figs. 2a–k. Here the δD values are plotted as a function of the semi-major axes of the hailstones. In the case of the Coffeyville hailstone δD measurements were made on samples taken along both sides of the major axis. The two sets of values so obtained agreed to 1–2‰ and the mean of the values has been plotted in Fig. 2k. In some stones the δD values show only a small variation at the larger radii while others show more marked variations. As pointed out above, although the δD measurements have a standard deviations of ±0.5‰, the δD values possibly fluctuate due to fluctuations in the initial values in the subcloud vapor, the magnitudes of which are unknown. The Coffeyville hailstone has more negative δD values than the other stones. In fact, the values are somewhat more negative than those in other Coffeyville hailstones reported by Knight *et al.* (1975), which range typically from −60 to −80‰. This implies that it grew at slightly lower temperatures.

The types of embryos in the hailstones were classified visually in a manner similar to that of Knight and Knight (1970). Bubbly spherical embryos (hailstones S1, S3 and S5) had δD values ranging from −62 to −65‰, while frozen drop embryos (YS22 to 25) had values ranging from −43 to −47‰. Consequently, the former are formed at relatively low temperatures while the latter are formed at relatively high temperatures, as indicated by the crystal structures analyses discussed below.

b. *Crystal structure*

The lengths of the crystals were measured in the directions of the semi-major and semi-minor axes of the sections and the ambient temperatures were obtained using the data of Rye and Macklin (1975). The two sets of ambient temperatures so obtained differed by only 1–2°C. These temperatures are also shown in Fig. 2. No systematic measurements could be made on the Yellow Springs hailstones. The crystals were small indicating that they were formed by re-orientation (see Rye and Macklin, 1975) and had aspect ratios close to unity indicating that they had undergone recrystallization, as discussed by Macklin *et al.* (1976). Also, no reliable temperature measurements could be obtained for the hailstone embryos

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from the crystal sizes. Based on the assumption of adiabatic ascent, the fall of δD with the temperature over the range −15 to −35°C is approximately 1‰ $°\mathrm{C}^{-1}$ (see Macklin *et al.*, 1970, Fig. 6). Thus the scales of the δD and the temperature values in Fig. 2 are related.

To facilitate comparison between the two sets of data, the δD and ambient temperature scales in Fig. 2 have been drawn so that the mean of the δD values lies opposite the mean of the ambient temperature values. Given the experimental error in the ambient temperature measurement from the crystal size (±2°C) and that the δD values are subject to fluctuations in the D/H ratio in the subcloud vapor, there is a reasonable degree of correlation between the variations in δD and the temperature values for the hailstones studied. Hailstone S2 shows the best correlation with both the δD and temperature values rising and falling in unison. The data for hailstones S3, S4, S5 and S6 are also correlated but to a lesser degree while the data for S1 are poorly correlated. In the case of hailstone CK21 the δD values are more scattered than the temperature values. It is evident from the ambient temperature values that, at the larger radii, the hailstones grew in the updraft at ambient temperatures between about −17 and −30°C, as found by Macklin *et al.* (1976). The bubbly spherical embryos were evidently formed at temperatures around −30°C. If it is assumed that the same temperature scale applies to the Yellow Springs hailstones, then the embryos were formed at temperatures between −10 and −15°C. The stones then rose in the updraft and grew at temperatures between about −20 to −25°C which is consistent with the Sterling hailstones. The crystal sizes in the Coffeyville hailstone suggest that it grew at temperatures slightly below −20°C. This indicates that major portions of the Coffeyville hailstones analyzed by Knight *et al.* were formed at temperatures somewhat higher than −20°C because of their more positive δD values.

c. Air bubble structure

The air bubble concentrations and size distributions in the transparent hailstone layers showed characteristics similar to those exhibited by layers formed under wet growth conditions (see Carras and Macklin, 1975). The bubble concentrations had values less than 10^6 cm^{-3} and the size distributions were broad. The opaque layers showed characteristics that were intermediate between dry and wet growth. The bubble concentrations varied from 10^6 to 6×10^6 cm^{-3}, while the mean bubble radius of each distribution varied from 3 to 8 μm. The bubble concentrations suggest that the opaque layers were formed at surface temperatures within a degree or two of 0°C. Overall, the bubble data indicate that the hailstones grew at or close to the wet growth limit.

TABLE 1. Values of the effective liquid water concentrations *EW* and the actual liquid water concentrations for a droplet diameter of 10 μm.

Hailstone	Radius (cm)	*EW* ($\mathrm{g\ m^{-3}}$)	W ($\mathrm{g\ m^{-3}}$)
S1	1.3	2.6	5.3
	1.9	2.0	4.4
	2.5	2.2	5.1
	4.2	2.3	6.3
S2	1.1	2.5	4.7
	1.5	2.8	5.5
	2.7	2.5	5.4
	4.4	2.3	6.0
S3	1.2	2.6	4.9
	1.6	2.4	4.9
	2.5	2.1	4.5
	4.1	2.2	5.8
CK21	0.7	3.2	5.0
	1.7	1.9	4.0
	2.3	1.9	4.3
	3.3	1.9	5.0

Assuming that this was the case, values for *EW* have been calculated for the layers of those hailstones having approximately spherical symmetry by inserting $T_d=0°\mathrm{C}$ in the heat balance equation (see, e.g., Macklin and Payne, 1967) with the result that

$$\begin{aligned}\tfrac{1}{4}EWV[L_f+c_w(T_a-T_m)+c_i(T_m-T_d)]\\ =\chi\,\mathrm{Re}^{0.5}[\mathrm{Pr}^{\frac{1}{3}}K(T_d-T_a)\\ +\mathrm{Sc}^{\frac{1}{3}}L_vD(\rho_s-\rho_e)](2R)^{-1},\quad(2)\end{aligned}$$

where E is the collection efficiency, W the liquid water concentration, V the hailstone fallspeed, L_f the latent heat of fusion of water, c_w the specific heat of water, c_i the specific heat of ice, T_a the ambient temperature, T_m the melting point of ice (0°C), T_d the hailstone temperature, χ the heat transfer coefficient, Re the Reynolds number, Pr the Prandtl number, K the thermal conductivity of air, Sc the Schmidt number, L_v the latent heat of vaporization of ice, D the coefficient of diffusion of water molecules in air, R the hailstone radius, and ρ_s and ρ_e are the water vapor densities at the hailstone surface and in the environment, respectively. The two sides of the equation represent the rate of liberation of heat by the freezing droplets and the rate of dissipation of this heat to the environment by forced convection processes, respectively. In calculating *EW*, values of χ determined experimentally by Macklin and Bailey (1968) have been used.

The *EW* values are listed in Table 1; they range from about 2–3 $\mathrm{g\ m^{-3}}$ which are consistent with those obtained by Macklin *et al.* (1976). Calculations of the actual liquid water concentrations require a knowledge of the collection efficiency of hailstones for cloud droplets. Macklin and Bailey (1968) have measured

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the collection efficiencies of smooth spheres and artificial hailstones having realistic surface roughness held stationary in an icing tunnel, while Carras and Macklin (1973) determined the collection efficiencies from the growth rates of artificial hailstones growing freely suspended in a vertical icing tunnel. Both sets of values cover the range of Reynolds numbers appropriate to large hailstone growth and both are 10–15% lower than the collision efficiencies calculated by Langmuir and Blodgett (1946). Macklin *et al.* have argued that the median volume radius of the cloud droplets is about 10 μm and approximate values for the actual liquid water concentrations have been calculated from the values of *EW* using this value for the droplet size and the collision efficiencies of Langmuir and Blodgett reduced by 10%. These are also given in Table 1. Most of the values range from about 4 to 5.5 g m^{-3} which are about the values to be expected in severe storms. Some of the values, particularly at the larger radii, are over 6 g m^{-3} which may be somewhat high. This may be because the droplet radius chosen is too small. Alternatively, it indicates that the outer layers of the hailstones were formed in the wet growth regime, since the liquid water concentrations listed in Table 1 are essentially the critical values. The latter alternative is supported by the air bubble structure of these layers.

4. Conclusions

The deuterium concentrations and the ambient temperatures deduced from the crystal structure of large hailstones generally show similar variations even though there are completely different assumptions underlying the two types of analysis. However, it is not in general possible to determine the actual temperatures from the deuterium values alone. In some cases the crystal structure analysis provides a useful way of placing an absolute temperature scale against the deuterium values. This obviates the necessity of making any explicit assumptions in the isotopic method of analysis.

The results indicate that, throughout most of their growth, the hailstones grew in the updraft between ambient temperature levels of ~ -17 to $-30°$C. This corresponds to a height interval of about 2 km. The deuterium values indicate that the bubbly spheriical embryos were formed at high temperatures (-10 to $-15°$C). The air bubble analyses show that the transparent layers in the hailstones were formed in the wet growth regime, while the opaque layers were formed near the wet growth limit. On this basis calculations of the effective liquid water concentrations *EW* give values of 2–3 g m^{-3}. On the assumption that the median volume radius of the cloud droplets is 10 μm, the actual liquid water concentrations are then about 4 to 5.5 g m^{-3}.

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