

Stable Isotopes in Hailstones. Part I: The Isotopic Cloud Model

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ABSTRACT

Equations describing the isotopic balance between five water species (vapor, cloud water, rainwater, cloud ice and graupel) have been incorporated into a one-dimensional steady-state cloud model. The isotope contents of the various water substances were calculated as a function of height. From these profiles, the isotopic concentration δ_H of the water accreted by hailstones growing in this model cloud is determined. The results are compared with those obtained from the adiabatic model (δ_{AM}). The differences are substantial. A sensitivity study shows that the δ_H profiles for two different soundings are dependent on the changes in the droplet-size distributions, which influence the conversion of cloud water to rainwater, and, to a lesser degree, on entrainment. If a reasonable concentration of raindrops is allowed, the results are remarkably stable if all the other adjustable parameters are varied. The application of the model to isotopic data of hailstones and its comparison with the adiabatic model in Part II shows that it gives realistic results.

1. Introduction

The measurement of the deuterium (D) and oxygen 18 (O^{18}) content of hailstone growth layers has been used since the early sixties (Facy *et al.*, 1963) to derive information about hail growth trajectories. The method is based on the fact that during phase transitions within a cloud, the condensed phases acquire a greater concentration of the heavy HDO and H_2O^{18} molecules than the vapor phase. This is based on the fact that each molecular species has a different equilibrium vapor pressure. At equilibrium the relative enrichment is determined by the fractionation factors $\alpha = C(\text{condensed phase})/C(\text{vapor})$, where C is the isotope content, i.e., the concentration N of D (O^{18}) atoms to that of H (O^{16}) atoms, i.e., $C = N_D/N_H$ or N_{18}/N_{16} (see list of symbols in the Appendix). This isotope content is usually expressed as the deviation δ from Standard Mean Ocean Water (SMOW):

$$\delta = (C - C_{SMOW})/C_{SMOW} \quad (1)$$

For the interpretation of isotopic results of hailstone samples it is assumed that the isotope content of the condensed water, from which hailstones grew, can be calculated as a function of height. So far the calculations have been based on the adiabatic assumption.

In adiabatic ascent, δ (condensate) changes with height because the mixing ratio of water vapor de-

creases substantially and α increases from 0 to -35°C (by 6.6% for D and 0.6% for O^{18}). In a discussion of the adiabatic model (AM), proposed by Merlivat *et al.* (1965), Jouzel *et al.* (1975) gave a number of stringent conditions for its exact validity. Some of the assumptions which must be made are 1) no mixing of the ascending parcel with environmental air, 2) no depletion by drops and hailstones, 3) no ice, 4) no fallout, 5) absence of raindrops, i.e. isotopic equilibrium between the condensed phase and the water vapor, 6) no isotopic fractionation upon freezing, 7) spatial and temporal constancy of δ_0 and Q_{v0} , and 8) no shedding of excess water in hailstone growth.

In cases where the range of the isotopic content in the hailstones is large, the application of the AM can lead to contradictions. Roos *et al.* (1977) found it impossible to reconcile the observed D -range with the model without assuming a significant variation of δ_0 during the storm. The same problem occurred in the interpretation of the samples to be described in Part II.

New calculations of the isotope content of the five different forms of water (vapor, cloud water, rainwater, cloud ice and graupel) as a function of altitude, are presented using a refined version of the one-dimensional (1D), steady-state cloud model of Hirsch (1971). A description of this model has recently been published by Orville *et al.* (1980). It is a variation of the Pennsylvania State University parameterized

cloud model used by Weinstein and McCready (1969). A sensitivity analysis of the cloud microphysical parameters of the time-dependent version of the model has been given by Weinstein (1970) and the merits and limitations of 1D models have been widely discussed by Warner (1970), Simpson (1971), Cotton (1971) and Weinstein (1971). Despite their limitations, the 1D models are extremely convenient for both operational purposes and for theoretical studies, because of their ability to incorporate rather complicated microphysics while maintaining the ease and rapidity of a computer simulation. Equations dealing with isotopic exchange processes between the various water substances considered are incorporated into the model, to study the effects of their interactions and of entrainment on the isotope content of the hailstones.

A major impetus for developing this model has been the fact that in many Swiss storms the presence of supercooled raindrops is suggested by the high frequency of frozen drop hailstone embryos (Federer and Waldvogel, 1978). Since the AM excludes raindrops [condition 5) above], it was felt worthwhile to investigate the effect of their presence (and that of ice particles) on the isotope content profiles using a simple numerical cloud model. While conditions 1)–5) of the adiabatic assumption can be dropped in the isotopic cloud model (ICM), a new set of assumptions is introduced by the many disposable parameters contained in the 1D models. It is therefore necessary to test the model predictions against observations. The main test employed has been the accuracy with which one can predict the heights reached by clouds grown in a given environment. Although this is not a particularly critical test, it has been used to predict the intensity of hailbearing clouds with good success in Colorado (Foote and Mohr, 1979). The ICM has been calibrated in a similar fashion with Swiss soundings and radar measurements of cloud tops. The “tuned” model was then used to investigate the sensitivity of the computed isotopic profiles to the various adjustable parameters within their range of observational limits. Although such a model cannot represent the microphysical and dynamical interactions in a hailstorm correctly, its use is justified in order to try to answer the following questions: 1) How do the interpretations of the isotope content of hailstones based on the ICM compare with those based on the simple AM? 2) Which model gives more consistent results when used in conjunction with the crystallography of hailstone sections, D-content determinations of subcloud water vapor, and radar data for selected well-documented storms?

In the present paper the ICM is described and then subjected to tuning by adjusting the key parameters, to achieve optimal predictions of radar cloud tops. Finally, the results of the sensitivity analyses will be reported. In Part II the two questions

above will be addressed using a large data set of field observations, and it will be shown (among other things) that some contradictions of the interpretations of isotopic data obtained with the AM can be eliminated with the ICM.

2. The cloud model and its calibration

In contrast to the adiabatic model (AM), in the numerical cloud model the mixing ratios of five water substances are calculated at each height step of the ascending plume. Taking entrainment and fallout into account, the rate of change with height of the total water is subdivided into the contributions from vapor, cloud water, rainwater, ice and graupel particles and is given by the mass continuity equation

$$\frac{dQ_v}{dz} + \frac{dQ_c}{dz} + \frac{dQ_r}{dz} + \frac{dQ_i}{dz} + \frac{dQ_g}{dz} = -\mu(Q_v + Q_c + Q_r + Q_i + Q_g - Q_c) - F_r - F_g. \quad (2)$$

The differences between this equation and that given by Hirsch (1971) are the alternate treatment of the entrainment (the mixing ratios of rainwater and graupel are also diluted during entrainment), and a corrected distribution of the newly condensed water between Q_c and Q_i [see Eq. (3)].

The set of equations giving the rate of change with height of the various mixing ratios may be written

$$\left. \begin{aligned} \frac{dQ_c}{dz} &= P_{vc} - P_{1ci} - P_{1cr} - P_{2cr} \\ &\quad - P_{2cg} - \mu[\chi(Q_v - Q_c) + Q_c] \\ \frac{dQ_i}{dz} &= P_{vi} + P_{1ci} - P_{1ig} - P_{2ig} \\ &\quad - \mu[(1 - \chi)(Q_v - Q_c) + Q_i] \\ \frac{dQ_r}{dz} &= P_{1cr} + P_{2cr} - P_{1rg} \\ &\quad - P_{2rg} - F_r - \mu Q_r \\ \frac{dQ_g}{dz} &= P_{2cg} + P_{1ig} + P_{2ig} + P_{1rg} \\ &\quad + P_{2rg} - F_g - \mu Q_g \end{aligned} \right\} \quad (3)$$

In these equations the formation and loss of each water species is expressed. Rainwater, for instance, is formed by autoconversion and coalescence from cloud water (P_{1cr} and P_{2cr} , respectively) and is lost by freezing of drops and their accretion on preexisting graupel (P_{1rg} and P_{2rg}), as well as by fallout F_r and by dilution through mixing (μQ_r).

The freezing function for rainwater P_{1rg} is modeled according to the stochastic freezing of supercooled drops given by Bigg (1953). Translating his equation for the freezing probability of drops with volume V

TABLE 1. Results of the sensitivity analysis for seven adjustable parameters in the cloud model for 14 July 1978 and 6 August 1977 (values in parentheses). The variation of rainwater content Q , [g kg^{-1}] is given for the extreme values of each parameter indicated, at four relevant temperatures. The other parameters were held constant at the following values: $N_c = 300 \text{ cm}^{-3}$, $D_c = 0.1$, $a = 0.1$, $w_0 = 5 \text{ m s}^{-1}$, $f(T) = -15$ to -25°C , $A = 0.5 \text{ g kg}^{-1}$. If, for instance, the entrainment constant a is increased from 0.0 to 0.2, the Q value at $T = -15^\circ\text{C}$ is reduced by 0.636 g kg^{-1} on 6 August 1977. Columns are identified for discussion in text.*

Parameter	N_c and D_c (cm^{-3})	a	w_0 (m s^{-1})	$f(T)$ ($^\circ\text{C}$)	A (g kg^{-1})	A^* ($^\circ\text{C}^{-1}$)	B^* ($\text{m}^{-3} \text{ s}^{-1}$)
Extreme values	300 / 2000 and / and 0.2 / 0.1 (1)	0.0/0.2 (2)	2.0/8.0 (3)	-15, -25/-10, -25 (4)	0.5/1.5 (5)	0.6/0.8 (6)	$10^{-10}/10^{-12}$ (7)
$T_c = -10^\circ\text{C}$	-1.85 (-1.26)	-0.63 (-0.31)	0.00 (-0.49)	0.00 (0.00)	-0.04 (-0.07)	-0.03 (-0.04)	-0.12 (-0.13)
$T_c = -15^\circ\text{C}$	-1.88 (-1.05)	-0.72 (-0.64)	+0.12 (+0.49)	-0.15 (-0.11)	-0.40 (-0.02)	-1.93 (-1.76)	-2.36 (-2.24)
$T_c = -20^\circ\text{C}$	-0.08 (-0.03)	-0.08 (-0.07)	+0.04 (0.04)	+0.01 (0.00)	0.00 (-0.01)	-0.91 (-0.35)	-2.06 (-0.93)
$T_c = -25^\circ\text{C}$	0.00 (0.00)	0.00 (0.00)	0.00 (0.0)	0.00 (0.00)	0.00 (0.00)	-0.03 (-0.01)	-0.01 (-0.02)

* In column (1) the variation is calculated for the pairs $N_c = 300 \text{ cm}^{-3}$ and a dispersion of 0.2 vs. $N_c = 2000 \text{ cm}^{-3}$ with smaller $D_c = 0.1$. The freezing intervals in column (4) are -15 to -25°C vs. -10 to -25°C .

at temperature T for a time t into terms of the concentration of raindrops and integrating, yields for the production term P_{irg} (Wisner *et al.*, 1972, p. 1165):

$$P_{\text{irg}} = [20\pi^2 B^* N_0 \rho_w \{ \exp[A^*(273 - T)] - 1 \}] / (w - v_i) \lambda^7.$$

The constants A^* and B^* in Bigg's equation may vary considerably depending on the nature of the rain samples considered. The influence of varying these

constants on the development of the mixing ratios and the isotope concentrations of the different forms of water was calculated. As examples the sensitivities of Q_r and δ_H to variations of A^* and B^* are given in Tables 1 and 2. Values of A^* between 0.6 and 0.8 ($^\circ\text{C}^{-1}$) and of B^* between 10^{-10} and $10^{-12} \text{ m}^{-3} \text{ s}^{-1}$ are used in models, based on laboratory measurements for rainwater and doubly distilled water (Wisner *et al.*, 1972; Cotton, 1972). Since in a hailcloud the nucleus concentration might be fairly high, the values $A^* = 0.66 \text{ (}^\circ\text{C}^{-1}\text{)}$ and $B^* = 10^{-10} \text{ m}^{-3} \text{ s}^{-1}$

TABLE 2. Results of the sensitivity analysis for nine adjustable parameters in the ICM for 14 July 1978 and 6 August 1977 (values in parentheses). The variation of δ_H (‰) for deuterium is given for the extreme values of the parameter indicated, at four relevant temperatures. The other parameters were held constant at the following values: $N_c = 300 \text{ cm}^{-3}$, $D_c = 0.1$, $a = 0.1$, $d\delta_e/dz = -25.0\text{‰ km}^{-1}$, $w_0 = 5 \text{ m s}^{-1}$, $f(T) = -15$ to -25°C and $A = 0.5 \text{ g kg}^{-1}$. If, for instance, a very continental spectrum is chosen for 6 August 1977, the D value of a hailstone increases by 10.3‰ at $T_c = -20^\circ\text{C}$, whereas it decreases (becomes poorer) by 9.3‰ if a freezing constant $B^* = 10^{-12}$ is chosen instead of $B^* = 10^{-10} \text{ m}^{-3} \text{ s}^{-1}$.*

Parameter	N_c and D_c (cm^{-3})	a	$d\delta_e/dz$ (‰ km^{-1})	w_0 (m s^{-1})	$f(T)$ ($^\circ\text{C}$)	A (g kg^{-1})	A^* ($^\circ\text{C}^{-1}$)	B^* ($\text{m}^{-3} \text{ s}^{-1}$)	n
Extreme values	300 / 2000 and / and 0.2 / 0.1 (1)	0.0/0.2 (2)	-12.8/-25.0 (3)	2.0/8.0 (4)	-15, -25/-10, -25 (5)	0.5/1.5 (6)	0.6/0.8 (7)	$10^{-10}/10^{-12}$ (8)	0/1 (9)
$T_c = -10^\circ\text{C}$	-1.9 (-1.7)	+2.1 (+4.5)	-0.4 (-0.4)	-1.1 (-2.2)	0.0 (0.0)	-0.1 (-0.2)	-0.1 (-0.1)	-0.2 (-0.3)	+0.1 (+0.2)
$T_c = -15^\circ\text{C}$	-1.2 (-0.9)	+2.3 (+3.9)	-0.5 (-0.6)	-1.0 (-1.4)	-0.3 (-0.3)	-0.1 (-0.1)	-5.5 (-6.8)	-7.0 (-8.4)	-0.1 (+0.2)
$T_c = -20^\circ\text{C}$	+9.0 (+10.3)	+3.4 (+2.3)	-0.7 (-1.0)	+1.0 (+4.3)	+2.5 (+4.5)	+0.3 (+0.5)	-6.4 (-4.07)	-11.5 (-9.3)	-0.4 (-0.8)
$T_c = -25^\circ\text{C}$	+13.1 (+14.9)	+3.8 (+0.7)	-0.8 (-1.3)	+0.5 (+6.6)	+3.8 (+6.0)	+0.2 (+1.3)	-0.7 (+1.0)	-1.45 (+0.8)	-0.6 (-1.0)

* See Table 1 footnote.

from Wisner *et al.* (1972) were chosen in the present work.

Formation of rain by the shedding of drops from existing hailstones and melting of precipitating ice are not taken into account. The influence of those two processes on the isotopic profiles is thought to be minor for the following reasons. Shedding takes place without isotopic fractionation and the drops formed are isotopically not very different from those resulting from autoconversion and coalescence at the same level. Melted graupel can be recirculated, but the extent of this contribution to the rainwater content is probably rather small and it is therefore assumed that it does not significantly influence the mass balance at the injection level.

It was thought that the freezing function for cloud water $f(T)$ had a decisive influence on the profiles. However, the change from a linear $f(T)$ to an exponential one showed that only the freezing temperature plays an important role, so that the original linear function was retained.

Latent heat of freezing leads to a temperature increase $\Delta T = T' - T_c$ of the parcel, where T_c is the cloud temperature before freezing and T' the temperature resulting from the freezing. This warming is expressed by the formula for isobaric freezing given by Saunders (1957), but multiplied by the proportion of water which actually freezes $Q^*/(Q_c + Q_r)$ at this height. The formula reads

$$\Delta T = \frac{(Q_c + Q_r)L_f + \frac{\epsilon L_s}{p}(\bar{e}_w - e_i)}{c_{p,a} + (Q_c + Q_r)c_w + (Q_i + Q_g)c_i + \frac{\epsilon L_s^2 e_i}{pR_v T^2}} \times \left(\frac{Q^*}{Q_c + Q_r} \right), \quad (4)$$

where

$$\bar{e}_w = \frac{e_w(Q_c + Q_r) + e_i(Q_i + Q_g)}{Q_c + Q_r + Q_i + Q_g}$$

is the actual water vapor pressure in the parcel before the freezing of Q^* at temperature T . Eq. (4) leads to cloud top heights which are 5–10% higher than in the original Hirsch formulation. The approximation \bar{e}_w is a weighted mean between e_w and e_i that becomes e_i at total glaciation.

The model was calibrated by calculating the maximum cloud top height H_{max} (where $w = 0$) with different sets of adjustable parameters, using the official Swiss (1200 GMT) soundings at Payerne, which is about 80 km to the southwest of the experimental area. This was done for 68 days with strong convection in the area covered by the Grossversuch IV radars (Federer, 1977). The results were compared with observed radar echo top heights. The

parameter set yielding the best correlation was adopted for subsequent analyses. The quality of the model output depends on the representativeness of the sounding, which is difficult to judge. To avoid subjective bias, most soundings were used without modifications, except in cases with pronounced air mass changes. Cloud base observations made in the Geneva region support the use of the convective condensation level (CCL) as a cloud base. The model was then run with a mixing constant $a = 0.1$ and a starting velocity $w_0 = 5 \text{ m s}^{-1}$, linear freezing for cloud water between -15 and -25°C , a step $\Delta z = 20 \text{ m}$ and an updraft radius R_{up} , which was computed from PPI radar data for the largest cell on each day. The area of the largest 45 dB(Z) contour S was determined and R_{up} was taken as its equivalent radius

$$R_{up} = (S/\pi)^{1/2}. \quad (5)$$

The results are depicted in Fig. 1. The correlation coefficient of 0.78 for the Swiss storms is satisfactory and similar to that obtained by Hirsch (1971) for storms in South Dakota.

3. The isotopic cloud model (ICM)

The adiabatic model (AM) was extensively used in the past (Merlivat *et al.*, 1965; Macklin *et al.*, 1970; Jouzel *et al.*, 1975; Knight *et al.*, 1981). It was assumed that in an adiabatic ascent the water vapor condenses to form cloud droplets which are in isotopic equilibrium with the vapor at all heights. The isotopic content of the condensed phase can then be expressed by

$$\delta_c + 1 = \alpha(T) \cdot (\delta_v + 1), \quad (6)$$

where $\alpha(T)$ is the equilibrium fractionation factor whose numerical values were published for D and O^{18} by Majoube (1971) for liquid-vapor equilibrium.

The mixing ratios Q_v and Q_c of vapor and cloud water in the ascending parcel are determined from a thermodynamic chart, so that $\delta_c(z)$ is obtained analytically as

$$\delta_c(z) + 1 = \frac{(\delta_0 + 1)Q_{v0}}{Q_c(z) + \frac{Q_v(z)}{\alpha(T)}}, \quad (7)$$

provided δ_0 is known. The isotope content of each hailstone layer is then considered to correspond to δ_c , so that its growth temperature (height) can be derived unequivocally.

In the ICM, the equation of conservation of the isotopes in the parcel is written similarly to Eq. (2), replacing the mixing ratios and the fallout terms by primed symbols, i.e.,

$$Q' = jQC_{SMOW}(\delta + 1), \quad (8)$$

$$F' = jFC_{SMOW}(\delta + 1), \quad (9)$$

with Q' and δ having indices v, c, r, i, g and e and F' , r and g. The coefficient j is the ratio of the molecular weights [$j = 2(19/18)$ for HDO/H₂O and $j = 20/18$ for H₂O¹⁸/H₂O¹⁶] and is introduced by the definition of $C = N_D/N_H$ (or $N_{O^{18}}/N_{O^{16}}$).

The equations for the rate of change of Q' with height dQ'/dz are formed for each water species separately, using an equation of the type [similar to (3)]

$$\frac{dQ'}{dz} = \sum P'. \quad (10)$$

The terms on the right side are production/loss rates of isotopes given by

$$P' = jPC_{SMOW}(\delta + 1), \quad (11)$$

where the δ 's have to be evaluated individually for each P , depending on the physics involved in the process. The rates of change of Q'_i and Q'_c with height are treated differently, since equilibrium between these two phases is always established (see Sections 3d-3e).

To calculate the rate of change with height $d\delta/dz$ of the deviation of the isotope content from SMOW for the various water substances, the derivative of (8) with respect to height is formed, i.e.,

$$\frac{d\delta}{dz} = \frac{1}{Q} \left[\frac{1}{jC_{SMOW}} \frac{dQ'}{dz} - (\delta + 1) \frac{dQ}{dz} \right], \quad (12)$$

where dQ'/dz and dQ/dz are expressed in Eqs. (10) and (3), respectively. It can easily be verified that all the terms describing isotopic losses cancel when (10) and (3) are inserted into (12).

a. Isotope content of the cloud ice δ_i

Cloud ice is formed by deposition on ice nuclei and freezing of cloud water. In the deposition process fractionation occurs due to the higher saturation vapor pressure of H₂O¹⁶ compared with that of HDO and H₂O¹⁸, and the kinetic effects which are important in non-equilibrium situations (at relative humidities other than 100%). These kinetic effects reduce the isotopic fractionation in the case of supersaturation with respect to ice, due to the larger diffusivity of H₂O¹⁶ compared with those of HDO and H₂O¹⁸. To determine the isotope content of the ice formed by deposition on the surface of an ice crystal, Jouzel *et al.* (1980) derived a fractionation factor $\alpha_{ci} = \alpha_k \alpha_i$, which takes into account both kinetic and vapor pressure effects. Since at $T < -15^\circ\text{C}$, $0.97 \leq \alpha_k \leq 1$ for D and $0.994 \leq \alpha_k \leq 1$ for O¹⁸, the influence on the end results was negligible (see Section 4). Therefore, α_i as given by Jouzel *et al.* (1980)

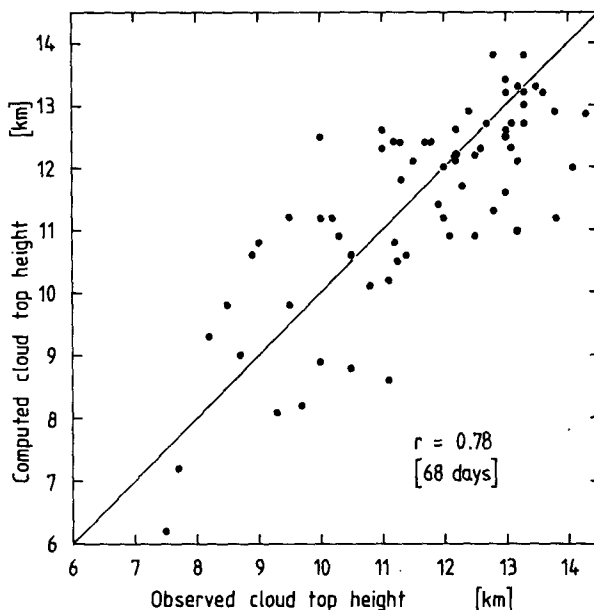


FIG. 1. Scatter diagram of computed versus observed cloud top heights for 68 days. The correlation coefficient is 0.78.

will be used in the treatment of the deposition process according to

$$\delta^{(s)} + 1 = \alpha_i(\delta_v + 1). \quad (13)$$

In ice crystal growth, only the newly deposited ice is subject to fractionation since there is no homogenization of the isotopes in ice. This is due to the low diffusivities d'_i of HDO and H₂O¹⁸ in ice. Kuhn and Thürkauf (1958) gave $d'_i = 10^{-14} \text{ m}^2 \text{ s}^{-1}$ at $T = -2^\circ\text{C}$, which corresponds to a diffusion distance $l = (2d'_i t)^{1/2} = 4.5 \text{ }\mu\text{m}$ for $t = 1000 \text{ s}$, which is ~ 300 times smaller than in water at $T = 0^\circ\text{C}$. In freezing of cloud droplets, given by P_{1ci} in (3), fractionation does not occur, since the freezing time of droplets is so short that isotopic exchange does not take place. The isotope content (δ_c) is therefore conserved during the transition to the ice phase. Considering the discussion above, dQ'_i/dz in (10) is formed and inserted, together with (3), (6) and (13), into (12) yielding

$$\begin{aligned} \frac{d\delta_i}{dz} = \frac{1}{Q_i} \{ & [\alpha_i(\delta_v + 1) - (\delta_i + 1)]P_{vi} \\ & - \mu(1 - \chi)[Q_v(\delta_v - \delta_i) - Q_c(\delta_c - \delta_i)] \\ & + [\alpha(\delta_v + 1) - (\delta_i + 1)]P_{1ci} \}, \quad (14) \end{aligned}$$

which gives the isotopic profile of ice crystals.

b. Isotope content of the rainwater δ_r

Autoconversion and coalescence do not influence the isotope content of the exchanged mass, which

remains δ_c . But there is continuous exchange between drop and water vapor through diffusion processes which modify the isotope content of drops even if this term is negligible in the rainwater mass balance. This isotopic exchange term due to diffusion, P'_{vr} , has been calculated considering the drop size spectrum and the fact that different drop sizes have different δ -values (see Appendix 2). In this treatment, the parameter n is introduced to describe the distribution of the δ 's as a function of r . The study of individual drops has shown that $0 \leq n < 1$. A sensitivity analysis of n was made over this range of values (Table 2). It is seen that the influence of n on the final result for δ_H is less than 1‰, although the variation of δ_r of the different drops in the spectrum amounts to ~6‰. Therefore in all the model runs n was set equal to zero, in which case $\delta_r \equiv \bar{\delta}_r$. Together with (3), (6) and (17), Eq. (10) is then inserted into (12), which leads to the formula

$$\frac{d\delta_r}{dz} = \{[\alpha(\delta_v + 1) - (\delta_r + 1)] \times (P_{1cr} + P_{2cr}) + P'_{vr}\} / Q_r, \quad (15)$$

for the rate of change of δ_r with height.

c. Isotope content of the ice particles δ_g

Ice particles are produced by accretion of cloud droplets, freezing and accretion of rainwater, and autoconversion and accretion of cloud ice. It is assumed that the freezing of cloud droplets in the accretion process is fast enough at cloud temperatures below -8°C , so that the graupel growth can be considered to be dry. The isotopic exchange described by Bailey *et al.* (1969) is therefore neglected, and it is assumed that no evaporation and no fractionation takes place during this process. The freezing time t_f of raindrops is much shorter than the isotopic relaxation time t_r . According to Murray and List (1972) and Stewart (1975) the ratio of t_f/t_r is < 0.05 . It is therefore assumed that the frozen raindrops keep the isotope content δ_r .

The accretion of rainwater by graupel particles can lead to formation of a water-ice mixture (soaking), but only in the case of porous graupel grown by accretion of cloud droplets or cloud ice and not in the case of frozen raindrops. Soaked graupel evidently exchange isotopes with the environment, as did the raindrops. The influence of these ice-water mixtures on δ_g has been neglected, however, since the production term P'_{2rg} proved to be three orders of magnitude smaller than those for the other graupel formation mechanisms.

During autoconversion and accretion of cloud ice by graupel, no phase change takes place. Therefore the isotope content of the ice crystals remains δ_i dur-

ing this process. As above, these effects were combined into Eq. (10) for graupel which is inserted into (12) together with (3) and (6) leading to the formula

$$\frac{d\delta_g}{dz} = \{[\alpha(\delta_v + 1) - (\delta_g + 1)]P_{2cg} + (\delta_r - \delta_g) \times (P_{1rg} + P_{2rg}) + (\delta_i - \delta_g)(P_{1ig} + P_{2ig})\} \frac{1}{Q_g}, \quad (16)$$

for the rate of change of δ_g with height.

d. Isotope content of the cloud water δ_c

The cloud water is treated as in the AM [Eq. (6)], since the relaxation time t_r for the exchange of isotopes with the vapor is ≤ 1 s at all temperatures and pressures of interest (Jouzel *et al.*, 1975). It is therefore of the same order of magnitude as the time step in the model ($\Delta t = \Delta z/w = 20 \text{ m}/20 \text{ m s}^{-1} = 1 \text{ s}$). Since vapor and cloud droplets are always in isotopic equilibrium, the rate of change with height of δ_c , $d\delta_c/dz$, is obtained from (6) simply by taking its derivative with respect to height, i.e.,

$$\frac{d\delta_c}{dz} = \alpha \frac{d\delta_v}{dz} + (\delta_v + 1) \frac{d\alpha}{dz}, \quad (17)$$

where $d\alpha/dz$ is calculated from the temperature dependence of α , and the known temperature gradient inside the model cloud, and $d\delta_v/dz$ is given below.

e. Isotope content of the water vapor δ_v

Water vapor is involved in isotopic exchanges with cloud water, rainwater and cloud ice (see above). It is therefore necessary to calculate δ_v from the isotopic balance equation [Eq. (2) with primed symbols, designated 2'], into which Eqs. (8)-(11) for Q_v , Q_r and Q_g are inserted. As mentioned above, dQ'_v/dz and dQ'_c/dz , between which equilibrium is established, are determined by using the derivative of (8) and inserting (3), (6) and (20). The resulting equations both contain the unknown $d\delta_v/dz$. Inserting them into (2') leads to

$$\frac{d\delta_v}{dz} = - \frac{\delta_v + 1}{Q_v + \alpha Q_c} \left\{ \frac{dQ_v}{dz} + \alpha P_{vc} + \alpha_i P_{vi} + Q_c \frac{d\alpha}{dz} - \chi \mu \left[Q_v(\alpha - 1) - Q_c \left(\alpha - \frac{\delta_c + 1}{\delta_v + 1} \right) \right] + \frac{P'_{vr}}{1 + \delta_v} \right\}, \quad (18)$$

which gives the rate of change of δ_v with height. This equation is a very general one, valid for any cloud model. Neglecting the ice phase ($P_{vi} = 0$), entrainment ($\mu = 0$) and isotopic exchange by diffusion ($P'_{vr} = 0$), Eq. (18) becomes identical to the equation proposed by Merlivat and Jouzel (1979) for isotopic

transfer on a global scale. If $Q_c = 0$, Eq. (18) reduces to the well-known Rayleigh distillation formula (all the condensate is immediately removed from the parcel). If all the water is retained in the parcel, $dQ_c + dQ_v = 0$ and integration of (18) leads to Eq. (7) used in the AM.

f. Isotope content of hailstones δ_H

To determine the isotope content of a hailstone inserted into a parcel of the cloud, simultaneous accretion of all the water substances is simulated.

Knowledge about collection efficiencies for the interactions treated in the model is generally incomplete. The usual procedure is to set $E_c = E_r = 1$ (English, 1973). A reasonable procedure for E_i would be to let it vary from zero to one, depending on whether the surface temperature is below -20°C or near 0°C . Due to the absence of information on this temperature (which is not calculated in the model), and its small effect in a very limited temperature interval E_i is set equal to unity. Another justification for this can be found in the relatively high frequency with which wet growth occurs (see Part II). We put E_g to zero, since ice particles are not likely to be collected by hailstones, although their presence is indispensable because they influence the isotope content of the other species. This is due to the fact that freezing of large drops to form ice particles, is one of the main removal processes for rainwater. Model runs with $E_g = 0.01$ to account for the small concentration of large ice crystals did not change the results. The isotope content δ_H at each height interval finally reads

$$\delta_H = \frac{1}{jC_{SMOW}} \left(\frac{E_c Q'_c + E_r Q'_r + E_i Q'_i + E_g Q'_g}{E_c Q_c + E_r Q_r + E_i Q_i + E_g Q_g} \right) - 1. \tag{19}$$

The model is initiated at cloud base by prescribing a δ_0 , determined after the procedures indicated in Part II for each day. This δ_0 gives the lowest point of the isotope profiles according to $(\delta_H + 1) = \alpha(\delta_0 + 1)$, using Eq. (8). This is equivalent to the result obtained with the AM. Above cloud base $\delta_H(z)$ cannot be calculated analytically, but is obtained step by step and depends on the development and interactions of the various water species. Note that at each step the isotope concentration of the entrained air δ_e also influences the various δ 's. The determination of the δ_e profiles which have to be prescribed, is discussed in Part II, Section 2.

δ_H is the isotope content of a hailstone layer grown in an environment as defined by the model without taking into account the surface conditions of the growing hailstones, which are not modeled in the ICM. This simple approach is a first step toward a

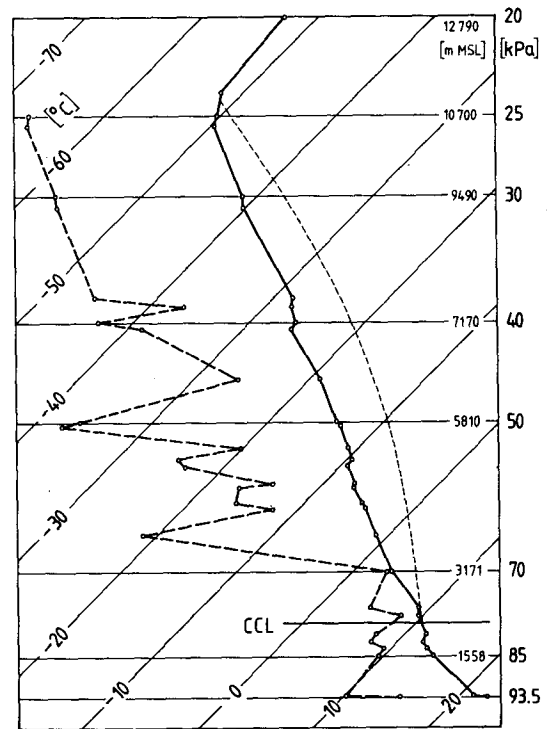


FIG. 2. Atmospheric sounding of 1340 GMT 14 July 1978 taken from Gormund (11 km from the radar site). Solid line is the temperature, thick dashed line the dew point, and thin dashed line the pseudo-adiabat through the convective condensation level (CCL) on a skew T - $\log p$ diagram.

more sophisticated model. Since such effects as wet and dry growth, soaking of rimed structures, shedding, etc. play an important role in the analysis of hailstone growth layers, their influence on the isotopic profiles should be checked by a model containing explicit microphysics.

The isotope profiles were calculated for all days discussed in Part II. As examples, the two soundings of 14 July 1978 and 6 August 1977 are depicted in Figs. 2 and 3 in skew T - $\log p$ diagrams. The 071478 sounding is very unstable and resembles the mean sounding obtained from the typical hail days described in Federer and Waldvogel (1978). The 080677 sounding, on the other hand, is only marginally unstable. The two days also differ in cloud base temperature and δ_0 , which both have a strong influence on the isotopic profiles. This fact is well known from the applications of the AM. In Fig. 4a the deuterium content of the five water substances are shown as a function of cloud temperature, based on the ICM. The O^{18} -profiles are almost identical to the D-profiles. A check of δD and δO^{18} values calculated in the model showed that they are linearly related over the entire range of formation of hailstones. The slope in the δD - δO^{18} diagram, which has a mean value of 8.5, is practically insensitive to the

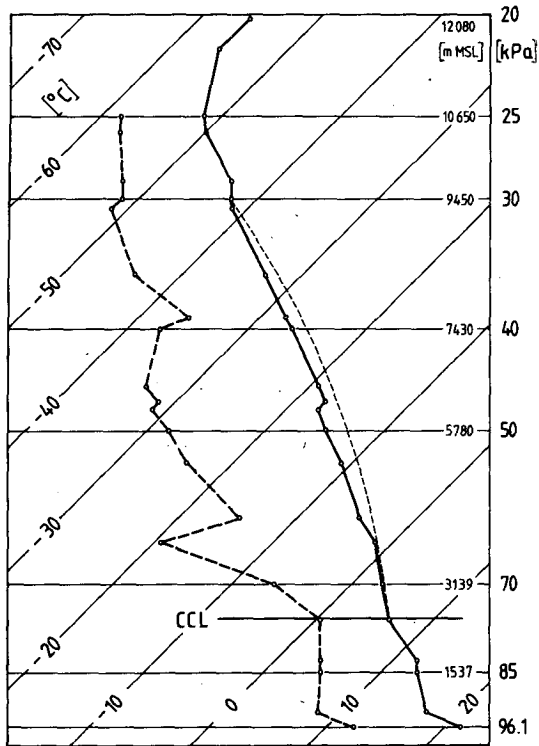


FIG. 3. As in Fig. 2, but for 1200 GMT 6 August 1977 at Payerne (80 km from the radar site).

choice of parameters in the model and independent of the storm considered. In Fig. 4b the δ_H profiles are compared to those based on the AM (δ_{AM}). For ease of comparison the same δ_0 has been used for both days.

It is seen that the differences between the δ_H and δ_{AM} profiles are appreciable, especially at lower temperatures. At high temperatures the isotope content of hail is 1–3‰ higher, but below the point of intersection of δ_H and δ_{AM} at around -17°C , it becomes considerably lower (up to 20‰ at -30°C). The relatively higher isotope content of hail with respect to cloud water is due to the interaction with rainwater, which is seen to be substantially enriched with regard to the other species (relaxation effect). Above the intersection, the difference between δ_H and both δ_c and δ_r becomes very large due to the increasing importance of the ice phase (δ_i). In the AM all the cloud droplets condensed are retained in the parcel, and since no freezing is taken into account, the vapor gradient δ_v is small. In the ICM, however, liquid particles which contribute to the isotopic exchange are frozen at an increasing rate at lower temperatures. The removal of particles which exchange isotopes leads to a considerable depletion of δ_v (ICM) with respect to δ_c (AM), indicated in the inset of Fig. 4b. In the limit of no isotopic exchange when all the liquid is frozen at low temperatures, a δ_v gradient

similar to the one in a Rayleigh process is obtained. Therefore, according to (6), the gradients of δ_c for the two models vary widely, resulting in the large impact on δ_H seen in Fig. 4b. Thus, by taking into account the various microphysical interactions which reduce the cloud water concentration in the ICM, the δ_H curve differs appreciably from the AM below about -15°C , and this effect is responsible for the smaller growth ranges of hailstones obtained.

4. Sensitivity analyses

The adjustable parameters in the numerical model are impossible to determine in each case, so that it is necessary to investigate their influence on the δ_H profile calculated from the soundings. Simple changes in the cloud parameterization are made to simulate processes which may alter the interactions among particles. In this way the effects of each parameter can be isolated and the important differences between the ICM and the AM discussed. In a first step a sensitivity analysis of the model itself was made. The changes of the mixing ratios of all the five water species upon variation of the influential parameters were calculated. As an example the variation of Q_r is given in Table 1. Second, the differences between the ICM and the AM are discussed. Since the δ_H profile is the most sensitive to the specification of the parameters, the analyses for the δ profiles of the other water substances are omitted.

A summary of the variation of δ_H (in ‰) is given in Table 2 for different parameters. The tables give the results for the two soundings in Figs. 2 and 3 at

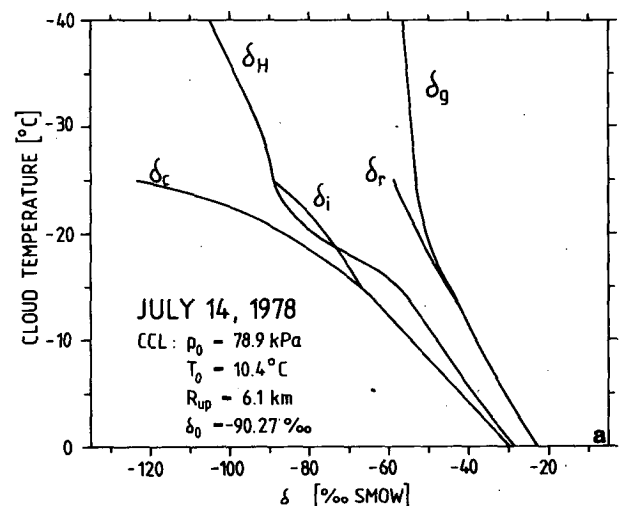


FIG. 4a. Deviation of the deuterium content from SMOW for cloud water (δ_c), cloud ice (δ_i), rainwater (δ_r), ice particles (δ_v) and hail (δ_H) as a function of cloud temperature calculated by the ICM. The sounding of Fig. 2 served as input and the adjustable parameters listed in Table 2 were used in the model.

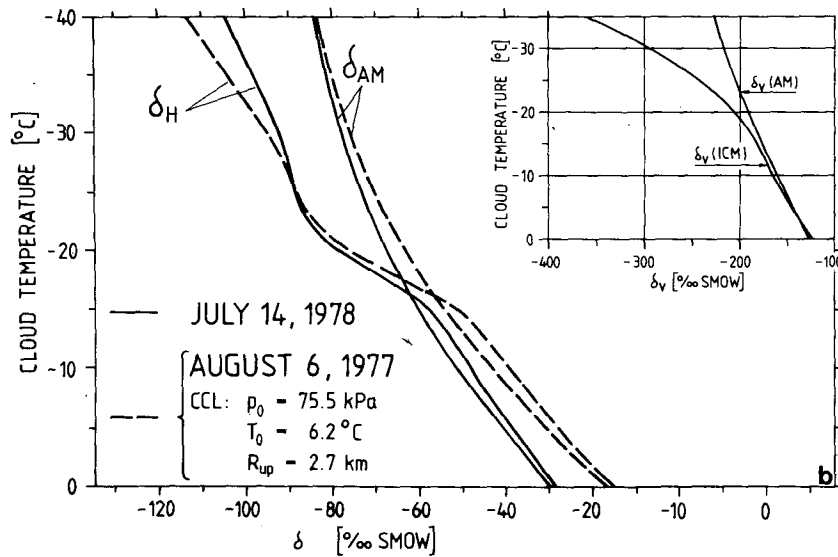


FIG. 4b. Comparison of the calculated deuterium profiles for hail based on the isotopic cloud model (δ_H) and on the adiabatic model (δ_{AM}) for the soundings of Figs. 2 and 3 and the parameters of Table 2. The curves for 080677 were calculated for the same $\delta_0 = -90.27\text{‰}$ valid for 071478. The inset shows the isotopic profiles of the vapor for both models on 14 July 1978.

four cloud temperatures. The cloud base parameters T_0 and δ_0 are not included in the sensitivity analysis, since they influence the δ_H profile in exactly the same way in the ICM as in the AM, i.e., a mere parallel shift of the δ curves to lower values with increasing T_0 and decreasing δ_0 (for a discussion of problems related to the δ_0 determination, see Part II).

The model was first run with a droplet spectrum of $N_c = 300 \text{ cm}^{-3}$ and a dispersion of $D_c = 0.2$ in the Berry (1968) autoconversion formulation. The resulting Q_r and δ_H profiles were compared with those obtained using $N_0 = 2000 \text{ cm}^{-3}$ and $D_c = 0.1$, a very continental spectrum. At $T_c = -20^\circ\text{C}$ for instance, the δ_H values increase by 9.0‰ on 071478 and even by 10.3‰ on 080677. The reason is that the narrow continental spectrum reduces the concentration of large drops in the model cloud to such an extent that the profile becomes similar to that in the adiabatic case (as it should be). Droplet spectra in individual Cb are generally unknown. In the extremely continental clouds of northeast Colorado, measured peak droplet concentrations are between 700 and 1300 cm^{-3} in Cu cong (Heymsfield *et al.*, 1978). For our hailclouds $N_c = 300 \text{ cm}^{-3}$ and $D_c = 0.2$ was chosen.

As expected, the mixing parameters a and $d\delta_e/dz$ also influence the Q_r and δ_H profiles. The mixing ratios change by as much as 30% upon variation of the entrainment constant, but in most cases the δ_H profiles are simply shifted toward slightly higher values. If a is varied from 0 to 0.2 the difference between the two resulting δ_H profiles increases with height on

071478 (Fig. 2), while it decreases on 080677 (Fig. 3). The latter is quite unexpected, but can be explained by the smaller R_{up} derived for 6 August [Eq. (5)] and by the larger difference between the vapor mixing ratios in cloud and environment for this day. This results in a considerably smaller rate of change of δ_v with height, because the last term of Eq. (18) is approximately four times larger on 6 August than on 14 July, while the other terms turned out to be approximately equal. Thus, entrainment increases the slope of the δ_H profiles in marginally unstable situations, while in very unstable situations with large updraft radii the slope is decreased. A mean entrainment constant of $a = 0.1$ was chosen for use in the model.

The assumed gradient of the δ_e (column 3) has a small influence on δ_H . Doubling $d\delta_e/dz$ decreases δ_H by only 1.3‰ in the less unstable sounding. In the model runs which form the basis of the discussion in Part II, the larger gradient of -25‰ km^{-1} was chosen, since in most cases precipitation was observed before the occurrence of the hailstorms from which the hail collections were obtained.

The vertical velocity impulse at cloud base w_0 , was varied from 2 to 8 m s^{-1} . Here again, the influence of the degree of instability is drastically demonstrated. In the unstable sounding, where high updraft speeds ($w_{max} = 42 \text{ m s}^{-1}$) are obtained, w_0 does not influence $w(z)$ to any extent. In the case of marginal instability, however, the value of w_0 is important for the development of the updraft profile. Since the con-

version rates are proportional to w^{-1} , a larger w initially leads to a reduced formation of large drops due to the reduced growth time (Table 1) and to an appreciable increase of Q_c (0.9 g kg^{-1} at -15°C). This results in a deviation of the δ_H profile of 6.6‰ at -25°C in the marginally unstable case, but has much less influence on the profiles from the sounding producing large updrafts anyway. Since in Section 2 the highest correlation was obtained with $w_0 = 5 \text{ m s}^{-1}$, this value was retained for all isotope calculations. The freezing function $f(T)$ has no influence on Q_r but has the expected large effect on the hail profile δ_H . The usual procedure in similar models is to convert linearly from all water at a temperature $T_f = -15^\circ\text{C}$ to all ice at -25°C , since measurements indicate that the LWC drops to zero below that temperature (Heymsfield *et al.*, 1978). If $T_f = -10^\circ\text{C}$ is chosen, the increase of the freezing interval from 10 to 15°C increases the isotope content of hailstones below about -18°C . At the end of the freezing interval, the increase of δ_H amounts to 6‰ for the marginally unstable sounding. The resulting increase of the δ_v and δ_c gradients is overcompensated by the formation of isotopically richer cloud ice. If the freezing interval is -20 to -40 , the deviation in δ_H are appreciable, but only below -28°C .

The conversion threshold A (column 6) in the autoconversion equation of cloud water to rainwater has only a small influence on the δ_H profile, if it is varied from 0.5 to 1.5 g kg^{-1} . This is observed despite a relatively large variation of Q_r (Table 1) and it results from the simple fact that the isotopic content of liquid remains practically unchanged by the transfer of cloud water to rainwater.

As mentioned previously, the constants A^* and B^* in the rainwater freezing function were varied from values corresponding to distilled water to those more representative for rainwater. This results in large changes of Q_r and δ_H , since A^* and B^* determine the onset and rate of freezing. Since no freezing experiments are available with water from our storms, the values for A^* and B^* were chosen as in the original version of the cloud model.

The microphysical effects specific to the isotopic exchange, mentioned in Section 3, namely, the isotopic fractionation at the surface of the ice crystals (α_{ci}) and the vapor exchange term P'_{vr} for raindrops have only a small influence on the profiles. As an

example, the variation of n in Eq. (A4) between 0 and 1 changes δ_H at all temperatures and for all storm days by less than 1‰. This justifies the choice of $N = 0$ in the model.

Table 2 shows that only if the highest values of the ranges indicated are used throughout, the slope of the δ_H curves in Fig. 4 increases, so that the profiles are similar to those of the AM. Only if an extremely continental droplet spectrum is assumed in the Berry conversion formula, can the slope increase to such an extent that the ICM curve lies close to the AM curve. Taking this extreme as basis of the calculations would essentially exclude the raindrops from consideration, which is contrary to observation.

5. Conclusion

A one-dimensional steady-state model (Hirsch, 1971) has been used to calculate profiles of the isotopic concentration of various water substances as a function of height. The differences with respect to the profiles obtained on the basis of the adiabatic assumption are substantial. This is mainly due to the presence of raindrops in the isotopic cloud model (ICM). This finding is in agreement with the estimates of Jouzel *et al.* (1975) that even a very thin layer (100 m) containing raindrops can change the deuterium profile drastically. If a reasonable concentration of raindrops is allowed in the model, a reasonable variation of all the other adjustable parameters does not change the results appreciably. This relative stability of the model with respect to parameters, which are difficult to measure, warrants its application to real data and to compare the results with those obtained from the AM (Part II). However, the sensitivity of the isotopic content of hailstones to the concentrations of large drops indicates the necessity of measuring their spectrum *in situ*. Finally, a more sophisticated time-dependent model would be needed to take into account some of the more subtle interactions, especially those leading to wet and dry hailstone growth, soaking of graupel and shedding of drops from growing hailstones.

Acknowledgments. The comments of Randy Koenig, WMO, Geneva and the very helpful review of Charles Knight and James Drake are gratefully acknowledged.

APPENDIX A

List of Symbols

Indices

a	air
c	cloud water
e	environment

- g graupel (ice particles)
 - i cloud ice
 - r rainwater
 - v water vapor
 - w bulk water
 - 0 value at cloud base
 - 1 means autoconversion or freezing in the production terms
 - 2 means coalescence or accretion in the production terms
- Primed symbols are used for the stable isotopes HDO or H₂O¹⁸.

Symbols

	<i>Units</i>	
<i>a</i>	mixing constant	
<i>A</i>	autoconversion threshold for rainwater	g kg ⁻¹
<i>A*</i>	constant in freezing equation for drops	0.66 (°C) ⁻¹
<i>B*</i>	constant in freezing equation for drops	10 ⁻¹⁰ m ⁻³ s ⁻¹
<i>C</i>	isotope content	ppm
<i>C_{SMOW}</i>	isotope content of Standard Mean Ocean Water (155.76 ppm for HDO and 2005.2 ppm for H ₂ O ¹⁸)	ppm
<i>d'</i>	diffusivity of HDO or H ₂ O ¹⁸ molecules in air	m ² s ⁻¹
<i>d'_i</i>	diffusivity of HDO or H ₂ O ¹⁸ molecules in H ₂ O ¹⁶ ice	m ² s ⁻¹
<i>D_c</i>	dispersion of a droplet spectrum	
<i>E</i>	collection efficiency	
<i>e</i>	vapor pressure	nM ⁻²
<i>f(T)</i>	freezing function	
<i>f'_v</i>	ventilation coefficient for HDO or H ₂ O ¹⁸ vapor diffusion in air	
<i>F</i>	fallout mixing ratio	g kg ⁻¹
<i>j</i>	coefficient used in the definition of the isotope content <i>C</i> [<i>j</i> = 2(19/18) for HDO and <i>j</i> = 20/18 for H ₂ O ¹⁸]	
<i>L_f, L_s</i>	latent heats of fusion and sublimation	J kg ⁻¹
<i>m</i>	drop mass	kg
<i>M</i>	parcel mass	kg
<i>n</i>	parameter in Eq. (A4)	
<i>N</i>	number of atoms	
<i>N_c</i>	number concentration of cloud droplets	cm ⁻³
<i>N₀</i>	intercept in the MP-distribution	m ⁻⁴
<i>P</i>	production terms for different water species	g kg ⁻¹ m ⁻¹
<i>Q</i>	mixing ratio	g kg ⁻¹
<i>Q*</i>	mixing ratio of the water frozen in one height step [<i>Q</i> = (<i>P</i> _{1ci} + <i>P</i> _{2cg} + <i>P</i> _{1rg} + <i>P</i> _{2rg})Δ <i>z</i>]	g kg ⁻¹
<i>r</i>	drop radius	m
<i>R_v</i>	specific gas constant for water vapor	461.5 J kg ⁻¹ K ⁻¹
<i>R_{up}</i>	updraft radius	km
<i>t_f</i>	freezing time of a raindrop	s
<i>t_r</i>	isotopic relaxation time	s
<i>T</i>	absolute temperature	K
<i>T_f</i>	freezing temperature	K
<i>v_t(<i>r</i>)</i>	terminal fall velocity of drops	m s ⁻¹
<i>w</i>	updraft velocity	m s ⁻¹
<i>z</i>	vertical coordinate	m
<i>α(T)</i>	fractionation factor at the interface liquid/vapor	
<i>α_i(T)</i>	equilibrium fractionation factor at the interface solid/vapor	
<i>α_{ci}(T)</i>	fractionation factor with kinetic effects [<i>α</i> _{ci} = <i>α_kα_i</i>]	
<i>δ</i>	deviation of the isotope content <i>C</i> from <i>C_{SMOW}</i> [<i>δ</i> = <i>C/C_{SMOW}</i> - 1]; also called isotope content	‰
<i>δ^(s)</i>	isotope content of the surface phase of ice particles	‰
<i>δ_H</i>	isotope content of a hailstone	‰

λ	slope of the MP-distribution	m^{-1}
μ	entrainment parameter [$=a/R_{up}$]	m^{-1}
ρ	density	$kg\ m^{-3}$
χ	weighting factor of Q_c and Q_i [$\chi = Q_c/(Q_c + Q_i)$]	
ϵ	departure of δ_r from isotopic equilibrium (A4)	
Γ	gamma function	

APPENDIX B

Isotope Content of a Drop Spectrum

The isotopic exchange term P'_{vr} due to diffusion of water vapor on drops of different sizes is calculated. For one drop the isotopic mass exchange is given by (Jouzel *et al.*, 1975)

$$\frac{1}{jC_{SMOW}} \frac{\partial m'}{\partial z} = \frac{4\pi r d' f'_v \rho_a Q_v}{w - v_r(r)} \left[(\delta_v + 1) - \frac{\delta_r + 1}{\alpha} \right]. \quad (A1)$$

For a drop distribution of the Marshall-Palmer (MP) type, (A1) can be written in terms of the mixing ratios as

$$\frac{1}{jC_{SMOW}} P'_{vr} = - \frac{8\pi d' Q_v}{\alpha(w - v_r)} \int_0^\infty \epsilon(r) N_0 r f'_v \exp(-2\lambda r) dr, \quad (A2)$$

where v_r is the mass-weighted mean velocity of the drops (Hirsch, 1971) and

$$\epsilon(r) = (\delta_r + 1) - \alpha(\delta_v + 1) \quad (A3)$$

is the difference between the actual δ -value of a drop of radius r and that corresponding to isotopic equilibrium.

The distribution of δ values for a population of drops is not computed in the present model, but it can be estimated by studying the δ_r changes with radius of individual drops taking into account both coalescence and diffusion. Since ϵ increases with r at a slow rate, it can be fairly well estimated by the general law (see Jouzel *et al.*, 1975)

$$\epsilon(r) = kr^n, \quad \text{with } 0 \leq n \leq 1, \quad (A4)$$

where k and n depend on the relative importance of the coalescence and diffusion processes. It appears reasonable to assume that the same distribution of $\epsilon(r)$ also holds for a distribution of drops at a given level of the model cloud.

For a MP distribution the coefficient k is related to the mean $\bar{\delta}_r$. Combining (A2), (A3) and (A4) we have

$$\begin{aligned} [(\bar{\delta}_r + 1) - \alpha(\delta_v + 1)] \int_0^\infty r^3 \exp(-2\lambda r) dr \\ = \int_0^\infty kr^{3+n} \exp(-2\lambda r) dr. \quad (A5) \end{aligned}$$

Integration leads to

$$k = 6[(\bar{\delta}_r + 1) - \alpha(\delta_v + 1)](2\lambda)^n / \Gamma(n + 4), \quad (A6)$$

where Γ is the gamma function.

Inserting $f'_v(r) = 1 + 1.16(rv_r/2\pi d')^{1/2}$ with $v_r = 184r^{1/2}$ and $\lambda^4 = (N_0\pi\rho_w/Q_r\rho_a)$ (Kinzer and Gunn, 1951; Kessler, 1969), (A2) becomes

$$\begin{aligned} \frac{1}{jC_{SMOW}} P'_{vr} = - \frac{12d'Q_v}{(w - v_r)\alpha} \left(\frac{\pi N_0 Q_r \rho_a}{\rho_w} \right)^{1/2} \\ \times \frac{(\bar{\delta}_r + 1) - \alpha(\delta_v + 1)}{\Gamma(n + 4)} \times \left[\Gamma(n + 2) \right. \\ \left. + 3.732\Gamma(n + 2.75) \left(\frac{Q_r \rho_a}{N_0 \pi \rho_w} \right)^{3/16} (d')^{-1/2} \right], \quad (A7) \end{aligned}$$

which allows P'_{vr} to be calculated as a function of Q_v , Q_r , ρ_a , w , $\bar{\delta}_r$ and δ_v .

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