A stochastic model of ice particle multiplication by drop splintering

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SUMMARY

A stochastic model is developed which describes the increase in ice particle concentrations resulting from the splintering of supercooled drops on freezing. Analysis of the experimental evidence suggests that significant splintering occurs at least for drops with diameters in the range 50 to 200 μm. At time \( t = 0 \) one such drop freezes and ejects \( r \) ice splinters. Each of these splinters may be captured by another water drop in the same size range at any subsequent time, which in turn will freeze and eject a further \( r \) splinters, and so on. Consideration of the probability that a particular capture will take place at a particular time yields an expression for \( m(t) \), the estimated number of particles existing at time \( t \),

\[
m(t) = (r^2 e^{(r-1)t}) - 1)(r - 1),
\]

where \( r \) is the mean lifetime of an ice splinter. It is stressed that by considering the probability of a collision occurring at any time after the birth of a splinter, far higher estimates of the population are obtained than from a non-stochastic model.

Calculations show that the largest ice particle multiplication factors measured by Mossop et al. (1972), of order \( 10^3 \), can be produced in the available time if the value of \( r \) for the splintering size range is 5 or 6. Analysis of the experimental evidence suggests that such a value for \( r \) is possible.

1. Introduction

There now exists a substantial body of evidence (reviewed by Mossop 1970, Mason 1971 and others) for the occurrence in some cumulus clouds of concentrations of ice crystals which exceed, by factors of up to about \( 10^4 \), the measured ice nucleus concentrations effective at the cloud top. Two particularly detailed studies have been reported recently by Mossop, Ruskin and Heffernan (1968) and Mossop, Cottis and Bartlett (1972). In the former case the enhancement factor was at least \( 10^3 \) in a long-lived cumulus cloud whose lowest temperature was \(-4^\circ C\); and in the latter case, where the clouds studied never had summit temperatures lower than \(-13^\circ C\), the ice crystal concentrations sometimes exceeded the ice nucleus concentration by between \( 10^2 \) and \( 10^4 \).

Mossop et al. (1972) reviewed a large number of possible explanations for the multiplication process which appears to exist in these clouds but concluded that none of them was adequate, with the possible exception of ice splinter production during rimming. They found that the multiplication factor was high when the cloud contained large concentrations (~1 per litre) of precipitation sized drops and rimed particles, probably formed originally by the freezing of some of the drops. Clearly, ice particle production during rimming will be particularly effective when substantial concentrations of large ice hydrometeors are present in the cloud. However, Brownscombe and Goldsmith (1972) and Mossop (private communication) have concluded from their recent detailed laboratory experiments that the rate of production of ice splinters during rimming is far too low to explain the high concentrations of ice crystals found in cumulus clouds. Mason (1973) has calculated that these rates are also much too low, even if it is postulated that the tops of the clouds studied by Mossop et al. (1972) had penetrated, at an earlier stage of their development, to considerably colder levels than those at which they were located when flown through.
In the present article a re-examination is presented of the possibility that the operative multiplication mechanism may be the ejection of ice splinters by precipitation sized supercooled drops during freezing, nucleation being initiated by the capture of an ice splinter. Thus the splinters from one freezing drop may cause several further drops to freeze and splinter, and in this way an avalanche process may be initiated. The cloud will become glaciated at a rate governed by $r$, the average number of splinters produced per freezing event, and $\tau$, the mean lifetime of an ice splinter before it is captured by a supercooled drop capable of ejecting splinters on freezing.

The particular feature of the present discussion is the recognition of the stochastic nature of the capture of ice splinters by the drops. A statistical pure birth process is assumed, i.e. the probability of a capture is proportional to the number of ice splinters present. The details of this stochastic process are given in Section 2, but we here demonstrate the inadequacy of a non-stochastic process. At time $t = 0$ a freezing drop is assumed to produce $r$ ice splinters. If each splinter is captured at the end of the mean lifetime $\tau$, then $r^2$ splinters are produced at time $\tau$ and $r^3$ at $2\tau$. At time $2\tau$ there will also be $1 + r + r^2$ residual frozen drops, giving a total population of $(r^4 - 1)/(r - 1)$ ice particles. A discussion of experimental evidence presented in Section 3 shows that the time available to produce the observed degree of multiplication in cumulus clouds is about $2\tau$ (40 to 50 min) and that, within the size range that exhibits appreciable splintering, several splinters may be ejected per freezing event. If we take $r = 5$ we see that this non-stochastic process has produced only 156 particles at time $2\tau$. To achieve $10^4$ in time $2\tau$, as reported by Mossop et al. (1972) a value of $r = 22$ is required, which is unrealistic. Alternatively, if we take the more reasonable splintering number, $r = 5$, the time required to produce $10^4$ ice particles is $5\tau$, which is inconsistent with the experimental evidence. This model is therefore seen to give an inadequate number of particles in the available time. However, as is shown later, the introduction of the stochastic element into these calculations produces estimates of numbers of ice particles which can be orders of magnitude greater than those just described.

2. A STOCHASTIC MODEL OF THE SPLINTERING PROCESS

In this Section a stochastic model is used to obtain the estimated number, $m(t)$, of ice particles existing at time $t$ as the result of the splintering of one freezing drop at time $t = 0$. This initial event may have resulted from a water drop capturing or containing a freezing nucleus. The model is a consequence of two assumptions, the validity of which are discussed in detail in Section 4.

The key assumption concerns the probability of capture of an ice splinter by a water drop in a specified time interval. Let $\lambda dt$ denote the probability that a particular ice splinter existing at time $t$ is captured in the time interval $t$ to $t + dt$ by a water drop capable of splintering. The assumption is that $\lambda$ is independent of the time. This is used by Jeans (1940) in the context of the kinetic theory of a gas to calculate the probability that a molecule has a free path of a given length. Following Jeans, let $f(t)$ denote the probability that an ice splinter formed at time $t = 0$ by the splintering of a drop has not been captured by time $t$. Then the probability that the splinter, having escaped capture at time $t$, will still have eluded capture at time $t + dt$ is

$$f(t + dt) = f(t)(1 - \lambda dt) . \quad . \quad . \quad . \quad . \quad (1)$$

giving

$$\frac{df}{dt} = -\lambda f . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$
The solution of this equation satisfying \( f(0) = 1 \) is

\[
f(t) = e^{-\lambda t},
\]

which yields a mean lifetime for the splinter of

\[
\tau = \int_0^\infty f(t) dt = \lambda^{-1}.
\]

In order for this key assumption to be valid it is necessary that the concentration of drops capable of splintering on nucleation is not significantly depleted by the splintering process. We also assume that the same \( \lambda \) applies to each splinter. From this it follows that the probability of the capture of an ice splinter is proportional to the number of ice splinters; this is an example of a pure birth process, as described in chapter seven of Karlin (1966). These secondary assumptions are also discussed in Section 4.

The second major assumption used in the development of this model is that at each capture of an ice splinter by a water drop \( r \) splinters are instantaneously ejected and that the residual frozen drop takes no further part in the splintering process. In the following Section experimental evidence is advanced to show that within a particular diameter range (50 ≤ \( d \) ≤ 200 \( \mu m \)) an average of several splinters may be ejected per freezing event. This second assumption requires that the capture of ice splinters by drops outside this diameter range, which may not eject splinters on freezing, does not significantly deplete the ice splinter population of the cloud. This point is fully discussed in Section 4. The capture of an ice splinter by a cloud droplet is assumed to produce no splinters and the combined particle is treated as a splinter. The time delay between nucleation of a drop and the ejection of splinters is only about \( 10^{-3} \tau \) and is ignored.

The production of splinters as dictated by the second assumption is an example of a continuous time branching process as described in chapter eleven of Karlin (1966). The integral equation description usually associated with branching processes may be avoided because of the particularly simple form of the probability density function given in Eq. (3).

We now determine the estimated number \( m(t) \) of ice particles existing at time \( t \). Let \( p_q(t) \) denote the probability that \( q \) successful collisions have taken place between ice splinters and water drops after time \( t \). As a consequence of each collision an incident ice splinter and a water drop are transformed into \( r \) ice splinters and one residual frozen drop. Thus after \( q \) collisions there are

\[
r + q(r - 1) = (q + 1)(r - 1) + 1
\]

ice splinters and \( (q + 1) \) frozen drops. The probability that \( q \) collisions have taken place at time \( t + dt \) is the sum of the probabilities

(i) that \( q \) collisions have taken place at time \( t \) and none of \( \{(q + 1)(r - 1) + 1\} \) splinters collide with drops in the time interval \( t \) to \( t + dt \), and

(ii) that \( (q - 1) \) collisions have taken place at time \( t \) and one of the existing \( \{q(r - 1) + 1\} \) splinters experiences a collision in time \( t \) to \( t + dt \). Thus, using the pure birth process embodied in the first assumption,

\[
p_{q}(t + dt) = p_{q}(t)(1 - \lambda dt)^{(q + 1)(r - 1) + 1} + p_{(q - 1)}(t)(q(r - 1) + 1)\lambda dt.
\]

Hence \( p_q(t) \) satisfies the differential difference equation

\[
\frac{dp_{q}(t)}{dt} + \lambda((q + 1)(r - 1) + 1)p_{q}(t) = \lambda(q(r - 1) + 1)p_{(q - 1)}(t).
\]

Before the first collision occurs, Eq. (3) applied to each of the \( r \) initial ice splinters gives

\[
p_{0}(t) = e^{-\lambda t},
\]
giving \( p_0(0) = 1 \) and hence \( p_q(0) = 0 \) for \( q = 1, 2, 3, \ldots \). The solution of Eq. (6) which satisfies \( p_q(0) = 0 \) for \( q \geq 1 \), is

\[
p_q(t) = e^{-\lambda t} \left( \frac{r}{r-1} \right) \left( \frac{r}{r-1} + 1 \right) \cdots \left( \frac{r}{r-1} + q - 1 \right) \left( 1 - e^{-\lambda(t-1)} \right) q! / q!.
\]

These probabilities are the coefficients of \( s^q \) in the expansion of the generating function

\[
G(s, t) = e^{-\lambda t} \left[ 1 - s(1 - e^{-\lambda(t-1)}) \right] \left( \frac{r}{r-1} \right)
\]

in positive powers of \( s \). The estimated number of collisions that have occurred by time \( t \) follows as

\[
\sum_{q=1}^{\infty} q p_q(t) = \left\{ \frac{\partial}{\partial s} G(s, t) \right\}_{s=1} = \frac{r}{(r-1)} \left( e^{\lambda(t-1)} - 1 \right).
\]

As there are initially \( r + 1 \) ice particles and each collision increases the total population by \( r \), the estimated number of ice particles at time \( t \) is

\[
m(t) = r + 1 + r \sum_{q=1}^{\infty} q p_q(t) = \frac{1}{(r-1)} \left( r^2 e^{\lambda(t-1)} - 1 \right).
\]

Using Eqs. (5) and (10) this estimated total number of particles may be separated into \( r e^{\lambda(t-1)} \) ice splinters and \((re^{\lambda(t-1)} - 1)/(r-1)\) residual frozen drops.

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Figure 1. The variation of \( m(t) \), the expected number of ice particles, with time \( t/\tau \) for three values of \( r \), as deduced from Eq. (11). The six individual points plotted correspond to the numbers predicted at \( t = \tau \) and \( t = 2\tau \) on the non-stochastic treatment given in Section (1) – the numbers labelling each point indicate the value of \( r \). Curve A, \( r=8 \); Curve B, \( r=5 \); Curve C, \( r=3 \).
Fig. 1 shows the variation of $m(t)$ with $t$, for various values of $r$, while Fig. 2 presents the predicted variation with $r$ of the time $t$ required to achieve selected values of $m(t)$. It is evident from these figures that the rate of increase of the number of ice particles is explosive over the ranges of $r$ and $t$ considered, and much greater than that given by a non-stochastic treatment of this problem. However, a full discussion of the numerical results illustrated in Figs. 1 and 2 is reserved for Section 4.

Figure 2. The time $t/\tau$ taken to achieve selected values of $m(t)$, the expected number of ice particles, as a function of $r$, according to Eq. (11). Curve A, $m = 10^4$; Curve B, $m = 10^3$; Curve C, $m = 100$.

It is instructive to compare the behaviour of this model for small values of $t$ with the non-stochastic model in which the initial $r$ ice splinters produce $r^2$ splinters at the mean lifetime $\tau = 1/\lambda$. Eq. (11) gives for the stochastic model

$$\frac{dm}{dt} = \lambda r^2 e^{-2t(r-1)},$$

which even at time $t = 0$ has a growth rate ensuring equality with the non-stochastic model at time $\tau$. The exponential factor in $dm/dt$ ensures significantly more ice particles from the stochastic model at time $\tau$, and at larger values of the time this effect is considerably more pronounced.

3. VALUES OF $r$ AND $\tau$

Despite the large number of experiments which have been conducted into the freezing and fragmentation of supercooled drops there exists no conclusive quantitative picture of the splintering behaviour over the entire ranges of drop-sizes and temperature $T$ of interest. However, if we neglect the early work in which, generally, insufficient care had been taken to eliminate the meteorologically spurious effects of dissolved CO$_2$, non-representative heat flow and the absence of thermal and solution equilibrium it is possible to make certain definitive statements. The carefully conducted experiments of Kuhns (1966) and Hobbs
and Alkezweeny (1968) agree in demonstrating that drops of diameter $d$ less than 50 $\mu$m will not eject secondary particles on freezing. Also, even though it was not possible accurately to simulate the effects of free-fall on the symmetry of the heat transfer and freezing, the comprehensive series of experiments by Johnson and Hallett (1968) suggest strongly that supercooled drops of diameter greater than about 1 mm will not shatter in conditions similar to those occurring in clouds. They concluded that a primary condition for drop fracture on freezing is the establishment of a strong shell of ice around the drop, which is governed by the spherical symmetry of the heat transfer to the environment. Since, for these large drops, ventilation produces highly asymmetric heat transfer, shell formation and associated shattering or splintering, will not occur.

However, within the intermediate range of sizes $50 \mu m < d < 1$ mm there exists good evidence for the ejection of ice splinters during freezing in conditions of meteorological relevance, although precise values of the ranges of $d$ and $T$ which produce appreciable splintering and the associated values of $r$ remain to be established. The major difficulty in determining accurate values of $r$ lies in detecting the very small ice splinters which may be produced on freezing. If the air is subsaturated with respect to ice, as has been the case in most experiments, the splinters are likely to evaporate prior to collection. This point is exemplified by the experiments of Hobbs and Alkezweeny, which constitute perhaps the most thorough investigation of the freezing of drops within this intermediate size range. They studied the freezing behaviour of falling drops in the diameter range 20 to 150 $\mu$m at temperatures $T = -8^\circ$C and $-32 < T < -20^\circ$C, and established that at least for $d < 100$ $\mu$m the important meteorological conditions were accurately reproduced. They report that drops of all sizes within the range 50 to 150 $\mu$m were observed to shatter during freezing. Unfortunately, in their experiments they were able to view a drop only over a small fraction (about 10%) of its fall, in which they observed that about 5% of the frozen or freezing drops shattered, so they could not determine precisely what fraction of freezing drops ejected ice particles; the above facts would suggest, however, that a substantial proportion did so. They found that 'when the air in the chamber was subsaturated with respect to ice, only a few pieces of ice thrown from a fragmenting droplet could be discerned on the screen. However, on several occasions a tray of supercooled sugar solution was placed inside the chamber to one side of and just below the point at which the droplets were freezing. The growth of many ice crystals in the sugar solution indicated that more ice particles were being ejected by the freezing drops than could be observed visually. This conclusion was confirmed by raising the humidity in the chamber until the air was saturated with respect to water. Under these conditions many ice particles could be seen in the vicinity of a droplet which fragmented during freezing.'

Several other workers have performed recent experiments on the freezing of drops in this intermediate size range. Brownscombe and Thorndike (1968a) found that 9% of the falling drops in the diameter range 100 to 180 $\mu$m and 6% in the range 160 to 240 $\mu$m shattered on freezing at a temperature of $-5^\circ$C. In a separate experiment (1968b) little splintering accompanied the freezing of drops of similar sizes and temperatures. Takahashi and Yamashita (1970) found that 17% of drops in the diameter range 75 to 175 $\mu$m falling in thermal equilibrium with their environment shattered on freezing at $-6^\circ$C, but they had no provision, in their experiments, for detecting the presence of any ice splinters produced. Pitter and Pruppacher (1973) studied the freezing of water drops of diameter $400 < d < 700$ $\mu$m suspended in the UCLA wind tunnel at temperatures between 0 and $-20^\circ$C and relative humidities ranging from 60% to ice saturation. They found that the moment ice nucleation began the freezing particle experienced an abrupt decrease in its terminal velocity - this could, of course, be due solely to evaporative loss - and often began to drift horizontally or to move in a circle describing a rather erratic trajectory. Photographs of the frozen
drops often displayed protuberances and irregularities indicative of the bursting of an ice shell; unfortunately, however, the humidities were too low to permit any ejected splinters to be seen. Finally, Gay (private communication, to be published) has made a detailed study of the freezing of drops in the diameter range 60 to 150 μm levitated electrically at temperatures between 0 and −20°C in an environment which is slightly subsaturated with respect to ice. These drops were in good thermal and solution equilibrium with their environment. He finds that many drops recoil suddenly from their equilibrium positions during the later stages of their freezing, showing that their charge-to-mass ratio has changed and thereby suggesting that ice has been ejected; they also exhibit a recoil immediately after nucleation, but this can be attributed to evaporative loss of mass. Again, the humidity was too low to render visible any ejected material.

It is impossible to draw firm quantitative conclusions from the experiments reviewed above but it appears reasonable to suggest, particularly in view of the experiments of Hobbs and Alkezweeny, that an average of several ice splinters may be ejected during the freezing of supercooled drops in the diameter range from about 50 μm to perhaps a few hundred microns at temperatures around −5°C.

It is also difficult to estimate precisely the rate at which drops which eject ice splinters on freezing sweep out volume within a cloud; and thereby to calculate τ, the mean lifetime of a splinter before it is captured by such a drop. For example, in the very thorough study of ice crystal concentrations in cumulus and strato-cumulus clouds conducted by Mossop et al. (1972) the instrument used to detect precipitation particles — an aluminium foil impactor — did not give impressions for particle diameters below 250 μm, and the only information on smaller precipitation particles was obtained from occasional use of the Squires cloud droplet gun, which recorded some drops of diameter up to about 100 μm. Even for particles of d > 250 μm the concentrations determined by the foil impactor must be considered to be very uncertain because of its small sampling area; the average horizontal spacing of the hydrometeors detected was several metres. Nevertheless, by interpolating between the limits of the devices used by Mossop et al. (1972) on the basis of size-distributions determined by other workers for similar situations, and also by studying the sampler records obtained by Mossop et al. (1968) for a cumulus cloud which also possessed substantial concentrations of ice particles at temperatures close to 0°C it is possible to determine some reasonably accurate values of τ, as is shown below.

If d_{min} and d_{max} are the lower and upper limits of the range of diameters of drops which exhibit significant splintering on freezing, then the precipitation water content per unit volume of cloud comprised of drops within this range is given by the equation

\[ C = \frac{\pi \rho_w}{6} \sum_{d_{\text{min}}}^{d_{\text{max}}} d^3 N(d), \]  

where \( \rho_w \) is the density of water and \( N(d) \) the concentration of drops of diameter \( d \). The probability \( \frac{dt}{\tau} \) of capture of an ice splinter in time interval \( dt \) is estimated by assuming that

(i) the ice splinters have negligible fall velocities
(ii) a splinter occupies negligible volume and will be captured if it lies within the direct path of a drop. In time \( dt \) a drop of diameter \( d \) and fall velocity \( U(d) \) sweeps out a volume

\[ \frac{4}{3} \pi d^2 U(d) dt. \]  

Hence for a concentration \( N(d) \) the fractional volume swept out in \( dt \) is

\[ \frac{4}{3} \pi \sum_{d_{\text{min}}}^{d_{\text{max}}} d^2 N(d) U(d) dt = \frac{dt}{\tau}. \]
Hence
\[
\frac{4}{\pi} = \sum_{d_{\text{min}}}^{d_{\text{max}}} d^2 U(d) N(d) = \sum_{d_{\text{min}}}^{d_{\text{max}}} d^3 N(d) \times U(d)/d.
\]  
(16)

Values of \(\tau\) applicable to clouds which exhibit ice-particle multiplication can therefore be calculated from Eq. (16) and the values of drop-sizes and concentrations determined by Mossop et al. (1968, 1972) and others. In general, however, there exists insufficient information from which to construct a detailed size distribution of drops within the diameter range \(d_{\text{min}} \to d_{\text{max}}\) and the calculation of \(\tau\) can be simplified, with reasonable accuracy, by noting that the ratio \(U(d)/d\) appearing in Eq. (16) is fairly insensitive to \(d\) over most of this size range. We may therefore use an average value \(\overline{U(d)/d}\) for this ratio. In this situation Eqs. (13) and (16) can be combined to give
\[
\tau = \frac{2\rho_w}{3C} \times \frac{\overline{d}}{\overline{(Ud)}}.
\]  
(17)

If we take \(\overline{d} = 0.15\) m, \(\overline{(Ud)} = 0.47\) m s\(^{-1}\) and \(\rho_w = 10^6\) gm m\(^{-3}\) we see from Eq. (17) that
\[
\tau \approx \frac{200}{C},
\]  
(18)

where \(\tau\) is measured in seconds and \(C\) in gm m\(^{-3}\). It is therefore necessary, in order to calculate \(\tau\), only to know the precipitation water content within the splintering range. Analysis of the records of Mossop et al. (1968) shows that in the regions of the cloud containing high concentrations of drops – which is where he finds appreciable multiplication to occur – the values of \(C\) were in the region of 0.1 gm/m\(^3\); no very large drops were reported in this study, suggesting that virtually all the precipitation drops collected lay within the diameter range of about 60 to 200 \(\mu\)m. In this case Eq. (18) predicts a value of \(\tau\) of about 33 minutes. It is difficult to determine precise values of \(\tau\) from the more detailed measurements of Mossop et al. (1972) because precipitation drops of \(d < 250\ \mu\)m were generally not detected, but if we make the assumption that such drops contributed about 0.1 gm/m\(^3\) to the total, (this figure agrees quite well with estimates based on the Squires’ gun measurements, flights F5, F15, F17 of 3 June 1970), the calculated values of \(\tau\), if we assume that drops of diameter above 500 \(\mu\)m will not splinter on freezing, are typically in the range 20 to 26 min; if we discard all drops of diameter greater than 250 \(\mu\)m our estimate of \(C = 0.1\) gm/m\(^3\) gives a value of \(\tau\) around 28 minutes. The time available to produce the highest degree of multiplication (\(10^4\)) reported by Mossop et al. (1972) can be estimated, from the time variations of their sampling records, the known rates of development of cumulus clouds and the times which must have been necessary for ice crystals to grow from the vapour at about \(-5\)°C to produce the largest crystals shown in their photographs to be in the region of 30 to 50 minutes. It may be assumed, therefore, that a period of about \(1.5\tau\) to \(2\tau\) is available to produce the large multiplication factors recorded.

4. Results of the Stochastic Calculations

Eq. (11) has been solved for various values of \(r\) to provide, in Table 1, the expected numbers of ice particles (frozen drops plus splinters), \(m(t)\), which have resulted at selected times \(t/\tau\) from the freezing of one drop at time \(t = 0\) and the subsequent stochastic multiplication process already described. The concentrations of drops capable of ejecting splinters on freezing are many orders of magnitude greater than the measured concentrations of freezing nuclei in clouds which exhibit ice particle multiplication. It seems reasonable
TABLE 1. THE MULTIPLICATION FACTOR $f$ EXISTING AT VARIOUS TIMES $t/\tau$ FOR SELECTED VALUES OF $r$, ACCORDING TO Eq. (11)

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<th>$r = 4$</th>
<th>$r = 5$</th>
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</tbody>
</table>

Therefore, to assume that the majority of nuclei will quickly be activated on reaching their temperature threshold. We may consequently assume that $m(t)$, the expected number of ice particles at time $t$, is approximately equal numerically to the multiplication factor $f$ determined by Mossop et al. (1968; 1972) and others, and in this Section we use formula (11) for $m(t)$ to predict $f$.

The rate of increase of the numbers of ice particles is seen from the Table 1 and Fig. 1 to be explosive, particularly for the larger values of $r$. After one mean lifetime ($t/\tau = 1$) the values of the multiplication factor $f$ are seen to be 33, 341 and $10^4$ for $r$ values of 3, 5 and 8 respectively; the corresponding values of $f$ for the non-stochastic treatment, outlined in Section 1 are 13, 31 and 73. We see, therefore, that the introduction of the stochastic element into this multiplicative process has increased the predicted number of ice particles existing at $t = \tau$ by factors of about 2.5, 11 and 137 respectively; for $t/\tau = 2.0$ these ratios are much larger, having values of about 6.2 for $r = 3$, 25 for $r = 4$ and 119 for $r = 5$. Table 2 gives more detailed values of this ratio.

TABLE 2. VALUES OF THE RATIO OF THE MULTIPLICATION FACTOR PREDICTED BY THE STOCHASTIC TREATMENT TO THAT BY THE NON-STOCHASTIC TREATMENT, FOR VARIOUS VALUES OF $r$ AT TIMES $t/\tau$ AND $2t/\tau$

<table>
<thead>
<tr>
<th>$r$</th>
<th>$t/\tau = 1.0$</th>
<th>$t/\tau = 2.0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>2.5</td>
<td>6.2</td>
</tr>
<tr>
<td>4</td>
<td>5.1</td>
<td>25</td>
</tr>
<tr>
<td>5</td>
<td>11</td>
<td>119</td>
</tr>
<tr>
<td>6</td>
<td>25</td>
<td>614</td>
</tr>
<tr>
<td>8</td>
<td>137</td>
<td>1.39 x 10^4</td>
</tr>
</tbody>
</table>

In order to assess whether this process might be responsible for the ice particle concentrations reported by Mossop et al. (1968; 1972) and other workers, it is illustrative to calculate, using Eq. (11), the times $t/\tau$ taken to achieve particular values of $f$ for selected values of $r$. We see from Fig. 2 and Table 3 that a multiplication factor of $10^2$ is easily achieved in less than one lifetime by values of $r$ in excess of about 4; $10^3$ can be attained in less than one lifetime by values of $r$ greater than about 6 and in less than two lifetimes by $r$ greater than about 4; while values of $10^4$, the highest degree of multiplication reported by Mossop et al. (1972) takes less than two lifetimes for $r = 5$. Consequently, it appears that since the time available to produce the highest concentrations of ice particles may be about 1.5$\tau$
TABLE 3. \textit{The time $t/\tau$ taken to achieve selected values of the multiplication factor $f$ for various values of $r$, according to Eq. (11)}

<table>
<thead>
<tr>
<th>$r$</th>
<th>$f = 100$</th>
<th>$f = 10^3$</th>
<th>$f = 10^4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>3.21</td>
<td>5.52</td>
<td>7.82</td>
</tr>
<tr>
<td>3</td>
<td>1.55</td>
<td>2.70</td>
<td>3.85</td>
</tr>
<tr>
<td>4</td>
<td>0.98</td>
<td>1.75</td>
<td>2.49</td>
</tr>
<tr>
<td>5</td>
<td>0.69</td>
<td>1.26</td>
<td>1.84</td>
</tr>
<tr>
<td>6</td>
<td>0.53</td>
<td>0.99</td>
<td>1.45</td>
</tr>
<tr>
<td>7</td>
<td>0.42</td>
<td>0.80</td>
<td>1.19</td>
</tr>
<tr>
<td>8</td>
<td>0.34</td>
<td>0.67</td>
<td>1.04</td>
</tr>
<tr>
<td>9</td>
<td>0.29</td>
<td>0.57</td>
<td>0.86</td>
</tr>
<tr>
<td>10</td>
<td>0.24</td>
<td>0.50</td>
<td>0.76</td>
</tr>
</tbody>
</table>

To $2\tau$, as discussed in Section 3, this stochastic process of drop splintering can explain all the observations on ice multiplication in cumulus clouds if small raindrops in the diameter range of about 50 to 200 $\mu$m produce an average of not less than about 5 or 6 ice splinters on freezing. The earlier discussion of relevant experiments, particularly those of Hobbs and Alkezweeny, suggests that this requirement is quite possible, although not definitively proven.

It is now necessary to consider in more detail some of the assumptions on which the stochastic model is based. The assumption that $\tau$ is independent of time is reasonably valid in a simple treatment if the concentration of splinter-producing drops that have frozen is much less than that of those that have not. To a first approximation we may assume that the concentration of frozen drops is equal to the observed concentration of ice particles divided by the average number of splinters produced per freezing event. In both studies referenced by Mossop \textit{et al.} (1968; 1972) ice particle concentrations of up to 100 per litre were observed, so that with $r = 5$ the concentration of frozen drops would be about 20 per litre. This figure is several times less than the concentrations of small raindrops ($50 < d < 200$ $\mu$m) measured by Mossop \textit{et al.} (1968) in an extensive traverse through a region containing large amounts of precipitation water. It appears therefore that our assumption of insignificant depletion of drops which can splinter on freezing is reasonably accurate throughout the period of ice multiplication.

We now consider the assumption that drops which do not splinter on freezing do not significantly deplete the ice splinter population of the cloud. It is difficult to assess this point accurately because the drop diameter above which insignificant splintering occurs is not known. However, even if we assume that drops of diameter greater than 250 $\mu$m do not produce splinters we can see from the paper of Mossop \textit{et al.} (1972) that the concentrations of such drops, about one per litre, are about two orders of magnitude less than that of those that do splinter on freezing. These large drops will, however, be nucleated by ice splinters relatively quickly because of their high sweepout rates, but once frozen are unlikely to collect more than a fairly small fraction of the splinters which they encounter subsequently. This problem may be further alleviated by noting that the freezing of large drops needs to produce only one splinter per event to maintain the rate of multiplication; the experiments of Pitter and Pruppacher, mentioned previously, suggest that this is quite likely, at least for drops of diameter below 700 $\mu$m. Indeed, it is quite possible that interactions between large frozen drops and smaller drops within the splintering size range may also produce ice particles. Overall, therefore, it appears unlikely that any appreciable slowing down of the rate of production of ice splinters will result from the capture of some of them by large drops.

The assumption that growth of the hydrometeors, which will be particularly rapid for the ice particles, will not affect the value of $\tau$ is clearly an over-simplification. In fact, the
photographs of Mossop et al. (1968) show that some crystals apparently grown almost completely from the vapour can achieve dimensions greater than those of the drops which are considered to produce splinters on freezing. However, almost all of the ice particles collected were substantially smaller than the drops and in view of the explosive nature of the multiplication process considered a large proportion of the splinters existing at any time during the process of glaciation will have been produced only slightly earlier. This assumption, although crude, appears therefore to be reasonable. The growth of drops, which will influence the value of $\tau$ by changing their sweepout rate and moving drops into and out of the splintering size-range as time progresses, is neglected in the present treatment. We assume that the drop sizes and concentrations abstracted from the experiments are representative values over the time span.

Finally, we discuss the assumption that the probability $\lambda dt$ that an ice splinter is captured by a drop in a time-interval $dt$ is independent of time and is the same for each splinter. This assumption strictly requires a random distribution of all particles and will be weak if the $r$ ice splinters ejected from a freezing drop spend a significant fraction of their time preferentially localized near to their parent or each other. It is easily shown that, because of the substantial velocities and high concentrations of the drops which may produce splinters, the time for which the $r$ splinters remain in the proximity of their parent is a small fraction of a second. The rate of dispersion of the splinters from each other will be a somewhat slower process because of the lower velocities involved, but bearing in mind that with $N \sim 100 \text{ l}^{-1}$ the mean spacing of the splinter-producing drops is only about 20 mm so that only a few seconds may be expected to elapse before the splinters, which are probably ejected with a wide range of velocities, sizes and directions, have drops rather than other splinters as their nearest neighbours. Since the mean lifetime of a splinter is around 20 or 30 min this localization effect is unlikely to be of significance.

It therefore appears that, despite the many simplifications made in this treatment, the stochastic calculations of the rate of increase of ice particle concentrations may be considered to be reasonably accurate.

5. Discussion

The foregoing analysis demonstrates that the introduction of the stochastic element into a consideration of ice particle multiplication through drop splintering greatly reduces the required average value of $r$ for those drops which produce secondary ice particles on freezing. To underline this point we note that on the stochastic model the values of $r$ required to produce a multiplication factor $f = 10^4$ in times $\tau$, $2\tau$ and $3\tau$ are about 8-0, 4-7 and 3-5 respectively; the corresponding values in the non-stochastic case are about 100, 22 and 10. Since, in calculating values of $\tau$ from field observations, we have considered only that range of drop-sizes which is known to exhibit splinter production; and in view of the experimental results on drop freezing, described earlier, it appears quite possible that the observed multiplication factors are a consequence of splinter ejection from freezing drops.

This suggestion fits in well with the observation, described in both papers by Mossop et al. (1968; 1972) that significant multiplication occurs only when there exist within the cloud significant concentrations of drops of precipitation dimensions. In their earlier paper they find large concentrations of drops ($\sim 100 \text{ l}^{-1}$) in the approximate diameter range 50 to 150 $\mu$m but few larger ones. This observation is consistent with our suggestion that such drops – present in concentrations adequate to explain the observed multiplication – are of primary importance in the production of ice splinters. It is also possibly relevant to mention a point raised by Pitter and Pruppacher (1973). They report that Weickmann,
Katz and Steele (1970) concluded from laboratory investigations, and Auer (1971) from field observations in cap clouds, that a large proportion of hexagonal shaped ice crystals contained in the crystal centre a small frozen drop which consequently must have frozen as a single crystal of ice. These results are consistent with the freezing experiments of Pruppacher and Pitter, who suggest on the basis of this work that glaciation in atmospheric clouds may often result from the freezing of supercooled drops. The observation by Mossop et al. (1968; 1972) and other workers that rimed particles were present in clouds which exhibit ice particle enhancement and absent or rare in those which do not is also consistent with the drop-splintering hypothesis, since many splinters will have time to grow by diffusion to a sufficiently large size to commence effective collection of supercooled droplets; also, frozen drops will rapidly become rimed. The observation by Mossop et al. (1972) that shallow stratocumulus clouds, with similar top temperatures to the cumuli, exhibit a very small degree of ice particle enhancement – 10 being the upper limit for \( f \) – is also consistent with the drop splintering mechanism. In these clouds the concentrations of drops of diameter greater than 50 \( \mu m \) are very low so there will probably be a sufficient number of freezing nuclei available to nucleate them. In this case each drop will produce \( 1 + r \) ice particles – giving a multiplication factor of somewhat less than 10, according to our earlier discussion – and no more splintering will occur.

The stochastic model described in Section 2 may be capable of considerable extension, to take account of (i) the capture of splinters by larger drops which do not splinter on freezing and (ii) changes with time of the value of \( \tau \); it can be shown that the value of \( m(t) \) is unaffected by the distribution of \( r \). However, it would probably be premature to undertake such a detailed study until more specific information is available concerning the numbers and sizes of ice splinters produced by the freezing of drops over a wide range of conditions; and more detailed measurements exist of the particle size distributions within clouds which exhibit this multiplication phenomenon. At present, it may be stated that drop splintering – when considered stochastically – appears to provide a possible explanation for the reported high concentrations of ice crystals in cumulus clouds.


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