Three Models for the Coalescence Growth of Cloud Drops

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ABSTRACT

In an attempt to resolve the confusion over the "stochastic completeness" of the stochastic coalescence equation, an analysis is made of an idealized cloud consisting of a number of large "drops" falling through very many small, equal-size "droplets" with a constant drop-droplet collection kernel. It is shown how three superficially equivalent physical interpretations of the drop-droplet collection kernel lead to three quite different models for the growth of the drops. The implications of and relationships between the models are drawn out in detail. The stochastic completeness controversy is apparently a consequence of a failure to distinguish clearly between two of these three conceptual approaches.

1. Introduction

A widely held view in cloud physics is that there are two models which can be used to mathematically describe cloud drop growth by coalescence—the *continuous* model and the *stochastic* model (Mason, 1971). The continuous model is simpler, but it yields a uniform, relatively slow rate of growth of raindrops which is uncharacteristic of real clouds. The more sophisticated stochastic model predicts a non-uniform growth of the cloud drops with a more rapid formation of a few large raindrops, in closer agreement with observations.

The stochastic model was introduced into cloud physics by Telford (1955). The heart of this approach is the stochastic coalescence equation (SCE),[1] which purports to describe the time evolution of the drop-size distribution. During the past few years the "stochastic completeness" of the SCE has been the subject of some discussion. The cause for concern on this point has been well expressed by Chin and Neiburger (1972), who point out that in the integration of the SCE "no randomness has generally been introduced either through the initial conditions or through the collection rate kernel. In the formulation of the collision kernel, the probability of collision between droplets of different sizes has been invoked. But once this is done, the collection process is allowed to proceed according to this mean rate . . . [with] no fluctuation of this process [being] possible." Chin and Neiburger concluded that the well-defined drop-size spectrum which is predicted by the SCE probably ought to be interpreted as some sort of "expected" spectrum.

In a previous paper (Gillespie, 1972), this writer gave an analysis of cloud drop growth by coalescence which *does* allow for randomness, both in the initial conditions and in the collection process.[2] It was shown there that, *if* certain correlations can be neglected, then the SCE is indeed the time evolution equation for the mean or expected drop-size spectrum; in addition, the random fluctuations about this mean were accounted for quantitatively. In the present paper we shall argue that the approach taken in that previous paper can be regarded as a *third* model for coalescence growth, which we shall call the "pure stochastic" model, and that what is currently thought of as the stochastic model might be more aptly termed the "quasi-stochastic" model. By making a detailed analysis of the continuous, the quasi-stochastic and the pure stochastic models as applied to a highly idealized cloud, we shall try to clarify the role played by the SCE in the description of a coagulating aerosol.

2. An idealized "drop-droplet" cloud

We consider an idealized cloud of constant volume V, which at time $t=0$ consists entirely of L "drops" each having mass m_0 and Γ "droplets" each having mass μ. The drops are supposed to be much larger than the droplets, but very much less numerous:

$$m_0 > \mu, \quad L \ll \Gamma.$$

In this cloud any drop and any droplet may coalesce, but we suppose that drop-drop and droplet-droplet coalescences do not occur. In a drop-droplet coalescence the drop "collects" the droplet, the droplet disappears,

[1] This equation is also called by various writers the stochastic collection equation, the coagulation equation, the scalar transport equation and the kinetic equation.

[2] Another rigorous treatment of this problem, from a slightly different point of view, was given earlier by Marcus (1968).

and the mass of the drop increases by μ. Thus, the masses of the drops are increasing, but the number of drops L remains constant. The masses μ of the droplets remain constant, and although the number of droplets is decreasing due to their collection by the drops, *we suppose the droplets to be so numerous that this decrease is negligible*, i.e., the number of droplets Γ remains essentially constant.

Our object is to describe the growth of the L drops. In order to do this it is first necessary to specify in detail the process whereby a drop collects a droplet. Mathematically this is done through the "collection kernel" $C(m)$, which, loosely speaking, describes the droplet collecting ability of a drop of mass m. Now, the type of model we get for the growth of the cloud drops depends upon precisely *how* $C(m)$ describes this collecting ability. More specifically, the type of model we get depends upon what *physical interpretation* we choose to give to the quantity $C(m)\Gamma dt$, where dt is an infinitesimal interval of time. As will be shown momentarily, three possible interpretations are:

(i) *Continuous model*

$$C(m)\Gamma dt = \text{number of droplets that any drop of mass } m \text{ will collect in time } dt \tag{1}$$

(ii) *Quasi-stochastic model*

$$C(m)\Gamma dt = \text{fraction of the drops of mass } m \text{ which will collect a droplet in time } dt \tag{2}$$

(iii) *Pure stochastic model*

$$C(m)\Gamma dt = \text{probability that any drop of mass } m \text{ will collect a droplet in time } dt. \tag{3}$$

Let us first show that it is indeed reasonable to use the *same quantity* $C(m)$ in all three contexts above. We assume that the L drops and Γ droplets are scattered randomly and uniformly throughout the cloud volume V. In time dt each drop of mass m will sweep out, relative to each droplet of mass μ, a "collection volume"

$$dV_{\text{col}} = [\pi(r_m + r_\mu)^2(v_m - v_\mu)dt]E(m,\mu) \tag{4}$$

in the sense that if the center of a droplet lies inside this collection volume that droplet will be collected by the drop in time dt. In (4), r_m and v_m denote the radius and terminal fall velocity of the drop, r_μ and v_μ the corresponding quantities for the droplet, and $E(m,\mu)$ is the "collection efficiency" which accounts for the fact that, owing to hydrodynamic and other effects, the collection volume will differ somewhat from the geometrical sweep-out volume. [The form of $E(m,\mu)$ is not relevant to our arguments here.] In terms of dV_{col}, our problem is now what to write down for each of the quantities on the right-hand sides of (1), (2) and (3) [which quantities we shall refer to as RHS(1), RHS(2) and RHS(3), respectively].

We first consider RHS(3). The probability that an m-drop will collect a droplet in time dt is evidently equal to the probability of finding a droplet inside the collection volume dV_{col} of the m-drop. Since the Γ droplets are distributed randomly and uniformly over the cloud volume V, then the probability that a *particular* one will be found inside dV_{col} is just dV_{col}/V. Since dV_{col} is infinitesimal the probability for finding two or more droplets inside dV_{col} is negligible, so the probability of finding *any* one of the Γ droplets inside dV_{col} is $\Gamma \times dV_{\text{col}}/V$. Thus,

$$\text{RHS}(3) = \Gamma dV_{\text{col}}/V. \tag{5}$$

Comparison with (4) and (3) yields the result

$$C(m) = \pi(r_m + r_\mu)^2(v_m - v_\mu)E(m,\mu)/V. \tag{6}$$

However, our main interest here is to show that we get the same result (5) for RHS(2) and RHS(1).

We now consider RHS(2). Since RHS(3) is the probability that any m-drop will collect a droplet in time dt, then if there are a total of N_m m-drops it follows that *approximately* $N_m \times \text{RHS}(3)$ of these will each collect a droplet in time dt. The approximate nature of this number is due to the fact that we cannot make a precise prediction because the droplets are distributed randomly throughout the cloud and the collections will be probabilistic.[3] Hence, the *fraction* of the N_m m-drops which will collect a droplet in time dt is given approximately by

$$\text{RHS}(2) \approx [N_m \times \text{RHS}(3)]/N_m = \text{RHS}(3), \tag{7a}$$

thus showing that it is reasonable to use the same quantity $C(m)$ in (2) as in (3).

Finally, to calculate RHS(1) we reason as follows. The number of droplets collected by a given m-drop in time dt will be equal to the number of droplets which lie inside the collection volume dV_{col} of the m-drop. This number will thus be equal to dV_{col} multiplied by the *local* droplet density. However, the local droplet density is a randomly fluctuating quantity, owing to the random positions of the droplets. The best we can do is to *approximate* the local volume density by the *average* droplet density, Γ/V. Thus, we obtain

$$\text{RHS}(1) \approx dV_{\text{col}}(\Gamma/V) = \text{RHS}(3), \tag{7b}$$

where the second equality is from (5). This shows that it is reasonable to use the same quantity $C(m)$ in (1) as in (3).

We have thus demonstrated that we are justified in using the same mathematical expression to represent

[3] For example, if the probability of obtaining heads in one coin toss is $\frac{1}{2}$, then ten tosses should yield *approximately* $10 \times \frac{1}{2} = 5$ heads; however, it is clearly *possible* to obtain anywhere from 0 to 10 heads in ten tosses.

the quantities RHS(1), RHS(2) and RHS(3). Notice, however, the *intrinsically approximate nature* of RHS(2) and RHS(1) in (7a) and (7b). This reflects the fact that, by choosing *not* to specify the positions of all the drops and droplets *exactly*, we are forced to specify these positions *probabilistically*, with the result that the collection process is properly described only in probabilistic terms. Of the three definitions (1), (2) and (3), it is clear that only (3) is framed in terms of a true probability. Therefore, to the extent that these three physical interpretations of the drop-droplet collection kernel lead to different results, the pure stochastic model should be the most realistic.

In the next three sections we shall examine the consequences of each of the models defined in (1)–(3) under the assumption that the collection kernel $C(m)$ is a constant:[4]

$$C(m)=C \quad \text{(a constant)}. \tag{8}$$

Although this simplifying assumption makes our drop-droplet cloud even more artificial, it will allow us to carry out the mathematical analysis of each model completely analytically without any approximations. We emphasize again that our concern in this paper is not so much to model a realistic cloud, but rather to compare and contrast three different approaches to cloud modeling.

3. The continuous model

In the continuous model it is implied by (1) that all drops having the same mass collect droplets at the same rate. Since all the drops start out with the same mass m_0, it follows that at any later instant every drop will have the same mass. Consequently, in the continuous model we may specify the *state* of the drops at time t by means of the function

$$M(t)\equiv\text{mass of any drop at time } t. \tag{9}$$

With assumption (8) it is easy to derive a formula for $M(t)$. Since (1) and (8) imply that each drop collects $C\Gamma dt$ droplets in time $(t, t+dt)$, and since the mass of each collected droplet is μ, then in time $(t, t+dt)$ the mass of each drop will increase by

$$dM(t)=\mu(C\Gamma dt).$$

Hence, the time evolution equation for $M(t)$ is

$$\frac{d}{dt}M(t)=\mu C\Gamma, \tag{10}$$

which is to be solved subject to the initial condition

$$M(0)=m_0. \tag{11}$$

Since μ, C and Γ are all constants the solution is trivial:

$$M(t)=m_0+\mu C\Gamma t. \tag{12}$$

Thus, the mass of each drop increases *continuously* (and in this case, linearly) with time.

4. The quasi-stochastic model

The implication in (1) that all the drops grow at the the same continuous rate is obviously unrealistic. Owing to the random positions of the drops and droplets, some drops of mass m will collect more droplets than other drops of mass m in the same period of time; furthermore, since only whole droplets are collected the masses of the growing drops will take on only the discrete values m_0, $m_0+\mu$, $m_0+2\mu$, ..., instead of the continuum of values implied by (12). The quasi-stochastic model (2) represents a first step toward taking these features of the coalescence process into account.

According to (2) only a certain *fraction* of the drops of mass m will collect a droplet in time dt. This implies that *drops having the same mass do not grow in unison*; consequently, in the quasi-stochastic model the definition of $M(t)$ in (8) makes no sense. However, we *can* legitimately define the function

$$N(m;t)\equiv\text{number of drops of mass } m \text{ at time } t, \tag{13a}$$

where

$$m=m_0, m_0+\mu, m_0+2\mu, \ldots. \tag{13b}$$

In progressing from the continuous model to the quasi-stochastic model, the function $N(m;t)$ replaces the function $M(t)$ as the quantity which describes the *state* of the drops at time t.

To find a formula for $N(m;t)$ we proceed as follows: According to (2) and (8), in time $(t, t+dt)$ exactly $N(m-\mu;t)\times C\Gamma dt$ drops of mass $m-\mu$ will each collect a droplet and so become drops of mass m, while exactly $N(m;t)\times C\Gamma dt$ drops of mass m will each collect a droplet and so become drops of mass $m+\mu$. Hence, the net increase in the number of drops of mass m in time $(t, t+dt)$ is[5]

$$dN(m;t)=N(m-\mu;t)C\Gamma dt-N(m;t)C\Gamma dt.$$

Therefore, the time-evolution equation for $N(m;t)$ is[6]

$$\frac{\partial}{\partial t}N(m;t)=C\Gamma[N(m-\mu;t)-N(m;t)], \tag{14}$$

which is to be solved subject to the prescribed initial condition

$$N(m;0)=\begin{cases}L, & \text{for } m=m_0\\ 0, & \text{for } m\neq m_0\end{cases} \tag{15}$$

[4] Our "drop-droplet" cloud is essentially the same as that considered by Telford (1955), except that Telford does not insist upon the simplifying assumption (8).

[5] The number of drops collecting two or more droplets in time $(t, t+dt)$ is of order >1 in dt, and hence need not be considered here.

[6] Eq. (14) is the same as Eq. (2) of Telford (1955) for a constant collection kernel.

Eq. (14) is precisely the SCE as it would be written for our simple drop-droplet cloud. Indeed, our derivation of (14) uses the same kind of reasoning as is employed in the conventional derivation of the general SCE. For a more realistic cloud, in which drops of all sizes interact via a size-dependent collection kernel, the SCE is a nonlinear integro-differential equation which is quite difficult to solve. However, for our drop-droplet cloud with a constant collection kernel, the SCE evidently takes the form of a system of coupled, first-order, linear differential equations [cf. Eq. (13b), and notice that $N(m_0-\mu;t)\equiv 0$ since m_0 is the minimum drop size]:

$$\frac{\partial}{\partial t}N(m_0;t)=-C\Gamma N(m_0;t),$$

$$\frac{\partial}{\partial t}N(m_0+\mu;t)=C\Gamma[N(m_0;t)-N(m_0+\mu;t)],$$

$$\frac{\partial}{\partial t}N(m_0+2\mu;t)=C\Gamma[N(m_0+\mu;t)-N(m_0+2\mu;t)],$$

etc.

Using the initial conditions (15), the first equation is easily solved, and its solution can then be used to obtain the solution to the second equation, which in turn can be used to solve the third equation, and so on. This procedure yields the result

$$N(m_0+k\mu;t)=\frac{L(C\Gamma t)^k\exp(-C\Gamma t)}{k!},\qquad k=0,1,2,\ldots, \quad (16)$$

which may easily be verified by direct substitution into (14) and (15).

Eq. (16) gives the number of drops of mass $m=m_0+k\mu$ in the cloud at time t, and we see that at any time $t>0$ there will be drops of many masses (i.e., a mass *spectrum*), in contrast to the prediction of the continuous model that all drops will have the *same* mass $M(t)$. Writing $x\equiv C\Gamma t$, we have for the total number of drops at time t

$$\sum_{m=m_0}^{\infty}N(m;t)=\sum_{k=0}^{\infty}\frac{Lx^ke^{-x}}{k!}=Le^{-x}\sum_{k=0}^{\infty}\frac{x^k}{k!}=Le^{-x}e^{x}=L$$

as expected.

In order to discover some of the important features of the mass spectrum $N(m;t)$, it is instructive to calculate its normalized power moments

$$M_j(t)\equiv\frac{1}{L}\sum_{m=m_0}^{\infty}m^jN(m;t),\quad j=1,2,\ldots. \quad (17)$$

Physically, $M_j(t)$ is just *the average (mass)^j of the drops at time t.* In particular (again abbreviating $x\equiv C\Gamma t$), we have for the average drop mass at time t

$$M_1(t)\equiv\frac{1}{L}\sum_{m=m_0}^{\infty}mN(m;t)=\frac{1}{L}\sum_{k=0}^{\infty}(m_0+k\mu)\frac{Lx^ke^{-x}}{k!}$$

$$=e^{-x}\left[m_0\sum_{k=0}^{\infty}\frac{x^k}{k!}+\mu x\sum_{k=1}^{\infty}\frac{x^{k-1}}{(k-1)!}\right]$$

$$=e^{-x}(m_0e^x+\mu xe^x)=m_0+\mu x,$$

so

$$M_1(t)=m_0+\mu C\Gamma t. \quad (18)$$

Comparing this result with (12) we see that $M(t)$ and $M_1(t)$ are *mathematically identical.* However, it is important to keep in mind that $M(t)$ and $M_1(t)$ represent *different physical quantities*: $M(t)$ is the mass of each and every drop at time t in the continuous model, whereas $M_1(t)$ is only the *average* drop mass at time t—i.e., the "center" of the graph of $N(m;t)$ vs m—in the quasi-stochastic model. $M(t)$ describes the instantaneous state of the drops completely in the continuous model, whereas $M_1(t)$ does *not* completely describe the instantaneous state of the drops in the quasi-stochastic model. For example, in addition to knowing the "center" of the mass spectrum, it is also important to know its "width." This width is given roughly by the rms deviation,

$$\Delta(t)=[M_2(t)-M_1^2(t)]^{\frac{1}{2}}. \quad (19)$$

We calculate $M_2(t)$ directly from (17) and (16) in essentially the same way that we calculated $M_1(t)$ above [7], and we find

$$\Delta(t)=\mu(C\Gamma t)^{\frac{1}{2}}. \quad (20)$$

Thus we see that, while the "center" of the graph of $N(m;t)$ vs m grows linearly with t, the "width" of this graph is proportional to $t^{\frac{1}{2}}$. The fact that $M_1(t)=M(t)$ tells us that all the drops *together* collect droplets at the same rate in the quasi-stochastic model as they do in the continuous model. However, the fact that $\Delta(t)$ increases with time tells us that the drops do not grow in unison in the quasi-stochastic model: some drops grow slower than predicted by the continuous model, while other drops grow faster.

5. The pure stochastic model

Although the quasi-stochastic model does not require a *given* m-drop to collect a definite number of droplets in a given time interval, as the continuous model does, it does require *all* m-drops *together* to collect a definite number of droplets in a given time interval. This feature is obviously inconsistent with the probabilistic nature of the collection process: The random locations of the drops and droplets should introduce stochastic

[7] The calculation of $M_2(t)$ is facilitated by writing $[k(k-1)+k]$ for k^2.

fluctuations in the number of droplets collected by any *group* of drops as well as by any individual drop. Such fluctuations will be permitted if we start with the purely probabilistic formulation (3).

One consequence of the pure stochastic collection process defined by (3) is that *we cannot predict exactly how many drops of a particular size there will be at any time $t>0$*; consequently, in the pure stochastic model the definition of $N(m;\,t)$ in (13a) makes no sense. However, (3) ought to allow us to predict the *probability* of finding a given number of drops of a particular size at time t. This suggests that an appropriate function to describe the instantaneous state of the drops might be

$$P(n,m;\,t)\equiv\text{probability that exactly } n \text{ drops have mass } m \text{ at time } t, \tag{21a}$$

where

$$n=0,\,1,\,2,\,\ldots,\,L, \tag{21b}$$

$$m=m_0,\,m_0+\mu,\,m_0+2\mu,\,\ldots. \tag{21c}$$

Just as we changed the state function of the drops from $M(t)$ to $N(m;\,t)$ in progressing from the continuous model to the quasi-stochastic model, so now we change the state function from $N(m;\,t)$ to $P(n,m;\,t)$ in progressing from the quasi-stochastic model to the pure stochastic model.

Under the assumption that the collection kernel is independent of m, we may derive a formula for $P(n,m;\,t)$ by the following line of reasoning. First, we calculate the auxiliary quantity

$$\mathcal{P}(k,t)\equiv\text{probability that any given drop will collect exactly } k \text{ droplets in time } t. \tag{22}$$

Since $C\Gamma dt$ is by (3) the probability that a given drop will collect one droplet in time dt, then for $k=0$ we have

$$\mathcal{P}(0,\,t+dt)=\mathcal{P}(0,t)[1-C\Gamma dt],$$

or

$$\frac{d\mathcal{P}(0,t)}{\mathcal{P}(0,t)}=-C\Gamma dt.$$

Integrating with the obvious initial condition $\mathcal{P}(0,0)=1$, we find

$$\mathcal{P}(0,t)=\exp(-C\Gamma t) \tag{23a}$$

Now, for any $k\geqslant 1$ the probability that a given drop will collect exactly k droplets in time $(0,t)$ can be expressed as

$$\mathcal{P}(k,t)=\int_0^t \mathcal{P}(k-1,t')\times C\Gamma dt'\times\mathcal{P}(0,t-t'). \tag{23b}$$

For this is just the *product* of [the probability that the drop will collect exactly $k-1$ droplets in $(0,t')$] times [the probability that the drop will collect one more droplet in dt' at t'] times [the probability that the drop will collect no more droplets in (t',t)], *summed* over all t' from 0 to t. Eq. (23b) constitutes a recursion relation for $\mathcal{P}(k,t)$: With the formula for $\mathcal{P}(0,t)$ in (23a) we can calculate first $\mathcal{P}(1,t)$, and then $\mathcal{P}(2,t)$, and then $\mathcal{P}(3,t)$, and so on. Proceeding in this way, it is not difficult to show that

$$\mathcal{P}(k,t)=\frac{(C\Gamma t)^k\exp(-C\Gamma t)}{k!},\quad k=0,\,1,\,21\ldots. \tag{24}$$

Now, since each drop collects droplets *independently* of the other drops, the probability that a *particular arrangement* of n drops will collect exactly k droplets in time $(0,t)$ while the remaining $L-n$ drops will not is

$$\mathcal{P}^n(k,t)[1-\mathcal{P}(k,t)]^{L-n}.$$

But the number of distinct ways of arranging L drops into two groups of n drops and $L-n$ drops is

$$\frac{L!}{n!(L-n)!}.$$

The probability that exactly n of the L drops will collect exactly k droplets in $(0,t)$ is just the product of these last two quantities. Therefore,

$$P(n,\,m_0+k\mu;\,t)=\frac{L!}{n!(L-n)!}\mathcal{P}^n(k,t)[1-\mathcal{P}(k,t)]^{L-n},$$

$$n=0,\,1,\,\ldots,\,L;\ k=0,\,1,\,2,\,\ldots. \tag{25}$$

Eqs. (24) and (25) constitute a complete and exact solution to the pure stochastic model for our simple drop-droplet cloud. It is seen that, at any time $t>0$, it is possible for there to be anywhere from 0 drops to L drops of a given mass m, in contrast to the prediction of the quasi-stochastic model that there will be *exactly* $N(m;\,t)$ drops of mass m. Writing $g\equiv\mathcal{P}(k,t)$, we have for the probability of having *some* number $0\leqslant n\leqslant L$ of drops of mass m at time t

$$\sum_{n=0}^{L}P(n,m;\,t)=\sum_{n=0}^{L}\frac{L!}{n!(L-n)!}g^n(1-g)^{L-n}$$

$$=[g+(1-g)]^L=1,$$

as expected.

In order to discover some of the important features of $P(n,m;\,t)$, it is instructive to calculate its moments with respect to n:

$$N_j(m;\,t)\equiv\sum_{n=0}^{L}n^jP(n,m;\,t),\quad j=1,\,2,\,\ldots. \tag{26}$$

Physically, $N_j(m;\,t)$ is just *the average (number)^j of m-drops in the cloud at time t*. To understand precisely what we mean by "average" here, imagine making a series of very many experiments or "runs," in which the ith run consists of starting out with L drops of mass m_0 and letting the collection process proceed according to (3) until time t, at which time we count

the number of drops having mass m, $n_i(m,t)$. For fixed m and t the numbers $n_1(m,t)$, $n_2(m,t)$, ... will vary from run to run owing to the stochastic nature of the collection process. However, in the limit of very many runs we may expect that the average of the jth power of these numbers will be precisely the quantity $N_j(m;t)$ in (26). In particular, we have for the average number of drops found to have mass m at time t [again abbreviating $g \equiv \mathcal{P}(k,t)$]

$$N_1(m;t) \equiv \sum_{n=0}^{L} nP(n,m;t) = \sum_{n=1}^{L} n \frac{L!}{n!(L-n)!} g^n (1-g)^{L-n}$$

$$= Lg \sum_{n=1}^{L} \frac{(L-1)!}{(n-1)![(L-1)-(n-1)]!} \times g^{n-1}(1-g)^{(L-1)-(n-1)}$$

$$= Lg \sum_{n'=0}^{L-1} \frac{(L-1)!}{n'![(L-1)-n']!} g^{n'}(1-g)^{(L-1)-n'}$$

$$= Lg[g+(1-g)]^{L-1}$$

$$= Lg \equiv L\mathcal{P}(k,t),$$

or, with (24),

$$N_1(m_0+k\mu;t) = \frac{L(C\Gamma t)^k \exp(-C\Gamma t)}{k!}, \qquad k=0,1,2,\ldots. \quad (27)$$

Comparing this result with (16) we see that $N(m;t)$ and $N_1(m;t)$ are *mathematically identical*. However, it is important to keep in mind that $N(m;t)$ and $N_1(m;t)$ represent *different physical quantities*: $N(m;t)$ is the number of drops having mass m at time t in the quasi-stochastic model, whereas $N_1(m;t)$ is only the *average* number of drops having mass m at time t—i.e., the "center" of the graph of $P(n,m;t)$ vs n—in the pure stochastic model. $N(m;t)$ describes the instantaneous state of the drops completely in the quasi-stochastic model, whereas $N_1(m;t)$ does *not* completely describe the instantaneous state of the drops in the pure stochastic model. For example, in addition to knowing the "center" of the graph of $P(n,m;t)$ vs n, it is also important to know its "width." This width tells us by roughly how much we may expect the actual number of drops found to have mass m at time t in any particular experiment or run, to deviate from $N_1(m;t)$; in other words, this width estimates the uncertainty associated with $N_1(m;t)$. A convenient measure of this width is the rms deviation

$$\Delta(m;t) = [N_2(m;t) - N_1^2(m;t)]^{\frac{1}{2}}. \quad (28)$$

We calculate $N_2(m;t)$ directly from (26) and (25) in essentially the same way that we calculated $N_1(m;t)$ above[8], and we find

$$\Delta(m;t) = [N_1(m;t)]^{\frac{1}{2}}[1-N_1(m;t)/L]^{\frac{1}{2}}. \quad (29)$$

The second factor in (29) is slightly less than unity, and in fact approaches unity as $t \to \infty$, as may be proved from (27). Therefore, we conclude that the graph of $P(n,m;t)$ vs n is mainly located between $[N_1(m;t) - \sqrt{N_1(m;t)}]$ and $[N_1(m;t) + \sqrt{N_1(m;t)}]$, so we may "reasonably expect" the number of m-drops at time t found in any *particular* experiment or run to lie within this interval.

6. Summary and discussion

The mode of description of the coalescence growth of cloud drops depends upon the precise physical interpretation given to the collection kernel. We have demonstrated this by giving, for an idealized "drop-droplet" cloud, three possible and intuitively plausible interpretations [Eqs. (1)–(3)] and then tracing the consequences of these interpretations under the simplifying assumption that the collection kernel is constant.

For interpretation (1), the *continuous model*, the state of the drops was described by the function $M(t)$, which gives the mass of every drop at time t. The formula for $M(t)$ is given by Eq. (12).

For interpretation (2), the *quasi-stochastic model*, the state of the drops was described by the function $N(m;t)$, which gives the number of drops having mass m at time t. We can also calculate $M_j(t)$, the normalized jth moment of $N(m;t)$ with respect to m [see Eq. (17)]. In particular, $M_1(t)$ is the average drop mass at time t, and defines the "center" of the graph of $N(m;t)$ vs m; and $M_2(t)$ allows us to calculate the rms dispersion $\Delta(t)$, which defines the "width" of the graph of $N(m;t)$ vs m. Thus, if we allow the cloud to evolve from time 0 to time t according to (2), then we may reasonably expect that *the mass of a randomly chosen drop* at time t will lie somewhere between

$$M_1(t) - \Delta(t) \quad \text{and} \quad M_1(t) + \Delta(t).$$

The formulas for $N(m;t)$, $M_1(t)$ and $\Delta(t)$ are given by Eqs. (16), (18) and (20), respectively.

For interpretation (3), the *pure stochastic model*, the state of the drops was described by the function $P(n,m;t)$, which gives the probability that exactly n drops will have mass m at time t. We can also calculate $N_j(m;t)$, the jth moment of $P(n,m;t)$ with respect to n [see (26)]. In particular, $N_1(m;t)$ is the average number of drops having mass m at time t, and defines the "center" of the graph of $P(n,m;t)$ vs n; and $N_2(m;t)$ allows us to calculate the rms dispersion $\Delta(m;t)$, which defines the "width" of the graph of $P(n,m;t)$ vs n. Thus, if we allow the cloud to evolve from time 0 to time t according to (3), then we may reasonably expect that *the number of drops having mass* m at time t will lie

[8] The calculation of $N_2(m;t)$ is facilitated by writing $[n(n-1)+n]$ for n^2.

somewhere between

$$N_1(m;t)-\Delta(m;t) \quad \text{and} \quad N_1(m;t)+\Delta(m;t).$$

The formula for $P(n,m;t)$ is given by Eqs. (24) and (25); the formulas for $N_1(m;t)$ and $\Delta(m;t)$ are given by (27) and (29), resepectively.

It is found that $M_1(t)$ is mathematically the same as $M(t)$. Therefore, if we measure the masses of many randomly selected drops at time t, the continuous model predicts that *every* drop will have mass $M(t)$, whereas the quasi-stochastic model predicts that the masses of the drops will be scattered over an interval of $\sim 2\Delta(t)$ about $M(t)$. From the point of view of the quasi-stochastic model, the continuous model is thus deficient in that it ignores fluctuations in the drop mass.

Similarly, it is found that $N_1(m;t)$ is mathematically the same as $N(m;t)$. Therefore, if we make a series of repeated "runs" from time 0 to time t, the quasi-stochastic model predicts that every run will produce $N(m;t)$ drops of mass m, whereas the pure stochastic model predicts that the numbers of drops of mass m found in these runs will be scattered over an inverval of $\sim 2\Delta(m;t)$ about $N(m;t)$. From the point of view of the pure stochastic model, the quasi-stochastic model is thus deficient in that it ignores fluctuations in the drop mass spectrum.

The stochastic coalescence equation (SCE) determines the function $N(m;t)$ in the quasi-stochastic model. If this were *all* one could say, then one could *not* assert that the SCE is "stochastically complete." However, according to the pure stochastic model *we may reasonably expect to find roughly between*

$$N(m;t)-[N(m;t)]^{\frac{1}{2}} \quad \text{and} \quad N(m;t)+[N(m;t)]^{\frac{1}{2}} \tag{30}$$

drops of mass m in the cloud at time t, where $N(m;t)$ is the solution to the SCE. So it turns out that the SCE allows us to calculate not only the mean mass spectrum, but also the approximate size of the stochastic fluctuations which may be expected to occur in the mass spectrum about this mean. Therefore, the SCE is more "stochastically complete" than its usual derivation via the quasi-stochastic interpretation would suggest.

Common derivations of the SCE are usually rather confused mixtures of the quasi- and pure stochastic approaches. Some derivations start out with a pure stochastic definition of the collection kernel along the lines of (3), then tacitly (but erroneously) assume that this is equivalent to the quasi-stochastic definition (2), and so proceed with a derivation equivalent to that presented in Section 4. Frequently, in an attempt to make this assumption of equivalence more plausible, $N(m;t)$ is initially defined to be the *mean* mass spectrum instead of the mass spectrum; in that case, an argument along the lines of Section 4 is not really a derivation at all, but merely a plausibility argument which begs the whole issue. Another confusing point is that the quantity $f(m;t)\equiv N(m;t)/V$ (where V is the total cloud volume) is usually introduced at the outset, and is referred to as "the average mass spectrum per unit volume"; in this case, however, "average" refers to variations in the number of drops contained in different specific unit volumes at a given instant, and *not* to the fluctions caused by the stochastic nature of the collection process which have been the subject of our discussion here.

In conclusion, we should comment on the extent to which these results for the simple "drop-droplet" cloud carry over to a more realistic cloud, in which drops of all sizes interact with each other via a size-dependent collection kernel. In a previous paper (Gillespie, 1972) we presented an analysis of such a cloud from the viewpoint of the pure stochastic model. We showed there that *if* (i) certain correlations can be neglected, and *if* (ii) coalescences between droplets of exactly the same size do not occur, then the standard SCE indeed determines the mean drop-size spectrum $N_1(m;t)$; moreover, the probability function $P(n,m;t)$ tends to the Poisson form

$$\lim_{t\to\infty} P(n,m;t)=\frac{N_1^n(m;t)\exp[-N_1(m;t)]}{n!}, \tag{31}$$

and in particular $\Delta(m;t)$ approaches the value $\sqrt{N_1(m;t)}$. Therefore, *subject to conditions (i) and (ii), statement* (30) *is valid for the general aerosol coalescence process.*

On the other hand, if one does not impose the simplifying conditions (i) and (ii), the situation is not at all clear. This is mainly due to the fact that the "state" of the aerosol in the pure stochastic model cannot be specified completely by the probability $P(n,m;t)$ *alone*; one also needs the "conditional probabilities"

$P^{(1)}(n,m|n',m';t)$
$\equiv$ probability of having n drops of mass m at time t, given that there are n' drops of mass m', (32a)

$P^{(2)}(n,m|n',m';n'',m'';t)$
$\equiv$ probability of having n drops of mass m at time t, given that there are n' drops of mass m' and n'' drops of mass m," (32b)

etc.

For our drop-droplet cloud, $P^{(1)}(n,m|n',m';t)$ would be given by (25) with L replaced by $L-n'$, and $P^{(2)}(n,m|n',m';n'',m'';t)$ by (25) with L replaced by $L-n'-n''$. But as yet no way has been found to calculate the complete set of functions P, $P^{(1)}$, $P^{(2)}$, ... for a general aerosol. Indeed, the "no-correlation approximation" (i) mentioned above simply amounts to approximating the conditional probabilities in (32) by

$P(n,m;t)$. Just how much this approximation invalidates (30) is an important question. A significant step toward answering this question has been taken recently by Bayewitz *et al.* (1974), who have succeeded in solving the pure stochastic problem for a full range of drop sizes with a size-independent collection kernel. Their results indicate that correlations produce significant deviations from the SCE spectrum when the aerosol population is either small or poorly mixed. The conjecture of Bayewitz *et al.* that these correlation effects would probably be accentuated for a more realistic size-dependent collection kernel seems to mark the limit of our present understanding of this problem.

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