FLUCTUATIONS AND LUCK IN DROPLET GROWTH BY COALESCENCE

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Simple theoretical arguments quantify the "luck" required for warm rain initiation, regardless of whether that luck comes from giant nuclei or occasional "fast eddies."

In a recent article in this magazine (Stevens et al. 2003) describing a major field campaign devoted to the study of stratocumulus clouds, the authors noted:

> One of the remarkable impressions left on the investigators was how the apparent uniformity of the cloud top viewed from above could mask enormous variations in the microphysical structure within the cloud layer.

How can such "enormous variations" arise from seemingly identical mean-field thermodynamic conditions? For example, why does one such stratocumulus cloud drizzle while another one does not? Part of the answer, we think, is in pronounced fluctuations (deviations from the mean), and methods of statistical physics may be helpful. In particular, the initiation of drizzle or rain is reminiscent of colliding gas molecules insofar as to form a 1-mm raindrop in a warm (no ice) cloud requires that a million 10-μm droplets coalesce. The case for fluctuations in cloud physics is, of course, not new. In fact, the notion of statistically fortunate droplets (ones that grow much faster than average) is an important part of the current understanding as indicated by many texts in cloud physics (e.g., Houghton 1985, 272–277; Mason 1971, 145–155; Pruppacher and Klett 1997, section 15.3; Rogers and Yau 1989, 134–136; Wallace and Hobbs 1977, 172–181; Young 1993, 180–185). Yet, a quantitative theory of warm rain initiation remains elusive. For example, Blyth et al. (2003) write

> A significant part of precipitation that falls in the Tropics is warm rain formed by coalescence of cloud droplets. Despite 50 years of research on this topic we still do not possess a quantitative understanding of the production of warm rain.

Indeed, most results are not easy to interpret, even in the simplest collector drop scenario of one drop falling through a cloud of identical, smaller droplets, used in Telford's (1955) classic treatment. All of the

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above-mentioned textbooks resort to computer simulations when examining the role of fluctuations. For example, Rogers and Yau (1989) refer to Robertson (1974) who used a Monte Carlo simulation to calculate the number of collisions required before the continuous coalescence equation can be used without serious errors. Because of this almost exclusive reliance on computers, statements made in many cloud physics texts concerning fluctuations are often obscure. Consider, for example, the following statement from a recent cloud physics text (Young 1993, p. 185) in the section on pure stochastic versus quasi-stochastic models (collection refers to collision and coalescence):

> This represents an average drop concentration of 1000 drops per m^3. What is the expected range in the concentration of drops of this size due to the stochastic nature of the process? . . . 1000 ± 10^{-3} drops m^{-3}. It may be concluded that the differences due to the stochastic nature of the collection processes are negligible and that the cloud behavior may be adequately described by the quasi-stochastic treatment.

Natural clouds do not, of course, have each cubic meter containing exactly a thousand droplets (with radius within 48–50 μm) to within 10^{-3} droplets.[1] Such confusion about droplet number fluctuations is vexing and may be caused, in part, by the rather complicated formalism that surrounds stochastic coalescence (even the terminology is obscure: stochastic, pure stochastic, quasi stochastic, discrete versus continuous collection, collection versus coalescence, etc.). This has convinced us to adopt, at least as a first step, the simpler collector drop scenario in favor of the Smoluchowski integro-differential equation approach. The latter has been the primary focus of research on droplet coalescence in clouds since the 1960s but it does not provide simple solutions for realistic coalescence rates (kernels) (Drake 1972). Thus, we agree with Cotton and Anthes (1989, p. 90), who wrote

> However, for complicated cloud or mesoscale models, there remains a strong desire to develop simplified techniques for predicting the evolution of the droplet spectrum to form rain along with its sedimentation through the cloud.

This paper represents our attempt to gain physical insight into the role of fluctuations in droplet growth by coalescence. In order to develop simple closed-form expressions allowing such insight, throughout the paper we seek clarity of the final approximate expressions, sometimes at the expense of details. In particular, we wish to separate effects of fluctuations in droplet growth from those of the average droplet growth. Recall from the opening paragraph that the typical fraction of large drops required to initiate warm rain is about 10^{-6} of all cloud droplets. Therefore, from the outset we focus on the one-in-a-million fraction of fastest growing (henceforth called lucky) droplets, and ask about their growth time rather than the average droplet growth time. Indeed, in the long run the latter is irrelevant because the remaining droplets are collected by the lucky ones anyway. Our task, then, requires clean separation of fluctuation effects from the mean growth of the collector drop. To that end, we begin with the probability distribution of intercoalescence times.

DISTRIBUTION OF TIMES TO COALESCENCE. Consider a statistically homogeneous cloud containing droplets of the same radius (r). The droplets are assumed to be perfectly randomly distributed in space. Now, a collector drop of twice the volume (a result of coalescence of two droplets) is introduced and allowed to fall through the cloud of droplets. The growth of such a droplet is punctuated by coalescence events.

In accordance with the assumption of perfect spatial randomness, the consecutive random time intervals between coalescence events are statistically independent random variables. Let the mean time to the first coalescence be τ. Again, because of the perfect spatial randomness, the probability density function of times to first coalescence (describing an ensemble of test drops) is given by the exponential distribution

$$p(t) = \frac{1}{\tau}\exp\left(-\frac{t}{\tau}\right). \tag{1}$$

This is, perhaps, best understood by noting that this distribution is memoryless.[2] This lack-of-memory property is both fundamental and ubiquitous in applications; for example, see the cover of the text by Balakrishnan and Basu (1995).

[1] The confusion, we think, is caused by misinterpreting the variance–mean (Poisson) relation $\sigma_n^2 = \mu$, which does not simply scale with volume because μ and σ_n^2 are both unitless (unlike concentration).

It is important to note the skewness of (1) from the outset because it is associated with the lucky droplets that grow more rapidly. When exponentially distributed random numbers are displayed as time series, there are frequent excursions below the mean and rare but large excursions above the mean. For example, the 10^{-6} fraction of fastest droplets undergo the first coalescence in one-millionth of the mean time to coalescence because $P(t) = \int_0^t p(s)ds = 1 - \exp(-t/\tau) \approx t/\tau$, for $t \ll \tau$. These are the lucky drops as they begin to grow more rapidly. What about the 10^{-6} fastest growing droplets over, say, six consecutive coalescence intervals? How is the luck redistributed? One possibility is to be among the fastest 10% in each of the six consecutive (and independent) collisions, yielding $(0.1)^6$. In other words, the one-in-a-million fraction of droplets can be expected to grow roughly 10 (versus 10^6) times faster than the average, over the six consecutive collisions with equal τ. These estimates are not exact, of course, because after each coalescence the expected time to the subsequent event must be recalculated. However, insofar as the first few coalescence events dominate cumulative growth time, as we shall see in the next section, the rough estimate is not far from the exact result (see the sidebar on the "Distribution of Times for *N* Encounters"). We now proceed to obtain an expression for τ, the mean time to coalescence, as a function of the droplet radius.

MEAN TIME TO NEXT COALESCENCE. As in the kinetic theory of gases, the mean time to collision τ satisfies $\tau = L/u$ where L is the mean free path and u is the relevant speed (e.g., see Reif 1965, chapter 12). For gravitational sedimentation, the expected time to first coalescence is

$$\tau = (c\sigma uE)^{-1}, \tag{2}$$

where c is the number concentration of droplets, σ is the effective cross section, u is the relative velocity, and E is the coalescence efficiency (e.g., Rogers and Yau 1989). As the collector drop volume changes after each coalescence, so does the expected time for the subsequent collision because σ, u, and E have changed (given the 10^{-6} fraction, c is assumed constant throughout the paper). Our next task, therefore, is the development of a simple approximation for τ as a function of the droplet size r.

To that end, let us start with a cloud of identical droplets (single size) but pick a collector drop that starts the process by undergoing a single coalescence event and acquiring volume $v_1 = 2v$, where v is the initial volume for all of the cloud droplets. The subscript 1 in the volume refers to the drop having undergone one coalescence event.[3] Then, the collector drop volume after n coalescence events is simply $v_n = (n + 1)v$. For drops between 10 and 50 μm in radius, σ, u, and E each scale approximately as r^2 yielding r^6 dependence for the collision rate (Pruppacher and Klett 1997, p. 618). Thus, $\tau^{-1} \propto r^6$, and, therefore, $\tau_n \propto r_{n-1}^{-6}$; that is, the expected time between the $(n-1)$th and nth coalescence, τ_n, decreases approximately as the inverse square volume: $\tau_n \propto v_{n-1}^{-2} = v^{-2} \times n^{-2} = (nv)^{-2}$ where the notation is such that $\tau_1 = \tau$ involves the original volume $v_0 \equiv v$, and τ_n depends on the $(n - 1)$th volume.

Observe that for a droplet to grow from 10 to 50 μm (drizzle size is about 100 μm in diameter and above), the radius must increase fivefold. This implies $5^3 = 125$ coalescence events. Hence, the mean cumulative time to nth coalescence, $T_n \equiv \langle t_1+t_2+ \ldots +t_n \rangle = \tau_1+\tau_2+ \ldots +\tau_n$, can be approximated by an infinite sum, using Euler's $\sum_{n=1}^{\infty} n^{-2} = (\pi^2)/6$, as

$$\begin{aligned} T_n &= \sum_{n=1}^{125} \tau_n \approx \sum_{n=1}^{\infty} \tau_n \\ &= \tau\left(1+\frac{1}{2^2}+\frac{1}{3^2}+\ldots+\frac{1}{n^2}\right) \\ &= \tau\left(\frac{\pi^2}{6}\right) \approx 1.6\tau, \end{aligned} \tag{3}$$

where angular brackets denote averaging over the drops.[4] The relative importance of early collisions is

[2] Denote the cumulative distribution function by $P(t)$ so that $p(t) = dP(t)/dt$ and define the distribution tail $\bar{P}(t_0) = 1 - P(t_0) = \text{prob}(t > t_0)$, that is, the probability that an observed time interval is larger than t_0. A random variable X is memoryless if $P(X > s + t|X > t) = P(X > s)$ where the vertical bar denotes conditional probability. There is no memory because waiting does not affect the current probability. Lack of memory is the defining feature of the exponential distribution because $\bar{P}(t_1 + t_2) = \bar{P}(t_1)\bar{P}(t_2)$ is satisfied when $\bar{P}(t)$ is exponential. The sufficient part is clear from $e^{(x+y)} = e^x e^y$. An exponential $\bar{P}(t)$ yields negative exponential $p(t)$ because the latter is the negative derivative of the former. Hence, pure randomness and additivity yield lack of memory.

[3] The collector drop formation is addressed in a later section but for now the reader can imagine particles being advected into each other by the airflow, or condensing on giant nuclei.

[4] The first 125 terms account for 99.5% of the infinite sum (3).

appreciated by noting that the sum of the first five terms is 89% of the infinite sum ($\frac{\pi^2}{6}$) or 90% of the growth time from 10 to 50 μm, while the first 11 terms account for over 95% of the total growth time. This dominance of the early terms, combined with the skewness of the exponential distribution, suggests that droplets must not just be lucky but must be lucky early, for example, within the first six or so coalescence events.

But how are the collisions initiated if the droplets are all the same size? After all, the first term contributes more than one-half of the entire series but this is the only term where the relative settling velocity is infinite if all droplets are exactly the same size. We propose an explanation in a subsequent section, but typically this problem of initiation of the process is bypassed by postulating a larger collector drop introduced by outside factors. For example, if we invoke giant nuclei or hygroscopic seeding and increase the collector drop volume to $2v$ or 12.5 μm, the series (3) can be recalculated by subtracting the first term from $\pi^2/6$, that is, subtracting unity from both sides. In this case, the (new) first term is not quite as dominant, accounting for about 39% of the cumulative time while the first two terms contribute about 56%. One can proceed in this manner to any size collector drop, subtracting more leading terms from the series (3), with progressively diminishing importance of the early collisions because the "luck" is now supplied externally by other means. In other words, the presence of giant nuclei or precipitation seeds reduces the stochastic element of the coalescence process, by supplying the luck externally.[5]

We are now in a position to pose and answer Robertson's (1974) question analytically: At what n can one switch to the continuous coalescence equation without a significant loss of accuracy? In other words, when can fluctuations in the time to nth coalescence be neglected? To that end, we make use of the following observations:

i) cumulative growth time is a sum of exponentially distributed random variables;
ii) the variance of the exponential probability density function equals the square of its mean ($\sigma^2 = \tau^2$); and
iii) variances due to independent causes add.

Hence, from Eq. (3), the variance of the (random) cumulative time to Nth coalescence, $\sigma^2(T_N)$, is given by

$$\begin{aligned}\sigma^2(T_N) &= \sum_{n=1}^{N} \tau_n^2 \\ &= \tau^2\left(1+\frac{1}{2^4}+\frac{1}{3^4}+\ldots+\frac{1}{n^4}+\ldots\right) \\ &\approx \tau^2 \times \frac{\pi^4}{90},\end{aligned} \tag{4}$$

where, again, we have approximated the finite sum (e.g., 125 terms) with the infinite one. The relative fluctuation for the entire growth (σ/τ, cumulative standard deviation relative to the mean) is $6/\sqrt{90} \approx 0.63$.[6]

The same quantity for the first 15 collisions is about 0.66 (within 5% of the limit) so the continuous approach can be tolerated, but recall that these 15 collisions contribute over 96% of the total growth time. Thus, while the continuous coalescence approach is valid for the later part of the series on a term-by-term basis, this is the part of the series that contributes negligibly to the total growth time. In other words, early fluctuations in coalescence times can never be neglected.

As an example of the physical insight to be gained from the simple approach considered here, consider the redistribution of liquid water content [in the spirit of Twomey (1966)]. What happens if we hold cloud liquid water content constant but vary droplet size, for example, let all droplets combine into doublets of twice the volume ($2v$), but with half the concentration? This may help in studies of the secondary aerosol indirect effect or can be viewed as an extreme case of droplet clustering when the collector drop encounters two droplets at once.

[5] The rate of decay of τ_i with increasing coalescence number in (3) determines the importance of early collisions, but the numerical value of the exponent is not critical to arguments that follow. Should the r^{-6} collision rate be deemed unsuitable, for example, in other collision mechanisms such as turbulence or Brownian motion (Seinfield and Pandis 1998, section 12.3), our method is readily generalized to another polynomial fit, resulting in $\sum_{n=1}^{125} n^{-s}$, where $s \neq 2$. This sum can be obtained by looking up corresponding values of the Reimann zeta function $f(s) \equiv \sum_{n=1}^{\infty} n^{-s}$. For s ≤ 1 the infinite series diverges, hinting perhaps that such a collision process is not dominated by early events.

[6] This is unlike a single exponential probability distribution with $\sigma/\tau = 1$. This is also in contrast to a sum of exponential distributions with equal τs, where the central limit theorem predicts $\sigma/\tau \sim 1/\sqrt{N}$ and σ/τ approaches zero with increasing N. Hence, the memory of the early collisions persists.

The coalescence rate changes because the concentration decreases by a factor of 2 while droplet cross sections, settling velocities, and efficiencies go up by a factor of $2^{2/3}$ each. The net result is a decrease by a factor of 2 in τ. Otherwise, the series (3) remains unchanged, except for the number of terms needed to reach the 50-μm radius (64 versus 128). We see, then, that the redistribution of liquid water content only results in rescaling the characteristic time scale τ, but does not alter the essential structure of the series (3) and, therefore, the role of fluctuations in early collisions. This is the benefit of the decoupling of the mean behavior from fluctuations. Hence, while we expect the doublets to grow on average twice as fast as the singlets, the fastest 10^{-6} doublets will be just as lucky as the fastest 10^{-6} singlets, relative to their own mean time τ_{doublet}. While such analysis is helpful, further progress requires the probability distribution of the time required for several encounters (coalescence events), generalizing (Eq. 1). This is done in the next section.

DISTRIBUTION OF TIMES TO DRIZZLE: THE LUCK FACTOR. Rain or drizzle formation requires hundreds of droplet coalescence events and we therefore consider the cumulative time to nth coalescence, $T_n = t_1 + t_2 + \ldots + t_n$. When viewed as a sum of independent random variables, the probability distribution of T_n might be expected to approach the bell-shaped (Gaussian) form, whose variance diminishes with the increasing number of terms as $1/\sqrt{N}$, as implied by the famous central limit theorem. However, the theorem conditions are violated because the early terms dominate so that our sum of random variables is unequally weighted. Yet, it is still possible to derive the distribution of times for a test drop to undergo N collisions as outlined in the sidebar Distribution of Times for N Encounters. The resulting (generalized Erlang) probability distributions are shown in Fig. 1 for several N (number of coalescence events), where we see rapid convergence to the asymptotic limit. These distributions are discussed in greater detail in the sidebar on the Distribution of Times for N Encounters. We note here, however, that the variance of the distributions is still given by the expression in Eq. (4), confirming our earlier and simpler arguments.

Next, to explore the role of fluctuations, we ask: how fast is the one-in-a-million fastest fraction of (lucky) droplets? To that end, we need an expression for the fraction of drops at a given time that have experienced N encounters: this fraction is the cumulative probability distribution, shown in Fig. 2 (the blue

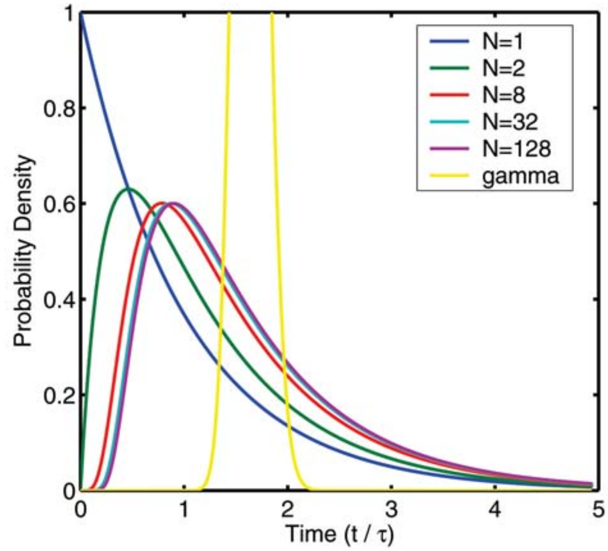

FIG. 1. A family of generalized Erlang probability distributions (density functions). The unitless abscissa is a fraction of the mean time to first coalescence and the curves are distributions of dimensionless times required for the collector drop to undergo N = 1, 2, 8, 32, 128 coalescence events. The N = 1 curve is an exponential distribution, and the other curves may be thought of as convolutions of exponentials with mean τs varying according to series (3). Rapid convergence with increasing number of coalescence events (N) is observed. For comparison, an ordinary Erlang (gamma) distribution is shown whose mean time to coalescence is set to $\frac{(\pi^2/6)\tau}{128}$ so that the cumulative growth time is equal to that of the N = 128 curve. The one-in-a-million fastest (lucky) droplets for the constant-coalescence-time scenario (gamma) are seen to be far less lucky than the corresponding droplets with realistic, rapidly decreasing coalescence rates (generalized Erlang with N = 128).

curve) for N = 128 coalescence events, with τ varying as the series (3). The unitless abscissa, corresponding to a given droplet fraction, may be regarded as a luck factor ϕ for that droplet fraction. For example, the blue curve intersects the 10^{-6} line at about $\phi \approx 0.1$, corresponding to $\phi^{-1} = 10$ or 10 times faster than the average pace of growth (regardless of the mean time τ). Thus, the 10^{-6} lucky drops are expected to reach 50 μm in time $\phi\tau$ rather than τ or 10 times faster than typical droplets.[7] The changing role of luck as quantified by ϕ can be seen in the other three curves in Fig. 2, which correspond to series (3) with the first one, two, and

[7] It is often of interest to obtain the drop size distribution at a given time rather than the distribution of times for a given number of collisions as considered here. The former is obtained in the online supplement.

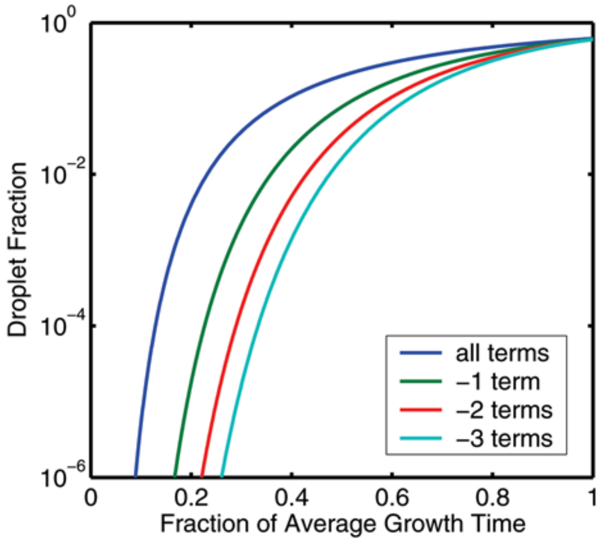

FIG. 2. The fraction of droplets that have experienced 128 encounters (cumulative probability distribution) corresponding to the generalized Erlang distribution (density function) of Fig. 1. Each curve is for 128 coalescence events but with a varying number (0–3) of the leading terms of series (3) removed. Logarithmic coordinates are used in order to clearly show the lucky (10^{-6}) fraction. The abscissa is a unitless fraction: the ratio of actual cumulative growth time to the average cumulative growth time. For fair comparison, time for each curve is normalized by its average growth time [$(\pi^2/6)\tau$, $(\pi^2/6 - 1)\tau$, $(\pi^2/6 - 1 - 1/4)\tau$, and $(\pi^2/6 - 1 - 1/4 - 1/9)\tau$, respectively, from left to right]. For all curves, the abscissa of a given fraction may be thought of as a luck factor ϕ for that fraction. For example, the blue curve crosses the 10^{-6} fraction at about $\phi \approx 0.1$, corresponding to $\phi^{-1} = 10$ times faster than the average pace of growth (regardless of the mean time τ). Thus, the 10^{-6} lucky drops are expected to reach 50 μm in time $\phi\tau$ rather than τ or 10 times faster than typical droplets. Also, we see that the 10^{-6} luckiest fraction of drops is progressively luckier, or farther from the mean, as the collector drops approach the size of the collected drops. As expected, the presence of large collector drops, such as giant nuclei, reduces the importance of fluctuations in the formation of precipitation.

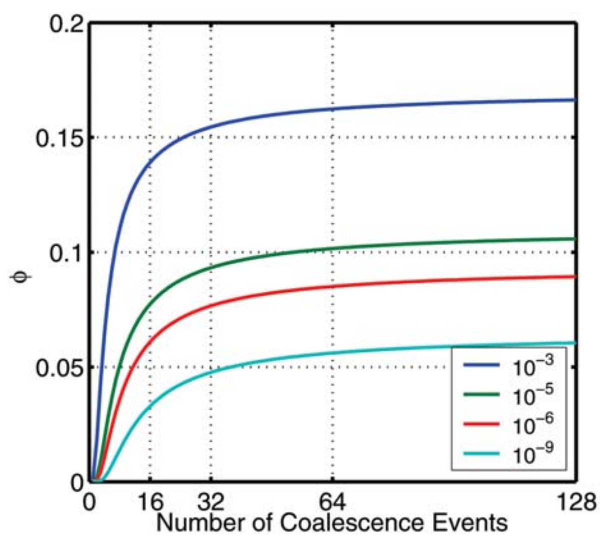

FIG. 3. The luck factor ϕ vs the number of coalescence events for the generalized Erlang distribution. The actual speedup is given by ϕ^{-1} so that $\phi = 0.1$ implies a factor of 10 faster than the mean growth rate. An asymptotic approach to a limit with the increasing number of coalescence events is observed. The curves correspond to four different fractions of the lucky particles. The 10^{-5} and 10^{-6} fractions are in agreement with observations of relative drop and droplet concentrations. The one-in-a-billion droplet (associated with occasional drizzle or a lonely raindrop) is only about 2.5 times faster than the one-in-a-thousand droplet. The dependence on the number of coalescence events is rather weak, (e.g., in order to half the luck factor, the number collisions must be decreased from 125 to about 9). These curves are a result of the clear separation of fluctuations (random variable times) from the mean behavior (parameter of the distributions τ). Yet, this "decoupling" is rather subtle because, say, a seventh term in the mean growth series (3), is not entirely negligible for the growth of the 10^{-6} fastest droplets (despite contributing only 1.2% to the total average growth).

three terms removed: meaning the collector drops start with volumes $2v$, $3v$, and $4v$, respectively. The growth time of the 10^{-6} fraction of lucky droplets steadily approaches the mean growth time, confirming that fluctuations in growth time are less prominent when the growth process is short-circuited by the initial presence of larger droplets. The luck factor ϕ versus the number of coalescence events is shown in Fig. 3 for four different lucky fractions. We see that ϕ approaches a limit as the number of coalescence events increases. Simply taking the asymptotic value ($\phi = 0.087$ for the one-in-a-million curve) allows a simple discussion of rain initiation versus droplet size solely in terms of the physics of the mean collision time τ, as discussed next.

Our examination of the role of fluctuations in stochastic coalescence is now concluded. It culminates with Fig. 3 and a rather robust conclusion that the fastest 10^{-6} cloud droplets grow about 10 times faster than the average droplets ($\phi \approx 0.1$).

DISCUSSION: CAN IT RAIN IN 30 MINUTES?

While the average growth time from 10 to 50 μm is about $(\frac{\pi^2}{6})\tau$, the lucky one-in-a-million droplets (sufficient to initiate rain) accomplish this in time $\phi(\frac{\pi^2}{6})\tau$. Henceforth, the discussion is concerned with the av-

DISTRIBUTION OF TIMES FOR *N* ENCOUNTERS

Our goal is to obtain the probability distribution for the (random) cumulative growth time $T_n = t_1 + t_2 + \ldots + t_n$. Recall that a sum of independent random variables has a probability distribution that is a convolution of the individual distributions. For identically distributed (exponential) random variables with equal averages (τ), the Erlang probability density results. This is also known as the Gamma density for an integer number of events, originally obtained by A. K. Erlang, when considering the distribution of waiting times for telephone networks in the early twentieth century (Evans et al. 2000; Porter and Ogilvie 2000).

The Erlang distribution becomes progressively narrower as the number of terms increases and approaches an extremely narrow bell-shaped curve in the limit. Not so in our case because of the unequal weighting (different τs) of the terms in the sum $T_n = \tau_1 + \tau_2 + \ldots + \tau_n$. The inhomogeneous Poisson process provides the proper framework because of statistical independence and variable but known mean times. The individual exponential distributions $p(t_1) \ldots p(t_N)$ with different τs can then be convolved to yield the generalized Erlang distribution [Ventzel and Ovcharov (1988, pp. 359, 367); see also Syski (1986 p. 51), but note the omission of the alternating sign]:

$$p(t_{1:N}) = (-1)^{N-1} \prod_{i=1}^{N} \lambda_i \sum_{j=1}^{N} \frac{e^{-\lambda_j t}}{\prod_{k=1,k\neq j}^{N} (\lambda_j - \lambda_k)}, \qquad \textbf{(SB1.1)}$$

where $\lambda_i \equiv \tau_i^{-1}$. Figure 1 provides several illustrations of the Erlang distribution. Closer inspection reveals similarity to expressions derived by Telford (1955), as detailed in the online supplement, but connection to work done by Erlang and others in the fields of queuing theory and statistical signal processing does not appear to have been appreciated (e.g., Gross and Harris 1998, especially section 6.2.1).

In order to evaluate the fastest one-in-a-million droplet growth rate, we require the cumulative probability density for the generalized Erlang distribution, which is given by

$$P(t_{1:N}) = (-1)^{N-1} \prod_{i=1}^{N} \lambda_i \sum_{j=1}^{N} \frac{1 - e^{-\lambda_j t}}{\lambda_j \prod_{k=1,k\neq j}^{N} (\lambda_j - \lambda_k)}, \qquad \textbf{(SB1.2)}$$

where again $\lambda_i \equiv \tau_i^{-1}$. This function is shown in Fig. 2 for a variety of scenarios. In the limit of $t \ll \tau$, which is valid for coalescence initiation, these distributions reduce to the simple form $P(t_{1:N}) \approx N!(t/\tau)^N$ (see the online supplement).

To gain an intuitive appreciation of these distributions, let us return to the case of equal τs. Since individual probabilities are equal and independent, the 10^{-6} probability (lucky fraction) is a product of equal fractions for each individual step; or $p_1 p_2 p_3 \ldots p_N = p^N = 10^{-6}$. For the 128 collisions considered in Fig. 2 this suggests a crude estimate of $\phi = (10^{-6})^{(1/128)} \approx 0.9$ or 90%. The fact that the one-in-a-million fastest droplet is only 10% faster than the mean for the 128 collisions reflects the narrowness of the corresponding bell-shaped probability density function [this distribution, with a mean collision time set to $(\frac{\pi^2}{6})\frac{\tau}{128}$ for consistent comparison, is shown in Fig. 1]. The relative fluctuations decay as $1/\sqrt{N}$ and are quite small for $N = 128$ coalescence events.

The actual coalescence growth, however, is dominated by the early history where the droplets must invest most of the available luck. Crudely, one can divide the series into two parts: the first, say, six events and the rest of the series, with the former accounting for 90% of the cumulative mean time. Then, the 10^{-6} fraction is distributed as $(0.1)^6$ over the first part of the series and the remaining 10% are simply ignored. This is in rough agreement with the factor-of-10-speedup of the lucky droplets, undergoing 128 coalescence events. The speedup (ϕ^{-1}) is shown in Fig. 3 and discussed in the main text.

erages only. Therefore, all that remains for an explanation of warm rain initiation in cumulus clouds is that the lucky droplets grow to raindrops within commonly observed times (~30 min). The essential point here is that it is $\phi(\frac{\pi^2}{6})\tau$, rather than $(\frac{\pi^2}{6})\tau$, that needs to be set to 30 min = 1800 s. Is the acceleration factor of 10 (ϕ^{-1}) sufficient to explain the apparently often observed warm rain initiation in 30 min? To that end, we require

$$\phi \frac{\pi^2}{6} \tau \leq 30 \text{ min} \quad \text{or} \quad \tau \leq 3 \text{ hr}, \qquad (5)$$

which, combined with Eq. (2), yields a bound on the efficiency–velocity product (denoted u_{bound}):

$$Eu \geq \frac{10^4 \text{ s}}{c\sigma} \equiv u_{\text{bound}}. \qquad (6)$$

COALESCENCE INITIATION IN A FAST EDDY

Consider the traditional turbulent energy cascade, with energy dissipation occurring on length and velocity scales of λ_o and $u_{\lambda o}$. This viscous dissipation (Kolmogorov) scale, $\lambda_o \sim 1$ mm, is 10–100 times larger than the droplet size scale λ, at which collisions occur. Smoothness of the viscosity-dominated velocity field allows one to expand the velocity difference of the droplets, separated by distance λ, in powers of λ. Keeping the first term in the series yields $u_\lambda = u_{\lambda o}\lambda/\lambda_o$ as shown in Landau and Lifshitz (1959, section 32) where another argument leading to this result is also given. Traditional energy cascade arguments then relate u_λ to large-scale variables as

$$u_\lambda = \frac{\lambda}{L}\Delta U R^{\frac{1}{2}}, \tag{SB2.1}$$

where, again, the quantity of primary interest to us, u_λ, is the velocity difference at points separated by distance λ. The distance L is the large sale of the energy-injecting eddies, ΔU is the mean velocity difference at points separated by L, and $R \equiv L\Delta U/\nu$ is the turbulence Reynolds number.

One expects the length scales of energy injection in cumulus clouds to be buoyancy driven and associated with up- and downdrafts and we pick the typical $L = 200$ m, $\Delta U = 2$ m s^{-1} (e.g., see Houghton 1985, p. 286, Fig. 8.2; Emmanuel 1994, 194–196), corresponding to $\varepsilon = 400$ cm^2 s^{-3} and turbulent diffusivity $K = 400$ m^2 s^{-1}. For $\lambda = 4 \times r = 40$ μm, these numbers yield $u_\lambda \approx 2$ mm s^{-1}.

As shown in the main text, the velocity difference needed for warm rain initiation is $u_{bound} = (10^4 \text{ s})/n\sigma$, which is about 300 μm s^{-1}, so that turbulence provides $u_\lambda \approx 2$ mm s^{-1}, which is about seven times larger. This means that the coalescence efficiency of about 0.15 is sufficient to initiate rain in 30 min, even for a cloud of a single droplet size. Furthermore, increasing ΔU by a factor of $2^{2/3}$ or, say, doubling the liquid water content (w) to 2 g m^{-3}, lowers the required coalescence efficiency to 0.07 (while holding the time to rain to 30 min). Nevertheless, the actual efficiency of shear-induced coagulation may be lower still and we must examine the question explicitly versus the droplet size.

To that end, we return to the redistribution-of-liquid-water-content scenario by keeping w constant but varying the concentration and (single) droplet size accordingly. Observe that, for fixed w, $u_{bound} = (10^4 \text{ s})/c\sigma$ depends linearly on the droplet size r. Indeed, for fixed w, $c \sim r^{-3}$, $\sigma \sim r^2$, and therefore, $u_{bound} \sim r^1$. On the other hand, $u_\lambda = \frac{\lambda}{L}\Delta U R^{1/2}$ also depends linearly on r because we set $\lambda = 4r$. Therefore, an important conclusion is that the ratio u_{bound}/u_λ, which is the lower bound on the coalescence efficiency, is independent of the droplet size. We are now able to express this threshold efficiency (E_{bound}) in terms of the observed time for rainfall (T_{obs}), the fractional water volume ($\omega \equiv w/\rho$), and a large eddy turnover time ($\tau_L \equiv L/\Delta U$):

$$E_{bound} \equiv \frac{u_{bound}}{u_\lambda} = \frac{\pi^2}{72}\frac{\phi}{\omega}\frac{1}{\sqrt{R}}\frac{\tau_L}{T_{obs}} \sim \frac{10}{72}\times\frac{10}{10^{-3}}\times\frac{1}{\sqrt{10^6}}\times\frac{\tau_L}{T_{obs}} \sim \frac{\tau_L}{T_{obs}}. \tag{SB2.2}$$

In contrast, the actual efficiency is likely to have a strong dependence on r (e.g., r^2 for gravitational coagulation between 10 and 50 μm). Hence, only those clouds will rain whose droplet sizes correspond to coalescence efficiency exceeding u_{bound}/u_λ. Thus, for a given (macroscopic) $\Delta U(L)$, warm rain is initiated by droplet sizes whose coalescence efficiency exceeds E_{bound}, given by (10). Seen from this perspective, rare but vigorous vortices (fast eddies) could promote droplet growth even more efficiently than giant condensation nuclei.

This bound can be viewed as the minimal droplet relative velocity necessary for warm rain to occur in cumulus clouds within 30 min. As an illustration, inserting numerical values for 10-μm droplets and 1 g m^{-3} liquid water content (concentration of about 240 droplets cm^{-3}) yields u_{bound} of 265 μm s^{-1}. Thus, in order to produce rain in 30 min with the help of the factor-of-10 growth acceleration of the lucky droplets, we require u_{bound} on the order of 300 μm s^{-1}, over separation distances of 40 μm or so (i.e., two droplet diameters).

It is now time to face the problem of collision initiation. The equation $\tau = (c\sigma uE)^{-1}$ involves relative settling velocity u. The latter, technically, is zero when collector drop and droplet are equal in size. There are several ways around this difficulty. Realistic droplet size distributions have some dispersion caused by the initial aerosol size distribution and by fluctuations in the condensation growth process. Also, one may assume (as is usually done) that the collector is somewhat larger—perhaps because of a giant cloud condensation nucleus. Increasing the collector drop density (mineral or salt is twice as heavy as water and so is the corresponding terminal speed) or, say, doubling the volume (12.6 μm) removes the difficulty, whereupon series (3) can be resummed by subtracting the first term from $\pi^2/6$, that is subtracting unity from both sides, as discussed earlier. Nevertheless, in order to explore the fundamental lower bound for a variety of growth scenarios, we will continue with the assumption of a single droplet size and focus on the crucial genesis stage of coalescence.

Can the required u_{bound} be attributed to a physical mechanism different from gravitational coagulation? For example, can the mean shear in a cumulus cloud supply sufficient relative velocities (spatial velocity differences) at interdroplet distances on the order of 10–100 μm via the traditional turbulent energy cascade? Our general conclusion is that for sufficiently vigorous (yet realistic) turbulence the answer is yes and the supporting physical arguments are given in the sidebar on Coalescence Initiation in a Fast Eddy. Thus, even a cloud of single-size droplets can rain within 30 min. Note that we are invoking neither still-controversial turbulence-induced inertial effects nor any other kind of spatially clustered droplets, merely the traditional turbulent energy cascade. Seen from this perspective, rare but vigorous vortices could promote growth just as efficiently as giant condensation nuclei.

CONCLUDING REMARKS. The importance of the stochastic element in growth by coalescence has been known at least since Telford (1955) and in the online supplement we discuss how our expressions relate to those of Telford for the droplet size distribution, modified for realistic collision rates. Taking the stochastic element as a starting point, we decoupled the effects of fluctuations from those of the mean growth, doing so with a plausible functional form for the collision rate, obtaining nevertheless simple analytical expressions. Our approach is readily generalized to coalescence time distributions other than exponential, thereby allowing the introduction of droplet clustering, negative spatial correlations, etc. Furthermore, decoupling of fluctuations from the mean allows simple exploration of the effect of liquid water redistribution with droplet size. Finally, it allows us to focus on the mechanisms for those critical initial collisions via Eq. (6).

The generalized Erlang distribution function permits straightforward consideration of the "lucky" fraction, whether this is the fastest 10^{-6} droplets required for warm rain initiation, the 10^{-9} fraction needed for patchy drizzle formation, or the 10^{-12} fraction of lonely drops occasionally falling from seemingly thin clouds. The role of the initial size of collector drops can be clearly traced by deleting the corresponding terms from the coalescence time series. This has implications for the importance of the stochastic treatment when large particles are present in sufficient quantities to act as precipitation seeds (e.g., hygroscopic seeding, ultragiant nuclei, etc.).

Finally, a compelling conclusion is that the factor-of-10 acceleration in the growth of the lucky droplets, combined with traditional turbulent cascade ideas puts us, at least, within striking distance of initiating rain from turbulent clouds in 30 min with 10-μm droplets and 1 g m^{-3} liquid water content. This is despite having no size dispersion, no clustering, and no giant nuclei. Indeed, rare, fast eddies may be more effective in initiating warm rain than rare, giant condensation nuclei.

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