Secular growth of droplet size variance due to condensation in turbulent clouds

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We use a stochastic model and direct numerical simulation to study the impact of turbulence on cloud droplet growth by condensation. We show that the variance of the droplet size distribution increases in time as $t^{1/2}$, with growth rate proportional the large-to-small turbulent scale separation and to the turbulence integral scales but independent of the smallest turbulence scale. Direct numerical simulations confirm this result and produce realistically broad droplet size spectra over time intervals of 20 minutes, comparable with the time of rain formation.

Many multi-scale processes—including nutrient foraging of plankton, gas/dust accretion disks in astrophysics and spray evaporation and combustion in engines—involve the interaction between small particles and tracers transported in a turbulent flow. Here we focus on the case of droplet condensation in turbulent warm (i.e. ice-free) clouds. Clouds are a leading source of uncertainty in climate modeling due to the difficulty of accurately parameterizing the macro-scale effects of micro-scale physical processes, such as the effect of droplet size distribution on precipitation rates and cloud albedos.

The role of turbulence in cloud microphysics presents a range of open questions, particularly as a possible solution for the “bottleneck” problem of rain formation. All cloud droplet populations evolve through a sequence of steps: (1) nucleation/activation of cloud condensation nuclei, (2) droplet growth by condensation and (3) growth to raindrop size by collision and coalescence. The passage from (2) to (3) presents a bottleneck because collisional growth is only triggered when the droplet population acquires a sufficiently broad size distribution, but condensational growth is inversely proportional to droplet radius squared (droplet surface area) in terms of the thermodynamics and turbulence characteristics, modeling the fluctuations as stochastic processes. Understanding the mechanisms that break the condensational bottleneck is a longstanding and still unresolved problem in atmospheric physics.

Turbulence has often been invoked as a key process in this context since it can enhance collision rates via inertial clustering and the so-called “sling effect.” Turbulence also induces fluctuations in the supersaturation field that can potentially broaden droplet spectra in the condensational stage. Early studies using analytical models and direct numerical simulations (DNS) generally showed too little broadening as compared with observations. Later work attempting to simulate large-scale turbulence in an $O(100 \text{ m})$ cloud showed a dramatic broadening of the droplet size spectrum but only with ad-hoc assumptions about the small-scale supersaturation fluctuations. Lanotte at al. modelled both small- and large-scale effects on the droplet size distribution with simulations of a cloud of 70 cm and pointed out the importance the Taylor Reynolds number, $Re_{\lambda}$, the non dimensional parameter measuring the large to small scale separation in homogeneous isotropic turbulence. In particular, they suggested that droplet spectral broadening should scale with $Re_{\lambda}$.

A question that has not been addressed so far is the long-term fate of the droplet spectrum: does it reach a steady state, or does it continue to evolve? The large range of scales involved makes DNS very computationally demanding, and all existing simulations consider time spans between few seconds and two minutes, well below the observed rain formation timescale.

Here, we approach this question by first deriving an analytical expression for the standard deviation of the droplet radius squared (droplet surface area) in terms of the thermodynamics and turbulence characteristics, modeling the fluctuations as stochastic processes. We demonstrate that the droplet size distribution increases monotonically with time as $t^{1/2}$. We validate this analytical result by extending previous numerical results with DNS and large eddy simulations (LES) to timescales comparable with those of rain formation, about 20 minutes. Our results imply that every warm cloud would precipitate if given enough time. The broadening rate is determined by the large scale turbulent motions and by the positive correlation between droplet surface area and local supersaturation.

Our physical model is analogous to that in \cite{francesco}. The homogeneous isotropic turbulence obeys the incompressible Navier-Stokes equations

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{\nabla p}{\rho} + \nu \nabla^2 \mathbf{u} + \mathbf{f},$$

where $\mathbf{u}$ is the divergence-free fluid velocity, $p$ the pressure, $\rho$ the air density, $\mathbf{f}$ an external forcing to maintain a statistically stationary state and $\nu$ the kinematic viscosity. These approximations are valid for clouds smaller than $L \approx 100 \text{ m}$ to safely neglect the spatial inhomogeneity and large scale variations of the thermodynamic parameters. The supersaturation field $s$ is transported...
by the fluid according to
\[
\frac{\partial s}{\partial t} + u \cdot \nabla s = \kappa \nabla^2 s + A_1 w - \frac{s}{\tau_s},
\]
a generalization of the Twomey model \[10\]. The diffusivity of the water vapor in air is denoted by \(\kappa\), \(w\) is the velocity component in the gravity direction, \(A_1w\) is a source/sink term of supersaturation resulting from the variation in temperature and pressure with height. The supersaturation relaxation time \(\tau_s\) depends on droplet concentration and dimensions \[20\], \(\tau_s^{-1} = 4\pi \rho_w A_2 A_3 \sum R_i/V\) where \(R_i\) are the radii of the droplets in the volume \(V\), \(\rho_w\) the water density, \(A_1, A_2\) and \(A_3\) thermodynamic coefficients \[13\]. The droplets are assumed to behave as rigid spheres smaller than the Kolmogorov turbulent scale, at low mass fraction to neglect feedback on the flow. The only forces governing the droplet motion are gravity and the Stokes drag (nucleation/activation is not considered):
\[
\frac{dv_d}{dt} = \frac{u(x_d, t) - v_d}{\tau_d} - g e_z, \quad \frac{dx_d}{dt} = v_d
\]
with \(x_d\) and \(v_d\) the droplet position and velocity, \(u(x_d, t)\) the fluid velocity at droplet position, \(\tau_d = 2 \rho_w R_i^2/(9 \nu)\) the droplet relaxation time, \(g\) the gravitational acceleration. The supersaturation at the droplet position, \(s(x_d, t)\), determines the droplet evolution via
\[
\frac{dR_i^2}{dt} = 2A_3 s(x_d, t)
\]
An exact equation for the average of the droplet radius fluctuations is obtained from \[4\],
\[
\frac{d\langle R_i^2 \rangle}{dt} = \frac{d\sigma_{R_i}^2}{dt} = 4A_3 \langle s' R_i^2 \rangle \tag{5}
\]
showing that \(\langle (R_i^2) \rangle \) increases linearly with time only if the correlation \(\langle s' R_i^2 \rangle \) reaches a non-zero statistical steady state.

To quantitatively estimate the droplet growth, we adopt a 1-D stochastic model for the velocity fluctuations \(w_i\) and the supersaturation field \(s'_i\) of the \(i\)-th droplet. Such an approach implicitly assumes that the small-scale turbulent motions have a negligible influence on the macroscopic observables. This assumption is motivated by previous results revealing the broadening of the droplet size distribution with \(Re\) \[13\] and fully justified a posteriori by the numerical data presented below.

Homogeneous isotropic turbulence and supersaturation are modelled with two Langevin equations \[21\]:
\[
\dot{w}'_i(t) + \frac{w'_i(t)}{T_0} dt + v_{rms} \sqrt{ \frac{2 \langle s \rangle}{T_0} \xi_i(t), \tag{6}
\]
where \(\xi(t)\) is \(\delta\)-correlated noise and \(T_0\) the turbulence integral time scale, and
\[
\dot{s}'_i(t + dt) = s'_i(t) - \frac{s'_i(t)}{\tau_s} dt + A_1 w'_i dt - \frac{s'_i(t)}{\tau_s} dt + \sqrt{(1 - C_{ws}^2)(s'^2)/T_0} \eta_i(t) + C_{ws} \sqrt{(s'^2)/T_0} \xi_i(t) \tag{7}
\]
for the supersaturation with forcing from the velocity field. Here \(C_{ws} = \langle u w' \rangle/(v_{rms} \sqrt{(s'^2)})\) is the normalized velocity-supersaturation correlation, \(\tau_s\) is the supersaturation relaxation time based on the mean droplet radius and \(\eta(t)\) a Gaussian random variable, \(\delta\)-correlated in time. Equation \(7\) represents the stochastic version of Twomey model. The mean updraft is zero as the mean supersaturation (the mean droplet radius does not change); entrainment effects \[22\], collisions and inhomogeneities are also not considered to analyze the conservative case when the droplet spectral broadening is only induced by supersaturation fluctuations.

From \[1\], \[6\] and \[7\], assuming \(\tau_s \ll T_0\) as in real clouds, the fluctuation correlations become
\[
\frac{d\langle s'^2 \rangle}{dt} = A_1 \langle w'^2 \rangle + 2A_3 \langle s'^2 \rangle - \frac{\langle s'^2 \rangle}{\tau_s} \tag{8}
\]
\[
\frac{d\langle w'^2 \rangle}{dt} = 2A_3 \langle w'^2 \rangle - \frac{\langle w'^2 \rangle}{T_0} \tag{9}
\]
\[
\frac{d\langle s'^2 \rangle}{dt} = 2A_1 \langle w'^2 \rangle - 2 \frac{\langle s'^2 \rangle}{\tau_s} \tag{10}
\]
\[
\frac{d\langle w'^2 \rangle}{dt} = A_1 v_{rms}^2 - \frac{\langle w'^2 \rangle}{\tau_s} \tag{11}
\]
Assuming statistical quasi-steady state \((\frac{d}{dt} = 0)\) we find that \(\langle s'^2 \rangle_q s = A_1^2 v_{rms}^2 (\tau_s)^2\) and
\[
\langle s'^2 \rangle_q s = 2A_3 A_1^2 v_{rms} (\tau_s)^2 T_0 = 2A_3 \langle s'^2 \rangle_q s T_0 \tag{12}
\]
which give, using \[6\],
\[
\sigma_{R^2} = \sqrt{8A_3 A_1 v_{rms} (\tau_s) (T_0)^{1/2} = \sqrt{8 \langle s'^2 \rangle_q s A_3 (T_0 t)^{1/2}}. \tag{13}
\]
The model shows that the variance of the droplet distribution increases monotonically in a turbulent flow even if the supersaturation fluctuations reached a statistical steady state \(s_{qs}\). The correlation \(\langle s'^2 \rangle_q s\), directly responsible for the radius growth rate, is proportional to the level of fluctuations of the supersaturation field and to the integral scale of the turbulence, see \[12\]. Expression \[13\] can be formulated in terms of Kolmogorov scales since \(v_{rms} \sim (\lambda)^{1/2} v_\eta\) and \(T_0 \sim 0.06 (\lambda) \tau_\eta \) \[21\]:
\[
\sigma_{R^2} \approx 0.7A_3 A_1 v_{rms}^{1/2} (\tau_s) R e^{1/2} \tag{14}
\]
Note that for \(t = T_0\) (short times) the lower limit proposed in \[13\] is recovered, \(\sigma_{R^2} \approx R e^{1/2}\). From \[14\] we
note that $\sigma_{R^2}$ at a fixed time depends only on the scale separation represented by $Re_{\lambda}$ and not on the value of the mean dissipation inside the clouds. This implies that clouds with different dissipation rate and same Reynolds number have an identical behavior in terms of droplet growth by condensation. The droplet/turbulence condensation dynamics does not depend on the turbulent small scales: the correlation between the supersaturation field and the droplet surface area, governing the distribution broadening, is determined by the large flow scales. This result is in contrast with the belief that the variance of the droplet distribution should not grow indefinitely as turbulence tends to decorrelate the particle size from the local saturation field [8].

To test our predictions, we run simulations by gradually increasing the size of the computational clouds from few centimeters to 100 m. The governing equations (1-4) are solved with a classical pseudo-spectral code for the fluid phase coupled with a Lagrangian algorithm for the droplets [23]. All cases share the same turbulent kinetic energy dissipation $\varepsilon = 10^{-3} m^2 s^{-3}$, a value typically measured in stratocumuli. This corresponds to the same small-scale dynamics, with Kolmogorov scale $\eta = (\nu^3/\varepsilon)^{1/4} \approx 1 mm$, Kolmogorov time $\tau_\eta = (\nu/\varepsilon)^{1/4} \approx 0.1 s$ and velocity $v_\eta = \eta/\tau_\eta \approx 1 cm/s$. We examine droplets with 2 different initial radii, 13$\mu$m and 5$\mu$m, denoted as case 1 and 2, with supersaturation relaxation time $\tau_\sigma = 2.5$ and 7$s$, and same concentration (130 droplets per cm$^3$). The reference temperature and pressure are $T = 283 K$ and $P = 10^5 Pa$, with $A_1 = 5 E-4 m^{-1}$, $A_2 = 350 m^3/kg$, $A_3 = 50 \mu m^2/s$. The simulation parameters are reported in Table I. Note that simulation DNS D1 represent the largest Direct Numerical Simulation of a warm cloud up to now.

The time evolution of $\sigma_{R^2} = \sqrt{\langle (R^2)^2 \rangle}$ is shown in Figure 1 for all cases investigated. The data confirm the predictions from [13], i.e. that $\sigma_{R^2} \propto t^{1/2}$. The agreement between the model and the numerical data is remarkable for the largest domain sizes where scale separation is significant and viscous effects can be neglected. For small $Re_{\lambda}$, viscous effects are important and the stochastic inviscid model overestimates the correct behavior.

The correlation $\langle s'R^2 \rangle$ is displayed in the inset of Figure 1 (thin solid line): in all cases, $\langle s'R^2 \rangle$ do reach a statistical steady state, fluctuating around the value determining the growth of $\sigma_{R^2}$. The turbulence creates a positive correlation between supersaturation and droplet surface area fluctuations that increases by increasing the turbulent scale separation, i.e. the cloud size.

To test the model for a larger cloud size, we perform a large eddy simulation (LES E1) of a cloud of about 100 m size. LES can be seen as a good model for our problem since it fully resolves the larger flow scale, those relevant

| Label | N$^3$ | $L_{box}$[m] | $v_{rms}$[m/s] | $T_L$[s] | $T_d$[s] | $Re_{\lambda}$ | $N_d$ |
|-------|-------|--------------|---------------|---------|---------|---------------|-------|
| DNS A1/2 | 64$^3$ | 0.08 | 0.035 | 2.3 | 0.64 | 45 | 6E4 |
| DNS B1/2 | 128$^3$ | 0.2 | 0.05 | 4 | 0.95 | 95 | 9.8E5 |
| DNS C1/2 | 256$^3$ | 0.4 | 0.066 | 6 | 1.5 | 150 | 9E6 |
| DNS D1 | 1024$^3$ | 1.5 | 0.11 | 14 | 3 | 390 | 4.4E8 |
| LES E1 | 512$^3$ | 100 | 0.7 | 142 | 33 | 5000 | 1.3E14 |

TABLE I: Parameters of the simulations. The resolution $N$, the cloud size $L_{box}$, the root mean square of the turbulent velocity fluctuations $v_{rms}$, and $T_L = L_{box}/v_{rms}$ an approximation of the large turbulent scales. $T_d$ indicates the integral time $T_d = (\pi/2 v_{rms}^2) \int E(k)/dk$ with $k$ the wavenumber and $E(k)$ the turbulent kinetic energy spectra [21]. The total number of droplets is indicated by $N_d$. |
to droplet condensation/evaporation, as shown above. We model the small scales with a classic Smagorinski model [24] and use the method of droplet renormalization described in [18] to evolve a feasible droplet number. The Taylor Reynolds number is 5000. The time evolution of $\sigma_{R^2}$ and of $(s^2/R^2)$ are depicted in [2] together with the analytical predictions from [12] and [13]. The agreement between the two fully validates our model.

To motivate the use of the variance $\sigma_{R^2}$ to define the droplet size distribution we show in Figure 3 that its probability distribution is Gaussian, in agreement with measurements in real stratocumuli [25]. The data in the figure refer to the final simulation time (about 20 minutes) and are compared to Gaussian distributions of equal variance. The size distribution from the LES of the large cloud (see inset) reveals that the Gaussian can be fitted just using the value of $\sigma_{R^2}$ from the proposed model also at this higher Reynolds number.

In summary, we have derived an analytical expression for the role of turbulence on the dynamics of droplet condensation and validated it against large-scale numerical simulations. We show that the standard deviation of the square droplet radius fluctuations $\sigma_{R^2}$ increases in time as $t^{1/2}$; the growth rate depends linearly on the turbulent scale separation parametrized by $Re_{\lambda}$. As shown in Figure 3, for a cloud with $Re_{\lambda} = 10000$—a typical value estimated in cumuli with integral scale of 100 m [8]—our expression predicts numerical results requiring $10^{17}$ degrees of freedom. From a practical viewpoint, this indicates the promising potential of modeling approaches based on PDFs.

Our results represent a lower limit for the impact of turbulence on warm rain formation since real clouds typically exceed 100 m in scale and are in general non-homogeneous, with vigorous entrainment of relatively dry air from outside the cloud which leads to enhanced supersaturation fluctuations within the cloud. These additional effects would lead to even larger values of $\sigma_{R^2}$, more than sufficient to explain the spectral broadening observed in real clouds.

FIG. 3: Probability density functions (PDF) of the square radius fluctuations after a simulation time corresponding to about 20 minutes (symbols). The lines represent Gaussian distributions with the same variance. The inset reports data from the LES of a large cloud.

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