The role of turbulent fluctuations in aerosol activation and cloud formation

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Aerosol indirect effects are one of the leading contributors to cloud radiative properties relevant to climate. Aerosol particles become cloud droplets when the ambient relative humidity (saturation ratio) exceeds a critical value, which depends on the particle size and chemical composition. In the traditional formulation of this problem, only average, uniform saturation ratios are considered. Using experiments and theory, we examine the effects of fluctuations, produced by turbulence. Our measurements, from a multiphase, turbulent cloud chamber, show a clear transition from a regime in which the mean saturation ratio dominates to one in which the fluctuations determine cloud properties. The laboratory measurements demonstrate cloud formation in mean-sub saturated conditions (i.e., relative humidity < 100%) in the fluctuation-dominant activation regime. The theoretical framework developed to interpret these measurements predicts a transition from a mean-to-a fluctuation-dominated regime, based on the relative values of the mean and standard deviation of the environmental saturation ratio and the critical saturation ratio at which aerosol particles activate or become droplets. The theory is similar to the concept of stochastic condensation and can be used in the context of the atmosphere to explore the conditions under which droplet activation is driven by fluctuations as opposed to mean supersaturation. It provides a basis for future development of cloud droplet activation parameterizations that go beyond the internally homogeneous parcel calculations that have been used in the past.

Significance

Formation of cloud droplets is a threshold phenomenon; droplets form only when the local relative humidity (RH) exceeds a critical value which depends on the size and chemical composition of preexisting aerosol particles which serve as the seeds for droplets. In the traditional view of this process, only the average value of RH is considered. However, clouds are ubiquitously turbulent, meaning they are characterized not by a single value of relative humidity, but by a distribution. We show, with laboratory experiments and theory, that cloud formation occurs in three regimes, characterized by the relationship between the distribution of RH in the environment and the critical value. Our results show that fluctuations in RH must be considered, as well the average.

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relative dispersion of the DSD and thus affect the cloud optical properties (3).

In a turbulent environment the activation of an aerosol particle would depend on the critical saturation ratio $S_c$ and the instantaneous saturation ratio $S$. The saturation ratio $S$ can be decomposed into a mean $\bar{S}$ and a fluctuation $s'$ characterized by its SD $\sigma_s = \langle s'^2 \rangle ^{1/2}$. Based on the relative importance of $\bar{S}$ and $\sigma_s$ compared to $S_c$, we identify three different activation regimes. The three regimes are best understood by considering the distribution of $S$ and a population of monodisperse aerosol with $S_c$, as schematically shown in Fig. 1. Regime 1 is referred to as “mean-dominated activation” and corresponds to $\bar{S} > S_c$. In this case, we expect that essentially all aerosols will activate and the fluctuations do not play any role in the activation process. This may be considered as consistent with the homogeneous-parcel approach in cloud physics. Regime 2 is referred to as “fluctuation-influenced activation” and corresponds to $\bar{S} - S_c \lesssim \sigma_s$ with $\bar{S} > S_c$. Because a small but significant fraction of $S < S_c$, then the aerosols may not always activate depending on the local supersaturation they encounter and may remain as a haze droplet until a supercritical ($S > S_c$) environment is encountered. Similarly, an activated droplet, small in diameter, may encounter a locally sub-saturated parcel and deactivate as a consequence. Under these conditions the role of supersaturation fluctuations is to either deactivate cloud droplets or suppress activation of aerosols and maintain the haze droplet population. Regime 3 is referred to as “fluctuation-dominated activation” and corresponds to $|\bar{S} - S_c| \lesssim \sigma_s$ with $\bar{S} < S_c$ (subcritical). In this case, a significant fraction of the distribution of $S$ is subcritical such that the mean saturation ratio is less than $S_c$. Under these conditions, cloud droplets can activate only if the haze droplets encounter a significantly large positive fluctuation in $\bar{S}$. In this regime we have active cloud droplets only due to positive fluctuations in $\bar{S}$. In other words, under subcritical conditions cloud droplets may activate only due to large supersaturated turbulent fluctuations (15). In this paper we demonstrate and explore these three regimes through controlled laboratory experiments.

**Experimental Method**

Experiments were conducted in the Pi chamber, operated under an unstable thermal gradient to create a supersaturated turbulent environment (23). This volume was seeded with dry, nearly monodisperse sodium chloride aerosols (diameter ≈130 nm) to generate a cloud. Over time, the system reaches a steady state in number concentration of aerosols and cloud droplets by striking a balance between the injection of aerosols and the removal of cloud droplets through sedimentation. The mean saturation ratio and the intensity of its fluctuation are altered by varying the temperature and vapor pressure gradient across the chamber and also by varying the aerosol injection rates. The size distribution of the droplets in the chamber averaged over 100 s at a flow rate of 5 L per min was measured using the WELAS 2000 Digital optical particle counter in the size (diameter) range of 0.6 to 40 μm (24, 25). This is a measurement capability not available in previous published results from the Pi chamber, providing the diameter of both unactivated haze droplets and activated cloud droplets, which is essential to understand the activation process. In the experiments the smallest scales of turbulence, namely the Kolmogorov length and time scales, are ~1 mm and 0.1 s, respectively (23, 26, 27), and are comparable to the smallest scales in stratocumulus clouds and cumulus clouds with moderate updrafts (28, 29). Since the activation of aerosols is a small-scale process, the understanding gained from these experiments is directly relevant to atmospheric clouds. The experimental conditions and the microphysical properties of the steady-state clouds are summarized in Table 1. Additional details about the experiments are discussed in SI Appendix. The data used in this paper are available in ref. 30.

**Experimental Observations**

Fig. 2 A and B shows the size distribution of the droplets (both haze droplets, $d \leq d_c$, and cloud droplets, $d > d_c$, where $d_c = 2 \left( \frac{2}{3} \right)^{0.5}$ is the critical diameter of the aerosol; see Eq. 7 for details on $a$ and $b$) for five different aerosol injection rates $I$. The $d_c$ for the aerosols in the experiment is ≈1.8 μm and is marked in Fig. 2. We observe that as the injection rate $I$ is increased, the size distribution becomes bimodal with one peak in the haze droplet size range and another in the cloud droplet size range (Fig. 2B). Furthermore, we note that as the injection rate is increased the size distribution develops a minimum at about 2 μm which is very close to $d_c$. In Fig. 2A and B, all of the size distributions have a peak in the cloud droplet size range ($d > d_c$). The behavior of this peak and its tails have been discussed extensively in ref. 20. In Fig. 2A and B, and is very close to $d_c$. In this case, essentially every injected aerosol is activated as a cloud droplet, leaving no haze droplets. A similar conclusion is applicable to case 2, although the slope of the distribution in case 2 is greater than that in case 1. This is related to the difference in mean supersaturation between these two cases, $\bar{S}_c < \bar{S}_c$. More on this is discussed in Theory and Discussion. Since nearly every injected aerosol was activated in cases 1 and 2, we conclude that at each instant, $S > S_c$. Thus, these experiments are in regime 1 (mean-dominated activation), experiencing mean-dominated activation where every injected aerosol is activated.

In cases 3 to 5 (Fig. 2B), there are a sizable number of haze droplets. Specifically, there is a peak in the haze droplet size range and a minimum near the critical size of the injected aerosols. The height of the haze peak and the magnitude at the

![Saturation ratio](https://www.pnas.org/cgi/doi/10.1073/pnas.2006426117)

**Fig. 1.** Schematic representation of the saturation ratio distributions depicting three CCN activation regimes, as shown. $S_c$ indicates the critical saturation ratio for a monodisperse aerosol. Regime 1 is mean-dominated activation, regime 2 is fluctuation-dominated activation, and regime 3 is fluctuation-dominated activation. The gray region indicates the subcritical zone of regime 2 and the light red color indicates the supercritical zone of regime 3.
Table 1. Microphysical properties of steady-state clouds

<table>
<thead>
<tr>
<th>Exp. ID</th>
<th>l, cm⁻³ min⁻¹</th>
<th>n₀, cm⁻³</th>
<th>nᶜ, cm⁻³</th>
<th>D, μm</th>
<th>σdc, μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>7</td>
<td>0.2</td>
<td>18</td>
<td>25</td>
<td>8</td>
</tr>
<tr>
<td>Case 2</td>
<td>20</td>
<td>0.4</td>
<td>71</td>
<td>18</td>
<td>8</td>
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<tr>
<td>Case 3</td>
<td>67</td>
<td>30</td>
<td>378</td>
<td>12</td>
<td>5</td>
</tr>
<tr>
<td>Case 4</td>
<td>109</td>
<td>66</td>
<td>697</td>
<td>11</td>
<td>5</td>
</tr>
<tr>
<td>Case 5</td>
<td>172</td>
<td>158</td>
<td>1,082</td>
<td>9</td>
<td>4</td>
</tr>
<tr>
<td>6 K FW</td>
<td>33</td>
<td>1,073</td>
<td>771</td>
<td>4</td>
<td>2</td>
</tr>
<tr>
<td>7 K SW</td>
<td>33</td>
<td>1,327</td>
<td>1,038</td>
<td>5</td>
<td>3</td>
</tr>
</tbody>
</table>

Shown are time-averaged injection rate l, number density of haze droplets n₀, number density of cloud droplets nᶜ, mean cloud droplet diameter D, and SD of cloud droplet diameter σdc. All of the experiments had a layer of liquid water approximately 1 cm in height at the chamber’s bottom boundary and a mean temperature of 20 ± 0.5 °C. Temperature difference between the bottom and top plate, ΔT: cases 1 to 5, 18 °C; 6 K FW, 6 °C; and 7 K SW, 7 °C. Abbreviations: Exp. ID, experiment identification number; FW, fresh water; SW, salt water. (See Experimental Observations and SI Appendix for details.)

minimum increase with the injection rate, unlike in the experiments in cases 1 and 2 where the concentration of haze droplets is negligible. It appears that as the injection rate is increased from cases 1–2 to cases 3–5, the aerosol–cloud system undergoes a qualitative transition. It is important to note that the classical ideas based on mean saturation ratio alone cannot explain the coexistence of both activated cloud droplets and unactivated haze droplets formed from monodisperse aerosols with uniform chemical composition and thus call for further discussion. Since not even aerosol was activated in these experiments, we conclude that at least a fraction of the saturation ratio distribution is subcritical (S < Sc) which is essential to maintain a steady haze droplet population. Thus, the cases in Fig. 2B are in regime 2 (fluctuation-influenced activation), and the observations therefore suggest that the fluctuations in saturation ratio are important in determining the concentration of activated cloud droplets and, as a result, the concentration of interstitial haze droplets.

We also conducted additional experiments at a lower temperature gradient compared to the experiments in Fig. 2 A and B to reduce the mean saturation ratio. These experiments were conducted at two different temperature gradients such that the mean source contribution to supersaturation was kept the same in both cases by modulating the vapor pressure at the bottom boundary. Thus, different combinations of vapor pressure and temperature gradients allow us to maintain the same mean saturation ratio, but alter its fluctuation intensity. We discuss the details of this procedure in SI Appendix. Fig. 2C shows the size distribution of the droplets from these experiments. The experiment with higher temperature gradient (7 K salt water [7 K SW]) has higher turbulence intensity and therefore higher variability in saturation ratio. Consistent with the conceptual picture of regime 3, the higher-turbulence case has a broader size distribution with a greater number of cloud and haze droplets compared to the pure water experiment (6 K fresh water [6 K FW]), although both the experiments have the same injection rate (Table 1).

Interestingly, in these experiments we observe only one peak in the haze droplet size range, and also we observe no minimum in the neighborhood of the critical diameter unlike in the experiments in Fig. 2B. These observations indicate yet another regime change between Fig. 2 B and C and thus should be in regime 3, fluctuation-dominated activation. In Theory and Discussion we provide a detailed explanation for why these experiments are in regime 3.

Before we start interpreting these observations it is important to understand how the supersaturation s = S – 1 in the experiment varies from case to case. The evolution equation for supersaturation can be written as a combination of sink and source terms where the sink is due to condensation to the droplet population and the source is from the flux of water vapor from the bottom to the top boundaries, which is proportional to the temperature difference across the two plates. In the absence of cloud droplets the boundaries act as both source and sink to supersaturation. The supersaturation is described in the same way as for natural clouds, where the source term could be from the vertical updraft (33); i.e.,

\[ \frac{ds}{dt} = \text{source} - s \tau_c. \]

where \( \tau_c = (2\pi n_c D_s)^{-1} \) is the phase relaxation time, \( n_c \) is the number density of cloud droplets, \( D_s \) is the effective diffusivity of water vapor in air, and \( D \) is the mean diameter of cloud droplets. Under steady-state conditions, \( \tau \propto (n_c D)^{-1} \). For the experiments shown in Fig. 2 A and B, the source for mean supersaturation is nearly constant as the temperature gradient is fixed and \( n_c D \) increases as the injection rate \( I \) increases. Thus, in the experiments reported in Fig. 2 A and B, the mean supersaturation

Fig. 2. (A and B) Size distribution \( \bar{F} = \frac{dN}{dD} \) of droplets observed under steady-state conditions for five different aerosol injection rates. Injection rates averaged over the volume of the chamber (number per cubic centimeter per minute) are varied over five levels: l. In A the blue curve is 7 and the red curve is 20, and in B the yellow, purple, and green curves are 67, 109, and 172, respectively. The temperature difference \( \Delta T \) between the top and bottom plate is 18 °C, and mean temperature \( T_{\text{mean}} \) is 20 °C. (C) For two experiments with the same mean supersaturation forcing but with different levels of supersaturation fluctuations. Blue, \( \Delta T = 6 \) K and pure water boundary at the bottom; red, \( \Delta T = 7 \) K and salt water boundary at the bottom. Distributions in A correspond to mean-dominated activation (regime 1), those in B to fluctuation-influenced activation (regime 2), and those in C to fluctuation-dominated activation (regime 3).
decreases from case 1 to case 5. And in the experiments reported in Fig. 2C, the mean source contribution to supersaturation was matched between the two experiments, although significantly lower than in the experiments in Fig. 2A and B. We now develop a theoretical model to explain these observations in the next section.

Theory and Discussion

In this section, we provide an Eulerian framework for studying the activation of cloud droplets. Based on the stochastic condensation framework (e.g., refs. 34 and 35), the instantaneous evolution equation for the size distribution $F(r, x)$ of the droplets (including both activated cloud droplets and haze droplets) is written as

$$\frac{\partial F}{\partial t} = -\nabla \cdot (F \mathbf{u}) + \frac{\partial}{\partial z} (w_d F) - \frac{\partial}{\partial r} (F \dot{r}) + I \delta (r - r_i, x - x_i),$$

where $r$ is the radius of the droplet, $u$ is the local velocity vector, $w_d(r)$ is the settling velocity, $\dot{r}$ is the growth rate of the droplets of size $r$, and $I$ is the injection rate of dry aerosols of size $r_i$ into a volume of cloud droplets at the location $x$. The instantaneous flow variables and size distribution fluctuate in a turbulent environment. Using Reynolds decomposition, each of these variables can be decomposed into a mean and a fluctuation, represented with an overbar and a prime, respectively; e.g., $F = \bar{F} + F'$.

Thus, the Reynolds averaged size distribution equation is written as

$$\frac{\partial \bar{F}}{\partial t} = -\nabla \cdot (\bar{F} \mathbf{u}) - \nabla \cdot (\bar{F} \dot{r}) + \frac{\partial}{\partial z} (w_d \bar{F}) - \frac{\partial}{\partial r} (\bar{F} \dot{r}') + I \delta (r - r_i, x - x_i).$$

The terms on the right side of Eq. 3 represent the various contributions to the size distribution. The first five terms are mean-advective transport, turbulent transport, transport due to sedimentation, growth due to mean supersaturation, and growth due to supersaturation fluctuation, respectively. The fluctuation covariance term $\bar{F}' \dot{r}'$, is the Reynolds transport term in “radius space” and is analogous to the Reynolds stress term in turbulent evolution equation for the size distribution $\bar{F}(r, x)$ of the droplets (including both activated cloud droplets and haze droplets) is written as

$$\frac{\partial \bar{F}}{\partial t} = -\nabla \cdot (\bar{F} \mathbf{u}) - \nabla \cdot (\bar{F} \dot{r}) + \frac{\partial}{\partial z} (w_d \bar{F}) - \frac{\partial}{\partial r} (\bar{F} \dot{r}') + I \delta (r - r_i, x - x_i).$$

and $\dot{r}$ is the growth rate of the droplets of size $r$, and $I$ is the injection rate of dry aerosols of size $r_i$ into a volume of cloud droplets at the location $x$. The instantaneous flow variables and size distribution fluctuate in a turbulent environment. Using Reynolds decomposition, each of these variables can be decomposed into a mean and a fluctuation, represented with an overbar and a prime, respectively; e.g., $F = \bar{F} + F'$. Thus, the Reynolds averaged size distribution equation is written as

$$\frac{\partial \bar{F}}{\partial t} = -\nabla \cdot (\bar{F} \mathbf{u}) - \nabla \cdot (\bar{F} \dot{r}) + \frac{\partial}{\partial z} (w_d \bar{F}) - \frac{\partial}{\partial r} (\bar{F} \dot{r}') + I \delta (r - r_i, x - x_i).$$

and regime 3, as cloud droplets in a given volume. This allows us to express the mean size distribution as

$$I = F \bar{r} + \bar{F}' \dot{r}' + F \bar{\varepsilon}.$$  

Eq. 5 shows that the net flux of droplets in radius space is a constant and suggests a dynamic balance between the rate at which the aerosols are injected and the rate at which they are processed as cloud droplets in a given volume. This allows us to express the mean size distribution as

$$\bar{F} = \frac{I - \bar{F}' \dot{r}'}{\bar{r}}.$$  

In the size range that we are interested in, the curvature and solute effects which are relevant to activation are important in the growth term in Eq. 2. The growth term can be expressed in terms of saturation ratio $S = \bar{S} + s'$ as

$$\dot{r} = \frac{1}{\tau} \left( \frac{S - (1 + \frac{s}{b} - \frac{a}{\bar{r}})}{F_k + F_d} \right) + \frac{1}{\tau} \frac{s'}{F_k + F_d} + \frac{a}{\bar{r}}.$$  

Thus, the saturation field, i.e., all injected aerosols activate as cloud droplets, similar to Fig. 2. The mean saturation ratios for these cases can be obtained using Eq. 11. And from Eq. 4, cases 3 and 4, in Fig. 2A and B, are $S > S_c$, and $S < S_c$, where $S_c$ is the critical supersaturation for the injected aerosols.

In regime 1, for mean-dominated activation, since the contribution from the turbulent covariance term is negligible, we can express the mean size distribution as

$$\bar{F} = \frac{I \bar{r}}{\bar{S} - B}.$$  

From Eq. 9, we note that the slope of the distribution decreases as the mean supersaturation increases. This suggests that the injected aerosols rapidly grow to larger sizes as $S$ increases. As a consequence, in this regime we have very few haze droplets; i.e., all injected aerosols activate as cloud droplets, similar to cases 1 and 2 in Fig. 2A. The mean saturation ratios for these cases can be obtained using Eq. 9 and the experimentally measured $\bar{F}$ (Fig. 2A). The saturation ratios for cases 1 and 2 are $S_1 \approx 1.04$ and $S_2 \approx 1.01$, respectively. Although $S_2 < S_1$, which is evident from the difference in their respective slopes in Fig. 2A, it is not low enough for the fluctuations to play a significant role in activation. From the calculated $\bar{S}$ we obtain the ratio between the supersaturation in these two cases $\frac{\Delta \bar{S}}{\bar{S}} \approx 4$.

And from Eq. 1, under steady-state conditions and assuming identical source terms in cases 1 and 2, we obtain $\frac{\Delta \bar{S}}{\bar{S}} = \frac{\tau a}{\tau \bar{r}} \approx 3$ (using data from Table 1), which is comparable to the prediction from Eq. 9. This shows that Eq. 9 is a good representation of regime 1. The size distributions obtained using Eq. 9 and the experimental parameters for cases 1 and 2 are shown in Fig. 3A.

In cases 3 to 5, there is a minimum in the size distribution near $d_c$ and a sizable concentration of haze droplets as opposed to regime 1. In regime 2, $S > S_c$, but $S$ is small enough that the covariance between the supersaturation fluctuation and the size distribution fluctuation is important. Let us consider a positive deviation in $F'$. This would consume the local supersaturation
through condensation, thus reducing it toward the equilibrium value. Now, if we consider a negative deviation in $F''$, i.e., very few droplets locally, it would result in restoring the supersaturation to near moist (no cloud droplets) conditions, thus resulting in a positive deviation in $s'$. The discussion so far suggests that $F'$ and $s'$ are negatively correlated; i.e., in this regime $F' s' < 0$. Thus, the formation of a minimum near $d_1$ must be due to the competing contributions from the covariance term and the injection term $I r$ (increasing with $r$) in Eq. 8. This would require the covariance term to decrease in magnitude with $r$ and, therefore, would explain the formation of a minimum near $d_1$ and a peak in the haze droplet size range in Fig. 2B.

We now attempt to derive the shape of the covariance term. We infer from Eq. 1 that under steady-state conditions, $s \propto (n, \tau)^{-1}$. If we consider a hypothetical situation with monodisperse cloud droplets of size $r$, then the product of number concentration and supersaturation would decrease as $r^{-1}$ as the radius increases. Similarly, if we consider a perturbed system of monodisperse droplets, then the magnitude of $F' s'$ should also decrease with radius as $r^{-1}$. Let us consider an instant where $s' < 0$ locally. Since in this regime $F' s' < 0$, the negative fluctuation in $s'$ is on average an outcome of a positive fluctuation in $F'$ and should be inversely proportional to the increase in cloud droplet numbers and their radius. This suggests that the fluctuation in $F'$ required to create an equivalent negative deviation in $s$ should decrease with $r$. Thus, we deduce that $F' s'$ in Eq. 8 can be expressed as $C_1/r$, where $C_1 > 0$ and is a function of $s'$ and the turbulence intensity in the system. Although the discussion so far is for a monodisperse cloud, the qualitative behavior of a polydisperse cloud should be similar. Thus, in regime 2, the mean size distribution can be expressed as

$$F = \frac{I r - \xi (C_1/r)}{\xi (S - B)}. \tag{10}$$

The shape of $F$ obtained in this regime using Eq. 10 is shown in Fig. 3B. It should be noted that we have not used the parameters from the experiments as the values of $F'$ and $F' s'$ are unknowns. We thus focus on the qualitative behavior of the predicted distribution and the experiments. Fig. 3B shows the shape of the distribution for three different values of the covariance coefficient $C_1$ for a fixed $S$ and injection rate. We see a peak in the distribution in the haze droplet size range and also a minimum in the distributions. We note that as the covariance term increases in magnitude for a fixed $S$, the concentration of haze droplets appears to be increasing with the magnitude of $F' s'$. A negative value of $F' s'$ (since $F' s' < 0$) in Eq. 5 indicates a backward flux in number concentration from the cloud droplets to haze droplets. Although the behavior of the predicted $F$ in Fig. 3B is qualitatively similar to the experiments in Fig. 2B, there are some quantitative differences between the observations and predictions. The location of the minimum $d_{min}$, predicted by the model, is different from that of the minimum observed in the experiments. The minimum in the model is located at $d_{min} \approx 2$ to 3 times $d_1$, whereas in the experiments $d_{min} \approx d_1$. Furthermore, to test whether $d_1$ played any role in determining the location of the minimum, additional experiments were conducted with aerosols of diameter 70 nm (SI Appendix). It was observed that the location of the minimum was not strongly affected by the critical radius of the aerosols. This suggests that the formation of the minimum in the size distribution is independent of the critical diameter of the injected aerosols and is a property of the turbulent fluctuations in the saturation ratio in this regime. A lower value of $d_{min}$ in the experiments compared to the theoretical predictions suggests that the decay of the covariance term should be faster than $d^{-1}$. This is evident by normalizing the distributions in cases 3 to 5 with the injection rate as depicted in Fig. 3C. Here, we note that the height of the minimum in regime 2 is independent of the injection rate, but the peak in the haze droplet size range is increasing with $I$. This suggests that as $S$ is decreased (from case 3 to case 5), the decay rate of the covariance term with $d$ increases. Additional analysis and experiments are required to further characterize the behavior of the covariance term. Despite these limitations the fact that in a turbulent environment all of the injected aerosols do not activate is evident from the formation of a minimum between haze and cloud droplet population and is captured by Eq. 10. Thus, the role of turbulent fluctuations in this regime is to deactivate cloud droplets (evaporate until $d < d_1$) or suppress the activation of CCN and thus increase the population of haze droplets in a mean supersaturated environment. This indicates that a certain fraction of the saturation ratio distribution is subsaturated or at least subcritical.

In regime 3, $S < S_c$, and $F' s'$ is an important term like in regime 2, but with very different behavior. Since $S < S_c$, the term $S - B$ in Eq. 8 would be negative in a certain range of $r$. In this range of $r$, the only admissible solution would be if $\xi F' s' > I r > 0$, which is in stark contrast to the behavior of the covariance term in regime 2. The requirement of a positive covariance...
suggests that this size range can be populated only if there is a positive fluctuation in $S$, which is consistent with the fact that $\mathcal{S}$ is subcritical. Thus, in this regime the positive fluctuations in saturation ratio play the prominent role in activating the aerosols as droplets. If we consider the saturation ratio distribution, this would suggest that only a fraction of the distribution is supersaturated. These supersaturated fluctuations provide the additional impetus required to overcome the growth barrier potential $B$. In the size range where $\mathcal{S} - B > 0$, similar to regime 2, we expect $\mathcal{F}S_\mathcal{S} < 0$. Compared to regime 2, a larger fraction of the injected aerosols are unactivated haze droplets in this regime. This is due to the fact that haze droplet populations have contributions from both mean saturation ratio and turbulent fluctuations. No additional information about the shape of $\mathcal{F}S_\mathcal{S}$ can be predicted at this point.

In the size distributions shown in Fig. 2C, we have only one peak in the haze droplet size range, and also we have no minimum in the neighborhood of the critical diameter or an additional peak in the cloud droplet size range unlike in the experiments in Fig. 2. The hallmark of a cloudy system with $\mathcal{S} > S_0$ is the formation of a peak in the cloud droplet size range. The slope of the size distribution in the cloud droplet size range of interest is inversely proportional to the mean supersaturation in the system (Eq. 8). The absence of a peak in the cloud droplet size range suggests that $\mathcal{S} < S_0$. This implies that the cloud droplets were activated through positive fluctuations in saturation ratio. Thus, the experiments shown in Fig. 2C are in regime 3. We have two experiments with the same mean supersaturation forcing, but different supersaturation fluctuation intensity. The experiment with higher temperature gradient has higher variability in $S$. This experiment has a broader size distribution with a greater number of cloud and haze droplets compared to the lower-gradient experiment, although both the experiments have the same aerosol injection rate. The higher variability in turbulent fluctuations would suggest a higher value for the covariance term compared to the 6 K FW experiment, thus increasing the flux to both haze and cloud droplets, as the covariance term is positive in the cloud droplet size range and negative in the haze droplet size range. These experiments are a part of a unique set of controlled experiments that shows the activation of CCN and the broadening of the cloud droplet size distribution under subcritical conditions. For the aerosols in the experiments, $S_e - 1 \approx 0.1\% < \alpha_s \approx 1\%$, and as a consequence, in regime 3 the subcritical regime can be considered to be subsaturated. Furthermore, in the experiments in this regime, the absence of a peak in the cloud droplet size range indicates that $\mathcal{S} < 1$.

The 7 K SW experiment has higher $n_o$ and $\mathcal{d}$ compared to the 6 K FW experiment. Since $\mathcal{S} < 1$ in these experiments, the source term in Eq. 1 on an average is negative; i.e., the source term in Eq. 1 is considered as a sink for supersaturation. Thus, the formation of cloud droplets due to the supercritical fluctuations counters the sink term in Eq. 1 through the evaporation of cloud droplets. It follows that the mean saturation ratio in a cloudy case would be higher than the cloud free case.

In regimes 2 and 3 there are instances when $S < S_c$. The presence of haze droplet populations in these regimes is indicative of equilibrium solutions in the stable region of the Köhler curve. For a time-varying saturation ratio, however, the kinetics of droplet growth can limit the response of a haze droplet and its ability to activate as a cloud droplet (36). The model presented here is based on an Eulerian framework, which has the advantage of being consistent with the measurements. The limitation of this model is the inability to make predictions for the time history of an individual particle and its response to supersaturation. For instance, the time-scale $\tau$ for a particle to respond to the supersaturation fluctuations can be expressed as $\tau = r/(dr/dt) \propto r^2/(S - B)$. This requires the knowledge of the history and the nature of the supersaturation (amplitude, correlation, etc.) experienced by the particle. In the current framework this information is stored in the $\mathcal{F}S_\mathcal{S}$ term in Eq. 8. Additional numerical simulations are required to carry out a Lagrangian analysis and are part of future work.

**Atmospheric Implications**

The theoretical model discussed in the previous section is not specific to the experiments discussed here. It utilizes the stochastic condensation framework, which is general, and can therefore be extended to the atmospheric context. The theoretical approach is successfully able to describe essential features of the experimental observations, including both forward and backward activation fluxes in the presence of supersaturation fluctuations. It therefore provides a basis for the development of physically based parameterizations of CCN activation in turbulent clouds. In the theory, the injection rate $I$ can be interpreted as the rate at which new aerosols are entrained into or created in a given cloud volume, and the source and removal terms can be adapted to the specific circumstances. The theory also points to the importance of quantifying the Reynolds flux $\mathcal{F}S_\mathcal{S}$. It is that closure problem that will identify under what conditions the three regimes, from mean dominated to fluctuation dominated, are expected to occur in natural clouds. The experiments presented here are in a steady state, which facilitates the comparison to the theoretical framework. Atmospheric clouds, while sometimes in approximate steady state (e.g., stratuscumulus), are also often transient in nature (e.g., cumulus). Even in the transient case, however, a quasi-equilibrium supersaturation is established for the large-scale fluctuations that are slow compared to the phase relaxation time, resulting in a dynamic balance between updraft and cloud droplet growth (14, 33). The mid- to small-scale fluctuations are therefore of most interest, and these are captured in the chamber experiments. For example, the process of activation of a CCN is steady from the perspective of individual aerosols, and the typical time scales for activation are within the range of turbulence time scales in our experiments (18, 36). Furthermore, in clouds where the quasi-steady-state approximation is not valid (e.g., close to the cloud edges) the time averaging used here can be replaced with spatial averaging or ensemble averaging, which is a standard procedure in turbulent flows. Thus, the insights gained from these steady-state experiments are applicable to the activation processes in atmospheric clouds.

Field measurements show that saturation ratio is a fluctuating quantity and hence may play an important role in the activation/deactivation of cloud droplets (14, 22). For example, in an entraining cloud system like a cumulus cloud, the three regimes can be identified depending on the distance from the cloud core (e.g., in analogy with the cloud entrainment regions identified by ref. 29). In the cloud core, the effects of mixing and entrainment are negligible and thus can be considered to be in regime 1 where the entire saturation ratio probability density function...
is supersaturated. Near the edge of the cloud where the effects of entrainment are strongly felt by the cloud droplets, the system could be said to be in regime 3. The significant entrainment of dry air may saturate the cloud parcel on an average, but the fluctuations can activate or deactivate cloud droplets. The volume between the cloud core and the edge can be said to be regime 2 where the effects of entrainment are important to form a locally subsaturated environment responsible for the deactivation of cloud droplets, but the mean saturation ratio is above $S_c$. In this region, one may observe the presence of haze droplets. Similarly, it will be of interest to extend the parcel-based studies of aerosol-limited versus updraft-limited activation and growth regimes (8, 10), to account for turbulent fluctuations. One can anticipate that cloud regions with strong updrafts will experience mean-dominated activation, whereas clouds with more isotropic vertical velocity distributions such as stratocumulus may experience fluctuation-influenced activation.

It should be emphasized that a minimum between unactivated haze droplets and cloud droplets is present for a realistic CCN spectrum. This was attributed to the inhomogeneity in the chemical composition and polydispersity (37) and to polydispersity and supersaturation fluctuations (22). This usual natural condition disguises the role of fluctuations and the resulting two-way flux of aerosols considered here. From our controlled laboratory observations, it appears that turbulent fluctuations in saturation ratio have the potential to mimic the effects of chemical inhomogeneity and polydispersity in the CCN spectrum. Thus, assessing the importance of chemical heterogeneity and polydispersity of the CCN in a turbulent environment is an opportunity for future work. Our results highlight the utility of idealized experiments and models for understanding the relevance in the atmospheric context. An example would be the similar distinction between haze and cloud droplets in a recent simulation of a turbulent entraining parcel system (ref. 18, figure 4b) using the Lagrangian superdroplet method, with the objective of simulating cumulus clouds. These simulations show a clear minimum between the haze and cloud droplet populations and are similar to the regime 2 experiments in Fig. 2. Perhaps the most surprising observation is the laboratory demonstration that steady-state clouds can be formed under subsaturated conditions. This shows that supersaturation fluctuations can play a dominant role in the activation of cloud droplets and their subsequent growth in a turbulent environment like atmospheric clouds. The formation of cloud droplets under these conditions has been explored theoretically and numerically in the past under various limiting assumptions (15, 17, 38). One can imagine scenarios such as entrainment zones at a stratocumulus cloud top where the mean saturation ratio would be less than unity, but activation of interstitial or entrained CCN by fluctuations could sustain cloud formation. Fluctuation-dominant activation may also be relevant for weakly turbulent fog (39), where mean forcing of supersaturation is weak. The size distribution measurements in regime 3 reported here are similar to the field measurements close to the boundaries of cumulus clouds and fogs (ref. 40, figures 1b and 12b). In these measurements no peak was observed in the cloud droplet size range, which is suggestive of a fluctuation-dominant activation mechanism. In these situations the shape of the supersaturation distribution will be of great importance (41). The sensitivity to the shape of distribution tails may have compelling implications for turbulence intermittency. Furthermore, in clouds with low (regime 2) and negative (regime 3) supersaturation, positive fluctuations may play an important role in converting Aitken mode aerosols to large CCN through the absorption of SO$_2$ through in-cloud processing (42).

The experiments described here and the associated theoretical analysis, as well as the comparisons with existing simulation data (15, 18), suggest that there is a need to move beyond the uniform-parcel view of CCN activation. These turbulence-based approaches will improve our understanding of the interaction between aerosols, cloud droplets, and turbulence. Furthermore, these observations show that turbulent fluctuations are an integral part of cloud microphysics and should be represented appropriately in cloud models to improve their predictive capability.

Data used in this paper are available at https://digitalcommons.mtu.edu/data-files/33 (30).

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