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Broadening of Cloud Droplet Size Distributions and Warm Rain Initiation

Associated with Turbulence: An Overview

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Abstract

In the study of warm clouds, there are many outstanding questions. Cloud droplet size distributions are much wider, and warm rain is initiated in a shorter time and with a shallower cloud depth than theoretical expectations. This review summarizes the studies related to the effects of turbulent fluctuations and turbulent entrainment-mixing on the broadening of droplet size distributions and warm rain initiation, including observational, laboratorial, numerical, and theoretical achievements. Particular attention is paid to studies by Chinese scientists since the 1950s, since most results have been published in Chinese. The review reveals that high-resolution observations and simulations, and laboratory experiments are needed because knowledge of the detailed physical processes involved in the effects of turbulence and entrainment-mixing on cloud microphysics still remains elusive. The effects of turbulent fluctuations and entrainment-mixing processes have been unrealistically separated in most theoretical studies. They could be unified by further advancement of a systems theory into a predictive theory. Developing parameterizations for the effects of fluctuations and entrainment-mixing processes is still in its infancy, and more studies are warranted.

KEYWORDS: Cloud droplet size distribution, warm rain, turbulent fluctuations, entrainment-mixing, systems theory
1. Introduction

Warm rain refers to rain without the presence of the ice phase in clouds. Lau and Wu (2003) pointed out that warm rain accounts for 31% of the total rain amount and 72% of the total rain area in the tropics. Warm rain initiation affects the cloud life cycle and cloud macro and microphysics. Since clouds influence radiation (Zhao and Ishizaka 2004; Xin and Li 2016; Fu and Lei 2017; Zhang and Chen 2017), parameterizations of warm rain initiation are important for evaluating cloud-radiation-climate feedbacks. Understanding warm rain initiation and cloud droplet size distribution broadening (spectral broadening hereafter) is one of the main challenges of cloud physics (Hu 1979; Beard and Ochs 1993; Sun et al. 2012; Blyth et al. 2013; Cooper et al. 2013; Dagan et al. 2015; Selvam 2015; Seifert and Onishi 2016). First, some warm clouds are often observed to rain within a shorter time than theoretical expectations (Zhou and Gu 1963; McGraw and Liu 2004). Second, the theoretical cloud depth necessary to produce raindrops is greater than 2 km, but clouds with depths of less than 1 km have been observed to rain (Zhou and Gu 1963). Third, according to conventional condensation theory (Wallace and Hobbs 2006), cloud droplet size distributions are much narrower than those from in-situ observations (e.g., Lu et al. 2013c). Fourth, it is still unclear how embryonic rains around 20 µm in radius form.

This review focuses on the effects of turbulence on the broadening of cloud droplet size distributions and warm rain initiation. Due to the space limit, other mechanisms, like the effects of giant cloud condensation nuclei (e.g., Feingold et al.
1999; Yin et al. 2000; Fang and Zheng 2011; Yang et al. 2012; Guo et al. 2015a), are not included in this review. Besides, many Chinese scientists have made contributions to this topic since the 1950s, but most of those studies were published in Chinese and are not readily accessible to the majority of the international communities in this field. So, a purpose of this review is to introduce these studies in English.

The rest of the paper is organized as follows. Section 2 presents the effects of fluctuations on condensation and collision-coalescence, including the progress of studies conducted in and outside China. In section 3, we address turbulent entrainment-mixing mechanisms, including their definitions, methods (microphysics and dynamics), and parameterizations. In section 4, we give a brief overview of the systems theory and its application. Concluding remarks are presented in section 5.

2. Effects of turbulent fluctuations on condensation and collision-coalescence

2.1 Studies in China

Studies on cloud/fog and precipitation physics in China began in 1958 to meet the needs of weather modification (Gu 1963). As pointed out by Gu (1962), size distribution formation is not only a key topic in cloud/fog microphysics, but also critical in the examination of the effects of macro conditions on cloud/fog formation and development and the interactions between macro and microphysical processes. Chinese scientists conducted in-situ observations on Hengshan Mountain (112.86 °E, 27.25 °N) in Hunan Province and Taishan Mountain (117.13 °E, 36.18 °N) in
Shandong Province. Xu (1964) designed an instrument to measure cloud/fog droplet size distributions. Air containing cloud/fog droplets was drawn by a fan through the instrument where droplets were impacted on sampling plates. Photos of the plates were taken continuously and each plate provided one droplet size distribution. He found that the cloud droplet number concentration in stratocumulus clouds on Hengshan Mountain in September 1960 had fluctuation amplitudes in the range of 30%–100% and a fluctuation scale of 1–6 m over which the number concentration could be quite different. The cloud droplet size distributions also fluctuated and the fluctuation amplitude decreased with increasing sampling volume. Zhan et al. (1965) observed cloud droplet size distributions on Hengshan (April–May 1962) and Taishan (July–August 1962) Mountains. They realized that part of fluctuations were due to small sampling volume and removed this part of the fluctuations from the total fluctuations in the observed data. They found that cloud droplet size distributions were broader for more significant fluctuations even when the average liquid water content was the same, suggesting that fluctuations in liquid water content were associated with spectral broadening. Number concentrations with different droplet sizes had different fluctuation strengths: large droplets had the largest fluctuations and medium-sized droplets had the smallest fluctuations. Another important characteristic of fluctuations of droplet size distributions was that the size distributions could be bimodal. Hong and Huang (1965) concluded that bimodal distributions were found more frequently with stronger rain intensity and intensity variation, which could be related to collision-coalescence. Zhu et al. (1965) found that bimodal distributions
formed and disappeared gradually with the passage of a front. Lin and Gu (1965) examined the mechanism for the formation of the bimodal distribution and pointed out that turbulent collision-coalescence could be an important reason. Mao and Gu (1963) analyzed the observations on Hengshan Mountain and found that eight cases included super adiabatic liquid water content. Through theoretical analysis, they further revealed that when the vertical velocity decreased with increasing height, droplets would accumulate near the cloud base, which were responsible for the appearance of super adiabatic liquid water content.

In addition to observational studies, Chinese scientists also theoretically derived equations used to study the effects of fluctuations on spectral broadening. Gu (1962) derived and listed 20 equations in cloud/fog physics and discussed how fluctuations accelerated the condensation and collision-coalescence of droplets. Chinese scientists realized that many quantities in cloud/fog fluctuate (Jaw 1966) and the key studies are briefly summarized here.

(1) Fluctuations of supersaturation. Zhou (1963) developed a stochastic theory for cloud droplet growth and a microphysical mechanism for precipitation in warm clouds. He assumed that superstation fluctuations were in the form of a Gaussian distribution and found that condensation with fluctuating supersaturation and turbulent electric collision-coalescence were both important for the formation of droplets in the range of 1–20 µm (radius). Similar conclusions were drawn in Xu et al. (1966). Wen and Zhu (1987) investigated the condensation process under fluctuations of supersaturation in different clouds. For tropical warm clouds, the time needed for
droplets to grow from 20 µm to 25 µm in radius was short, even with low fluctuations; for mid-latitude warm clouds, the time was short only when fluctuations were strong.

(2) Fluctuations of number concentration and liquid water content. The growth of droplet size due to collision-coalescence between different droplets is closely related to the number concentration and liquid water content where collision-coalescence occurs. Assuming the cloud droplet size distribution was bimodal and the number concentration had fluctuations, Gu and Zhan (1962) found that collision-coalescence could produce droplets of 50 µm in radius or even larger than 100 µm. Wen and his coauthors found that fluctuations in liquid water content were feasible for the formation of large cloud droplets by collision-coalescence, no matter whether the fluctuation time/space scale was large or small (Wen 1964, 1966; Xu et al. 1966).

(3) Fluctuations of vertical velocity. This type of fluctuation could affect how droplets move in clouds. Based on theoretical analysis, Xu and Gu (1963) found that droplets facing different vertical velocities would have different destinies and trajectories due to vertical velocity fluctuations. Some droplets were transported out of clouds rapidly, while others could cycle several times in clouds to produce large raindrops, which explained why shallow clouds with small cloud depths could produce rain. Xu (1963) further considered the effects of vertical velocity fluctuations and the vertical profile of vertical velocity. Assuming the vertical velocity linearly decreased with increasing height, she found that the cloud depth necessary for rain formation was smaller than that assuming that vertical velocity was distributed uniformly in the vertical direction.
(4) Cell structure. Cumulus clouds are often observed to be composed of convective cells (Xu and Li 1980). Xu and Li (1980) assumed that liquid water content and vertical velocity were larger inside cells than outside cells, i.e., fluctuations of both liquid water content and vertical velocity. Raindrops tended to appear with stronger fluctuations in cells, appearing in more cells and larger cell scales. Li (1963) further examined the inhomogeneity of both vertical velocity and horizontal winds, in which cells transported cloud droplets back and forth in clouds. As a result, the cloud depth for precipitation formation decreased. Because of the importance of turbulent collision-coalescence, Xiao et al. (1988) included it in a one-dimensional cloud model, and simulations showed that turbulent collision-coalescence played a significant role in productions of big droplets.

In summary, by relating spectral broadening to various turbulent fluctuations, Chinese scientists and Russian scientists (Belyayev 1961; Levin and Sedunov 1966; Sedunov 1974) were probably the first to introduce the idea of stochastic condensation into cloud physics (Liu et al. 2002a). Chinese scientists derived the equations that describe the collision-coalescence processes, accounting for fluctuations of key microphysical properties such as number concentration and liquid water content. These equations were promising for analytical study.

### 2.2 Studies outside China

Many studies focused on stochastic condensation, which attributed spectral
broadening to turbulent fluctuations of supersaturation (Buikov 1961; Cooper 1989). Khvorostyanov and Curry (1999) revised the early equations of stochastic condensation by introducing two new features: 1) consideration of supersaturation as a nonconservative substance with differentiation between macroscale and microscale supersaturation and 2) consideration of the supersaturation fluctuations of various frequencies. Shaw et al. (1998) presented a mechanism for the spectral broadening: cloud droplets were preferentially concentrated in regions of low vorticity in the turbulent flow field. Regions of high vorticity (low droplet concentration) developed higher supersaturation than regions of low vorticity (high droplet concentration). Droplets growing in regions of high vorticity experienced enhanced growth rates, allowing some droplets to grow larger than predicted by the classic theory of condensational growth. Vaillancourt et al. (2002) discussed the first direct numerical simulation (DNS) of the interaction between the turbulent flow field and cloud droplet growth by the diffusion of water vapor. It was found that as a result of increasing dissipation rate, fluctuations of supersaturation increased, but the width of the cloud droplet size distribution decreased. The result was a consequence of the decrease in the decorrelation time of supersaturation perturbations as the dissipation rate increased. This conclusion is different from those of many other studies. Celani et al. (2005) showed that the presence of an underlying turbulent velocity field induced a correlation between droplet trajectories and supersaturation, which led to both the enhancement of the droplet growth rate and to the fast spreading of the droplet size distribution. With DNS, Lanotte et al. (2009) found that spectral broadening increased
with the Reynolds number of turbulence. Using a stochastic model and DNS, Sardina et al. (2015) found that the width of cloud droplet size distribution increased proportionally to the square root of time. Using a Brownian diffusion model, McGraw and Liu (2006) revealed the mechanisms of diffusive broadening of the droplet size distribution in a turbulent field and found that the stationary size distribution follows a Weibull distribution, consistent with the result from the systems theory, reviewed in Section 4.

Besides the effects of turbulence on stochastic condensation, turbulence also exerts several effects on droplet growth by collision-coalescence (Telford 1955; Shaw 2003), including increasing the droplet relative velocity (e.g., Arenberg 1939; Pinsky and Khain 1997; Wang et al. 1998), droplet clustering (e.g., Kostinski and Shaw 2001; Shaw et al. 2002), increasing settling velocity (Dávila and Hunt 2001; Ayala et al. 2008), increasing collision efficiency/probability between droplets (e.g., Pinsky et al. 2000; Kunnen et al. 2013), etc. The four effects are explained as follows. The increased relative velocity is caused by the viscous drag and differential inertial response of the droplets to local fluid acceleration (Ayala et al. 2008). As mentioned above, droplet clustering means that inertial particles tend to accumulate in local regions of low vorticity and high strain rate due to an inertial bias (Maxey 1987; Shaw et al. 2002). The average settling velocity of a small rigid spherical particle in homogeneous turbulence, subject to the Stokes drag force, has been shown to differ from that in still fluid (Maxey 1987; Wang and Maxey 1993). The collision efficiency is sensitive to the droplet relative velocity, so in a cloud, the efficiency must account
not only for differential sedimentation, but also for relative velocities due to turbulent acceleration (Pinsky et al. 1999).

Pinsky and Khain (2002) found that an increase in the collision kernel in turbulent surroundings was an important factor in the acceleration of large droplet and raindrop formation. Falkovich et al. (2002) derived a formula for the collision rate of small heavy particles in a turbulent flow and concluded that air turbulence could substantially accelerate the appearance of large droplets that trigger rain. Besides, DNS is becoming a useful tool to determine turbulent collision kernels, and the kernels from DNS have been widely used. For example, by solving the stochastic collection equation with the use of turbulent collision kernels, Franklin (2008) found that turbulence could significantly reduce the time to produce drizzle-sized drops. A similar conclusion was reached in Wyszogrodzki et al. (2013). Franklin (2014) further found that the amount of rainwater reaching the surface was six times larger in the simulation with the turbulent microphysics scheme. Grabowski and Wang (2008) defined the turbulent speedup factor as the ratio of the rain initiation time for the turbulent collection kernel to the corresponding time for the gravitational kernel. They found that the factor was in the range of 0.75–0.85 and 0.60–0.75 for the different turbulent dissipation rates. Using a Lagrangian cloud model, Hoffmann et al. (2017) found that raindrop formation easily occurred when turbulence-induced collision enhancement was considered, with or without any extra mechanisms of spectral broadening. Hsieh et al. (2009) evaluated eight autoconversion parameterizations against integration of the Kinetic Collection Equation and found that including
turbulence effects on droplet collection increased autoconversion by factors of 1.82 and 1.24 for different clouds.

3. Turbulent entrainment-mixing mechanisms

Section 2 reviews the turbulent effects in adiabatic clouds. Turbulence can affect cloud droplet size distributions through the entrainment of dry air into clouds and mixing between clouds and dry air. Stommel (1947) was the first scientist to realize the existence of entrainment of dry air into clouds; the cloud lapse rate was not wet-adiabatic, and the vertical gradient of liquid water content was different from that derived from adiabatic theory. Studies on entrainment-mixing include, at least, how dry air is entrained into clouds, how large the entrainment rate is, and how cloud droplets respond to entrained dry air (Devenish et al. 2012; Wood 2012; Cheng et al. 2015; Guo et al. 2015b; Lu et al. 2016; Guo et al. 2017). This review only focuses on the last question.

Homogeneous/inhomogeneous entrainment-mixing mechanisms have been the most studied for entrainment-mixing investigation. Figure 1 shows the schematic diagram. The circles represent cloud droplets. After dry air is entrained into clouds, dry air and cloudy air will mix together due to turbulence, causing the evaporation of droplets. The two processes (mixing and evaporation) occur at the same time. The question is which process is faster, mixing or evaporation? The answer would be different for different clouds or even different stages in one cloud.
If mixing is much faster than evaporation, mixing occurs first and all the droplets are distributed throughout the area, including cloudy air and dry air. After mixing, all the droplets face the same saturation deficit (relative humidity minus 100%) and evaporate at the same time, thus decreasing the droplet size. Number concentration also decreases because of dilution. So, droplet size and number concentration are positively correlated. This is termed the homogeneous entrainment-mixing mechanism, which was first studied by Warner (1973). He found that if dry air was nucleus-free, mixing broadened the cloud droplet size distribution only slightly. If dry air contained nuclei, which were activated to produce droplets after mixing, the size distribution was broadened considerably but in a way that was not observed in natural clouds. In natural clouds, he found that the dispersion of the size distribution was independent of the amount of mixing. Thus, from both theory and observation, he concluded that homogeneous mixing between cloudy and dry air was unimportant in determining the size distribution, at least in the early stages of cloud growth. The discouraging results from Warner (1973) did not stop other scientists from using the concept of homogenous mixing. Mason and Jonas (1975), Lee and Pruppacher (1977), and Jonas and Mason (1982) all assumed homogeneous mixing and were able to reproduce the bimodal cloud droplet size distribution observed by Warner (1969). Recently, Yang et al (2016) showed that there was a critical height above the mixing level under the assumptions of clean environment and homogeneous mixing, and super-adiabatic droplets existed above the critical height.

If evaporation is much faster than mixing, evaporation occurs first and the
droplets closest to the dry air evaporate completely to saturate the dry air. After evaporation, mixing occurs. Because the dry air is already saturated, no droplets evaporate during mixing. As a result, the number concentration decreases significantly while the droplet size does not change. This is termed the extreme inhomogeneous entrainment-mixing mechanism. If after extreme inhomogeneous entrainment-mixing, the diluted cloud parcel is lifted upward again, then the droplets in the diluted cloud parcel will grow faster than those in the adiabatic cloud. The reason is that the number concentration is lower, the surface area for condensation is smaller, and the supersaturation is larger in the diluted cloud parcel. It is expected that the droplet size and number concentration are negatively correlated. This is termed inhomogeneous mixing with subsequent ascent. The extreme inhomogeneous entrainment-mixing mechanism was first found by Latham and Reed (1977). They performed experiments in which droplets formed by condensation were drawn slowly down a cylindrical tube of length 5 m, and they found that some droplets of all sizes were completely evaporated, while others were not significantly affected. Baker et al. (1980) assumed extreme inhomogeneous entrainment-mixing mechanism in their model and found that a small proportion of droplets could grow several times faster than adiabatic theory predicted. The calculations produced spectral shapes which agreed well with those observed in cumulus clouds by Warner (1969).

The above concept shows two extremes of entrainment-mixing mechanisms. The mechanisms in real clouds are much more complicated. Many scientists have been focusing on dissecting the real entrainment-mixing mechanisms in natural clouds,
through theoretical analysis, in-situ observations, and high-resolution simulations. Baker and Latham (1979) and Baker et al. (1980; 1984) used two timescales to quantitatively describe the speeds of two processes, mixing and evaporation, i.e., $\tau_m$ and $\tau_r$, respectively. The parameter $\tau_m$ is the time needed for dry air to be completely mixed and $\tau_r$ is the response time between cloud droplets and dry air. The ratio of the two quantities, also known as the Damköhler number, $\tau_m/\tau_r$, has been widely used in entrainment-mixing studies (Burnet and Brenguier 2007; Jeffery 2007; Andrejczuk et al. 2009; Yum et al. 2015; Pinsky et al. 2016; Yeom et al. 2017). Lehmann et al. (2009) realized that the dry eddy size used in the calculation in $\tau_m$ was often unknown. They defined a transition length for the Damköhler number equal to one. Lu et al. (2011) and Kumar et al. (2013) further calculated the ratio of the transition length to the Kolmogorov microscale; Lu et al. (2011) named it the transition scale number. A larger transition scale number indicates that mixing is more homogeneous. Besides these dynamical quantities, mixing diagrams of droplet size vs number concentration or vs liquid water content were introduced (e.g., Pawlowska et al. 2000; Burnet and Brenguier 2007; Gerber et al. 2008; Freud et al. 2011; Lu et al. 2013a; Small et al. 2013; Tölle and Krueger 2014). Based on the definitions of homogeneous/inhomogeneous mixing mechanisms described above, the dominant mechanism could be identified through aircraft observations (e.g., Lehmann et al. 2009) and numerical simulations (Andrejczuk et al. 2006; Krueger 2008; Kumar et al. 2014; Gao et al. 2017). Many observational studies suggested that the entrainment mixing process could be close to homogeneous (e.g., Jensen et al. 1985; Lu et al.
extreme inhomogeneous (e.g., Freud et al. 2011), or neither homogeneous nor extreme inhomogeneous (Lehmann et al. 2009; Lu et al. 2013b). Usually, cloud droplet size distributions are measured by the Forward Scattering Spectrometer Probe, the Cloud and Aerosol Spectrometer, or similar instruments, which require averaging over long distances. Beals et al. (2015) overcame the large-scale spatial averaging problem by using an airborne digital in-line holographic system that imaged the three-dimensional (3D) structure within cloud volumes of ~15 cm³ and provided size distributions. The measurements revealed that inhomogeneous mixing dominated, with sharp transitions between cloud and clear air properties, persisting to dissipative scales (< 1 centimeter).

To quantitatively describe how much mixing is homogeneous, the homogeneous mixing degree or similar quantities have been defined (Gerber et al. 2008; Morrison and Grabowski 2008; Lu et al. 2013b). Based on in-situ observations and numerical simulations, parameterizations of entrainment-mixing mechanisms have been developed, which could be used to diagnose instantaneous entrainment-mixing mechanisms in models (Andrejczuk et al. 2009; Lu et al. 2013b). Effects of different entrainment-mixing parameterizations on cloud microphysics have been tested. Grabowski (2006), Chosson et al. (2007), and Sławinska et al. (2008) found that cloud microphysical properties were very sensitive to the entrainment-mixing parameterizations. However, Sławinska et al. (2012), Morrison and Grabowski (2008), and Hill et al. (2009) found that the effects of mixing on cloud microphysics were not as sensitive as those found by previous studies. Thus, this topic is still controversial.
There are several other ideas of entrainment-mixing mechanisms. In entity-type entrainment mixing (Telford and Chai 1980; Telford 1996), dry air is entrained above the cloud top and behaves as a continuing entity. One entity is like a parcel/cell in the clouds. During the descent of the entity, droplets evaporate completely to saturate the entity. Afterwards, the surrounding cloudy air is further mixed into the entity. The droplet size distribution in the entity is the same as that in the surrounding cloud, but the number concentration is lower. The entity goes up when buoyancy is restored. Due to the lower number concentration, droplets in the entity can grow larger than those in the adiabatic clouds. Thus, the number of recycling times of “entity” in clouds is key to generating a broader size distribution and raindrops. A major challenge is how to determine the number of the recycling times, which may be related to turbulence intensity, thermodynamic conditions (e.g., temperature, relative humidity in dry air), and microphysical properties (e.g., liquid water content in clouds).

The idea of vertical circulation mixing (Wang et al. 2009; Yum et al. 2015) is similar. As the cloud parcel descends from the cloud top, cloud droplets evaporate until complete evaporation is reached. This height would be the local cloud base during the reascension of the parcels. If some of descending parcels are more diluted than others, the local cloud base would have different heights. These parcels would mix together during ascension, which would broaden the cloud droplet size distributions. The idea of fluctuating cloud bases is similar to that proposed by Betts (1978; 1983), whereby they analyzed the different cloud bases for different parcels.
using saturation point diagrams.

Thus, despite decades of research, the effects of entrainment-mixing mechanisms on cloud microphysical properties and cloud droplet size distribution still remain elusive, full of uncertainty. Further studies are needed through theoretical analysis, observations, and numerical simulations.

4. Systems theory

As introduced above, clouds are turbulent and cloud microphysical properties fluctuate significantly; also, turbulent entrainment-mixing has significant effects on cloud droplet size distributions. In fact, turbulence-induced fluctuations and entrainment-mixing are closely intertwined, which makes it even more difficult to understand the formation of cloud droplet size distributions. Besides turbulent fluctuations and entrainment-mixing, there are many other factors affecting cloud droplet size distributions, e.g., aerosol. This complexity inspired Liu and his group to consider cloud droplets as a system, without concern for the details of each individual droplet affected by different factors (Liu 1995; Liu et al. 1995; Liu 1997). After realizing that the molecular system and the atmospheric particle system are both multi-body and polydisperse systems with stochastic fluctuations, Liu et al. (1995) applied Shannon’s maximum entropy principle to the cloud droplet system to develop a systems theory. Shannon’s maximum entropy principle, as a theory for studying stochastic multi-body systems, has been successfully applied to various complex
systems. If a stochastic system is governed by some restrictive conditions and characterized by a continuous random variable $y$ with probability density function $\rho(y)$, $\rho(y)$ has many possible solutions, due to the fluctuations, and $\rho(y)$ corresponds to Shannon’s entropy $H(y)$. Shannon’s maximum entropy principle indicates that when $H(y)$ reaches its maximum, its corresponding $\rho(y)$ will have the largest possibility of occurrence. The systems theory predicts that the most probable size distribution is a Weibull distribution. Liu (1995, 1997) further proposed a general framework to unify atmospheric particle systems, including the unification of atmospheric particle shapes into self-similar fractals and the generalization from self-similar to self-affine particles. Liu and Hallett (1997) further extended the systems theory for cloud droplet size distributions from clouds with conserved liquid water content to other cases, accounting for the degree of entrainment-mixing. From the generalized systems theory, a general “1/3” power law was then formulated between the effective radius and the ratio of liquid water content to number concentration.

Besides the most probable size distribution, Liu and Hallett (1998) obtained the least probable distribution (a delta distribution) by studying the functional relationship between a cloud droplet size distribution and the corresponding energy change to form such a droplet population. The Weibull distribution is much wider than the delta distribution, but theoretical analysis in Liu et al. (2002a) indicated that the Weibull distribution approached the delta distribution with decreasing fluctuations, because the width of the cloud droplet size distribution was closely related to fluctuation levels.
in clouds and increased with increasing fluctuations. Liu et al. (2002b) generalized the systems theory to allow for varying fluctuations. The theory indicates that there exists an important characteristic scale (saturation scale), below which droplet size distributions strongly depend on the scale over which they are sampled. However, beyond this scale, droplet size distributions do not change with further increases in the averaging scale. An important prediction of the systems theory is that the observed discrepancy between the observed and predicted droplet size distribution may lie in the scale mismatch: in turbulent clouds, the observed size distribution represents the average over large scales and, thus, is closer to the broader most probable size distribution, whereas the delta function, like a narrow distribution, corresponds to the least probable distribution that is hardly observed, even at small local scales. The systems theory also provides a theoretical basis for considering microphysical parameterizations for large scale models as a general issue of statistical physics (Liu et al. 2002a).

A similar systems theory has been developed to explain the rain initiation by combining the systems theory for cloud droplet size distribution and nucleation theory (McGraw and Liu 2003; 2004). The theory treats rain formation as a barrier-crossing process and is used to derive a theoretical expression for the critical droplet radius that differentiates between cloud droplets and small raindrops (Liu et al. 2004).
5. Concluding remarks

We review the studies on spectral broadening and rain initiation with special attention paid to the early studies by Chinese scientists because they were written in Chinese and, thus, are not accessible to most non-Chinese scientists.

Chinese scientists carried out field campaigns over mountains to measure cloud droplet size distributions, using self-made instruments in the 1950s and 1960s. They observed fluctuations of cloud microphysical properties and cloud droplet size distributions. They also observed bimodal distributions and super adiabatic liquid water content. Compared with observations, they focused more on the theoretical work on cloud physics, in which equations governing cloud physics were derived. Based on the equations, the effects of fluctuations on stochastic condensation and collision-coalescence were studied, including fluctuations of supersaturation, number concentration, liquid water content, vertical velocity, and existence of cell structures. Turbulent fluctuations were found to be important for spectral broadening. This topic has also been widely studied throughout the world. Particularly with high-resolution DNS simulations, many studies concluded that stochastic condensation in a turbulent environment contributed to spectral broadening, while opposite effects were found in others. Different from stochastic condensation, there is more consensus that turbulent fluctuations play significant roles in the collision-coalescence process because turbulence increases the droplet relative velocity, collision efficiency/probability between droplets, and settling velocity, as well as causes droplet clustering.

Besides turbulent fluctuations, turbulence also causes entrainment of dry air into
clouds, and entrainment-mixing affects cloud microphysics. Several types of turbulent entrainment-mixing mechanisms are reviewed, such as homogeneous/inhomogeneous entrainment-mixing, entity-type entrainment-mixing, and vertical circulation mixing. The mechanism most studied is the homogeneous/inhomogeneous mixing. Different dynamical quantities—Damkohler number, transition length, and transition scale number—were defined to distinguish homogeneous and inhomogeneous mixing mechanisms. Mixing diagrams of droplet size vs number concentration or liquid water content were introduced to identify different mixing mechanisms from the point of view of microphysics. Homogeneous mixing degrees were defined to tell how much of mixing is homogeneous. How different mixing mechanisms affect cloud microphysics were tested in models.

As a theory for unifying turbulent fluctuations, entrainment-mixing and other factors, the systems theory provides a theoretical framework for explaining the shapes of cloud droplet size distributions. The theory predicts that the most probable size distribution is the Weibull distribution. The Weibull distribution approaches the delta distribution (least probable distribution) if turbulent fluctuations decrease. Cloud droplet size distributions are also scale-dependent; there is a saturation scale, over which the size distributions become stable.

Several challenges remain to be addressed. First, the detailed physical processes about the effects of fluctuations and entrainment-mixing on cloud droplet size distributions are still not clear, so more accurate and high-resolution observations, laboratory experiments, and high-resolution numerical simulations are needed. In
addition, there is room for improvement in technologies for measuring temperature, water vapor, and vertical velocity. Recently, Siebert and Shaw (2017) studied supersaturation fluctuations during the early stages of cumulus formation. The Pi cloud chamber developed at the Michigan Technological University has shown its advantages in examining the interactions between supersaturation fluctuations and aerosol, and their effects on spectral broadening (Chandrakar et al. 2016; Chang et al. 2016; Chandrakar et al. 2017). On the modeling front, models such as DNS (Gao et al. 2017), Explicit Mixing Parcel Model (EMPM) (Tölle and Krueger 2014), and large-eddy simulation models, etc., can be used to study the mechanisms of how fluctuations and entrainment-mixing affect cloud droplet size distributions and the scale dependence of the mechanisms (Lu et al. 2014b). Second, the effects of turbulent fluctuations and entrainment-mixing processes have been unrealistically separated in most theoretical studies, with the possible exception of the systems theory (Liu et al. 2002a). However, the current systems theory is largely descriptive, and, thus, further advancement into a predictive theory poses a challenge as well. Third, clouds play a significant role in weather forecasting and climate prediction. However, cloud parameterizations in models are far from complete. One critical problem hindering accurate parameterizations is the effects of fluctuations and entrainment-mixing on cloud physics. More development/improvement of parameterizations of fluctuations and entrainment-mixing, and testing in large-eddy simulations, cloud-resolving models, and even large-scale models are necessary to see how the new parameterizations affect cloud/precipitation and radiation.
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Caption List

Figure 1 Schematic diagram of homogeneous/inhomogeneous entrainment-mixing mechanisms (e.g., Baker et al. 1980; Yum 1998; Lu et al. 2011). See text for the explanations.
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