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Cloud Parameterizations, Cloud Physics, and Their Connections: An Overview

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ABSTRACT

This paper consists of three parts. The first part is concerned with the parameterization of cloud microphysics in climate models. We demonstrate the crucial importance of spectral dispersion of the cloud droplet size distribution in determining radiative properties of clouds (e.g., effective radius), and underline the necessity of specifying spectral dispersion in the parameterization of cloud microphysics. It is argued that the inclusion of spectral dispersion makes the issue of cloud parameterization essentially equivalent to that of the droplet size distribution function, bringing cloud parameterization to the forefront of cloud physics. The second part is concerned with theoretical investigations into the spectral shape of droplet size distributions in cloud physics. After briefly reviewing the mainstream theories (including entrainment and mixing theories, and stochastic theories), we discuss their deficiencies and the need for a paradigm shift from reductionist approaches to systems approaches. A systems theory that has recently been formulated by utilizing ideas from statistical physics and information theory is discussed, along with the major results derived from it. It is shown that the systems formalism not only easily explains many puzzles that have been frustrating the mainstream theories, but also reveals such new phenomena as scale-dependence of cloud droplet size distributions. The third part is concerned with the potential applications of the systems theory to the specification of spectral dispersion in terms of predictable variables and scale-dependence under different fluctuating environments.

1. INTRODUCTION

Clouds have attracted great interests from humans, as implied by the poem by Vollie Cotton [1], *"Clouds are pictures in the sky/They stir the soul/ they please the eye/They bless the thirsty earth with rain/which nurtures* *life from cell to brain/But no! They are demons, dark and dire/hurling hail, wind, flood, and fire/Killing, scarring, cruel masters/Of destruction and disasters /Clouds have such diversity/Now blessed, now cursed/the best, the worst/But where would life without them be?"* Clouds, which play a crucial role in regulating the energy cycle and water cycle of the Earth, have been a focus of the cloud physics community over the last few decades. With the increasing recognition of the importance of clouds in regulating climate and the growing concern over potential global climate change caused by human activities, clouds have recently come to the center stage of climate research. Cloud effects and cloud feedbacks have been identified as one of the largest uncertainties in current climate models [2]. Cloud processes/properties need to be parameterized in climate models as subgrid processes because they cannot be explicitly resolved by the state-of-art climate models. For the same reason, cloud microphysics has to be parameterized in cloud-resolving models.

Despite the great progress made over the last few decades, many issues regarding clouds remain unsolved. This work is concerned with two of them: microphysics parameterizations of warm clouds in climate models (cloud parameterizations hereafter for brevity), and theoretical studies of cloud droplet size distributions. In Section 2, we review and compare the existing schemes for cloud parameterizations in climate models, with emphasis on the effect of spectral shape of the droplet size distribution. We show that the spectral shape alone can cause substantial errors in calculation of cloud radiative properties, indicating the need to specify the spectral shape in addition to liquid water content and droplet concentration. We also argue that the same is true for cloud-resolving models, in which the effect of spectral shape is often ignored in the parameterization of cloud microphysics. The inclusion of spectral shape in cloud parameterizations makes the subject of cloud parameterizations essentially equivalent to the relatively old subject of specifying the droplet size distribution function. As a result, instead of being treated as separate subjects, the key to improving cloud parameterizations becomes the core of cloud physics.

In Section 3, we introduce some long-standing issues regarding the spectral shape of the droplet size distribution, and then briefly discuss the traditional mainstream theories proposed to address these issues and their deficiencies, including various entrainment and mixing models and stochastic models. In Section 4, we focus on a systems theory that has been recently formulated based on ideas of statistical physics and information theory to avoid the difficulties of the mainstream theories. This theory establishes a self-consistent theoretical framework, offers new insights into the issues that have been frustrating the mainstream theories, and reveals that droplet size distributions depend on the scale over which the size distributions are averaged (scale-dependence hereafter). This scale-dependence has important implications for virtually all cloud-related issues, including cloud parameterizations. Because the systems theory is relatively less known compared to mainstream models, it is a major focus of this contribution. In Section 5, we discuss new challenges ahead, address potential applications of the systems theory, and explore opportunities for a new theoretical framework to meet these challenges.

2. CLOUD PARAMETERIZATIONS

2.1. Effective Radius and Spectral Dispersion

Effective radius (defined as the ratio of the third to the second moment of a droplet size distribution) is one of the key variables used in calculation of the radiative properties of liquid water clouds [3-4]. The inclusion and parameterization of effective radius in climate models has proven to be critical for assessing global climate change. Slingo [5] studied the sensitivity of the global radiation budget to effective radius and found that the warming effect of doubling the CO_2 concentration could be offset by reducing effective radius by approximately 2 µm. Kiehl [6] found that a number of known biases of the early version of CCM2 were diminished, and important changes in cloud radiative forcing, precipitation, and surface temperature resulted if different values of effective radius were assigned to warm maritime and continental clouds. A high sensitivity to the method of parameterizing effective radius was also found in a recent study of the French Community Climate model [7].

Early parameterization schemes expressed effective radius as either a linear or a cubic root function of the liquid water content, implicitly assuming no dependence of effective radius upon the total droplet concentration [8, 9]. There has been increasing evidence for parameterizing effective radius as a "1/3" power law of the ratio of the cloud liquid water content to the droplet concentration [10-16]. The “1/3” power-law takes the form

$$r_e = \beta \left(\frac{3}{4\pi\rho_w} \right)^{1/3} \left(\frac{L}{N} \right)^{1/3}, \qquad (1)$$

where r_e is the effective radius in µm, L is the liquid water content in gm^{-3}, and N is the total droplet concentration in cm^{-3}. The dimensionless parameter β is a function of the spectral shape, and can be universally expressed as [13, 16]

$$\beta = \frac{\left(1+3\varepsilon^2+s\varepsilon^3\right)^{2/3}}{\left(1+\varepsilon^2\right)}, \qquad (2)$$

where ε is the spectral dispersion defined as the ratio of the standard deviation and the mean radius of the droplet size distribution, and s is the skewness of the droplet size distribution.

2.2. Relationship between β and ε

For clouds with a monodisperse droplet size distribution as described by a delta function $n(r)=N\delta(r-r_e)$, effective radius equals the volume mean radius, and $\beta_{MO} = 1$. This value of β was used by Bower and Choularton [10], and Bower et al. [11] to estimate the effective radii of layer clouds and small cumuli. In a study of the sensitivity of NCAR’s CCM2 to variations in r_e, Kiehl [6] used this scheme to provide support for choosing r_e of 5 µm and 10 µm for continental and maritime clouds respectively. Martin et al. [13] found that there are differences in the values of spectral dispersion between maritime and continental clouds, and derived estimates of $\beta_{MM} = 1.08$ for maritime stratocumulus, and $\beta_{MC} = 1.14$ for continental stratocumulus. By assuming a negligible skewness of the droplet size distribution, Pontikis and Hicks [12] analytically derived an expression that relates β to the spectral dispersion. This expression is hereafter referred to as Gaussian-like and denoted by GL, because the Gaussian distribution represents a typical form of such symmetrical distributions.

The assumption of either monodisperse or Gaussian-like distribution is clearly problematic for cloud physicists, because it has been long known in cloud physics that neither of them represent droplet size distributions observed in real clouds well (see Section 3 for details). These parameterizations are only appropriate for clouds with weak turbulent entrainment and mixing where droplet size distributions are rather narrow. For clouds exhibiting broad size distributions, the above-mentioned parameterizations underestimate β and therefore effective radii, though to different degrees. To allow for the spectral broadening processes such as turbulent entrainment and mixing in the parameterization of effective radius, Liu and Hallett [14] derived another parameterization for effective radius in the form of Eq. (1) based on a systems theory that is discussed in Section 4. The corresponding relationship between β and ε is hereafter referred to as the WB expression, because it corresponds to the Weibull form of cloud droplet size distributions. Besides the Weibull distribution, the gamma distribution and the lognormal distribution have also been widely used to represent droplet size distributions [17]. Similar to the derivation of the WB expression, expressions for β as a function of ε can be easily derived for the gamma [18, GM hereafter] and the lognormal [19, LN hereafter) droplet size distributions. These expressions are summarized in Table 1.

Table 1. The Commonly Used β–ε Expressions

MO	$\beta = 1$
MM	$\beta = 1.08$
MC	$\beta = 1.14$
GL	$\beta = \dfrac{\left(1+3\varepsilon^2\right)^{2/3}}{\left(1+\varepsilon^2\right)}$
WB	$\beta = 1.04\dfrac{\Gamma^{2/3}(3/b)}{\Gamma(2/b)}b^{1/3}$ (See Section 4 for the relationship between b and ε)
GM	$\beta = \dfrac{\left(1+2\varepsilon^2\right)^{2/3}}{\left(1+\varepsilon^2\right)^{1/3}}$
LN	$\beta = \left(1+\varepsilon^2\right)$

It is obvious from Table 1 that the only distinction between these different parameterizations for effective radius lies in the form of the dependency of β on spectral dispersion, which is determined by the functional form that is assumed for droplet size distributions. Therefore, given liquid water content and droplet concentration, the identification of the best parameterization of effective radius is essentially equivalent to the determination of the best mathematical expression for the droplet size distribution.

2.3. Comparison of the β-ε Expressions

Although the importance of β in the parameterization of effective radius was recognized in the early 1990s, most parameterizations assume a constant β and concern themselves mainly with liquid water content and droplet concentration (see a recent review for details [20]). To the best of our knowledge, the only systematic study where the effect of spectral dispersion is the focus was done by Liu and Daum [21]. In Ref. [21], the MO, MM, MC, GL and WB expressions were compared with observed data collected from continental stratocumulus clouds. It was found that the WB expression performs the best over the range of observed values of spectral dispersion (0 to ~ 1.2). For the dual purposes of identifying the best cloud parameterization and the best size distribution function, we compare in Fig. 1 all the expressions given in Table 1 with those calculated from the measured droplet size distributions collected in continental (crosses) as well as maritime (solid dots) clouds.

Significant differences between the dependencies of β on the spectral dispersion are exhibited in Fig. 1. The values of β derived from the measurements increase monotonically with the spectral dispersion for both continental and maritime clouds. The results indicate that the WB expression best fits the measurements over the range of observed values of spectral dispersion (the GM expression is close to the WB expression). The GL expression underestimates, while the LN overestimates β when droplet size distributions are broad. The GL, WB, GM and LN are almost equivalent for very narrow size distributions. The MO, MM and MC only represent cases with specific small values of spectral dispersion.

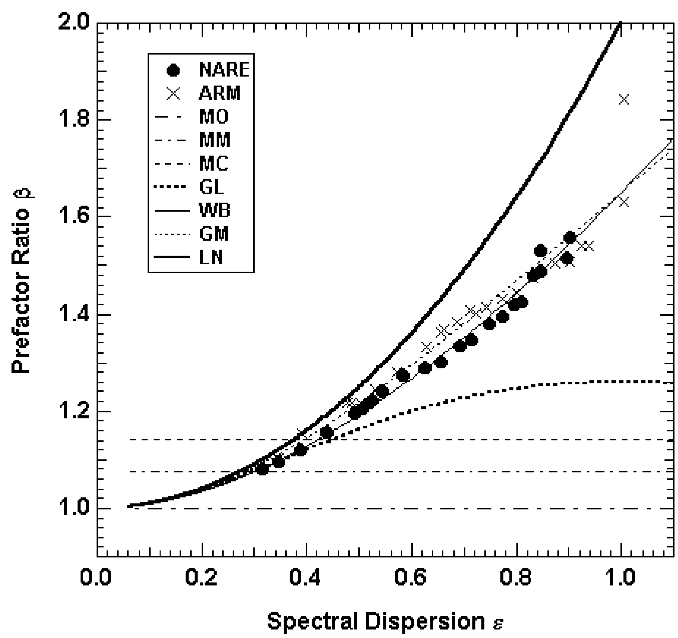

Figure 1. Comparison of different β-ε expressions given in Table 1 with measurements. NARE and ARM represents results derived from measured droplet size distributions collected in maritime clouds and continental clouds during two different projects, respectively. See the text for the meanings of the other symbols.

2.4. Comparison of Measured and Parameterized Effective Radius

Figure 2 further illustrates the performance of the different cloud parameterizations given in Table 1, including the GM and LN expressions. The measured values are calculated from droplet size distributions collected using the Forward Scattering Spectrometer Probe (FSSP). As expected, the WB (or GM) expression obviously outperforms the other schemes, which all underestimate or overestimate (LN) effective radii to different degrees.

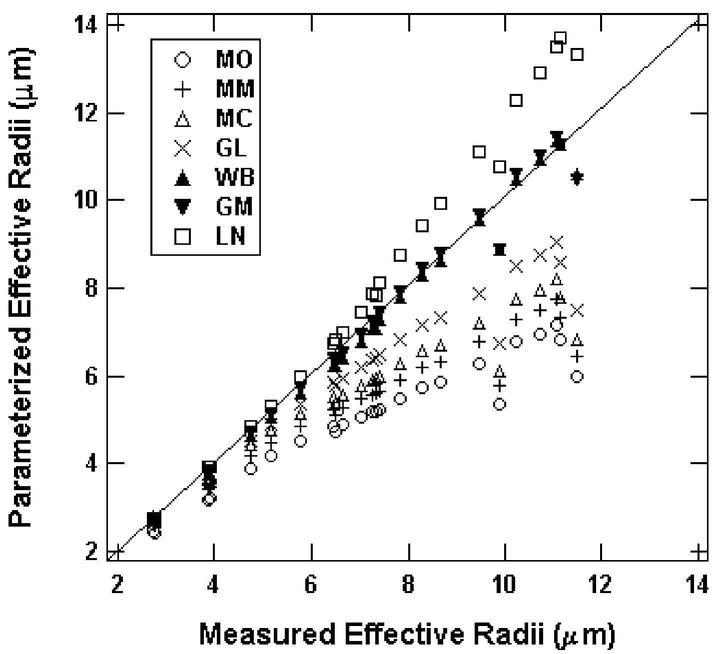

Figure 2. Comparison of measured effective radii with those derived from the different expressions given in Table 1.

The substantial differences in parameterized values of effective radius are due to the different treatments of β as a function of the spectral dispersion because the same values of L and N are used for all the parameterization schemes. This result can be better understood by examining the differences between measured and parameterized values of effective radii as a function of spectral dispersion. Figure 3 shows that except for the WB and the GM schemes, whose errors in parameterized effective radii are always within 1 μm and without obvious trend of change with the spectral dispersion, the biases in parameterized values of effective radii strongly increases with the spectral dispersion. At large spectral dispersions, the GL scheme could underestimate effective radius by over 2 μm; the underestimation is even larger for those schemes with fixed values of β (MO, MM, and MC). In the contrary, the LN scheme overestimates effective radius by similar amount.

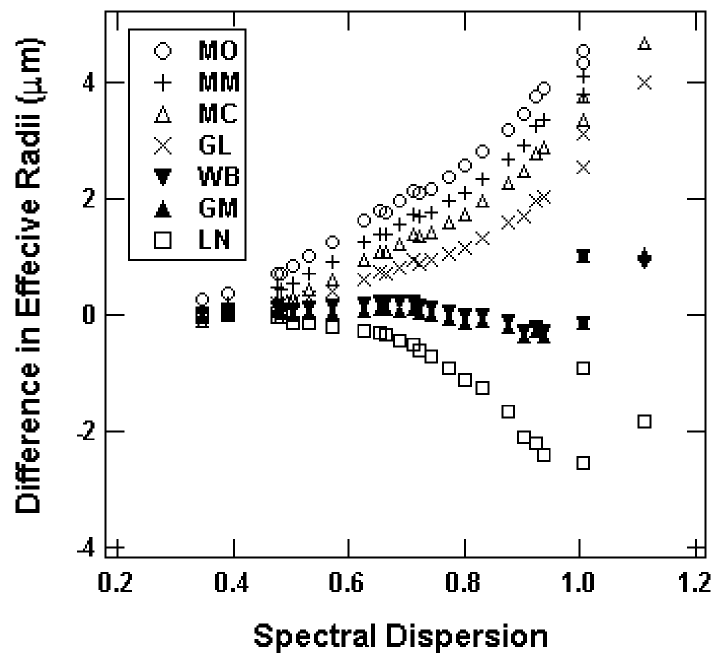

Figure 3. The difference between measured effective radii and those estimated from the expressions given in Table 1 as a function of spectral dispersion.

2.5. Section Summary

Briefly, this section demonstrates the following results. (1). Spectral dispersion is vitally important for the parameterization of effective radius, and therefore for the parameterization of cloud radiative properties. (2). The inclusion of spectral dispersion makes cloud parameterizations essentially equivalent to the choice of the droplet size distribution function. (3). The WB (or GM) scheme of cloud parameterization, which corresponds to the Weibull droplet size distribution, appears to be the most accurate among the different expressions given in Table 1. It has also been found that the Weibull distribution best fits droplet size distributions observed in cumulus clouds [22]. Because the close interactions between microphysics, dynamics and radiation, spectral dispersion is also important for microphysics parameterization in cloud-resolving models.

These results prompt us to ask two further questions: (1) why does the Weibull distribution well describe observed droplet size distributions, and (2) what is the best way to specify spectral dispersion in terms of predictable variables in climate models? These two questions lead the issue of cloud parameterizations to the core of cloud physics: the spectral shape of the cloud droplet size distribution and the physics behind it.

3. SPECTRAL BROADENING AND MAINSTREAM KINETIC THEORIES

3.1. Spectral Broadening

A wonderful historical review of cloud physics is given in Refs. [23] and [24]. Briefly, the study of clouds started in the 18th century, but most of the quantitative information on droplet size distributions was not available until the 1940s. Since 1940s, increasing attention has been devoted to cloud physics as a result of a number factors. For example, a surge of interest in doud physics was closely tied to the military-related research during World War II. After the war, interest was greatly stimulated by the discovery of the potential for weather modification in the late 1940s. A recent surge of interest comes from the recognition of the critical role of clouds in regulating climate. Although great progress has been made over the last few decades, the central problem of cloud physics—understanding and predicting droplet size distributions remains largely unsolved.

The condensational growth equation for individual droplets is well established and can be found in virtually every textbook on cloud physics [23, 24]. It is given by

$$\frac{dr}{dt} = \frac{G}{r}, \qquad (3)$$

where G is a function of the supersaturation. If all the droplets are exposed to the same supersaturation (this is the assumption of the classical uniform model), then this equation leads to droplet size distributions that tend to be monodisperse. This fact was realized long ago. For example, Houghton [25] in 1938 considered the asymptotic approach of droplet size distribution to monodisperse size distributions as a possible reason for the colloidal stability of clouds, and suggested an experimental verification.

Contrary to Houghton's expectation, it soon became evident in the late 1940s and early 50s that observed droplet size distributions are much broader compared to those predicted by uniform models [26]. The discrepancy between observations and uniform models (*spectral broadening*) has been a long-standing problem in cloud physics and is still awaiting physical explanations. In addition, it has also been observed that the discrepancy between observed and simulated droplet size distributions increases with increases in turbulence intensity. Even in adiabatic cores, it was recently found that observed droplet size distributions are still broader than those predicted by uniform models [27].

To explain the so-called spectral broadening, a number of models/hypotheses have been proposed over the last few decades. The mainstream models can be roughly grouped into two schools that are discussed below.

3.2. Entrainment and Mixing Models

a. Dynamical effects

Early studies of entrainment and mixing processes were mainly concerned with their effects on liquid water content, droplet concentration and dynamics. Stommel [28] noted in 1947 that cumulus clouds had to be significantly diluted by lateral entrainment and mixing of dry air in order to explain their internal temperature and liquid water content. He found that temperature profiles measured inside clouds were much closer to the soundings of the surrounding environment than to the moist adiabatic, and that about half the air in a cloud came from the surrounding environment. Malkus [29] had considerable success in interpreting observed liquid water deficits based on Stommel's entrainment process. Based on observations of the liquid water content of cumulus clouds, Warner and Squires [30] concluded, "it appears that the full adiabatic liquid water content in cumuli is realized, if at all, only in regions which are of negligible size in relation to the cloud as a whole. In most cases, the liquid water in the main body of the cloud is less than a quarter of the adiabatic value, and often considerably less."

Three features commonly observed in small cumulus clouds are a decrease in the ratio of liquid water content to adiabatic liquid water content with altitude above cloud base, the remarkably flat cloud base, and the horizontal uniformity of liquid water content at any one sampling altitude [31]. If entrainment occurred through the sides of the cloud, followed by lateral mixing, one would be facing the problem of explaining infinite horizontal diffusion and near-zero vertical diffusion. Based on these facts, Squires [32] in 1958 postulated the first cloud top entrainment model. However, his idea was not pursued by other scientists in this field until the mid-1970's. A thermal dynamic diagram was developed in 1979 by Paluch to locate the origin of the mixed cloudy air [33]. She used the wet equivalent potential temperature and the total water mixing ratio (liquid water and water vapor) as axes in her diagram. Since both of these parameters are conserved during adiabatic motions of a moist air parcel with or without condensation, an air parcel's position on the diagram will not change during adiabatic motions. She used this diagram to analyze data collected during the National Hail Research Experiment (NHRE) and found that clouds consisted of mixtures of air originating below cloud base and some level above the observation level, generally near cloud top. LaMontagne and Telford [34] analyzed observations of small cumulus clouds made in the South Dakota area using the Paluch diagram. They found that the clouds contained a mixture of air from cloud base and air from above cloud top. The portion of air from above cloud top increased with altitude. Blyth et al. [35], using the Paluch technique, investigated data obtained from the High Plain Experiment (HIPLEX) and the Cooperative Convective Precipitation Experiment (CCOPE), both conducted near Miles City, Montana. They concluded that the entrained air was generally close to, or slightly above the observation level of the aircraft. They presented a schematic model of a cumulus cloud with continuous entrainment into the surface of the thermal eroding the core, and the remaining undiluted core region continuing its ascent, leaving a turbulent wake of mixed air behind it. Based on thermodynamic analysis, Jonas [36] concluded, "many of the clouds contain evidence of entrainment of air originating significantly above the observation level which may result from penetrative downdraughts formed by entrainment at cloud top." He also found that cloud top entrainment was more common in deeper clouds (~ 4 km deep) than in shallower clouds. Carpenter et al. [37-39] presented results from their three-dimensional numerical model, and confirmed the above-mentioned shedding thermal model.

Entrainment at the top of the stratocumulus-topped marine boundary layer is important in the life cycle and the drizzle production of such clouds. Early studies in this field began with Lilly [40] in 1968 by modeling the cloud-topped mixed layers under an inversion. He, as well as Randall [41], assumed that cloud-top radiation was important in entrainment instability. Telford and Chai [42] proposed a hypothetical model of the formation of inversions, and fog, stratus and cumulus in warm air over cooler water. They claimed that if the air was cooler than the water (which is the case in most persisting cloud-topped marine boundary layers), an inversion could form which would then lead to the formation of stratus as well as cumulus depending on the entrainment instability. They found that radiation was not an important controlling factor in this process. Entrainment instability was controlled by the wet-bulb potential temperature difference across the sharp inversion at cloud top. The amount of air that could be entrained depended on the structure of the wet-bulb potential temperature above the inversion.

Recent research in this area has concentrated on two directions: airborne measurements of entrainment rates under various conditions, and improvements in the parameterization of the entrainment process in numerical models. Using three methods (thermodynamic budget of the boundary layer, turbulent flux observed near the inversion, and combining the observed rate of cloud top height change and the estimated subsidence rate), Boers et al. [43] obtained an entrainment rate of 4 mm s^{-1} over the Southern Ocean. The preliminary findings from the Dynamics and Chemistry of Marine Stratocumulus (DYCOMS-II) experiment [44] estimated an entrainment rate of 3 to 5 mm s^{-1} at the top of the stratocumulus during night times off the southern California coast.

On the modeling side, since large-eddy simulation (LES) models have great promise in the testing of entrainment closures, stratocumulus topped marine boundary layer simulations are more reliant upon these models [45, 46]. Scientists are not only testing different entrainment parameterization schemes but are also concentrating more on the effect of grid resolution on entrainment calculations [47]. The effect of radiation on entrainment rate has also raised more concern

[48]. The effect of entrainment and mixing processes on the parameterization of effective radius due to their impacts on liquid water content and droplet concentration in climate models was recently discussed [20].

b. Application to spectral broadening

The application of the idea of entrainment and mixing to explain spectral broadening started in the 1970s. In order to explain observed droplet size distributions, Warner in 1973 simulated entrainment in his model by assuming that the entrainment rate was either constant or varied with the updraft velocity [49]. He also assumed that the entrained air spread instantaneously throughout the whole lateral cross-section of the entrained level. In other words, all droplets at that level were exposed to the entrained dry air, and all drops reduced in size in order to maintain saturation. This process has been referred to as *homogeneous mixing* through the sides of a cloud. He found that spectral broadening was not significant if the entrained air was nuclei free, regardless of the entrainment rate. If the entrained air contained nuclei, the size distribution was broadened considerably; however, it often had a single broad peak that differed from those observed in natural clouds. Lee and Pruppacher [50] conducted a more complete simulation of the homogeneous mixing model in which they used two different nuclei [NaCl and $(NH_4)_2SO_4$]. They found that if entrained air contained no nuclei, size distributions were similar to those adiabatic cases with no entrainment. If both kinds of nuclei were considered, the size distributions were broadened, and sometimes bimodal spectra were obtained. If only one kind of nuclei was used, the broadening was somewhat less significant than in the previous case. In all cases, there were still some difficulties in explaining the observed droplet size distributions. Mason and Jonas [51] and Jonas and Mason [52] presented a spherical thermal model, each thermal rising through the residual of its predecessors. In these cases, the air entrained from outside the cloud is instantaneously mixed with that at the height of the center of the blob throughout the thermal. Unfortunately in a cloud of this size (500m radius or so), calculating the droplet size distribution at the center of the spherical thermal does not provide meaningful size distributions at other heights, particularly when the lifting condensation level is partway through the sphere.

Based on their laboratory experiments, Latham and Reed [53] found that entrainment left the shape of the spectrum and its mean diameter unchanged while the total number concentration of the droplets decreased. They hypothesized that, before spreading throughout the whole cross-section of the cloud at the level of entrainment, as assumed in the homogeneous mixing process, the entrained air would evaporate all the droplets in its immediate neighborhood until saturation was reached and then spread laterally throughout the level. This process is referred to as the *inhomogeneous mixing process*. Based on the findings from these experiments, Baker and Latham [54] and Baker et al. [55] simulated the inhomogeneous mixing process in a simple model, and discussed the time scales for turbulent diffusion, molecular diffusion, and droplet growth/evaporation. They concluded that the time scale for growth/evaporation is much shorter than for both turbulent and molecular diffusion if the scales are greater than 1 m for droplets of 10 μm radius. Their inhomogeneous mixing model produced broad bimodal spectra with large drops as well as smaller droplets of all sizes. However, the mechanism by which the entrained air quickly penetrated so deeply into the clouds remained unclear.

Telford [56] discussed his entity type entrainment mixing (ETEM) process through cloud tops. He suggested that once an air parcel is entrained, the parcel will maintain its identity and will mix with the cloudy air in its immediate neighborhood. The first droplets mixed into the entrained parcel will be totally evaporated until saturation is reached. Further mixing between the entrained parcel and cloudy air in its environment will only change the droplet concentrations but not their sizes. Evaporative cooling makes the entrained air parcel denser than the surrounding cloud and will cause it to descend to a level of neutral buoyancy. This parcel will rise again toward the cloud top at some later time. Continuous mixing with the surrounding cloudy air happens at all times due to turbulence. Telford and Chai [57] further presented a condensation model of the ETEM process. Because of the dilution effect in the entrained parcel, large drops can grow and because of the continuous mixing with its surrounding cloudy air, the entrained parcel has a continuous supply of smaller droplets of all sizes. This process broadens the droplet size distribution, reduces the adiabatic liquid water content and the total droplet concentrations.

3.3. Stochastic Theories

The idea of stochastic condensation, which considers the growth of a droplet population as a stochastic process and relates the spectral broadening to various fluctuations such as supersaturation associated with turbulence, was pursued from the 1960s especially by Chinese and Russian scientists [58-63]. These early theories often replaced the full growth equations by simplified versions amenable to analytic analysis, assumed Gaussian fluctuations, and claimed that turbulence fluctuations lead to spectral broadening. On the contrary, by numerically solving the full growth equations under Gaussian fluctuating environments generated by Monte-Carlo simulations, Warner [64], and Barlett and Jonas [65] predicted that turbulent fluctuations only slightly broaden droplet size distributions. They argued that the supersaturation and the updraft are so closely related to one another that a droplet that experiences a higher supersaturation, and therefore grows faster, is likely to be in a stronger updraft which will allow it a shorter time to grow on passing between any two levels in a cloud. Conversely, lower supersaturations are associated with smaller updrafts and longer growth times. Manton [66] demonstrated that turbulent mixing ignored in Refs. [64] and [65] can break the link between supersaturation and updraft, and that the modified stochastic theory can lead to spectral broadening. Nevertheless, the hypothesis of the breakdown of the correlation between supersaturation and updraft remains controversial [67, 68]. Khvorostyanov and Curry [69] pointed out that these early low-frequency theories of stochastic condensation generally yield droplet size distributions of the Gaussian type while observations tend to follow positively skewed distributions. They derived a more general mean-field equation, and showed that their equation has the analytical solution of the gamma distribution under certain assumptions in the low-frequency regime [70].

Considine and Curry [71] proposed a model based on the assumption that size distributions at a given level in a cloud are horizontal averages over a large number of air parcels that can have a different lifting condensation level. Shaw et al. [72] recently related spectral broadening to turbulence-induced preferential concentration of droplets. Srivastava [73] argued that the supersaturation that controls each individual droplet (microscopic supersaturation) differs from the commonly used macroscopic supersaturation. It was shown that, even without turbulence, the Poisson spatial distribution of droplets could cause droplet-droplet variations in the microscopic supersaturation, which in turn leads to some spectral broadening.

3.4. Section Summary

Significant progress in our understanding of formation of droplet size distributions has been made over the last few decades through the various mainstream theories. However, the details of the processes involved in the mainstream theories are poorly understood and highly controversial, which is self-evident from the diversity of the hypotheses discussed above. Furthermore, the school of entrainment and mixing is largely isolated from the school of stochastic condensation. Such isolation is problematic because entrainment, mixing and fluctuations, are actually acting together on droplets, and all these processes occur over a tremendous range of interacting scales between the largest eddy of a cloud size and the smallest eddy of the Kolmogorov microscale [74]. The mainstream theories do not provide explanations for the questions raised at the end of Section 2.

Despite their differences, the mainstream models have one feature in common: they attempt to follow each "eddy", or even each droplet. It has been increasingly recognized that the size of the model grid needs to be as small as ~ 1 mm so that the smallest eddies of turbulence, the mean distance between droplets, and microscopic supersaturation can be resolved and that the grid values of variables such as temperature and water vapor mixing ratio represent the ambient conditions for the growing droplet [74-76]. It is computationally prohibitive to numerically solve the associated equations. More importantly, the stochastic processes, the wide range of scales, and droplet interactions involved in turbulent clouds are so complex that it may be hopelessly difficult to completely know the path of each droplet/parcel, droplet interactions, and the initial and boundary conditions necessary for solving the growth equations. The difficulties are evident from the fact that the randomness of turbulence is no simpler than that of Brownian motions of molecules [77]. In fact, the subject of turbulence itself has been considered one of the unsolved problems of classical physics [78]. The mutual interactions between droplets and turbulence further complicate the problem [79, 80].

This vexing situation is similar to the early stage of the kinetic theory of gases in the late 19th and early 20th centuries. During that time period, scientists (e.g., Maxwell, Boltzmann and Gibbs) were frustrated by their inability to explain the macroscopic thermodynamic properties of gases, despite the fact that the Newtonian equations could accurately describe the motion of each individual molecule in a gas. By analogy to the kinetic theory of gases, these bottom-up models are generically referred to as kinetic theories.

4. THE SYSTEMS THEORY

In view of the insurmountable difficulties with kinetic models, a entirely different formalism, which considers cloud droplets as a system and studies them as a whole instead of following each droplet or eddy, has been recently developed by integrating into cloud physics the ideas from statistical physics and information theory [14, 81-86]. In this section, we elaborate on this theory and the major results derived from it, because it is relatively less known compared to the mainstream theories discussed in Section 3.

4.1. Basic Philosophy and Droplet Ensemble

The essence of the systems theory is to obtain useful information on droplet size distributions without concern with the details of each individual droplet. This philosophy is analogous to the idea used by Maxwell, Boltzmann and Gibbs, among others, to avoid the difficulties of following each molecule in a gas, and is still the backbone of modern statistical physics whose applications extend far beyond thermodynamic systems [87]. Let us start by quoting part of Gibbs' famous preface to his Elementary Principles in Statistical Mechanics [88]:

"*We may imagine a great number of systems of the same nature, but differing in the configurations and velocities.... And here we may set the problem, not to follow a particular system through its succession of configurations, but to determine how the whole number of systems will be distributed among the various conceivable configurations and velocities at any required time, when the distribution has been given for some one time.... The laws of thermodynamics, as empirically determined, express the approximate and probable behavior of systems of a great number of particles, or more precisely, they express the laws of mechanics for such systems as they appear to beings who have not the fineness of perception to appreciate quantities of the order of magnitude of those which relate to single particles, and who cannot repeat their experiments often enough to obtain any but the most probable results.*"

Similarly, the existence of turbulence and fluctuations in clouds leads us to assume that a droplet size distribution results from a large number of stochastic events and to consider a droplet ensemble that consists of an arbitrarily large number of different microstates (size distributions) satisfying the same macroscopic constraints (conservation laws). The choice of the ensemble depends on the conditions imposed on the systems (e.g., microcanonical ensemble for isolated systems, or canonical ensemble for systems in contact with a thermostat). The first key to the systems theory is to establish a droplet ensemble suitable for the droplet system.

The droplet ensemble that has been investigated so far is for the study of droplet systems having monomodal droplet size distributions [14, 83, 84, 86]. Briefly, it has two constraints:

$$\int r\,(\mathrm{x})\mathrm{dx} = 1, \quad (4a)$$

$$\int x r\,(\mathrm{x})\mathrm{dx} = \frac{\mathrm{X}}{\mathrm{N}}, \quad (4b)$$

where x, defined as the Hamiltonian variable (it was previously called restriction variable), is related to the physical processes controlling the droplet system; X is the total amount of x per unit volume; N is the total droplet concentration; ρ(x) = n(x)/N can be considered as the probability that a droplet of x occurs and n(x) is the droplet concentration per unit volume per unit x interval. It should be noted that the correspondence between x and the conservation law is a key to the ensemble. For example, for the special droplet system constrained by the conservation of liquid water content, x represents the mass of a droplet, X is the liquid-water content, and n(x) the droplet concentration per unit mass interval.

The current systems theory focuses on the following question: given X and N, what are the most and the least probable ways to distribute X among the N droplets?

4.2. The Most Probable Size Distribution

As the Boltzmann energy distribution describes the most probable energy distribution of a molecular system and the Maxwell velocity distribution characterizes the most probable velocity distribution of a molecular system, it is expected that there exists a characteristic droplet size distribution that occurs most probably among all the possible droplet size distributions.

By analogy with the Boltzmann entropy for molecular systems and the Shannon-Jaynes entropy generalized for complex systems, the spectral entropy H is introduced and defined as

$$H = -k\int r\,(x)\ln\left[r\,(x)\right]dx, \quad (5)$$

where k is a proportional constant that has no effect on the derivation of the most probable droplet size distribution. Maximizing the spectral entropy subject to the constraints described by Eqs. (4a) and (4b), we can easily derive

$$r^*(x) = \frac{1}{a}\exp\left(-\frac{x}{a}\right), \quad (6)$$

where α = X/N represent the mean amount of X per unit droplet. Note that the physical meaning of α is consistent with that of "$K_B T$" in the Boltzmann energy distribution (K_B is the Boltzmann constant, T is the temperature, and $K_B T$ essentially represents the mean energy per molecule in the gas). Therefore, the most probable droplet distribution with respect to the Hamiltonian variable x is

$$n^*(x) = \frac{N}{a}\exp\left(-\frac{x}{a}\right). \quad (7)$$

In cloud-related studies, droplet size distribution with respect to the radius (r) is preferred. It has been argued that x is related to r by the power-law relation [14, 83, 84, 86]

$$x = ar^b, \quad (8)$$

where the parameters a and b are related to physical mechanisms controlling the droplet system. For the special case of liquid-water content conservation, a = [1/(6$\pi\rho_w$], and b = 3. The symbol ρ_w denotes the water density. A combination of Eqs. (7) and (8) yields that the most probable droplet size distribution follows the Weibull distribution

$$n_{\max}(r) = N_0 r^{b\text{-}1} \exp(\text{-} l r^b), \qquad (9)$$

where the parameters N_0 = ab/α and λ = a/α.

4.3. The Least Probable Size Distribution

Cloud droplet systems are more complex than molecular systems. For a molecular system, the most probable state suffices to specify macroscopic thermodynamic properties such as temperature and pressure because of the enormous number of molecules involved (e.g., 10^{22} cm^{-3}). In other words, the most probable state virtually is the mean state. As a result, other states such as the least probable state have not been a concern in statistical physics. However, because of very limited concentrations of cloud droplets (e.g., 100 in cm^3), one cannot always equate the most probable size distributions with observed or modeled droplet size distributions. Therefore, a complete characterization of such a small system may also require knowing other possible droplet size distributions. Although determining the specific probability of each possible size distribution seems impossible at present, useful information can be obtained by knowing the least probable distribution. If the least probable distribution is identical with the most probable distribution, clouds are absolutely uniform and the uniform model suffices. An example would be a uniform updraft with all droplets exposed to the same supersaturation and identical cloud condensation nuclei (CCN). However, such idealized situations probably never occur in nature. If there are any differences between the most and least probable distributions, individual size distributions then depend on the scale over which they are averaged.

A new concept of spectral free energy, defined as the energy necessary for the formation of a specific droplet size distribution n(r), is introduced. Note that it was previously called the populational energy change, and we switch to the new term because of its analogy with the concept of free energy in thermodynamics. In Ref. [85], the spectral free energy E was expressed as

$$E = -\frac{p r_w L_v}{3}\int r^3 n(r)\,dr + p s \int r^2 n(r)\,dr + c, \quad (10)$$

where the first term on the right side is the latent energy with L_v representing the latent heat of the condensation of water vapor; the second term is the surface energy with σ representing the surface tension of water. The coefficient c is related to the activated CCN. Equation (10) was derived under the common assumption that other forms of energy (i.e., gravitational potential energy, the kinetic energy associated with droplet terminal velocities, and the solution effect) are negligibly small [24]. In Ref. [86], these minor terms were incorporated into the coefficients before the integrals as,

$$E = c_1 \int r^3 n(r)\,dr + c_2 \int r^2 n(r)\,dr + c, \qquad (11)$$

where the coefficient $c_1 = [(\pi\rho_w)/3 \ (-L + gh + 1/2\overline{V_t^2})]$ (g is the gravitational constant; h is the height over which the water molecules in droplets are displaced; $\overline{V_t^2}$ is the mean square terminal velocity of droplets) includes the effects of the latent heat, gravitational potential energy (gh), and the kinetic energy ($1/2\overline{V_t^2}$). The coefficient c_2 considers the solution effect on the surface tension. Maximizing E given by Eq. (11) subject to the constraints described by Eqs. (4a) and (4b), the least probable droplet size distribution is derived as

$$n_{\min}(r) = N\delta(r - r_b), \qquad (12a)$$

$$r_b = \left(\frac{\int r^b n(r)\,dr}{N}\right)^{1/b} = \left(\frac{X}{aN}\right)^{1/b} . \qquad (12b)$$

It is obvious that the least probable droplet size distribution is a monodisperse size distribution.

It is easy to show that the least probable size distribution also corresponds to the minimum spectral entropy H = 0. The state of the minimum free energy is expected to correspond to the most probable size distribution by analogy to molecular systems where the Gibbs free energy is minimum at the state of maximum entropy. However, a formal relationship between the spectral entropy and spectral free energy remains elusive.

4.4. Contrasts between the Most and Least Probable Size Distributions

As shown above, expressions for the two most and the least probable droplet size distributions are derived by use of the same mathematical technique — calculus of variation. On the other hand, the spectral shapes of the two characteristic size distributions are drastically different in general. The Weibull most probable droplet size distribution is much broader than the monodisperse least probable size distribution. Table 2 summarizes the major contrasts between the two characteristic size distributions.

Table 2. Contrasts between the Most and Least Droplet Size Distributions

	Most Probable Distribution	Least Probable Distribution
Probability of occurrence	Most	Least
Spectral width	Wide	Narrow
Function to be optimized	Spectral entropy	Spectral Free energy
Association	Observation	Uniform model

It is noteworthy that the most probable size distribution, the least probable size distribution, and their differences depend on the constraint characterized by the parameter b. An analysis of the Weibull distribution as described by Eq. (9) shows that it becomes narrower with increasing values of b, and eventually approaches the monodisperse distribution as described by Eq. (12a) when b approaches ∞ (See [86] for mathematical proof). This behavior can be understood by examining the relationship between b and spectral dispersion for the most probable Weibull distribution,

$$e = \left[{}^{2b\Gamma(2/b)} \Big/ {}_{\Gamma^2(1/b)} - 1 \right]^{1/2}, \qquad (12)$$

where Γ (.) represents the standard gamma function. Figure 5 helps to visualize the asymptotic approach of the most probable distribution to the monodisperse distribution when b increases.

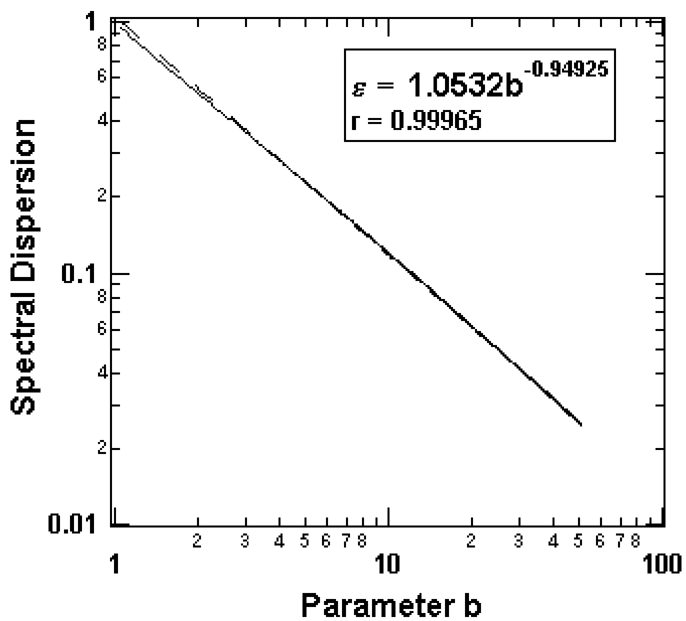

Figure 5. Relationship between spectral dispersion and the parameter b.

In Fig. 5, the solid line represents the relationship calculated from Eq. (12); the dashed line represents the fitting power-law relationship shown in the legend. The monodisperse size distribution has a spectral dispersion of 0. This result indicates that the most probable approaches the monodisperse distribution with decreasing fluctuations, because spectral dispersion is closely related to fluctuation levels in clouds, and increases with increasing fluctuations.

4.5. Explanations of Existing Puzzles

The unique properties of the most and least probable droplet size distributions, along with their dependence on fluctuations, can be used to explain the question raised at the end of Section 2, and virtually all the puzzles discussed in Section 3.1 in a unified way. First, the association of observed droplet size distributions with the most probable droplet size distribution explains why the Weibull distribution well describes observed droplet size distributions. This is best illustrated by an example. The PMS FSSP-100 is probably the most commonly used instrument for measuring droplet size distributions. The effective sample area of this instrument is approximately 0.004 cm^2. Suppose that at least 100 droplets are needed to establish a variance of less than 10%. The length that must be sampled is therefore (100 droplets/(0.004.N) = 25000/N (cm), where N is the droplet concentration in cm-3. This shows that for typical clouds of 100 - 200 cm-3, a path 125 to 300 cm long must be sampled, a path much longer than the typical Kolmogorov microscale of clouds (~ 1 mm). A much longer path is actually needed to get statistically meaningful droplet size distributions. FSSP measurements of 1 second (i.e., 100 m for a aircraft speed of 100 m/s) or longer are typically used in most studies. Therefore, observed droplet size distributions average many size distributions and, subsequently, look more like the most probable Weibull distribution.

Second, why is there spectral broadening? The agreement of the least probable monodisperse distribution with the prediction of the uniform growth model strongly suggests that the uniform model predicts a result that is least probable to occur when clouds are turbulent. The discrepancy between observations and model predictions may be due to comparing two entirely different characteristic droplet size distributions.

Third, very narrow droplet size distributions are indeed observed in clouds formed under "uniform" conditions, e.g., in lenticular clouds and in adiabatic cores of small cumulus. At first glance, the narrow size distributions observed under uniform conditions seem to indicate that these observed size distributions are associated with the least probable distribution. However, this is actually due to the fact that the most probable Weibull distribution approaches the monodisperse distribution when fluctuations decrease.

Finally, although droplet size distributions observed in the so-called adiabatic cores are very narrow, they are still broader than those predicted by uniform models [27]. In reality, real clouds are always in a more or less turbulent state. Even in non-turbulent, uniform clouds with uniform CCN, the Poisson random spatial distribution of droplets can cause fluctuations in the microscopic supersaturation [73], an essential variable controlling the condensation/evaporation of individual droplets. These facts suggest that the extreme monodisperse size distribution will probably never be observed, no matter how uniform the cloud.

It is worth noting in passing that the third law of thermodynamics assures that the state of absolute zero (or zero thermodynamic entropy) never occurs in nature. By analogy, we may speculate that the state of the zero maximum spectral entropy never occurs in atmospheric clouds. Coupled with the fact that the odds of observing the least probable distribution are extremely slim, the monodisperse droplet size distribution will never be observed!

4.6. Scale-dependence

The striking differences between the most and least droplet size distributions imply that individual droplet size distributions depend on the scale over which they are sampled/simulated. In particular, it has been argued [85, 86] that an individual size distribution approaches the most probable droplet size distribution with an increase in the averaging scale, and as a result, there exists a characteristic scale, defined as *saturation scale*, beyond which all size distributions are approximately the same and equal to the most probable droplet size distribution. When the averaging scale is less than the corresponding saturation scale, however, droplet size distributions are strongly dependent on the averaging scale, and therefore ill-defined without specification of the averaging scale. In this case, it is necessary to explicitly specify the scale and the dependence of droplet size distributions on the scale. The saturation scale and the details of the scale-dependence of droplet size distributions also depend on fluctuation intensities. The weaker the fluctuation, the smaller the difference between the most and the least probable droplet size distributions, the smaller the saturation scale, and the weaker the scale-dependency. The scale-dependence and spectral broadening disappear when there are no fluctuations in clouds.

Mandelbrot divided fluctuations into three categories: mild, slow and wild [77]. According to this proposal, the scale dependence with finite saturation scale corresponds to the classical mild fluctuations and is defined as the scale-dependence of the first kind. If the fluctuation is wild, there will be no saturation scale. In this situation, droplet size distributions are always dependent on the scale, and scale-dependence and the averaging scale are always key to understanding individual droplet size distributions, no matter how large the averaging scale. Even if the fluctuation is slow or the mild fluctuation is too strong, one may not be able to reach the saturation scale in practice. For now, we generically define the cases without saturation scales as the scale-dependence of the second kind. Figure 6 illustrates the two kinds of the scale-dependence. In reality, another complication comes from the fact that different droplet systems may be encountered during sampling processes in real clouds.

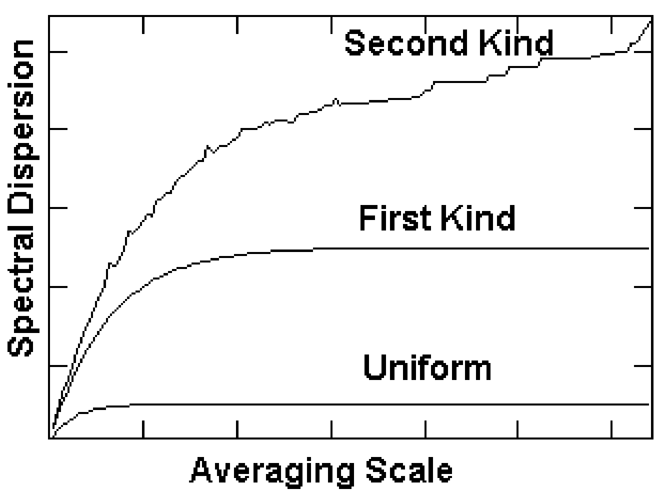

Fig. 6. A diagram illustrating the scale-dependence of droplet size distributions. Both axes are only qualitative. The bottom curve represents the simplest case of uniform clouds. The middle and top curves represent the scale-dependence of the first and second kind, respectively.

Scale-dependence and the associated ill-definedness have many practical implications as well. In general, question of the compatibility of the different scales will arise when comparing observations with models, when coupling models of different scales, and when comparing measurements collected using instruments with different sampling scales. For example, a suite of in-situ and remote sensing instruments (surface-, aircraft- and satellite-based) with a variety of scales are often coordinated in current field projects to address cloud-related issues. A key to such multi-instruments campaigns is the mutual comparison and validation of measurements made by instruments operated at different sampling scales.

To improve cloud parameterizations, it is increasingly common to couple climate models with microphysical models as detailed as allowed by computer resources. Such a direct coupling of models of different scales seems natural at first glance; but this coupling is questionable because of the scale-dependence of individual droplet size distributions. Droplet size distributions predicted by detailed microphysical models may not be compatible with those required in climate models because of the large scale-mismatch between the two kinds of models. It was recently proposed to replace the conventional cloud parameterization in climate models by the so-called super-parameterization which essentially means coupling cloud systems-resolving models to climate models [89, 90]. Similar scale-mismatch problems may exist in this idea too.

4.7. More Comparisons with Kinetic Models

The systems theory stands in stark contrast to kinetic models with regard to the philosophy of treating the problem of droplet size distributions. The philosophical differences between the systems theory and kinetic models in turn lead to differences in other aspects. Methodologically, kinetic models use a bottom-up approach, albeit different models involving different individual details; the systems theory uses a top-down approach. Mathematically, unlike kinetic models formulated using differential equations, the systems theory is built upon calculus of variations, integral equations and constrained optimization. Physically, the systems theory predicts the scale-dependence of droplet size distributions and accommodates spectral broadening as a manifestation of scale-dependency.

In the quest to understand and explain observed droplet size distributions, major efforts have been devoted to various kinetic models. The idea of the systems theory has received much less attention compared to its kinetic counterpart. At first glance, the systems theory seems to convey a "strange" impression that observed size distributions have little to do with the details of individual droplets and their interactions. It is interesting to note that physicists shared a similar impression regarding statistical mechanics during the early days of this discipline. However, such a view was refuted by the later success of statistical mechanics. The ideas of statistical mechanics have been successfully extended to study other complex systems [87]. Such widespread success provides indirect justifications for using the systems approach to study cloud droplet size distributions. The systems theory has also been justified by its successful explanations for many long-standing issues, including the Weibull droplet size distribution and spectral broadening.

4.8. Section Summary

By treating a droplet system as a whole instead of tracing individual details, the systems theory overcomes many difficulties confronting kinetic models, provides a theoretical framework that is able to explain existing puzzles in a unified fashion, and reveals many important points. First, observed droplet size distributions tend to follow the Weibull distribution because the probability of its occurrence is the highest. Second, the phenomenon that droplet size distributions observed in adiabatic cores of cumulus clouds are very narrow, yet still broader than those predicted by uniform models, is due to the small, yet inexorable fluctuations affecting cloud droplets even in adiabatic cores, which will cause small differences between the most and the least probable droplet size distributions. This result suggests that it is almost certain that monodisperse assumption will overestimate effective radius. Droplet size distributions depend on the scale over which they are observed or simulated, and the details of scale-dependence are related to fluctuation properties. Spectral broadening is a manifestation of scale-dependence, arising from scale-mismatch and incompatibility between models and observations.

The systems theory suggests that there are two "drivers" that compete to determine the spectral shape of cloud droplet size distributions: the deterministic driver as given in the uniform model tends to narrow the droplet distribution while the statistical driver tends to broaden the distribution. Observations seem to support the notion that the statistical driver prevails.

5. CONCLUDING REMARKS

We attempt to bring together the two traditionally separate subjects, cloud physics and cloud parameterizations in climate models and cloud-resolving models. Because each major topic has been briefly summarized in its own section, this section concentrates on the connections between these two subjects and the common problems confronting them.

It become evident now that spectral dispersion is the thread linking cloud parameterizations with cloud physics. In cloud physics, it had been recognized before the 1960s [17] that observed size distributions should be described by skewed distribution functions (e.g., gamma, Weibull, and lognormal distributions), rather than monodisperse or the Gaussian distributions as predicted by the uniform model and most stochastic theories. These skewed distribution functions have been assumed in the parameterization of cloud microphysics in cloud-resolving models. Unfortunately, cloud parameterizations corresponding to the monodisperse or the Gaussian droplet size distributions are still in wide use in current climate models. Furthermore, there is no agreement as to which distribution functions should be used to express droplet size distributions. Analysis of field data indicates that the Weibull distribution most accurately describes observed droplet size distributions among the commonly used distribution functions, and should be used in cloud parameterizations. However, mainstream kinetic theories have not yet succeeded in predicting this form of the droplet size distribution.

In fact, as discussed in Section 3, since the 1940s cloud physics has not been graced with major conceptual breakthroughs, but rather by a series of progressively more refined quantitative theories of previously identified microphysical processes. The reductionist approach inherent in the mainstream theories may be a reason for such a failure. On the other hand, the systems theory not only predicts the Weibull droplet size distribution, but also explains many other issues that have frustrated the cloud physics community over the last few decades. The successes of the systems theory furnish positive proof that a systems approach may hold the key to the understanding of droplet size distributions.

Several challenging questions remain to be solved. First, spectral dispersion needs to be further specified in terms of prognostic/ diagnostic variables in climate or cloud-resolving models. The issue of spectral dispersion subsists in the so-called super-parameterization. Spectral dispersion could become more important when precipitation is also a concern. The systems theory predicts that spectral dispersion is related to the parameter b that is involved in the constraints imposed on the droplet system and depends on fluctuation properties. One of the future challenges is to express b as a function of fluctuation properties. Based on previous studies, there are at least two major factors affecting spectral dispersion: dynamics and CCN properties. Dynamics can be further divided into updraft and turbulent fluctuations; CCN properties include size, chemical composition and concentration. To establish such relationships is also key to understanding indirect effects of anthropogenic aerosols on climate change, and this issue will be addressed elsewhere. A formal solution to this problem is not clear to us now. Nevertheless, an extension of the systems theory could be the answer. There are three distinct, complimentary disciplines to address issues regarding thermodynamic systems such as gases: kinetics, statistical physics, and thermodynamics. By analogy, this issue could be solved by establishing a new discipline, "thermodynamics for droplet systems" and linking it with the systems theory. Another way to find the constraints is to study the symmetry/invariance structure of the fundamental equations involved. According to Yang [91], symmetry reflects conservation laws and dictates interactions.

Second, the current systems theory only qualitatively reveals that individual droplet size distributions depend on the scales over which they are sampled/simulated. Furthermore, as discussed in Section 4.6, the details of the scale-dependence are critical for virtually all cloud-related problems, including cloud parameterizations. However, except for the uniform case, the saturation scale and the scale-dependence are largely unknown. Therefore, there is a dire need to quantify the scale-dependence and its relationship with fluctuation properties. Theoretically, the unique property of scale-dependence suggests the ultimate need for an entirely new theoretical framework that treats the scale as an independent variable, just as the variables of space and time are treated in the current framework. A combination of the systems idea with multiscale approaches seems to be a promising avenue. Actually, the need for a paradigm-shift from a scale-independent to a scale-dependent theoretical framework is emerging in many fields where a variety of fluctuations and scales are involved [92-94]. Current effort is concentrated on the search for the symmetry of scale-invariance (scaling models). Extensive scale-dependent analysis of data under a variety of fluctuating conditions can empirically provide crucial guidance to the establishment of the new theoretical framework.

Third, there seems little dispute on the role of fluctuations in determining droplet size distributions. A fundamental question is the origin of the "randomness". Practical schools think that randomness arise from our ignorance or incomplete information due to practical limitations, as well as from the prohibitive demand for computational resources. On the other hand, according to recent investigations into general dynamical systems [95], randomness is physically inherent in the dynamics of various processes occurring in clouds. Briefly, there are at least two types of situations in which dynamical motion physically generates randomness. The first corresponds to ergodic (mixing or K-flows) systems, and the second corresponds to the Poincare catastrophe (resonance). In both cases, the character of motion is such that two trajectories, regardless of how close together their starting points are, may diverge greatly in time. Although the quantitative details of kinetic equations of a droplet system as a dynamical system remain elusive, randomness is physically expected. Therefore, it should be stressed that in this case, probability and associated statistical laws are no longer a state of mind due to our ignorance, but the result of the laws of nature. Evidently, research into the physical origin of fluctuations in clouds is closely related to the determination of the nature of the droplet ensemble. However, to the best of our knowledge, there is no investigation into the physical origin of randomness along this line.

Finally, it is noteworthy in passing that new paradigm-shift poses mathematical challenges. Mainstream theories have established their framework within the familiar Hilbert functional space. In fact, the Hilbert space has been taken for granted in cloud physics. As implied by the least probable size distribution and its derivation, the systems theory or other new formalism needs to go beyond, moving the Hilbert functional space to the generalized functional space [96].

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