A Generalized Systems Theory for the Effect of Varying Fluctuations on Cloud Droplet Size Distributions

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ABSTRACT

A systems theory has previously been developed by Liu and Hallett to interpret droplet size distributions in turbulent clouds by utilizing ideas from statistical physics and information theory. The present paper generalizes that systems theory to allow for varying fluctuations. The generalized theory provides a self-consistent theoretical framework for a wide range of fluctuations. It reduces to that presented previously when liquid water content is conserved, and becomes consistent with the uniform growth models for nonturbulent, adiabatic clouds. The theory indicates that there exists an important characteristic scale, defined as the *saturation scale,* beyond which droplet size distributions do not change with further increases in averaging scale, but below which droplet size distributions strongly depend on the scale over which they are sampled and are therefore ill-defined without an adequate specification of scale. It is further demonstrated that the saturation scale and the details of scale dependence depend on the level of fluctuations; stronger fluctuations lead to larger saturation scales and stronger scale dependency of droplet size distributions. The potential scale mismatch leads to issues regarding the comparability between models and observations, and the direct coupling of numerical models of different scales, which in turn underscores the significance of understanding and quantifying the scale dependence of droplet size distributions. The importance of fluctuations suggests the need to measure and analyze turbulence simultaneously and at the same scales with measurements of droplet size distributions in order to provide a practical limit to the sample size required to reach the saturation scale, and to specify the effect of turbulence. The ideas presented in this paper have general applications to fields where fluctuations exist.

1. Introduction

Reliable knowledge of cloud droplet size distributions is crucial for many cloud-related areas such as precipitation, climate modeling, and remote sensing. A longstanding problem in cloud physics is that observed droplet size distributions are generally much broader than those predicted by the classical uniform model (e.g., Howell 1949). To explain this so-called spectral broadening has been a major focus of cloud physics over the last few decades, and a number of models have been proposed. Stochastic condensation theory considers the growth of droplet populations as a stochastic process and relates the spectral broadening to various fluctuations associated with turbulence (Zhou 1964; Sedunov 1974). Khvorostyanov and Curry (1999) derived from stochastic theory that gamma distributions could be used to represent ensemble-averaged droplet size distributions. Cooper (1989) derived equations for estimating the spectral width of droplet size distributions from the fluctuations of vertical velocity and integral radius, and their correlation by applying small perturbation analysis to the Lagrangian integral of the quasi-steady supersaturation averaged over an ensemble of droplets that encounter different growth trajectories through turbulent clouds. Considine and Curry (1996) 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. (1998) recently related spectral broadening to turbulence-induced preferential concentration of droplets. Much effort has been devoted to the processes of turbulent entrainment and mixing as causes of spectral broadening (Baker et al. 1980; Hicks et al. 1990; Telford and Chai 1980; Su et al. 1998). Srivastava (1989) 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

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could cause droplet–droplet variations in the microscopic supersaturation, which in turn leads to some spectral broadening. Despite their differences, all these models have one feature in common: they attempt to follow each droplet or each parcel and then take statistical averages one way or another. By analogy to the kinetic theory of gases, these models are referred to as kinetic theories of droplet size distributions throughout the rest of the paper. Although these kinetic models produce size distributions broader than those predicted by uniform models and improve the understanding of the formation of droplet size distributions, the details of the processes involved are poorly understood and highly controversial.

It has been generally accepted that the fundamental equations describing individual droplets have been well established. However, to numerically solve these equations, the size of the model grid needs to be as small as ~1 mm so that the smallest eddies of turbulence and the mean distance between droplets can be resolved and the grid values of variables such as temperature and water vapor mixing ratio represent the ambient conditions for the droplet (Vaillancourt 1998). In fact, the physical 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 kinetic equations. The difficulties are evident from the fact that the randomness of turbulence is no simpler than that of Brownian motions of molecules (Mandelbrot 1999), and that turbulent entrainment and mixing processes occur over a tremendous range of scales between the largest eddy of a cloud size and the smallest eddy of the Kolmogorov microscale (Su et al. 1998). In fact, the subject of turbulence itself has been considered one of the unsolved problems of classical physics (Nelkin 1992; Sreenivasan 1999). The mutual interactions between droplets and turbulence further complicate the problem (Malinowski and Jaczewski 1999; Vaillancourt and Yau 2000). This vexing situation is similar to the early stage of the kinetic theory of gases in the late Nineteenth and early twentieth 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.

In view of these insurmountable difficulties, a different formalism, which considers cloud droplets as a system and studies them as a whole instead of following each droplet/parcel, has been recently developed by integrating into cloud physics the ideas that have flourished in statistical physics and information theory (Liu 1992, 1995; Liu and Yang 1992; Liu et al. 1995; Liu and Hallett 1997, 1998, hereafter LH98). Just as classical statistical mechanics introduces statistical laws to remove the difficulties associated with random Brownian motions of individual molecules by concerning itself with the most probable state of a molecular system, the early systems theory focused mainly on the most probable droplet size distribution (defined as maximum likelihood size distribution and denoted by MXSD). The early systems theory predicts that the MXSD follows the Weibull distribution (Liu 1992, 1995; Liu et al. 1995; Liu and Hallett 1997). Observations are consistent with this prediction because a measured droplet size distribution averages many size distributions and, subsequently, looks more like the broad MXSD. LH98 recently generalized the systems idea by introducing and deriving the least probable size distribution (defined as minimum likelihood size distribution and denoted by MNSD) and coupling it with the MXSD. This generalization not only provides a reasonable framework for explanation of spectral broadening, but also reveals the scale dependence of droplet size distributions in turbulent clouds. Furthermore, it was argued that the discrepancy between observed and model-predicted droplet size distributions might be a manifestation of scale dependence because of the scale mismatch between theoretical models and observations.

The major objective of this paper is to further generalize the systems theory beyond that described in LH98 to allow for varying fluctuations as occur in real clouds, and thereby to lay the foundation for studying the effect of fluctuations using the systems theory approach. As will be shown, the newly generalized systems theory establishes a self-consistent theoretical framework, and offers new insights into the issues of spectral broadening and scale dependence of droplet size distributions as a function of fluctuations. For droplet systems with conserved liquid water content, it reduces to the framework presented in LH98, and for nonturbulent uniform clouds, its predictions are consistent with the uniform models. The theory also has important implications for the development of theoretical models and instrumentation, sampling strategies, and approaches to data analysis. We will further elaborate on these new challenges only briefly touched on in LH98.

2. The systems idea

The leitmotif of the systems theory approach is to obtain useful information on droplet size distributions without concern with the details of each individual droplet. This is analogous to classical statistical mechanics applied to molecular systems where properties of the system are derived without invoking the details of each individual molecule. The current systems theory mainly centers on two characteristic size distributions occurring with the maximum and minimum probability: MXSD and MNSD.

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, the MXSD as proposed, describes the most probable droplet size distributions. In this sense, the theory is better described as statistical cloud physics rather than the systems theory.

On the other hand, describing cloud droplet systems is more complex than describing 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}). However, because of very limited concentrations of cloud droplets (e.g., 100 cm^{-3}), one cannot equate the MXSD with observed or modeled droplet size distributions in many cases. Therefore, a complete characterization of a droplet system requires knowing the probabilities of all possible droplet size distributions. Although determining the specific probability of each possible size distribution seems prohibitive at present, useful information can be obtained by knowing the MNSD in addition to the MXSD. If the MNSD is identical with the MXSD, clouds are expected to be absolutely uniform, and the uniform model suffices. The equivalent model would be a uniform updraft with all droplets exposed to the same supersaturation and identical cloud condensation nuclei (CCN). However, such idealized situations hardly occur in nature. If there are any differences between the MXSD and MNSD, individual size distributions then depend on the scale over which they are averaged. As discussed in LH98, there are striking contrasts between the MXSD and the MNSD in turbulent clouds, and the resulting scale dependence of droplet size distributions, in fact, lies at the root of many unsolved problems of cloud physics, including the long-standing issue of so-called spectral broadening. The scale dependence poses new challenges to the development of numerical models as well as to instrumentation and sampling strategies. Furthermore, as will be addressed later in this paper, the involved complexity is closely related to fluctuations associated with turbulence.

3. Quantitative formulation

In LH98, the MXSD and the MNSD were derived for a special droplet system with conserved liquid water content. However, the assumption of conserved liquid water content does not always hold. A variety of fluctuations can occur in real clouds, for example, ranging from strongly turbulent cumulus to weakly turbulent stratocumulus, to laminar lenticular clouds. It is therefore desirable to incorporate varying fluctuations into the systems formulation. Liu and Hallett (1997) extended the MXSD from the special droplet system to a more general one that allows for different fluctuations. In this paper, we further extend the MNSD presented in LH98 to this general droplet system, and thereby establish a generalized systems framework applicable to a wide range of fluctuations by combining the general MNSD with the general MXSD presented in Liu and Hallett (1997).

For completeness, the general droplet system discussed in Liu and Hallett (1997) is briefly outlined here. The general droplet system is controlled by two constraints:

$$\int \rho(x)\, dx = 1, \tag{1a}$$

$$\int x\rho(x)\, dx = \frac{X}{N}, \tag{1b}$$

where x, defined as the restriction variable, is related to the physical processes controlling the droplet system, X is the total amount of x per unit volume, $n(x)$ is the droplet concentration per unit volume per unit x interval, N is the total droplet concentration, and $\rho(x) = n(x)/N$ can be considered the probability that a droplet of x occurs. It should be noted that the correspondence between x and the conservation law is a key to the generalization. For example, for the special droplet system discussed in LH98, which is constrained by the conservation of liquid water content, x represents the mass of a droplet of diameter D, X is the liquid water content, and $n(x)$ the droplet concentration per unit mass interval. The power-law relationship between the mass and the diameter of a droplet is readily generalized as

$$x = aD^b, \tag{2}$$

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.

Liu and Hallett (1997) derived the MXSD for this system. Briefly, by analogy with the Boltzmann entropy for molecular systems and the Shannon–Jaynes entropy generalized for complex multibody systems, spectral entropy H is defined as

$$H = -\int \rho(x) \ln[\rho(x)]\, dx. \tag{3}$$

The MXSD is the droplet size distribution that maximizes (3), subject to the constraints described by (1a) and (1b). By solving the corresponding variational problem, the MXSD was derived to be the Weibull distribution:

$$n_{\max}(D) = N_0 D^{b-1} \exp(-\lambda D^b), \tag{4}$$

where the parameters $N_0 = ab/\beta$ and $\lambda = a/\beta$, and $\beta = X/N$. Note that the β here is the inverse of that used in Liu and Hallett (1997) and represents the mean value of X per droplet. This change makes the physical meaning of β consistent with that of "K_BT" in the Boltzmann energy distribution (K_B is the Boltzmann constant, T is the temperature, and K_BT essentially represents the

mean energy per molecule in the gas). The derivation of (4) also uses the power-law relationship (2).

The MNSD is associated with the populational energy change (E) to form a population of droplets with $n(D)$. In LH98, E is expressed as

$$E = -\frac{\pi\rho_w L}{6}\int D^3 n(D)\, dD + \pi\sigma\int D^2 n(D)\, dD + c, \quad (5a)$$

where the first term on the right-hand side is the latent energy with L representing the latent heat of water; 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 (5a) is 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 (Pruppacher and Klett 1978). In fact, these minor terms can be incorporated into the coefficients before the integrals,

$$E = c_1\int D^3 n(D)\, dD + c_2\int D^2 n(D)\, dD + c. \quad (5b)$$

The coefficient $c_1 = [(\pi\rho_w)/6(-L + gh + 1/2\overline{V_t^2})]$ (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 (L), 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. Another assumption for both (5a) and (5b) is that the amount of energy change is completely exchanged with the environment by processes such as heat conduction. Maximizing E given by (5b) subject to the constraints described by (1a) and (1b), the general MNSD is derived as (see appendix A)

$$n_{\min}(D) = N\delta(D - D_b), \quad (6a)$$

$$D_b = \left(\frac{\int D^b n(D)\, dD}{N}\right)^{1/b} = \left(\frac{X}{aN}\right)^{1/b}. \quad (6b)$$

Similar to the special MNSD presented in LH98, the general MNSD is also a δ-function. However, the similarity ends here. Derivation of the general MNSD requires use of the general restriction variable satisfying (2); only the special case with $a = [1/(6\pi\rho_w)]$ and $b = 3$ was used in LH98. This generalization is not as trivial as it may seem. Mathematically, the derivation requires a combination of the calculus of variations and the generalized function theory, whereas the mathematics for the special case presented in LH98 is rather simple. Physically, as will become evident, the generalization permits us to study the effect of varying fluctuations on properties of droplet size distributions. Furthermore, an examination of (4), (6a), and (6b) reveals that the MXSD and MNSD presented in LH98 for the droplet system with conserved liquid water content are just special cases of the MXSD and the MNSD given above with $a = [1/(6\pi\rho_w)]$, $b = 3$, and X = liquid water content. Note that the general MNSD also corresponds to the minimum spectral entropy $H = 0$.

4. Important implications

The striking spectral differences between the MXSD and the MNSD in turbulent clouds have been discussed in LH98. Briefly, given X and N, the MXSD and the MNSD represent the most and the least probable way to distribute X among the N droplets, respectively. The MXSD is much broader than the MNSD. Based on the fact that very narrow droplet size distributions have indeed been observed under uniform conditions such as in adiabatic cores of cumulus clouds and in uniform lenticular clouds, we speculated in LH98 that the spectral differences between the MXSD and the MNSD depends on fluctuations in clouds and should decrease as fluctuations decrease. This speculation can now be justified by the generalized systems theory presented above.

An analysis of the Weibull distribution as described by (4) shows that it becomes narrower with increasing values of the parameter b, and eventually approaches the monodisperse MNSD as described by (6a) when b approaches ∞ (see appendix B for the mathematical proof). This result indicates that the MXSD approaches the MNSD with decreasing fluctuations, because b is closely related to fluctuation levels in clouds, and increases with decreasing fluctuations (Liu and Hallett 1997). The dependence on fluctuations of the MXSD and the MNSD, along with their other unique properties, have profound implications for many fundamental issues of cloud physics, for cloud-related models such as climate models, for instrument design, and for sampling strategies.

a. Scale dependence and spectral broadening

In LH98 we discussed the scale dependence of droplet size distributions in turbulent clouds because of the striking differences between the MXSD and the MNSD for the special droplet system where liquid water content is conserved. We also pointed out that the long-standing issue of spectral broadening might be just a manifestation of the scale dependence as a result of the scale mismatch between theoretical models and observations. These arguments are reinforced and extended by the generalized systems theory presented above in the following aspects.

First, the scale dependence of droplet size distributions indicates that an individual size distribution approaches the MXSD with an increase in the averaging

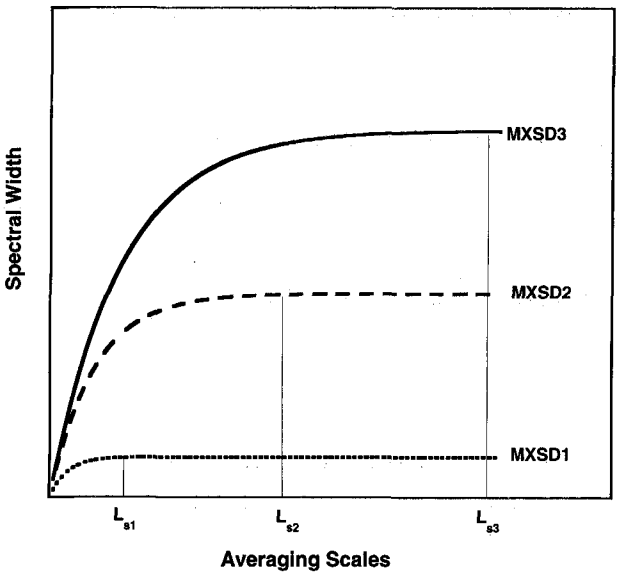

FIG. 1. A diagram illustrating the scale dependence of individual droplet size distributions at different levels of fluctuations. The meanings of both coordinates are only qualitative. The fluctuation increases from case MXSD1 to case MXSD3. The MXSD represent the maximum likelihood size distribution. This figure illustrates the following points addressed in the paper: 1) observed size distributions approach the MXSD with the increase in averaging scales; 2) there exists a characteristic scale, defined as *saturation scale* and denoted by L_s, beyond which droplet size distributions do not change much with further increases in averaging scales, but below which size distributions change rapidly with averaging scales; 3) the stronger the fluctuation the larger the macroscopic scale ($L_{s_3} > L_{s_2} > L_{s_1}$); 4) the spectral width of droplet size distributions increases with increases in averaging scales at a given level of fluctuations, and increases with increasing fluctuations at a given scale.

scale. It is anticipated from this behavior of scale dependence that there exists a characteristic scale, defined as the *saturation scale,* beyond which all size distributions are approximately the same and equal to the MXSD. However, droplet size distributions are strongly dependent on the averaging scale when it is less than the corresponding saturation 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. Similar findings were reported for raindrop size distributions in studying fluctuations of precipitation (Joss and Gori 1978; Liu 1993; Smith and Liu 1993; Ulbrich and Atlas 1998; Kostinski and Jameson 1997, 1999; Jameson and Kostinski 1998, 1999, 2000; Jameson et al. 1999).

Second, it is well known that the degree of spectral broadening depends on the processes of entrainment and mixing determined by turbulence. Very narrow droplet size distributions are often observed in clouds formed under uniform conditions, for example, 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 MNSD. However, as indicated by the generalized systems theory, this is actually due to the fact that the MXSD approaches the MNSD with decreasing fluctuations. Furthermore, real clouds are always in a more or less turbulent state. Even in nonturbulent uniform clouds with uniform CCN, the Poisson random spatial distribution of droplets can cause fluctuations in the microscopic supersaturation (Srivastava 1989), an essential variable controlling the condensation/evaporation of individual droplets. These facts suggest that the extremely narrow size distributions of the δ-function will seldom be observed, no matter how uniform the cloud is. Application of the generalized systems theory to the inevitable small fluctuations provides an explanation for the phenomenon that very narrow droplet size distributions, yet still broader than those predicted by uniform models, are often observed in the so-called adiabatic cores (Brenguier and Chaumat 2001). Similar to spectral broadening, the saturation scale and the details of the scale dependence of droplet size distributions also depend on fluctuations. The weaker the fluctuation, the smaller the difference between the MXSD and the MNSD, the smaller the saturation scale, and the weaker the scale dependency. The scale dependence and spectral broadening will eventually disappear when there are no fluctuations.

Figure 1 schematically illustrates the feature of scale dependence of droplet size distribution as a function of fluctuations derived from the generalized systems theory. For a given level of fluctuation, droplet size distributions approach the MXSD with increases in the averaging scale. The spectral change is fast and individual size distributions are ill-defined when the averaging scale is less than the saturation scale. The change slows down as the saturation scale is approached, and eventually becomes approximately zero beyond the corresponding saturation scale. Furthermore, the variability of a mean size distribution is expected to decrease with the averaging scale. For a given averaging scale, the MXSD–MNSD difference and the saturation scale increase with increasing fluctuations. Evidently, the averaging scale is an important quantity in defining droplet size distributions in a fluctuating environment. Because of the potential scale mismatches, the comparability is questionable between models and measurements having different scales, and so is the direct coupling of models of different scales. Although the issue of potential scale mismatch has been realized for sometime [see Hallett (1996) for instrumentation, Cotton et al. (1995) for model development, and LH98 for a general discussion], the issue of scale dependence has not been rigorously addressed. In the following, we will expand on the implications of, and challenges posed by, the scale dependence and associated ill definedness of droplet size distributions as a function of fluctuations.

b. Model development

Representation of droplet size distributions is crucial for a variety of cloud-related models (Cotton and Anthes 1989). Cloud parameterization in climate models has become a topic of intensive research because of the role of clouds in regulating the earth's radiation balance and the hydrological circle (Chahine 1992; Schwartz and Slingo 1996). To improve cloud parameterizations, it is increasingly common to couple dynamical and radiative models with microphysical models as detailed as allowed by computer resources (Ackerman et al. 1995). 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 in turbulent clouds. For example, 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. Even if the scales of droplet size distributions involved in models of both kinds are compatible (e.g., when clouds are uniform), detailed microphysical models are still computationally expensive. In fact, we would argue that the MXSD should be used to represent droplet size distributions in climate models because of the large scales involved. Recently, simple microphysical models have been developed based on the methods of moment by assuming size distributions follow an analytical form such as a gamma distribution (Mitchell et al. 1996; White and Hounslow 2000). Besides being simple and computationally cheap, such moment-based microphysical models could be more physically sound compared to complicated models with detailed microphysics, because the distribution functions are often obtained by empirically analyzing a large number of data and the resultant distributions could be close to the MXSD.

Assuming an analytical form for droplet size distributions is also a common practice in the parameterization of cloud microphysics in cloud models (Cohard and Pinty 2000). However, it is not clear whether the scales involved in such models are large enough to meet the requirement of the saturation scale. When the averaging scales involved are less than the saturation scale, treating droplet size distributions becomes a more complicated and vexing problem. As discussed above, size distributions are ill defined without specifying the scale involved under this condition. This unique property suggests the necessity of making explicit references to the averaging scale in numerical models. In other words, a new theoretical formalism is necessary to treat scale as an independent variable, just as the current formalism treats the variables of space and time. Similar challenges have been realized in other disciplines (Nottale 1996; Herrmann 1997; Glasser and Goldhirsch 2001). 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.

Furthermore, the vital role of fluctuations in determining the details of the scale dependence of droplet size distributions, and their close relationship with turbulence, demands better understanding and consideration of cloud turbulence. Even for the MXSD, this study strongly suggests that turbulence plays a critical role in determining its shape parameter b because of its close relationship to the spectral dispersion of the cloud droplet size distribution (Liu and Daum 2000b). Our recent studies have shown that the specification of turbulence is crucial for the parameterization of effective radius, a critical variable used to calculate radiative properties of clouds (Liu and Daum 2000a,b). Although it has been long speculated that turbulence affects cloud droplet size distributions, and the effect of turbulence has been studied rather extensively in cloud physics, turbulence and related scale issues are poorly represented (if at all) in current models.

c. Instrumentation, sampling strategy, and data analysis

The scale dependence and ill definedness of individual size distributions and their relationships with fluctuations also pose challenges to instrumentation, sampling strategies, and data analysis. On one hand, new instruments are needed that are capable of characterizing the finest physical structures of droplet size distributions (e.g., ~1 mm of the smallest turbulent eddies). This is particularly important in regions of sharp discontinuity (Hallett and Hallett 2000). On the other hand, the averaging scale needs to be large enough to obtain a well-defined droplet size distribution, in other words, to reach the corresponding saturation scale.

In general, instrument design defines a sampling volume, and instrument motion defines a sampling strategy in making any practical physical or chemical measurement in the atmosphere. Thus, an aircraft sweeps out a volume along its path; a ground instrument samples a volume of air defined by the local airflow. Both are dependent on the rate of processing of the fluid medium in the sensor itself. For a given instrument, further choice lies in not only the sampling time and but also the spatial scale of the sample. Is there motion relative to the fluid medium (atmosphere or ocean), or does the instrument follow the parcel in the Lagrangian sense? Superimposed on these issues is the time response of the sensor to change, and the rate at which the changing properties of the local environment are communicated to the sensor.

Measurement of particle size distributions raises particularly difficult problems because, in general, particle concentration is highly size dependent, there tends to be more smaller particles, and there may be spectral peaks that depend on different physical processes. Two separate questions are identified. First, even in a uni-

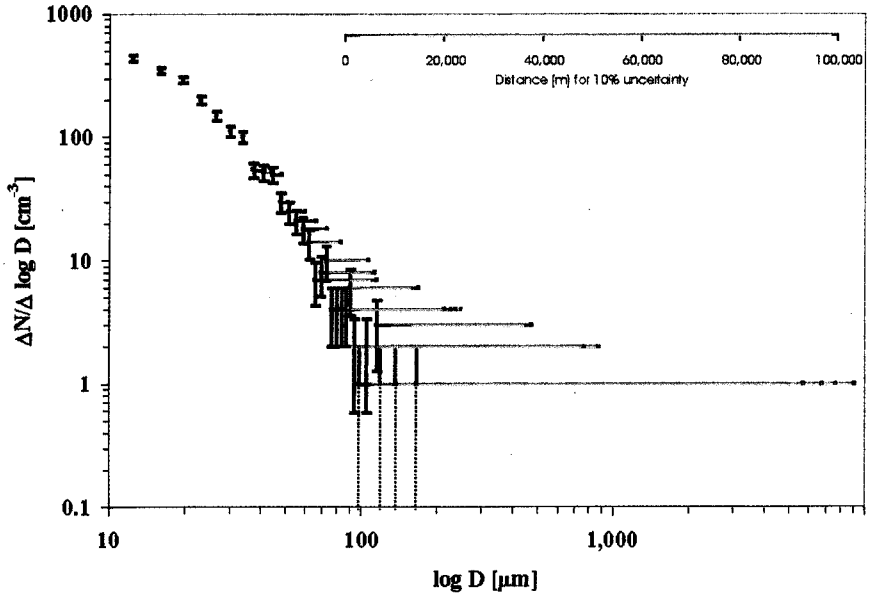

FIG. 2. Analysis routine for a particle size distribution collected by a cloudscope on the NASA DC-8 (sampling volume is 50 $cm^3 s^{-1}$) in outflow of Hurricane Earle approaching the Louisiana coast, 1998. The protocol is set by the selection of the number of size bins on a logarithmic scale. It is further set by the level of uncertainty—for example $\pm 10\%$. The uncertainty in the number of particles actually counted in each bin is given by Poisson statistics, represented by the vertical error bars on each point. The uncertainty is large for small concentrations of large particles. The horizontal line from each point represents a flight distance necessary to sample 100 ± 10 particles. The physical domain over which any set of measurements must be analyzed is then selectable. This could be (as this case) 10 km of a hurricane outflow; 1000 km of marine stratus or the whole North Pacific Ocean. In either case, the criterion for an overall sampling time or distance will be ultimately set by consideration of Fig. 1. It will be composed of a multitude of data selected according to the Poisson statistics of this figure but with a scale determined by the variability imposed by the limitation of the physical process considered—on the largest scale the whole atmosphere, on the smallest scale an adiabatic core with uniform everything, subject only to random spatial and time distribution of particles.

form, unchanging environment, particles are subject to the usual Poisson statistics of distribution. A size distribution is defined in terms of a number of particles in a selected bin width (linear or logarithmic) and therefore a measurement of N particles in a given bin is uncertain to $\pm N^{1/2}$. Somewhere in the spectrum a bin has but one particle, with an uncertainty of ± 1. These considerations apply also to higher moments as particle mass, area, or radar reflectivity. Obviously the statistics may be improved by sampling for a longer time but this merely extends the problem to a larger (lower concentration) size. A different approach lies in asking for how far or how long a sample must be taken to obtain an acceptable uncertainty—say, $\pm 10\%$. Figure 2 shows how such an approach may be used from a practical viewpoint in specifying and comparing spectra. The vertical bars on each point relate to the Poisson uncertainty of the actual number of particles in that bin; the horizontal lines from each point relate to how far the aircraft must fly for this particular instrument to achieve an uncertainty of $\pm 10\%$. Application is to the simplest adiabatic, uniform situation in which the MXSD is approximately equal to the MNSD of a δ-function. In this case, identifying the scale of satisfactory Poisson counting statistics may be enough.

The second problem lies in applying this analysis to the real, turbulent atmosphere. The length of time–distance to be sampled needs to first provide a 10% uncertainty for the largest size (smallest concentration) of interest, but, most important, to provide the saturation scale as shown in Fig. 1 so that the MXSD is approached. Note that recent studies have indicated Poisson statistics may not hold in turbulent clouds (Kostinski and Jameson 2000; Burnet and Brengier 2000). A sampling strategy exists for each case to obtain satisfactory measurements of spectra and to fulfill the criteria of Fig. 1. In practice this means that data for the cloud field with the same level of fluctuations would be used for the analysis. The prescribed field may be associated with different upper limit of the physical scale. If the upper limit of the physical scale is less than the saturation scale, repeated sampling of the same portion or portions of clouds defined by similar dynamics is then necessary to meet the requirement of saturation scale. Repeated sampling may be needed particularly in strongly turbulent, narrow regions of cumulus clouds in regions of

strong shear. When clouds or parts of clouds such as a growing turret are not stable enough to permit repeated penetrations, sampling of other similar clouds may be necessary to reach the saturation scale. Herein lies a decision of practicality for making an observation. Uniformity is to be translated from the Poisson uncertainty in a completely nonturbulent environment, to an environment whose fluctuations can be prescribed from physical or chemical considerations. Thus a cumulonimbus anvil, a field of marine stratocumulus cloud, or a widespread field of cumulonimbus may be chosen as the field of interest. The ultimate limit is evidently the whole atmosphere. The criterion for reaching the saturation scale should be investigated empirically by seeking the limit of measurement, which become invariant with further addition of data. In summary, not only are instruments needed that are capable of identifying the finest structure, but also sampling strategy has to meet the requirement of reaching the saturation scale.

Furthermore, even with satisfactory measurements available, extensive analysis is needed to fill in the details theoretically illustrated in Fig. 1. However, except for the uniform case, the saturation scale and the scale dependence are largely unknown. Application of such methodologies as multiscale analysis (Smith and Liu 1993; Liu et al. 1996; Davis et al. 1996; Kostinski and Jameson 1997, 1999; Jameson and Kostinski 1998, 1999, 2000; Jameson et al. 1999; Miles et al. 2000) to datasets sampled at different levels of fluctuations are necessary to fill this gap. After identifying the saturation scale, the relationship between the exponent b and the level of fluctuations can be empirically established by investigating the corresponding size distributions and their relationship with turbulent intensities (e.g., turbulent kinetic energy and dissipation rate).

Cloud properties and size distributions can also be measured by use of remote sensing techniques such as radars (Atlas et al. 1973; Heymsfield et al. 1996; Goddard et al. 1997). Because remote sensing techniques typically sample large volumes, measured size distributions are expected to be close to the MXSD. Recently, remote sensing techniques have been developed to measure droplet size distributions as well as turbulent properties such as velocity variance and turbulent dissipation rate (Babb and Verlinde 1999). This provides another way to relate the exponent b to turbulence properties. Furthermore, a variety of in situ and remote sensing instruments (surface-, aircraft-, and satellite-based) are often coordinated in current field projects to address cloud-related issues because of the wide range of scales involved. An important issue with this approach is the mutual comparison and validation of measurements made by these instruments operated at different sampling scales. Ideas presented here may be used for this purpose as well.

5. More comparisons with kinetic models

As discussed in the introduction, 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. Mathematically, unlike kinetic models formulated using differential equations, the systems theory is built upon calculus of variations, integral equations, and constrained optimization. Furthermore, averaging is used one way or another in kinetic models to explain observed droplet size distributions. Typical examples are those studies that generate many parcels and predict size distributions for each parcel and then average size distributions predicted for all the parcels (Feingold et al. 1998). The averaging processes of kinetic models are computationally extensive. A more fundamental difficulty arises from the wide range of scales involved in entrainment and mixing processes in turbulent clouds (Krueger et al. 1997). In contrast, as in statistical mechanics, the systems theory introduces statistical laws for fluctuations and simply relates the large-scale average to the MXSD, and provides an analytical way of averaging.

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 at 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 various disciplines to study complex systems (Haken 1977). 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 observed phenomena such as spectral broadening and scale dependence. The Weibull size distributions predicted as the MXSD by the systems theory have been demonstrated to be consistent with observed droplet size distributions (Costa et al. 2000; Liu and Daum 2000a,b). Miles et al. (2000) recently reported observational evidence supporting the scale dependence of droplet size distributions predicted from the systems theory.

6. Concluding remarks

The systems theory presented in LH98 is generalized to allow for varying fluctuations in clouds. The general MXSD and MNSD are, respectively, a general Weibull distribution and a general δ-function. The generalized theory provides a self-consistent theoretical framework for a wide range of conditions. It reduces to those presented in LH98 when liquid water content is conserved, and becomes consistent with the uniform growth models

for nonturbulent adiabatic clouds. The generalized systems theory reveals the following important points. First, droplet size distributions depend on the scale over which they are observed or simulated, and approach the MXSD with increases in averaging scale. Second, there exists a characteristic scale, defined as the *saturation scale,* beyond which observed size distributions do not change much because of their closeness to the MXSD. Third, individual droplet size distributions are strongly scale dependent when the sampling scale is less than the saturation scale, and therefore ill defined without specification of the averaging scale. Fourth, the difference between the MXSD and the MNSD, scale dependence, and the saturation scale decreases with decreasing fluctuations, and eventually disappear under uniform conditions. The absolute uniform state characteristic of the minimum spectral entropy and maximum populational energy change is analogous to the state of absolute zero for atomic systems, which assumes the minimum thermodynamic entropy and is prohibitively difficult to realize.

The generalized systems theory provides explanations for a number of long-standing issues. First, it is shown that spectral broadening can be understood in the framework of scale dependence, arising as a result of scale mismatch between models and observations. 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, are due to the small, yet inexorable fluctuations affecting cloud droplets even in adiabatic cores, which will cause small differences between the MXSD and MNSD.

Implications of the scale dependence and ill definedness of droplet size distributions in turbulent clouds are discussed. In general, caution is needed when comparing observed droplet size distributions with those simulated by models, when coupling models of different scales, and when comparing measurements collected using instruments with different sampling scales. The unique property of scale dependence in turbulent clouds suggests the necessity of making explicit references to the averaging scale in numerical simulations as well as in observational studies. In particular, it is argued that to represent clouds in climate models, the simple moment-based microphysical models could be more accurate than the complicated models with detailed microphysics because of the large scales involved in climate models. Furthermore, the unique property of scale-dependence suggests the ultimate need for a 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. It is not clear to us now how to realize this. But, it seems that a combination of the systems idea with multiscale approaches may be a promising avenue. In terms of instrumentation and sampling strategies, it is suggested that instruments capable of detecting structures at the resolution of the smallest turbulent eddies (e.g., 1 mm) are needed. Repeated or large volume sampling of the same cloud or similar clouds may be necessary to reach the saturation scale for clouds that have small physical scales, for example, small cumulus. Similarly, if regions of specific characteristics are selected for analysis—for example regions of strong shear between up- and down- drafts, such regions must have sufficient volume to satisfy the criterion for the saturation scale. Thus, the rationale for comparison of models and observations or intercomparison of models and observations themselves requires attention to not only the Poisson criteria for particle measurements, but also the scale dependence and the saturation scale in a turbulent environment.

The present work highlights the importance of fluctuations in determining droplet size distributions and their scale dependence. However, quantitative studies along this line are very limited. Because fluctuations are closely related to turbulence, simultaneous measurements and analysis of turbulence and droplet size distributions, at the same time, and spatial resolution are needed to provide some measure of the saturation scale and the form of the scale dependence. The term of "fluctuation" is used in this paper in a generic sense. No assumption on specific mechanisms causing fluctuations is made since we think there may exist a number of mechanisms that cause fluctuations and these mechanisms are not well understood. Such mechanisms include turbulence-related processes such as entrainment and mixing. The differences in CCN properties (e.g., sizes, chemical compositions, and concentrations) may cause fluctuations as well.

It is noteworthy that there may also be utility in applying the arguments presented in this paper for comparison of dynamical systems characterized by motions and particle distributions in an astronomical-scale setting where fluctuations exist.

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APPENDIX A

Derivation of the General Minimum Likelihood Size Distribution

To determine the MNSD, $n_{\min}(D)$, is a problem of the calculus of variation, and requires the generalized func-

tion theory because of the involvement of the δ function. The details of the generalized function theory are referred to in Gelfand and Shilov (1964), Zemanian (1965), and Keener (1995). Only the definitions and operations relevant to this paper are outlined here.

- The inner product of two functions $f(D)$ and $g(D)$ is defined as

$$\langle f(D), g(D)\rangle = \int f(D)g(D)\, dD.$$

- A test function, denoted by $\varphi(D)$, is a function such that it is continuously differentiable infinitely and has compact support.
- A generalized function is defined as a continuous linear function acting on $\varphi(D)$, denoted by the inner product $\langle f(D), \varphi(D)\rangle$.
- Two generalized functions $f_1(D)$ and $f_2(D)$ are said to be the same if, for a test function $\varphi(D)$, we have

$$\langle f_1(D), \varphi(D)\rangle = \langle f_2(D), \varphi(D)\rangle. \tag{A1}$$

- The derivative of a generalized function is defined by the equation

$$\left\langle \frac{df}{dD}, \varphi(D)\right\rangle = -\left\langle f(D), \frac{d\varphi}{dD}\right\rangle. \tag{A2}$$

The constraints as described by (1a) and (1b) for the general droplet system can be rewritten as

$$\int n(D)\, dD = N, \tag{A3}$$

$$\int D^b n(D)\, dD = ND_b^b, \tag{A4}$$

where $D_b = [\int D^b n(D) dD/N]^{1/b}$ is defined as the bth diameter. The MNSD is the characteristic distribution that maximizes the populational energy change defined by (5b) subject to (A3) and (A4). Following the general procedure of variational calculus, we construct the specific Lagrangian functional as

$$F[n(D)] = c_1 \int D^3 n(D)\, dD + c_2 \int D^2 n(D)\, dD - \lambda_1\left[\int n(D)\, dD\right] - \lambda_2\left[\int D^b n(D)\, dD\right].$$

Setting the first variation of $F[n(D)]$ with respect to the unknown $n(D)$ equal to zero, we have

$$\Delta F = \int (c_1 D^3 + c_2 D^2 - \lambda_1 - \lambda_2 D^b)\Delta n(D)\, dD = 0, \quad \text{or}$$

$$\Delta F = \int (c_1 D^3 + c_2 D^2 - \lambda_1 - \lambda_2 D^b)\frac{dn(D)}{dD}\, dD = 0. \tag{A5}$$

Solving (A5) utilizes the knowledge of the generalized function as introduced above. When the test function is chosen such that

$$\varphi(D) = \begin{cases} 0, & D > D_{\max} \\ c_1 D^3 + c_2 D^2 - \lambda_1 - \lambda_2 D^b, & D_{\min} \le D \le D_{\max} \\ 0, & D < D_{\min}, \end{cases} \tag{A6}$$

Eq. (A5) is equivalent to the following inner product:

$$\left\langle \frac{dn}{dD}, \varphi(D)\right\rangle = 0. \tag{A7}$$

Using (A2) for the derivative of a generalized function, we further have

$$\left\langle \frac{dn}{dD}, \varphi(D)\right\rangle = -\left\langle n(D), \frac{d\varphi}{dD}\right\rangle = 0. \tag{A8}$$

Therefore,

$$\int_{-\infty}^{\infty} n(D)\frac{d\varphi}{dD}\, dD = \int_{D_{\min}}^{D_{\max}} (3c_1 D^2 + 2c_2 D - b\lambda_2 D^{b-1})n(D)\, dD = 0. \tag{A9}$$

Given a generalized function

$$n^+(D) = K\delta(D - D_*), \tag{A10}$$

where $D \in (D_{\min}, D_{\max})$, we have

$$\int n^+(D)\frac{d\varphi}{dD}\, dD = K(3c_1 D_*^2 + 2c_2 D_* - b\lambda_2 D_*^{b-1}) = 0.$$

Without loss of generality, we can always choose a λ_2 such that

$$\left\langle n^+(D), \frac{d\varphi}{dD}\right\rangle = 0.$$

Therefore, we have

$$\left\langle n^+(D), \frac{d\varphi}{dD}\right\rangle = \left\langle n(D), \frac{d\varphi}{dD}\right\rangle. \tag{A11}$$

Based on the definition of equality of two generalized functions by (A1), the MNSD is

$$n_{\min}(D) = n^+(D) = K\delta(D - D_*). \tag{A12}$$

The constant K is determined by substituting (A12) into (A3):

$$\int n(D)\, dD = \int K\delta(D - D_*)\, dD = K = N, \tag{A13}$$

where D_* is given by substituting (A12) and (A13) into (A14):

$$\int D^b N\delta(D - D_*)\, dD = ND_*^b = ND_b^b, \quad \text{or}$$
$$D_* = D_b. \tag{A14}$$

Therefore the MNSD is given by

$$n_{\min}(D) = N\delta(D - D_b). \tag{A15}$$

APPENDIX B

The Approach of the MXSD to the MNSD

Equation (4) can be rewritten as

$$n_{\max}(D) = \frac{N(b-1)}{D_0^b} D^{b-1} \exp\left[-\frac{b-1}{b}\left(\frac{D}{D_0}\right)^b\right], \tag{B1}$$

where $D_0 = ((b-1)/b)^{1/b}(X/aN)^{1/b}$ is the mode diameter of the droplet (Deirmendjian 1964).

When $D = D_0$, (B1) becomes

$$n_{\max}(D_0) = \frac{N(b-1)}{D_0} \exp\left(-\frac{b-1}{b}\right) \tag{B2}$$

As b approaches ∞, the ratio $(b-1)/b$ approaches 1, and therefore $n_{\max}(D_0)$ approaches ∞. When $D \neq D_0$, it is obvious that $n_{\max}(D)$ approaches 0 as b approaches ∞. Together with the fact that D_0 approaches D_b as b approaches ∞, we have that $n_{\max}(D)$ approaches the MNSD, $n_{\min}(D) = N\delta(D - D_b)$ as b approaches ∞.

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