On the Generalized Theory of Atmospheric Particle Systems

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ABSTRACT

Unification is both necessary and challenging for studying atmospheric particle systems, which are polydisperse systems containing particles of different sizes and shapes. A general framework is proposed to realize the first order generalization. Within this generalized framework, (1) atmospheric particle shapes are unified into self–similar fractals; (2) a self–similar particle is characterized by various power–law relationships; (3) by combining these power–law relationships for a single particle with Shannon's maximum entropy principle and some concepts in statistical mechanics, unified maximum likelihoood number size distributions are of the Weibull form for atmospheric particle systems. Frontier disciplines (e. g., scaling, fractal, chaos and hierarchy) are argued to provide potential "tools" for such unification. Several new topics are raised for future research.

Key words: Unification, Self–similarity, Power–law relationships, Fractals, Shannon's entropy, Maximum likelihood distribution

I. INTRODUCTION

Further understanding of atmospheric particles and their systems calls for a unified treatment. It has been traditional to identify several elements in the atmosphere; air molecules, aerosol particles and hydrometeors (cloud and precipitation particles). However, many issues related to the atmosphere require an overall consideration of effects from different sources. For example, an ultimate understanding of the global warming requires taking green house gases such as water vapor and carbon dioxide, aerosol particles and hydrometeors into account. Moreover, all the atmospheric particles interact non–linearly and depend upon one another. Gas–to–particle conversions are important sources of atmospheric particles whereas particles are a link in the chain of the removal processes which returms gaseous pollutants to the earth's surface (Twomey, 1977). These interactions are complicated by the fact that some aerosol particles may work as cloud condensation nuclei (CCN) or ice nuclei (IN) to form cloud particles and evaporation of cloud particles may leave modified aerosol residues in the air. Small particles (small aerosol particles or cloud particles) may cluster to form large particles (large aerosol particles or precipitation particles) and large particles may break up or evaporate into small particles. Hydromteors also may scavenge aerosol particles (rainout and cloudout) (Pruppacher and Klett, 1978). A summary of these interactions is given in Fig. 1. Therefore, it is physically desirable to describe the atmosphere as a self–consistent whole, rather than a collection of neatly independent and ultimately simple pieces.

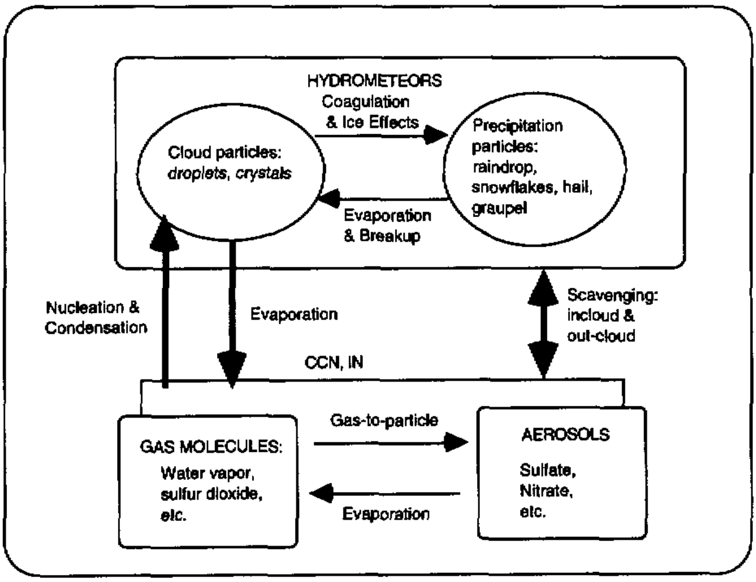

Fig. 1. Schematic show of interactions among different atmospheric particle groups.

On the other hand, as Maxwell's dictum states, "The great desideratum for any science is its reduction to the smallest number of dominating principles", the parsimony of basic concepts and models has been a target throughout the long history of mankind's search for an understanding of the physical world. Such economy, is a prerequisite for exploring complexity, dictates unification.

Atmospheric "particles" covers roughly 10 orders of magnitude in size ($\sim 10^{-8}$ cm of molecules and their clusters to $\sim 10^{1}$ cm of hailstones), with each group of "particles" within a limited range. At first glance, the wide size coverage, plus their shape multiplicities, seems to forbid a unified treatment. But, atmospheric scientists have to face this "complexity" just like physicists challenging the grand unification theory (a theory of unifying the four basic forces: gravity, electromagnetic, weak and strong forces). Previous studies have shown some promising results; details are referred to Liu et al. (1995), Liu (1994) and Liu, Mitchell and Arnott (1994). In those papers, discussed respectively were unified treatments of particle number distribution, of dynamical properties (terminal velocities) and of particle shapes. Also, rapid development of some frontier disciplines (e. g., self–similarity, scaling, fractals, chaos and non–linearity) provides the potential to realize this unification. The purpose of this paper is to propose a general unifying fremework by synthesizing and developing previous studies.

This paper is organized as follows. A summary of Liu, Mitchell and Arnott (1994) is given in Section 2; a summary of Liu (1994) is given in Section 3; a summary of Liu et al. (1995) is given in Section 4. The three parts are then synthesized and developed, yielding the general unifying framework in Section 5. Implications are made in Section 6. Qualitative reasoning for self–similar structures is made in Section 7, with emphasis on links with both self–similar atmospheric fields and models of colloidal aggregation. Section 8 is concluding remarks.

II. UNIFIED TREATMENT OF PARTICLE SHAPE

Atmospheric particles assume various geometrical shapes: Euclidean particles (e. g., spheres, spheroids and various symmetrical crystals) and "irregular" particles. Shapes of hydrometeors are more complicated than those of aerosols because the coexistence and interactions of both liquid phase particles such as cloud droplets and solid phase particles such as ice crystals. Such shape multiplicities are implicated by Lee–Mogono classification scheme (Lee and Mogono, 1968), in which about 80 solid hydrometeors are classified. However, such qualitative description does not meet the requirements of further developments, which calls for quantitative characterization. Furthermore, a unified treatment of particle shapes requires an approach which can deal with "irregular" as well as Euclidean particles.

Since the publication of fractal geometry (Mandelbrot, 1977; 1983), its ability for quantitatively characterizing irregularity has been demonstrated by numerous studies in diverse fields (Mandelbrot, 1983; Pynn and Skjeltorp, 1985; Milne, 1988; Peitgen and Saupe, 1988; Avnir, 1989; Falconer, 1990; Sugihara and May, 1990). Further, fractal dimension can be shown to be a generalization of Euclidean dimension by using self–similarity concept. This means that atmospheric particle shapes, to first approximation, can be unified into self–similar "fractals", with Euclidean particles as special cases (see Fig. 2). This issue was detailed in Liu, Mitchell and Arnott (1994); and is briefly introduced as follows.

1. *Unification of Particle Dimension*

Self–similarity, or invariance against changes in scale or size, is one of the central concepts in fractal geometry. Mathematically speaking, a power–law relationship, $Y = ax^b$, is self–similar: if x is rescaled (multiplied by a constant), then y is still proportional to x^b.

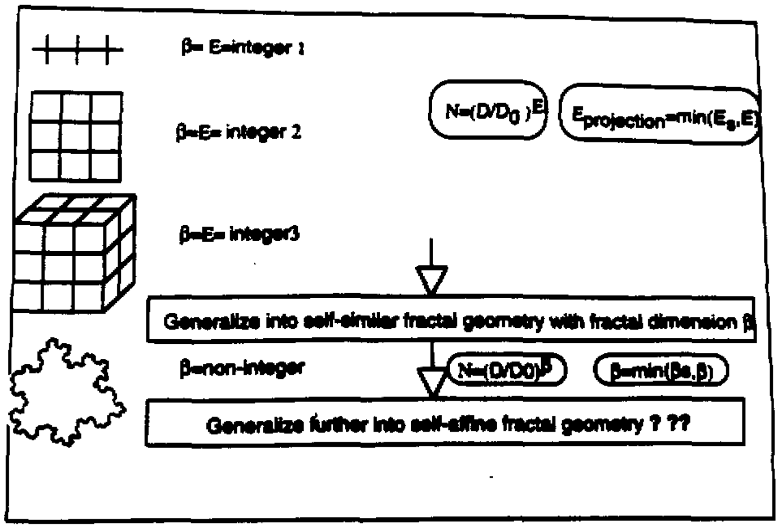

Fig. 2. This figure shows, from dimensional points of view, the possibility of unifying all atmospheric particles into self–similar fractals, with Euclidean particles as special cases. E represents an Euclidean dimension; β represents a general self–similar dimension. The subscript "projection" refers to the projection of a particle. The subscript "s" refers to the substrate on which a particle is projected.

The concept of self–similarity is Closely connected with our intuitive notion of dimension. A one–dimensional object, for example, a line segment, possesses such a scaling property. It can be divided into N identical parts each of which is scaled down by the ratio $D_0 = \frac{D}{N}$ (D is length) from the whole. A two–dimensional objects, such as a square, can be divided into N self–similar parts each of which is scaled down by a factor $D_0 = \frac{D}{\sqrt{N}}$. A three–dimensional object, like a solid cube, can be divided into N little cubes each of which is scaled down by a ratio $D_0 = \frac{D}{\sqrt[3]{N}}$. With self–similarity in mind, the generalization of Euclidean dimension to fractal dimension is straightforward. A β–dimensional self–similar object can be divided into N smaller copies of itself each of which is scaled down by a factor

$$D_0 = \frac{1}{\sqrt[\beta]{N}} \ , \tag{1a}$$

or

$$N = \left(\frac{D}{D_0}\right)^{\beta} \ . \tag{1b}$$

Further, given a self–similar object of N parts scaled down by a ratio r from the whole, its fractal dimension is given by

$$\beta = \frac{\log(N)}{\log\left(\frac{D}{D_0}\right)} \ . \tag{1c}$$

Eq.(1c) provides a practical method for determining the dimension of a fractal, and such defined fractal dimension is specifically called similarity dimension that equals to the common Eulidean dimension for the Euclidean objects (Mandelbrot, 1983). Note that "self–similar" is used to describe both the common self–similarity related to irregularity and the Euclidean that is called standard self–similarity by Mandelbrot (1983).

2. *Relationships among Projections*

A relationship between mass– and area–fractal dimensions will be very useful. Most atmospheric particles are embodied in three–Euclidean space. But most shape analyses are done for projected particle images. For Euclidean particles, it is obvious that dimensions of projections depend on both projected particles and projecting substrate. Plane–projections (projecting substrate is a plane) of a three–dimensional particle and a two–dimensional particle have dimension two; whereas a plane–projection of a one–dimensional particle still has dimension one. On the other hand, when a projecting substrate is one–dimensional curve, a three–dimensional, two–dimensional and one–dimensional particles have dimension one. It should be noted that in one unusual case, when a line is vertically projected into a plane or curve, the projection becomes a point and hence has dimension 0. This case is not treated. These can be summarized into Eq.(2).

$$E_{\text{projection}} = \min(E_s, E) \ , \tag{2a}$$

where $E_{\text{projection}}$ represents the dimension of its projection, E_s represents the dimension of a substrate, and E represents Euclidean dimension of original particles. This formula can be generalized for self–similar fractal particles as

$$\beta_{\text{projection}} = \min(\beta_s, \beta) \ , \tag{2b}$$

which indicates that the dimension of a particle projection will be the minimum of the

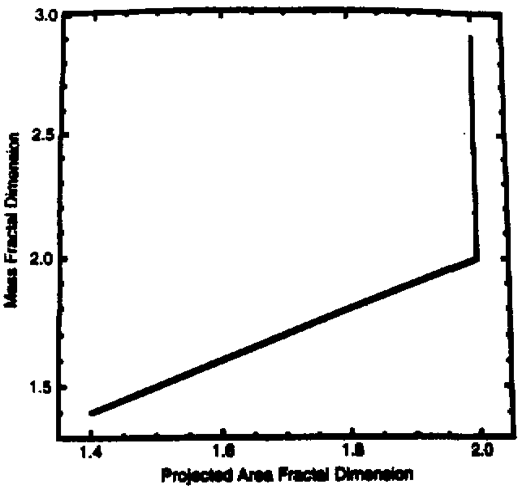

Fig. 3. Relationship between fractal dimension and dimension of the two–dimensional projection of a self–similar particle.

original particle and the substrate. For example, the plane–projection of a particle with dimension $\beta \geqslant 2$ has the same dimension with the substrate; a plane–projection of a particle with $\beta < 2$ has the same dimension with the original particle. This general result will be useful for inferring the original dimension of a particle from its projected dimension because most particles are analyzed based on their plane–projections. For the shape characterization of atmospheric particles, β_s is 2 since most of substrates are two–dimensional planes. Therefore, Eq.(2b) can be simplified as

$$\beta_{\text{projection}} = \min(2,\beta) \ , \tag{2c}$$

which is graphically shown in Fig. 3.

III. GENERALIZED POWER–LAW RELATIONSHIPS

A power–law relationship, $y = \alpha x^{\beta}$, has been empirically used to characterize an atmospheric particle; readers are referred to Liu (1994) and Liu, Mitchell and Arnott (1994) for a detailed review about empirical power–laws. The following outline the major theoretical derivations.

1. *Theoretical Basis for Mass–, Area–, and Perimeter–Dimensional Relationships*

Euclidean particles have power–law relationships between any pairs of mass (m), area (A), perimeter (P) and a characteristic length. Taking a soild sphere as an example, the following power–law relationships exist:

$$m \propto D^3 \ , \tag{3a}$$

$$A \propto D^2 \ , \tag{3b}$$

$$P \propto D \ , \tag{3c}$$

where D is the diameter of the sphere (note: D denotes the maximum diameter for nonspherical particles in the following).

Similar conclusions can be generalized into irregular particles in terms of self–similarity. For a self–similar particle with fractal dimension β_m,
we have

$$N=\left(\frac{D}{D_0}\right)^{\beta_m}=\frac{m(D)}{m(D_0)} \quad . \tag{4a}$$

Or

$$m(D)=\alpha_m D^{\beta_m} \quad , \tag{4b}$$

wher

$$\alpha_m=\frac{m(D_0)}{D_0^{\beta_m}} \quad . \tag{4c}$$

Similarly, for area we have,

$$A(D)=\alpha_a D^{\beta_a} \quad , \tag{4d}$$

$$\alpha_a=\frac{A(D_0)}{D_0^{\beta_a}} \quad , \tag{4e}$$

for perimeter

$$P(D)=\alpha_p D^{\beta_p} \quad , \tag{4f}$$

$$\alpha_p=\frac{P(D_0)}{D_0^{\beta_p}} \quad . \tag{4g}$$

The primary parts, $m(D_0)$, $A(D_0)$ and $P(D_0)$, are related to fractal generators (Mandelbrot 1983). For example, for chainlike aerosol particles the generator is selected as spherules whereas for ice crystals hexagonal shapes will be better.

2. Theoretical Basis for Other Power–Law Relationships

Terminal velocity V_t, defined by Eq.(5a) and Eq.(5b), is another important quantity in studying atmospheric particles.

$$mg=F_d \quad , \tag{5a}$$

$$F_d=\frac{1}{2}\rho_a C_d A v_t^2 \quad , \tag{5b}$$

where g is gravity; F_d is the drag force on the particle; ρ_a the air density; C_d drag coefficient. The following are several arguments which lead to power–law relationships among the terminal velocity and the characteristic diameters.

(1). *Re–WDB number approach*

Eq.(6) has been demonstrated for a particle moving in the atmosphere under gravity (Mitchell, 1994; Liu, 1994).

$$R_e=aW^b \quad , \tag{6}$$

where $R_e=\frac{Dv_t}{\mu}$ is Reynolds number; $W=C_d R_e^2=\frac{2g\rho_a}{\eta^2}\frac{m}{A}D^2$ is called WDB number (See Liu, 1994) for the reason called WDB number instead of Davis number or Best number); μ is kinematic viscosity of air; $\eta=\rho_a\mu$ is dynamic viscosity; a and b are constants depending on the range of flow regimes (Table 1). A combination of Eqs.(4b), (4d), (6) and $R_e=\frac{Dv_t}{\mu}$, yields the power–law relationship

Table 1. Values of a and a $R_e = aW^b$ over Different Regimes

a	b	W
6.1967×10^{-5}	0.52	$W \leqslant 10^{-6}$
0.01227	0.84	$10^{-6} < W \leqslant 0.01$
0.04	1	$0.01 < W \leqslant 10.0$
0.0604	0.83	$10.0 < W \leqslant 500$
0.2072	0.64	$500 < W \leqslant 10^5$
1.5052	0.50	$W > 10^5$

Note: Readers are urged to consult Liu (1994) on which this Table is based.

Table 2. Summary of Formulas Showing Generality

General formulas	Stokes' regime	Spherical particle
$R_e = aX^b$	$a = 0.04 \quad b = 1$	
$m = \alpha_m D^{\beta_m}$	$m = \alpha_m D^{\beta_m}$	$\alpha_m = \frac{\pi}{6}\rho_p \quad \beta_m = 3$
$A = \alpha_a D^{\beta_a}$	$A = \alpha_a D^{\beta_a}$	$\alpha_a = \frac{\pi}{6} \quad \beta_a = 2$
$v_t = \alpha_v D^{\beta_v}$ $\alpha_v = a\mu\left(\frac{2g\alpha_m}{\rho_a \mu^2 \alpha_a}\right)^b$ $\beta_v = b(\beta_m + 2 - \beta_a) - 1$	* $\alpha_v = 0.04\mu\left(\frac{2g\alpha_m}{\rho_a \mu^2 \alpha_a}\right)$ $\beta_v = (\beta_m + 2 - \beta_a) - 1$	* * $\alpha_v = \frac{52.27}{\rho_a \mu}$ $\beta_v = 2$
$\chi = \rho_p \left(\frac{D_{ve}}{D_{st}}\right)^{\frac{3b-1}{b}}$	$\chi = \rho_p \left(\frac{D_{ve}}{D_{st}}\right)^2$	$\chi = 1$
$\chi = \left(\frac{D_{ve}}{D_{st}}\right)^{\frac{3b-1}{b}}$	$\chi = \left(\frac{D_{ve}}{D_{st}}\right)^2$	$\chi = 1$
$\chi = \left(\frac{D_{ve}}{D_{me}}\right)^{3b-4}$	$\chi = \left(\frac{D_{me}}{D_{ve}}\right)$	$\chi = 1$
$D_{ae} = \rho_p^{\frac{b}{3b-1}} D_{st}$	$D_{ae} = \sqrt{\rho_p}\, D_{st}$	$D_{ae} = \sqrt{\rho_p}\, D_{st}$

* The commonly–used formula $v_t = \frac{g\rho_p}{72\rho_a \mu \chi} D_{ve}^2$ is a special case when substituting $\chi = \rho_p \left(\frac{2\alpha_a}{3\alpha_m}\right) \left(\frac{6\alpha_m}{\pi\rho_p}\right)^{2/3} D^{\frac{3\beta_a + 3 - \beta_m - 1}{3}}$ and $D_{ve} = \left(\frac{6\alpha_m}{\pi\rho_p}\right)^{\frac{1}{3}} D^{\frac{\beta_m}{3}}$.

* * Another existing terminal velocity formula $v_t \propto D^{0.5}$ in Newton's regime is also a special case of $b = 0.5$.

$$v_t = \alpha_v D^{\beta_v} \ , \tag{7a}$$

where

$$\alpha_v = a\mu \left(\frac{2g\alpha_m}{\rho_a \mu^2 \alpha_a} \right)^b \ , \tag{7b}$$

$$\beta_v = b(\beta_m + 2 - \beta_a) - 1 = b(\Delta + 2) - 1 \ . \tag{7c}$$

By expressing terminal velocity in different ways, general power–law relationships have been derived among characteristic diameters (e. g., volume equivalent diameter, dynamical equivalent diameter, mobility equivalent diameter) and the dynamic shape factor. A summarized table (Table 2) is given here. Further simplification of these formulas can be readily made for self–similar particles by a combination with the relationships among projected dimensions (Section 2.2).

(2). *Other equivalent approaches*

In addition to the Re–WDB number approach, there exist another two equivalent approaches: using the following power–law relationships:

$$R_e = a_0 C_d^{b_0} \ , \tag{8}$$

$$R_e = a_1 W_1^{b_1} \ , \tag{9a}$$

$$W_1 = C_d R_e^{-1} \ . \tag{9b}$$

Table 3. Relationships between Coefficients in $R_e = aW^b = a_0 C_d^{b_0} = a_1 W_1^{b_1}$

coefficient	a_0	a	a_1	b_0	b	b_1
a_0	$a_0 = a_0$	$a_0 = a^{\frac{1}{1-2b}}$	$a_0 = a_1^{\frac{1-3b}{1-2b}}$			
a	$a = a_0^{1-2b}$	$a = a$	$a = a_1^{1-3b}$			
a_1	$a_1 = a_0^{\frac{1-2b}{1-3b}}$	$a_1 = a^{\frac{1}{1-3b}}$	$a_1 = a_0^{\frac{1-2b}{1-3b}}$			
b_0				$b_0 = b_0$	$b_0 = \frac{b}{1-2b}$	$b_0 = \frac{b_1}{1+b_1}$
b				$b = \frac{b_0}{1+2b_0}$	$b = b$	$b = \frac{b_1}{1+3b_1}$
b_1				$b_1 = \frac{b_0}{1-b_0}$	$b_1 = \frac{b}{1-3b}$	$b_1 = b_1$

The equivalence of the three approaches can be readily seen from the relationships among the coefficients (Table 3), each having its advantages: W is not an explicit function of terminal velocity; W_1 is not a function of diameter for a spherical particle ($W_1 = \frac{32\rho_p g\mu}{3\rho_a} V_t^{-\frac{1}{3}}$, ρ_p is particle density); a relatively larger number of Cd–Re data is available. The physics underlining power–law relationships (6), (8) and (9) is explored in

Liu (1995) by establishing relationships between $b(b_0,b_1)$ and fractal dimensions of flows around a particle.

At this point, theoretical foundations for generalized power–law relationships have been established for a single self–similar particle, which include classical Euclidean particles as special cases. Next question is how to relate this single particle property to atmospheric particle systems in which particles interact with one another.

IV. UNIFIED PARTICLE NUMBER SIZE DISTRIBUTIONS

The Preceding sections concern a single particle. Then how about atmospheric particle systems? Looking carefully at number size distributions of atmospheric particles, one may ask the following questions. (1) Since atmospheric particles are produced by different mechanisms such as gas–to–particle and bulk–to–particle conversions, why do their number size distributions exhibit similar shapes? (2) While atmospheric particles cover a substantial size range, approximately 10 orders of magnitude, why do size distributions over limited ranges show similarity? Regarding the similarities, readers are referred to Liu et al. (1995), in which these questions were answered by introducing the Shannon's maximum entropy principle (SMEP), extending statistical mechanics and using generalized power–law relationships. Its outline is as follows.

1. *Shannon's Maximum Entropy Principle (SMEP)*

Information theory, especially SMEP, has increasingly found applications. Full details of this theory can be found in Guisu (1977) or other textbooks related to information theory. As an illustration, the continuous case is briefly introduced here.

Consider a stochastic system which is governed by some restriction conditions and characterized by a continuous stochastic variable y with the probability density function (PDF) $\rho(y)$. $\rho(y)$ has many possible realizations owing to fluctuations, and each $\rho(y)$ corresponds to one Shannon entropy $H(y)$. SMEP states that under the condition that the Shannon entropy $H(y)$ achieves its maximum value $H^*(y)$, its corresponding PDF, $\rho^*(y)$ will have the largest probability to occur. This means that maximum entropy $H^*(y)$ corresponds to the maximum likelihood PDF $\rho^*(y)$. Mathematically, we have,

$$\int \rho(y)dy = 1 \ , \tag{10a}$$

$$\int f_k(y)\rho(y)dy = F_k \ , \qquad (k = 1,2,\cdots,m) \tag{10b}$$

in which F_k is independent of y, m the total number of restrictions, Eq.(10a) means that $\rho(y)$ represents PDF; Eq.(10b) means that the quantities $f_k(y)$ $(k = 1,2,\cdots,m)$ obeys the dynamical equilibrium, suggesting that $f_k(y)$ has some fluctuations. The Shannon entropy is defined as

$$H(y) = -c\int \rho(y)\ln\rho(y)dy \ , \tag{10c}$$

where c is a constant related to the unit used.

In has been shown that under the conditions above, the maximum likelihood PDF has the form:

$$\rho^*(y)=\frac{1}{Z(q_1,q_2,\cdots,q_m)}\exp\left(-\sum_{i=1}^{m}q_if_i(y)\right), \tag{10d}$$

where $Z(q_1,q_2,q_3,\cdots,q_m)$ is defined as the generalized partition function, and can be obtained by:

$$Z(q_1,q_2,\cdots,q_m)=\int\exp\left(-\sum_{i=1}^{m}q_if_i(y)\right)dy\ , \tag{10e}$$

where $q_i(i=1,2,3,\cdots,m)$ are constants which can be calculated by means of Lagrange multipliers.

Table 4. Conservation Laws, Power–Laws and Weibull Distributions*

Conservation Law	Restriction Variable	Power–Law	Weibull Distribution
general, X	x	$x=\alpha_x D^{\beta_x}$	$n(D)=\alpha_x\beta_x XD^{\beta_x-1}\exp\left(-\frac{\alpha_x X}{N}D^{\beta_x}\right)$
mass, M	m	$m=\alpha_m D^{\beta_m}$	$n(D)=\alpha_m\beta_m MD^{\beta_m-1}\exp\left(-\frac{\alpha_m M}{N}D^{\beta_m}\right)$
momentum, T	$t=mv_t$	$t=\alpha_t D^{\beta_t}$ $\alpha_t=\alpha_m\alpha_v$ $\beta_t=\beta_m+\beta_v$	$n(D)=\alpha_t\beta_t TD^{\beta_t-1}\exp\left(-\frac{\alpha_t T}{N}D^{\beta_t}\right)$
kinetic energy, K	$k=\frac{1}{2}mv_t^2$	$k=\alpha_k D^{\beta_k}$ $\alpha_k=\frac{1}{2}\alpha_m\alpha_v^2$ $\beta_k=\beta_m+\beta_v^2$	$n(D)=\alpha_k\beta_k KD^{\beta_k-1}\exp\left(-\frac{\alpha_k K}{N}D^{\beta_k}\right)$
surface energy or surface area, A	a	$A=\alpha_A D^{\beta_A}$	$n(D)=\alpha_a\beta_a AD^{\beta_a-1}\exp\left(-\frac{\alpha_a A}{N}D^{\beta_a}\right)$

* These are only general results. Which conservation law used depends upon the specific problem in question.

SMEP relates the maximum likelihood PDF with the restriction conditions which are natural laws governimg the system in reality.

2. *New Concepts*

CONCEPT 1. Restriction Variable x. In reality, all the atmospheric particle systems are controlled by some natural laws (e. g., energy conservation, momentum conservation, mass conservation) and their combinations. Because these physical laws generally correspond to some restriction conditions in mathematical equations, the physical quantities that correspond to the conservation laws are called restriction variables and denoted by x here. In other words, the restriction variable x is analogous to the stochastic variable y in information theory.

CONCEPT 2. Atmospheric Particle Ensemble (APE). Similar to molecular ensemble in statistical mechanics, APE is defined to be composed of an arbitrarily large number of the systems satisfying the same macroscopic constraints but differing in their microscopic states (number distributions).

CONCEPT 3. Particle Number Spectrum Entropy Ep(x). Similar to Shannon entropy, if the particle number distribution with x is denoted by $n(x)$ which represents the particle number per unit volume per unit x interval and the total number per unit volume denoted by N, then probability density function is defined as:

$$\rho(x)=\frac{n(x)}{N} \quad . \tag{11}$$

The particle number spectrum entropy is defined as

$$E_p(x) = -\int \rho(x)\ln\rho(x)dx \quad , \tag{12}$$

in which the unit related constant c is omitted because it does not affect the result.

3. *Assumptions*

ASSUMPTION 1. Due to fluctuations and uncertainties, atmospheric particle systems may have different particle size distributions under the same initial conditions and governing laws. But there exists one distribution which occurs with the largest probability.

ASSUMPTION 2. It is postulated that SMEP is also suitable for the particle number spectrum entropy. This means that the maximum particle number spectrum entropy, $H_p^*(x)$, corresponds to the maximum likelihood PDF $\rho^*(x)$—hence the maximum liklihood distribution $n^*(x)$ under certain restrictions.

ASSUMPTION 3. As a first step, we consider the simplest case where only one physical law controls the studied atmospheric particle system. This means that only one restriction variable x is involved.

4. *Theory*

On the basis of above descriptions, a system theory on atmospheric particle systems can be expressed as follows.

The restrictions which govern the system are:

$$\int \rho(x)dx = 1 \quad , \tag{13a}$$

$$\int n(x)dx = N \quad , \tag{13b}$$

$$\int x\rho(x)dx = X' \quad . \tag{13c}$$

Or

$$\int xn(x)dx = NX' = X \quad , \tag{13d}$$

where the restriction variable x may be energy, mass, momentum, etc., depending on the natural laws governing the systems.

Combining Eqs.(13a) and (13c) with Eqs.(13d) and (13e) yields the following entities (Appendix A):

$$\rho^*(x)=\varepsilon_x\exp(-\varepsilon_x x)\ , \tag{14a}$$

$$\varepsilon_x=\frac{X}{N}\ , \tag{14b}$$

where ε_x represents the average X per particle, analogous to $\frac{1}{KT}$ (KT is energy per molecule; K Boltzmann constant; T temperature in Kelvin degree) in the Maxwell–Boltzmann distribution for ideal gases $\rho^*(e)=\frac{1}{KT}\exp\left(-\frac{e}{KT}\right)$. Or the maximum likelihood distribution (MLD) with x is

$$n^*(x)=N\varepsilon_x\exp(-\varepsilon_x x)\ . \tag{14c}$$

Furthermore, for atmospheric particle systems, the particle number size distribution is customarily used instead of the distribution with x. Generally speaking, the restriction variable x is a function of particle diameter D (for non–spherical particles, D means some equivalent diameter), and is denoted by $F(D)$:

$$x=F(D)\ . \tag{15}$$

Hence the maximum likelihood size distribution is

$$n^*(D)=N\varepsilon_x\frac{dF}{dD}\exp(-\varepsilon_x x)\ . \tag{16}$$

Atmospheric particle shapes can be unified in a self–similar fractal with various power–law relationships (Appendix B):

$$x=F(D)=\alpha_x D^{\beta_x}\ . \tag{17}$$

Substitution of Eq.(12) into Eq.(11) yields,

$$n^*(D)=N_0 D^{q-1}\exp(-\lambda D^q)\ , \tag{18a}$$

$$N_0=\varepsilon_x\alpha_x\beta_x N=\alpha_x\beta_x X\ , \tag{18b}$$

$$\lambda=\varepsilon_x\alpha_x\ , \tag{18c}$$

$$q=\beta_x\ . \tag{18d}$$

V. A GENERAL FRAMEWORK

Three unified aspects have been addressed. First of all, atmospheric particle shapes are unified as self–similar fractals, with Euclidean particles as special cases. Secondly, such self–similar particles have generalized power–law relationships, say $m(D)=\alpha_m D^{\beta_m}$. Finally, a system theory is established by introducing SMEP into atmospheric particle systems. This theory predicts an exponential form of particle number distribution with x. By using a power–law relationship, $x=\alpha_x D^{\beta_x}$, a Weibull distribution, $n^*(D)=N_0 D^{q-1}\exp(-\lambda D^q)$, is derived for any atmospheric particle systems controlled by a single conservation law. This can be generalized into a particle system which is governed by several physical laws. In this case, a multi–Weibull distribution will be derived. Although the first two aspects are for

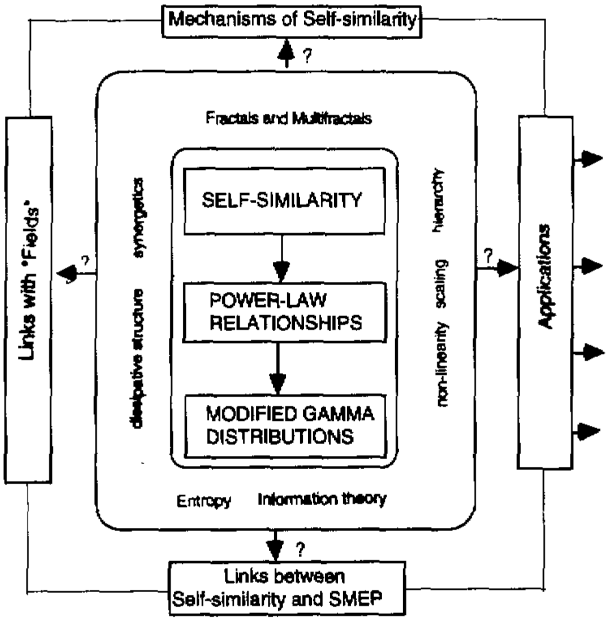

Fig. 4. Schematic show of general unification framework. The first rounded box represents what have been discussed; disciplines and / or concepts of potential usefulness are listed between the first and second rounded box; the four boxes represent new questions for future research.

a single particle whereas the last one is for a particle system, they can be synergistically combined to give a general unification framework (Fig.4). As shown in Fig. 4, within this generalized framework, basic assumptions are reduced to (1) a single particle is a self–similar fractal and (2) an atmospheric particle system obeys SMEP. Futher, involved are only two basic equation forms: power–law and Weibull. This will reduce equation numbers and hence computer time in modeling. Another advantage is its clear connection between macroscopic properties such as controlling laws and microscopic properties such as size distribution.

A further question is as to why and how self–similarity of a single particle and SMEP for a particle system are related to each other. In other words, is there a higher unifying principle to produce both self–similar geometry and SMEP? As pointed out by Schroeder (1991), self–similarity is a unifying concept underlining fractals, chaos, and power–law relationships. It is speculated thast the final resolution could be obtained by intuition and combining the disciplines and / or concepts listed between the first and second rounded box in Fig.4, each of which is a frontier in science. It is worth noting that it is just the scaling properties that make the unification possible, which is analogous to the well–known principle of dynamical similarity in fluid mechanics (Kundu, 1990). In the following, a qualitative arguments about self–similar structure will be made. But, what leads to the coupling between a single self–similar particle and SMEP for a atmospheric particle system remains open to explore.

VI. IMPLICATIONS

This generalized framework shows a number of encouraging points; results about size

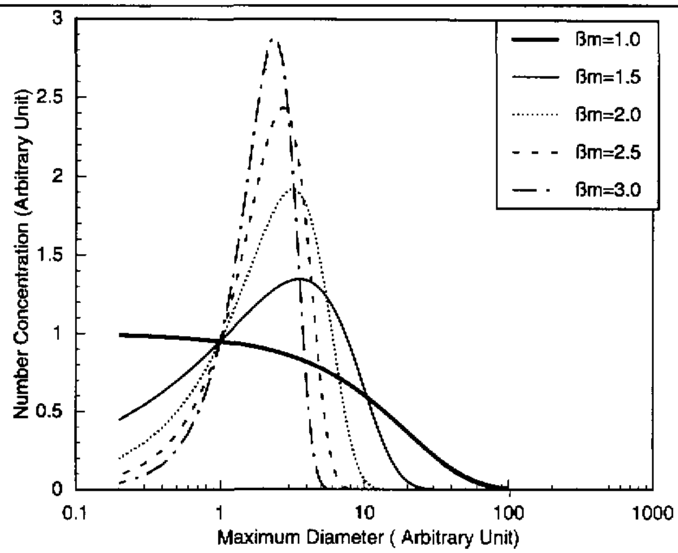

Fig. 5. Shape effects on size distributions. In this case ($n(D) = D^{\beta_m}$ $\exp(-0.5D^{\beta_x})$) it is shown qualitatively that size distributions will broaden with decrease in the mass fractal dimension β_m. This suggests a general result that size distributions widen with fractal dimensions of particles. For hydrometeors, this indicates a broadening trend from spherical droplets to thin plates, to dendrites, to chainlike crystals, to needles.

distributions are discussed. Table 3 gives a heuristic view of particle systems controlled by different physical laws. These equations reveal a close relationship between size distributions and the four factors: controlling physical law (via $x = F(D) = \alpha_x D^{\beta_x}$), particle shape (via α_x and β_x), total number concentration N and total X. This becomes more apparent when Eq.(18) is rewritten as

$$n(D) = \alpha_x \beta_x X D^{\beta_x - 1} \exp\left(-\frac{\alpha_x X}{N} D^{\beta_x} \right) . \tag{19}$$

1. *Shape Effects and Law Effects*

As an illustration, shape effect is further discussed as follows. For the same controlling law, N and X, size distributions vary with particla shapes. Fig. 5 is an idealized example of shape effect by assuming that the controlling law is mass conservation and that mass–dimensional relationships are employed. Regardless of its qualitativeness, it is evident that size distributions widen with decrease in β_m from 3 (such as spherical droplets) to 2 (such as thin plate snow crystals) to 1 (such as needle). This suggest that we must be careful of analyzing size distribution data, especially when comparing data from different instruments.

Duriny the processes, laws controlling a particle ensemble may change from one to another and this change will lead to change in size distribution. The law effects are similar to shape effects since both changes are embodied in power–law $x = \alpha_x D^{\beta_x}$.

2. *N Effects and X Effects*

By keeping other factors constant either N effects or X effects can be studied. N obviously determines "slope" of a distribution and slopes will decrease with increase in

N. X is related to both "slope" (with opposite effect to N) and "intercept".

Generalized framework also provides explanations for (1) empirical power–laws widely used for both aerosols and hydrometeors and (2) distribution similarity among atmospheric particle systems.

VII. QUALITATIVE JUSTIFICATIONS ABOUT SELF–SIMILARITY

Although, to the best knowledge of the author, no literature about theoretical aspects of hydrometeor's self–similarity has been published, promising research has been done in two related areas: one is about " fields" in which particles are located and the other is various aggregation models producing self–similar colloidal particles. Incorporation of results from these relevant areas and a coupled aggregation model with self–similar "fields" may provide a potential tool for exploring theoretically the self–similar structure of atmospheric particles.

1. *Self–similarity of "Fields"*

Atmospheric particles are engulfed in a hierarchy of scaling " elements". In terms of scales, molecular Brownian motion and turbulent motions consisting of eddies with different sizes are two "fields" directly related to atmospheric particles. Both phenomena are, in fact, typical examples of fractals or self–similarities and associated with randomness of different degrees. Further to larger scales in which Brownian molecules and turbulent eddies embedded, there has been considerable evidence that various atmospheric fields (e. g., rain, wind, clouds, temperature, and radiation fields) are fractals (Lovejoy, *1981*; Lovejoy and Mandelbrot, 1985; Lovejoy and Schertzer, 1985, 1986, 1990; Pflug et al., 1993; Schertzer and Lovejoy, 1987, 1988, 1989; Tessier et al., 1993. Hereafter, these will be denoted by McGill group since most of the work were done in McGill University, Canada). Schertzer and Lovejoy (1988) argue as follows. In geophysical fluid dynamics, the existence of scaling regimes can often be argued directly from the dynamical equations themselves: the only scales associated with the Navier–Stokes equations are a largest scale of energy injection and a small viscous scale where most of the dissipation occurs. In the atmosphere these scales (along the horizontal) are roughly of the order of thousands of kilometers and several mm, respectively, allowing the possibility of a scaling regime over nine orders of magnitude in scale. It is worth noting that scaling in atmospheric dynamics has been extended from self–similar scaling into a generalized scale invariance by MiGill group. This second order generalization in atmospheric particle systems is beyond this paper. In summary, in atmospheric processes, scaling features for molecular motions, turbulence and larger scale phenomena have been studied and identified. This means that atmospheric particles are existing in self–similar "fields".

Also, in terms of scale, self–similarities of atmospheric particles fill the "gap" between molecular Brownian motion and turbulence, i.e., the gap between molecules of $\sim 10^{-8}$ cm in size and the smallest turbulent eddy of $\sim 10^{-1}$ cm. In this sense, the atmospheric particle system is a level of the whole hierarchy (Fig. 6). This new "element" itself is composed of sub–levels (aerosol, cloud, etc.) and they must perform some work within the whole hierarchy. Also, elements of other levels especially the two nearest levels (molecules and turbulence eddies), must have influences on atmospheric particles. However, most theories on particle diffusional growth emphasize molecular effects, e. g., based on classical transport laws (Fick's first law for mass diffusion, fourier law for heat diffusion) whereas the effects of turbulence are "underestimated". The hierarchical structure dictates the importance of different turbulent eddies. This further suggests that the (non–linear) coupling bewteen statistical

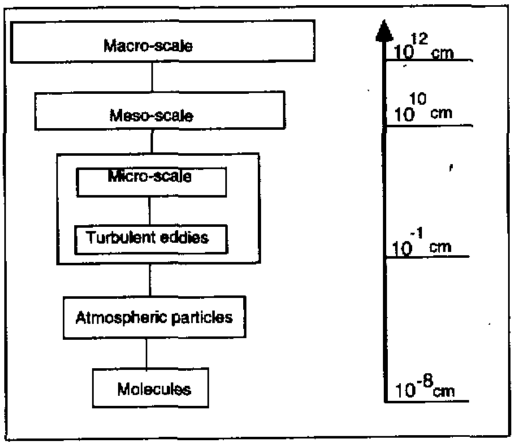

Fig. 6. The hierarchy of atmospheric "elements". It should be noted that each level consists of sub–levels. Scaling structures of other levels indicate self–similar structure of an atmospheric particle which is within an "in–between" level.

mechanics, particle microphysics and atmospheric dynamics be studied from scaling point of view. Readers are urged to consult Pattee (1973) and Wu (1991) for hierarchy theory.

2. *Self–similar Structure Predicted by Aggregation Models*

The other related area is aggregation of general colloidal particles. Research of fractal structure of particles was initiated by Forrest and Witten (1979), who studied smoke particle aggregates. Witten and Sander (1981) developed the diffusion–limited aggregation (DLA) model. Since then much effort has been made to develop various aggregation models producing self–similar structure. Readers are urged to consult Meakin (1989) for a detailed review. Potential physical mechanism underlying atmospheric particles of various shapes are related to various aggregation processes. Sander (1985) classified processes of growth by aggregation into three regimes. If the growing object is always near equilibrium despite the addition of new material then its internal structure and external shape will be describable by ordinary equilibrium considerations such as single crystals with the well–known equilibrium crystal shape. As we drive the system away from equilibrium we oftern find a new regime of morphology. In this case, new length scales associated with the steady–state growth give rise to intricate patterns which can persist even for large objects. Examples of these are the beautiful, feathery, dendrite shapes of snowflake growth. Even farther from equilibrium a whole new regime can appear—that of disorderly growth with no scales at all. The remarkable fact about this regime is that in some cases it is not merely amorphous growth but growth of scale–invariant fractals. Physically, atmospheric particles, which cover ordered fractal (e. g., various symmetrical crystals) and disordered fractals, e. g., "irregular" aerosol particles and hydrometeors, experience similar processes and hence theories and techniques can be introduced into investigation of atmospheric particles.

The striking geometry similarity (self–similar structure) and distribution similarity (Weibull form), among others, indicates that the functioning of atmospheric particles obeys some unifying principles. The close relationship of both self–similar fractal and SMEP with frontier disciplines further suggests a higher unifying principle.

VIII. CONCLUDING REMARKS

A Generalized framework for atmospheric particles is established based on two unifying principles. Characterization of particle shapes is unified by assuming that atmospheric particles can be generally considered as self–similar fractals. Particle number size distributions are unified by Weibull distributions using power–law relationships, which bridges a self–similar fractal and SMEP for a particle system, assuming that SMEP can be applied to atmospheric particle systems. The two unifying principles, in turn, reduce the number of equations: only power–law and Weibull equations are needed. Pasrsimony in underlining principles leads to economy in controlling equations, which in turn leads to higher efficiency of numerical modeling, and hence to feasibility of investigating complex interactions among different particle systems. Within this framework, four effects on size distributions are identified (shape effect, Law effect, N effect and X effect).

Qualitative arguments about the mechanisms of self–similarity suggest that introducing and coupling general aggregation models with a hierarchy of scaling "fields" may provide a tool for exploring the self–similar structure of a single atmospheric particle and the higher unifying principle. Identification of this self–similar structure will bridge the scaling gap beween molecular Brownian motion and turbulence and hence extend scaling regimes proposed by McGill group. Mechanisms behind such a hierarchy of self–similar structures and interaction between different levels are future challenges.

The following should be noted before conclusion.

(1). This paper focuses mainly on theoretical development. The unifying "principles" (self–similar geometry and SMEP) still awaitwait for observational evidences. An instrument which can simultaneously measure at least three quantities and a quantitative method to obtain the maximum likelihood distribution from a family of distribution data must be developed before such evidence can be obtained.

(2). New challenges are whether the two unifying "principles" can be further generalized into one higher unifying principle, and how the different levels of the whole hierarchy interact. I speculate that final solution should concern introduction of frontier disciplines and new concepts (e. g., self–similarity, scalling, fractals, chaos, hierachy and non–linearity).

Appendix A: Derivation of Equation (14)

Eq.(14) can be obtained by introducing two Lagrange multipliers q_1 and q_2, and maximizing the following Lagrange functional L:

$$L(\rho(x), q_1, q_2) = -\int_0^{\infty} \rho(x)\ln(x)dx + q_1\int_0^{\infty} \rho(x)dx + q_2\int_0^{\infty} x\rho(x)dx \ . \quad \text{(A1)}$$

Setting the first variation of L with respect to the unknown $\rho(x)$ equal to zero, the following result is obtained:

$$0 = \Delta L = \int_0^{\infty} [-\ln\rho(x) - 1 + q_1 + q_2 x]\Delta\rho dx \ . \quad \text{(A2)}$$

By noting the fact that $\Delta\rho$ is arbitrary, we have

$$-\ln\rho(x) - 1 + q_1 + q_2 x = 0 \ , \quad \text{(A3a)}$$

or

$$\rho(x) = \exp(q_1 - 1)\exp(q_2 x) \ . \tag{A3b}$$

A combination of Eq.(A3b) with Eqs.(13a) and (13c) yields:

$$\exp(q_1 - 1) = \frac{1}{X'} = \varepsilon_x \ ,$$

$$q_2 = -\frac{1}{X'} = -\varepsilon_x \ .$$

Therefore,

$$\rho^*(x) = \varepsilon_x \exp(-\varepsilon_x x) \ .$$

Appendix B: Semi–quantitative Methods for Vindicating Distribution Similarity and Obtaining MLD

Justification of distribution similarity and SMEP for atmospheric particle systems calls for a quantitative method to solicit MLD from a family of distribution data. Because no quantitative method is available, semi–quantitative methods are introduced briefly here.

Table A. Skewness Deviation Coefficient (C_s), Kurtosis Deviation Coefficient (C_k) and $C_s - C_k$ Relationship for Some Probability Density Functions (pdfs)

pdfs	C_s	C_k	$C_s - C_k$ relationship
normal	0	0	$C_s = C_k = 0$
exponential	1	1	$C_s = C_k = 1$
gamma	$\frac{1}{1+\mu}$	$\frac{1}{1+\mu}$	$C_s = C_k$
Weibull	$\frac{[3q^2 p_3 - 6qp_1p_2 + 2p_1^3]^2}{4(2qp_2 - p_1^2)^3}$	$\frac{4q^3 p_4 - 12q^2 p_1 p_3 + 24qp_1^2 p_2 - 12q^2 p_3^2 - 6p_1^4}{6(2qp_2 - p_1^2)^2}$	*
lognormal	$\frac{(\eta^3 + 3\eta)^2}{4}$	$\frac{\eta^8 + 6\eta^6 + 15\eta^4 + 16\eta^2}{6}$	* *

Non–dimensional Method

In a series of papers (Liu, 1992; Liu, 1993; Liu and Liu, 1993; Liu et al. 1995) a semi–quantitative method has been developed based on the skewness coefficient C_s and kurtosis coefficient C_k of number size distributions. As shown in Table A, for commonly used distributions (e. g, normal, exponential, gamma, Weibull, lognormal) there exist known C_s—C_k relationships which can be used as reference for judging the distribution pattern of statistical significance. Each measured distrbution maps into one point in C_s—C_k graph. Distribution similarity has been shown by means of C_s—C_k graph among number size distributions of raindrops (Liu, 1992; 1993), aerosol particles (Liu and Liu, 1994) and cloud droplets (Liu et al., 1995). The maximum likelihood cloud droplet distribution has been also identified (Liu et al, 1995). A Very similar method was proposed independently by Yee et al.

in studying probability distributions of pollutant concentrations in the atmospheric boundary layer (Yee et al., 1993; Yee et al., 1993). They used the plot of fluctuation intensity vs.skewness and kurtosis. It is interesting that distribution similarity was also found for concentration porbability distributions in their research. This agreement, in fact, suggests the possibility of unifying treatment from another point of view. Because both Liu's and Yee's methods use a pair of non–dimensional statistical parameters, they are called non–dimensional methods. It is worth noting that MLD will appear as a attraction point in the figure and can be determined from the coordinate values of this point; readers are urged to consult Liu et al. (1995) for detail.

General Scaling Method

Another candidate for studying distribution similarity study is general scaling method developed by Torres et al. (1994) on the basis of previous work (Sekhon and Srivastava, 1970, 1971; Willis, 1984). They gave a general expression for raindrop size distribution,

$$n(D,\Psi) = \Psi^{\alpha_\Psi} g\left(\frac{D}{\Psi^{\beta_\Psi}}\right) , \tag{B1}$$

where Ψ can be any integral rainfall variable although rainfall intensity R has generally been used. For a given Ψ, α_Ψ and β_Ψ are constants which keep Ψ^{β_Ψ} has dimension of $n(D,\Psi)$ and Ψ^{α_Ψ} has the dimension of D, g is a function that is independent of the value of Ψ and that will be called the general distribution function. Once α_Ψ and β_Ψ have been identified, an experimental function g is obtained by plotting the whole set of measured spectra on graph $y = \frac{n(D,\Psi)}{\Psi^{\alpha_\Psi}}$ vs. $x = \frac{D}{\Psi^{\beta_\Psi}}$. The use of these coordinates has a scaling effect on the spectra, making them comparable independently of both D and Ψ. In fact, $y = g(x)$ is a renormalized non–dimensional raindrop size distribution. It is obvious that this scaling technique is applicable to other particle size distributions. Furthermore, this non–dimensionality makes it useful for comparing distribution data from any atmospheric particle gruops. We must keep in mind that for all the compared distributions the same scaling variable Ψ must be used. However, this technique needs to be modified before used to identify specific distribution because it does not give any information about specific model distribution (e. g., gamma, lognormal). A combination of non–dimensional technique with this technique may provide better results.

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