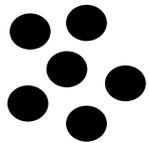


# Physics-Based Parameterization for Cloud Microphysics and Entrainment-Mixing Processes: Addressing Gaps

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**Yangang Liu**  
**(Brookhaven National Laboratory)**

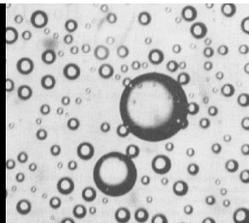
**EMC, NOAA**  
**August 15, 2017**



**Molecule**



**Aerosol**



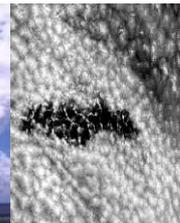
**Droplet**



**Turbulent Eddies**



**clouds**



**Clusters**



**Global**

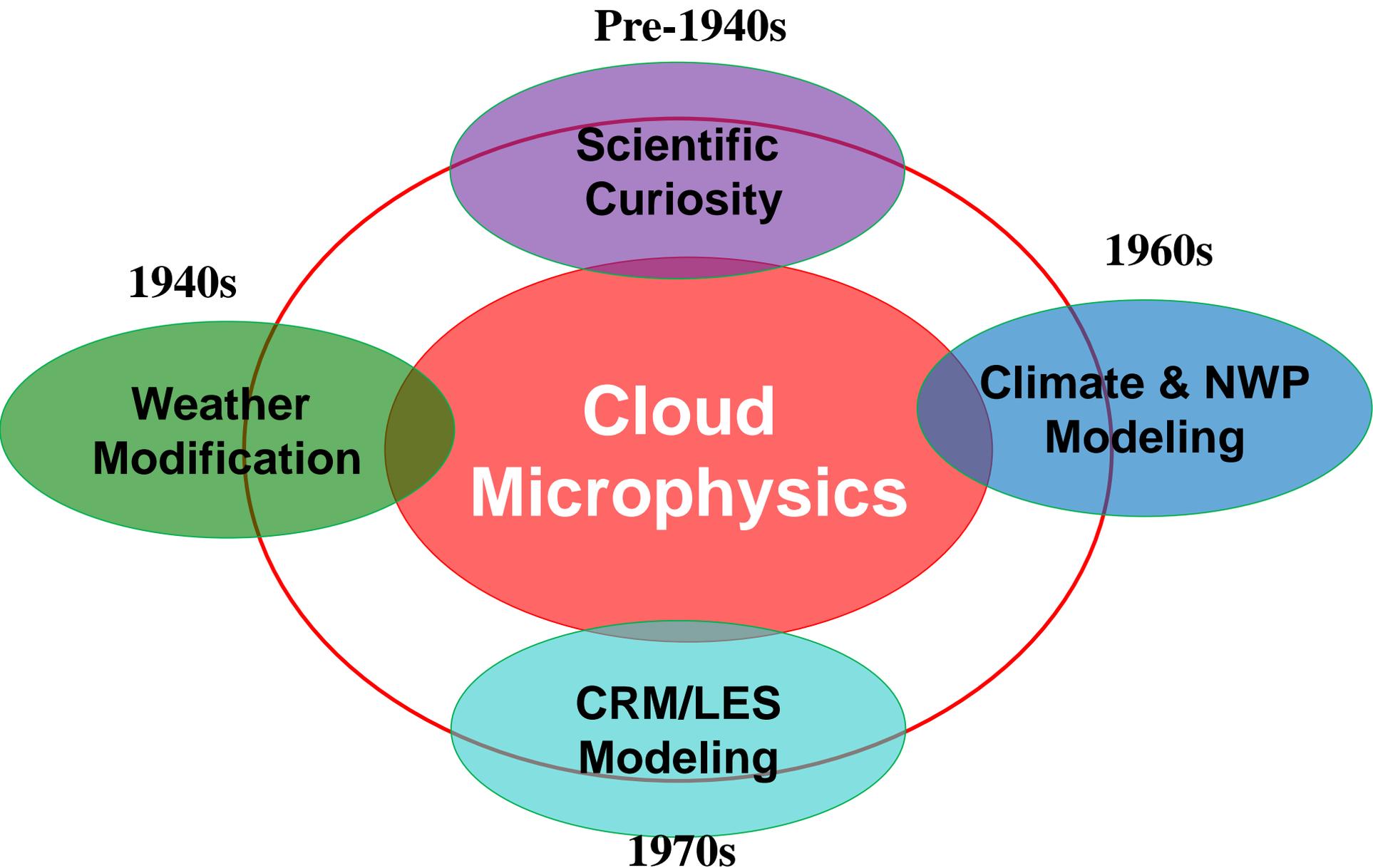
# Outline

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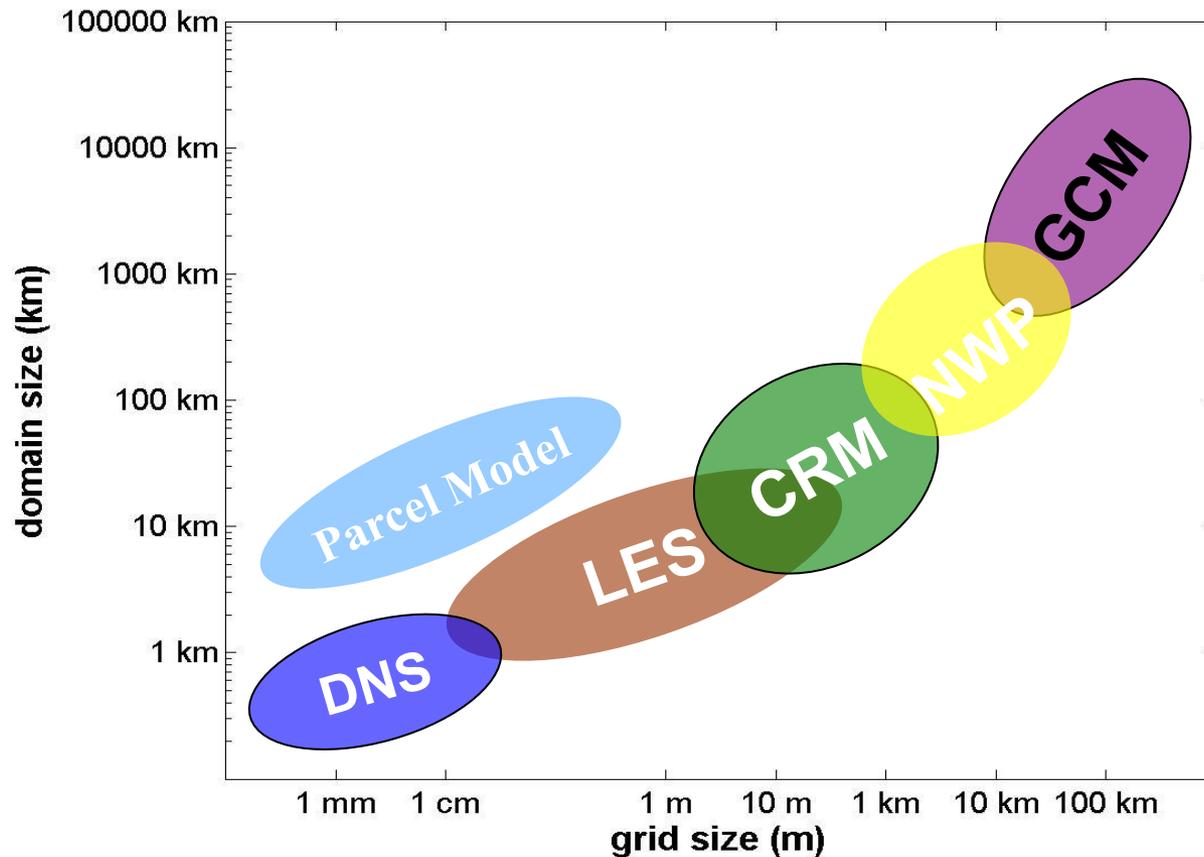
- **Background & Main Gaps**
- **Statistical Physics for Cloud Microphysics  
Parameterization**
- **Turbulent Entrainment-Mixing Process**
- **Particle-Resolved DNS**
- **Take-Home Messages**

# Four Fundamental Sci. Drivers

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# Microphysics parameterization is essential to virtually all major numerical models



**Except for DNS, microphysics is parameterized with different sophistications, e.g., single moment (L), double moment (L, N), three moment (L, N, dispersion), ..., bin microphysics.**

# Further improving $\mu$ -parameterization brings the issue to the heart of cloud physics

Uncertainty and Discrepancy

Microphysics Parameterization

- One moment scheme (LWC only)
- Two moment scheme (LWC & droplet concentration)
- Three moment scheme (LWC, N, & relative dispersion)
- ....

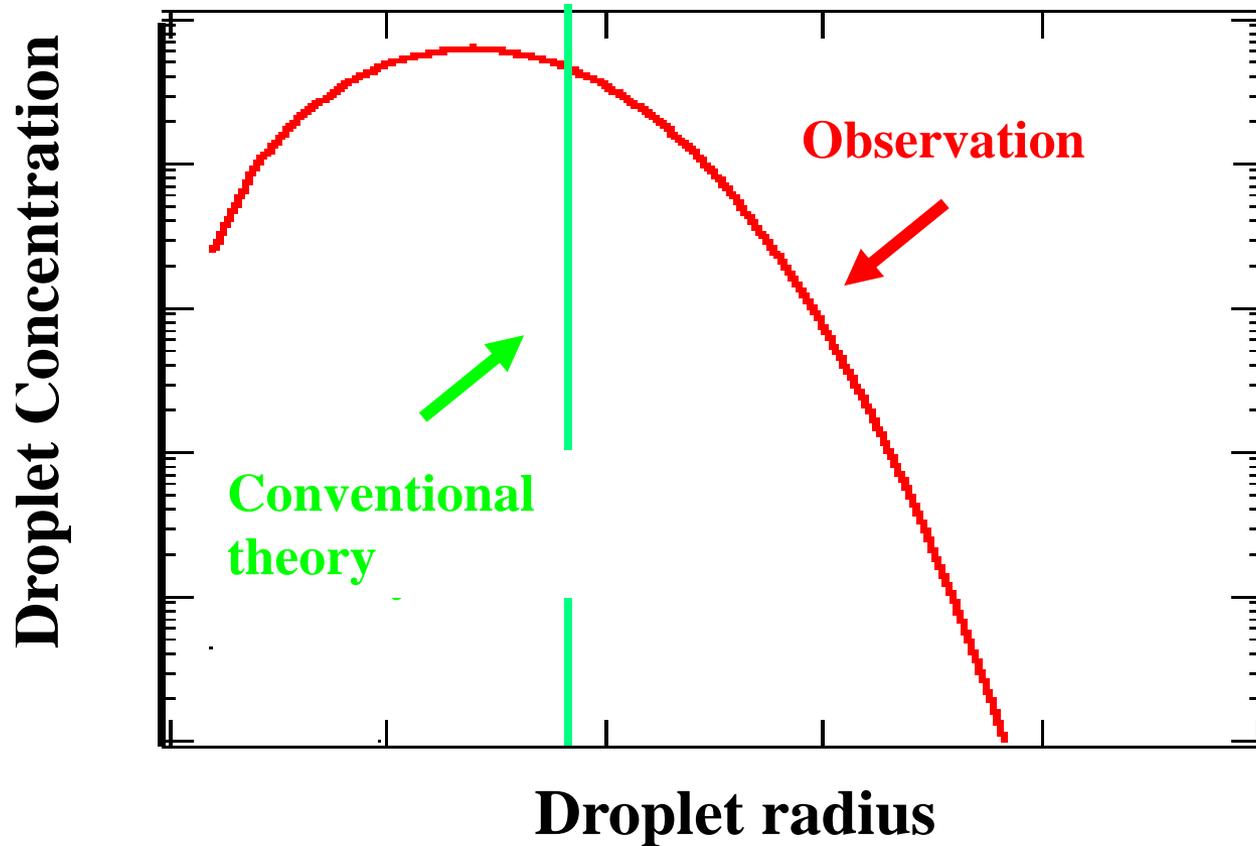
Spectra  
Broadening  
Rain  
Initiation



Cloud Physics

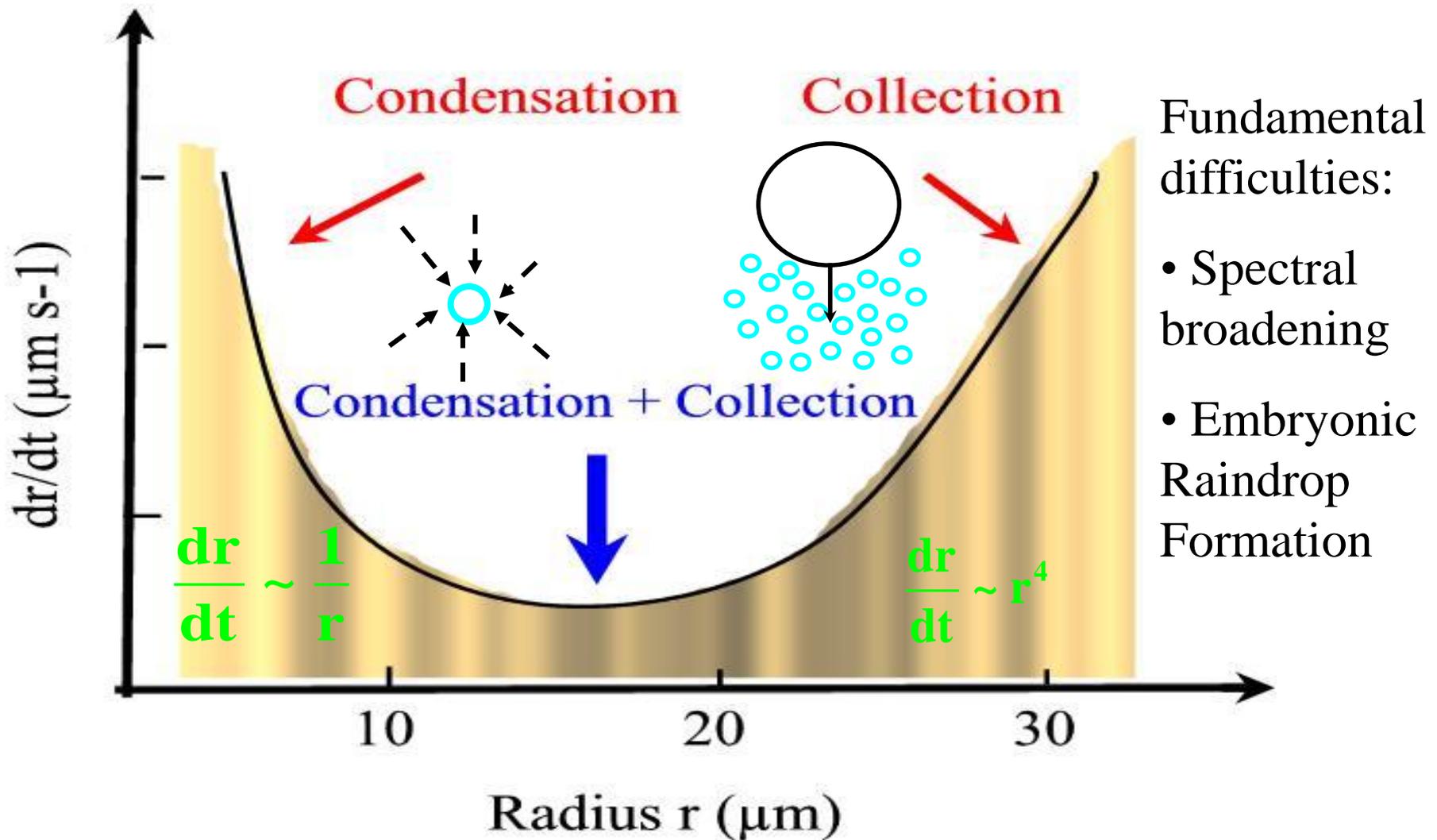


# Spectral broadening is a long-standing puzzle in cloud physics.



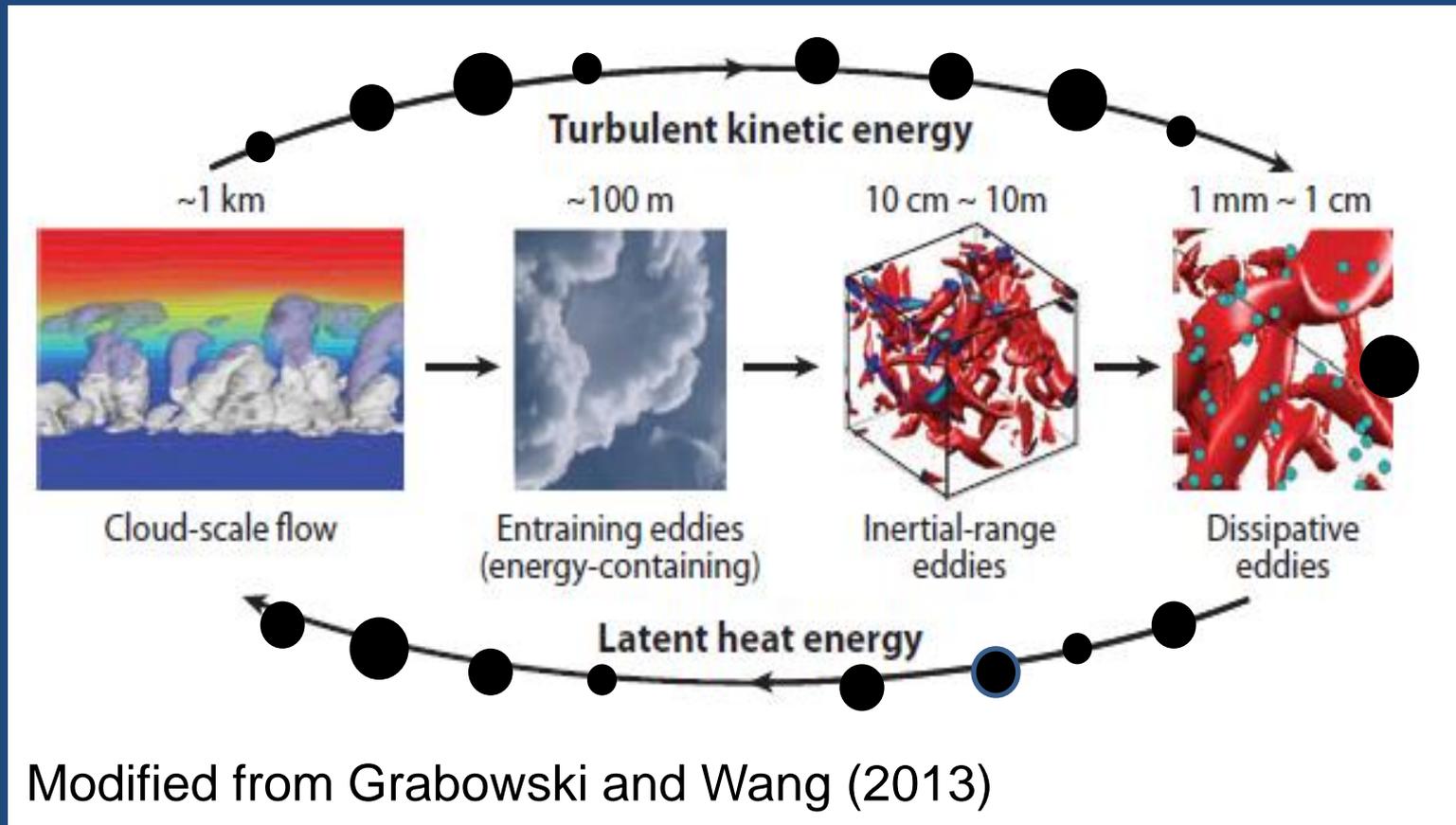
**The conventional condensational theory predicts a droplet size distribution much narrower than observation (Houghton, BAMS, 1938; Howell, J. Met, 1949). Key missing factors are turbulence, entrainment-mixing and associated processes.**

# Valley of Death and Drizzle Initiation



**Rain initiation has been another sticky puzzle in cloud physics since the late 1930s (Arenberg 1939). Key missing factors are related to turbulence as well.**

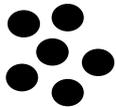
# Knowledge Gaps for Sub-LES Scale Processes



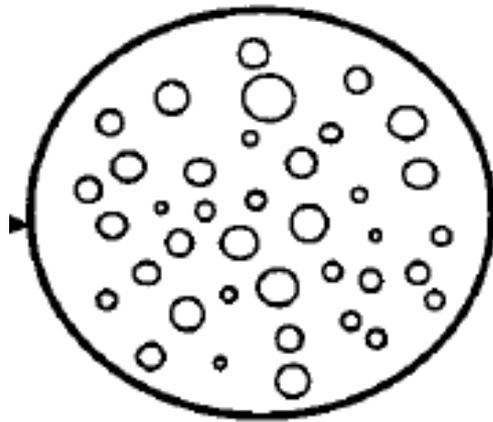
- Turbulence-microphysics interactions
- Entrainment-mixing processes
- Droplet clustering
- Rain initiation

# Fast Physics Parameterization as Statistical Physics

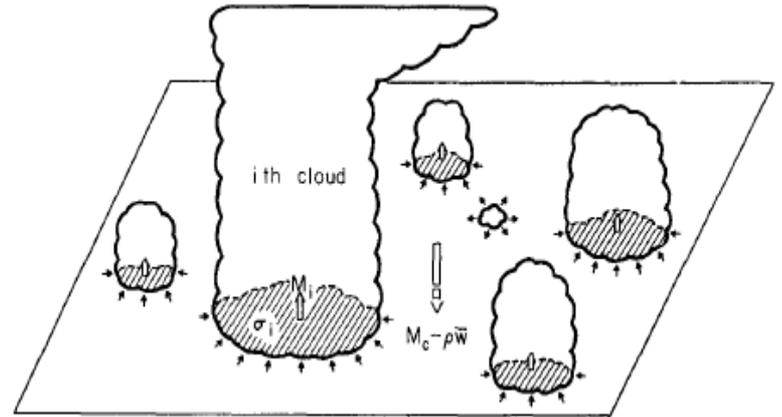
- “Statistical physics” is to account for the observed thermodynamic properties of systems in terms of the statistics of large ensembles of “particles”.
- “Parameterization” is to account for collective effects of many smaller scale processes on larger scale phenomena.



Molecule Ensemble  
Kinetics, Statistical  
Physics, Thermodynamics



Droplet Ensemble  
Systems Theory



Classical Diagram of Cloud Ensemble  
for Convection Parameterization  
(Arakawa and Schubert, 1974, JAS)

# **Statistical Physics for Microphysics Parameterization:**

## **Part I: Most Probable Size Distribution -- Theory for Gamma Size Distribution**

**(Liu et al., AR, 1994, 1995; Liu & Hallett, QJ, 1998; JAS, 1998, 2002; Liu et al, 2002)**

## **Part II: On Rain Initiation -- Autoconversion**

**(McGraw and Liu, PRL, 2003, PRE, 2004; Liu et al., GRL, 2004, 2005, 2006, 2007, 2008)**

# Commonly Used Size Distribution Functions

Table 1.1. Summary of Empirical Expressions for Size Distribution

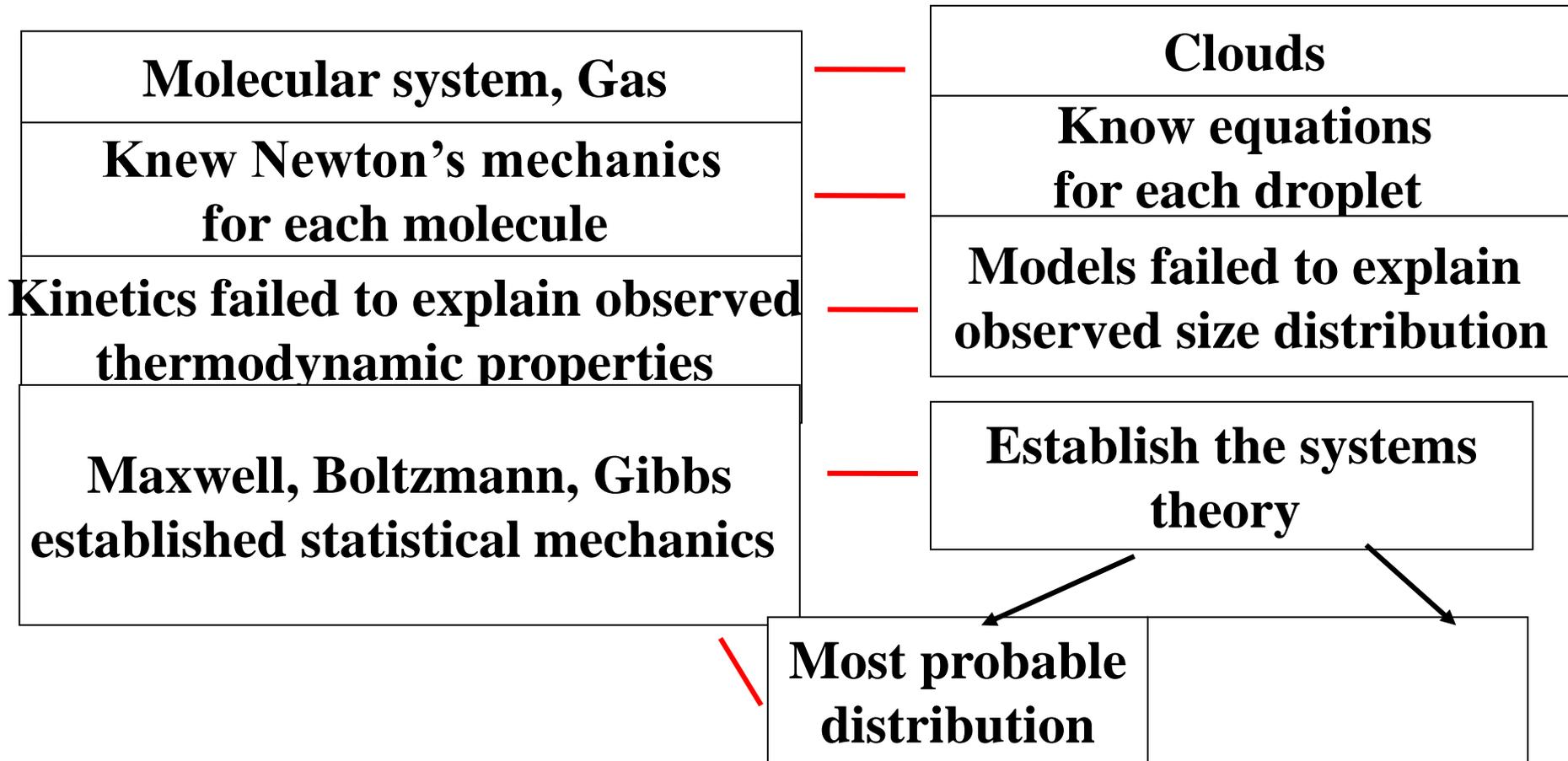
| Name           | Expression  | Parameters             |
|----------------|---|------------------------|
| Weibull        | $n(D) = N_0 D^{q-1} \exp(-\lambda D^q)$   | $N_0, \lambda, q$      |
| Gamma          | $n(D) = N_0 D^\mu \exp(-\lambda D)$   | $N_0, \mu, \lambda$    |
| Lognormal      | $n(D) = \frac{N}{\log \sigma \sqrt{2\pi}} \frac{1}{D} \exp\left(-\frac{\log^2(D/D_m)}{2\log^2 \sigma}\right)$ | $N, D_m, \sigma$       |
| Power-law      | $n(D) = aD^{-b}$  | $a, b$                 |
| Exponential    | $n(D) = N_0 \exp(-\lambda D)$   | $N_0, \lambda$         |
| Normal         | $n(D) = \frac{N}{\sigma \sqrt{2\pi}} \exp\left(-\frac{(D - D_m)^2}{2\sigma^2}\right)$                         | $N, D_m, \sigma$       |
| Modified gamma | $n(D) = N_0 D^\mu \exp(-\lambda D^q)$   | $N_0, \mu, \lambda, q$ |
| Delta function | $n(D) = N \delta(D - D^*)$  | $N, D^*$               |

(Most already summarized in “The Physics of Clouds” by B. J. Mason 1957)

Most microphysics parameterizations are based on the assumption that size distributions follow the Gamma or Weibull distribution >> theoretical framework for this?

# Droplet System vs. Molecular System

**Fluctuations associated with turbulence lead us to assume that droplet size distributions occur with different probabilities, and info on size distributions can be obtained without knowing details of individual droplets.**



# Droplet System

---

Consider the droplet system constrained by

$$\int \rho(\mathbf{x}) d\mathbf{x} = 1 \quad (1)$$

$$\int \mathbf{x} \rho(\mathbf{x}) d\mathbf{x} = \frac{\mathbf{X}}{\mathbf{N}} \quad (2)$$

$\mathbf{x}$  = Hamiltonian variable,  $\mathbf{X}$  = total amount of  $\chi$   
per unit volume,  $n(\mathbf{x})$  = droplet number distribution with  
respect to  $\mathbf{x}$ ,  $\rho(\mathbf{x}) = n(\mathbf{x})/\mathbf{N}$  = probability that a droplet of  $\mathbf{x}$   
occurs.

# Droplet Spectral Entropy

---

**Droplet spectral entropy is defined as**

$$E = - \int \rho(\mathbf{x}) \ln(\rho(\mathbf{x})) d\mathbf{x} \quad (3)$$

**Note the correspondence between the Hamiltonian variable  $\mathbf{x}$  and the constraint  $N \int \mathbf{x} \rho(\mathbf{x}) d\mathbf{x} = \mathbf{X}$**

# Most Probable Distribution w.r.t. $\mathbf{x}$

Maximizing the spectral entropy  
subject to the two constraints given by Eqs. (1) and (2)  
yields the most probable PDF with respect to  $\mathbf{x}$ :

$$\rho^* (\mathbf{x}) = \frac{1}{\alpha} \exp \left( -\frac{\mathbf{x}}{\alpha} \right) \quad (4)$$

The most probable distribution with respect to  $\mathbf{x}$  is

$$\mathbf{n}^* (\mathbf{x}) = \frac{N}{\alpha} \exp \left( -\frac{\mathbf{x}}{\alpha} \right) \quad (5)$$

where  $\alpha = X/N$  represents the mean amount of  $\mathbf{x}$  per droplet. Note that the Boltzmann energy distribution becomes special of Eq. (5) when  $\mathbf{x}$  = molecular energy. The physical meaning of  $\alpha$  is consistent with that of “ $k_B T$ ”, or the mean energy per molecule.

# Most Probable Droplet Size Distribution

---

Assume that the Hamiltonian variable  $x$  and droplet radius  $r$  follow a power-law relationship

$$\mathbf{x} = \mathbf{a}r^b$$

Substitution of the above equation into the exponential most probable distribution with respect to  $x$  yields the most probable droplet size distribution:

$$\mathbf{n}^*(\mathbf{r}) = \mathbf{N}_0 \mathbf{r}^{b-1} \exp(-\lambda \mathbf{r}^b)$$

$$\mathbf{N}_0 = \mathbf{a}b/\alpha; \lambda = \mathbf{a}/\alpha; \alpha = \mathbf{X}/\mathbf{N}$$

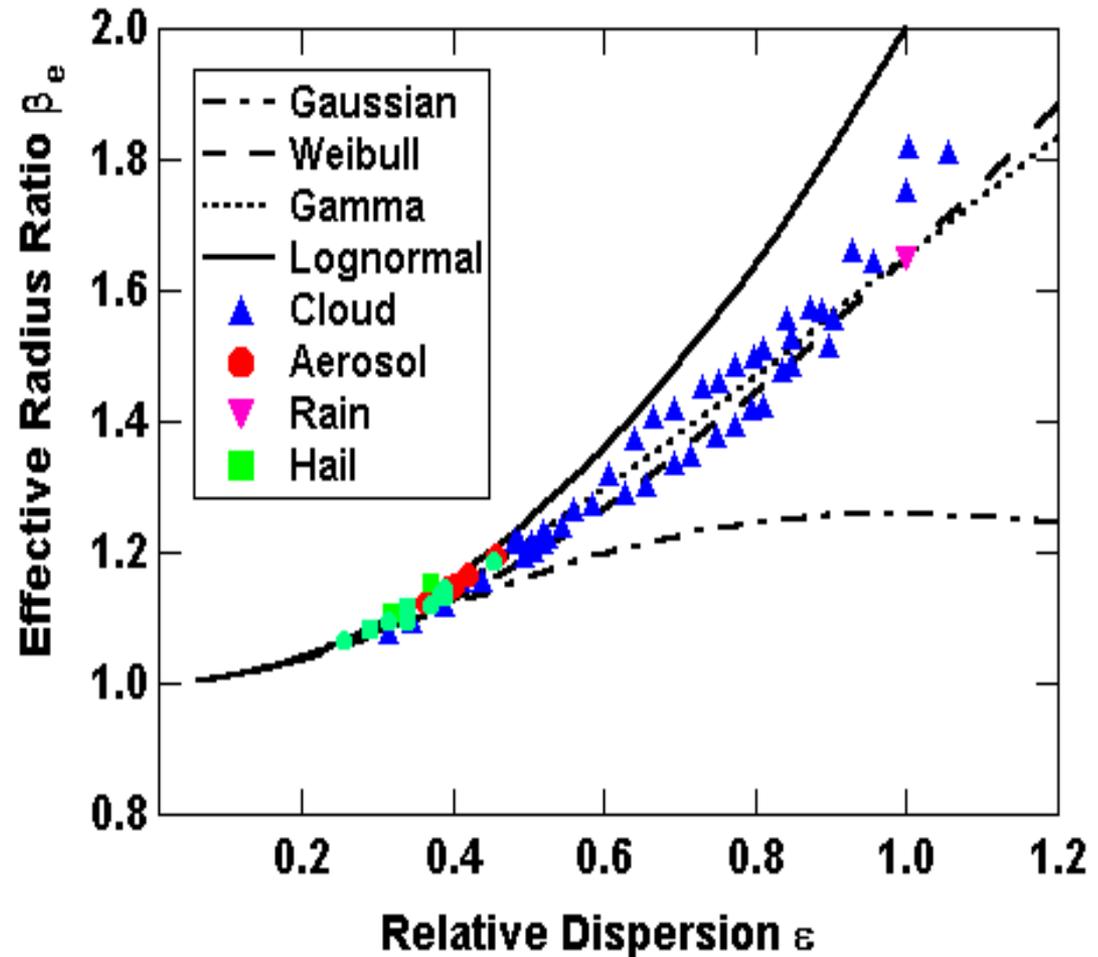
**This is a general Weibull distribution!**

# Observational Validation of Weibull/Gamma Particle Distribution

- Each point represents a particle size distribution
- $\varepsilon$  = Standard deviation/mean

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

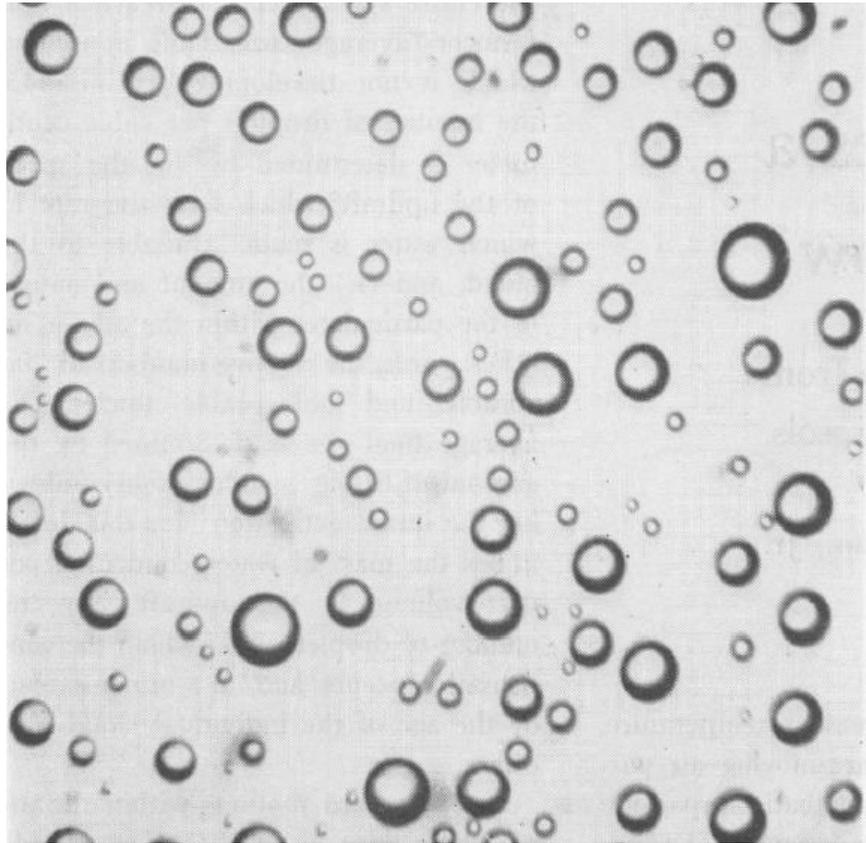
$$\beta = \frac{(1 + 2\varepsilon^2)^{2/3}}{(1 + \varepsilon^2)^{1/3}}$$



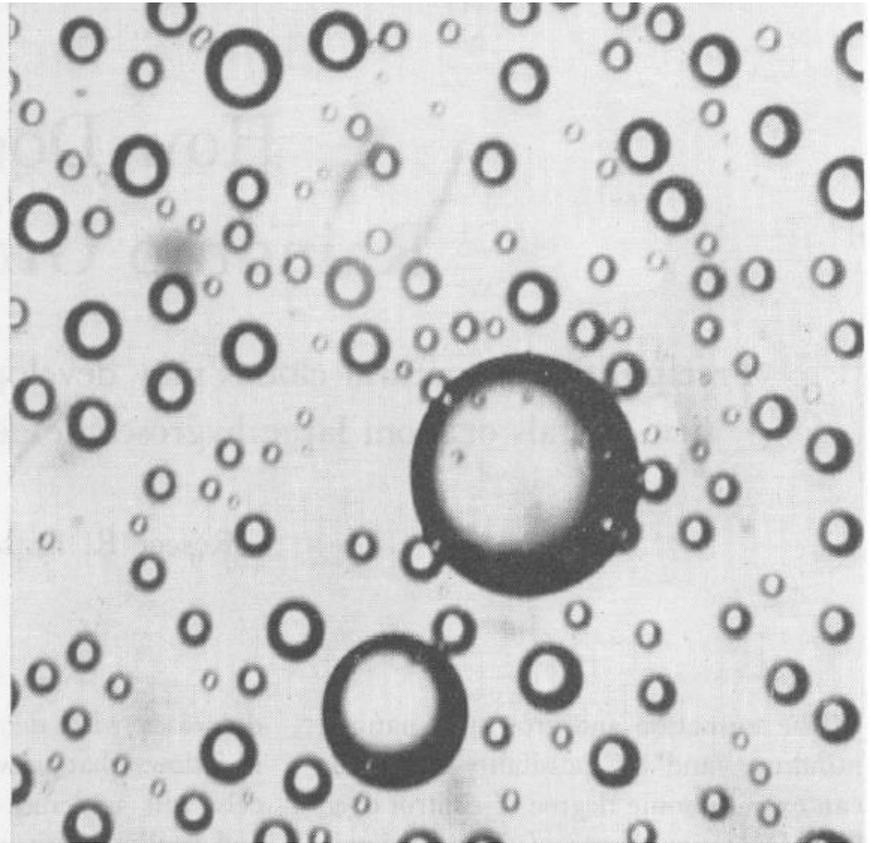
Aerosol, cloud droplet and precipitation particles share a common distribution form ---- Weibull or Gamma, suggesting a **unified theory on particle size distributions.**

# Autoconversion process is the 1<sup>st</sup> step for cloud droplets to grow into raindrops.

Nonprecipitating clouds



Precipitating clouds



Autoconversion was intuitively/empirically introduced to parameterize microphysics in cloud models in the 1960s as a practical convenience, and later has been adopted in models of other scales (e.g., LES, MM5, WRF, GCMs). The concept has been loose; I'll give a rigorous definition later.

# Autoconversion and its Parameterization

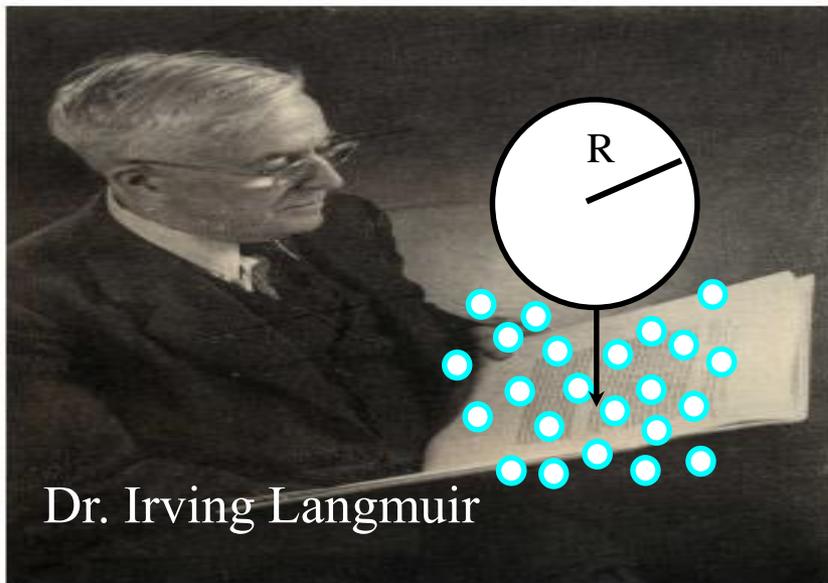
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- Autoconversion is the **first step** converting cloudwater to rainwater; autoconversion rate  $P = P_0 T$  ( $P_0$  is rate function &  $T$  is threshold function).
- Approaches for developing parameterizations over the last 4 decades:
  - \* educated guess (e.g., Kessler 1969; Sundqvist 1978)
  - \* curve-fit to detailed model simulations (e.g., Berry 1968)
- Previous studies have been primarily on  $P_0$  and existing parameterizations can be classified into three types according to their ad hoc  $T$ :
  - \* Kessler-type ( $T =$  Heaviside step function)
  - \* Berry-type ( $T = 1$ , without threshold function)
  - \* Sundqvist-type ( $T =$  Exponential-like function)
- Existing parameterizations have elusive physics and tunable parameters.

Our focus has been deriving  $P_0$  and  $T$  from first principles and eliminating the tunable parameters as much as possible.

# Rate Function $P_0$

Simple model: A drop of radius  $R$  falls through a polydisperse population of smaller droplets with size distribution  $n(r)$  (Langmuir 1948, J. Met).



Nobel prize winner & pioneer in weather modification in 1940s.

**Autoconversion = Collection of cloud droplets by small raindrops**

The mass growth rate of the drop is

$$\frac{dm}{dt} = \int \mathbf{k}(\mathbf{R}, \mathbf{r}) m(\mathbf{r}) n(\mathbf{r}) d\mathbf{r}$$

The rate function  $P_0$  is then given by

$$P_0 = \int \frac{dm}{dt} n(\mathbf{R}) d\mathbf{R}$$

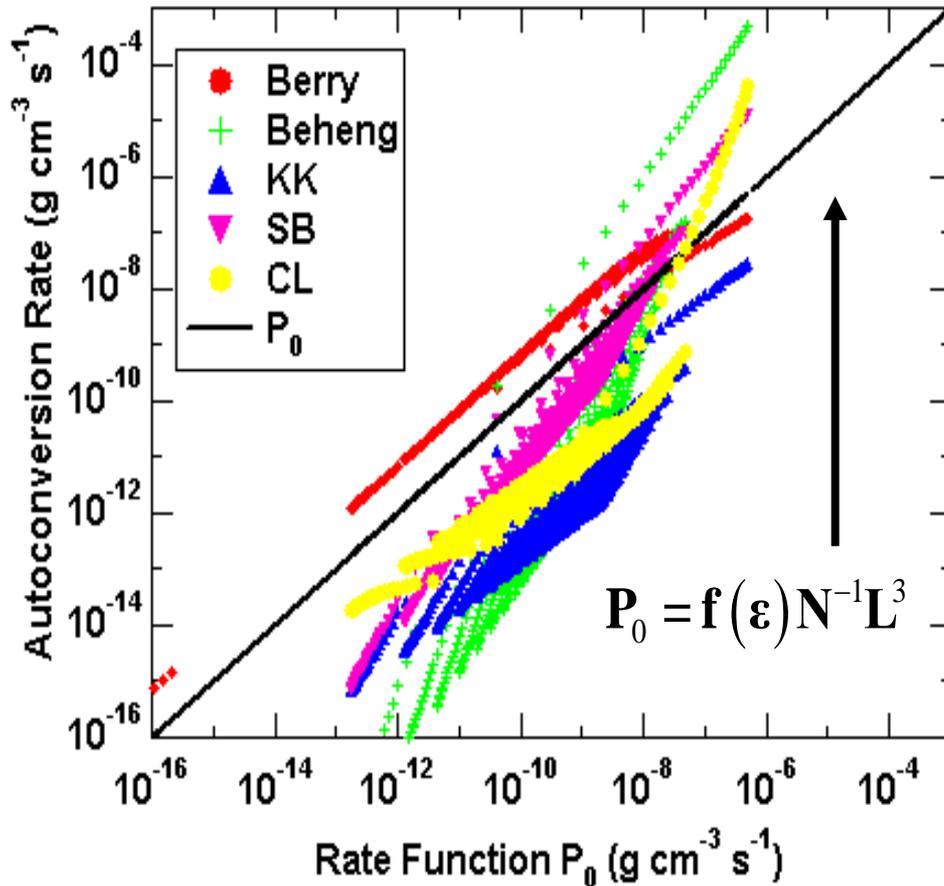
Generalized mean value theorem for integrals:

$$\int \mathbf{f}(\mathbf{x}) \mathbf{g}(\mathbf{x}) d\mathbf{x} = \mathbf{f}(\mathbf{x}_0) \int \mathbf{g}(\mathbf{x}) d\mathbf{x}$$

Application of the above equations with various collection kernels recovers existing parameterizations and yields a new one.

(Liu & Daum 2004; Liu et al. 2006, JAS)

# Comparison of New Rate Function with Simulation-Based Parameterizations



- Simulation-based parameterizations are obtained by fitting simulations to a simple function such as a power-law.

- Such a simple function fit distorts either P<sub>0</sub> or T (hence P) in P = P<sub>0</sub>T.

The rate function P<sub>0</sub> can be expressed as an analytical function of droplet concentration N, liquid water content L, and relative dispersion ε (Liu & Daum 2004; Liu et al. 2006, JAS).

# Kessler-Type Autoconversion Parameterizations

Table 1. Kessler-type Autoconversion Parameterizations

$$P = P_0 H(r_d - r_c)$$

|                 | <b>Expression</b>                                 | <b>Assumption</b>                      | <b>Features</b>   |
|-----------------|---|--|---|
| <b>Previous</b> | $P = \gamma N^{-1/3} L^{7/3} H(r_3 - r_c)$        | <b>Fixed collection efficiency</b>     | <b>Fixed <math>\gamma</math>, no <math>\varepsilon</math> effect, <math>r_d = r_3</math></b>                          |
| <b>New</b>      | $P_{LD} = f(\varepsilon) N^{-1} L^3 H(r_6 - r_c)$ | <b>Realistic collection efficiency</b> | <b>Has <math>\varepsilon</math>, stronger dependence on <math>L</math> and <math>N</math>, <math>r_d = r_6</math></b> |

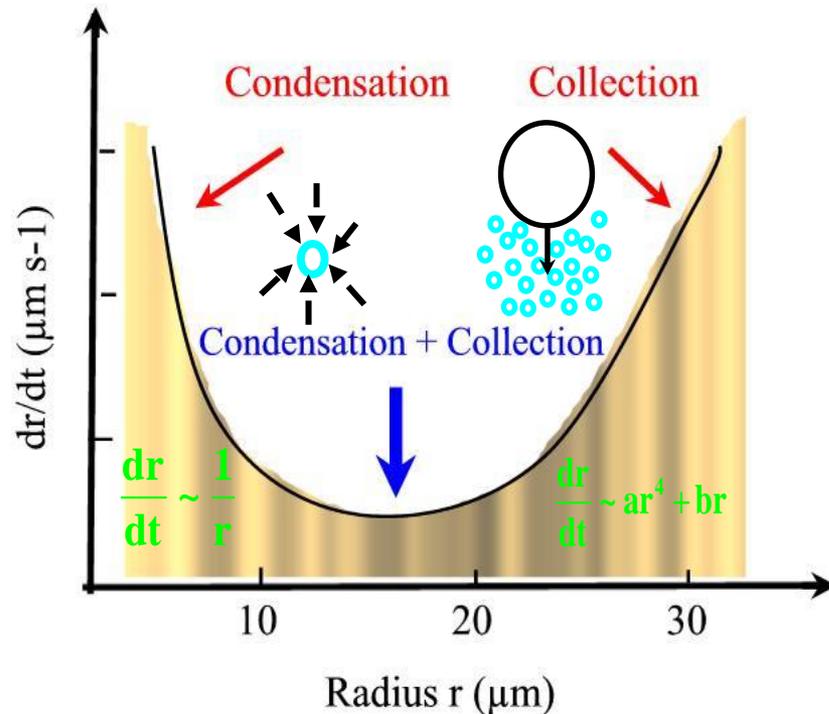
$r_3 = 3^{\text{rd}}$  moment mean radius;  $r_6 = 6^{\text{th}}$  moment mean radius

$H =$  Heaviside step function (Liu & Daum 2004, JAS).

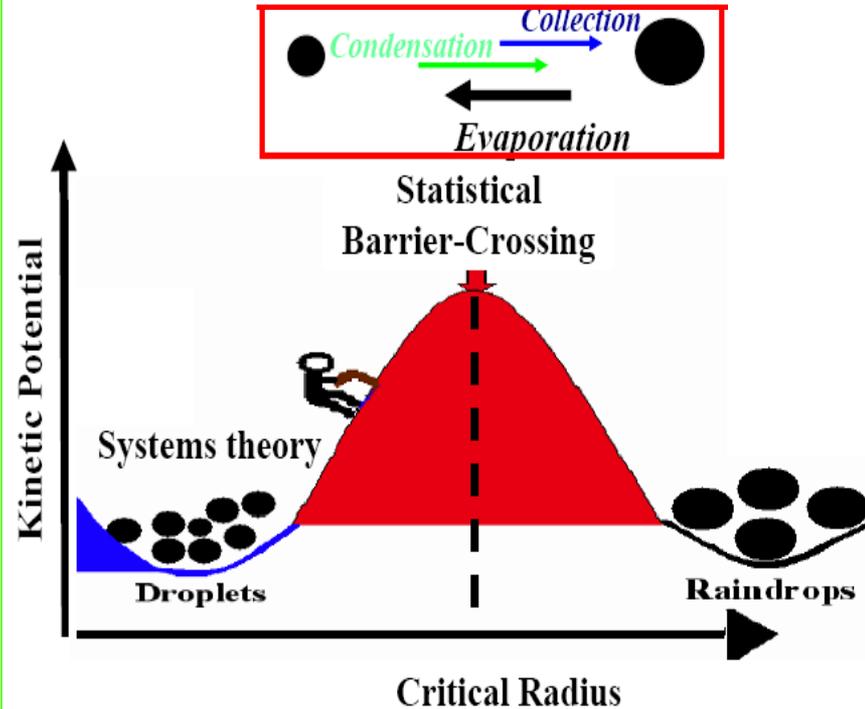
**What about the critical radius  $\gg$  rain initiation theory?**

# Systems Theory of Rain Initiation/Autoconversion

## Valley of Death



## Mountain of Life

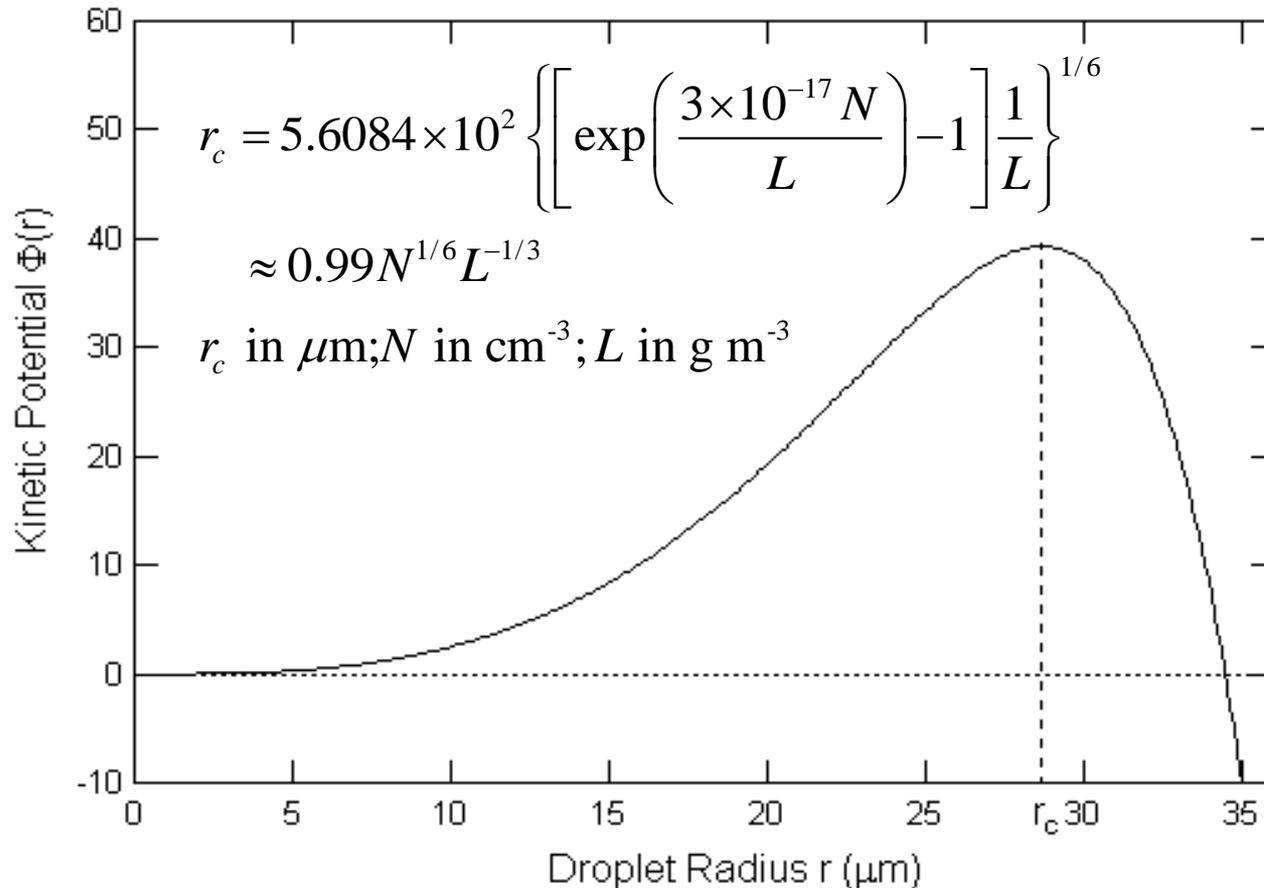


**The new theory considers rain initiation as a statistical barrier crossing process. Only those "RARE SEED" drops crossing over the barrier grow into raindrops.**

Rain initiation has been an outstanding puzzle with two fundamental problems of spectral broadening & formation of embryonic raindrop

**The new theory combines statistical barrier crossing with the systems theory for droplet size distributions, leading to analytical expression for critical radius (Phys. Rev. Lett., 2003; Phys. Rev., 2004; GRL, 2004, 2005, 2006, 2007).**

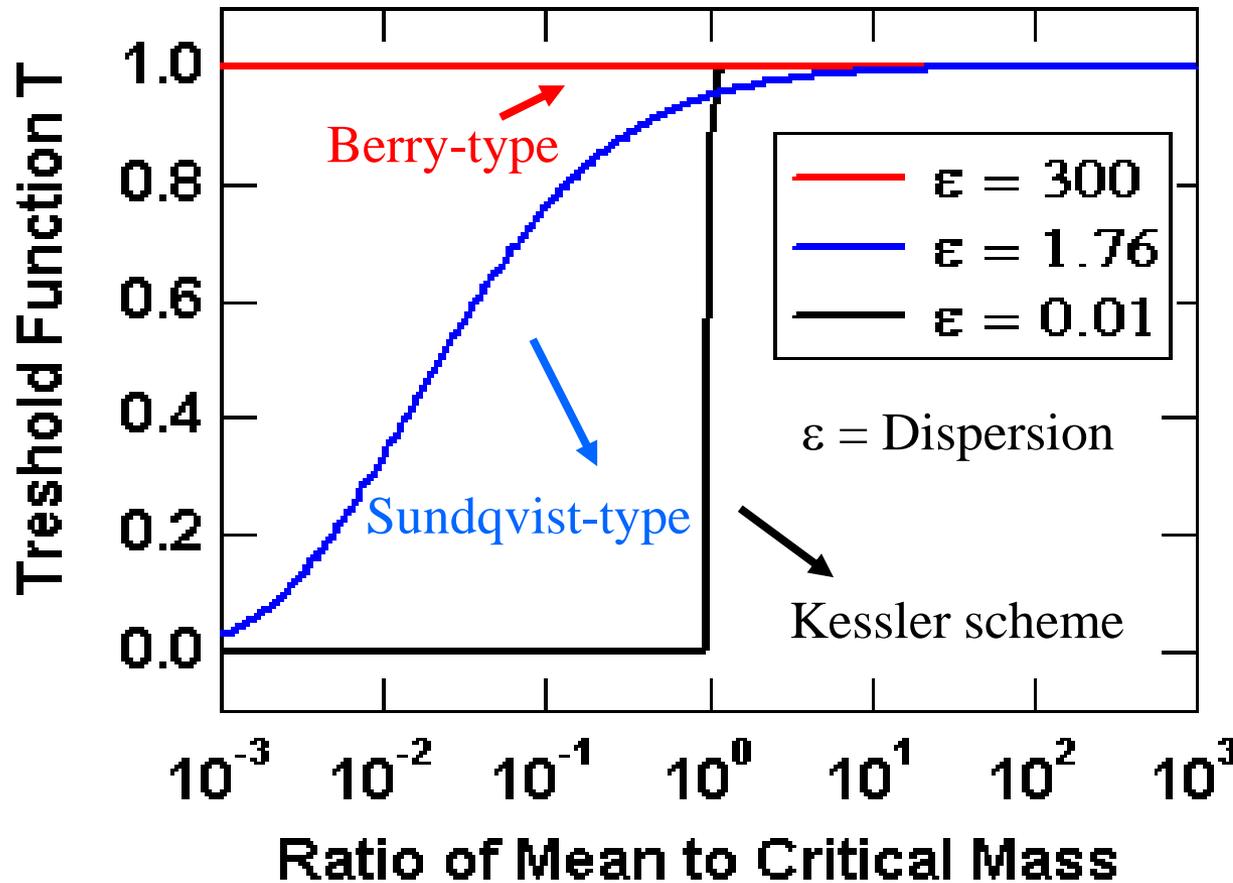
# Critical Radius & Analytical Expression



- Kinetic potential peaks at critical radius  $r_c$ .
- Critical radius & potential barrier both increase with droplet concentration.
- 2<sup>nd</sup> AIE: Increasing aerosols inhibit rain by enhancing the barrier and critical radius.

**Critical radius is the liquid water content and droplet concentration, eliminating the need to tune this parameter (McGraw & Liu 2003, Phys. Rev. Lett.; 2004, Phys. Rev. E; and Liu et al. 2004, GRL).**

# Relative dispersion is critical for determining the threshold function



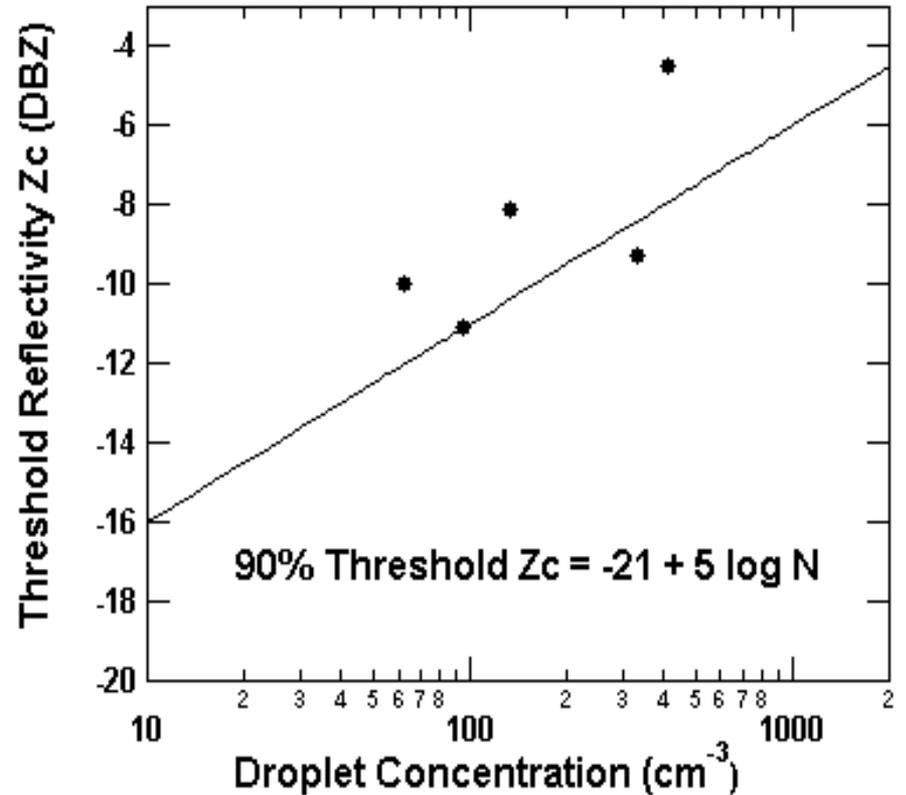
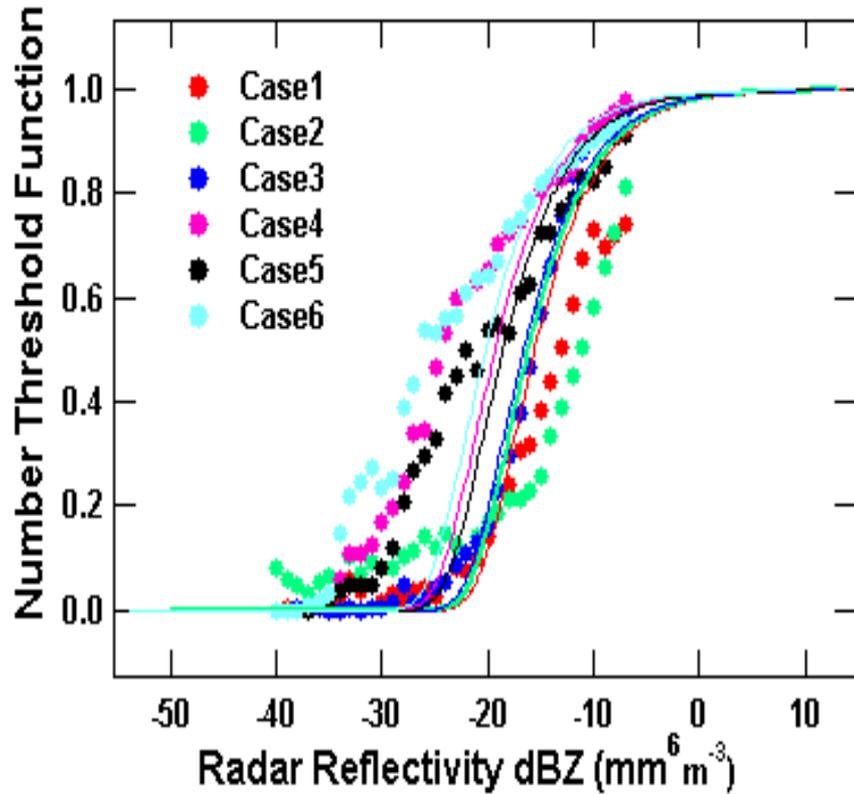
Truncating the cloud droplet size distribution at critical radius yields the threshold function:

$$T = \frac{P}{P_0}$$

Further application of the Weibull size distribution leads to the general  $T$  as a function of mean-to-critical mass ratio and relative dispersion.

The new threshold function unifies existing ad hoc types of threshold functions, and reveals the important role of relative dispersion that has been unknowingly hidden in ad hoc threshold functions (Liu et al., GRL, 2005, 2006, 2007).

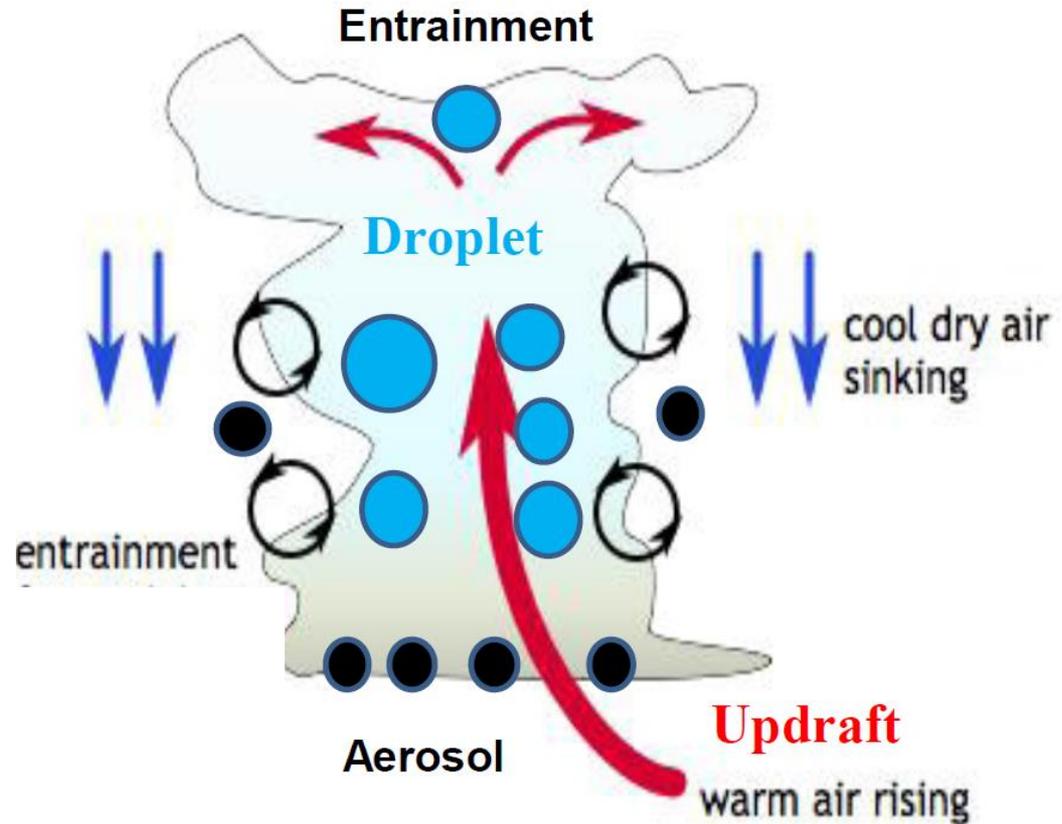
# Observational Validation of Threshold Function



The results explain why empirically determined threshold reflectivity varies, provides observational validation for our theory, and additional support for the notion that aerosol-influenced clouds tend to hold more water or a larger LWP (Liu et al., GRL, 2007, 2008).

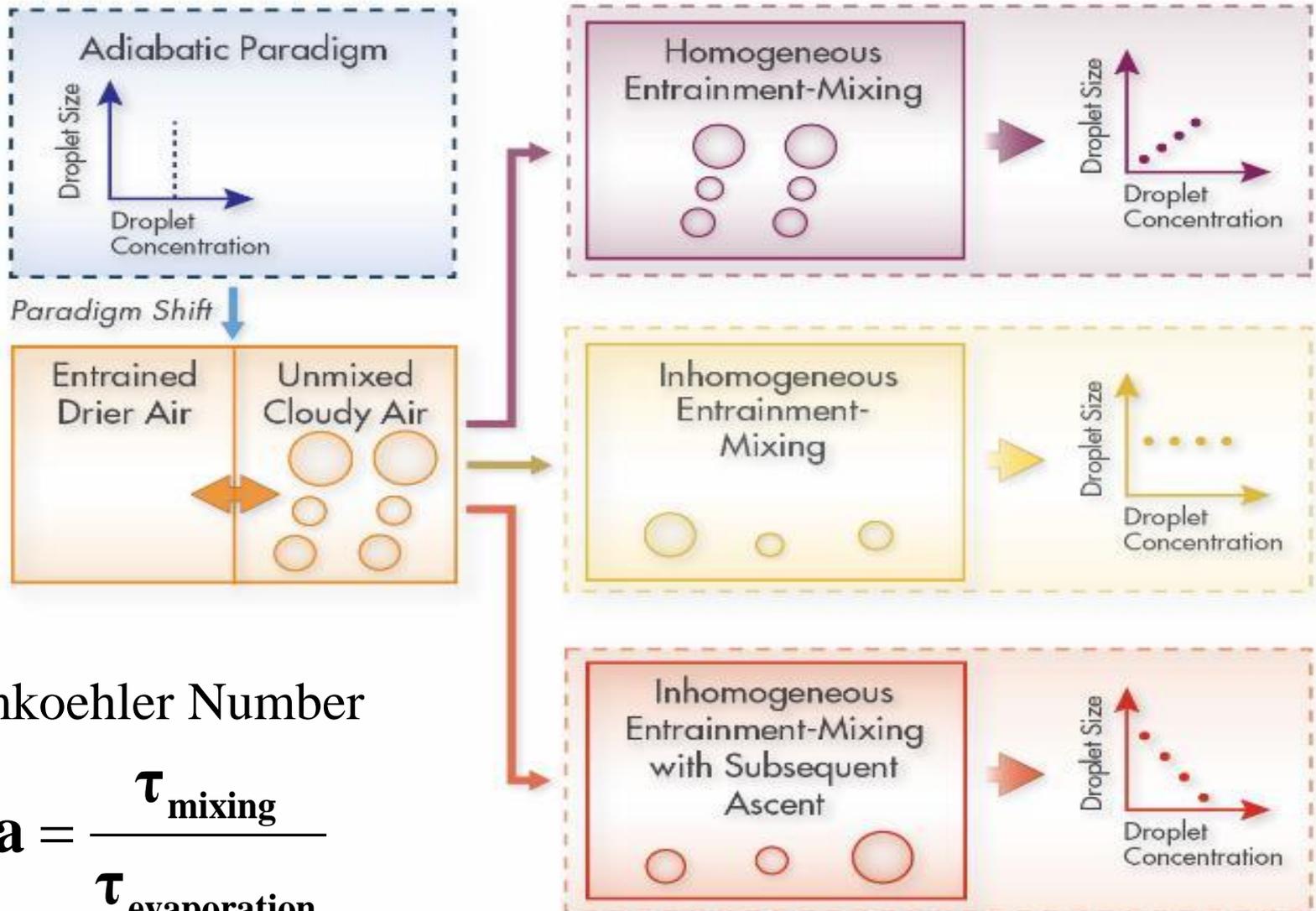
# Clouds are open multi-physics & multi-scale Systems

- Entrainment Rate
- Vertical velocity
- Buoyancy
- Dissipation
- Environment
- **Turbulent mixing**
- **Microphysics**
- Aerosol
- Couplings



**Turbulence, related entrainment-mixing processes, and their interactions with microphysics are key to the outstanding puzzles.**

# Different entrainment-mixing processes alter cloud properties significantly.

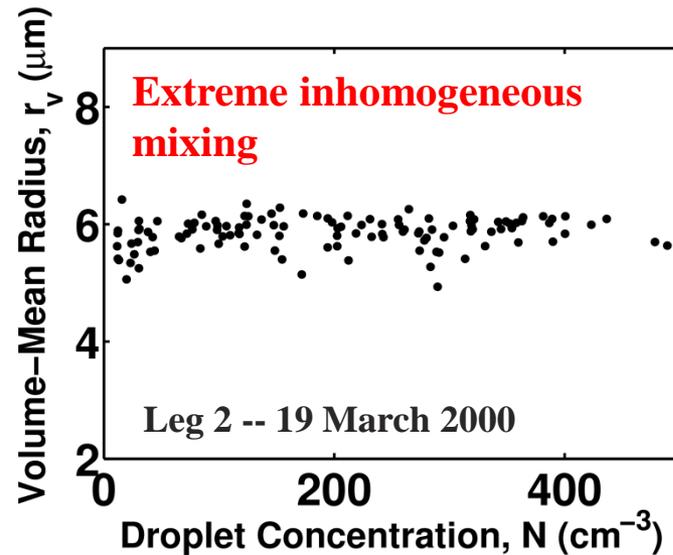
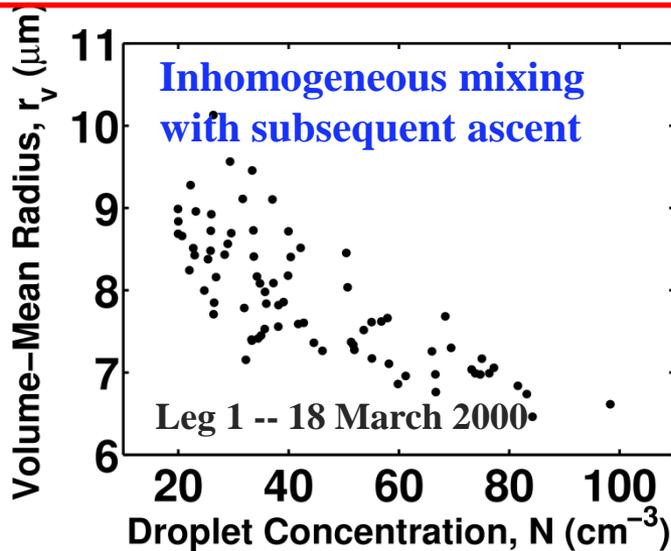
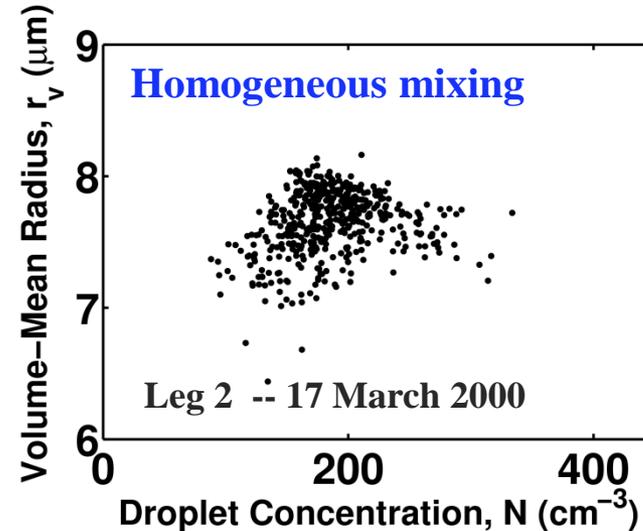
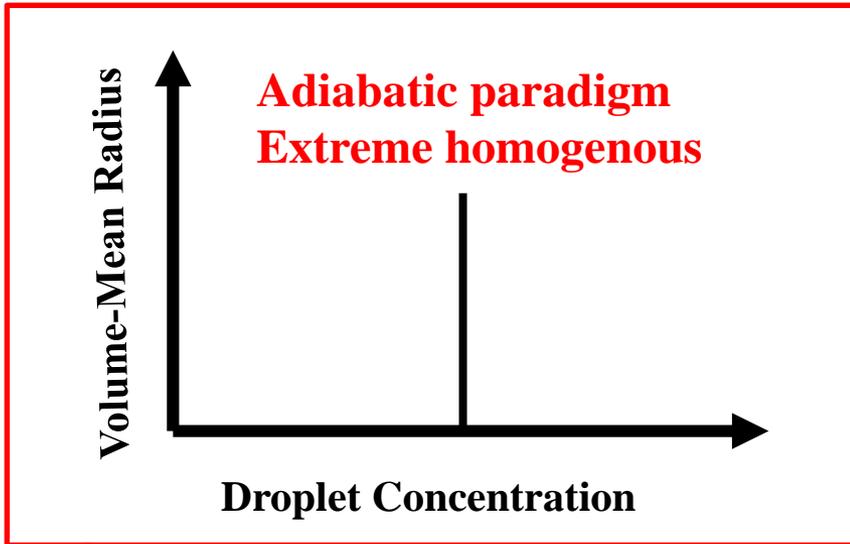


Damkoehler Number

$$Da = \frac{\tau_{\text{mixing}}}{\tau_{\text{evaporation}}}$$

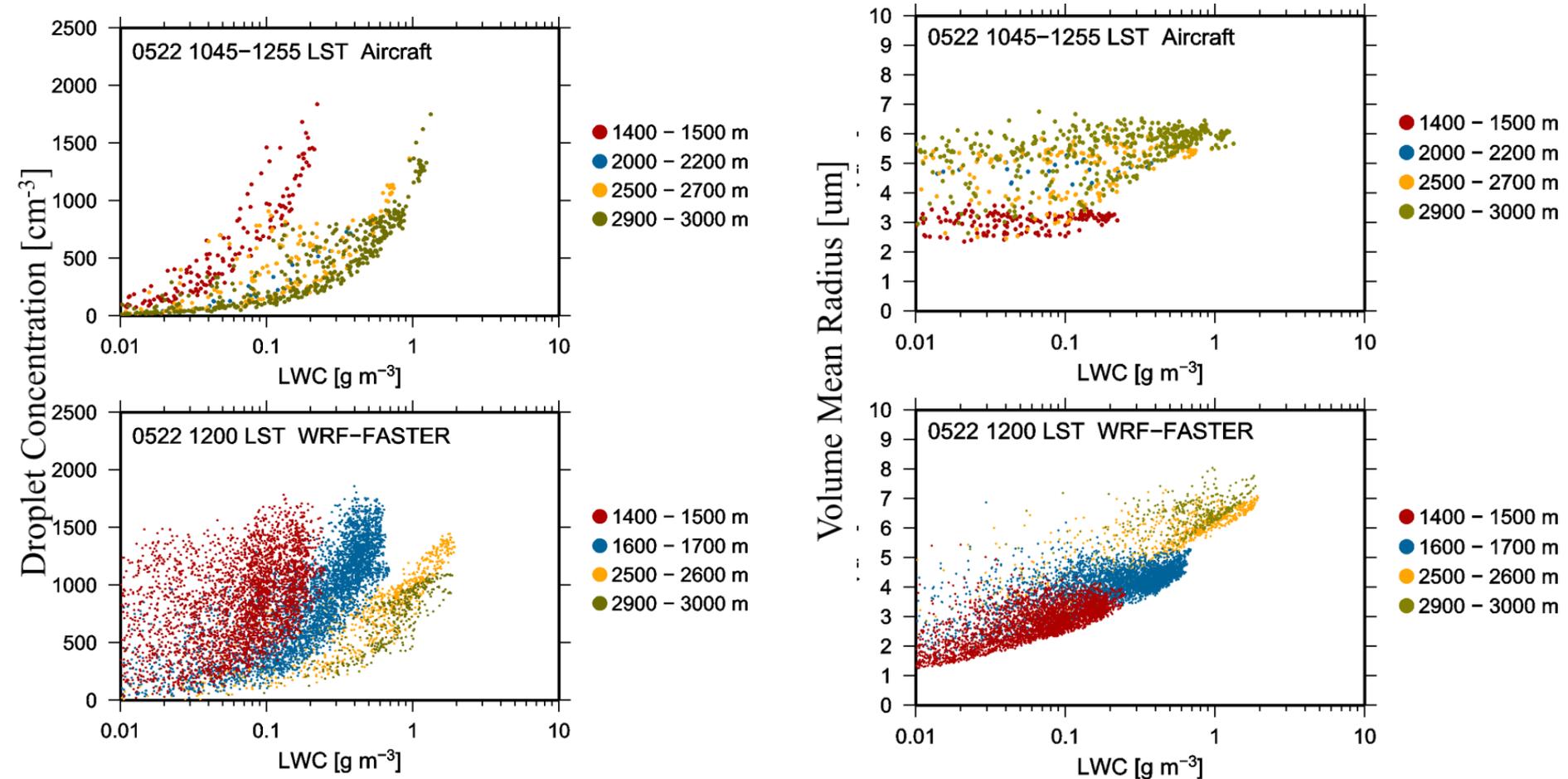
# Observational Examples

## March 2000 Cloud IOP at SGP



*A measure is needed to cover all!*

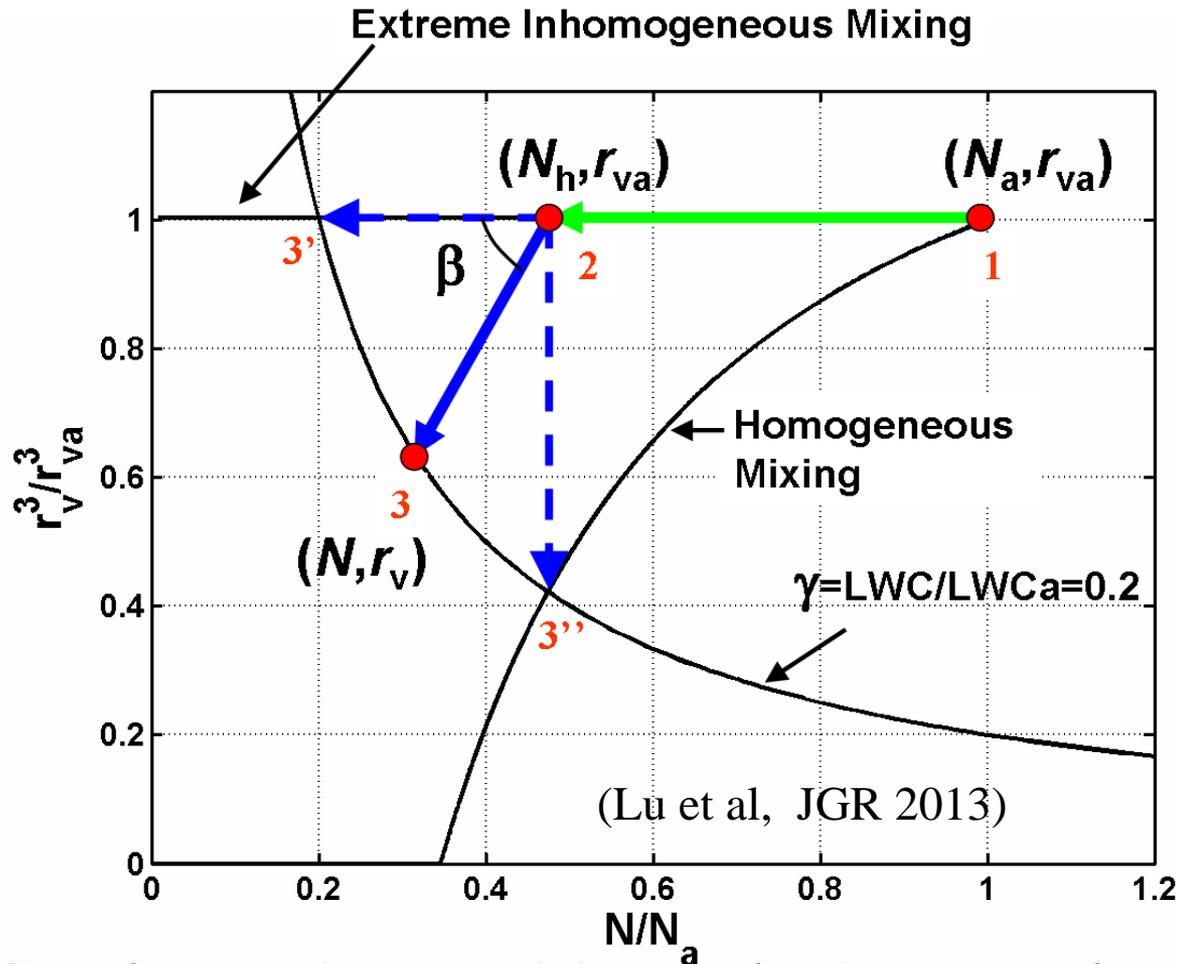
# LES Cannot Capture Observed Mixing Types



**LES captures the general trend of co-variation of droplet concentration and LWC; but the LES mixing type tend to be more homogeneous than observations (left panel).**

*(Endo et al JGR, 2014)*

# Microphysical Mixing Diagram & Homogeneous Mixing Degree



$$\Psi_1 = \frac{\beta}{\pi / 2}$$

A measure for all mechanisms

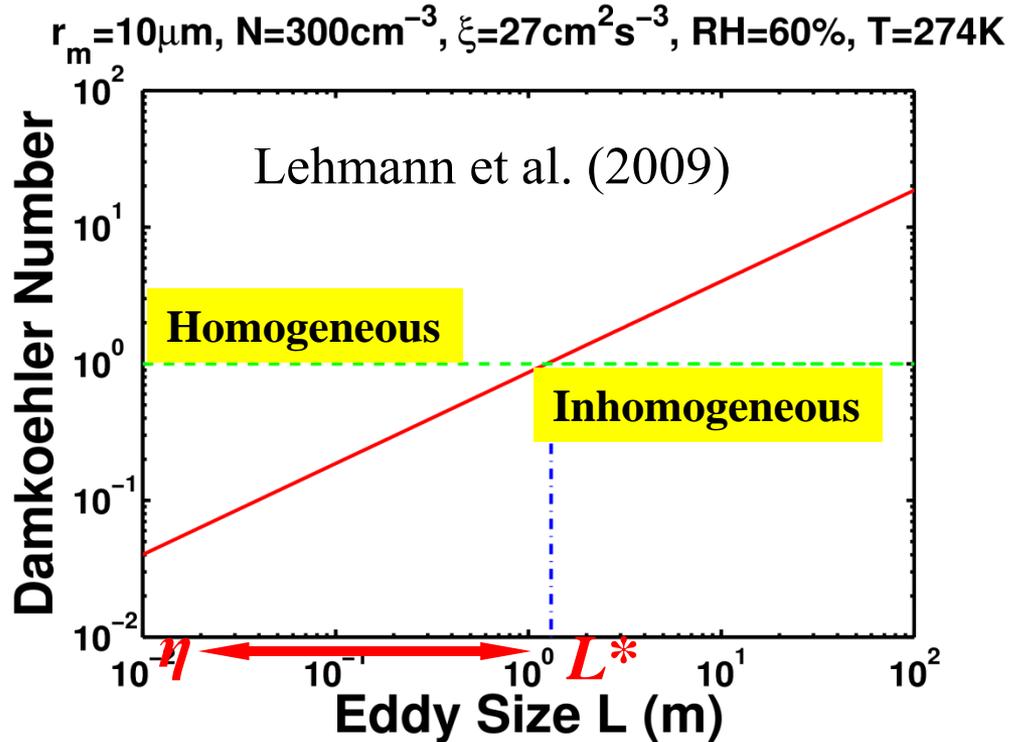
$\Psi_1 = 0$  for extreme inhomogeneous

$\Psi_1 = 1$  for extreme homogeneous

(Lu et al, JGR, 2012, 2013, 2014)

*Complex entrainment-mixing mechanisms are reduced to one quantity: slope (Andrejczuk et al., 2009), or homogeneous mixing degree (Lu et al., 2013).*

# Dynamical Measure: Damkholer Number vs. Transition Scale Number



$$\text{Da} = \frac{\tau_{\text{mixing}}}{\tau_{\text{evaporation}}}$$

A larger  $N_L$  indicates a higher degree of homogeneous mixing.

- Transition length  $L^*$  is the eddy size of  $\text{Da} = 1$ :

$$\tau_{\text{mixing}} = \tau_{\text{evap}}$$

$$\tau_{\text{mix}} \sim (L^2 / \xi)^{1/3}$$

$$L^* = \xi^{1/2} \tau_{\text{evap}}^{3/2}$$

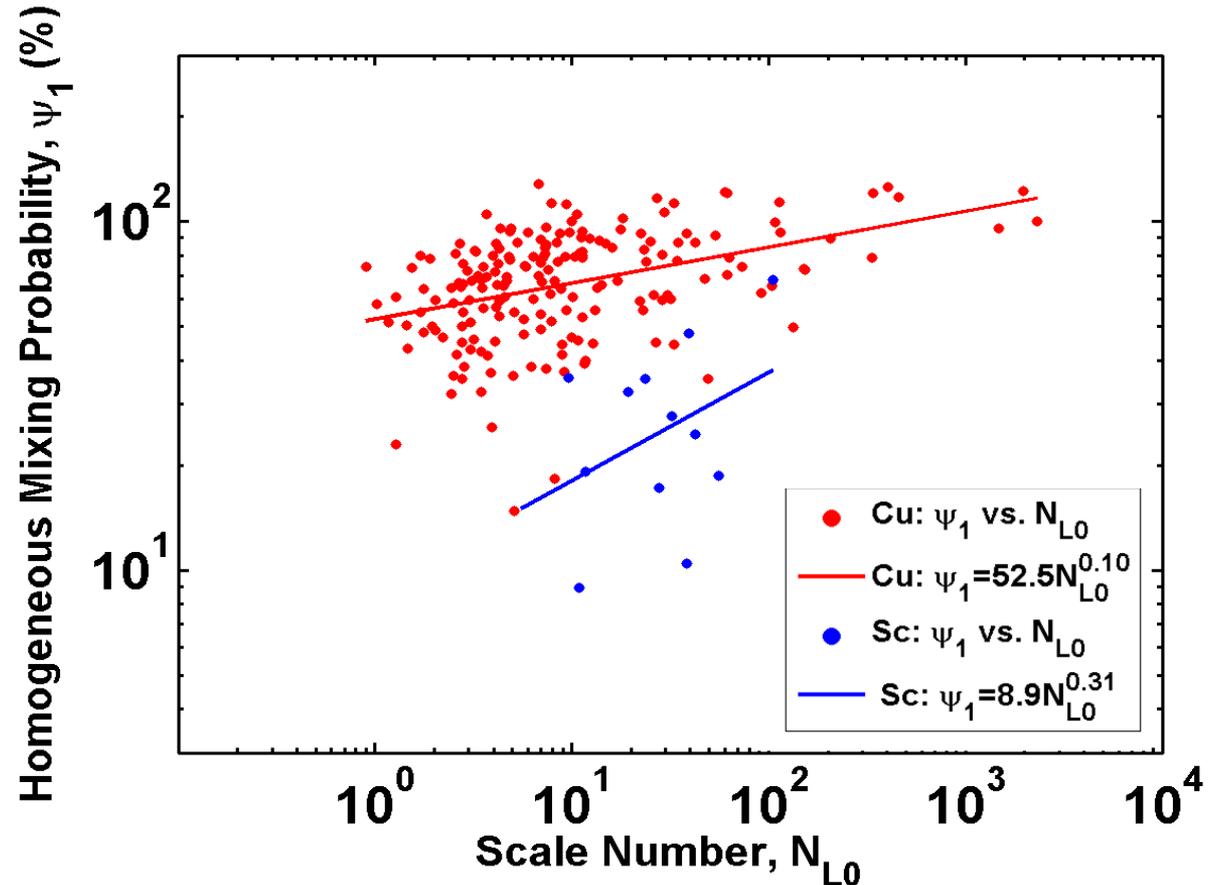
- Transition scale number:

$$N_L = \frac{L^*}{\eta} = \frac{\xi^{1/2} \tau_{\text{evap}}^{3/2}}{\eta} = \frac{\xi^{3/4} \tau_{\text{evap}}^{3/2}}{\nu^{3/4}}$$

$\eta$ : Kolmogorov scale;  $\xi$ : dissipation rate;  $\nu$ : viscosity

# Parameterization for Mixing Mechanisms

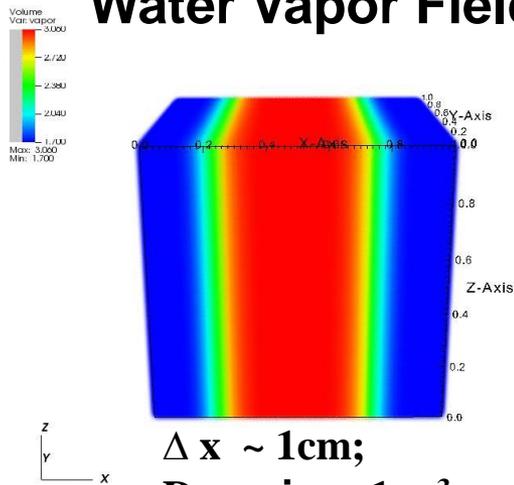
- Eliminate the need for assuming mixing mechanisms
- Scale number can be calculated in models with 2-moment microphysics
- Difference between Cu and Sc ?
- Limited sampling resolutions in obs.



*The parameterization for entrainment-mixing processes is further explored by use of particle-resolved DNS (Gao et al., JGR, 2017)*

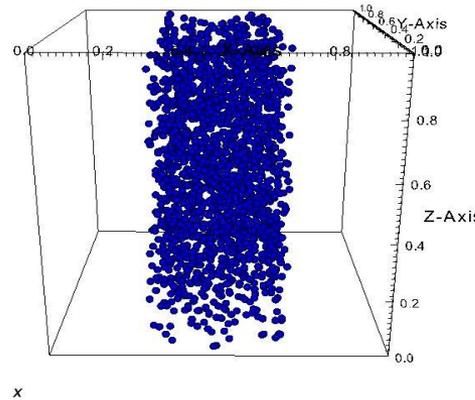
# Our Particle-Resolved DNS

## Water Vapor Field



$\Delta x \sim 1\text{cm};$   
Domain  $\sim 1\text{ m}^3$

## Droplets in Motion



**Turbulent motion and deformation at sub-LES grid scales can generate complex structures and droplet tracks.**

- LES does not resolve turbulent processes that occur at scales smaller than LES grid size and are critical for turbulence-microphysics (**knowledge gap**).
- Bridge the scales between LES grid size and smallest eddies (e.g., 1 mm  $\sim$  1 – 100 m), tracks individual droplets, and serve as a benchmark for spectral bin models
- Provide a powerful tool for studying turbulence-microphysics interactions and entrainment-mixing processes (**knowledge gap**), and informing related parameterization development (**parameterization gap**).

# Main DNS Equations

$$\nabla \cdot \vec{u} = 0$$

$$\partial_t \vec{u} + (\vec{u} \cdot \nabla) \vec{u} = -\frac{1}{\rho_0} \nabla p + \nu \nabla^2 \vec{u} + \vec{f}$$

$$\partial_t T + (\vec{u} \cdot \nabla) T = \kappa \nabla^2 T + \frac{L}{c_p} C_d$$

$$\partial_t q_v + (\vec{u} \cdot \nabla) q_v = \kappa_q \nabla^2 q_v - C_d$$

$$\vec{f} = g \left[ \frac{T - T_0}{T_0} + \epsilon (q_v - q_{v0}) - q_l \right] \vec{e}_z$$

**Fluid Dynamics**

$$S = \frac{q_v}{q_{vs}} - 1$$

$$C_d(\vec{x}, t) = \frac{1}{\rho_0 a^3} \sum_{\beta=1}^{\Delta} \frac{dm_l(\vec{X}_\beta, t)}{dt}$$

**Microphysics**

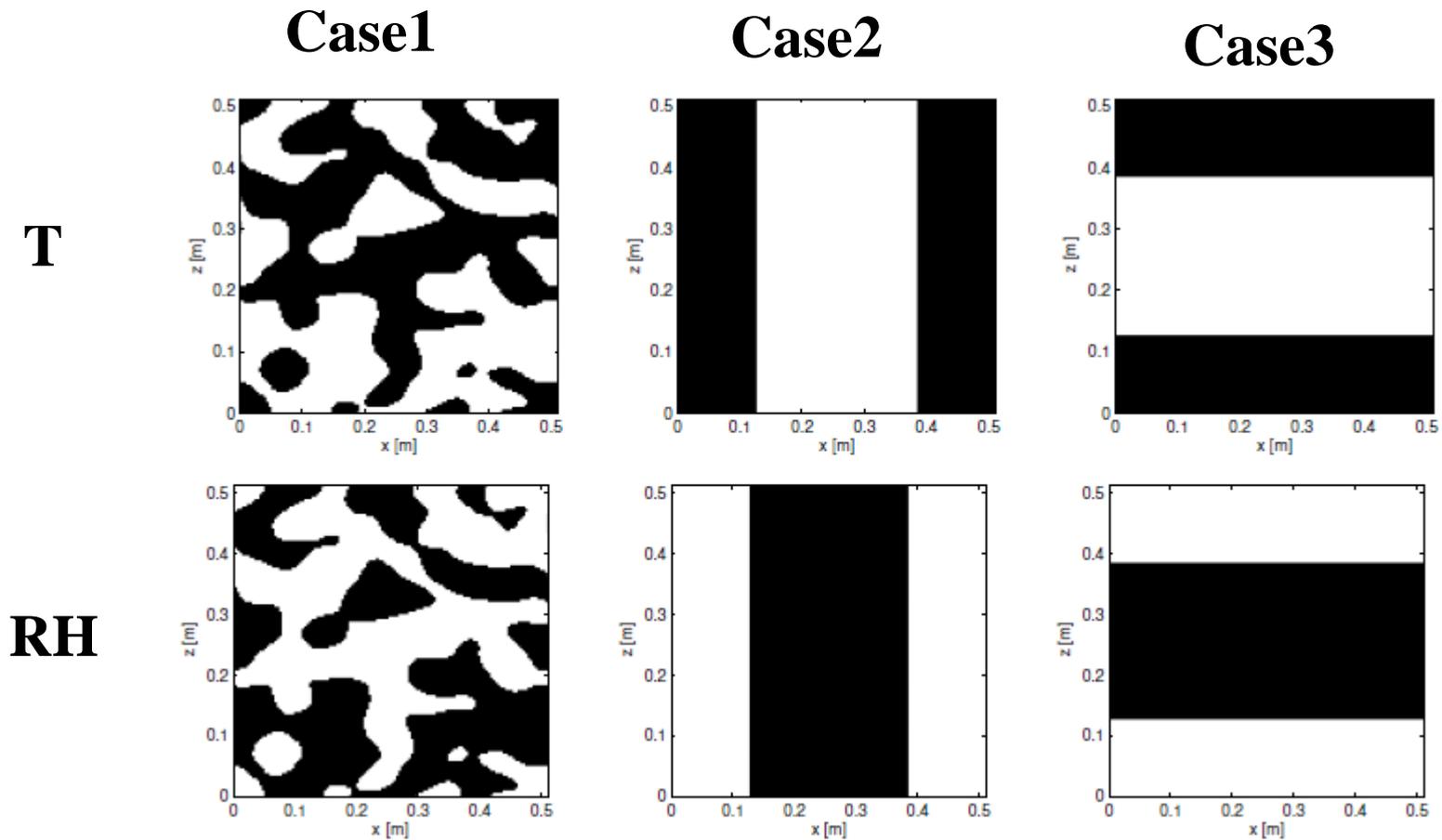
$$\frac{d\vec{X}}{dt} = \vec{V}(\vec{X}, t)$$

$$\frac{d\vec{V}}{dt} = \frac{1}{\tau_p} [\vec{u}(\vec{X}, t) - \vec{V}] + \vec{g}$$

$$r(\vec{X}, t) \frac{d\vec{r}(\vec{X}, t)}{dt} = KS(\vec{X}, t)$$

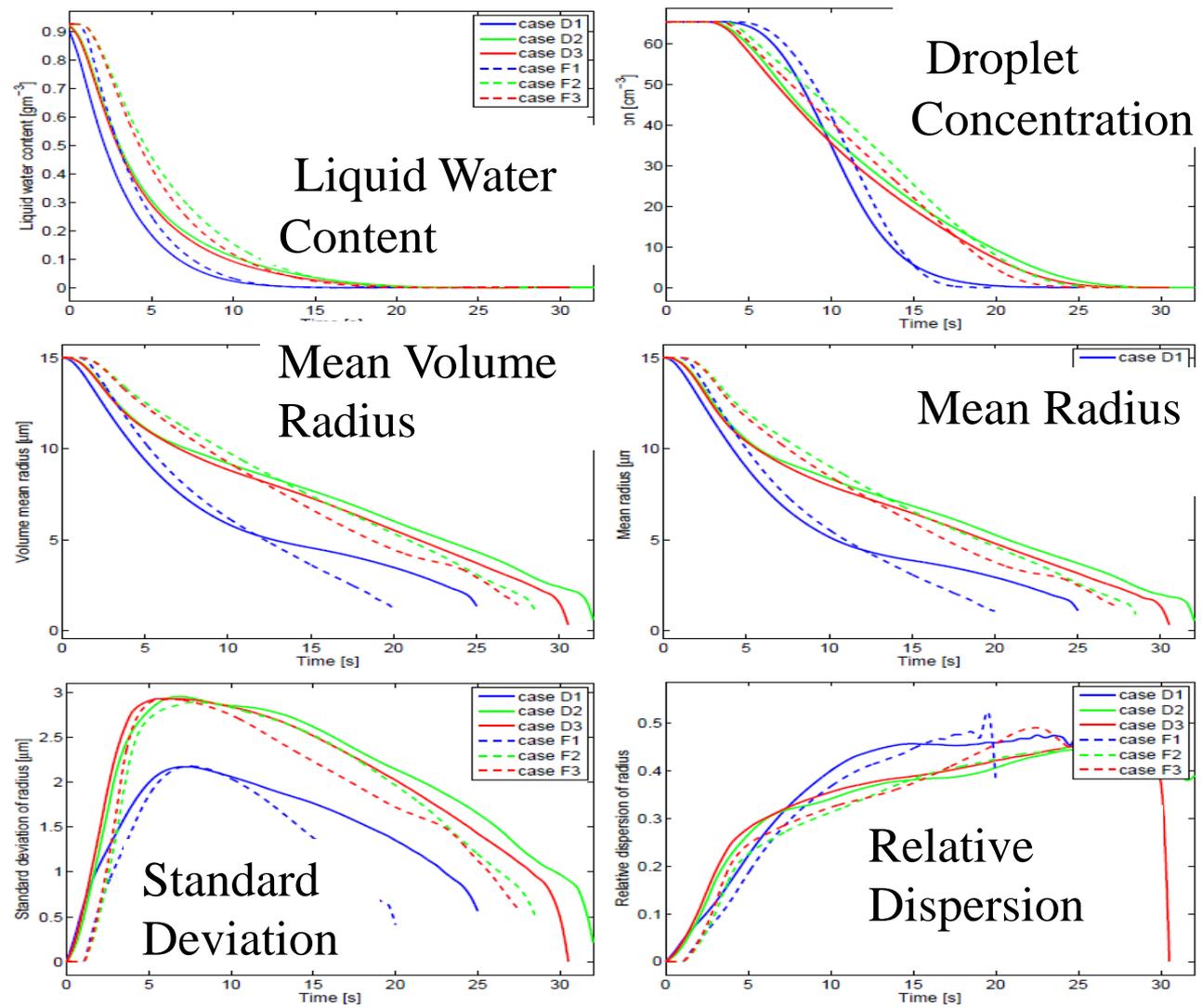
**Droplet Kinetics**

# Six Simulation Scenarios



**Two Turbulence Modes: Dissipating & Forced**

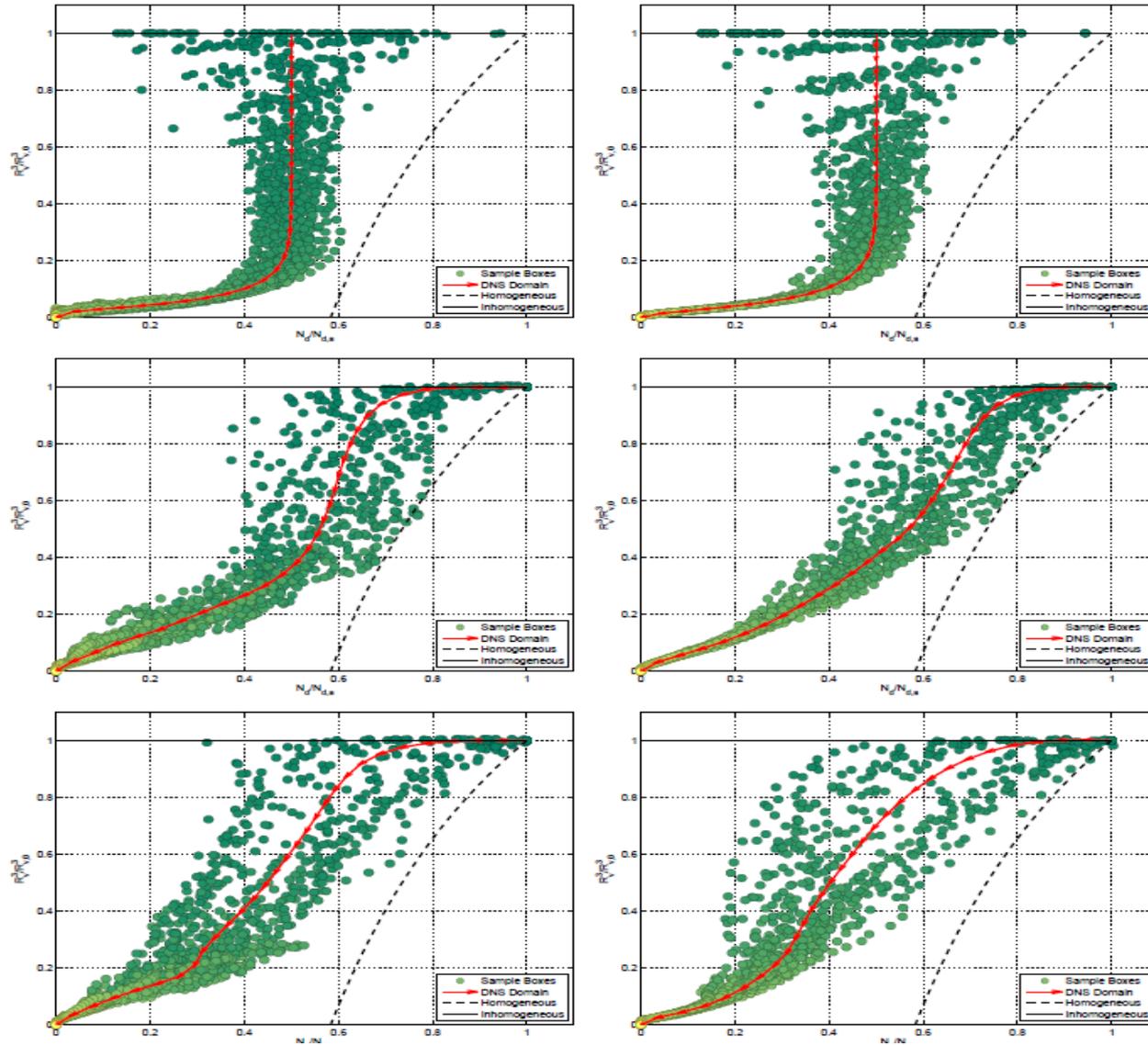
# Distinct Microphysical Properties for Different Scenarios at Different Times



Time (S)

# First Collapsing: Microphysical Mixing Diagram

Normalized Mean Droplet Volume



Normalized Droplet Concentration

# Unified Parameterization for Different Mixing Mechanisms

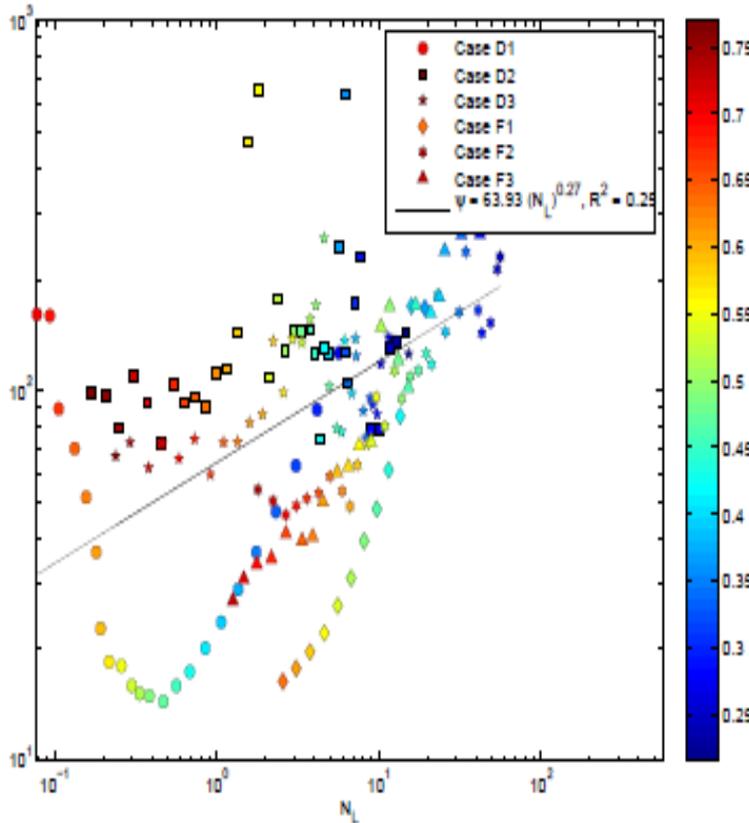
(Andrejczuk et al., JAS, 2009)

(Lu et al., JGR, 2013)

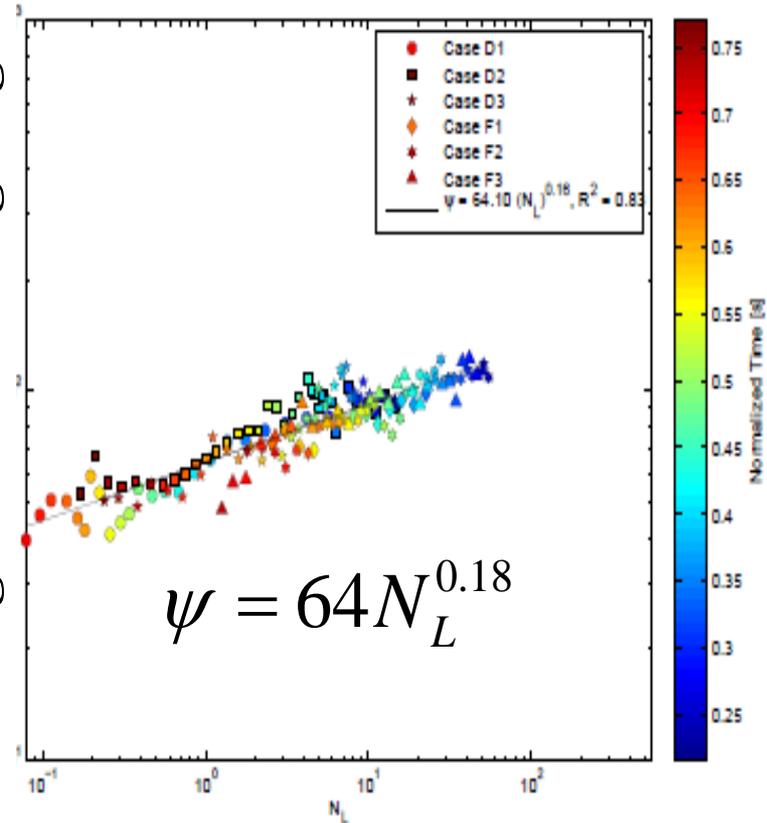
More Homogeneous Mixing



Slope Parameter



Homogeneous Mixing Degree

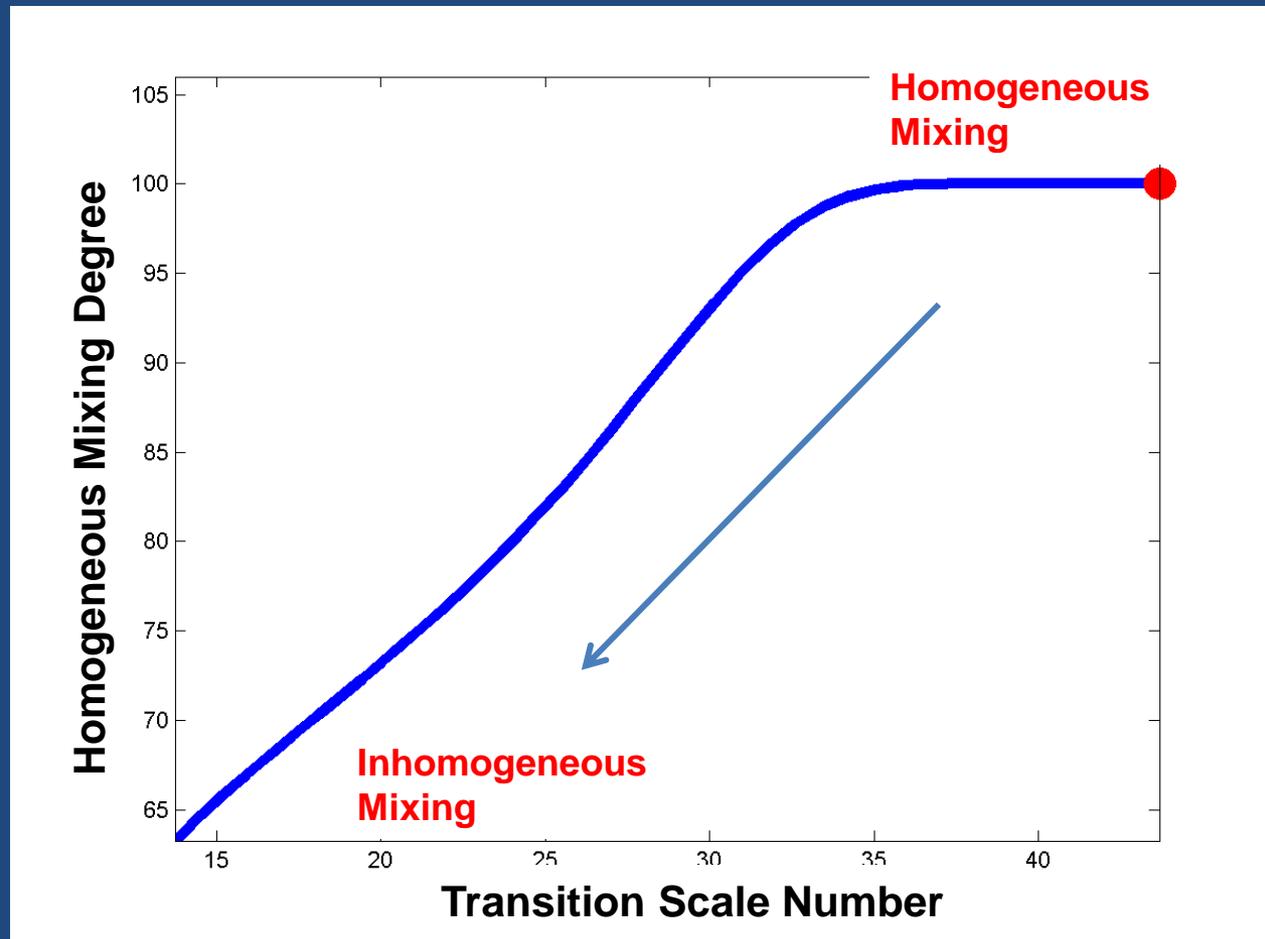


Transition Scale Number

Our measure of homogeneous mixing degree is clearly better than the previous slope parameter; the expression can be used to parameterize mixing types in two-moment schemes.

# Entrainment-Mixing Processes in P-DNS: Animation

- Different entrainment-mixing processes can occur in clouds and are key to rain initiation and aerosol-cloud interactions.
- Our knowledge on these processes is very limited.
- DNS can be used to fill in the knowledge gap and inform the development of related parameterization.



Droplets start with homogeneous mixing and evolve toward inhomogeneous mixing due to faster evaporation relative to turbulent mixing.

# Take-Home Messages

- Potentials of statistical physics (systems theory) as a theoretical foundation for microphysics parameterizations
- Potentials of unified parameterization for all turbulent entrainment-mixing processes
- Potentials of particle-resolved DNS to fill in the critical gaps between sub-LES and cloud microphysics
- Current is like the early days of classical physics when **kinetics, statistical physics, & thermodynamics** were established, full of challenges and opportunities:
  - Implement & test parameterization for entrainment-mixing processes
  - Consider relative dispersion (from two moment to three-moment scheme)
  - Small system, scale-dependence, and scale-aware parameterizations
  - Couple P-DNS with LES

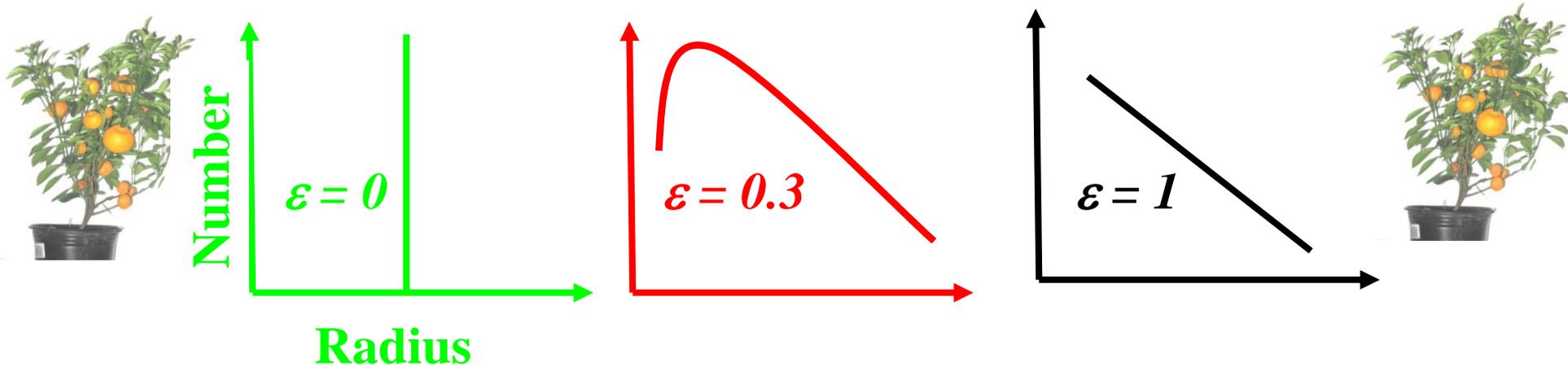
# Acknowledgment

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- **Collaborators: Chunsong Lu (NUIST), Zheng Gao (PhD student, SBU), Jingyi Chen (PhD student, SBU), Xin Zhou (PhD student, SBU), Bob McGraw (BNL), Pete Daum (BNL), John Hallett (DRI), ...**
- **Funding programs: DOE ARM, ASR, ESM and BNL LDRD**
- **Questions/comments/suggestions?**

**Thanks for your attention!**

# Long Ignored Quantity: Dispersion of Cloud Droplet Size Distribution



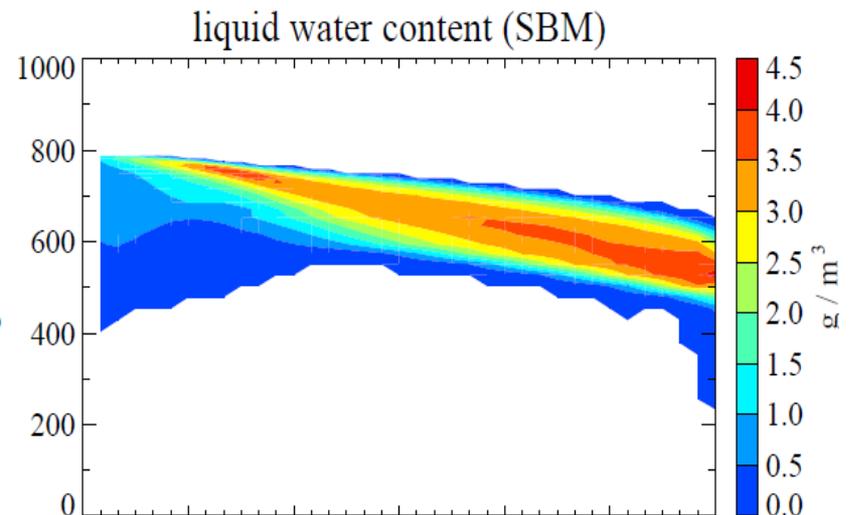
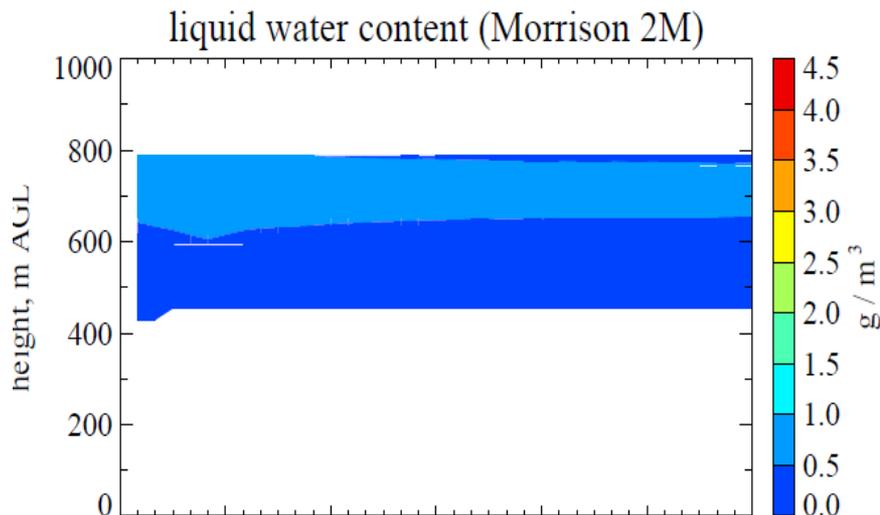
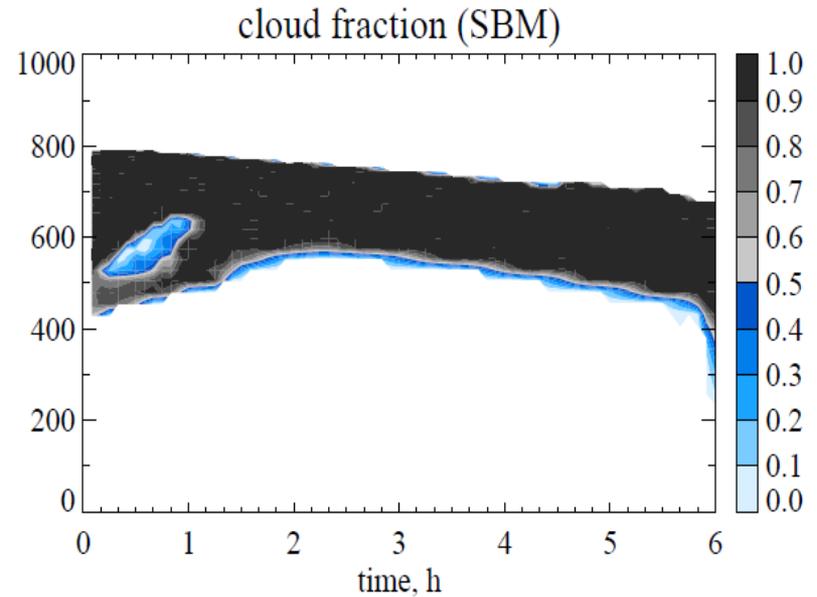
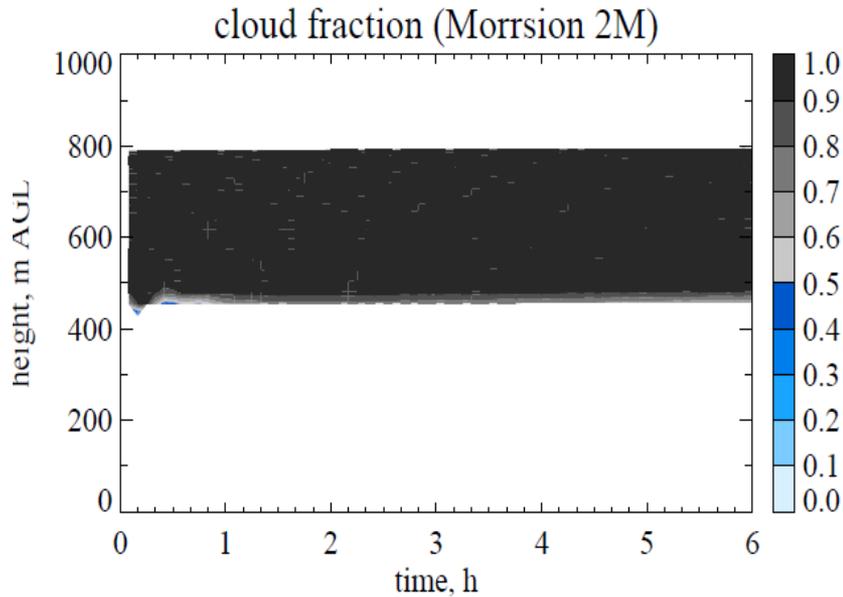
Dispersion  $\varepsilon$  is the ratio of standard deviation to the mean radius of droplet sizes, which measures the spread of droplet sizes.

Dispersion increases from **left** to right in above figures.

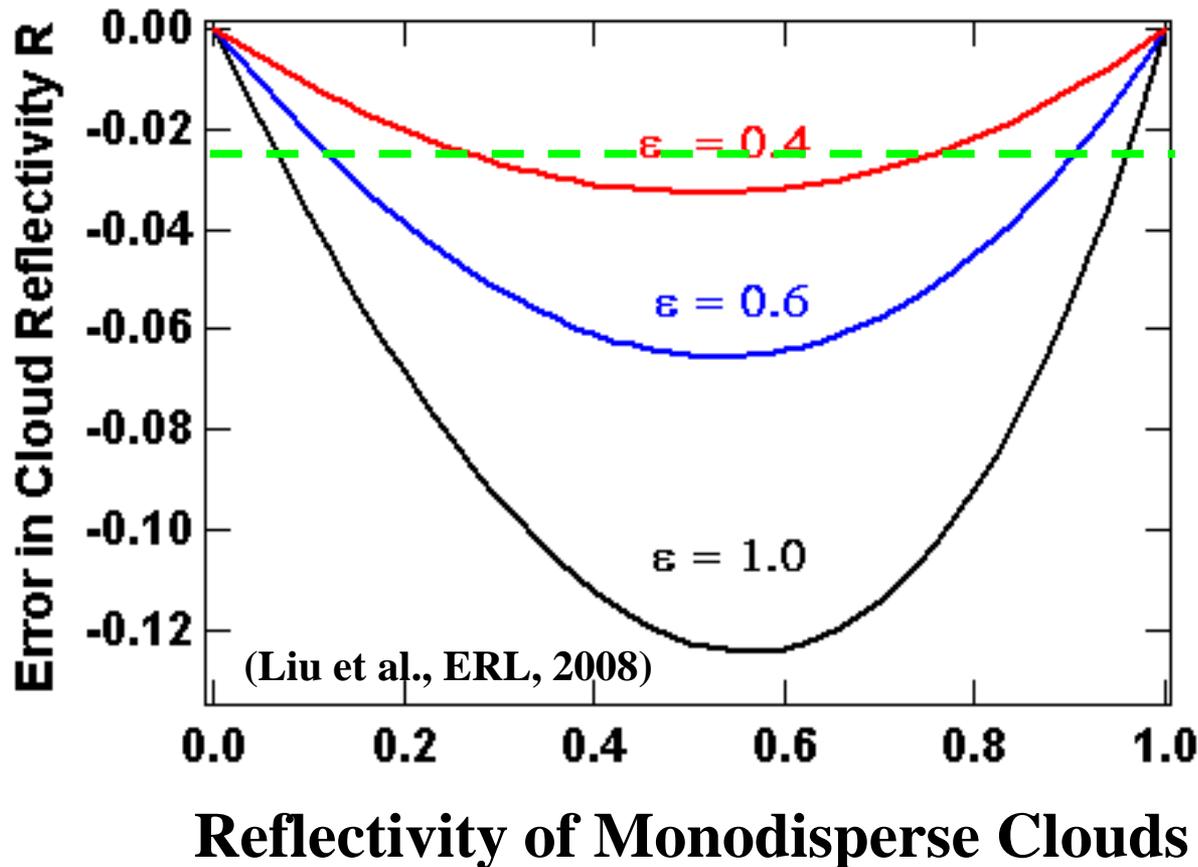
The three size distributions have the same L and N.

The necessity to consider the spectral shape in atmospheric models is bringing progress of atmospheric models to the core of cloud physics, converging with weather modification!

# Effect of Spectral Shape: Two Moment vs. SBM



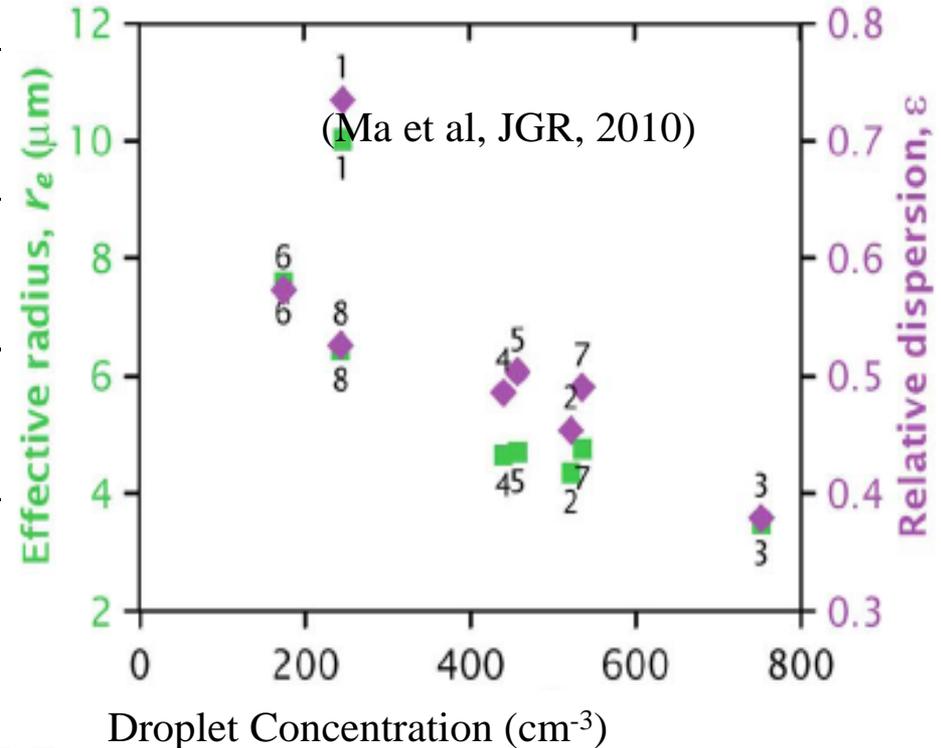
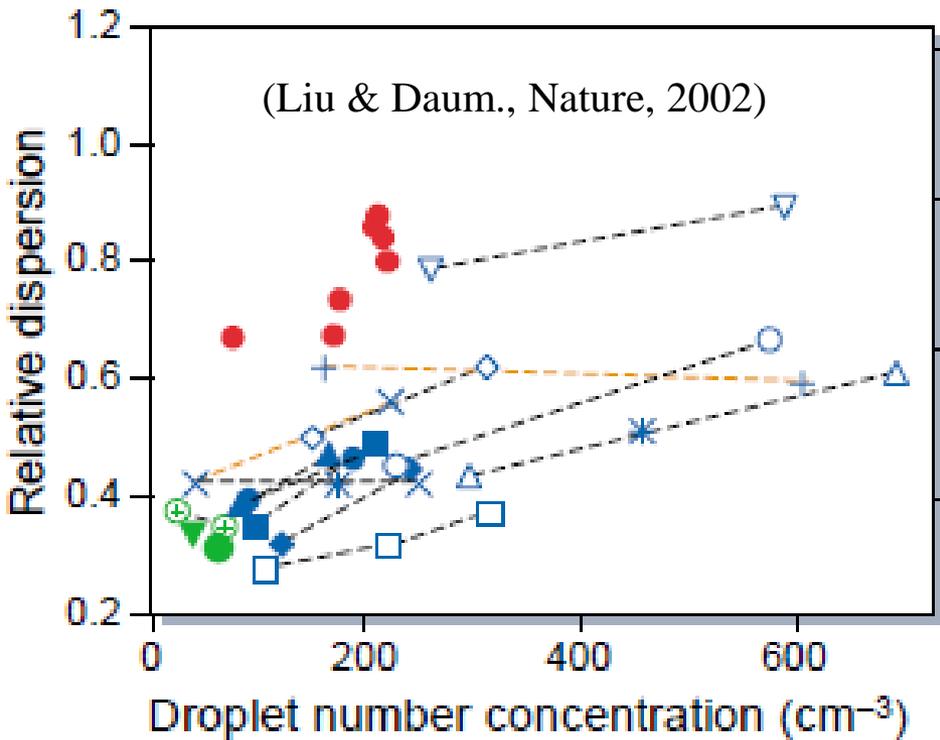
# Neglect of dispersion significantly overestimates cloud reflectivity



*Green dashed line indicates the reflectivity error where overestimated cooling is comparable to the warming by doubling CO2.*

**Neglecting dispersion can cause errors in cloud reflectivity, which further cause errors in temperature etc. Dispersion may be a reason for overestimating cloud cooling effects by climate models.**

# Conflicting Results since 2002



**Aerosol Increase**

## Warming dispersion effect:

(Lu et al, JGR, 2007; Chen et al, ACP, 2012; Pandithurai et al, JGR, 2012; Kumar et al ACP, 2016)

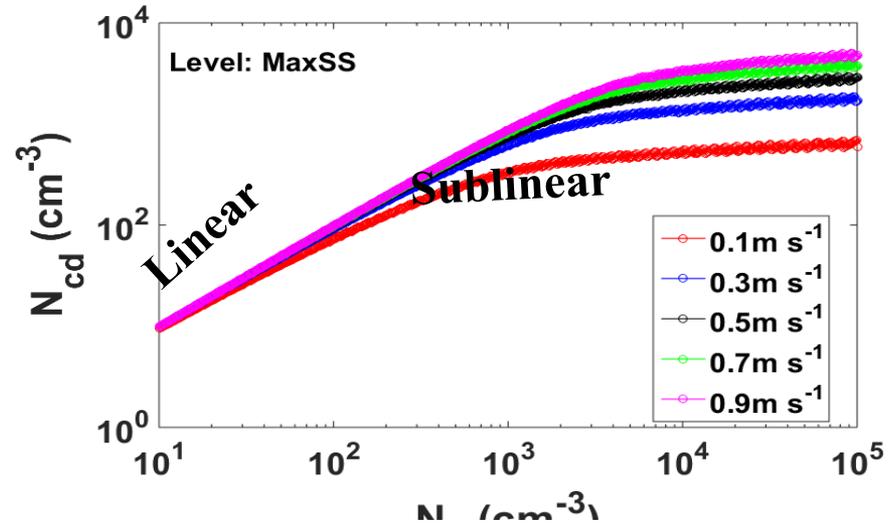
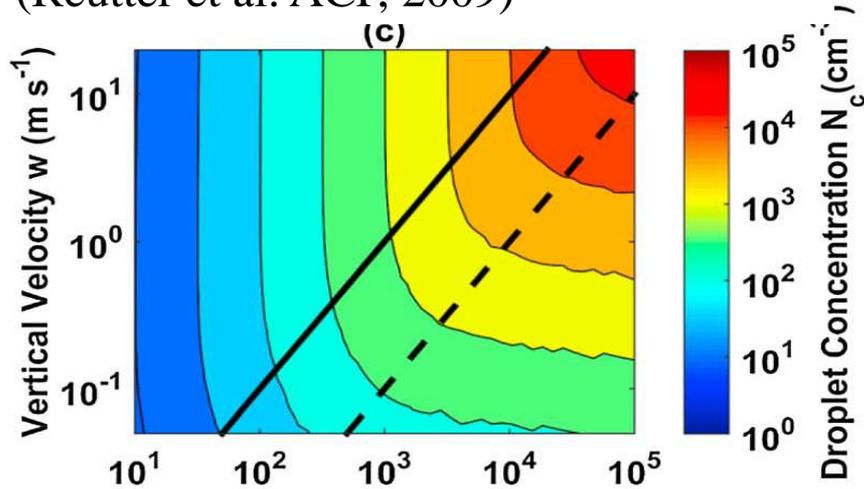
## Cooling Dispersion Effect:

(Martins et al, ERL, 2009; Hudson et al, JGR, 2012)

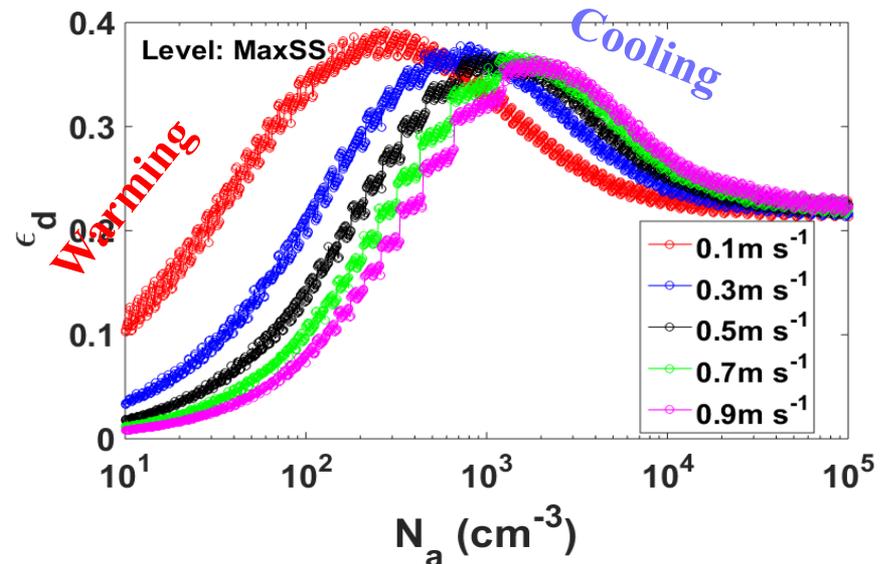
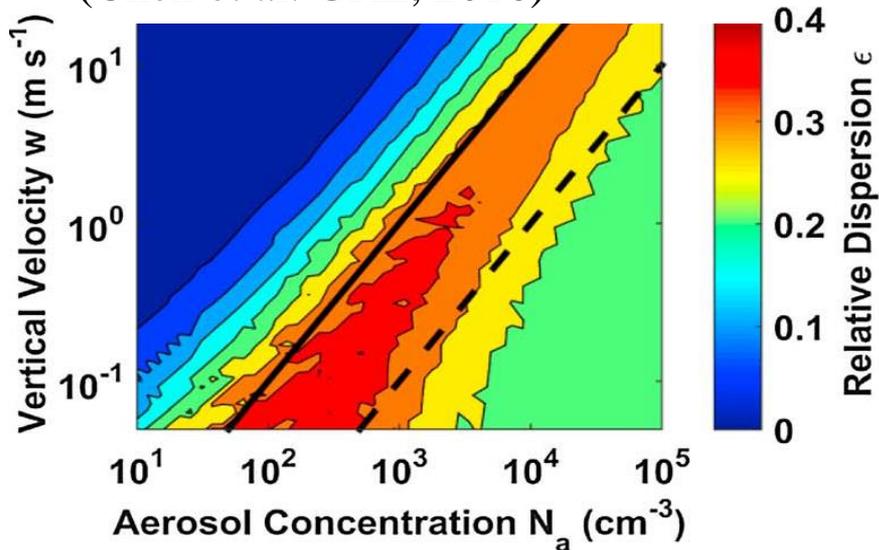
**These conflicting results suggest that dispersion effect exhibits behavior of different regimes, like number effect?**

# AIE Regime Dependence

(Reutter et al. ACP, 2009)

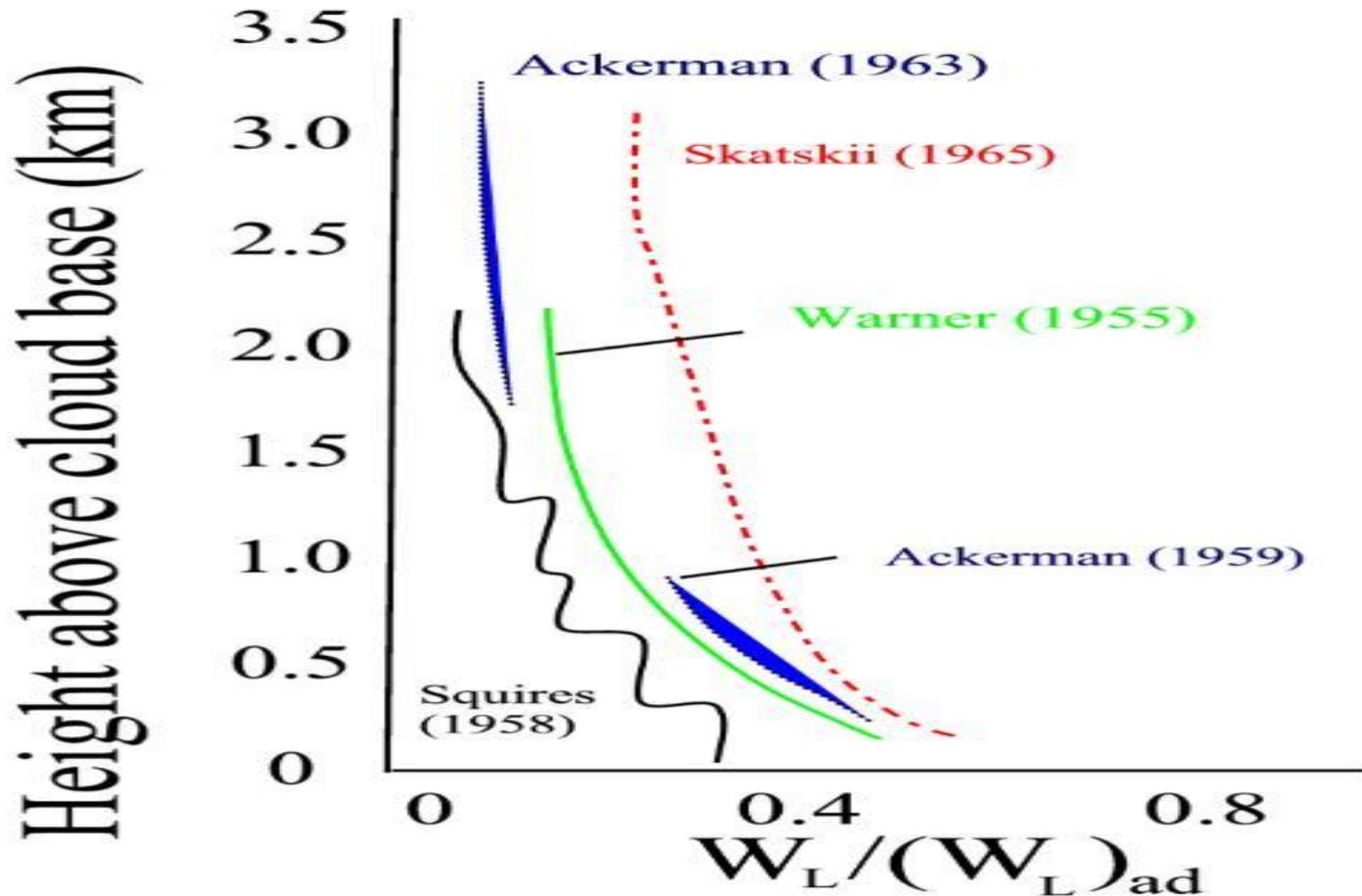


(Chen et al. GRL, 2016)



**Dispersion effect exhibits stronger regime dependence  
& works to “buffer” number effect!**

# Subadiabatic LWC Profile-Entrainment



**This figure shows that the ratio of the observed liquid water content to the adiabatic value decreases with height above cloud base, and less than 1 (adapted from Warner 1970, J. Atmos. Sci.)**

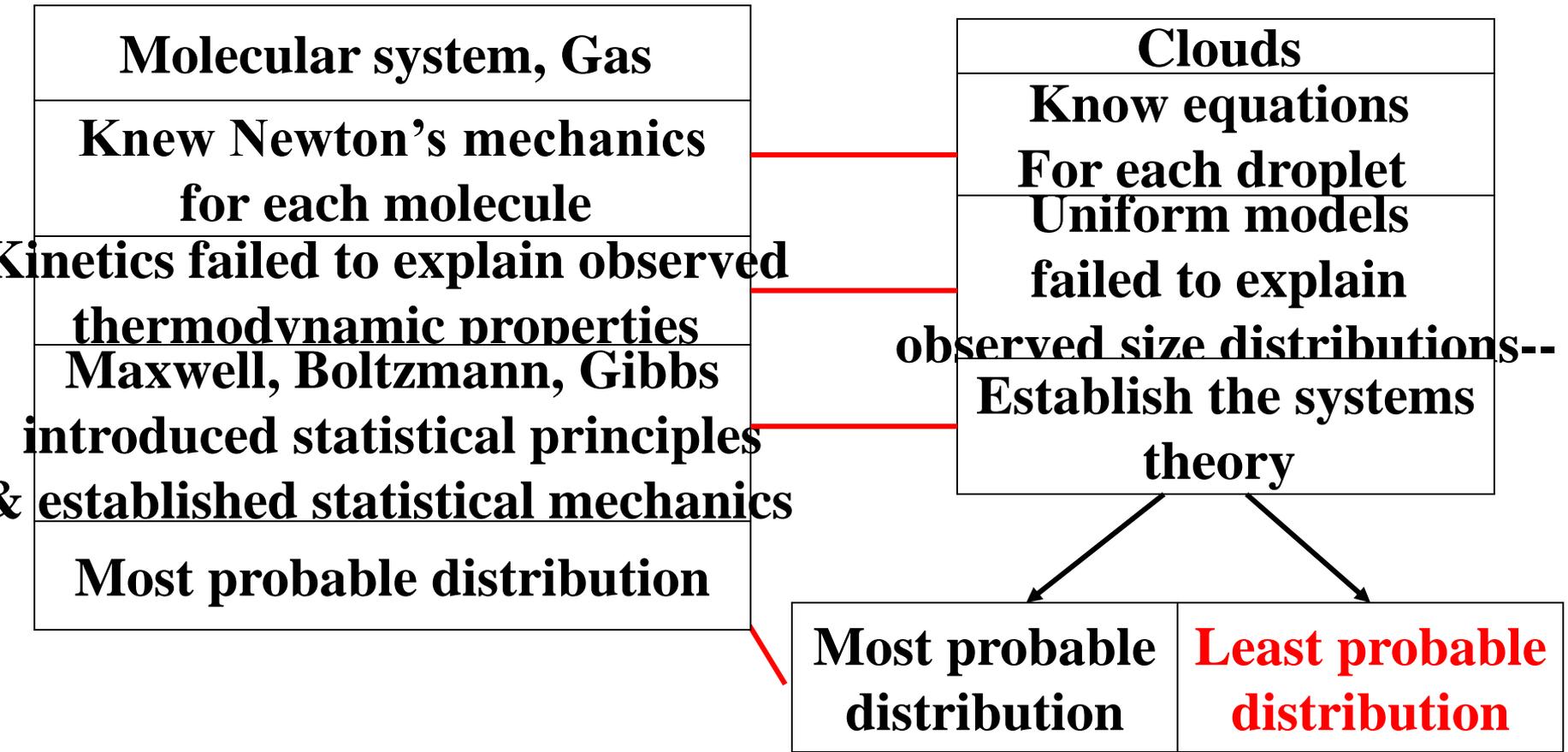
# Remaining Issues and Challenges

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- How to determine the parameters  $a$  and  $b$  in the power-law relationship  $\mathbf{x} = \mathbf{a}r^b$
- Establish a kinetic theory for droplet size distribution (stochastic condensation, Ito calculus, Langevin equation, Fokker-Planck equation).
- How to connect with dynamics?
- A grand unification with molecular systems?
- Application to developing unified and scale-aware parameterizations

# Difference of Droplet System with Molecular System

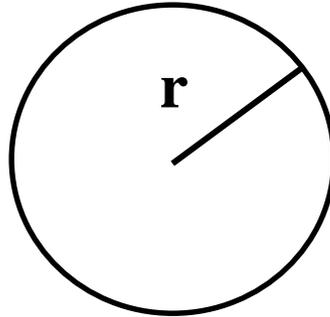
Big system vs. small system  
(Liu et al, JAS, 1998, 2002)



# Gibbs Energy for Single Droplet

$$V = \frac{4\pi}{3} r^3$$

$$A = 4\pi r^2$$



$\sigma$  = surface energy

$\rho_w$  = water density

$L$  = latent heat

The increase of the Gibbs free energy to form this droplet is

$$g = \left( 4\pi\sigma r^2 - 4\pi\sigma_c r_c^2 \right) - \frac{4\pi\rho_w L}{3} r^3$$

$$= c_1 r^3 + c_2 r^2 + c_3$$

$L$  – latent heat

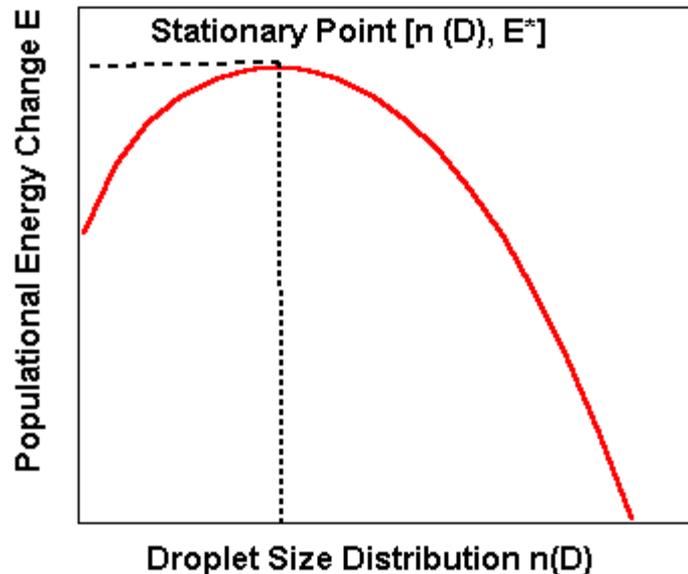
# Populational Gibbs Free Energy Change

To form a droplet population, Gibbs free energy change is

$$\begin{aligned} G &= \int g(\mathbf{r}) n(\mathbf{r}) d\mathbf{r} \\ &= c_1 \int r^3 n(\mathbf{r}) d\mathbf{r} + c_2 \int r^2 d\mathbf{r} + c_3 \end{aligned}$$

The larger the  $G$  value, the more difficult to form the droplet system. Therefore, the size distribution corresponds to the maximum populational Gibbs free energy subject to the constraints is the minimum likelihood size distribution (MNSD).

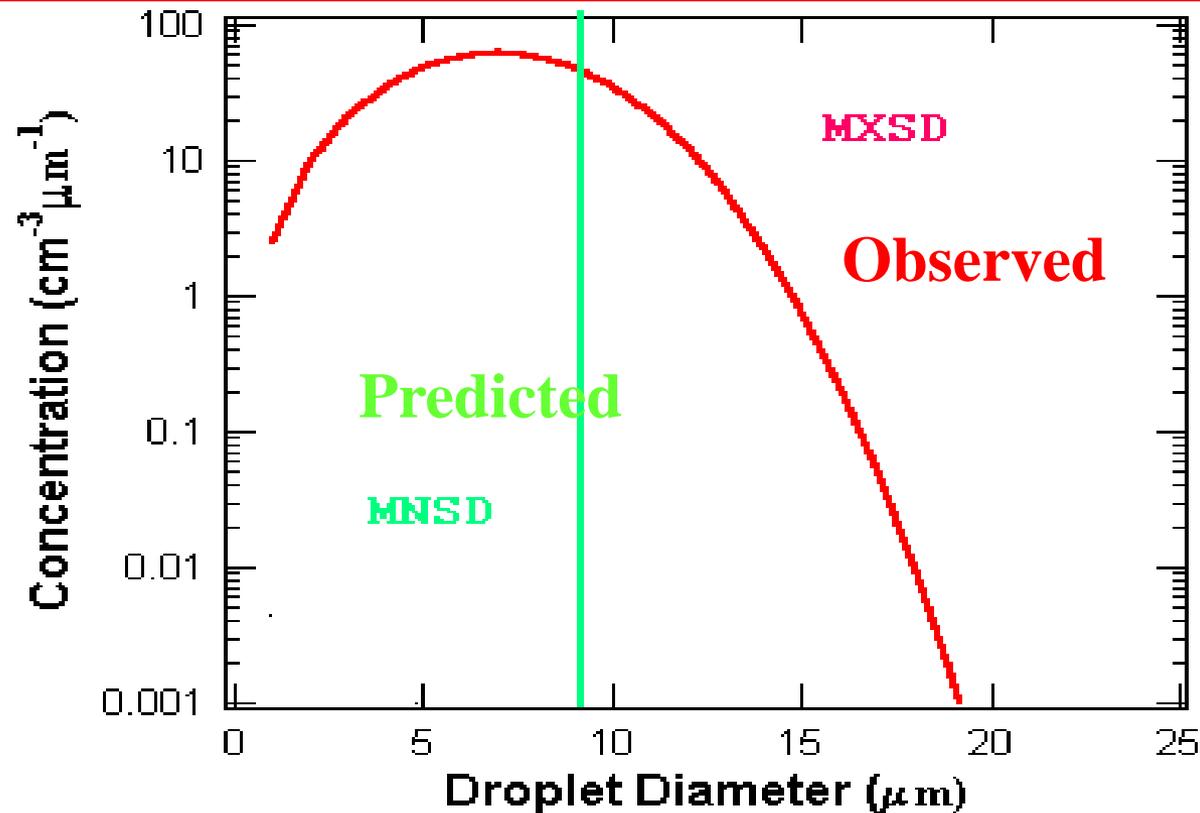
# Least Probable Size Distribution



The larger the  $G$  value, the more difficult to form the droplet system. Therefore, the size distribution corresponds to the maximum populational Gibbs free energy subject to the constraints is the least probable size distribution given by

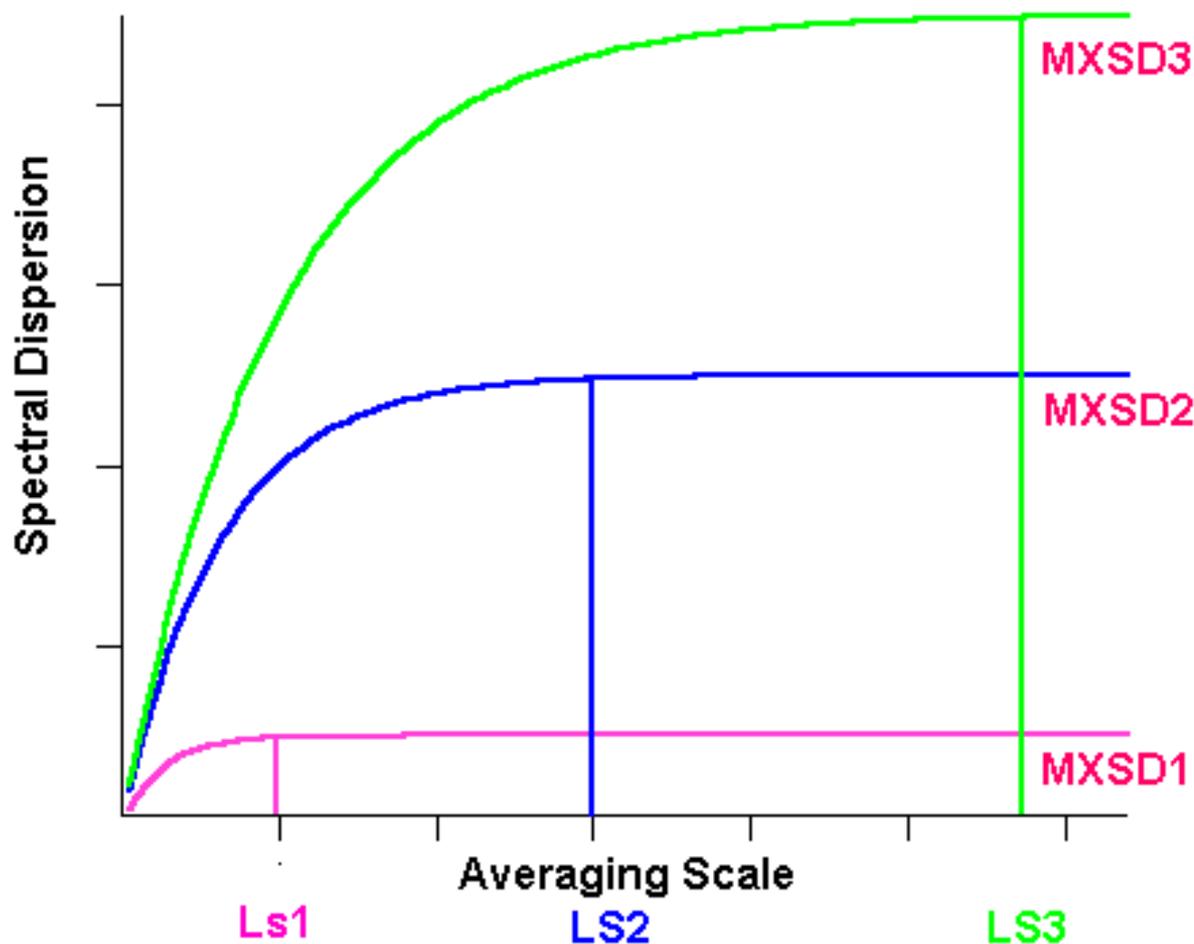
$$\mathbf{n}_{\min}(\mathbf{r}) = \mathbf{N}\delta(\mathbf{r} - \mathbf{r}_0)$$

# MXSD, MNSD and Further Understanding of Spectral Broadening



**Observed droplet size distribution corresponds the MXSD;  
the monodisperse distribution predicted by the uniform condensation  
model corresponds to the MNSD, seldom observed!  
Observed and uniform theory predicted are two totally different  
characteristic distributions!**

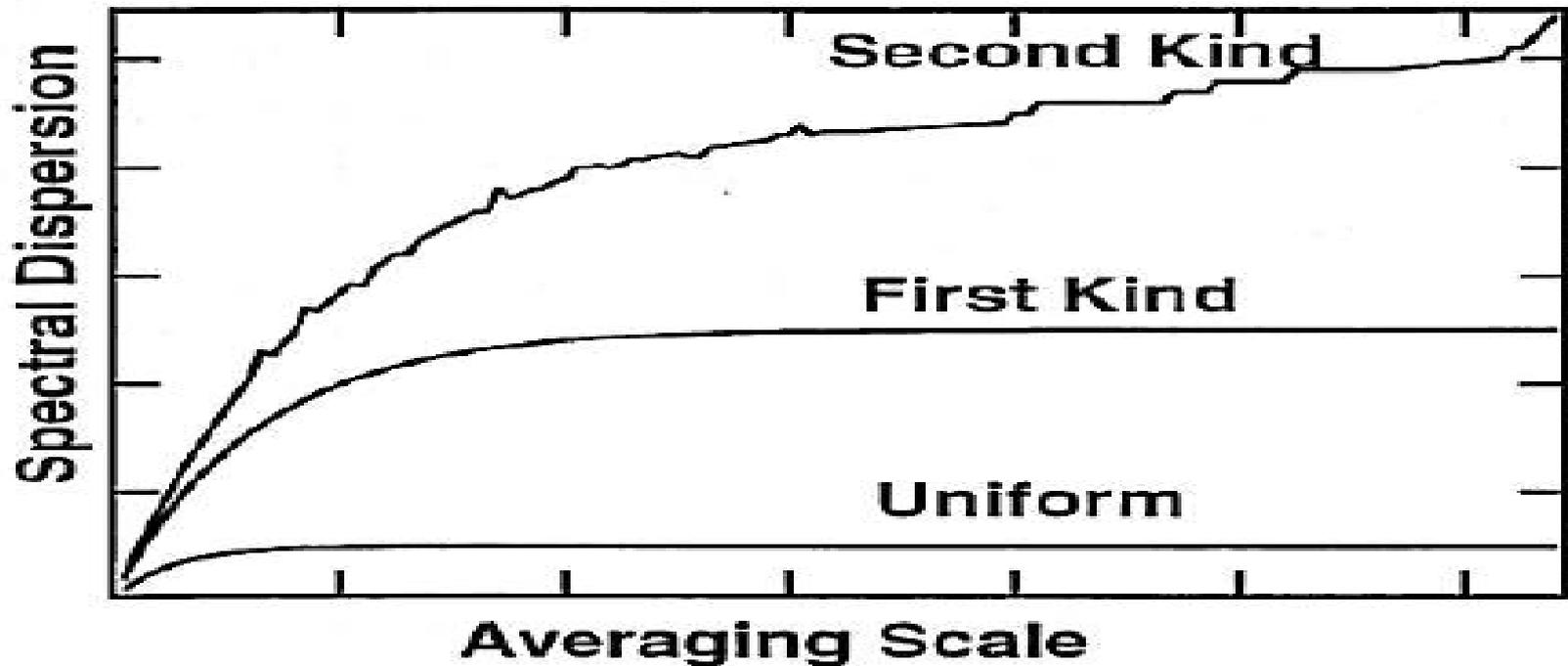
# Scale-Dependence of Size Distribution



- **Fluctuations increases from level 1 to 3.**
- **Saturation scale  $L_s$  is defined as the averaging scale beyond which distributions do not change.**
- **Distributions are scale-dependent and ill-defined if averaging scale  $< L_s$ .**

Diagram shows the dependence of size distributions (observed or simulated) on the averaging scale

# More Scale-Dependence of Size Distribution



**Figure 5.** A diagram illustrating the scale-dependence of droplet size distributions. Both axes are only qualitative. The bottom curve represents the simplest case of uniform clouds. The middle and top curves represent the scale-dependence of the first and second kind, respectively.

(Liu et al., 2002, Res Dev. Geophys)

# Entropy and Disorder



*"I blame entropy."*