Aerosol Indirect Effects, Buffering Mechanisms, and Connections to Small-Scale Dynamics

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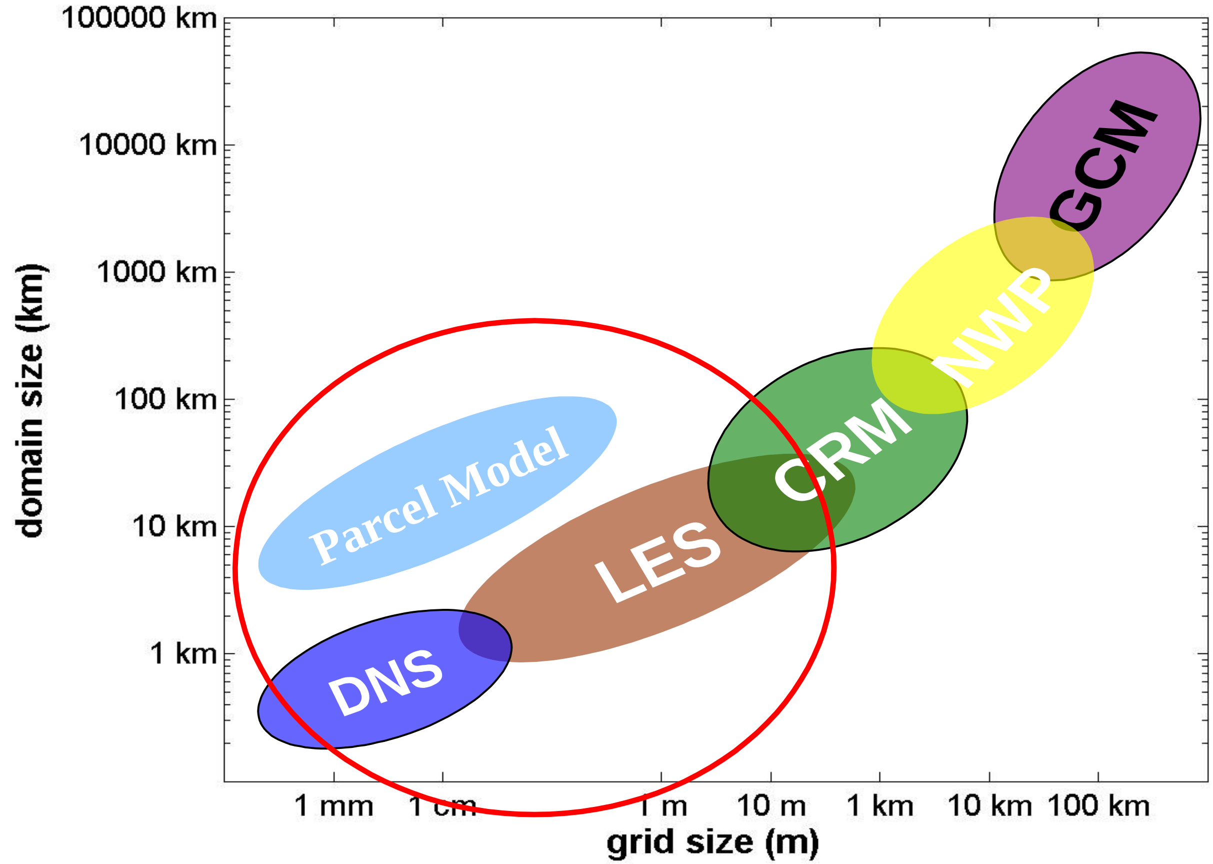
Outline

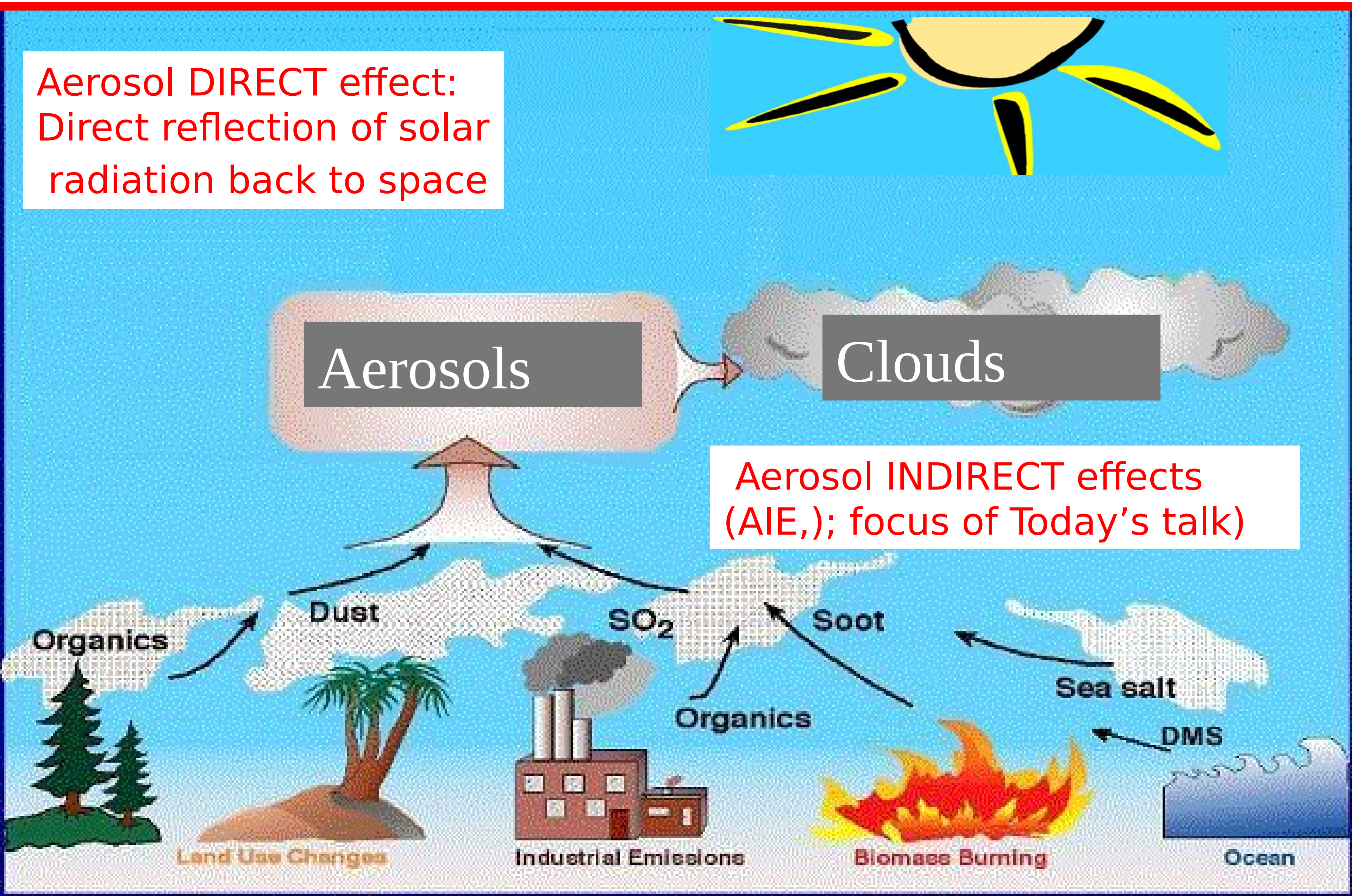
- **Background**
- **Dispersion Effect and ACI Regime Dependence**
- **Turbulent Entrainment-Mixing Process**
- **Particle-Resolved DNS**
- **Systems Theory for Microphysics Parameterization**
- **Take-Home Messages**

BNL-CMP use model hierarchy to address complex multiscale phenomena

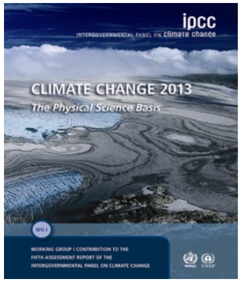


Except particle-resolved DNS, microphysics is parameterized with different sophistications, e.g., single moment (L), double moment (L, N), three moment (L, N, dispersion), …, bin microphysics.

Aerosol Effects on Climate



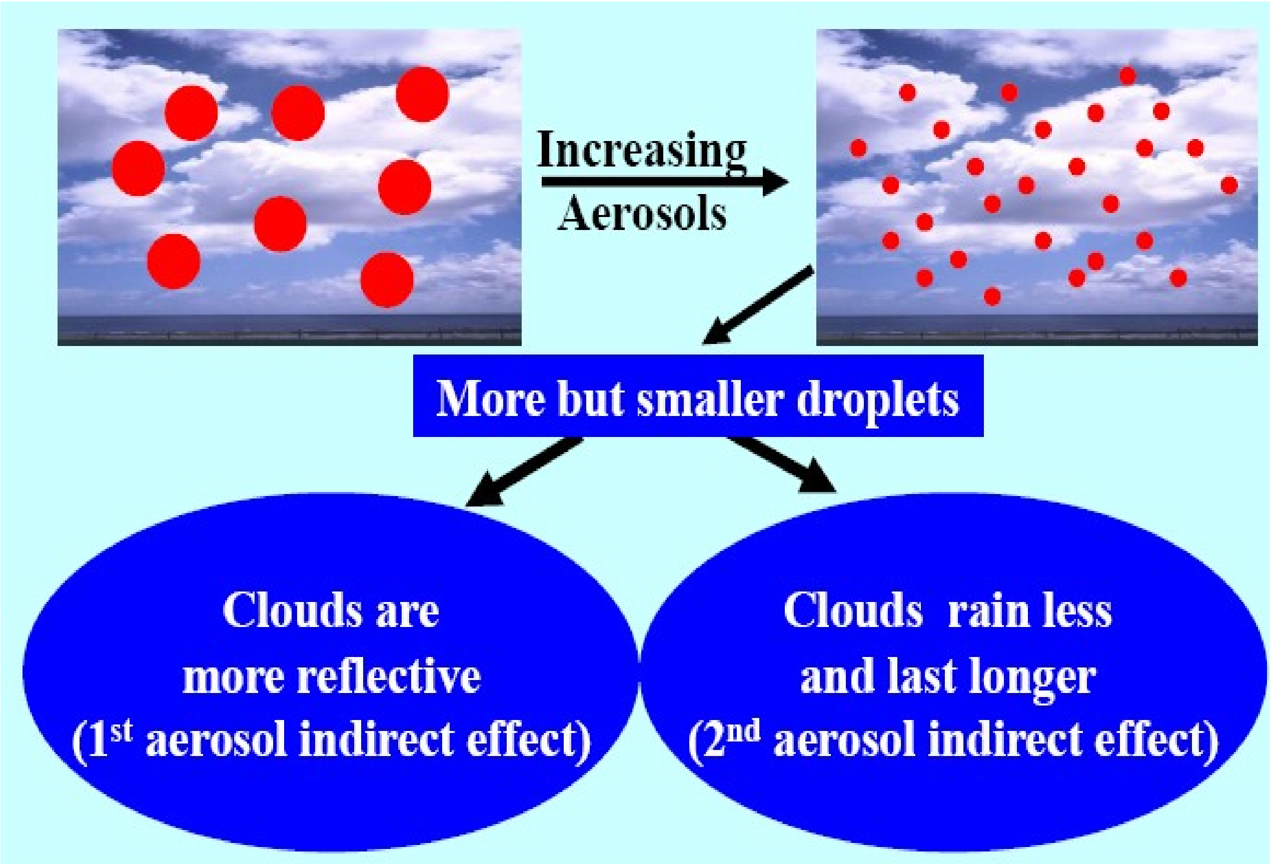
Science Drivers from IPCC-AR5



- **There is high confidence that aerosols and their interactions with clouds have offset a substantial portion of global mean forcing from well-mixed greenhouse gases. They continue to contribute the largest uncertainty to the total RF estimates.**
- **Ambient clouds seem less sensitive to aerosol perturbation than clouds in climate models, which do not represent well or mot represent at all buffering/compensating processes:**
  - **Dispersion effect**
  - **Non-monotonic dependence (regime dependence)**
  - **Turbulent Entrainment-mixing processes**
  - **Process couplings**

<u>Dominant View of AIE</u>: *Number Effect*

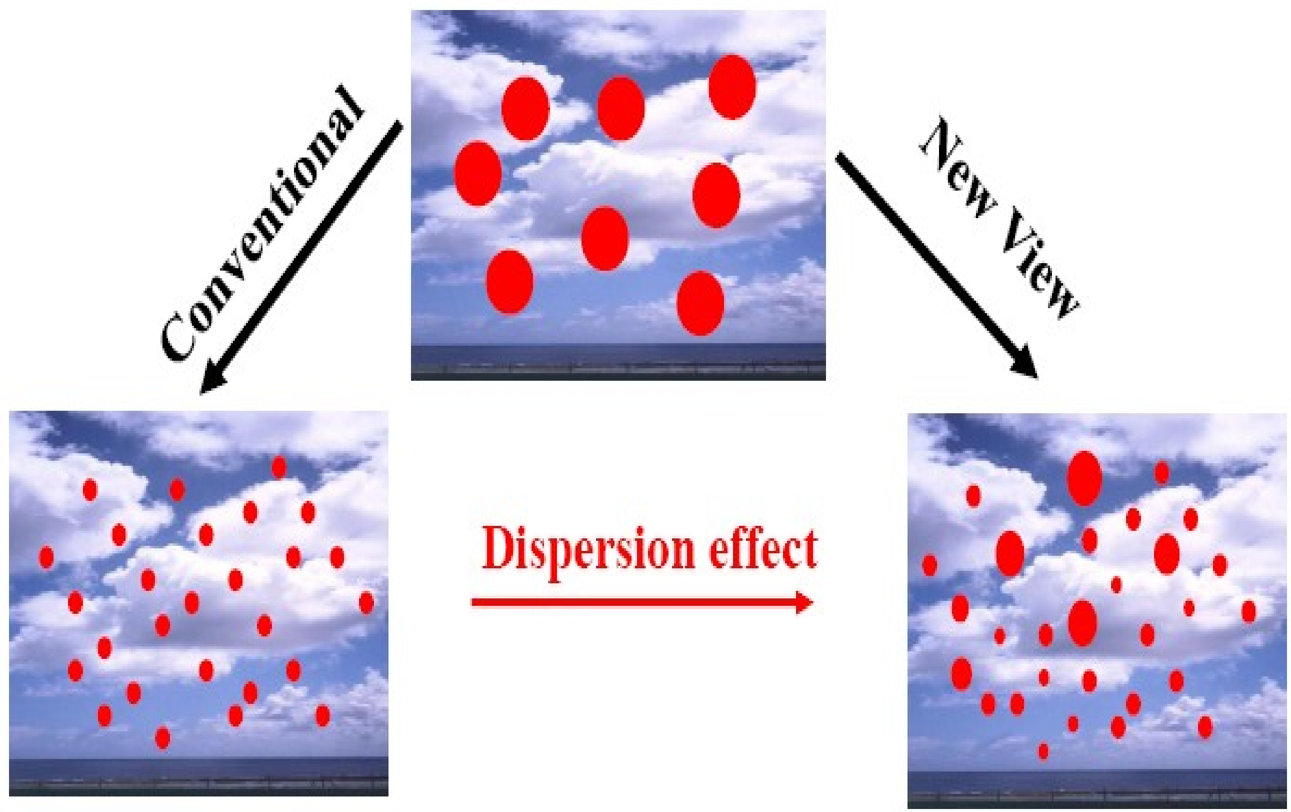
Twomey (1974, Atmos. Environ): "it is suggested that pollution gives rise to whiter (not darker) clouds -- by <u>increasing the droplet concentrations</u> and thereby the optical thickness (and cloud albedo) of clouds."



GCM estimates are full of uncertainties & tend to overestimate AIE cooling compared to obs. ➡ **Unrealistic assumptions and closely related buffering processes:** *Dispersion effect; Regime dependence; Entrainment-mixing processes; Couplings*

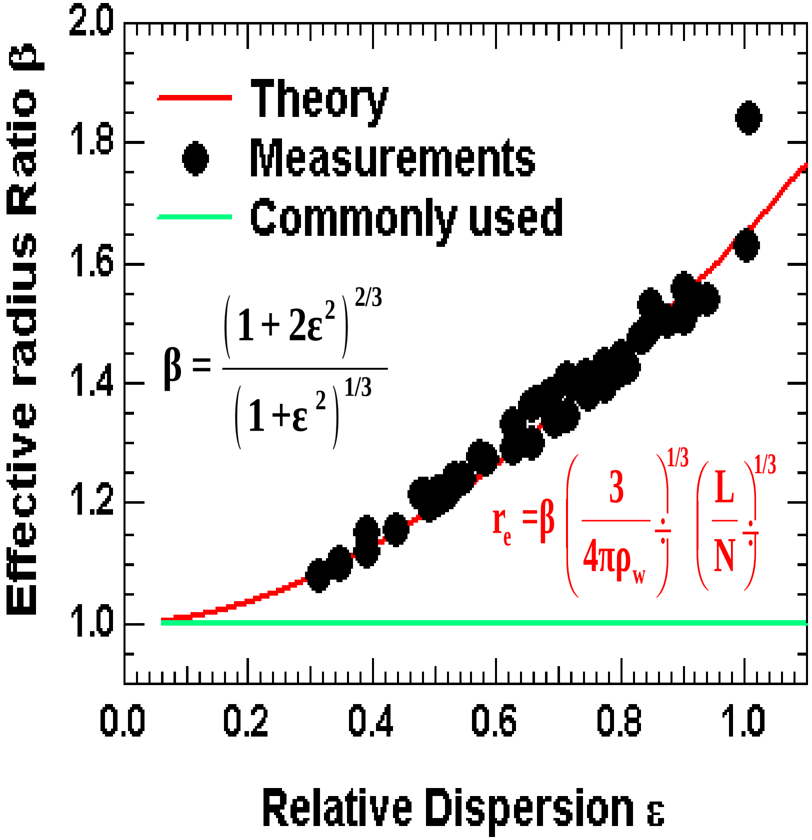
Modified View of AIE: *Dispersion Effect*

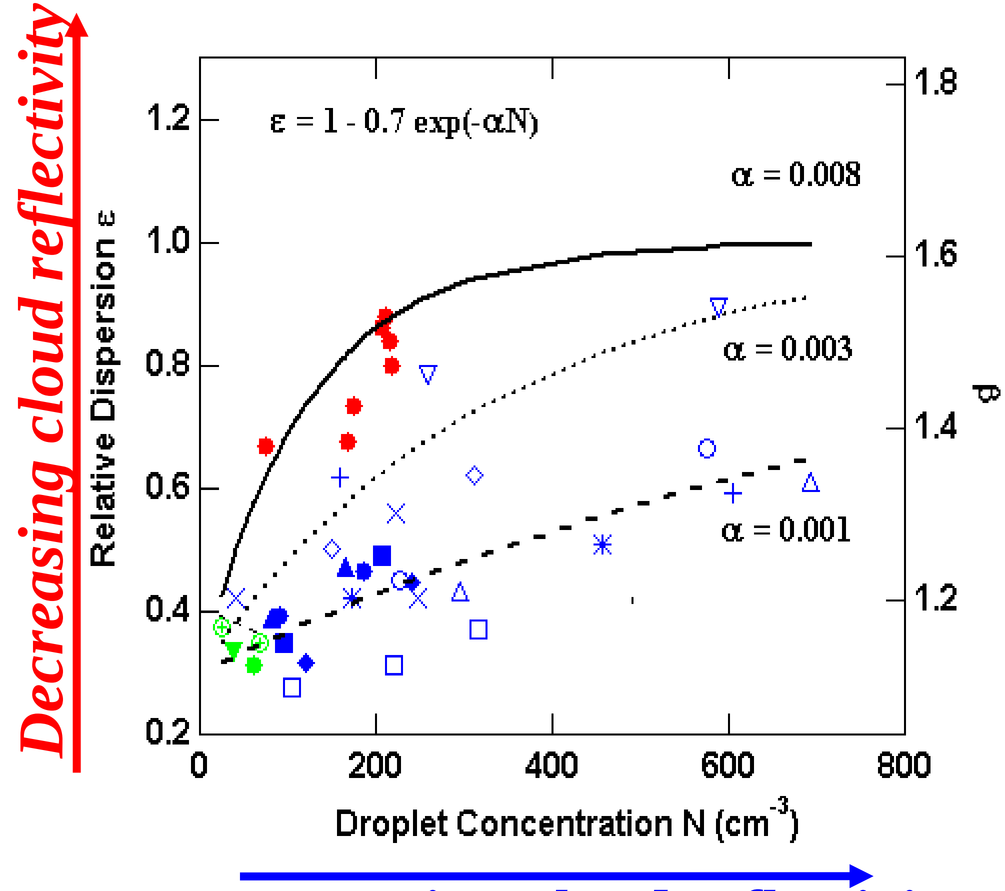
Liu and Daum (2002, Nature): "Anthropogenic aerosols exert an additional effect on cloud properties that is derived from changes in [the spectral shape of the size distribution of cloud droplets]in polluted air and acts to diminish the cooling of number effect by 10-80%.



AIE = Number Effect + Dispersion Effect

Warming Dispersion Effect





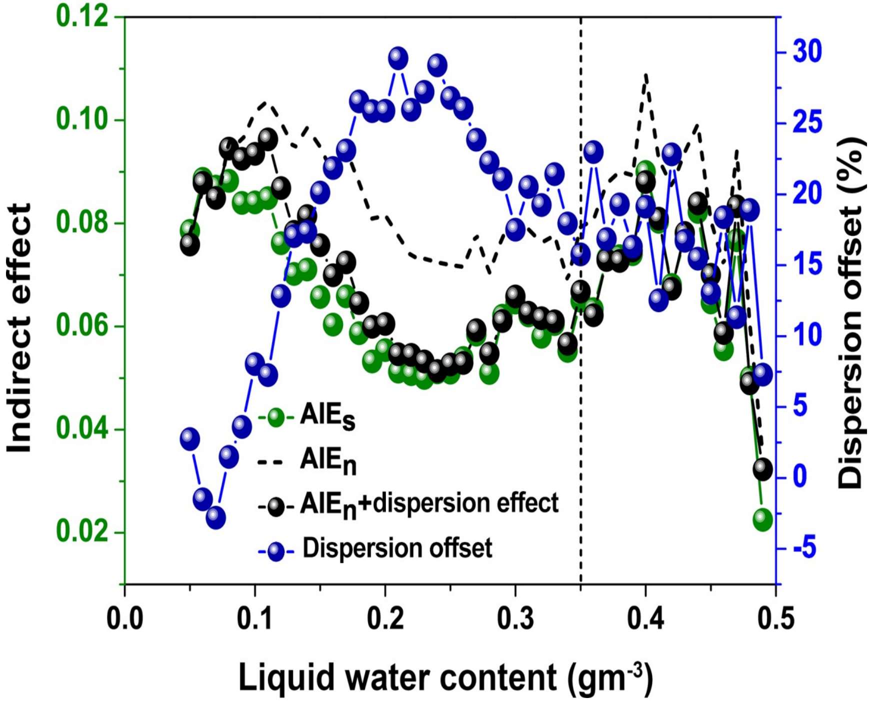
(ε = Standard Deviation/Mean Radius)

Increasing cloud reflectivity

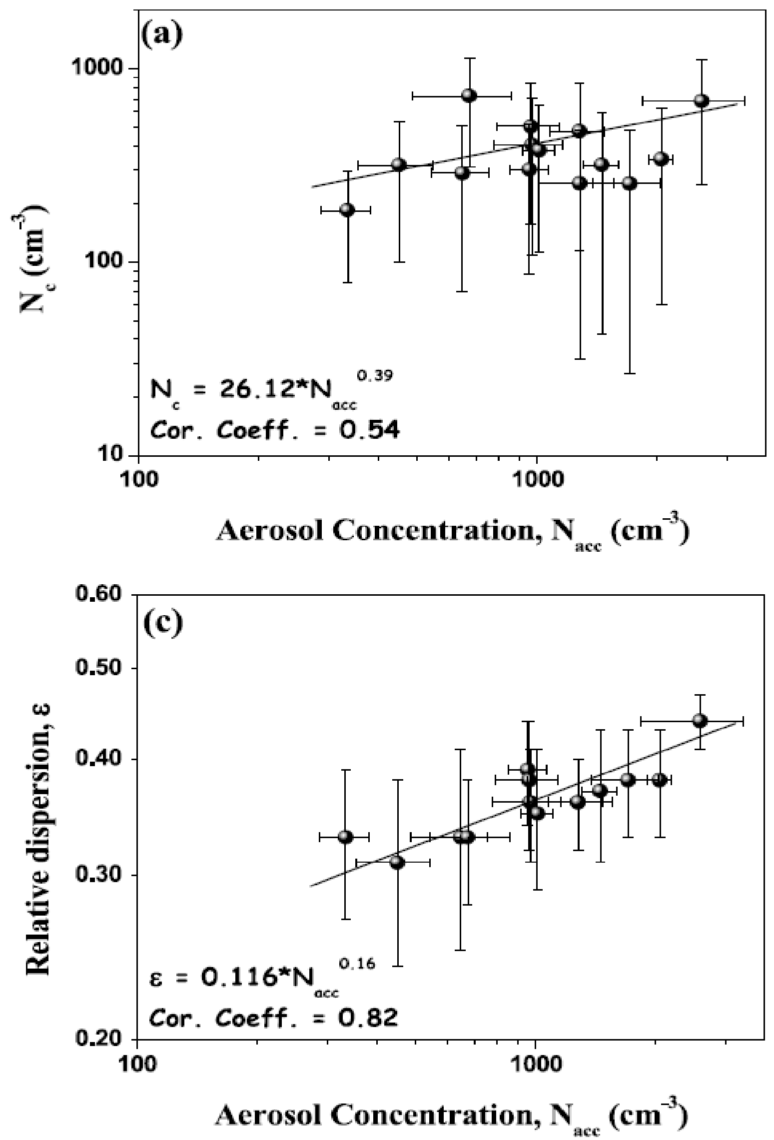
The parameter β is an increasing function of droplet relative dispersion ε, not a constant as implicitly assumed in the Twomey effect; furthermore, increasing aerosol enhances not just droplet concentrations, but also ε (hence β) (Liu & Daum 2000, GRL; 2002, Nature, Peng & Lohmann 2003, GRL; Liu et al. 2006; Lu et al. 2007).

Wonderful Observations in India

(Kumar et al., ACP, 2016)



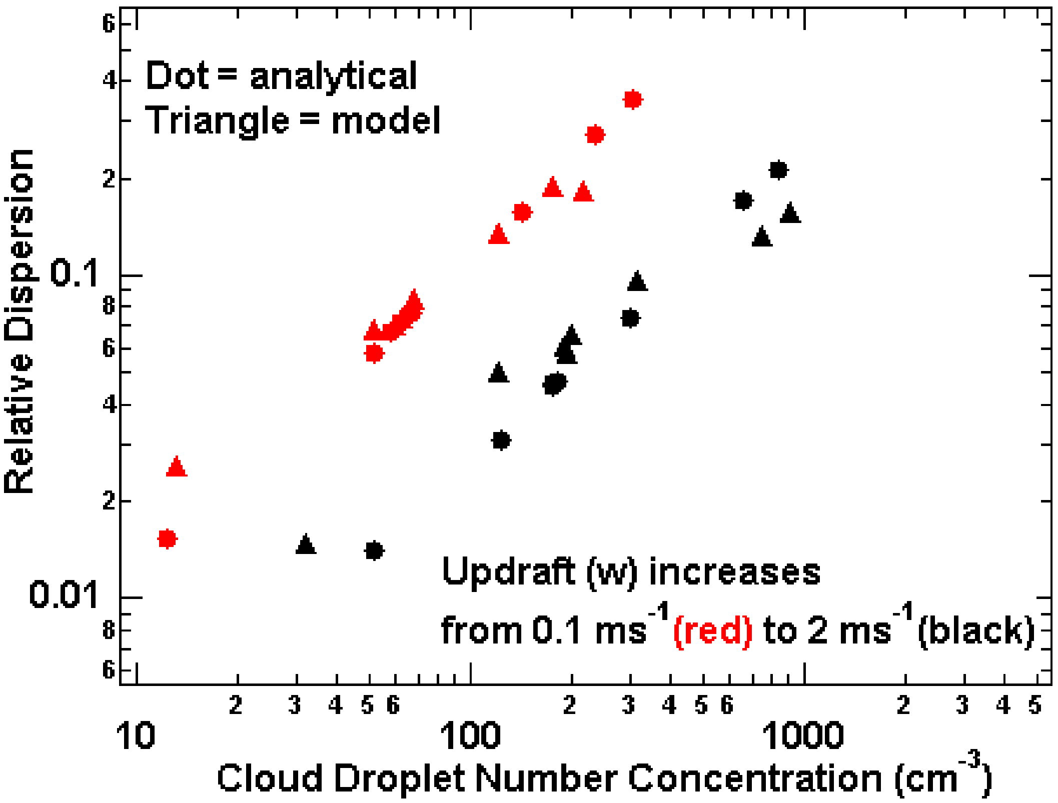
(Pandithurai et al., JGR 2012)



- Right: Aircraft measurements during Cloud Aerosol Interaction and Precipitation Enhancement EXperiment (CAIPEEX)
- Left: Ground-based measurements

Theoretical Expression for Dispersion

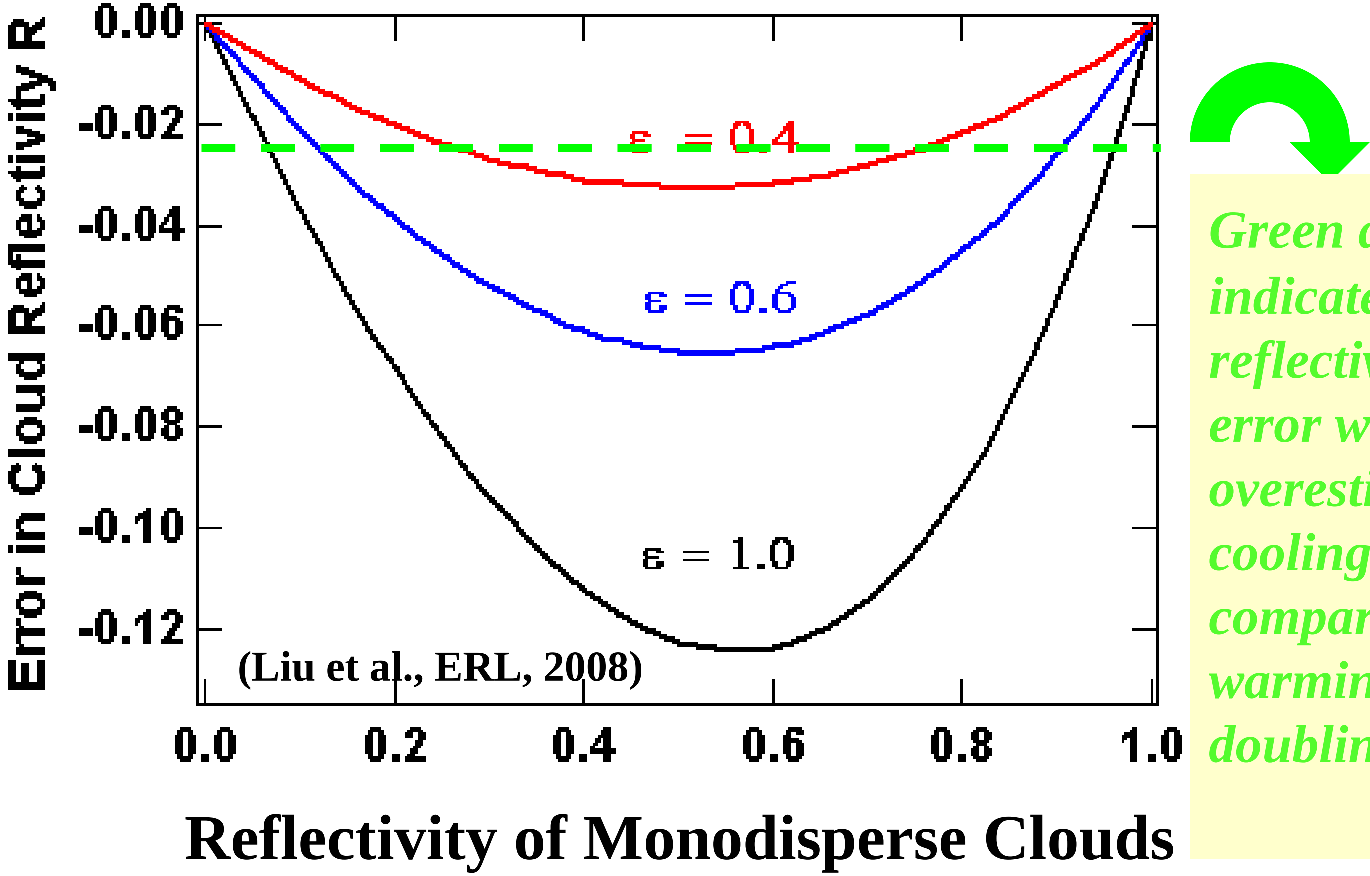
- **Generalized activation scheme considering droplet concentration & relative dispersion**
- **Analytical & use the same inputs as common schemes for droplet concentration**
- **Compares well with parcel model simulations**



(Liu et al. GRL, 2006)

Dispersion increases with increasing aerosols or decreasing updraft velocity due to competition for available water vapor.

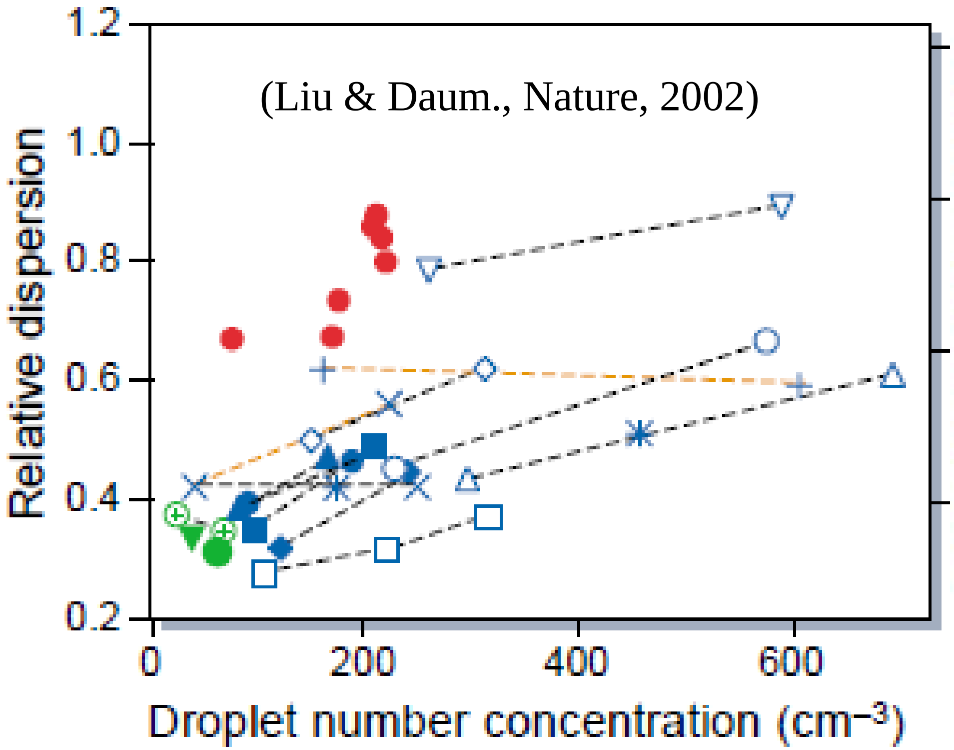
Neglecting dispersion significantly overestimates cloud reflectivity

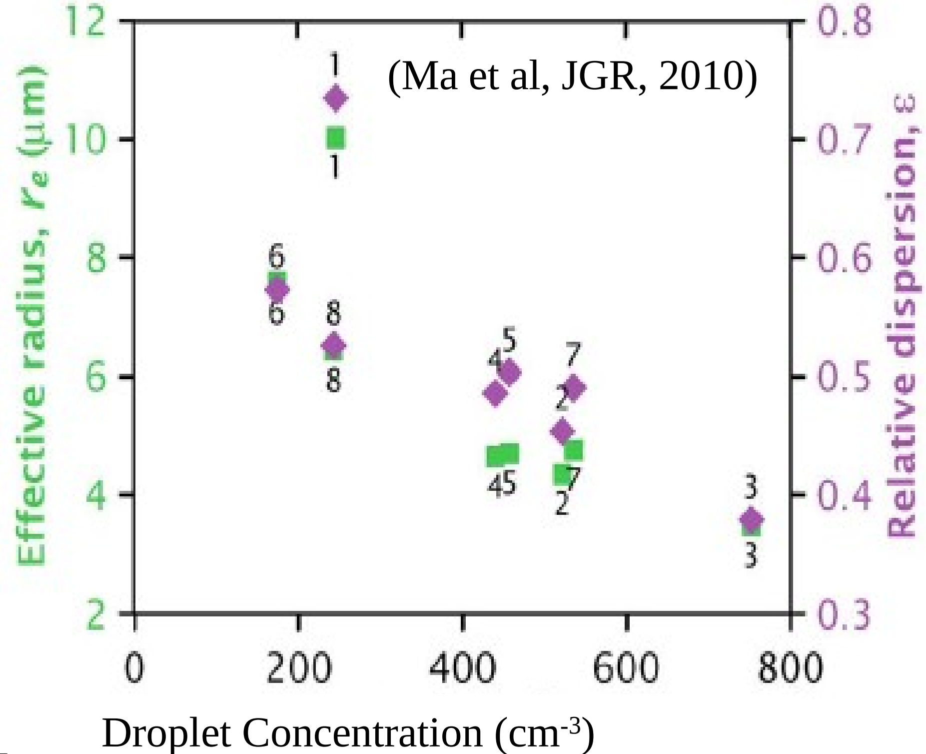


Green dashed line indicates the reflectivity error where overestimated cooling is comparable to the warming by doubling CO2.

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

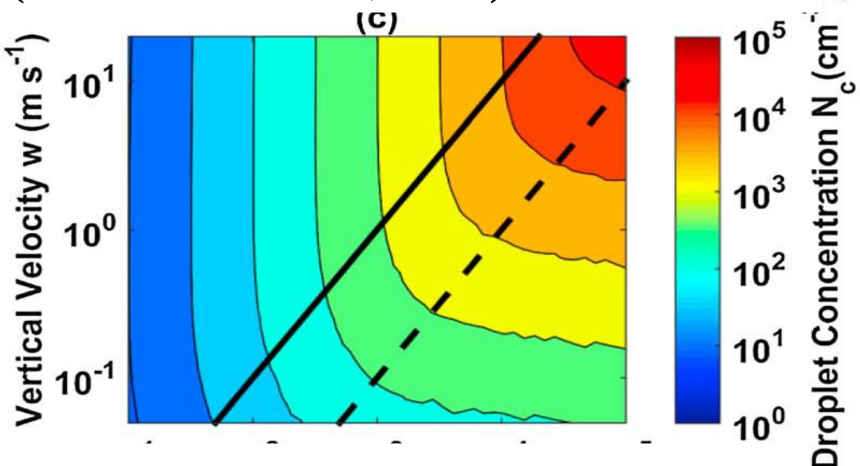
Warming dispersion effect:
(Lu et al, JGR, 2007; Chen et al, ACP, 2012; Pandithurai et al, JGR, 2012; Kumar et al ACP, 2016)

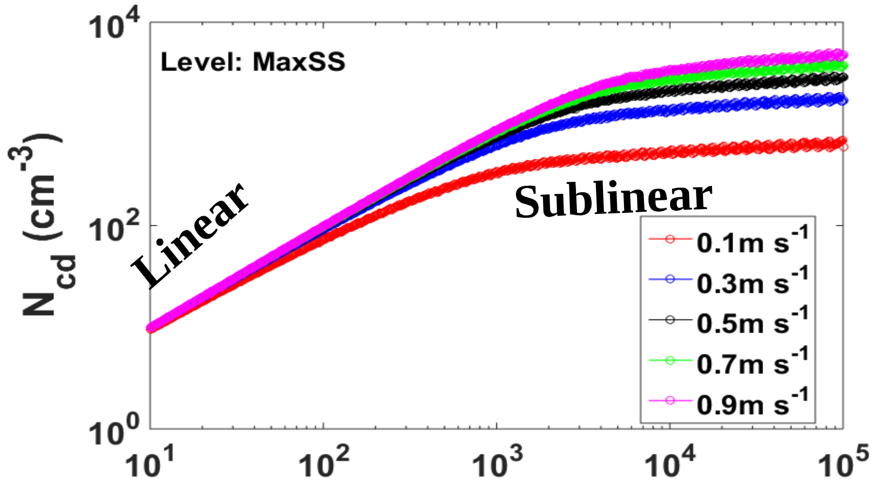
Cooling Dispersion Effect:
(Martins et al, ERL, 2009; Hudson et al, JGR, 2012)

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

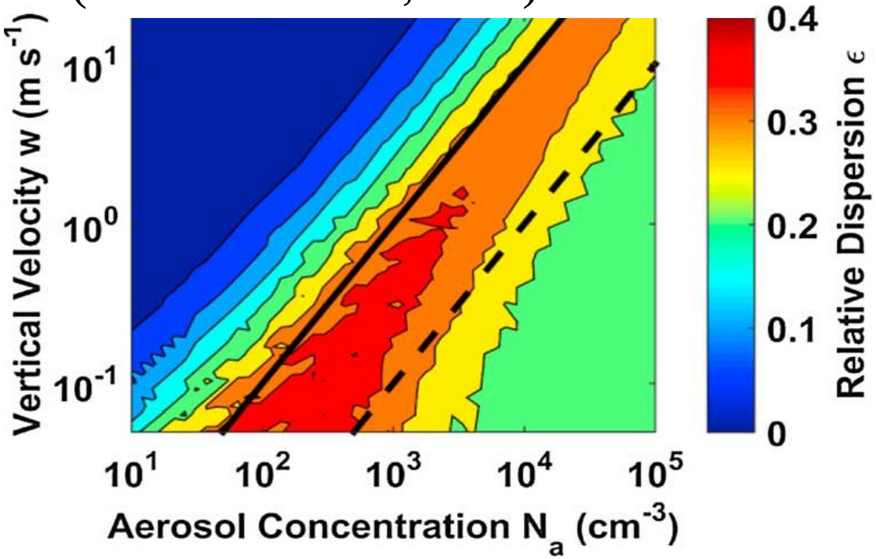
AIE Regime Dependence

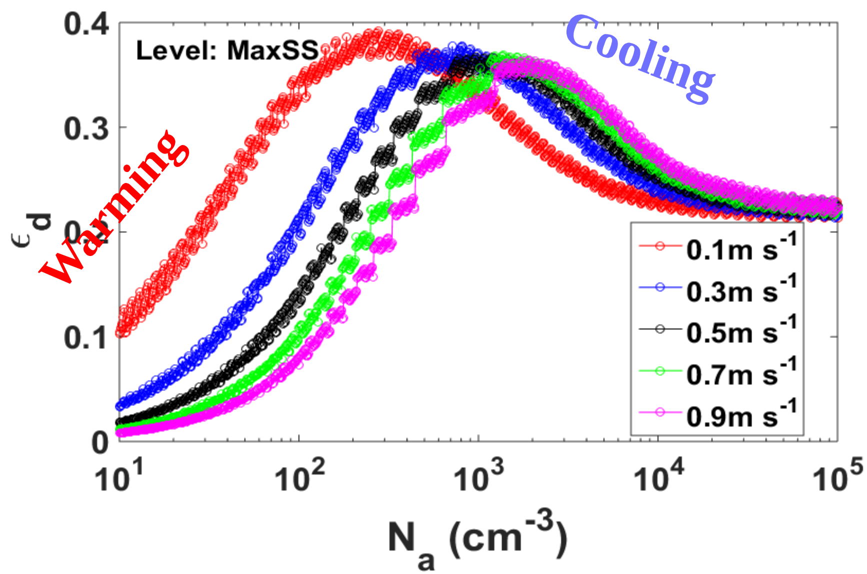
(Reutter et al. ACP, 2009)





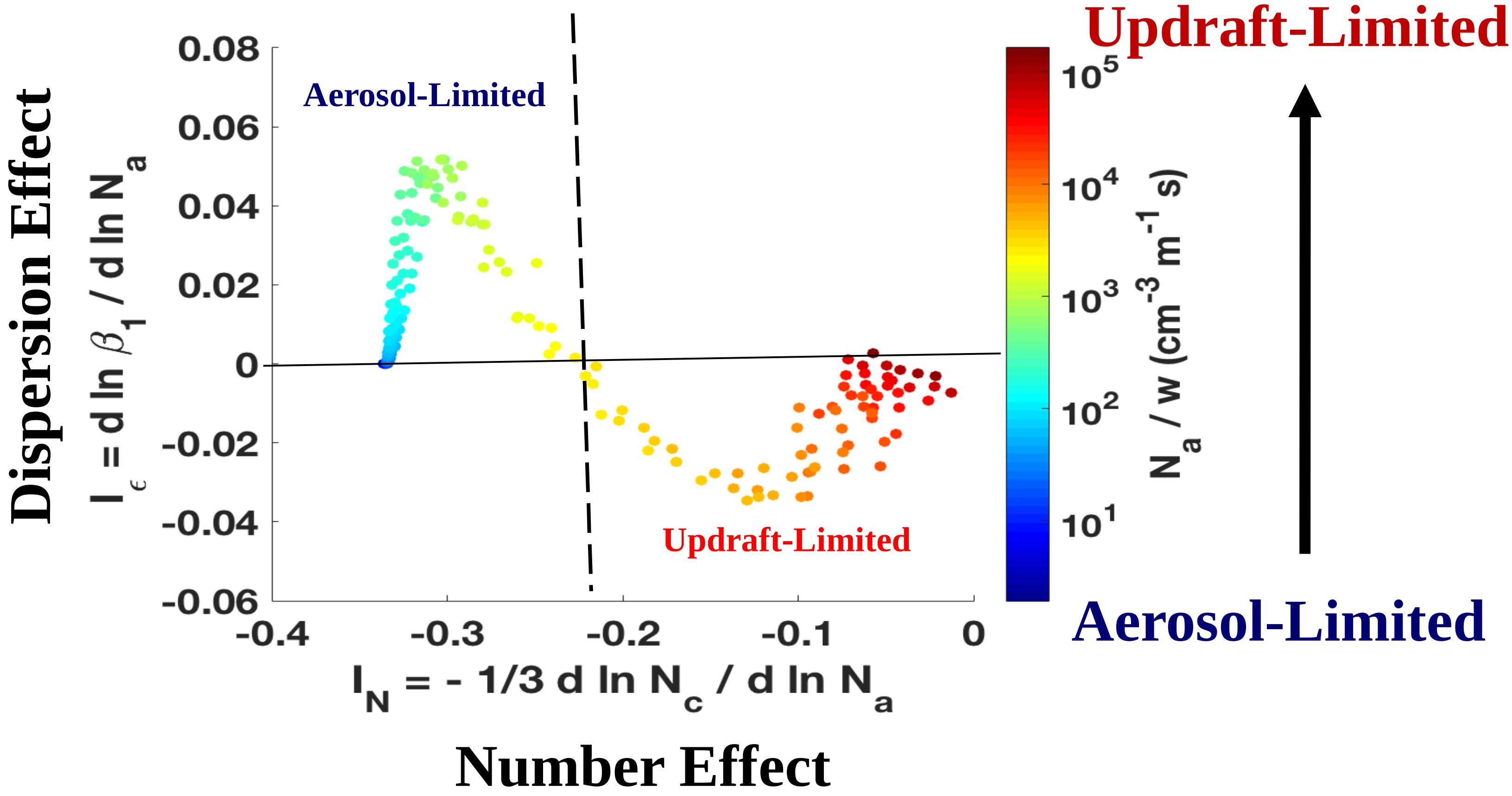
(Chen et al. GRL, 2016)





Dispersion effect exhibits stronger regime dependence & works to "buffer" number effect!

More Interesting Compensations between Dispersion Effect & Number Effect



- **Peaks in dispersion effect in aerosol-& updraft-limited regimes**
- **Entrainment-mixing processes alter this pattern? go beyond adiabatic paradigm,**

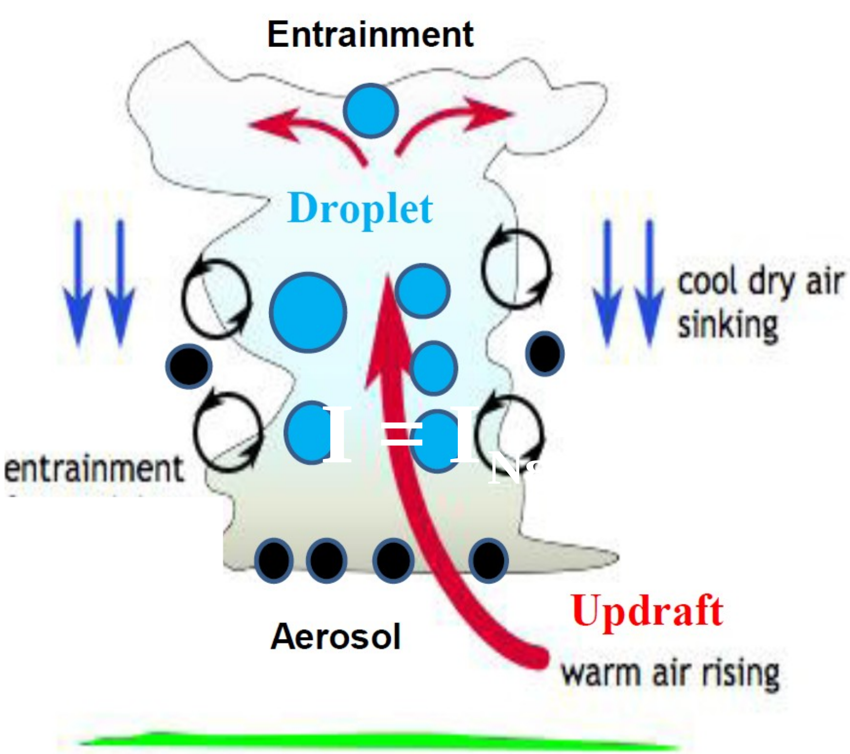
Summary I

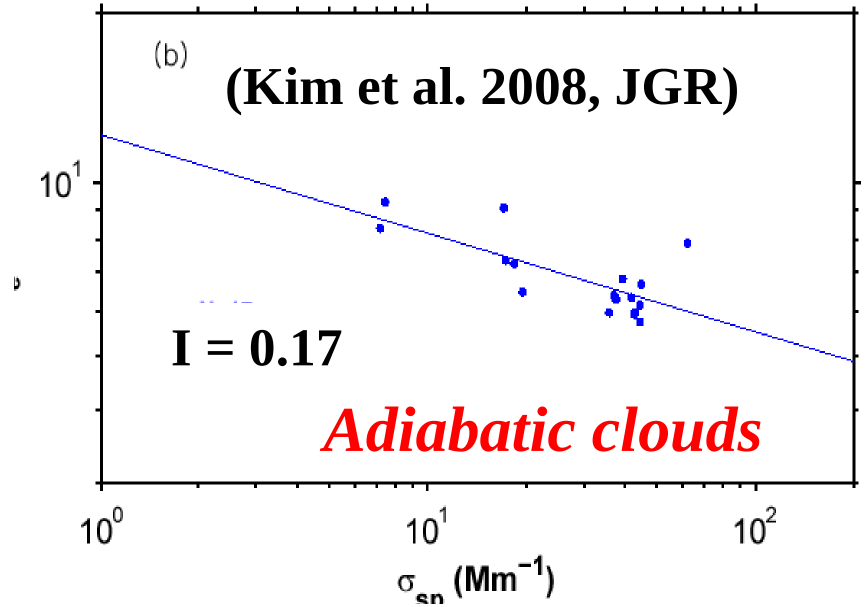
- **Dispersion effect can be warming or cooling, pending on relative impacts of updraft and aerosols (*aerosol-limited, updraft-limited, and transitional regime*).**

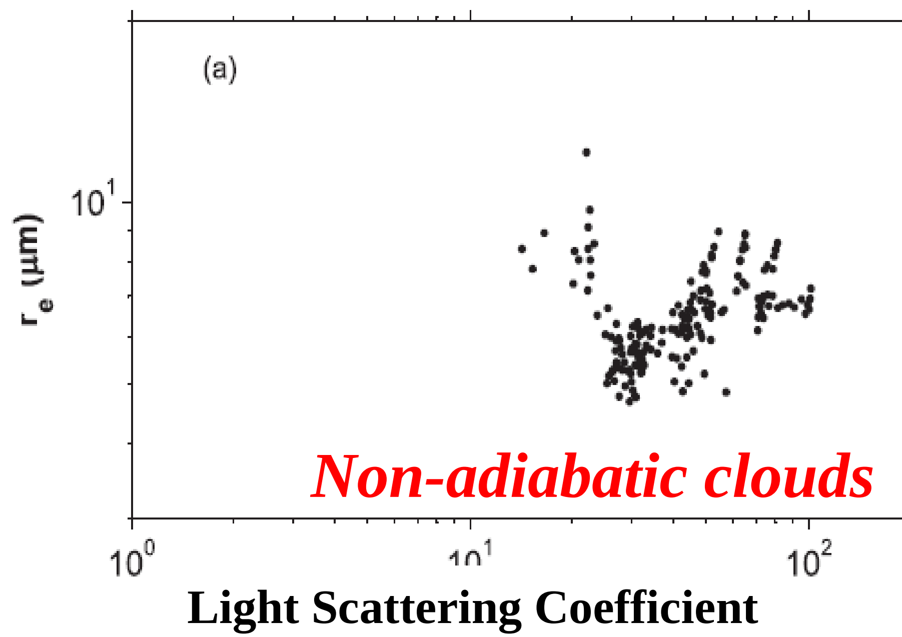
- **Dispersion effect mitigates cooling when number effect is large, but enhances cooling when number effect is small.**

- **Remaining puzzles:** ***overestimated number effect, but underestimated dispersion effect***
- **Go beyond adiabatic paradigm: turbulent *entrainment-mixing* offsets AIE cooling by reducing number effect but enhancing dispersion effect?**

Effect of Entrainment-Mixing Processes

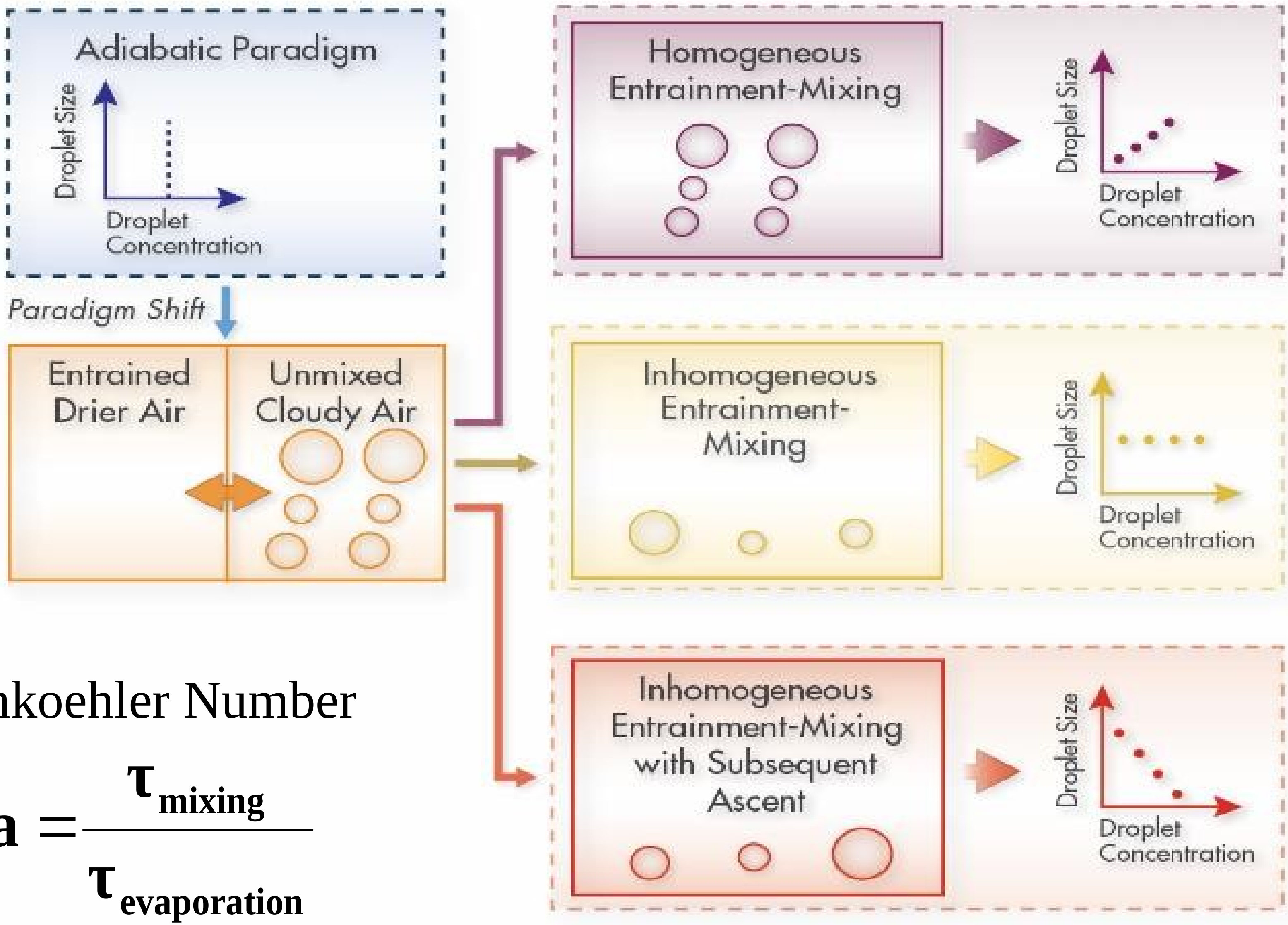






Entrainment-mixing processes may hold the key to the remaining puzzles.

Different entrainment-mixing processes alter cloud properties



Damkoehler Number

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

Economic Analogy

A Cloud

LWC= 0.01 g m^{-3}/Droplet*10 Droplets=0.1 g m^{-3}

Entrainment

LWC deceases from 0.1 g m^{-3} to 0.09 g m^{-3}.

Decrease the size of each droplet.

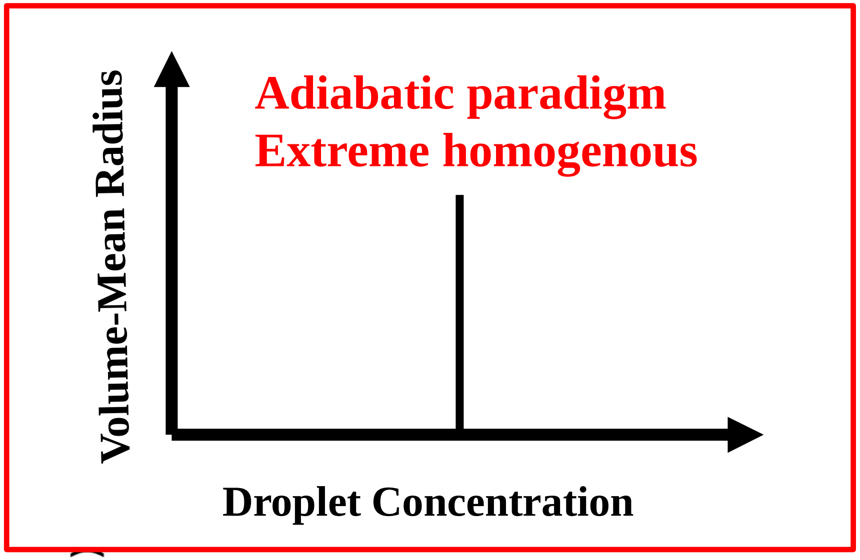
Decrease the number of droplets.

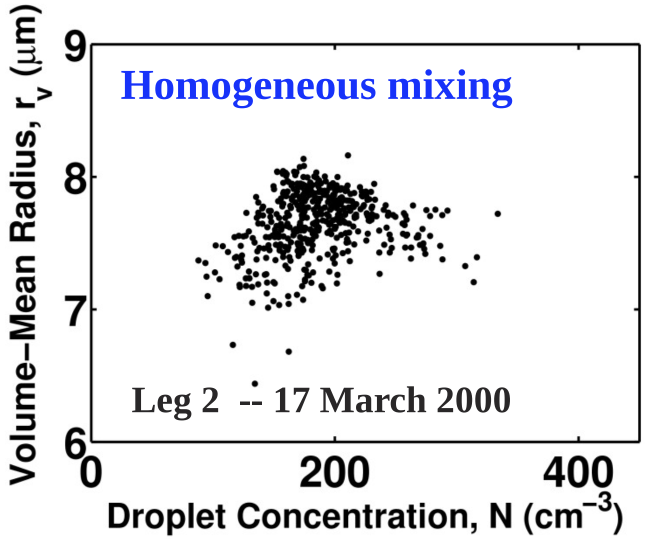
Homogeneous

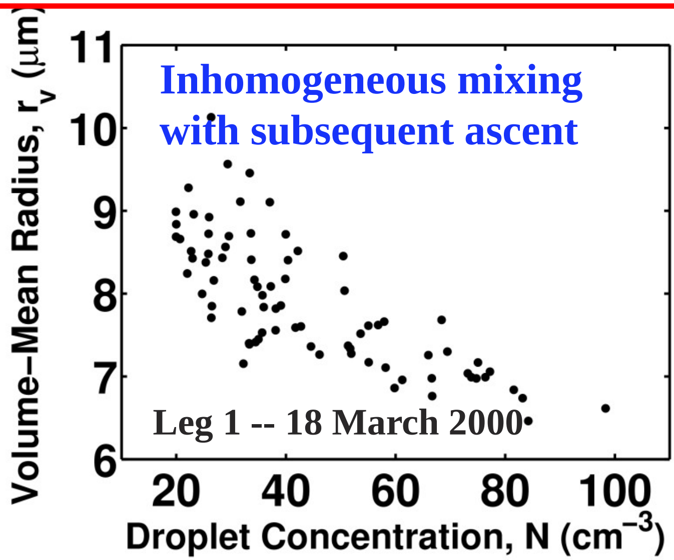
Extreme Inhomogeneous

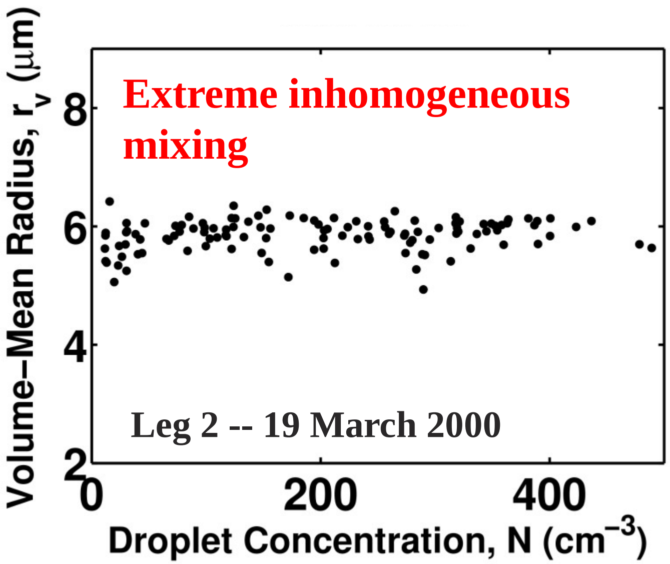
Observational Examples

March 2000 Cloud IOP at SGP



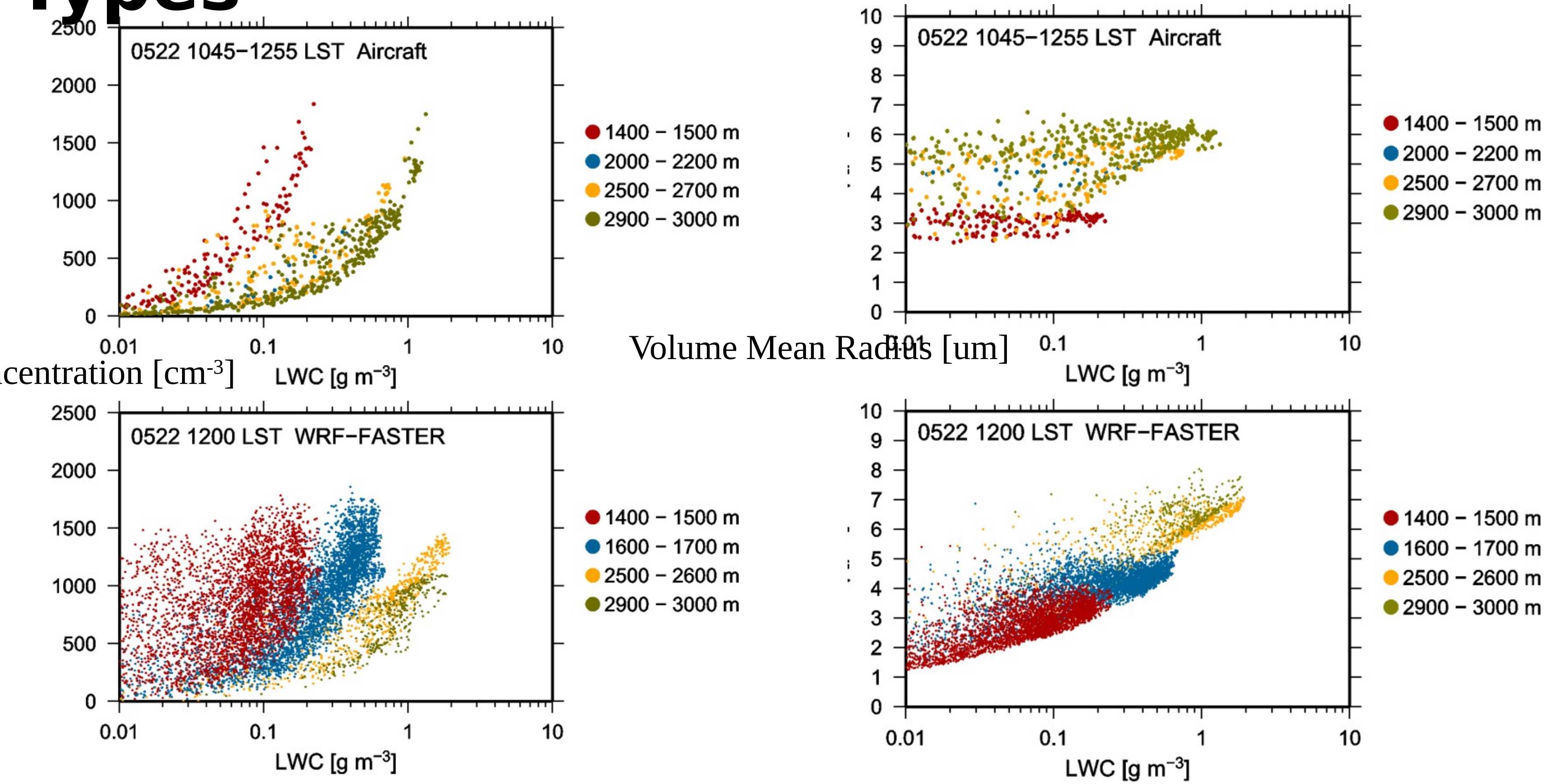






A measure is needed to cover all!

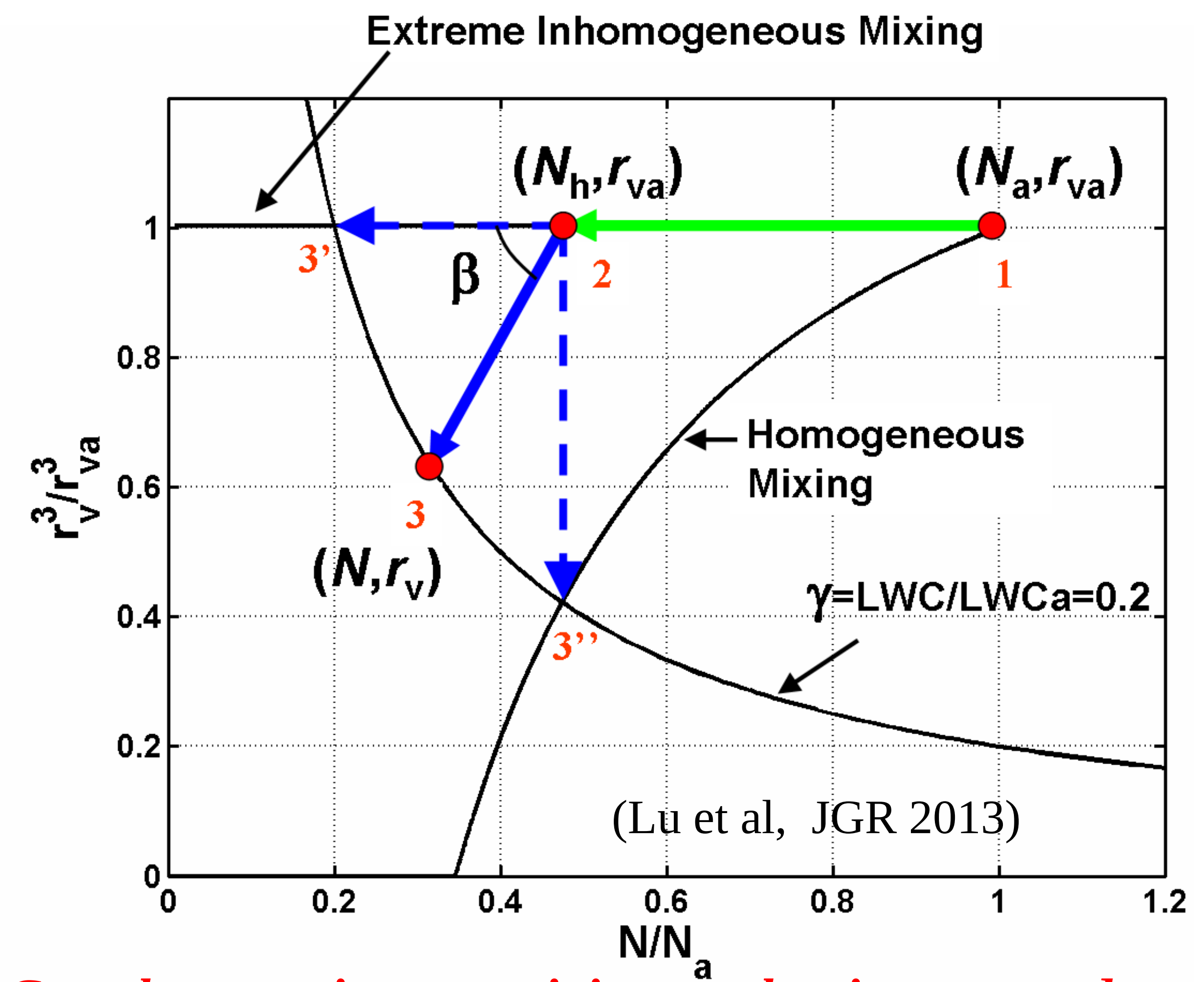
LES Cannot Capture Observed Mixing Types



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

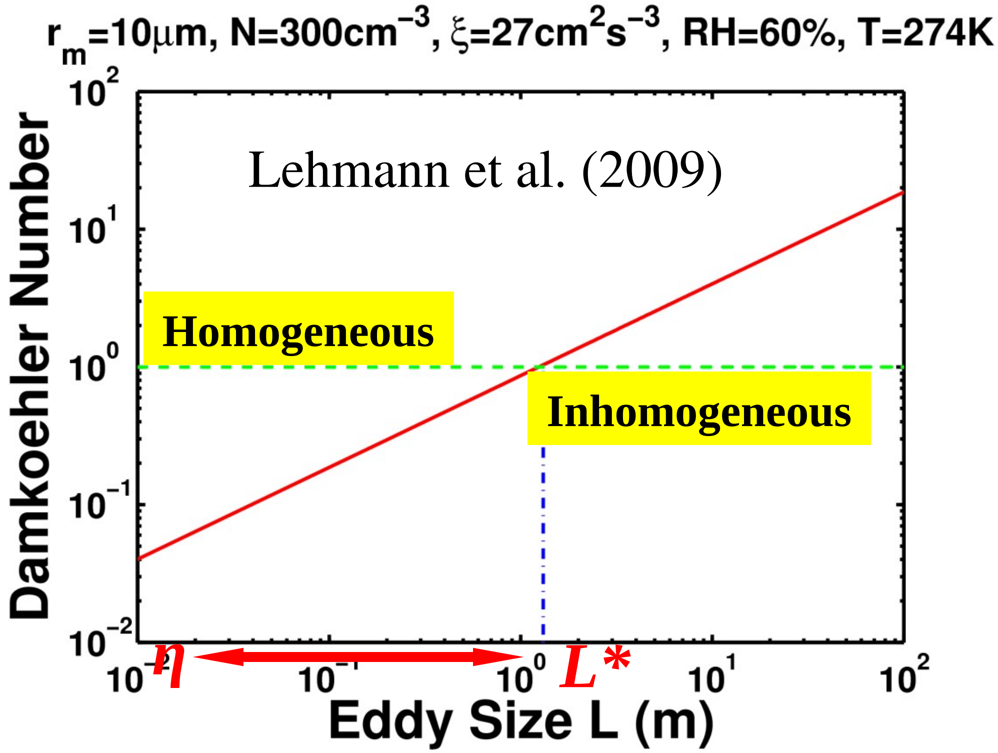
Ψ_1= 0 for extreme inhomogeneous

Ψ_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



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

A larger N_L indicates a higher degree of homogeneous mixing.

- **Transition length L^* is the eddy size of Da =1:**

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

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

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

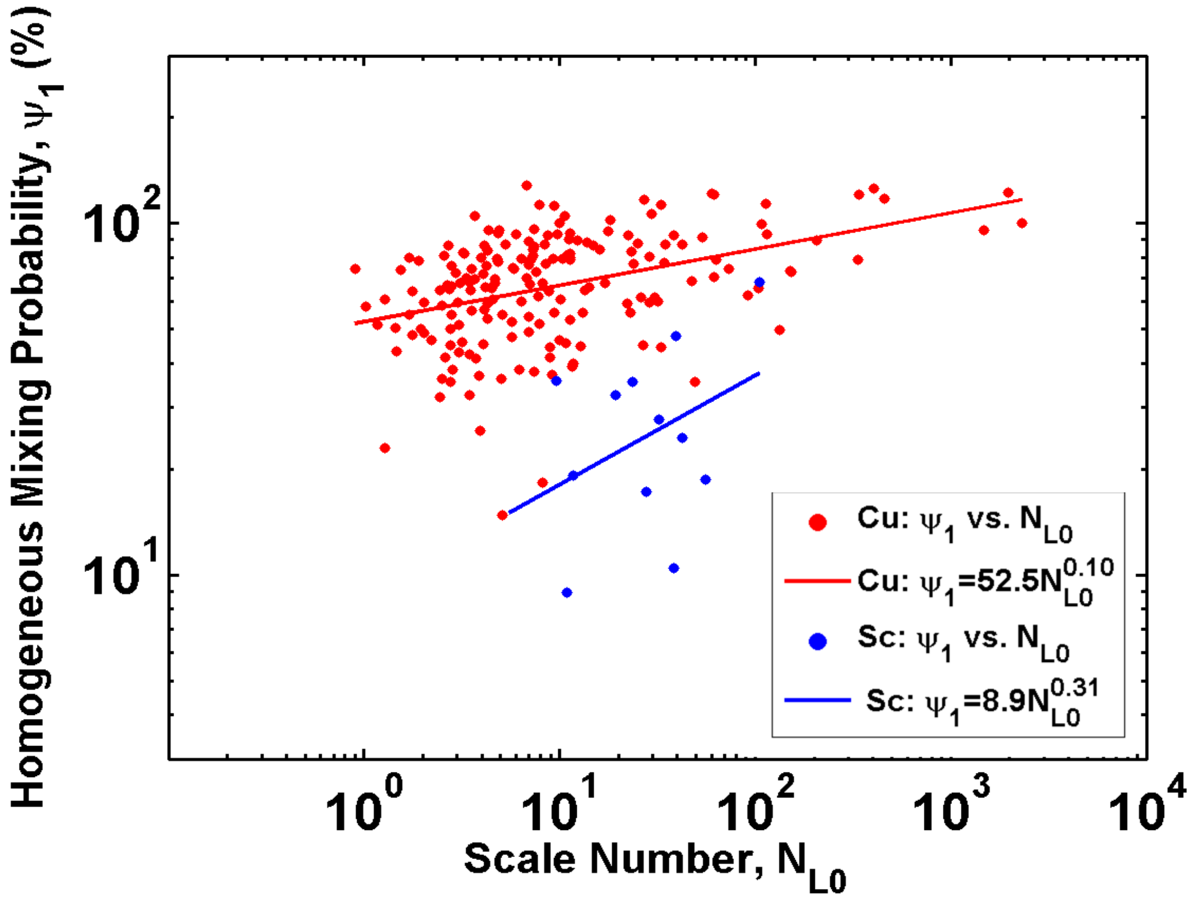
- **Transition scale number:**

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

η: Kolmogorov scale; ξ: dissipation rate; ν: viscosity

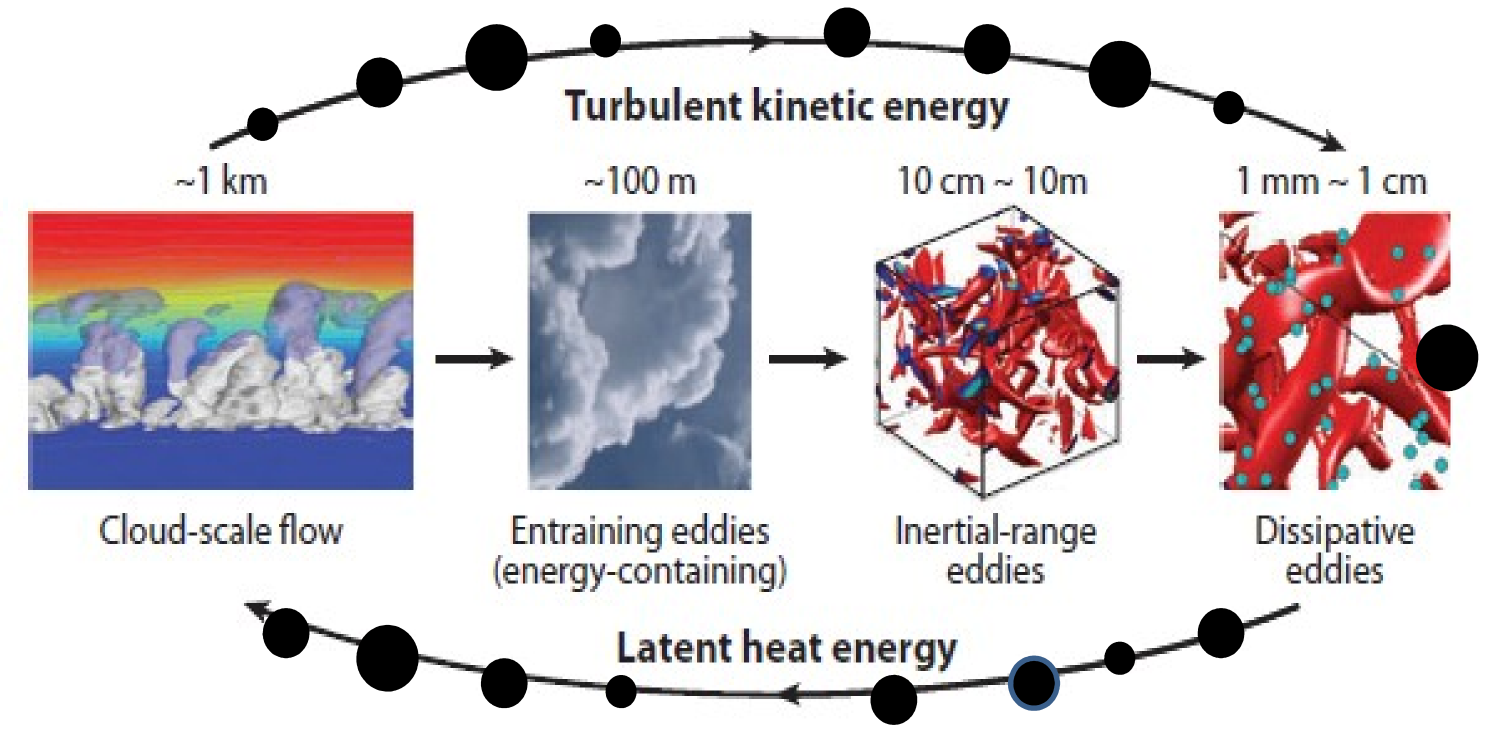
Parameterization for Mixing Mechanisms

- Eliminate the need for assuming mixing mechanisms
- Scale number can be estimated and thus homogeneous mixing degree 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, 2018)

Knowledge Gaps for Sub-LES Scale Processes

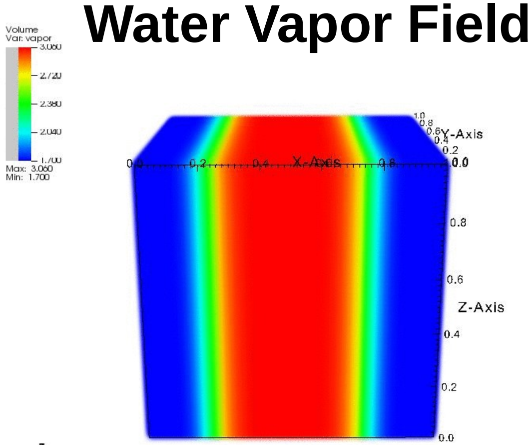


Modified from Grabowski and Wang (2013)

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

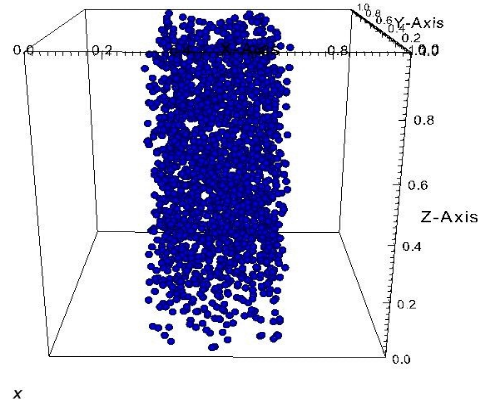
Our Particle-Resolved DNS

Water Vapor Field



$\Delta x \sim 1$cm;
Domain ~ 1 m^3

Droplets in Motion



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

- Provide a powerful tool for studying turbulence-microphysics interactions & entrainment-mixing processes, and for informing parameterization development **(of entrainment-mixing processes in our study shown**

AGU PUBLICATIONS
JGR
Journal of Geophysical Research: Atmospheres
RESEARCH ARTICLE
10.1002/2017JD027507
Special Section:
Fast Physics in Climate Models: Parameterization, Evaluation and Observation

Investigation of Turbulent Entrainment-Mixing Processes With a New Particle-Resolved Direct Numerical Simulation Model

Zheng Gao[1], Yangang Liu[1,2], Xiaolin Li[1], and Chunsong Lu[3]

Main DNS Equations

$$\begin{aligned}
\nabla \cdot \vec{u} &= 0 \\
\partial_t \vec{u} + (\vec{u} \cdot \nabla)\vec{u} &= -\frac{1}{\rho_0}\nabla p + \nu \nabla^2 \vec{u} + \vec{f} \\
\partial_t T + (\vec{u} \cdot \nabla) T &= \kappa \nabla^2 T + \frac{L}{c_p} C_d \\
\partial_t q_v + (\vec{u} \cdot \nabla) q_v &= \kappa_q \nabla^2 q_v - C_d \\
\vec{f} &= g \left[\frac{T - T_0}{T_0} + \epsilon (q_v - q_{v0}) - q_l \right] \vec{e}_z
\end{aligned}$$

Fluid Dynamics

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

$$C_d(\vec{x}, t) = \frac{1}{\rho_0 a^3} \sum_{\beta=1}^{\triangle} \frac{\mathrm{d} m_l(\vec{X}_\beta, t)}{\mathrm{d}t}$$

Microphysics

$$\begin{aligned}
\frac{\mathrm{d}\vec{X}}{\mathrm{d}t} &= \vec{V}(\vec{X}, t) \\
\frac{\mathrm{d}\vec{V}}{\mathrm{d}t} &= \frac{1}{\tau_p}\left[\vec{u}(\vec{X}, t) - \vec{V}\right] + \vec{g} \\
r(\vec{X}, t)\frac{\mathrm{d}\vec{r}(\vec{X}, t)}{\mathrm{d}t} &= K S(\vec{X}, t)
\end{aligned}$$

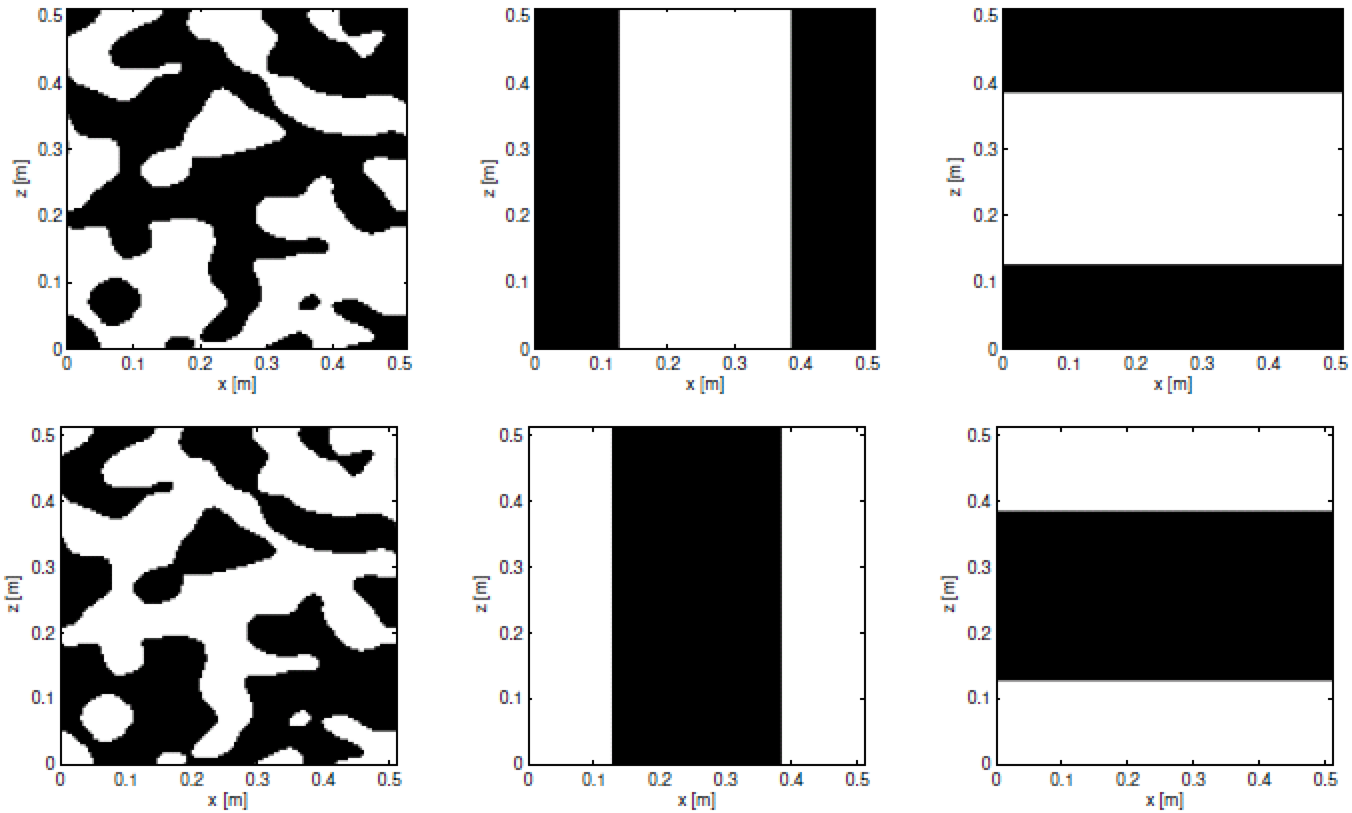
Droplet Kinetics

Six Simulation Scenarios

Case1 **Case2** **Case3**

T

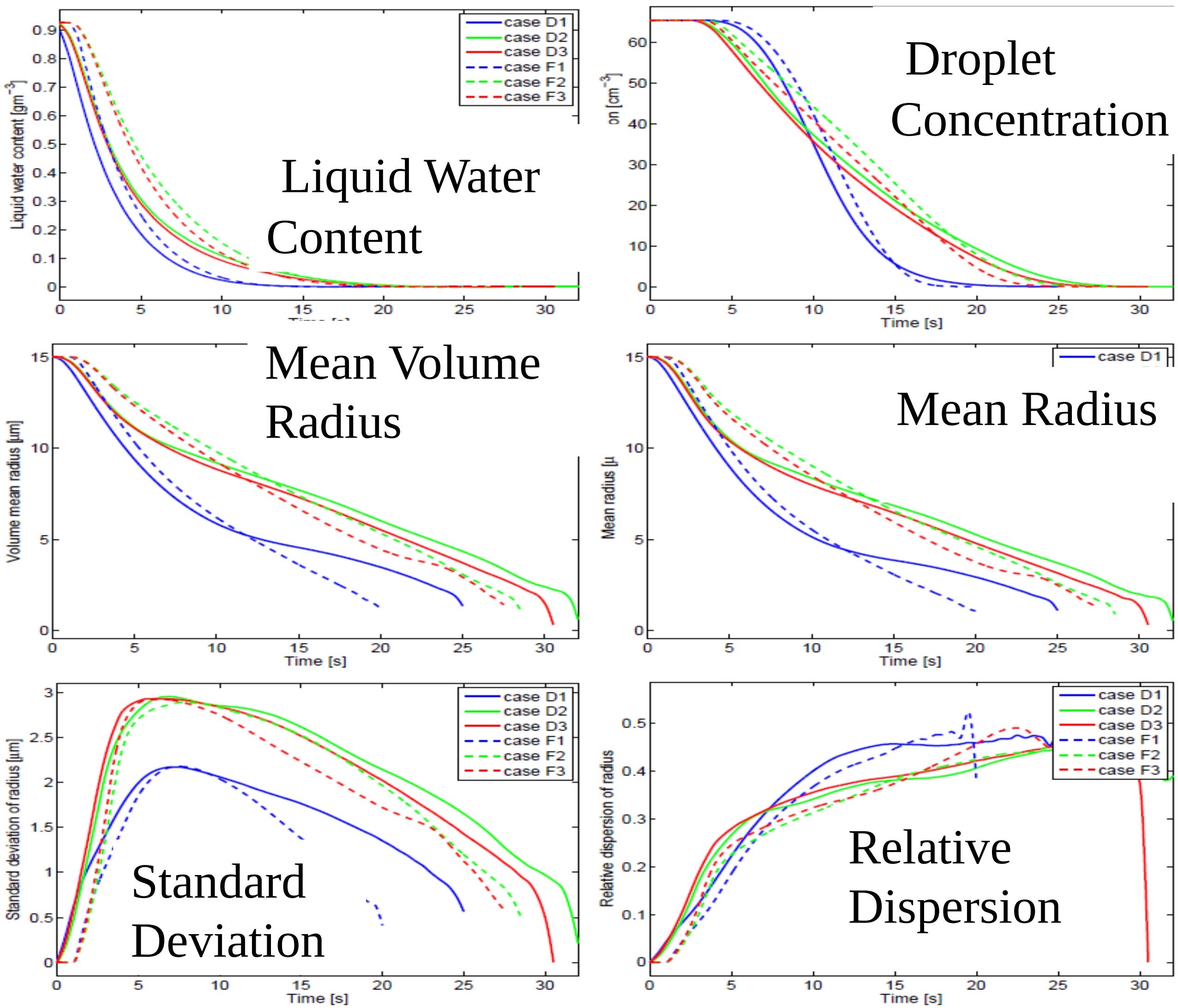
RH



(Andrejczuk et al., 2004) (Kumar et al, 2012) New

Two Turbulence Modes: Dissipating & Forced

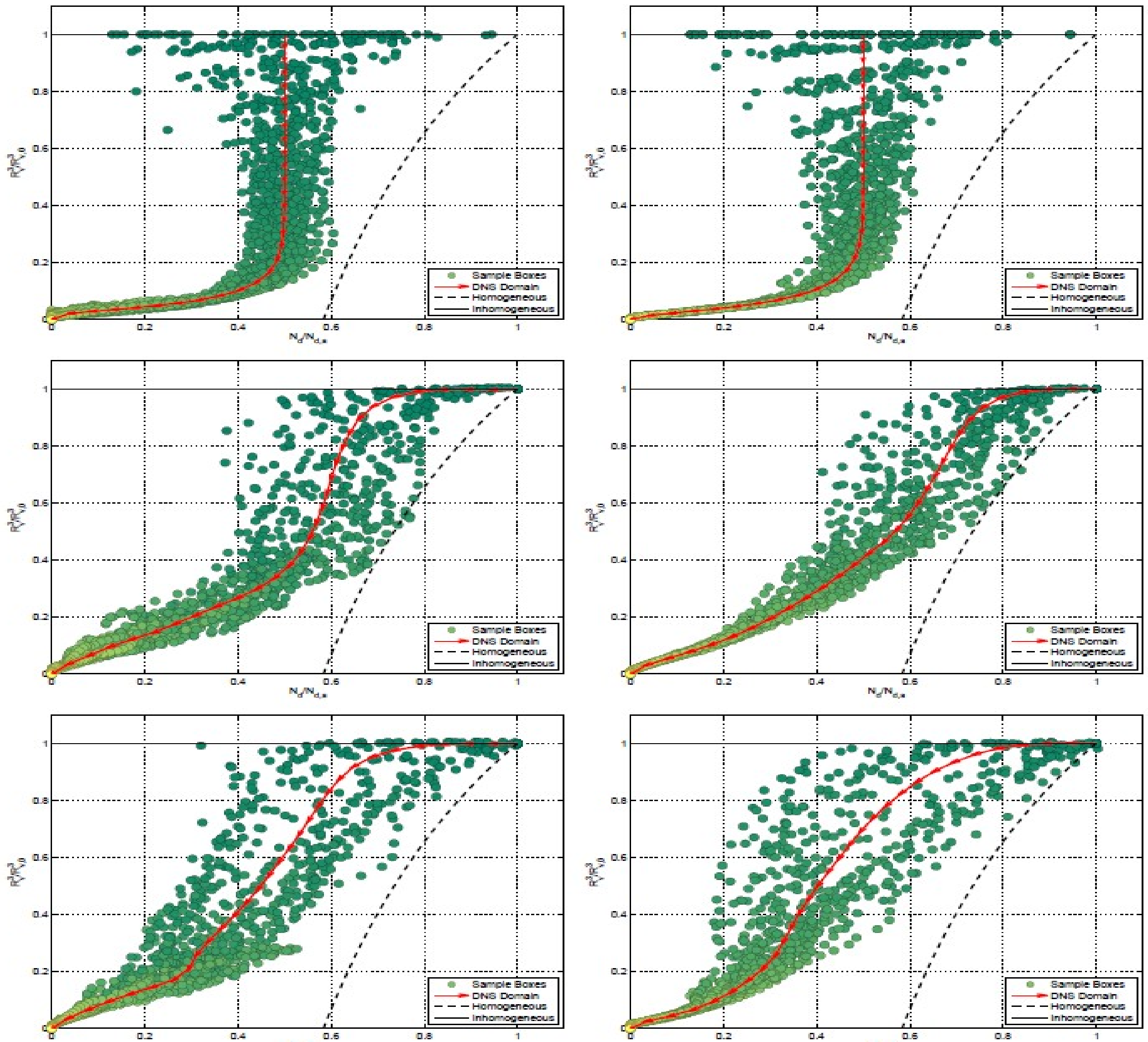
Distinct Microphysical Properties for Different Scenarios at Different Times



Time (S)

First Collapsing: Microphysical Mixing Diagram

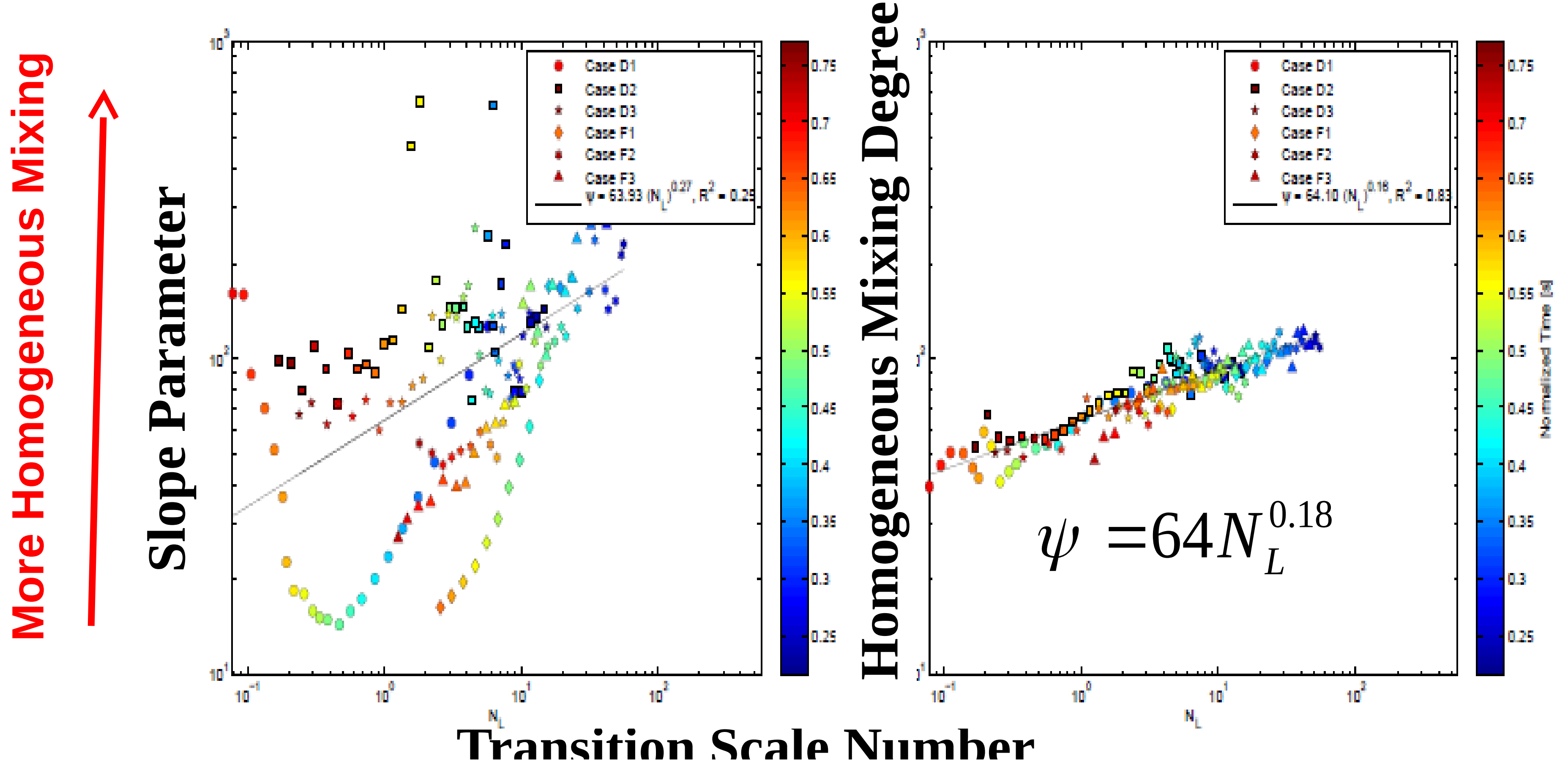
Normalized Mean Droplet Volume



Normalized Droplet Concentration

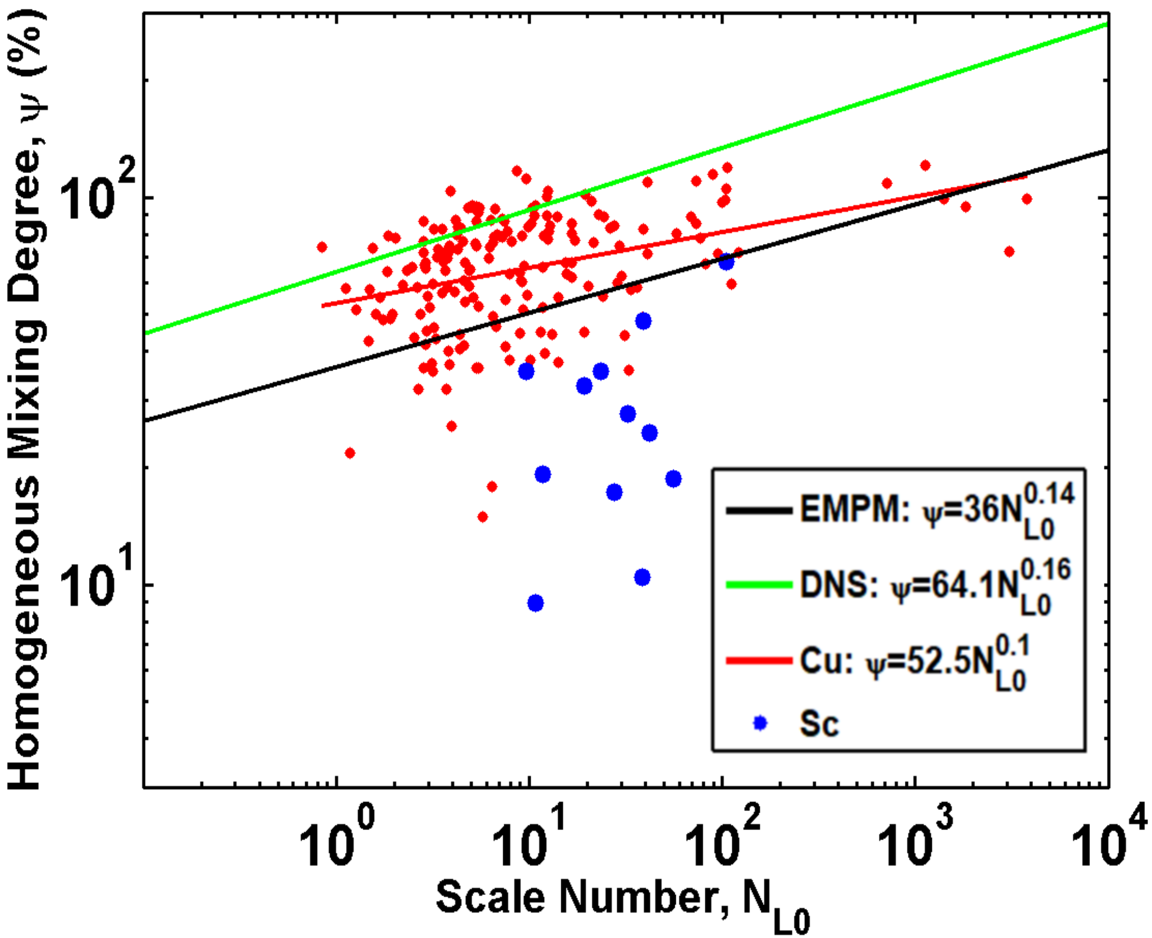
Unified Parameterization for Different Mixing Mechanisms

(Andrejczuk et al., JAS, 2009) (Lu et al., JGR, 2013)



Our measure is clearly better than the previous slope parameter; the expression can be used to parameterize mixing types in two-moment schemes. Recall the graduation normalization from original r-N mixing diagram!

Similar Mixing Parameterizations Derived from DNS, EMPM, and Observations



DNS-derived parameterization tends to be more homogeneous given transition scale number N_{L0}, suggesting possible scale-dependence?

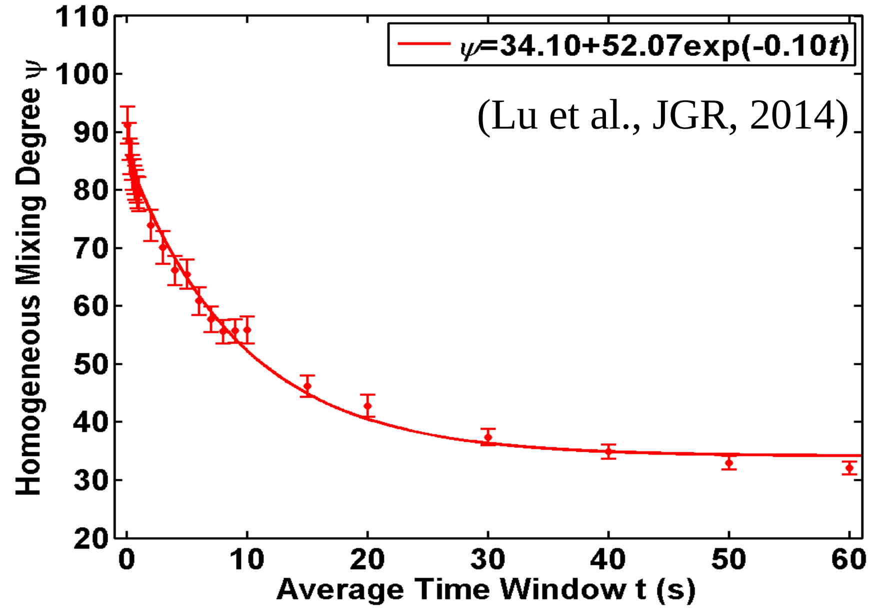
Scale-Aware Mixing Parameterization

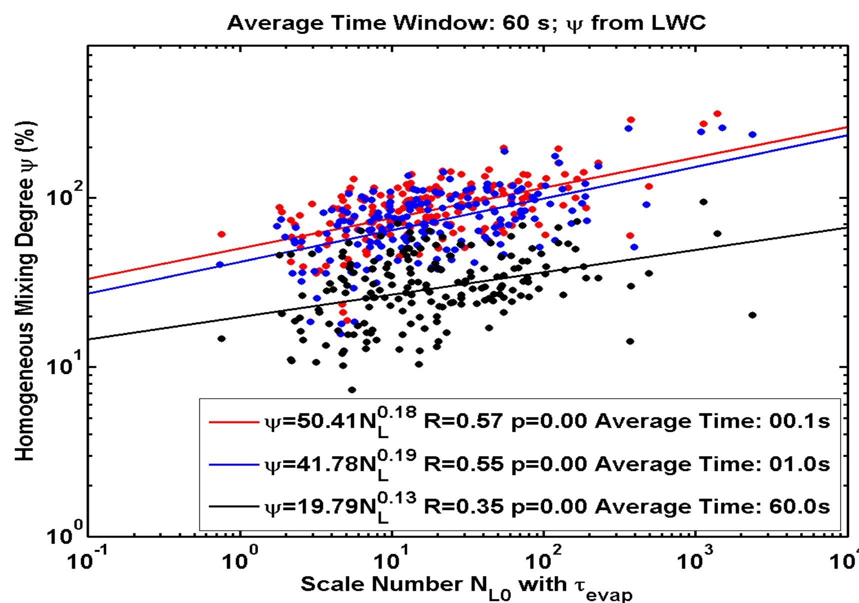
- Homogeneous mixing degree decreases with increasing averaging scales.

- Expect that transition scale number has less space-dependence, or the slope parameter varies little with averaging scale.

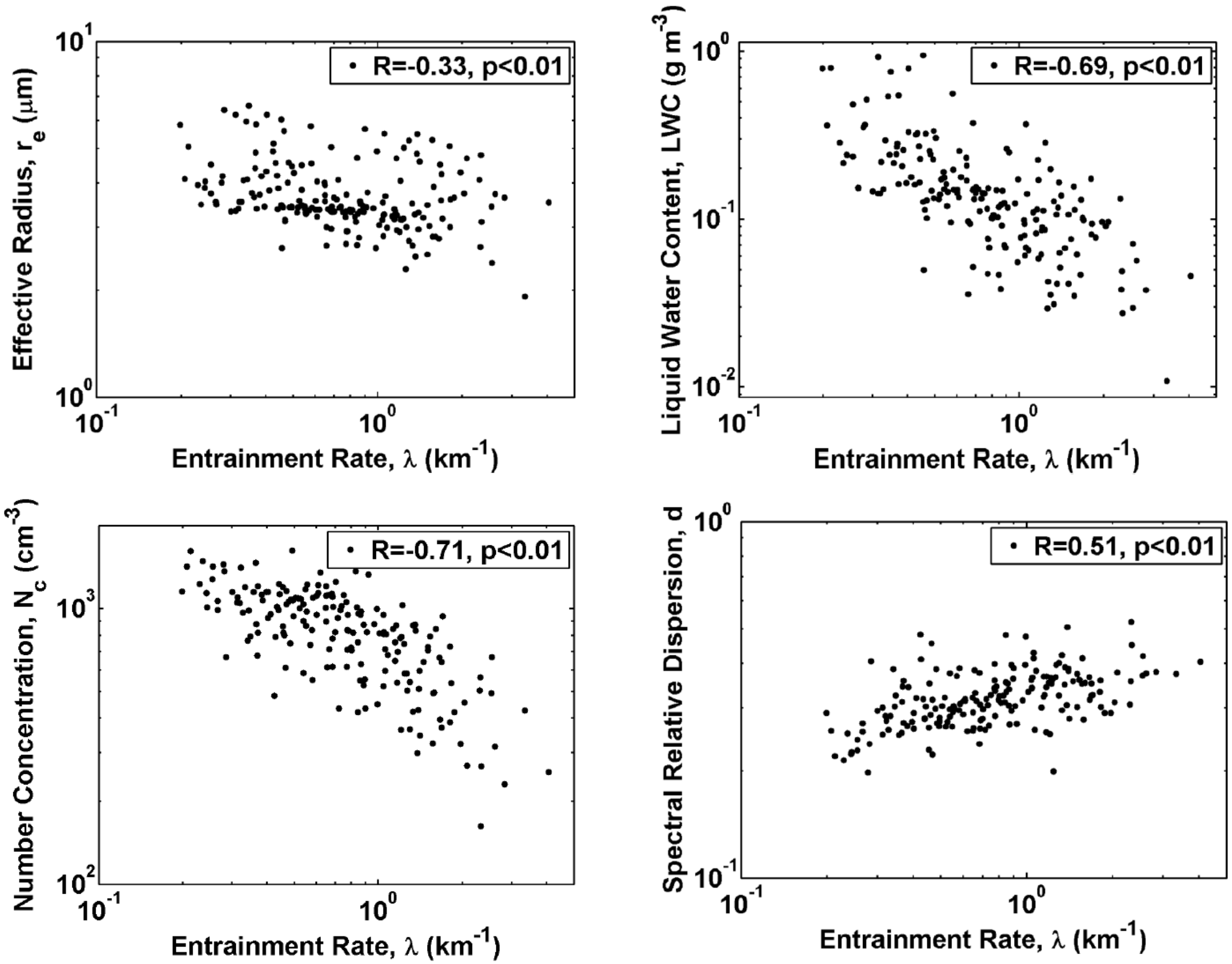
- New result confirms the expectation.

- Scale-aware mixing parameterization



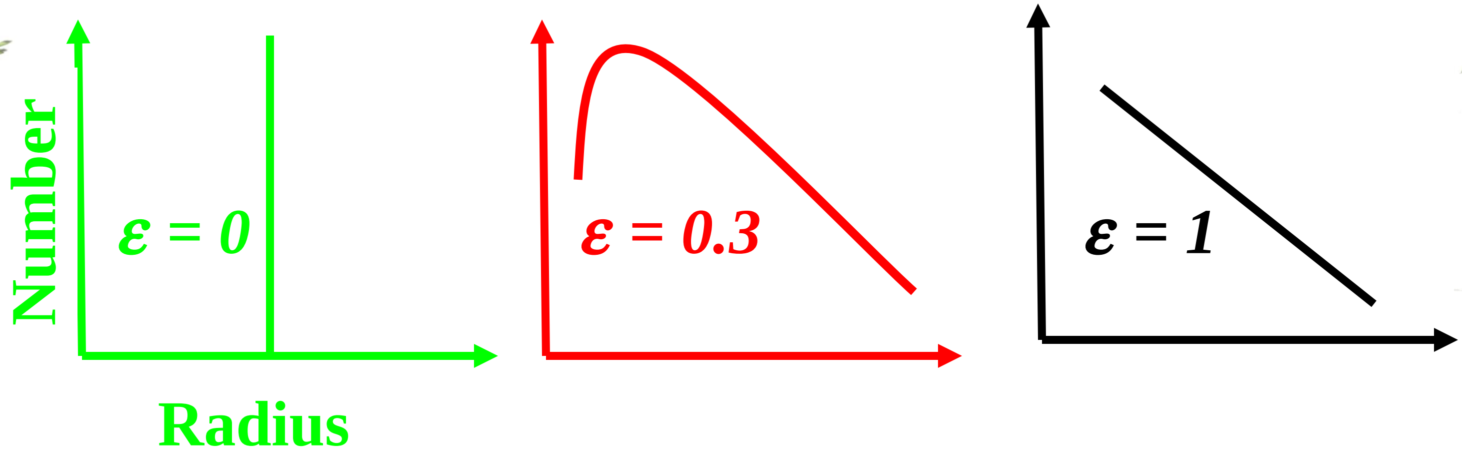


Entrainment Rate vs. Microphysics



An increase in entrainment rate corresponds to decreases in LWC, droplet concentration, and droplet size but an increase in relative dispersion, largely consistent with homogenous mixing mechanism.

Carton to Appreciate Relative Dispersion



Dispersion ε 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!

Dispersion Enhanced Economic Analogy

A Cloud

LWC= 0.01 g m^{-3}/Droplet*10 Droplets=0.1 g m^{-3}

Entrainment

LWC deceases from 0.1 g m^{-3} to 0.09 g m^{-3}.

Decrease t
each dropl

umber of

Adjust both the number and individual salary to make the company more cost-effective!?

Homogeneous **Extreme Inhomogeneous**

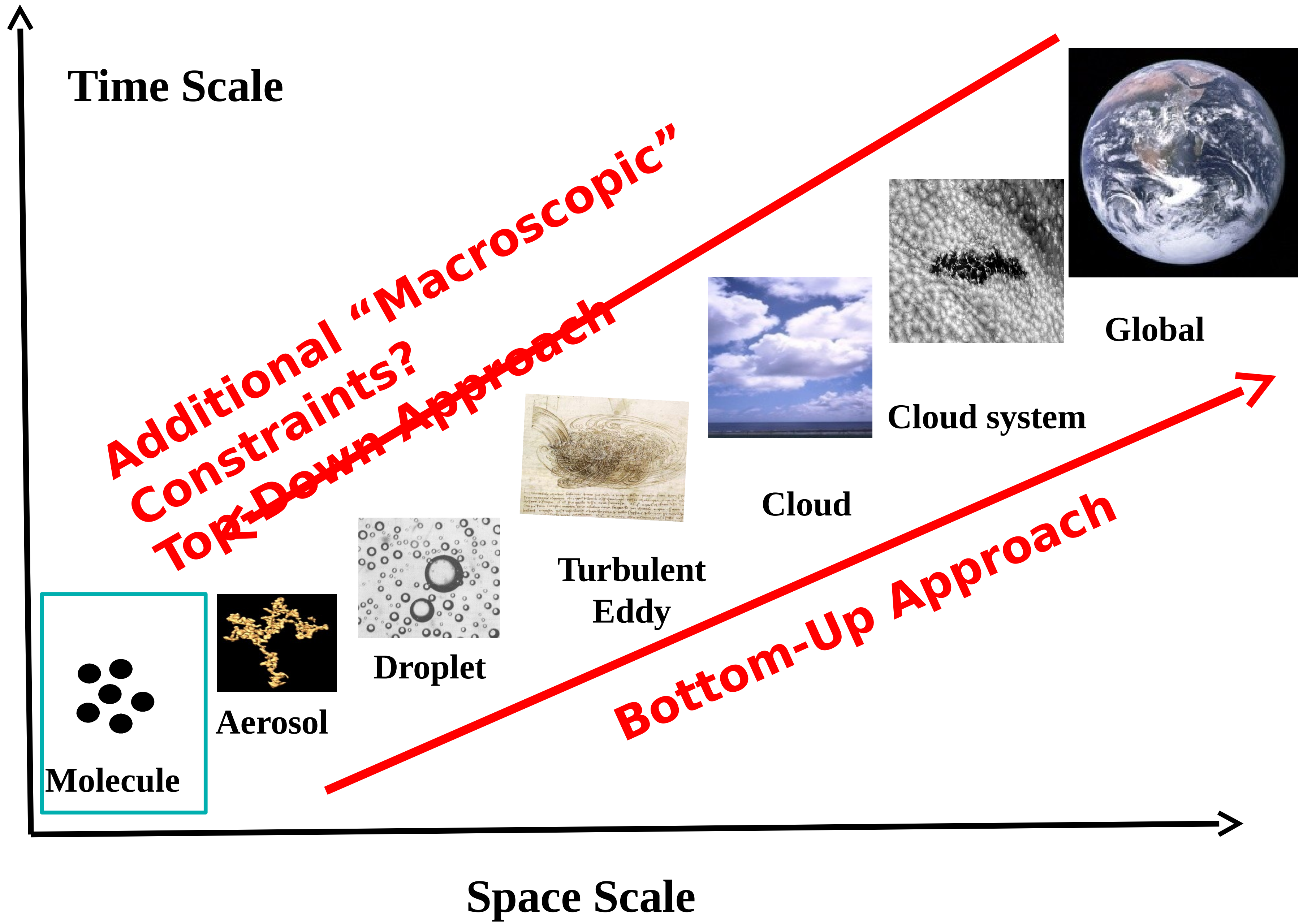
Summary II

- **Twomey and other pioneers identified the first order effects, leaving other detailed challenges to us.**

- **Dispersion effect & entrainment-mixing processes are two factors likely buffering the conventional AIE cooling.**
- **Consideration of spectral shape poses new challenges to parameterize entrainment-mixing processes.**

- **Other alternative ideas?**

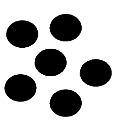
Multiscale Climate Hierarchy



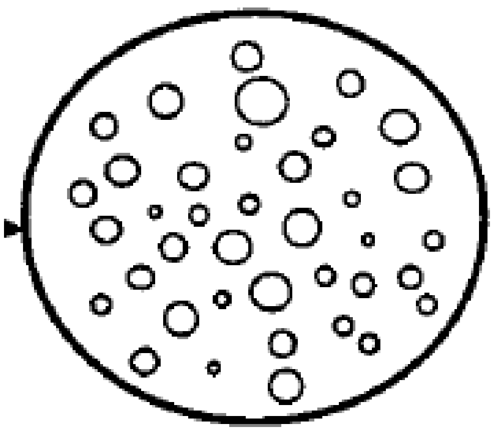
Fast Physics Parameterization as Statistical Physics

- “Statistical physics“ is to account for the observed thermodynamic properties of systems in terms of the statistics of large ensembles of “particles”.

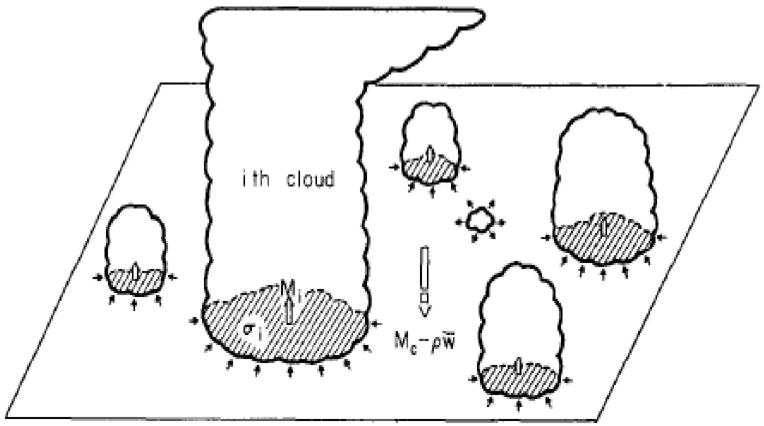
- “Parameterization” is to account for collective effects of many smaller scale processes on larger scale phenomena.



Molecule Ensemble
Kinetics, Statistical Physics, Thermodynamics

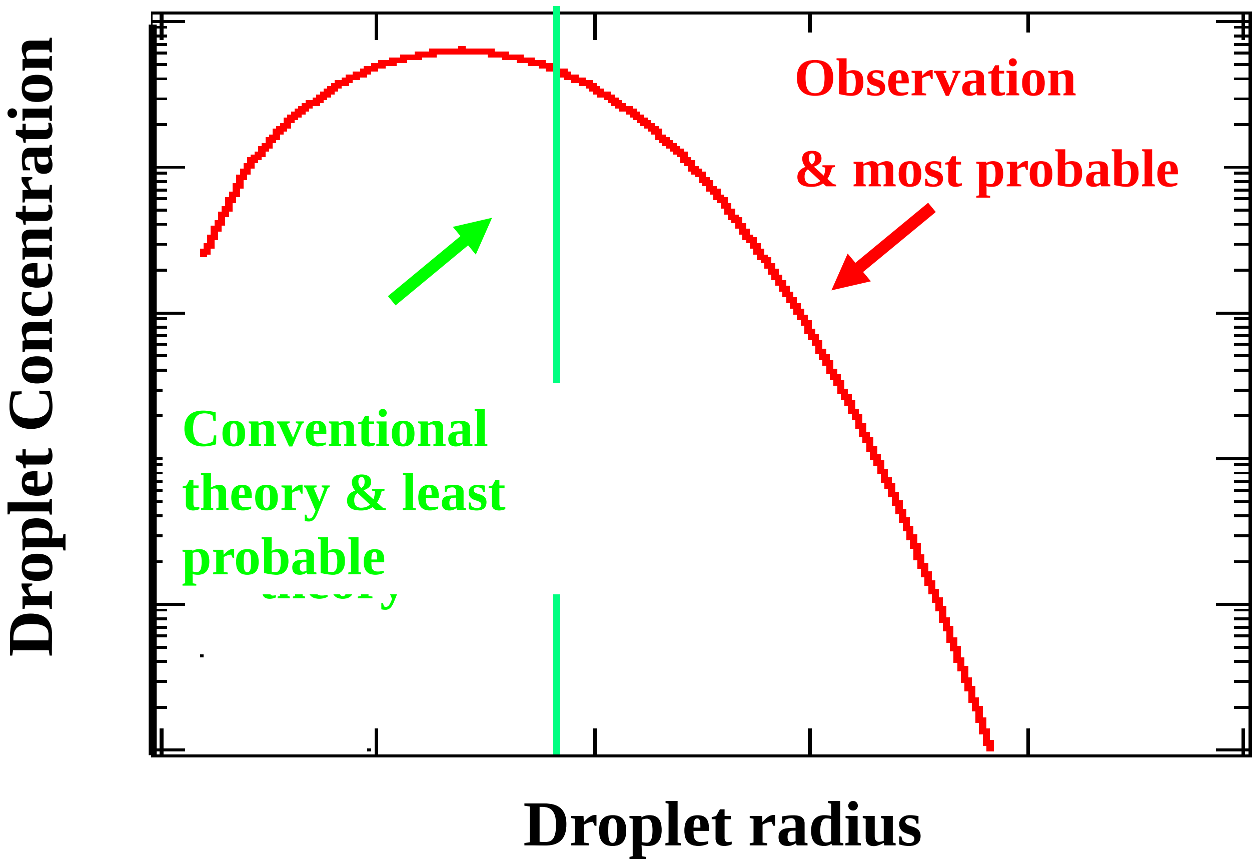


Droplet Ensemble
Systems Theory



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

Entropy-Based Systems Theory



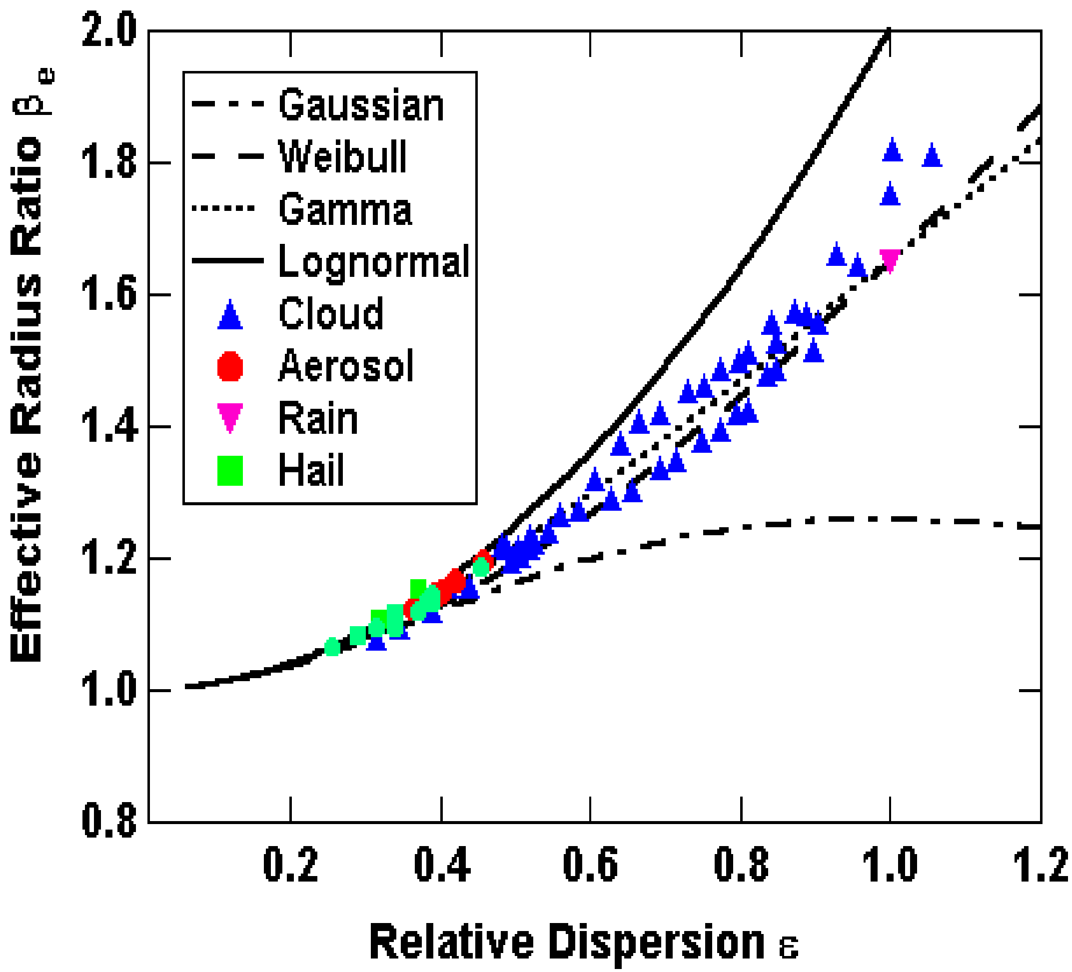
The systems theory predicts that Weibull (delta) distribution is the most (least) probable distribution given L and N (Liu et al., AR, 1994, 1995; Liu & Hallett, QJ, 1998; JAS, 1998, 2002; Liu et al, 2002).

Observational Validation of Weibull/Gamma Particle Distribution

- **Each point represents a particle size distribution**
- **ε = Standard deviation/mean**

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

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



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

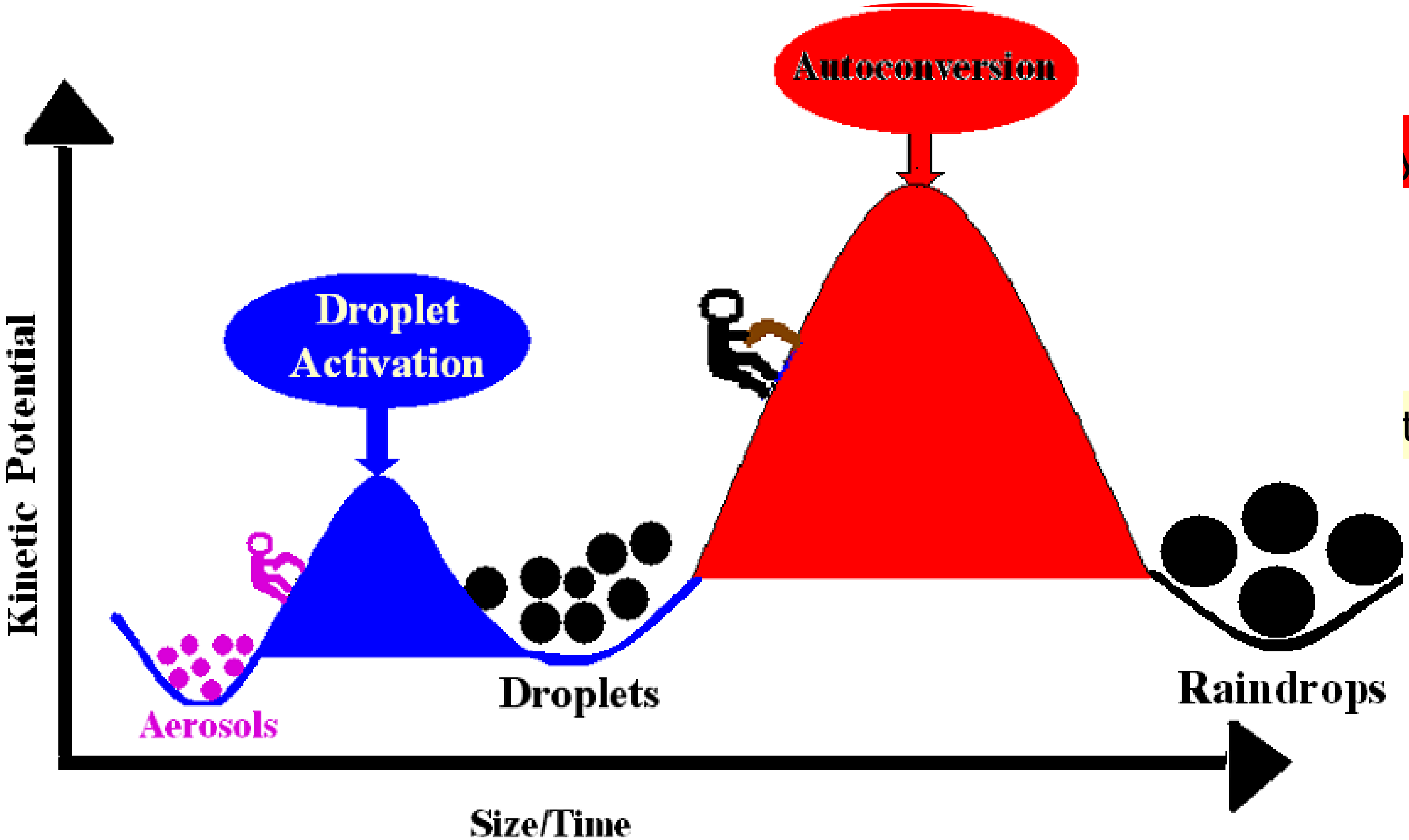
Take-Home Messages

- **Dispersion effect & entrainment-mixing are important AIE buffers**
- **Have expression predicting dispersion for adiabatic clouds**
- **Have a way to parameterize entrainment-mixing effect on droplet concentration and water content**
- **Have a theory on functional form of droplet size distribution influenced by entrainment-mixing**
- **Predicting entrainment-mixing-dispersion relationships remains a great challenge!**

Thanks for your attention!

Backup slides

Systems Theory
Unifying Microphysics Parameterizations



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Work in progress!

Commonly Used Size Distribution Functions

Table 1.1. Summary of Empirical Expressions for Size Distribution

Name	Expression	Parameters
Weibull	$n(D) = N_o D^q \exp(-\lambda D^q)$	No, λ, q
Gamma	$n(D) = N_o D^\mu \exp(-\lambda D)$	No, μ, λ
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, Dm, σ
Power-law	$n(D) = aD^{-b}$	a, b
Exponential	$n(D) = N_o \exp(-\lambda D)$	No, λ
Normal	$n(D) = \frac{N}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(D-D_m)^2}{2\sigma^2}\right)$	N, Dm, σ
Modified gamma	$n(D) = N_o D^\mu \exp(-\lambda D^q)$	No, μ, λ, q
Delta function	$n(D) = N\delta(D - D^*)$	N, D*

$$\tau = \frac{Q}{R} = \frac{Q}{kQ} = \frac{1}{k}$$

(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?

Statistical Physics for Microphysics Parameterization:

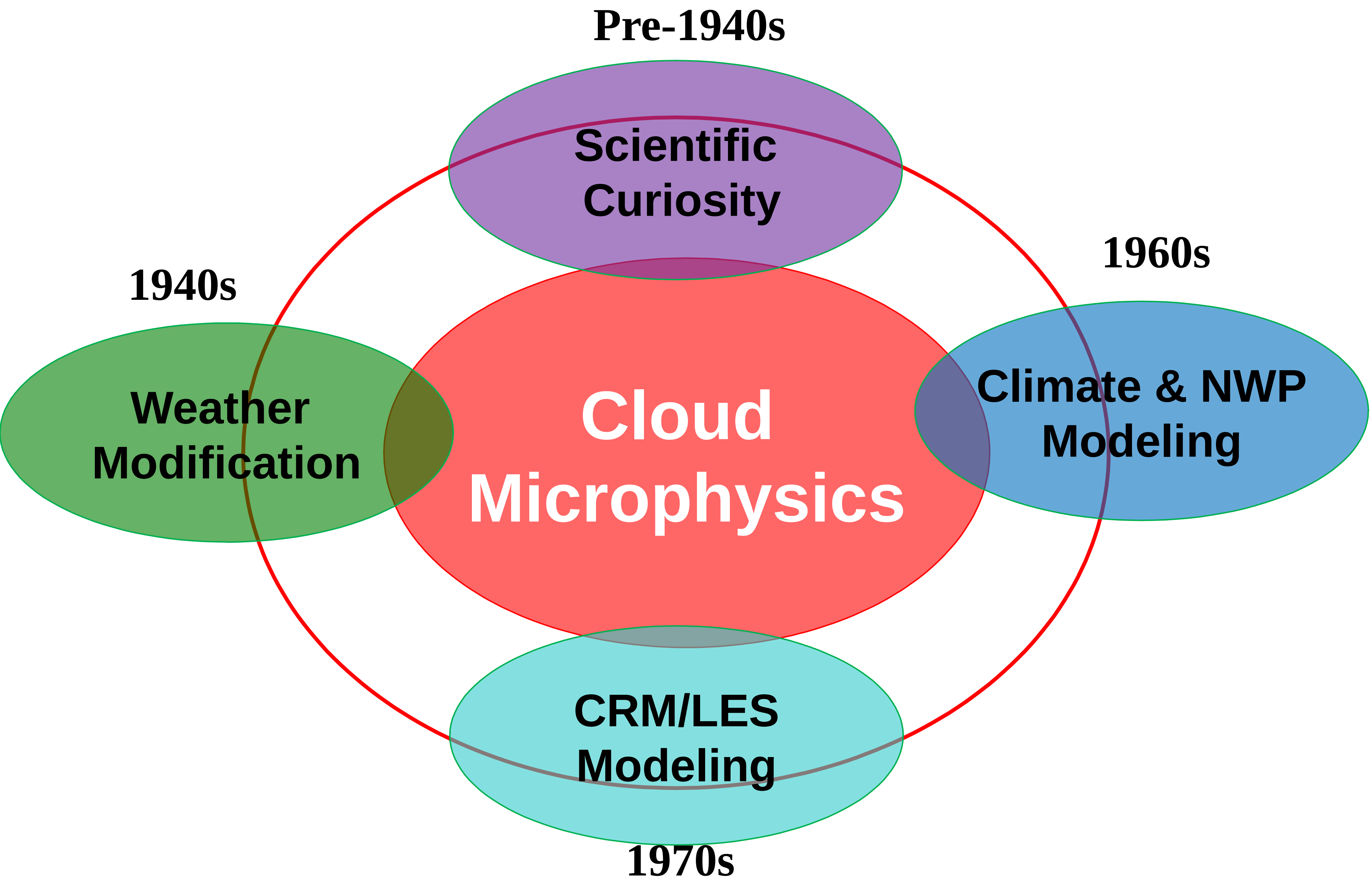
Entropy-Based Theory for Gamma/Weibull Size Distribution

(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)

Four Fundamental Sci. Drivers



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.

Molecular system, Gas	Clouds
Knew Newton's mechanics for each molecule	Know equations for each droplet
Kinetics failed to explain observed thermodynamic properties	Models failed to explain observed size distribution
Maxwell, Boltzmann, Gibbs established statistical mechanics	Establish the systems theory

Most probable distribution

Droplet System

Consider the droplet system constrained by

$$\int \rho(x)dx = 1 \qquad (1)$$

$$\int x\rho(x)dx = \frac{X}{N} \qquad (2)$$

x = Hamiltonian variable, X = total amount of x **per unit volume, n(x) = droplet number distribution with respect to x, ρ(x) = n(x)/N = probability that a droplet of x occurs.**

Droplet spectral entropy is defined as

$$E=-\int \rho(x)\ln(\rho(x))dx \qquad \textbf{(3)}$$

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

Liu et al. (1992, 1995, 2000), Liu (1995), Liu & Hallett (1997, 1998)

Most Probable Distribution w.r.t. x

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

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

The most probable distribution with respect to x is

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

where α = X/N represents the mean amount of x per droplet. Note that the Boltzman energy distribution becomes special of Eq. (5) when x = molecular energy. The physical meaning of α 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

$$x = ar^b$$

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

$$n^*(r) = N_0 r^{b-1} \exp\left(-\lambda r^b\right)$$

$$N_0 = ab/\alpha; \lambda = a/\alpha; \alpha = X/N$$

This is a general Weibull distribution!

Observational Validation of Weibull/Gamma Particle Distribution

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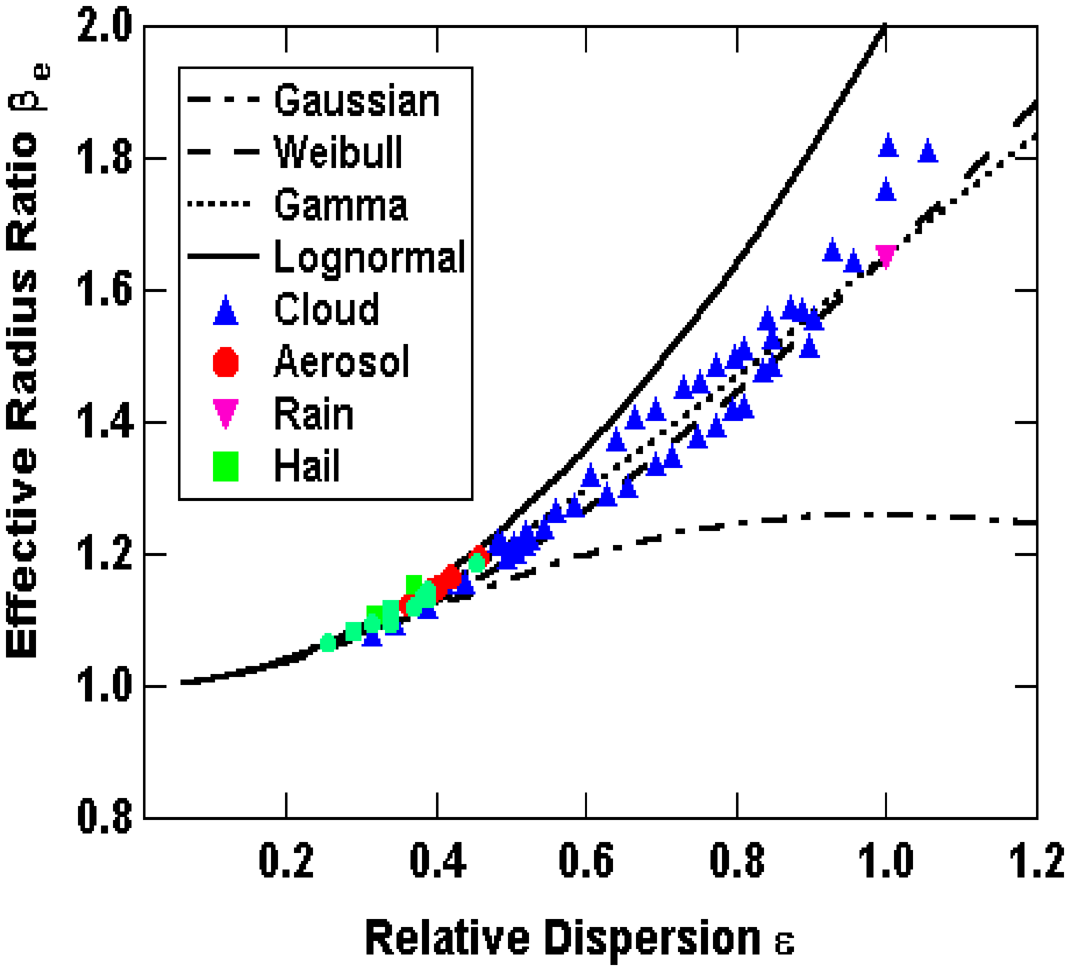
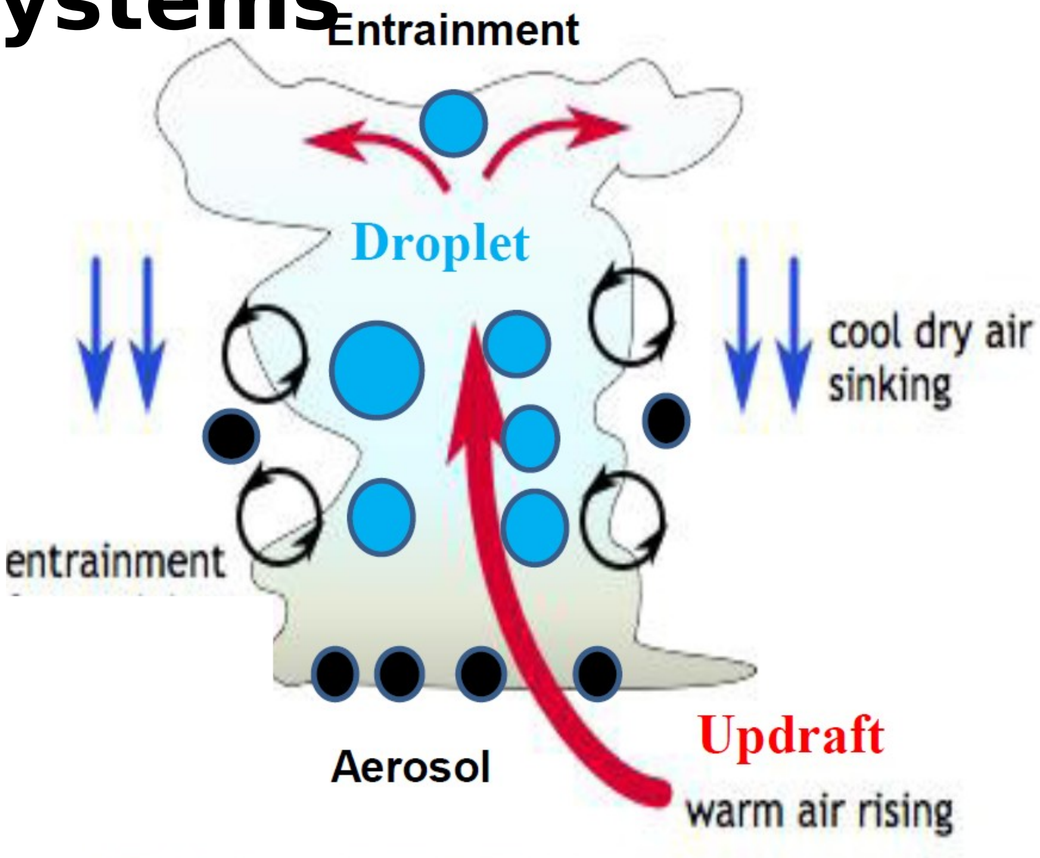


Aerosol, cloud droplet and precipitation particles share a common distribution form ---- Weibull or Gamma, suggesting a **unified theory on particle size distributions**. Talk to me about rain

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.

Lu et al (2011, 2012, 2013, 2014, 2016; Yum et al., 2015)

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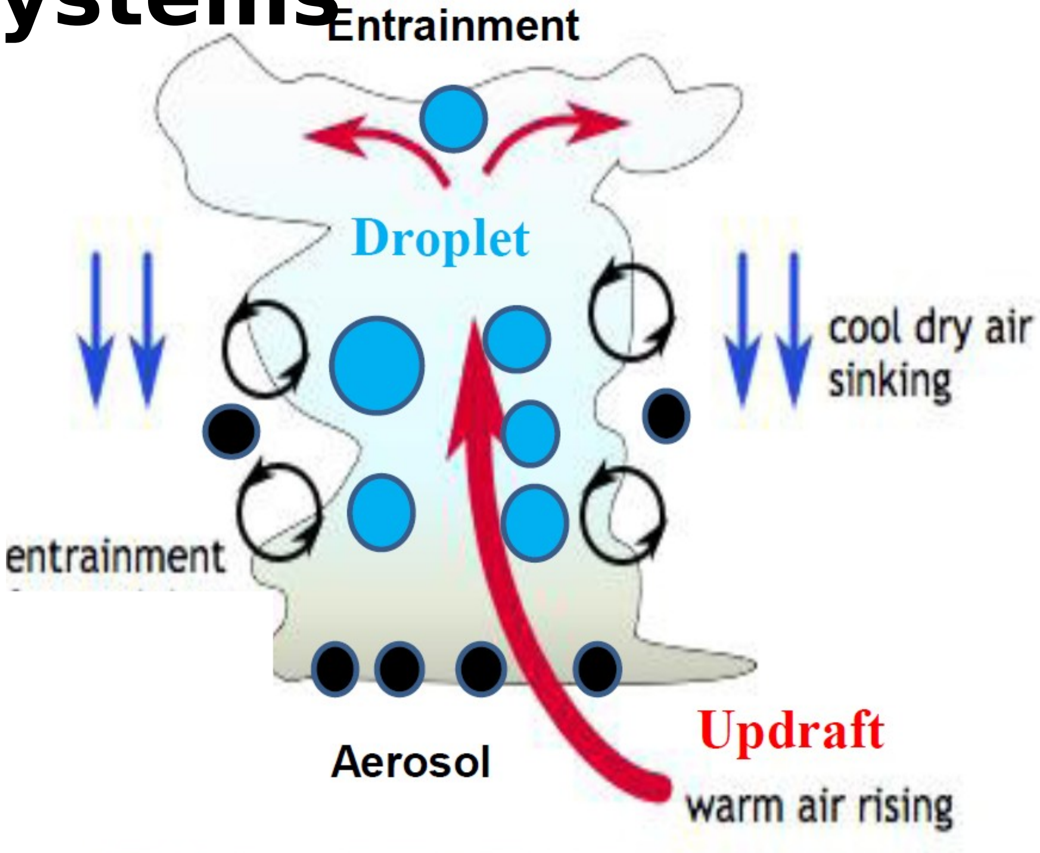
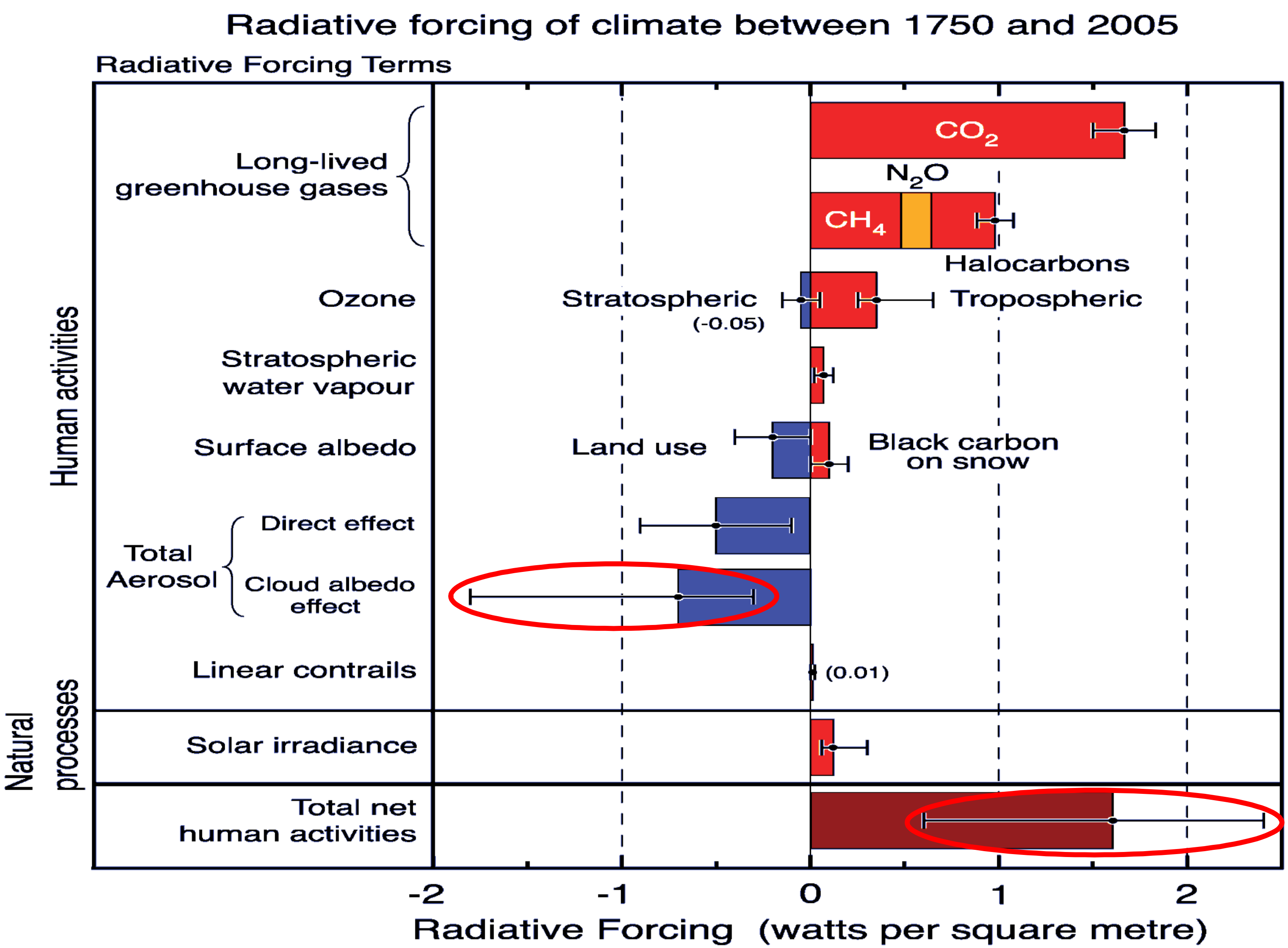


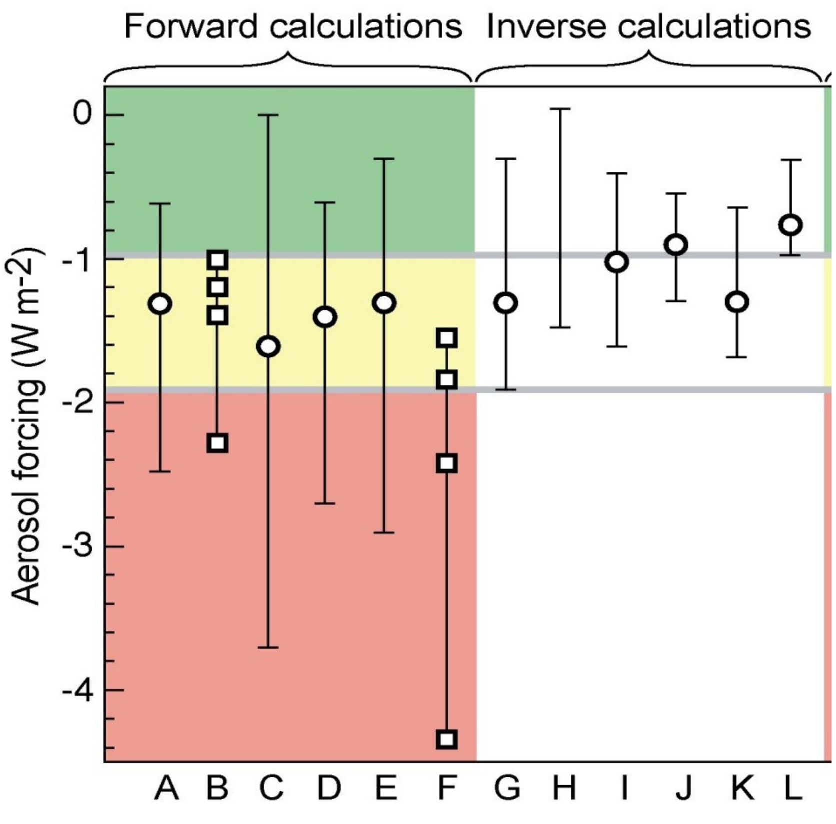
Turbulence, related entrainment-mixing processes, and their interactions with microphysics are key to the outstanding puzzles.

Lu et al (2011, 2012, 2013, 2014, 2016; Yum et al., 2015)

Aerosol indirect effects constitute the major uncertainty in climate forcing

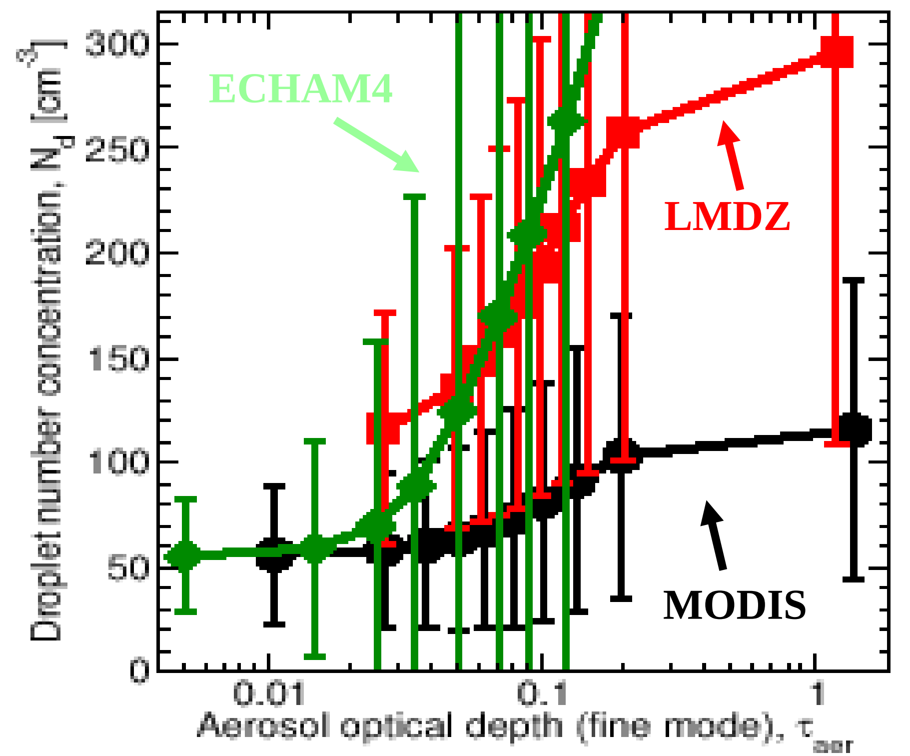


Forward GCM AIE estimates suffer from big uncertainty and discrepancy !



(Adapted from Anderson et al., Science, 2003)

Model Parameterizations vs. Satellite Results

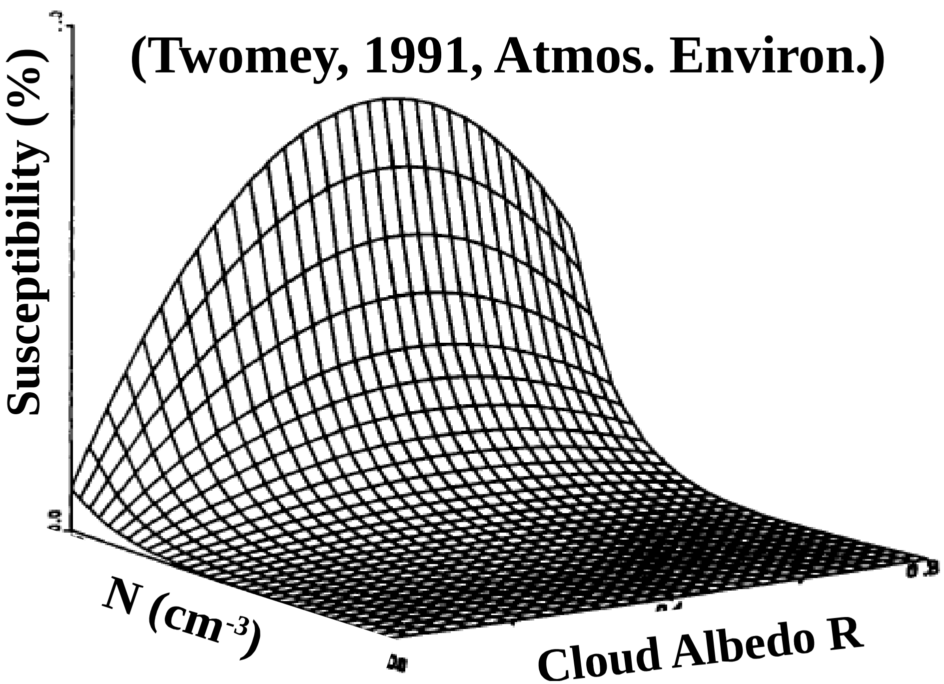


(Adapted from Quaas et al., ACP, 2006)

Forward GCM estimates are as good as the cloud parameterization used in GCMs, and the cloud parameterization poses a major problem to climate models (another driver of my research).

Twomey (Number) Effect

Twomey (1974, Atmos. Environ): "it is suggested that pollution gives rise to whiter (not darker) clouds ----- by <u>increasing the droplet concentrations</u> and thereby the optical thickness (and cloud albedo) of clouds."



(Twomey, 1991, Atmos. Environ.)

Cloud Susceptibility:

$$S \equiv \left(\frac{dR}{dN}\right)_x = \frac{R(1-R)}{3N}$$

- **R = Cloud albedo**
- **N = Droplet Concentration**
- **Explicit x: constant LWC**
- **Implicit x ?**

Later work links R (or other cloud properties) with aerosols using a relationship of N to aerosol loading (e.g., $N \approx \tau_a^{0.7}$ $N_a^{0.7}$, Kaufman and Fraser, 1997, Science).

Effective radius and Its Parameterization

- **Hansen & Travis (1974, Space Sci. Rev) introduced effective radius r_e to describe light scattering by a cloud of particles**

$$r_e = \frac{\int r^3 n(r) dr}{\int r^2 n(r) dr}$$

- **r_e is further parameterized as** $r_e = \beta \left(\frac{3}{4\pi\rho_w} \right)^{1/3} \left(\frac{L}{N} \right)^{1/3}$

Unrealistic assumptions in most GCMs:

- **β has been implicitly assumed to be a constant (only N effect)**
- **Clouds are adiabatic**

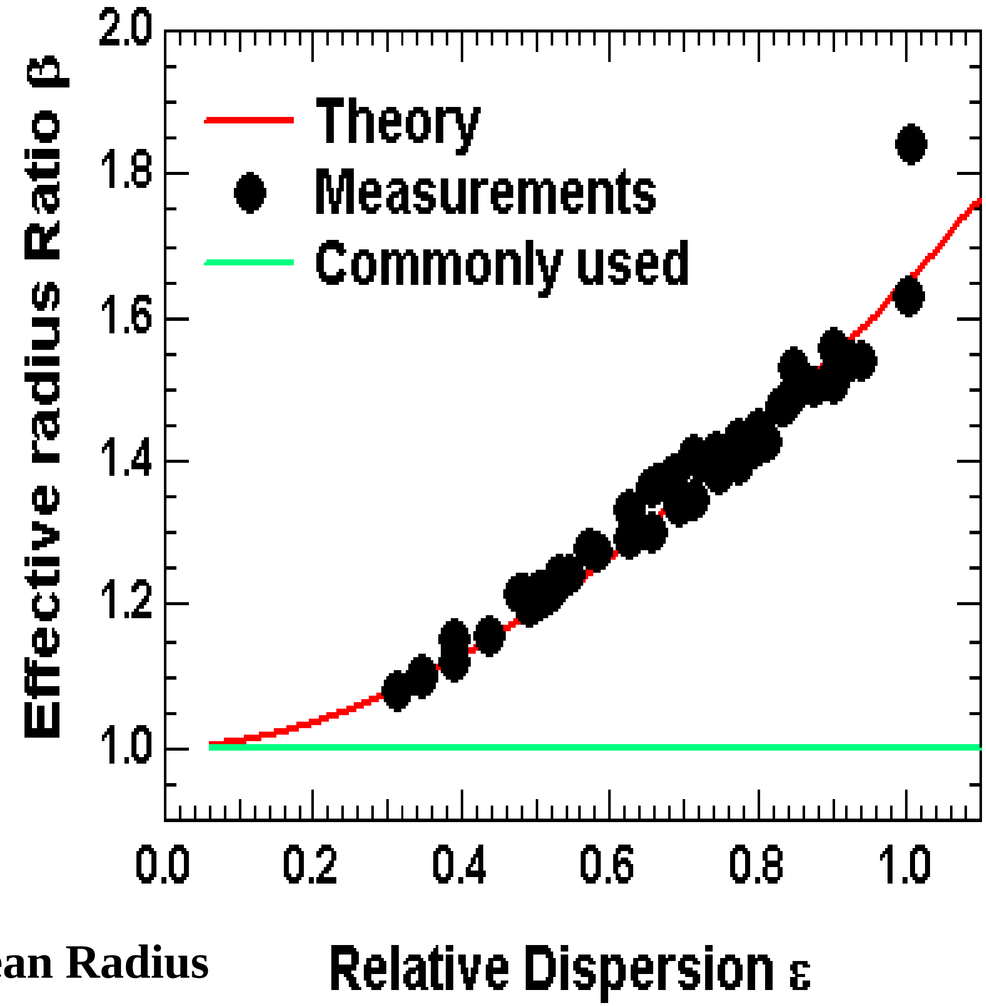
β in terms of Relative Dispersion

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

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

$$\varepsilon = \frac{\sigma}{\bar{r}}$$

ε = Standard Deviation/Mean Radius



Effective radius ratio β is an increasing function of relative dispersion.

Further improving μ-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)

….

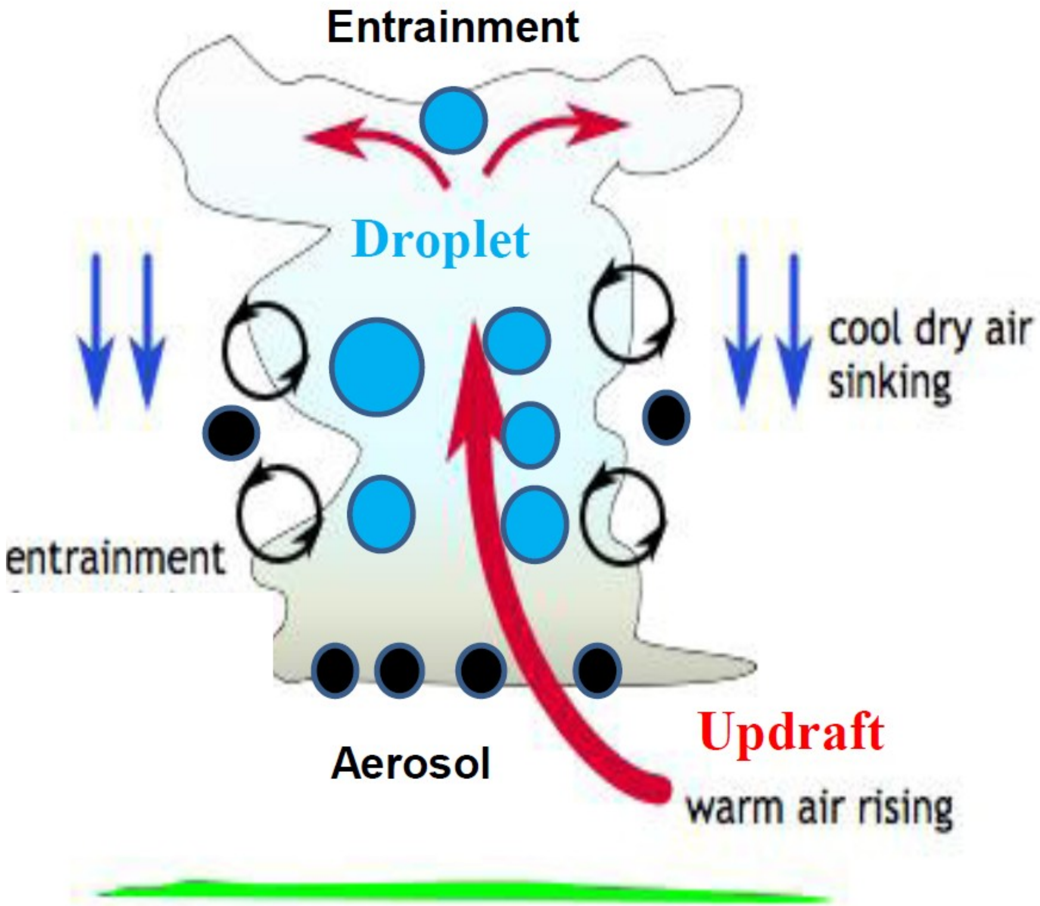
Spectral Broadening

Rain Initiation

Cloud Physics

Shallow Cumulus as an Open Multi-Physics System

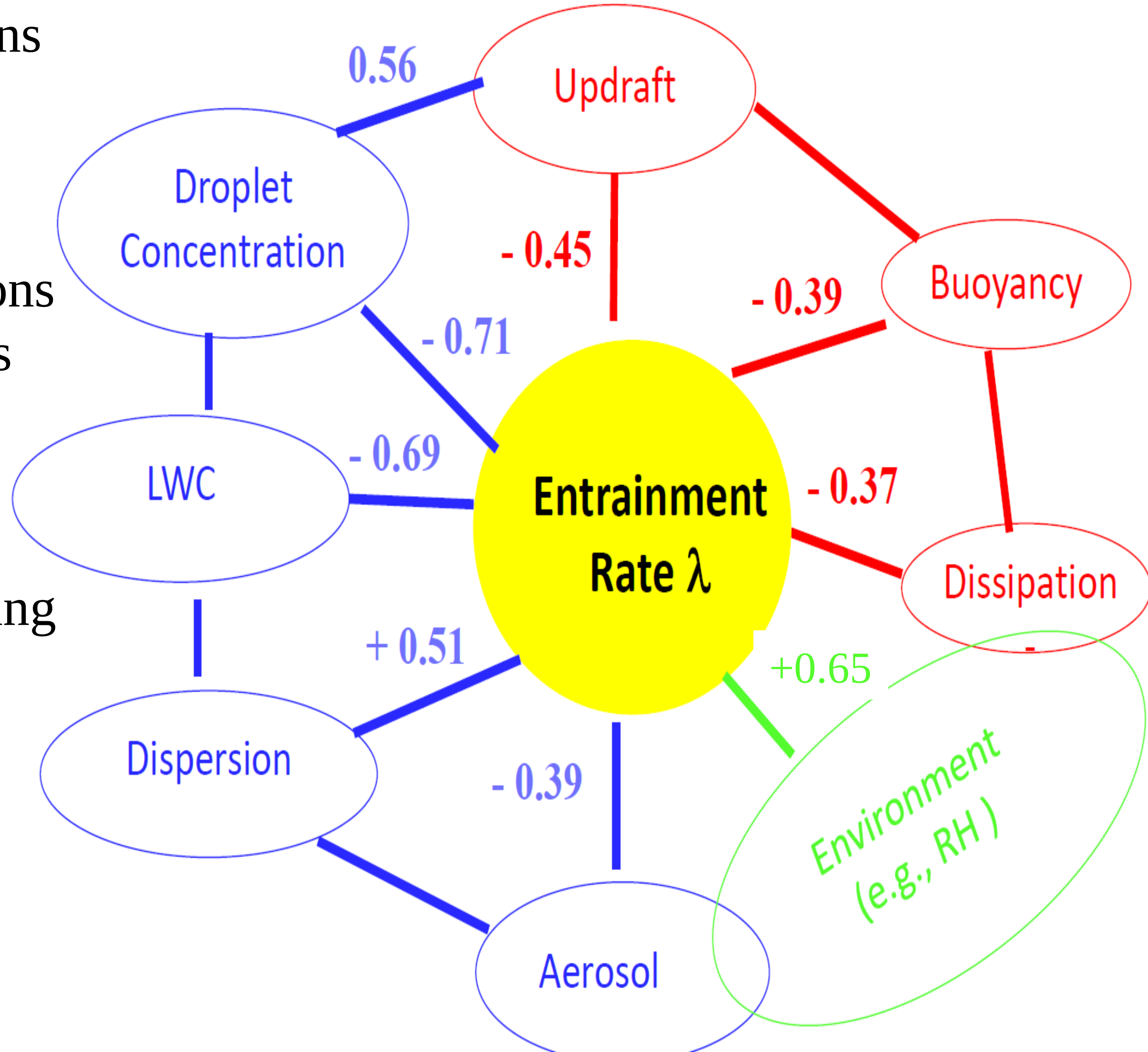
- **Entrainment Rate**
- **Vertical velocity**
- **Buoyancy**
- **Dissipation**
- **Environment**
- **Turbulent mixing**
- **Microphysics**
- **Aerosol**
- **Couplings**



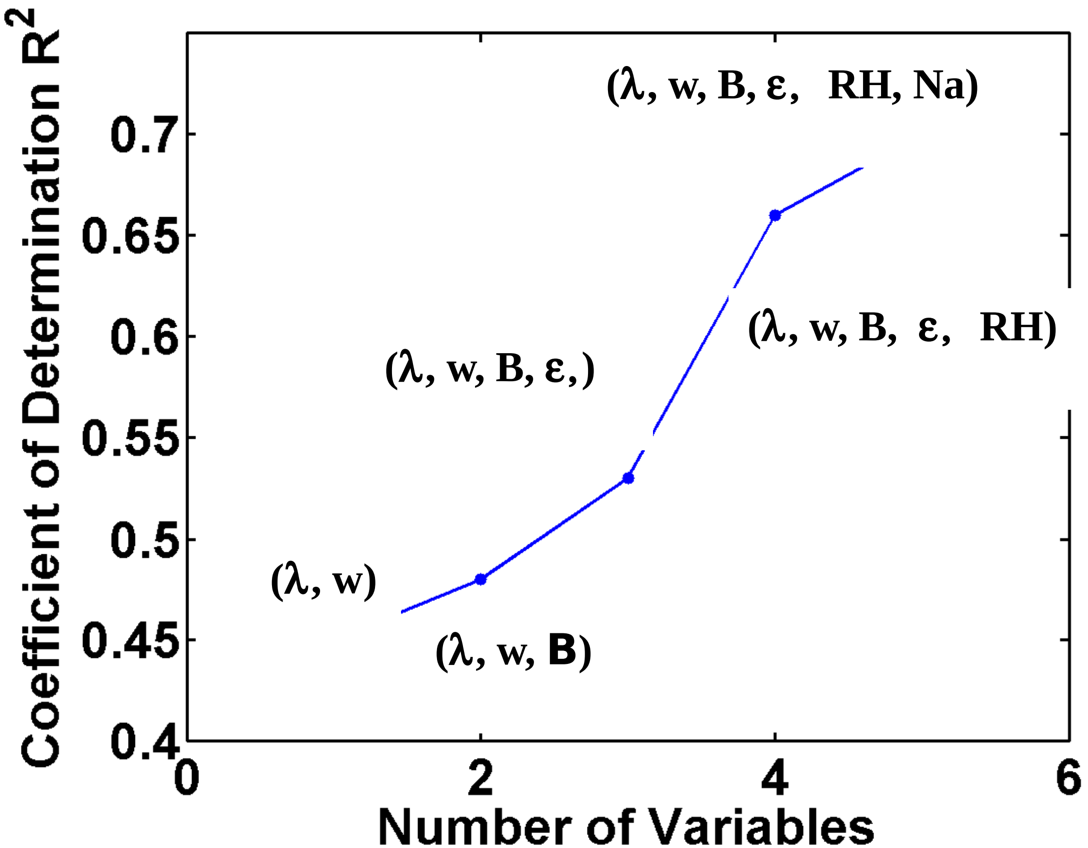
<u>Approach:</u> examine the relationships among these key variables in clouds (e.g., growing shallow cu) utilizing observations & models

Complex Coupling Web

- Similar correlations with dynamics & aerosols
- Similar correlations with microphysics & RH
- Consistent with homogenous mixing in updraft-limited regime
- Couplings reduce AIE as currently parameterized



Stepwise PCA Regression Confirms Similar Significance to Represent Entrainment rate



The unexplained variability is likely due to microphysical feedbacks on entrainment (work in progress)

Take-Home Messages

- **Potentials of statistical physics (systems theory) as a theoretical foundation for microphysics parameterizations**

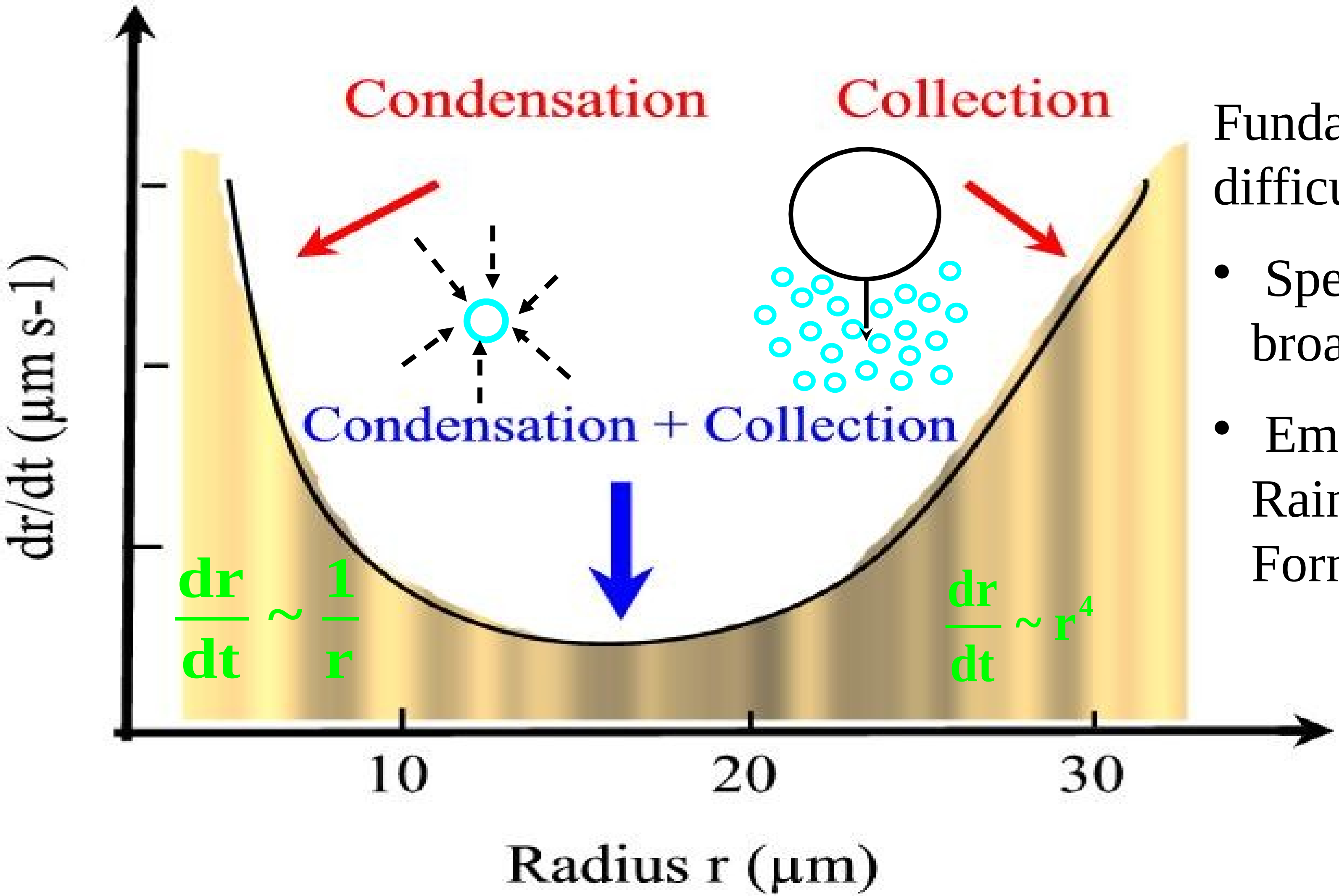
- **Potentials of unified parameterization for all turbulent entrainment-mixing processes**

- **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**

Valley of Death and Drizzle Initiation

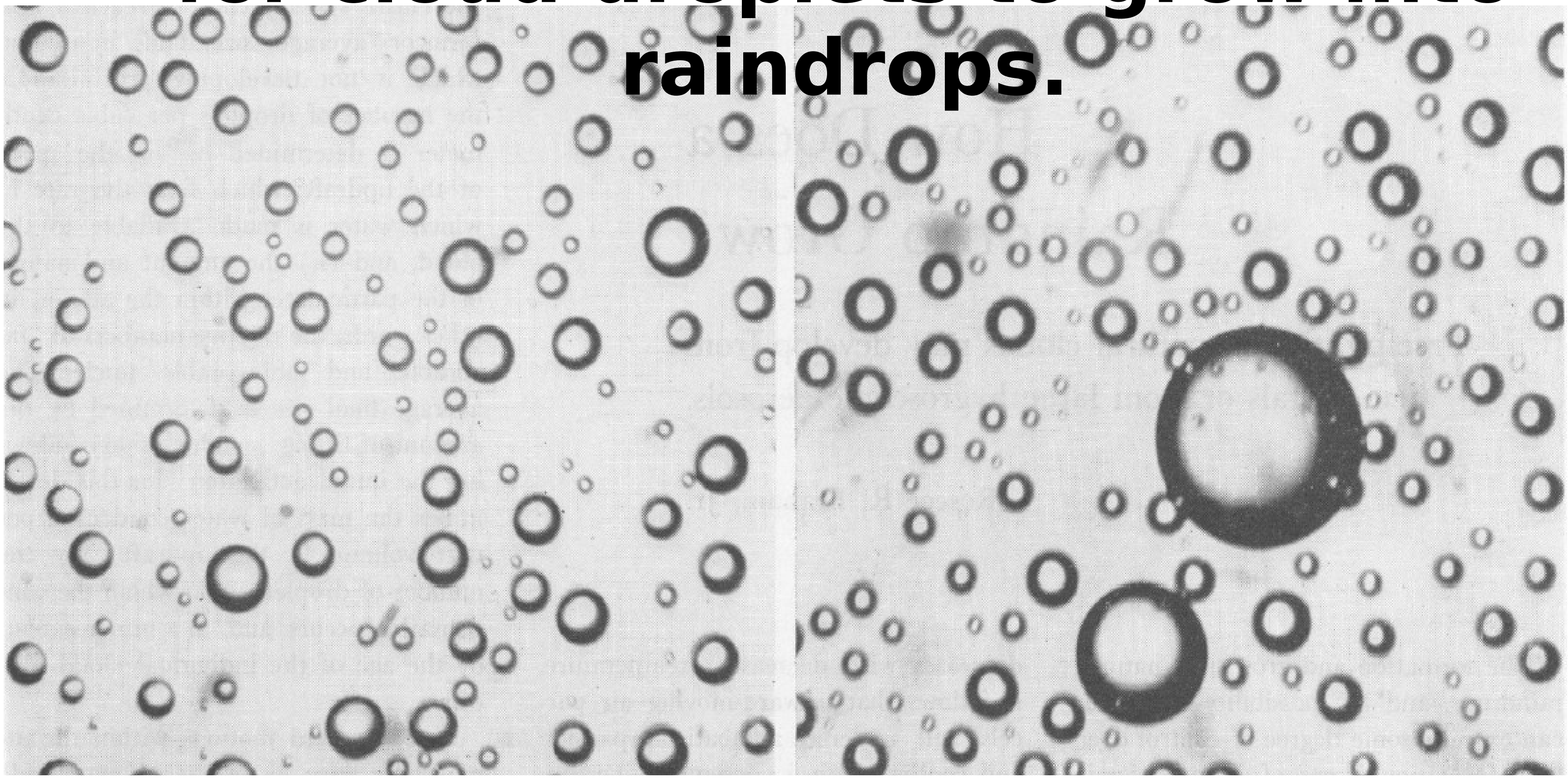


Fundamental difficulties:

- Spectral broadening
- Embryonic Raindrop Formation

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.

Autoconversion process is the 1st step for cloud droplets to grow into raindrops.



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

- 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)

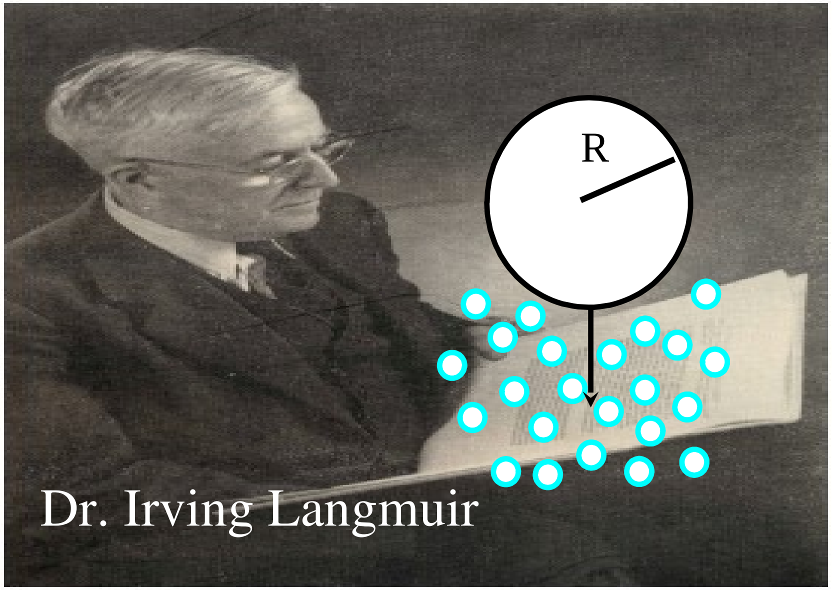
- Previous studies have been primarily on P_0 and existing parameterizations can be classified into three types according to their ad hoc T:
  * Kessler-type (T = Heaviside step function)
  * Berry-type (T = 1, without threshold function)
  * Sundqvist-type (T = Exponential-like function)

- 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

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

The rate function P_0 is then given by

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

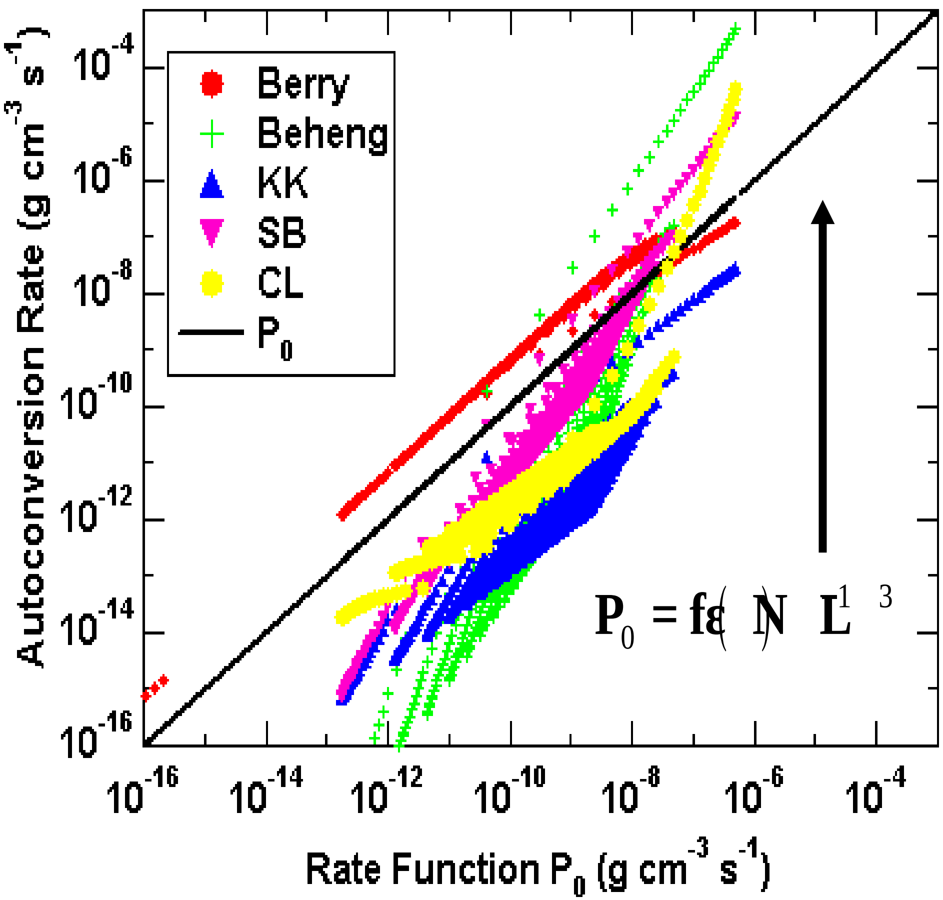
Generalized mean value theorem for integrals:

$$\int f(x)\,g(x)dx = f(x_0)\int g(x)dx$$

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
zations



- 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_0 or T (hence P) in $P = P_0T$.

The rate function P_0 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

$P = P_0 H(r_d - r_c)$

	Expression	Assumption	Features
Previous	$\mathbf{P} = \gamma \mathbf{N}^{-1/3} \mathbf{L}^{7/3} \mathbf{H}(\mathbf{r}_3 - \mathbf{r}_c)$	**Fixed collection efficiency**	**Fixed γ, no ε effect, $r_d = r_3$**
New	$\mathbf{P}_{LD} = \mathbf{f}(\varepsilon) \mathbf{N}^{-1} \mathbf{L}^{3} \mathbf{H}(\mathbf{r}_6 - \mathbf{r}_c)$	**Realistic collection efficiency**	**Has ε, stronger dependence on L and N, $r_d = r_6$**

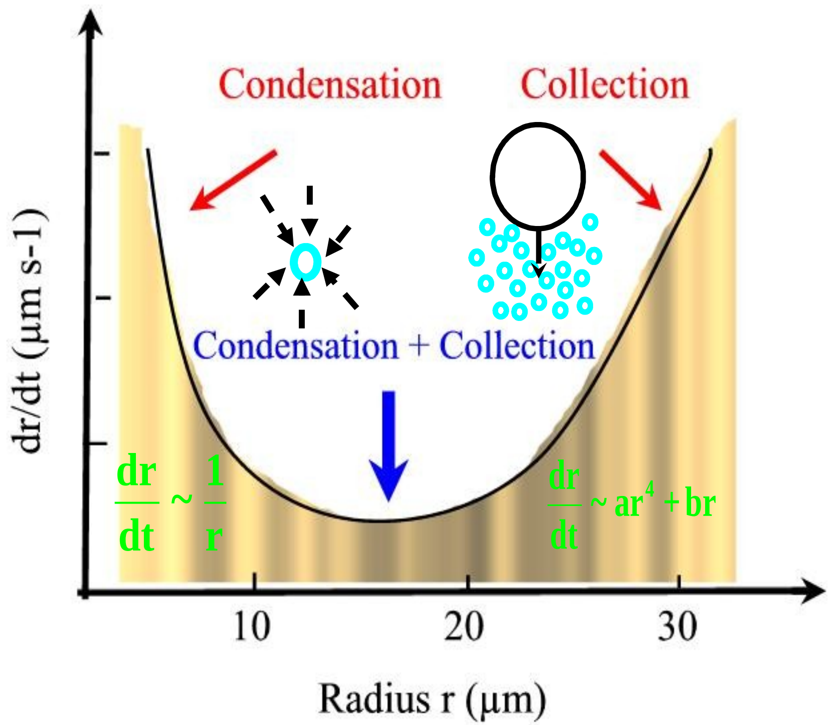
r_3 = 3rd moment mean radius; r_6 = 6th moment mean radius

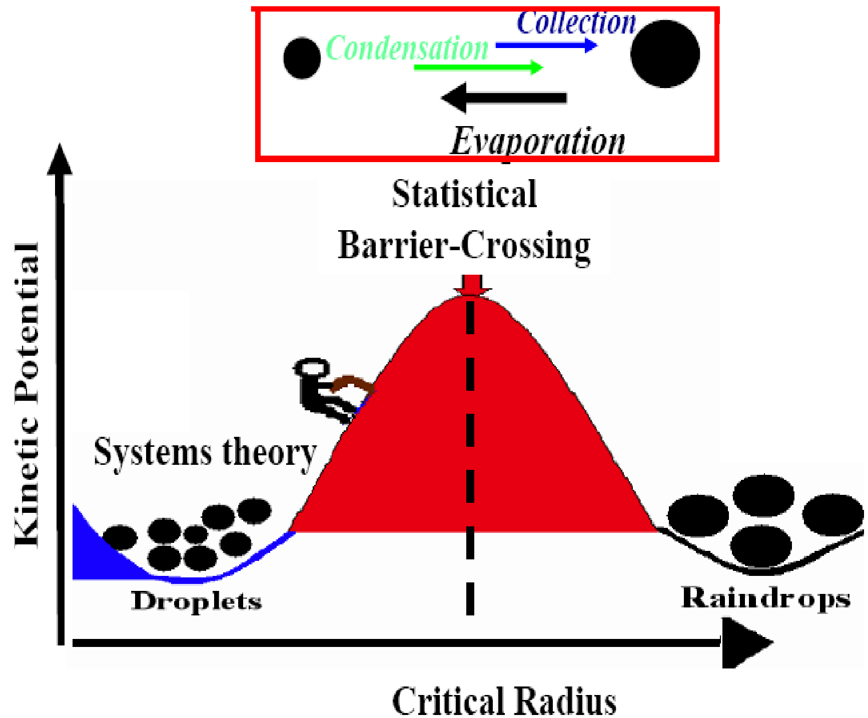
H = Heaviside step function (Liu & Daum 2004, JAS).

What about the critical radius >> rain initiation theory?

Systems Theory of Rain Initiation/Autoconversion

Valley of Death → **Mountain of Life**



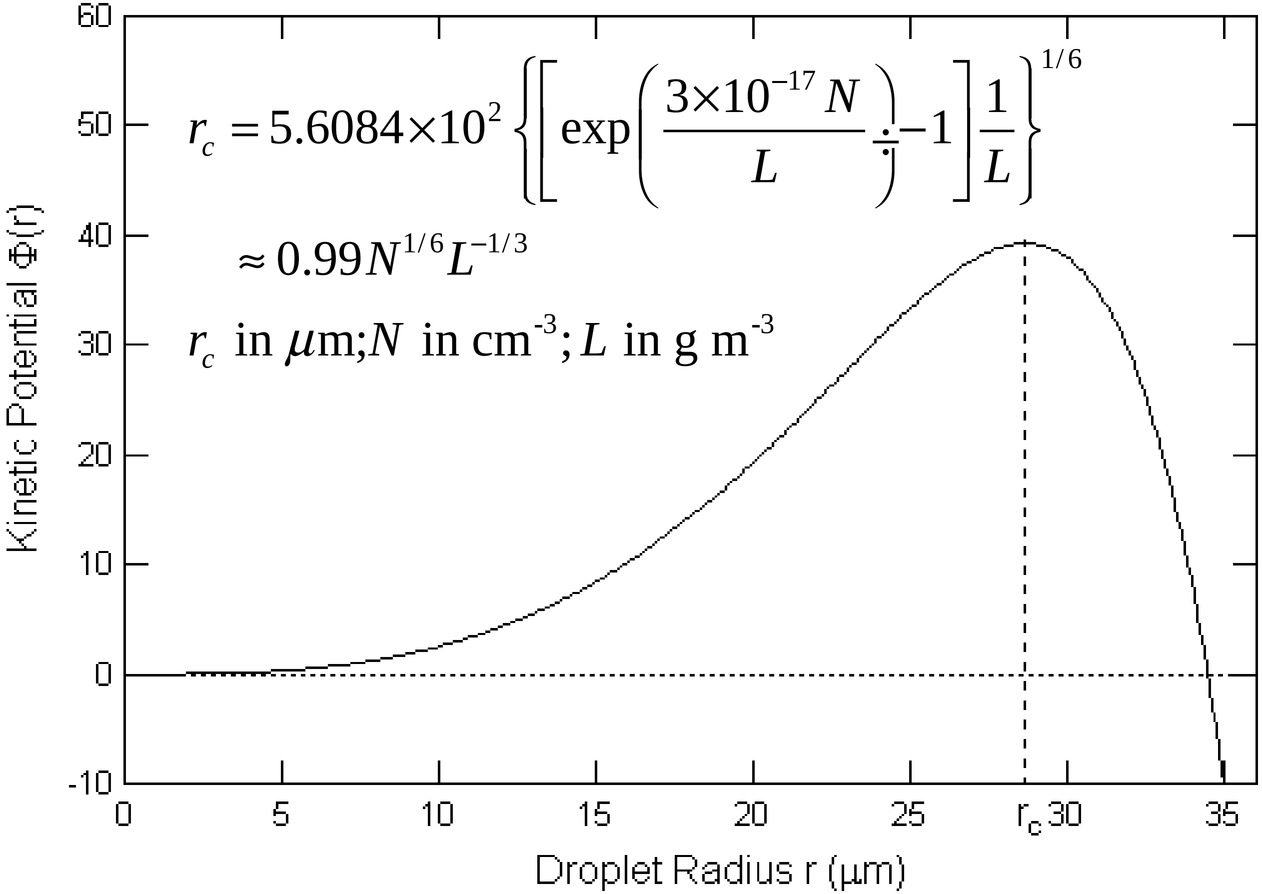


Rain initiation has been an outstanding puzzle with two fundamental problems of spectral broadening & formation of embryonic raindrop

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

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

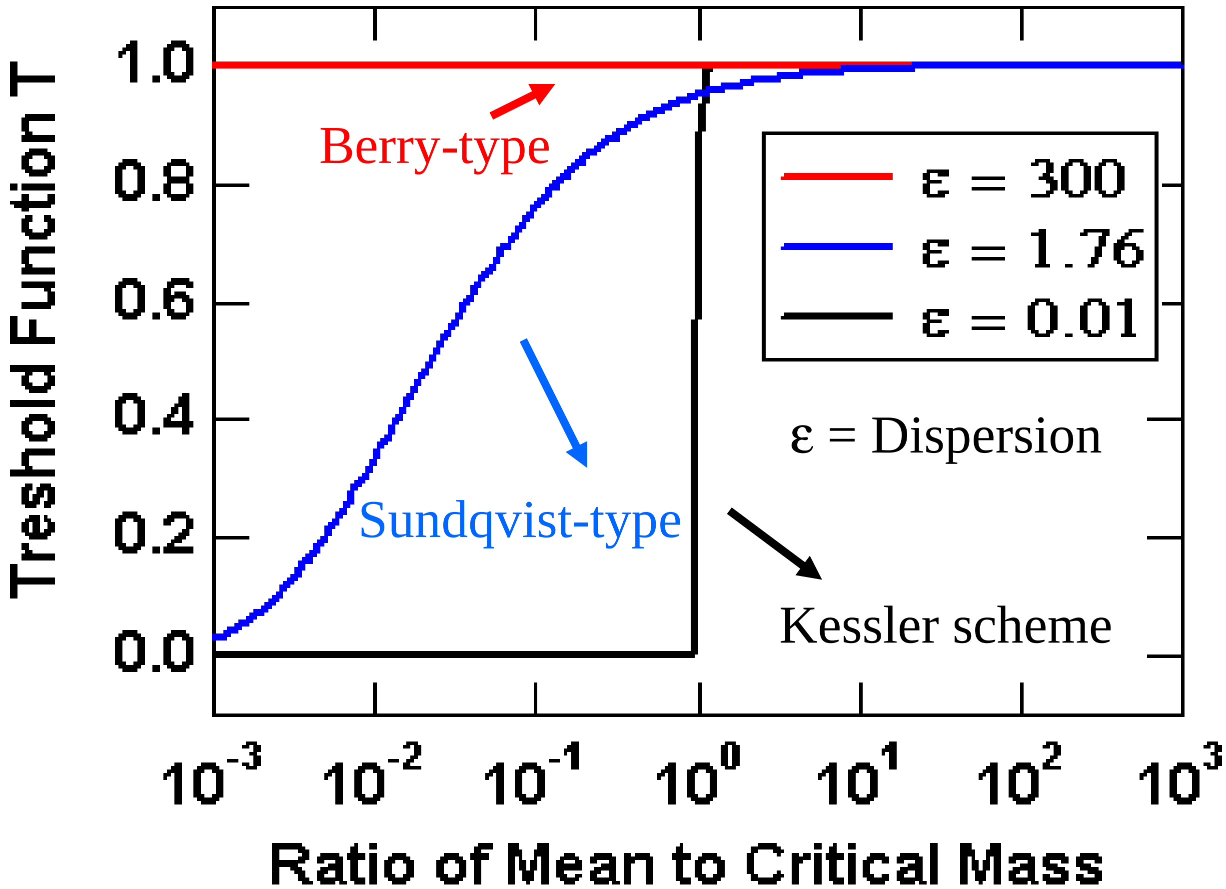


- Kinetic potential peaks at critical radius rc.
- Critical radius & potential barrier both increase with droplet concentration.

- 2nd AIE: Increasing aerosols inhibit rain by enhancing the barrier and critical radius.

Critical radius i 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



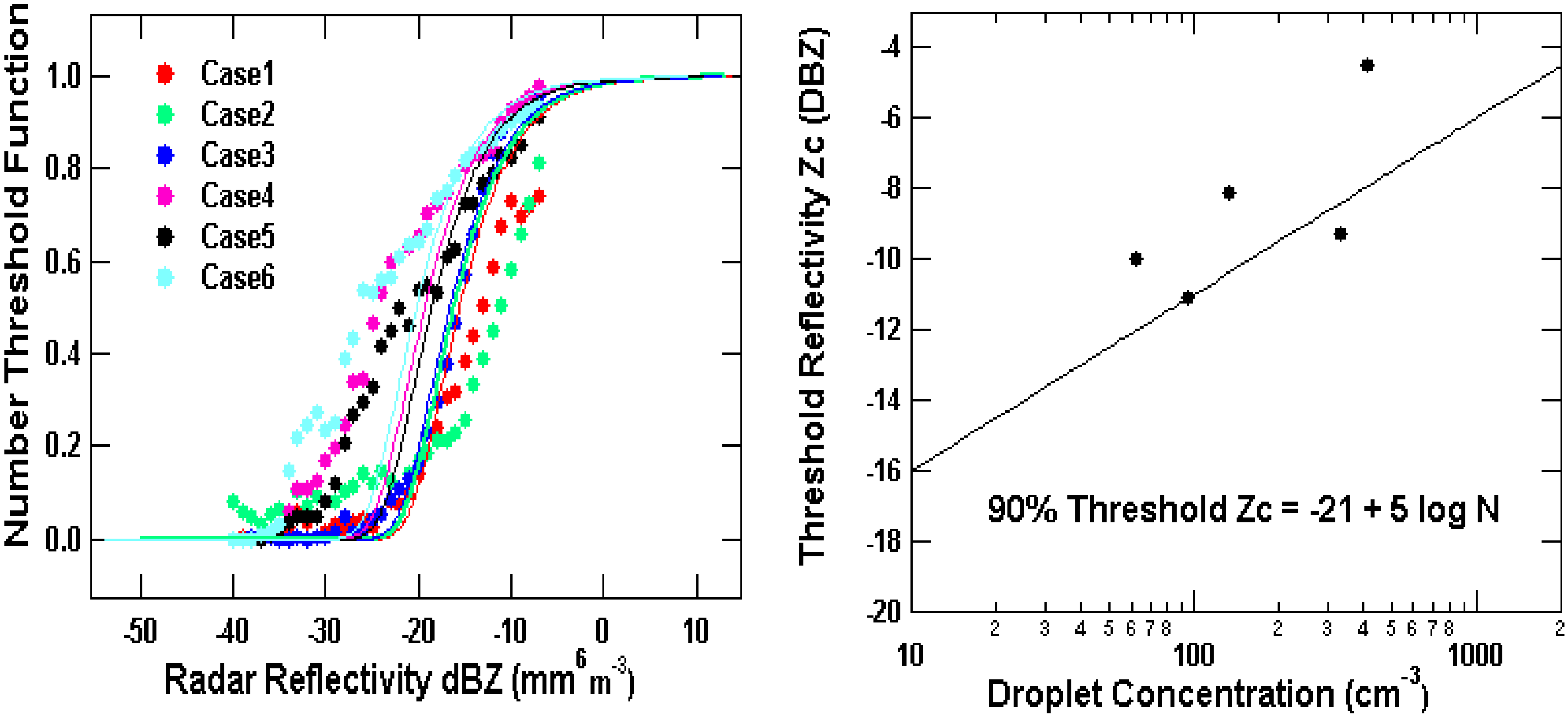
Truncating the cloud droplet size distribution at critical radius yields the threshold function:

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

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

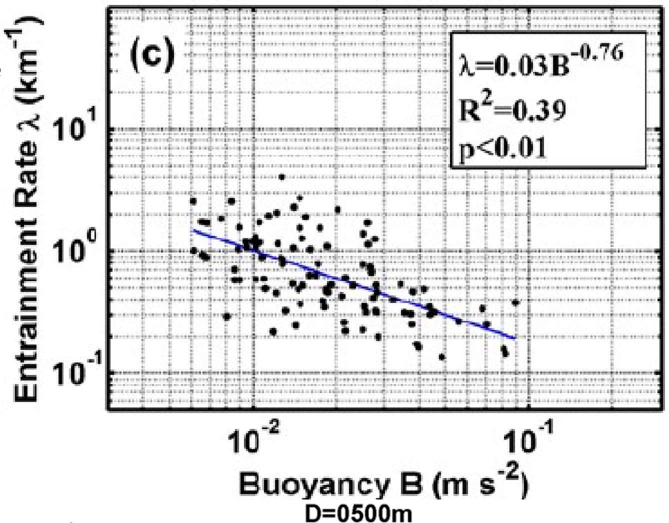
The new threshold function unifies existing ad hoc types of threshold functions, and reveals the important role of relative dispersion that has been unknowingly hidden in ad hoc threshold functions (Liu et al., GRL, 2005, 2006, 2007).

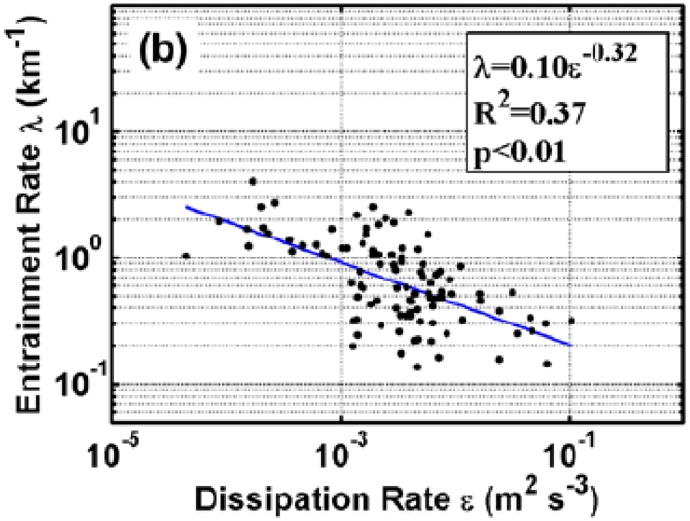
Observational Validation of Threshold Function

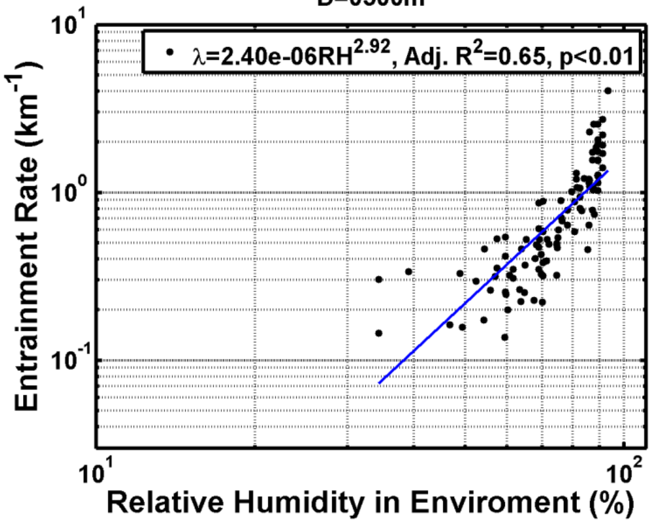


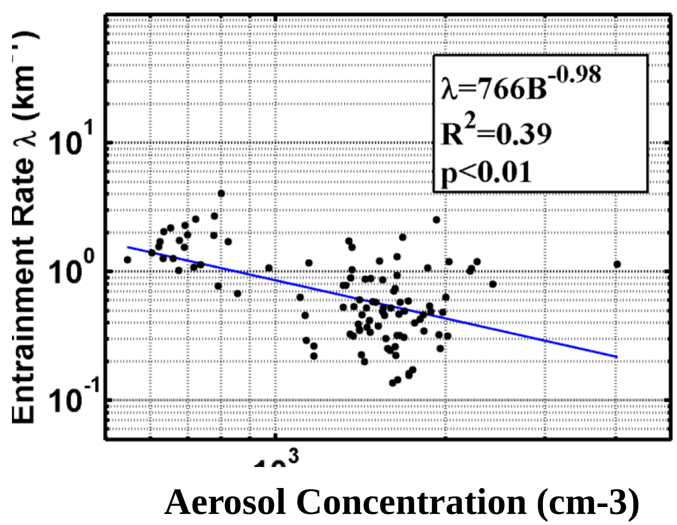
The results explain why empirically determined threshold reflectivity varies, provides observational validation for our theory, and additional support for the notion that aerosol-influenced clouds tend to hold more water or a larger LWP (Liu et al., GRL, 2007, 2008).

More Pairwise Relationships









These results suggest that shallow cumulus is a system in which variables are related to one another, *but only weakly, with ALL pair correlations < 0.5*.

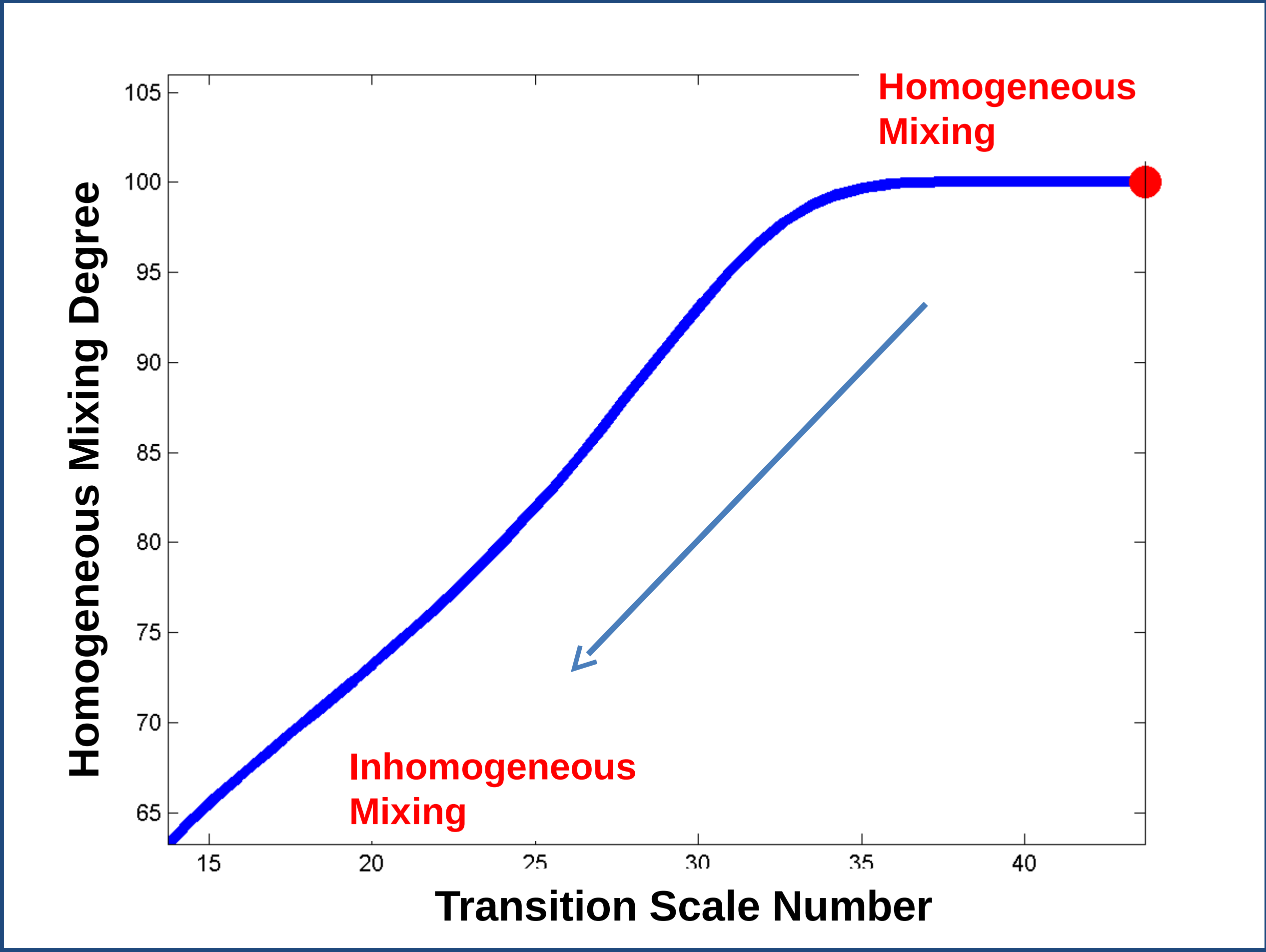
Entrainment-Mixing Processes in P-DNS: Animation

- 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.

-



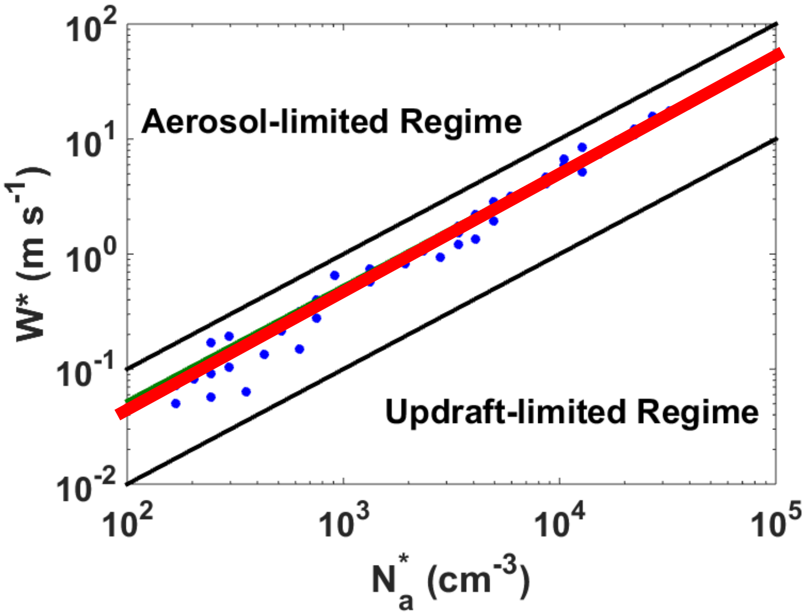
Droplets start with homogeneous mixing and evolve toward inhomogeneous mixing due to faster evaporation relative to turbulent mixing.

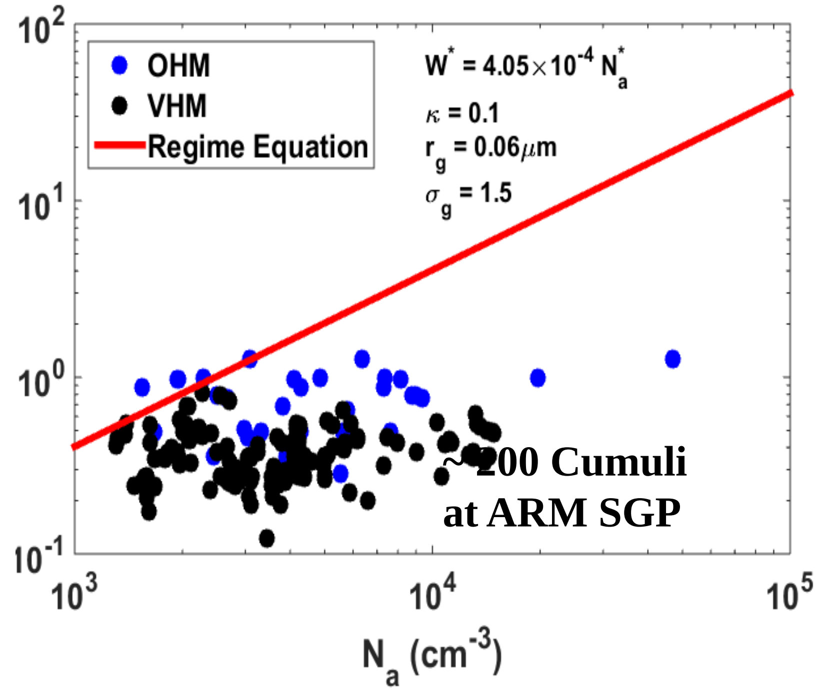
Ongoing and Future Work

- Examine causal relationships
- Develop coupled parameterization

Thanks for your attention!

New Equation for Regime Classification

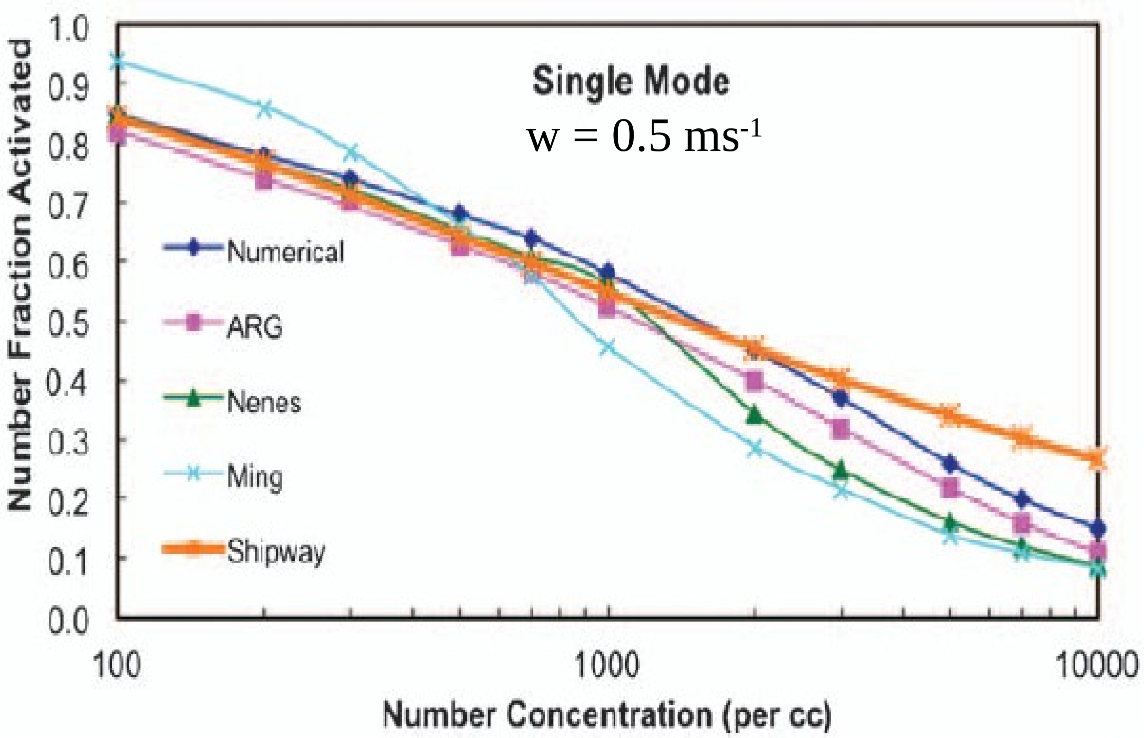




The regime equation can be applied to determine global distribution of AIE regimes, which calls for concurrently measuring/representing aerosols and <u>updraft velocity</u>.

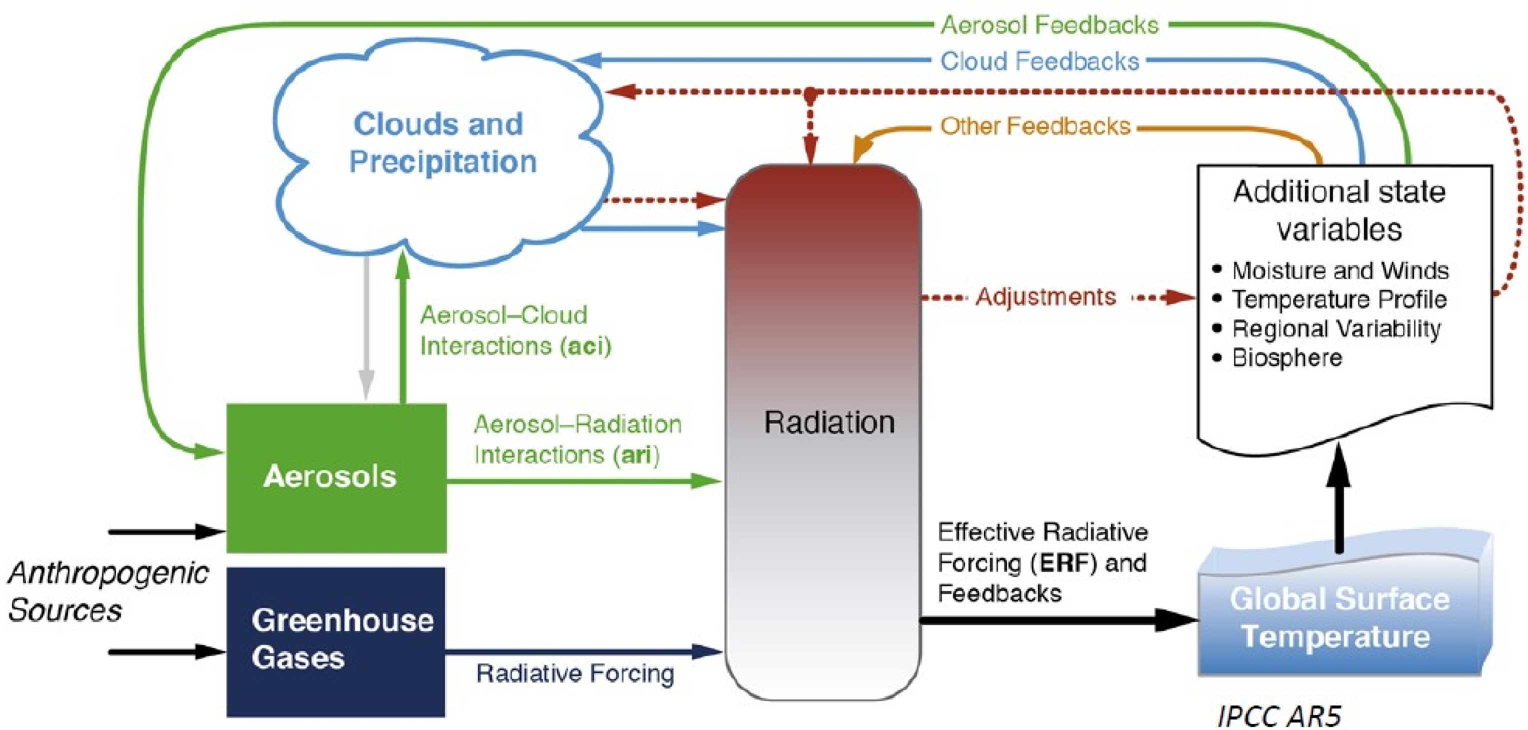
Parameterization for Droplet Concentration

(Fig. 4. Ghan et al. JAMES, 2013)



- Best in transitional regime but worst in the updraft-limited regime.
- No dispersion parameterization for updraft-limited regime yet.

Aerosols, Clouds and Climate

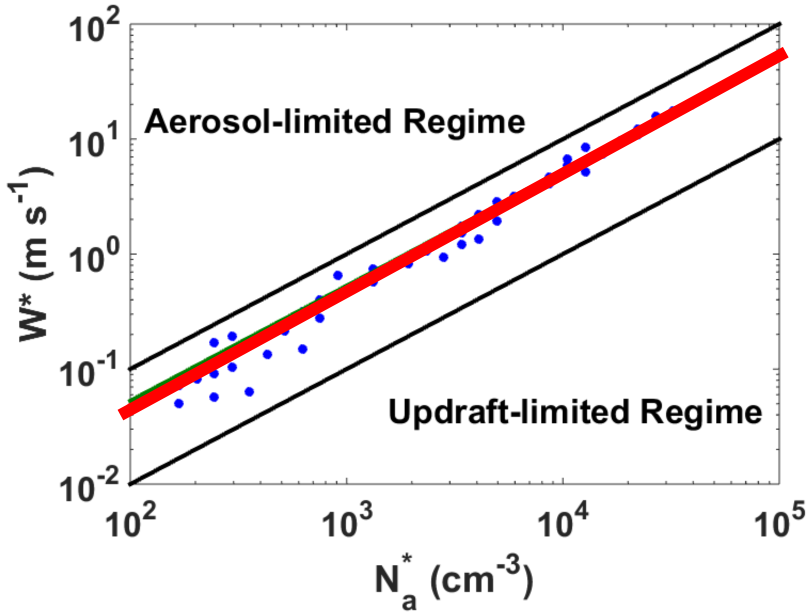


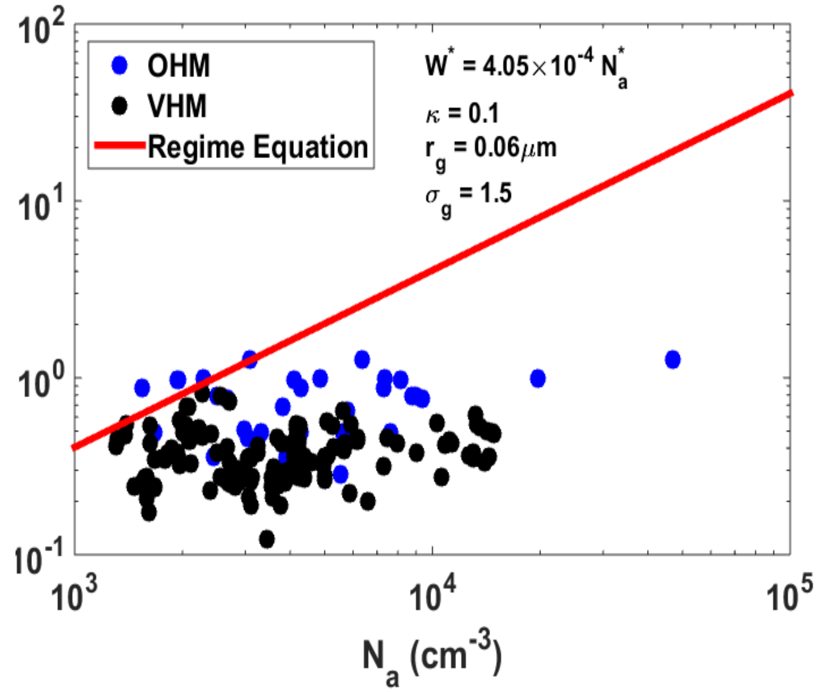
IPCC AR5
Boucher, Randall et al. 2014

"We propose that the difficulty in untangling relationships among the aerosol, clouds and precipitation reflects the inadequacy of existing tools and methodologies and a failure to account for processes that buffer cloud and precipitation responses to aerosol perturbations."

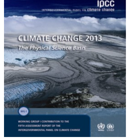
Stevens and Feingold, 2009, Nature

SGP Cu Most in Updraft-Limited Regime

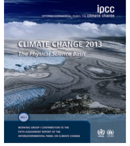




The regime equation can be applied to determine global distribution of AIE regimes, which calls for concurrently measuring/representing aerosols and <u>updraft velocity</u>.



Science Drivers



- **AIE estimates in climate models continue to suffer from large uncertainty & tend to be overestimated.**
- **Clouds in models may be oversensitive to aerosol perturbation, due to <u>buffering factors/processes</u> <u>that are either poorly represented or not at all (*Steven & Feingold, Nature, 2009*)</u>**

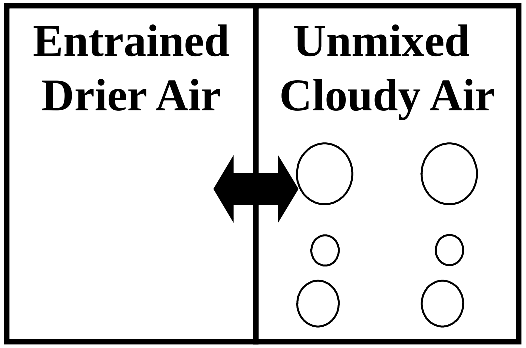
<u>Four Related Buffers:</u>

- *Dispersion effect*
- *Regime dependence*
- *Entrainment-mixing processes*
- *Couplings*

Dynamics: Damkoehler Number

- **Damkoehler number:**

$$Da = \tau_{\text{mix}} / \tau_{\text{react}}$$



- τ_{mix}: **the time needed for complete turbulent homogenization of an entrained parcel of size *L* (Baker et al., 1984):** $\tau_{\text{mix}} \sim (L^2/\xi)^{1/3}$

  ξ: dissipation rate

- τ_{react}: **the time needed for droplets to evaporate in the entrained dry air or the entrained dry air to saturate (Lehmann et al 2009):**

$$\tau_{phase} = \frac{1}{4\pi r D' n_d}$$

$$\begin{cases} \dfrac{dr_{\text{m}}}{dt} = -A \times \dfrac{s}{r_{\text{m}}} \\ \dfrac{ds}{dt} = -B \times s \end{cases}$$

r_{m}: mean radius

s: supersaturation

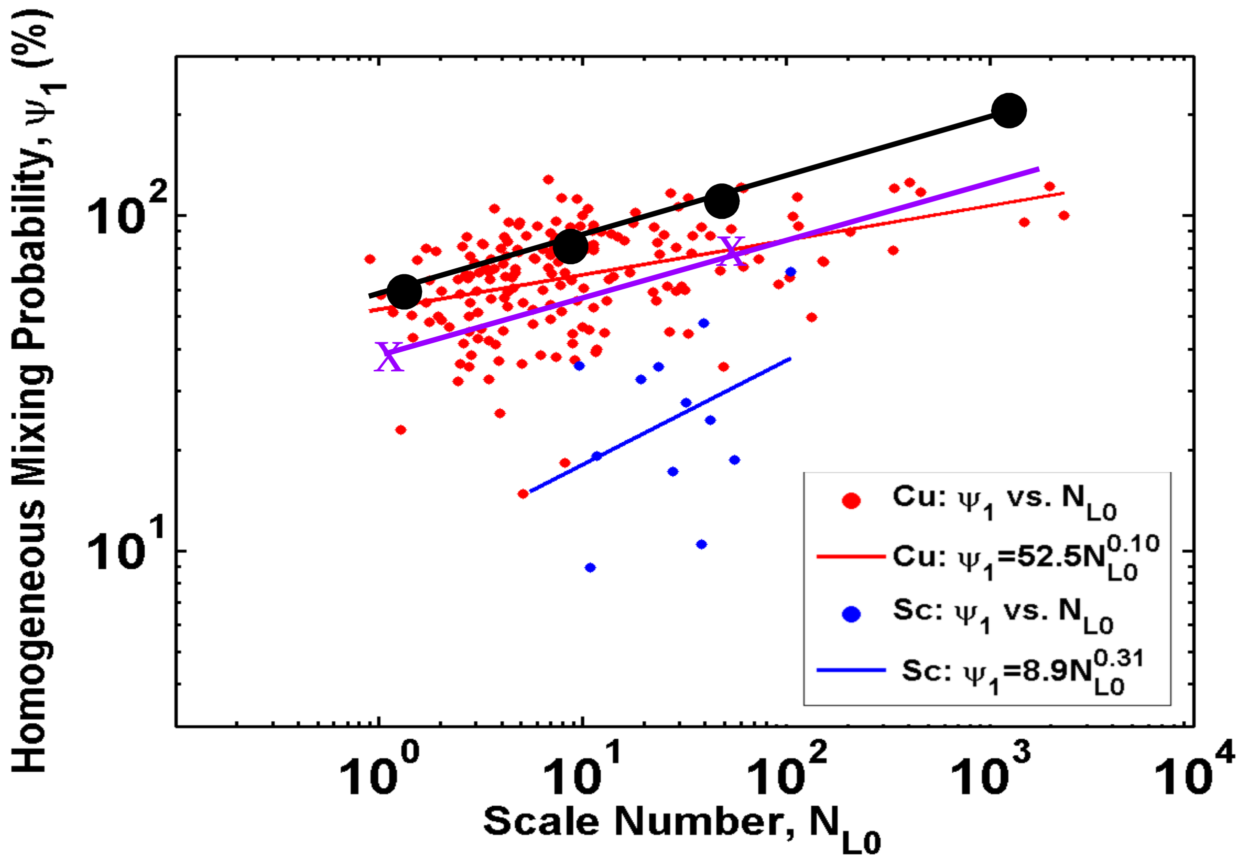
Parameterization for Mixing Mechanisms

- Eliminate the need for assuming mixing mechanisms

- Scale number can be calculated in models wit
2-moment microphysics

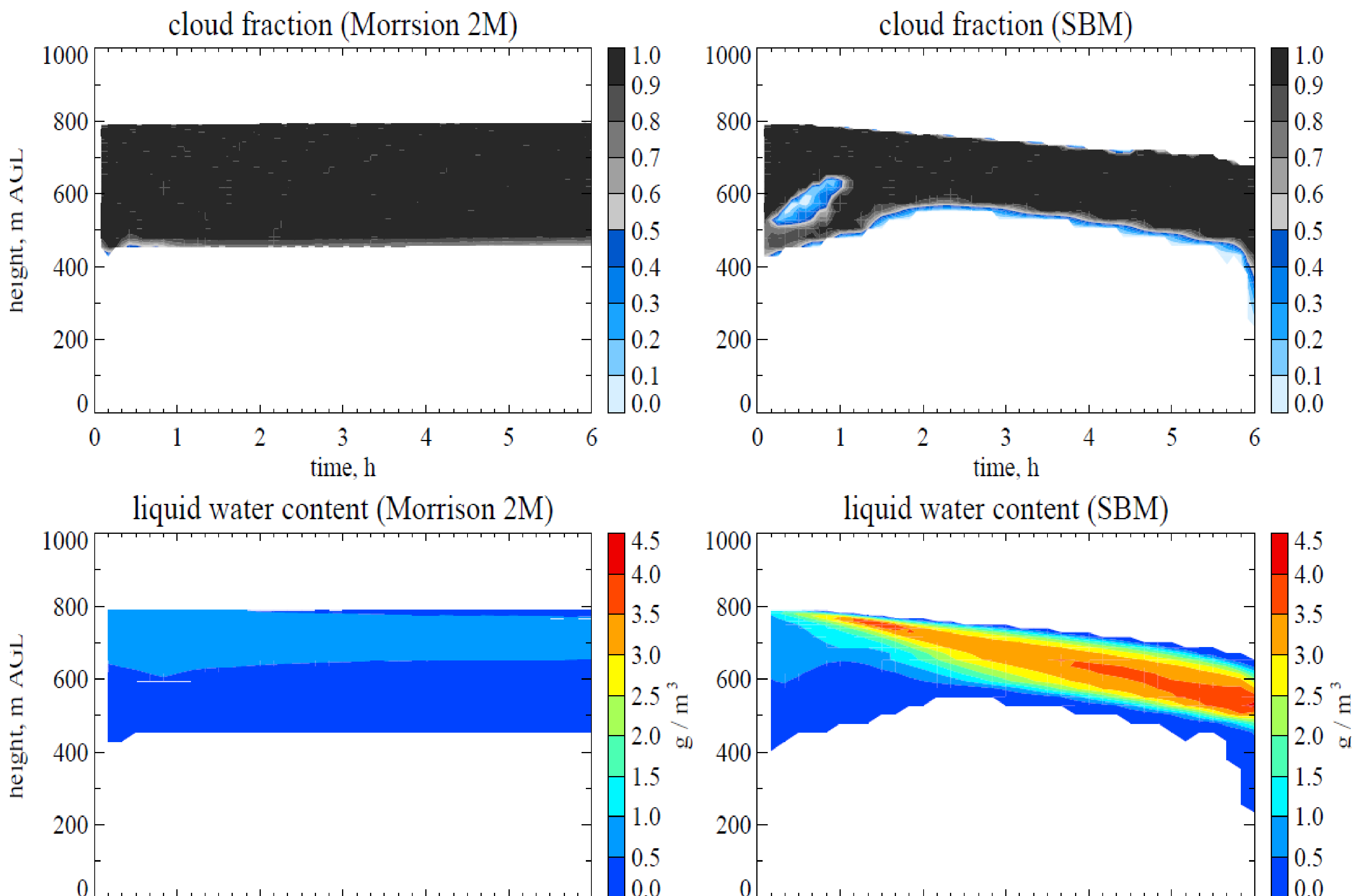
- Difference between Cu and Sc ?

- Evaluate, test, and improve

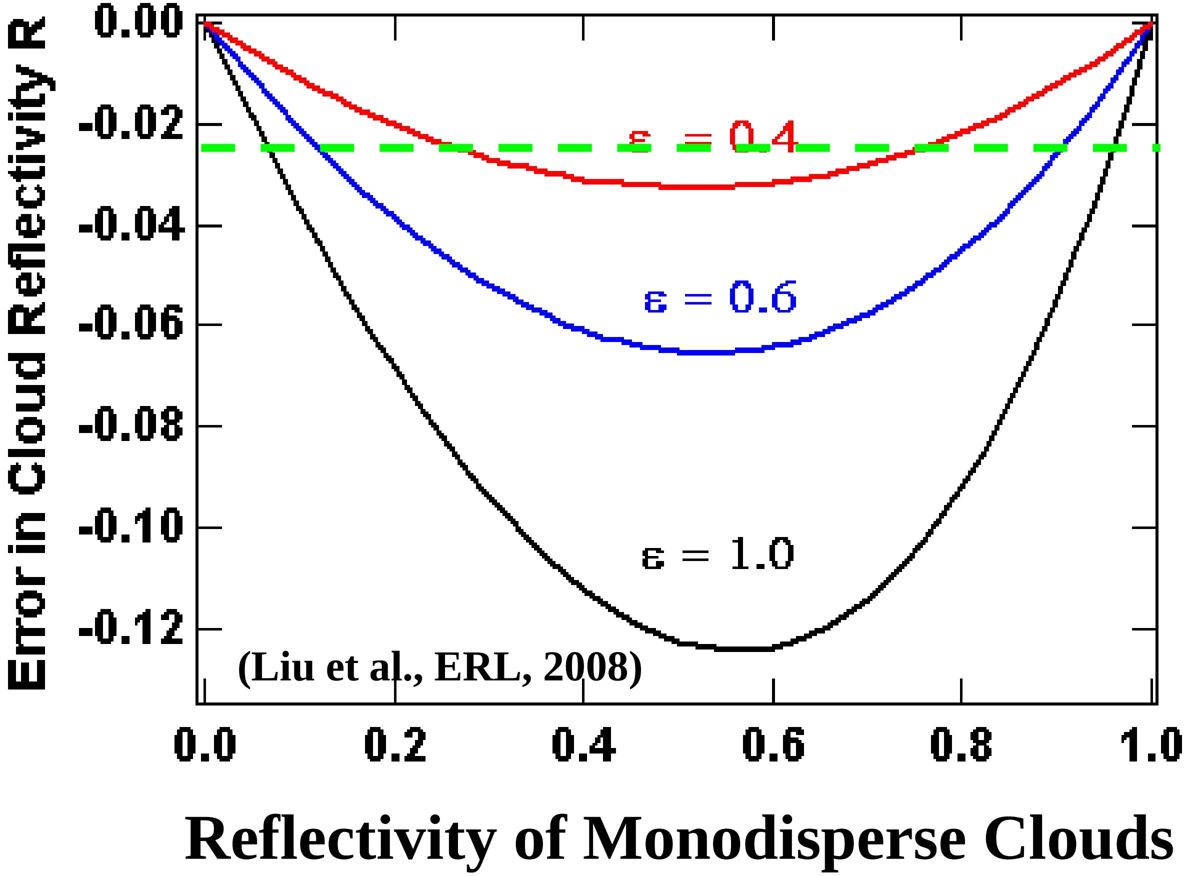


Combined with that for entrainment rate, we are exploring a parameterization that unifies entrainment-mixing-microphysics

Effect of Spectral Shape: Two Moment vs. SBM



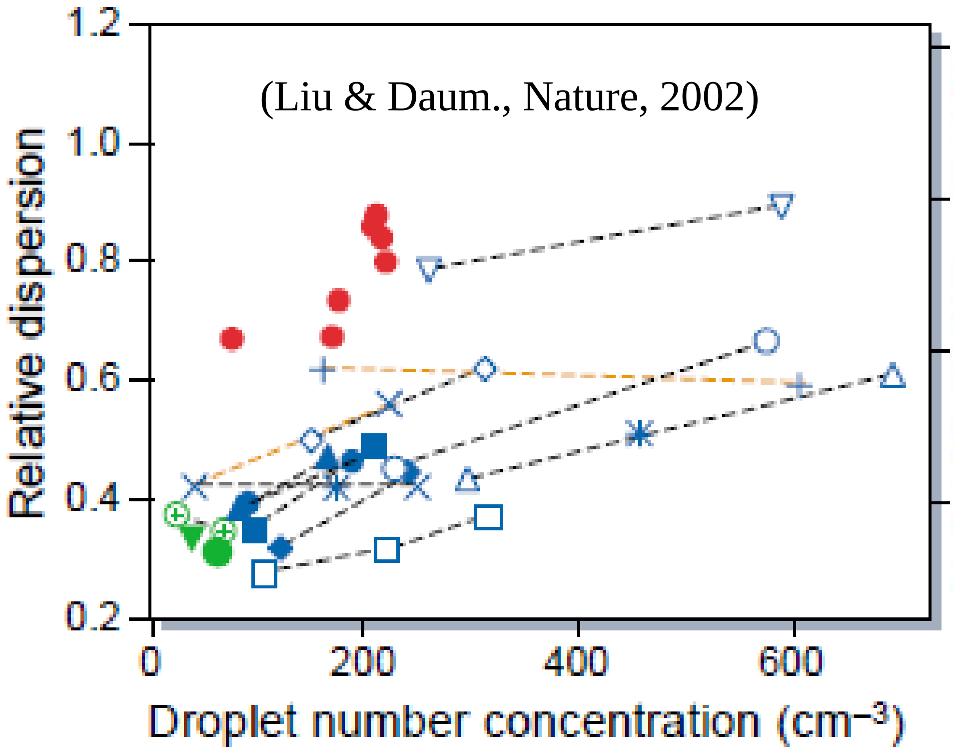
Neglection of dispersion significantly overestimates cloud reflectivity

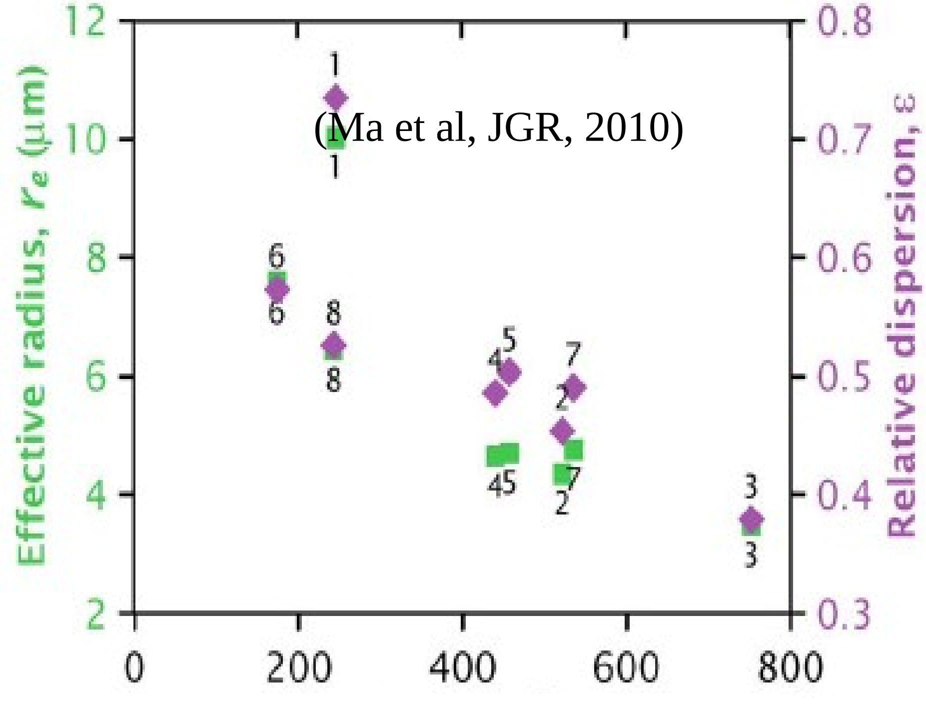


Green dashed line indicates the reflectivity error where overestimated cooling is comparable to the warming by doubling CO2.

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)

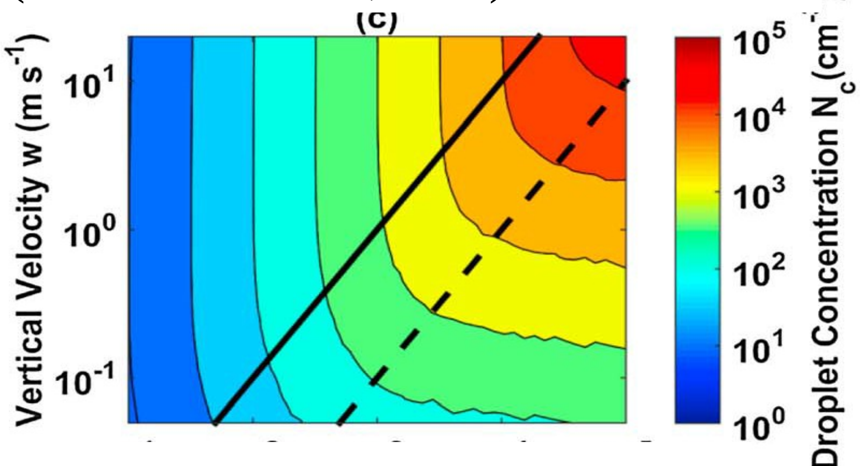
Cooling Dispersion Effect:

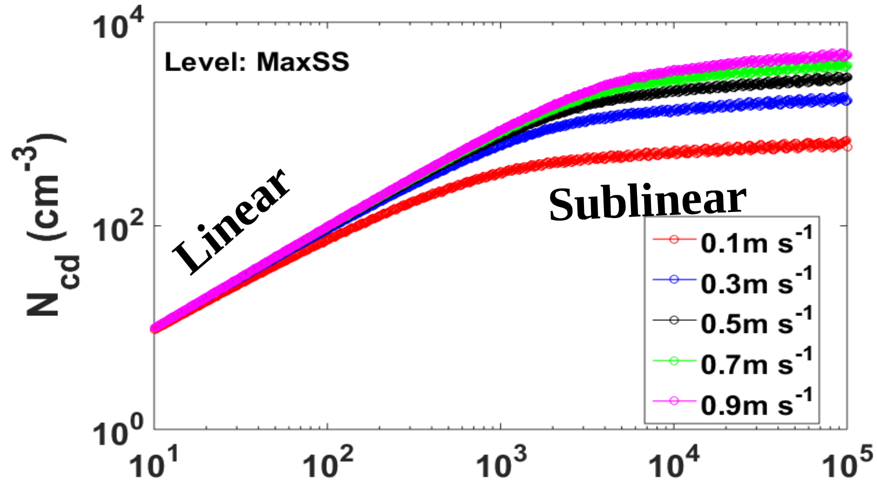
(Martins et al, ERL, 2009; Hudson et al, JGR, 2012)

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

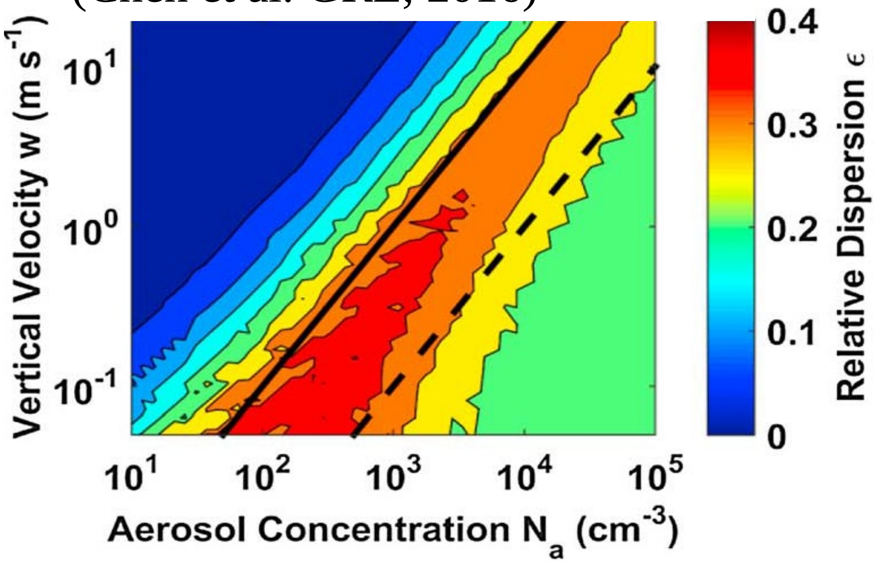
AIE Regime Dependence

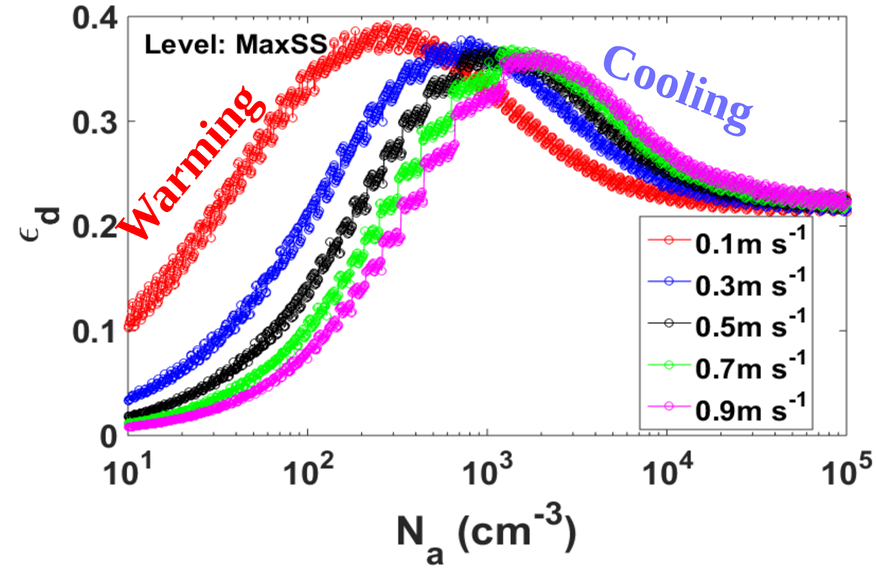
(Reutter et al. ACP, 2009)





(Chen et al. GRL, 2016)

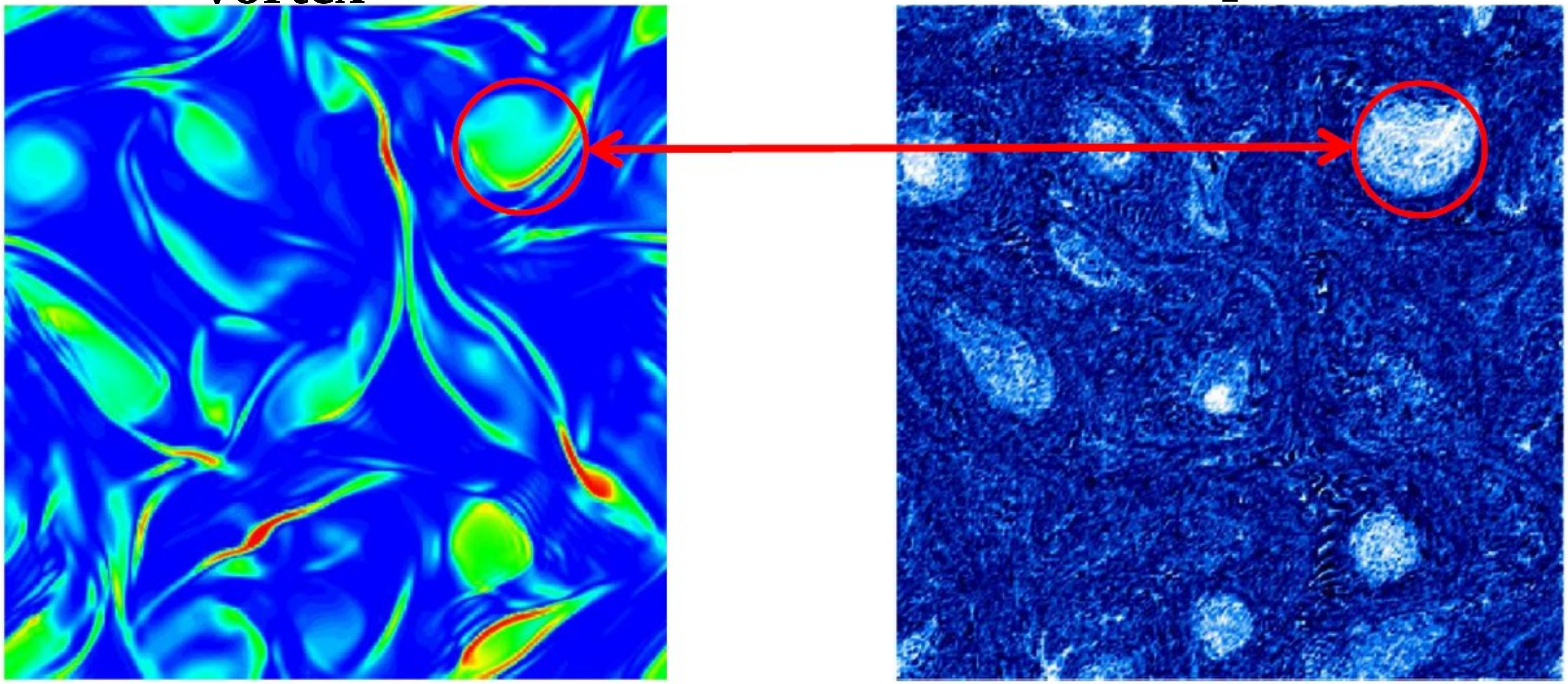




Dispersion effect exhibits stronger regime dependence & works to "buffer" number effect!

Preferential Concentration and Clustering

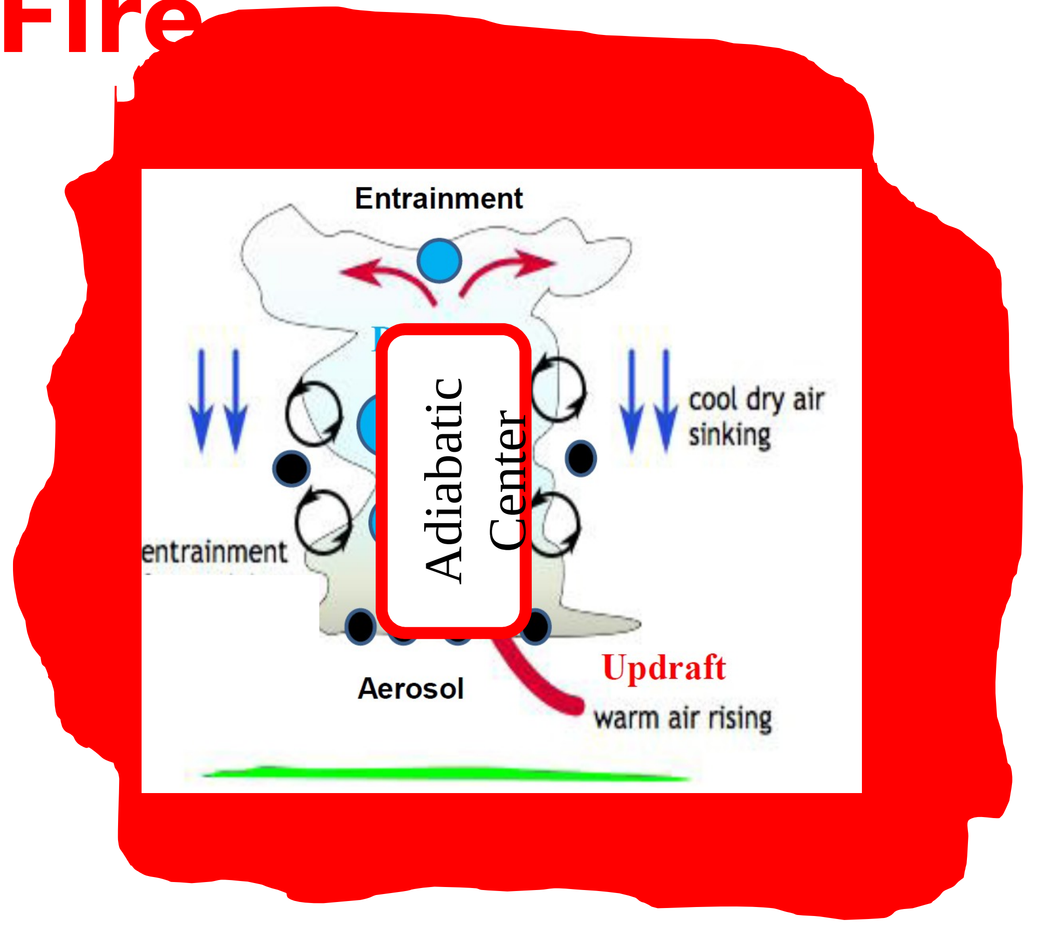
Vortex **Droplet Void**



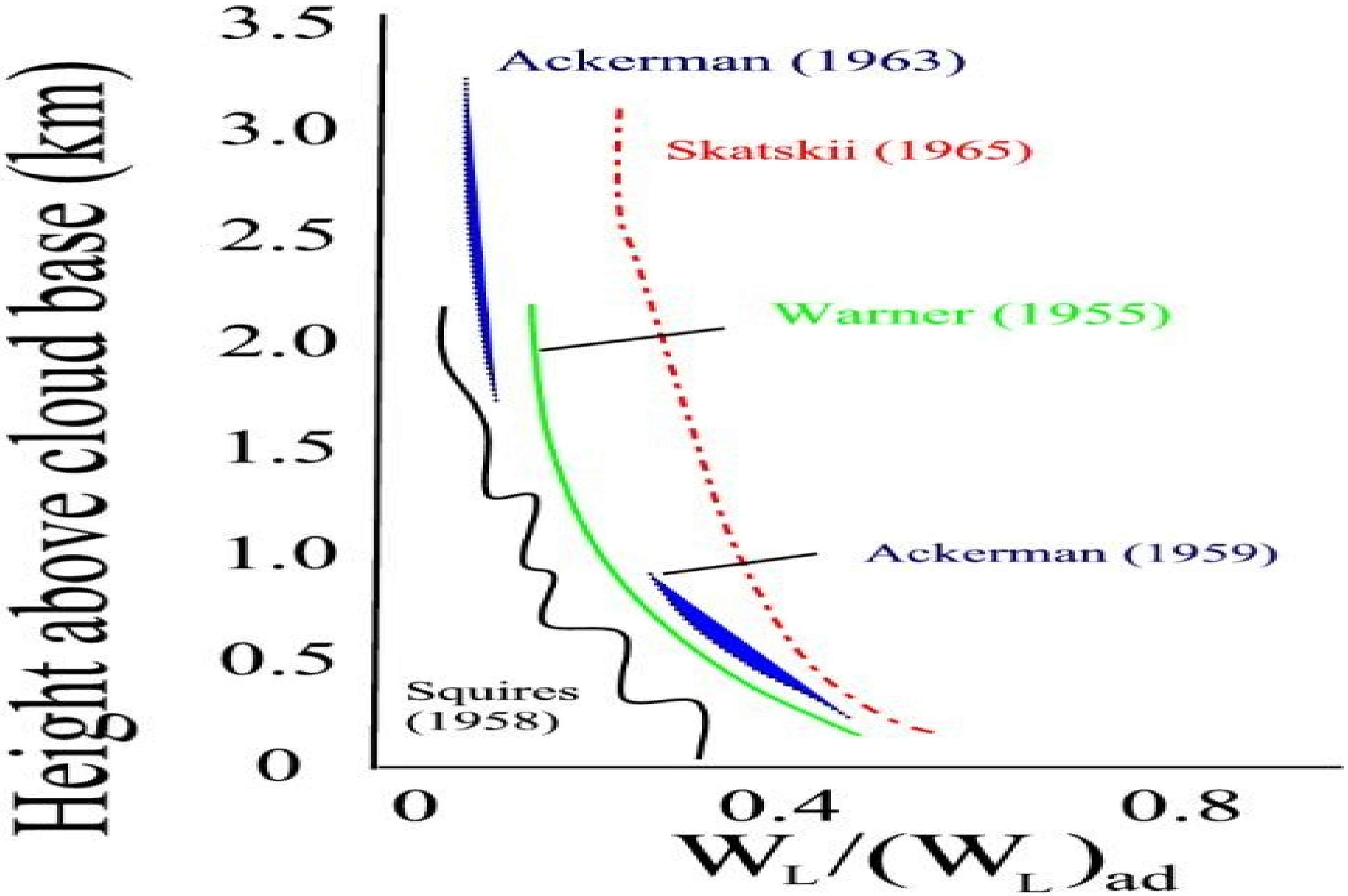
Combined effects of turbulent vortex and droplet inertial tend to concentrate droplets in regions of low vorticity. The so-called preferential concentration may be crucial for resolving long-standing puzzles.

Paradigm Shift – Cloud's Ring of Fire

- Near cloud edges (inward and outward)
- Paradigm shift from adiabatic center to diabatic edges
- Importance of updraft-limited regime
- Aerosol-cloud continuum
- 3D effect and radiation transfer
- More relevant and challenging to remote sensing?



Subadibatic LWC Profile-Entrainment



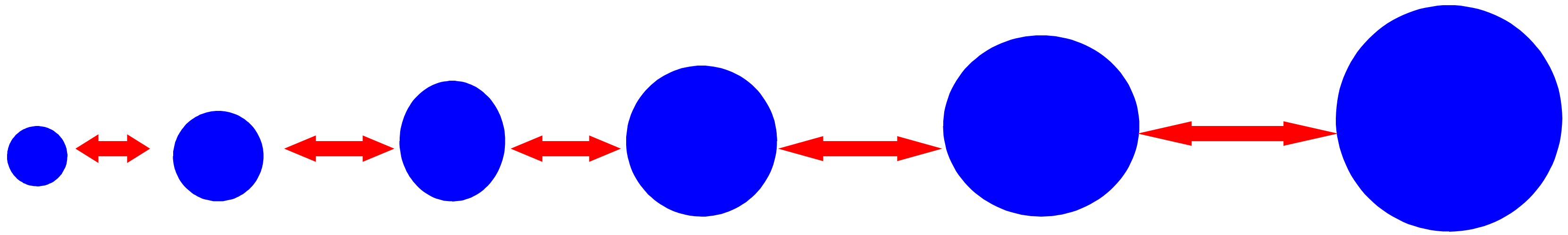
This figure shows that the ratio of the observed liquid water content to the adiabatic value decreases with height above cloud base, and less than 1 (adapted from Warner 1970, J. Atmos. Sci.)

Dynamic Equilibrium

Consider an ensemble of drops near the region of embroynic raindrops exchange water vapor molecules with surrounding environment at dynamic equilibrium (detailed balance):

$$\mathbf{A_g + A_1 = A_{g+1}}$$

A_g = a drop of size g; A_1 = a monomer



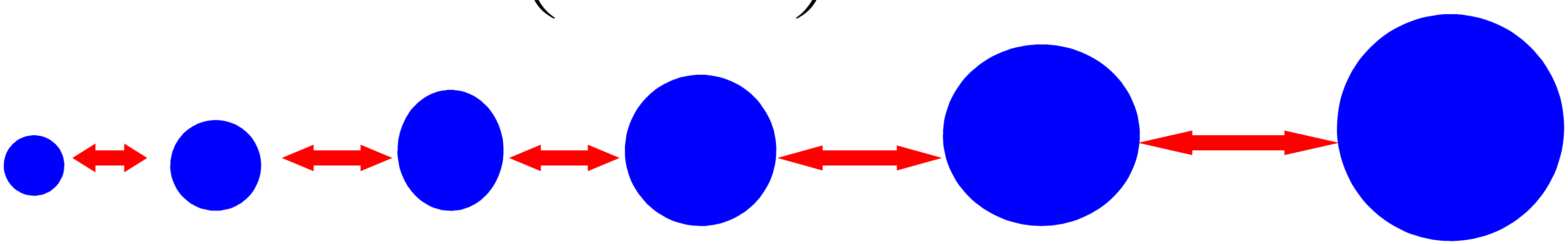
Kinetic Potential

Under equilibrium, ng can also be expressed in Boltzmann form

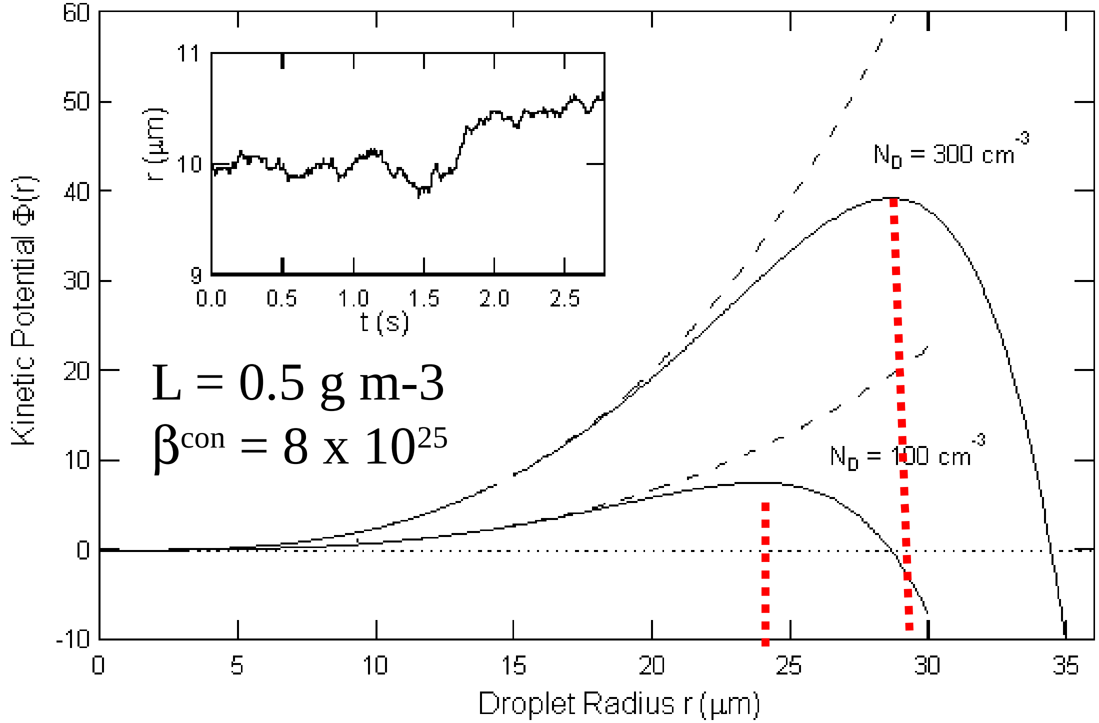
$$n_g = n_1 \exp\left[-\frac{w(g)}{kT} \right]$$

where “w/kT” is the reduced thermodynamics potential for droplet formation from vapor. Comparison of the two ng expressions yields the kinetic potential

$$\Phi(g) = -\ln\left(\prod_{i=1}^{g-1} \frac{\beta_i}{\gamma_{i+1}} \right) = \frac{w(g)}{kT}$$



Kinetic Potential Peaks at Certain Drop Size



- Rain initiation is a barrier-crossing process like nucleation.
- Both critical radius and potential barrier increases with increasing droplet concentration.
- The results suggest increasing aerosols inhibit rain by enhancing the barrier height and critical radius.

This figure shows the kinetic potential as a function of the droplet radius at different values of droplet concentration N calculated from the above equation for the kinetic potential. The dashed lines are without collection.

Remaining Issues and Challenges

- How to determine the parameters a and b in the power-law relationship $\mathbf{x = ar^b}$
- 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

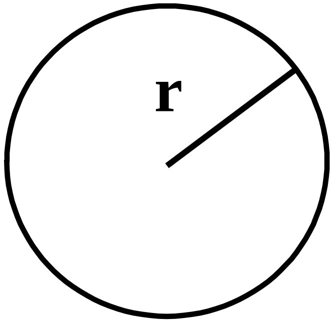
Big system vs. small system
(Liu et al, JAS, 1998, 2002)

Molecular system, Gas	Clouds
Knew Newton's mechanics for each molecule	Know equations For each droplet
Kinetics failed to explain observed thermodynamic properties	Uniform models failed to explain observed size distributions--
Maxwell, Boltzmann, Gibbs introduced statistical principles & established statistical mechanics	Establish the systems theory
Most probable distribution	Most probable distribution / Least probable distribution

Gibbs Energy for Single Droplet

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

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



σ = surface energy

ρ_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

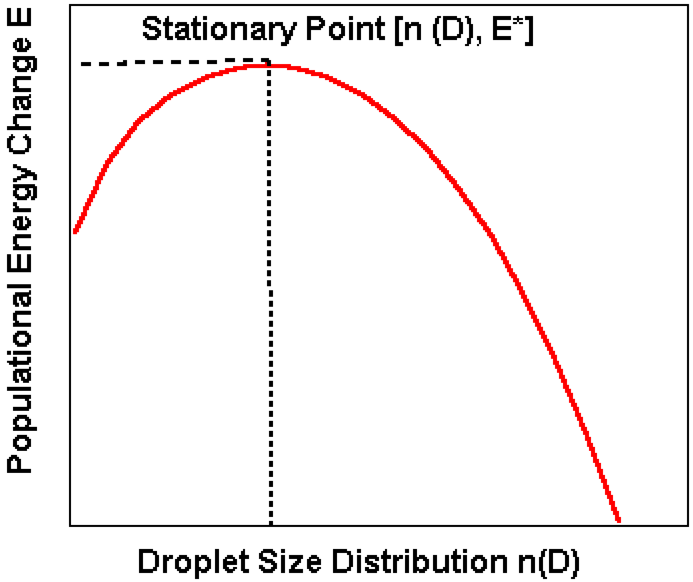
$$G = \int g(r)\, n(r)\, dr$$

$$= c_1 \int r^3 n(r) dr + c_2 \int r^2 dr + c_3$$

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).

Liu et al. (1993, 1995, 2000); Liu (1995); Liu & Hall

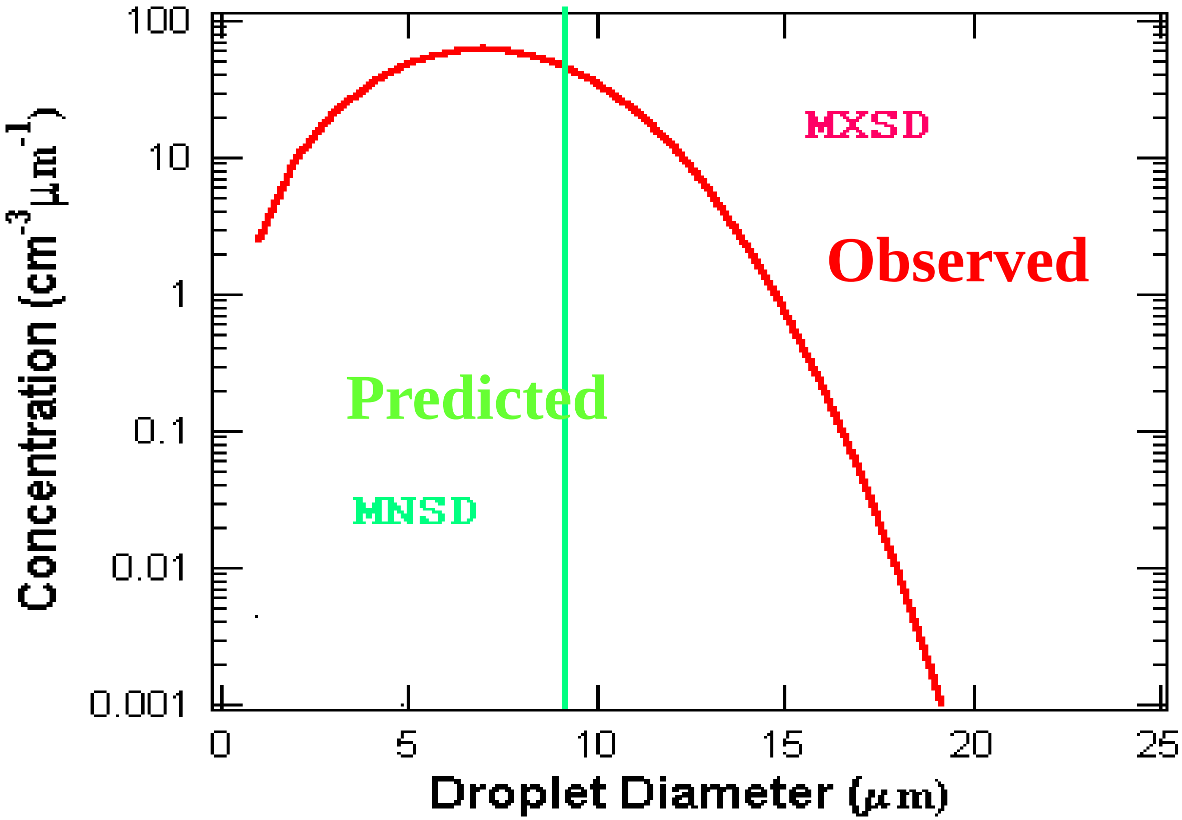
Least Probable Size Distribution



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

$$\mathbf{n_{min}(r) = N\delta(r - r_0)}$$

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

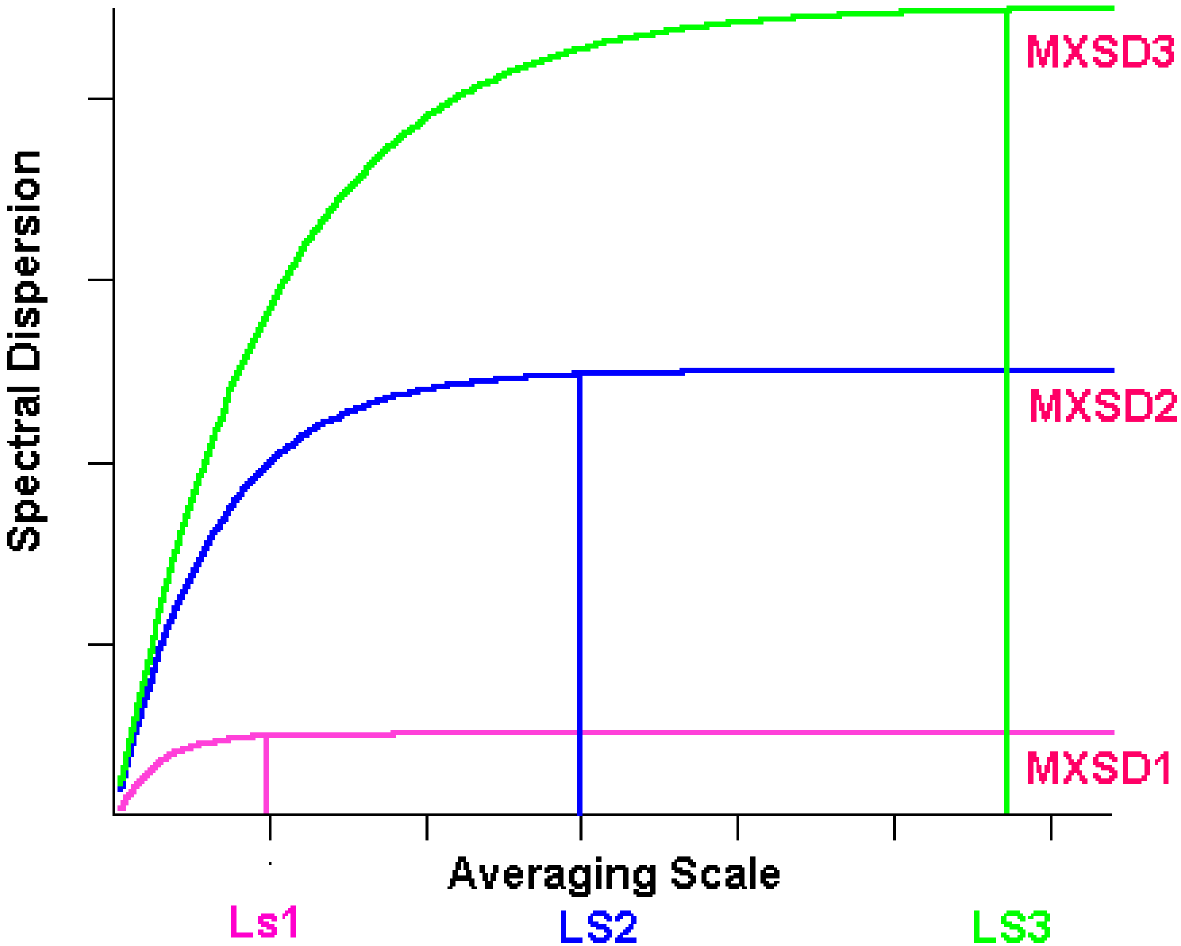


Diagram shows the dependence of size distributions (observed or simulated) on the averaging scale

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

More Scale-Dependence of Size Distribution

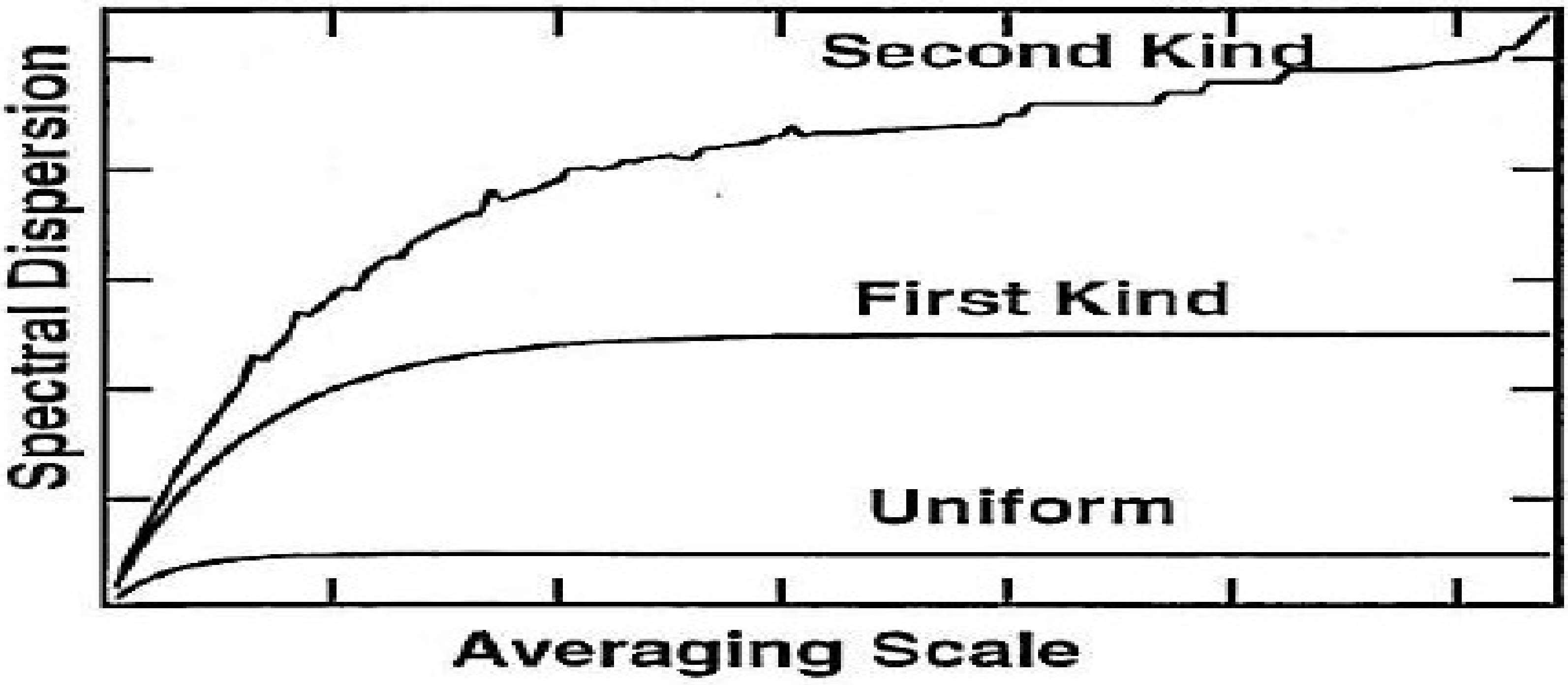


Figure 5. A diagram illustrating the scale-dependence of droplet size distributions. Both axes are only qualitative. The bottom curve represents the simplest case of uniform clouds. The middle and top curves represent the scale-dependence of the first and second kind, respectively.

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

Implications of Scale-Dependence for Microphysics Parameterizations

- **The scale-mismatch can make coupling of models at different scales challenging, if the issue of scale is not appropriately considered.**

- **Scale-dependent parameterizations are needed for models at different resolutions or adaptive-mess models.**

- **In view of cloud parameterizations in climate models, moment-based simple microphysical models may be physically better than sophisticated models with detailed microphysics.**

Systems Theory as a New Paradigm

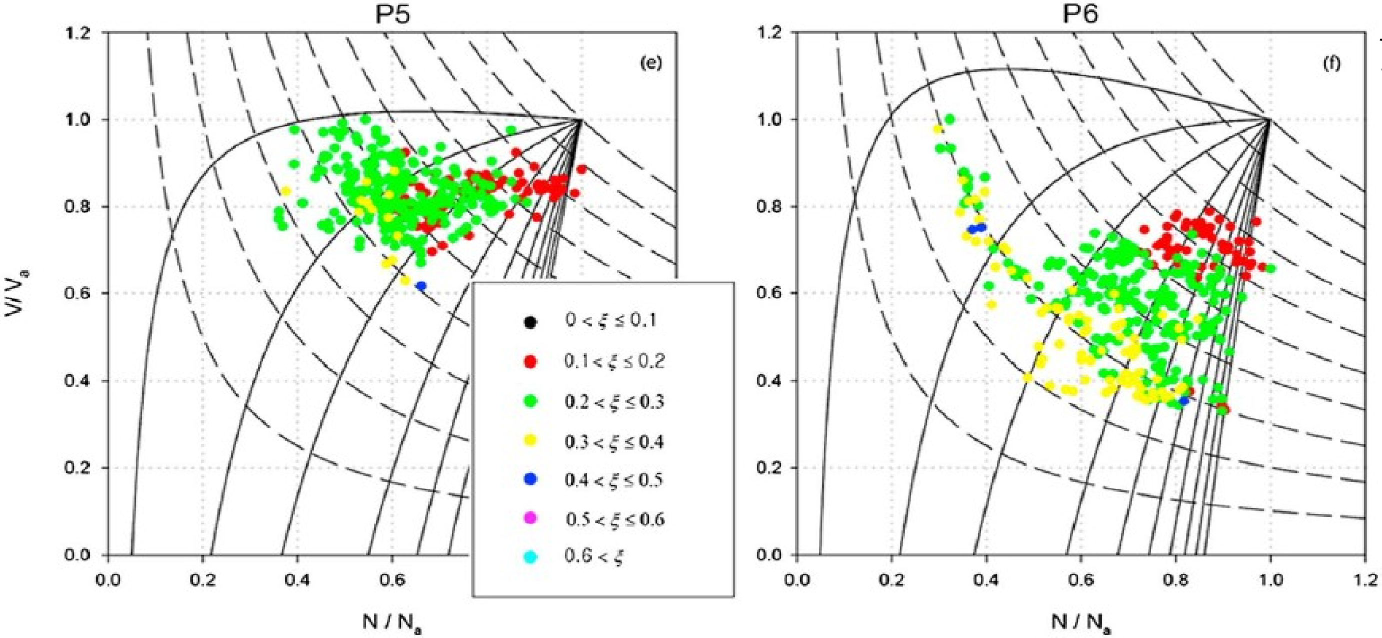
Fluctuations and interactions in turbulent clouds lead us to question the possibility of tracking individual droplets/drops and to consider droplets/drops as a system.

Molecular system, Gas	**Clouds**
Knew Newton's mechanics for each molecule	**Know equations For each droplet**
Kinetics difficult to explain thermodynamic properties	**Mainstream models difficult to explain size distributions**
Statistical mechanics; Phase Transition; Boltzmann equation	**Entropy principle; KPT; Fokker-Planck Equation**

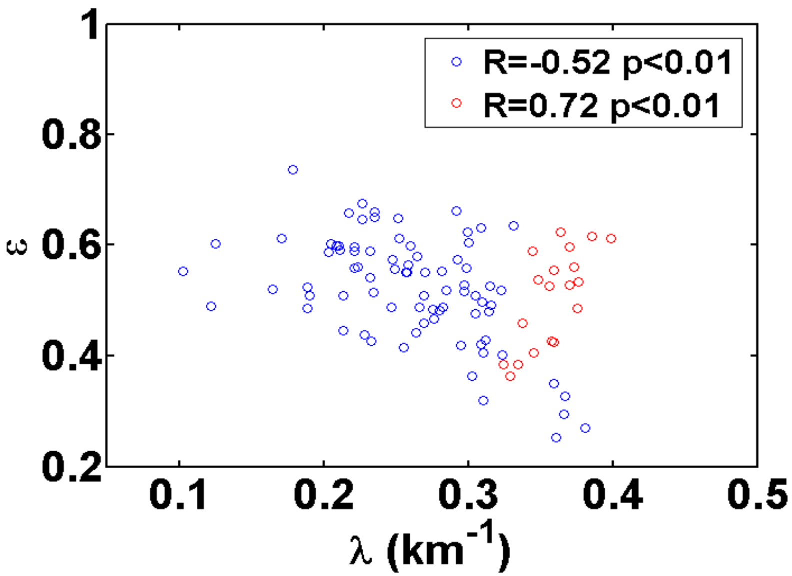
Entropy and Disorder

"I blame entropy."

Spectral Broadening with Entrainment



Yum et al JGR, 2015



Guo et al AE, 2016

Spectral narrowing

Entrainment Causes Multiscale Variability

- **Variation at ever finer scales**

(up to 1 cm)

- **Major progress in instrument**

-- Impact-based

-- Scattering-based since 1980 (e.g., FSSP)

-- Holographic – HOLODEC

~ CDSD at ~ 1 cm resolution

- **Aircraft speed**

-- DOE G-1 (100 m/s)

-- Helicopter (ACTOS)

- **Highest resolution**

ACTOS + HOLODEC

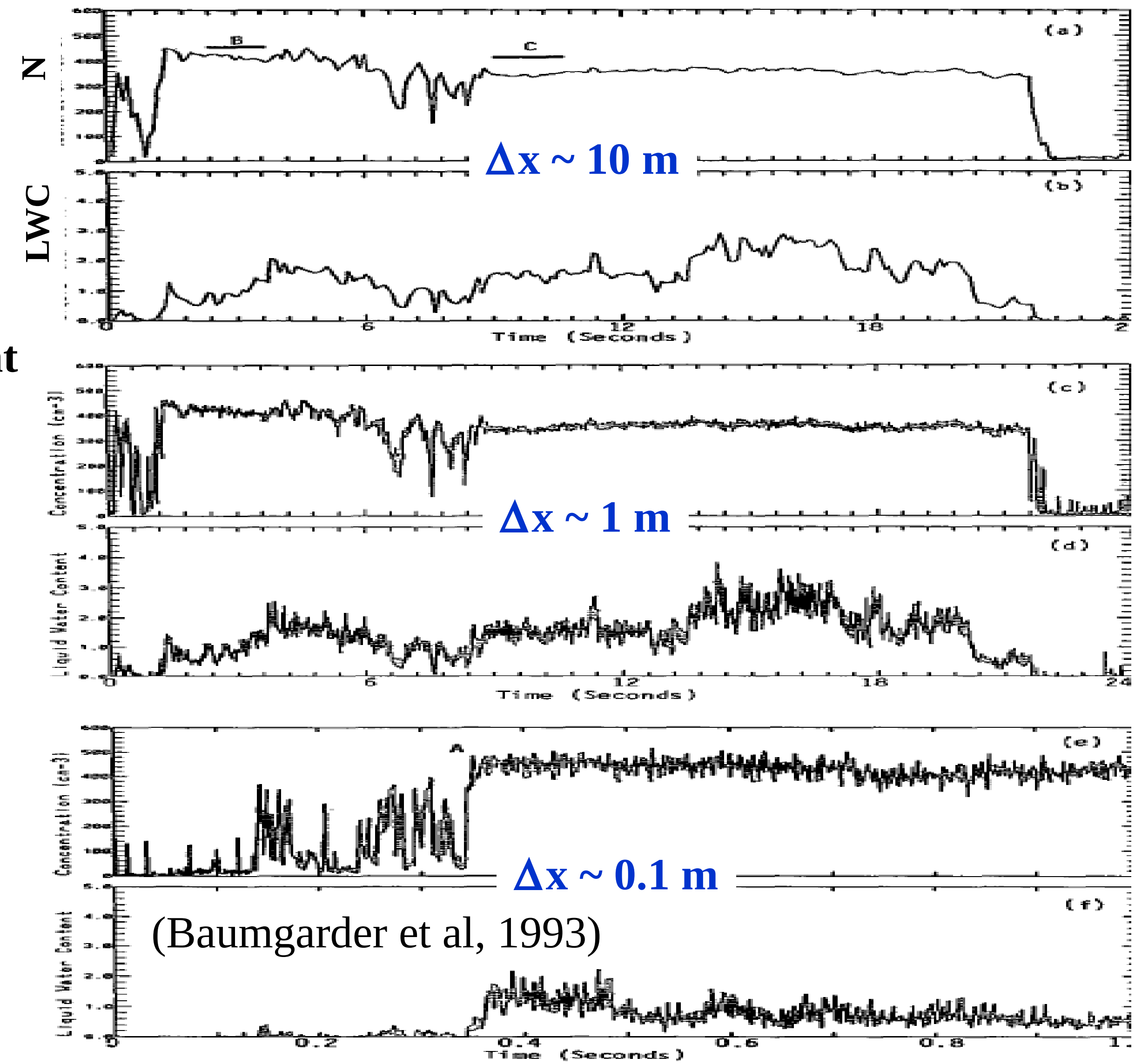


FIG. 2. The droplet concentrations and liquid water contents derived from the size distributions are shown plotted as a function of time for path averages of 10 m [(a) and (b)], 1 m [(c) and (d)] and 0.1 m [(e) and (f)].

CCN Effect and Squires Colloidal Instability

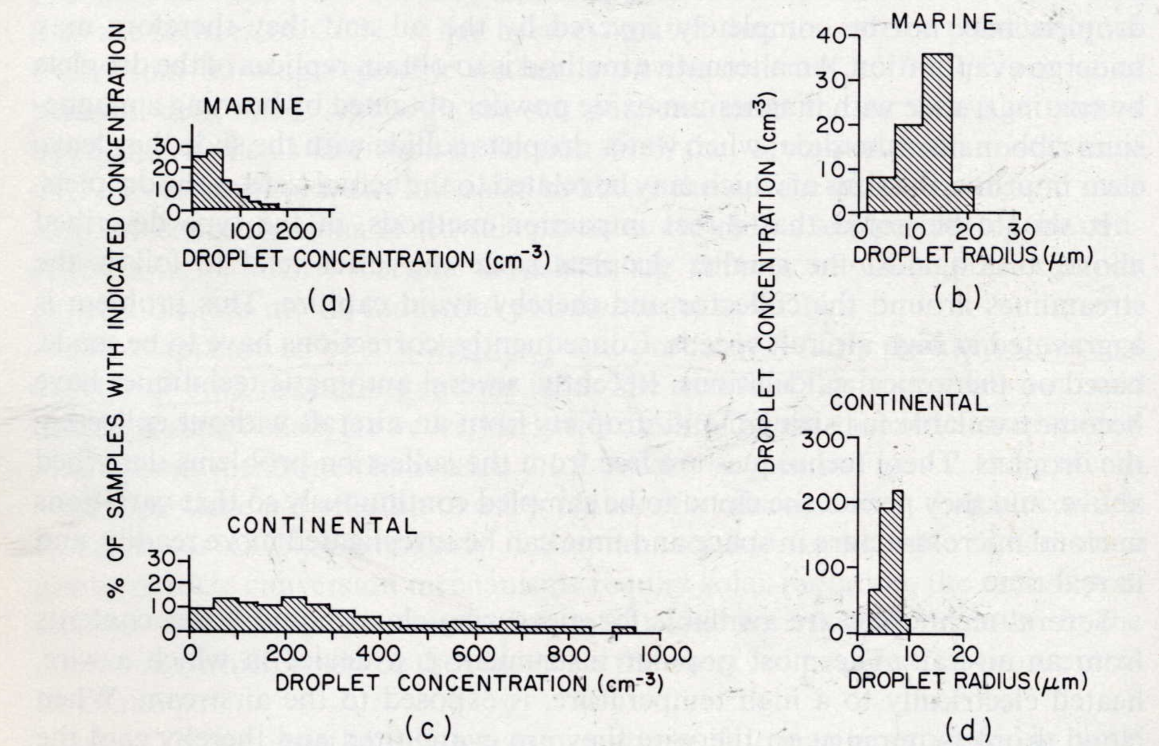


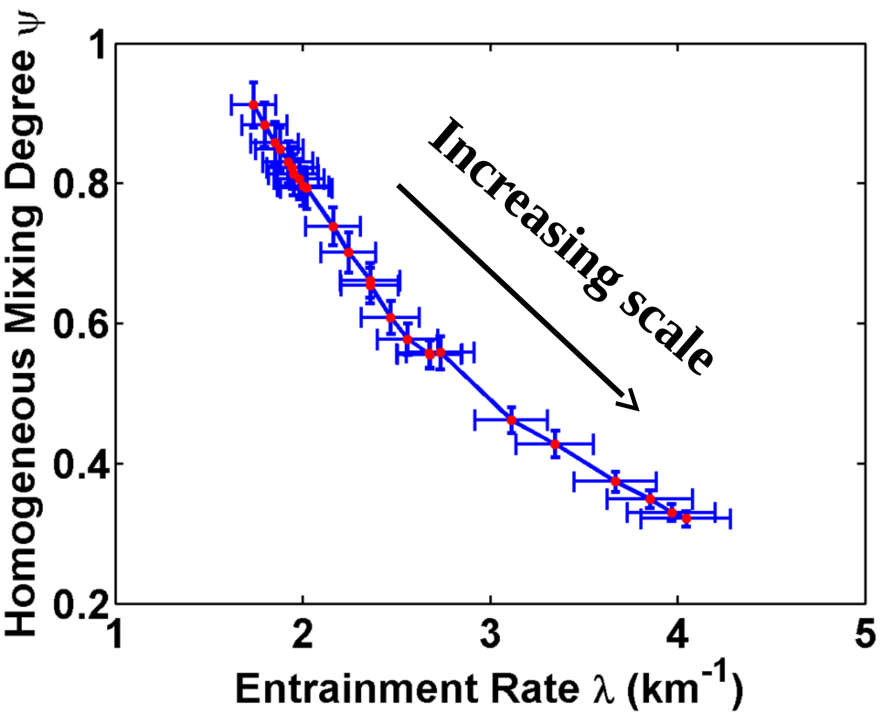
Fig. 4.15 (a) Percentage of marine cumulus clouds with indicated droplet concentrations. (b) Droplet size distributions in a marine cumulus cloud. (c) Percentage of continental cumulus clouds with indicated droplet concentrations. (d) Droplet size distributions in a continental cumulus cloud. Note change in ordinate from (b). [From *Tellus* 10, 258–259 (1958).]

nental clouds have more droplets, small

CCN or Turbulence Effects?

Scale-Induced Relationship between Entrainment Rate and Homogeneous Mixing Degree

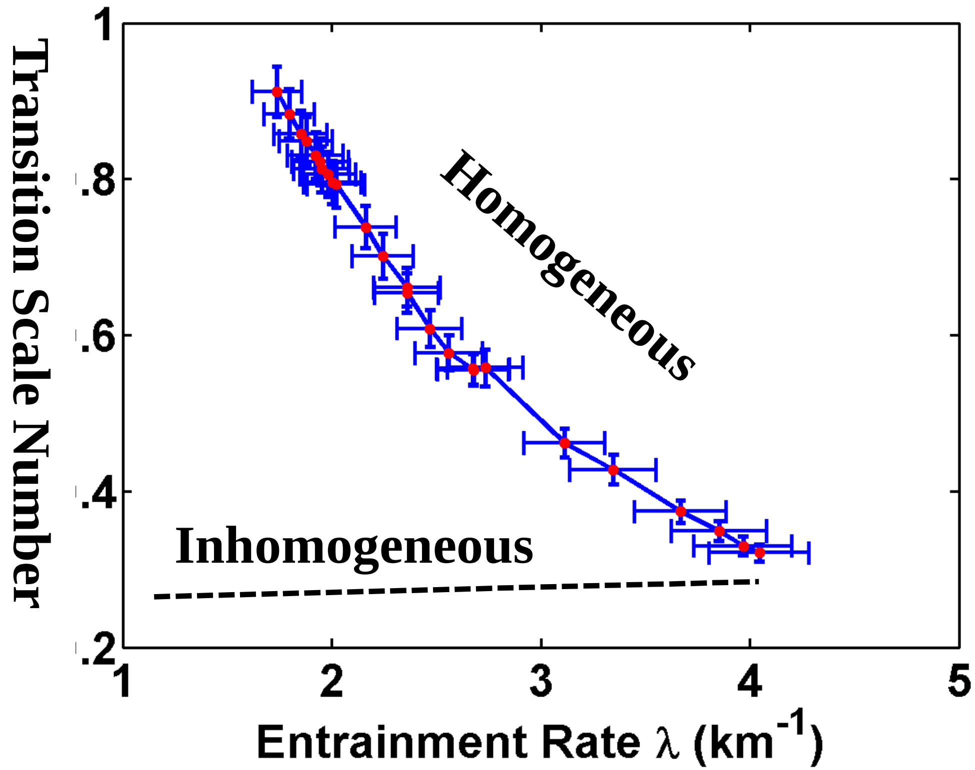
- Effect of dilution
- Effect of entrained eddy sizes/velocities
- Ill-defined without knowing scale
- Scale may be a reason for uncertainty



Can this relationship be used to diagnose mixing mechanisms from entrainment rate?

Dynamical Mixing Diagram for Parameterizations

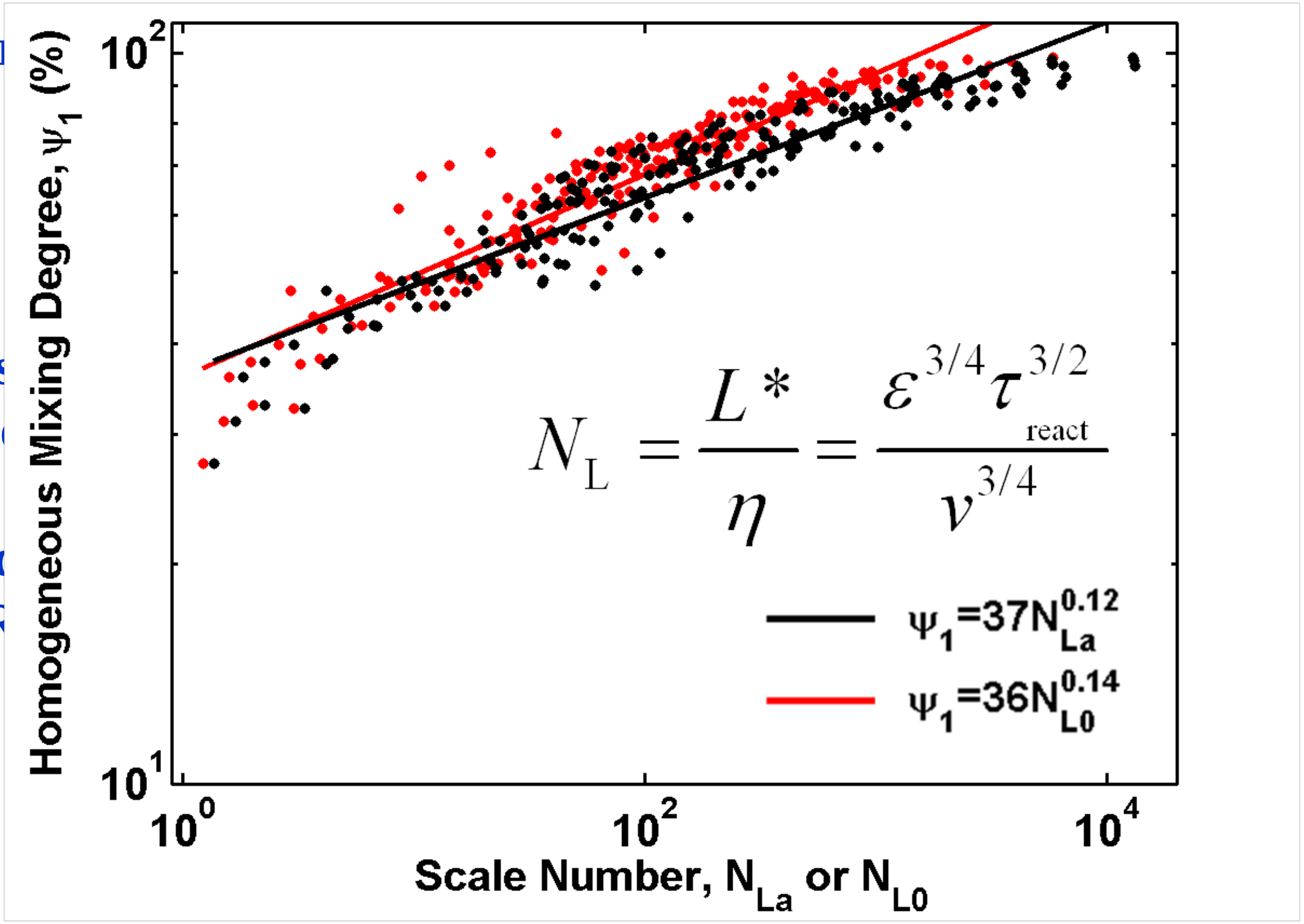
- Transition scale number can be used to parameterize homogeneous mixing degree (Lu et al., JGR, 2011, 2013) .
- The transition scale number at the highest resolution essential to the scale-dependence .



This dynamical mixing diagram can serve as a basis for developing scale-dependent parameterizations of entrainment rate and homogeneous mixing degree.

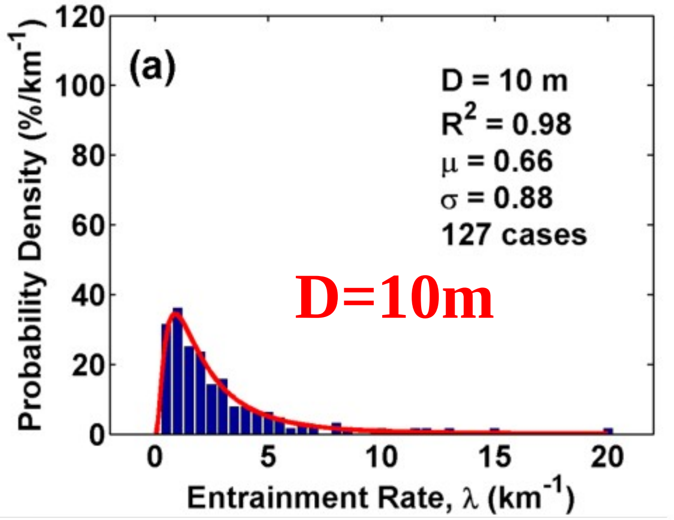
New Parameterization for Homogeneous Mixing Degree

- Eliminate the need fo assuming extreme inhomogenous or homogenous mixing;
- Work best for models with 2-moment schem
- Testing with SCM anc CRM/LES in FASTEF
- Integrating with entrainment rate



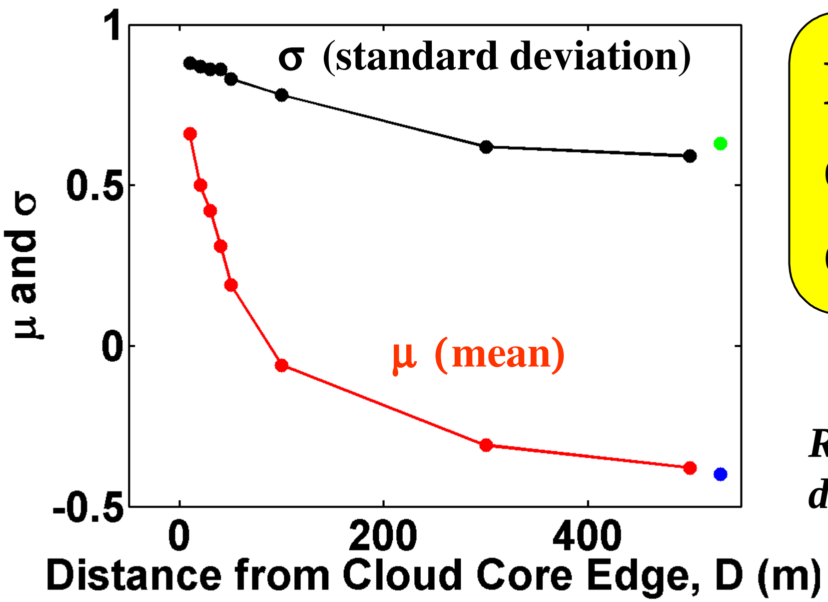
A new parameterization that unifies entrainment rate and mixing effects on cloud microphysics is on the horizon.

PDF and Distance Dependence



All the PDFs can be well fitted by lognormal distributions; $R^2 > 0.91$.

$$f_X(x;\mu,\sigma) = \frac{1}{x\sigma\sqrt{2\pi}} e^{-\frac{(\ln x-\mu)^2}{2\sigma^2}}, \quad x > 0$$

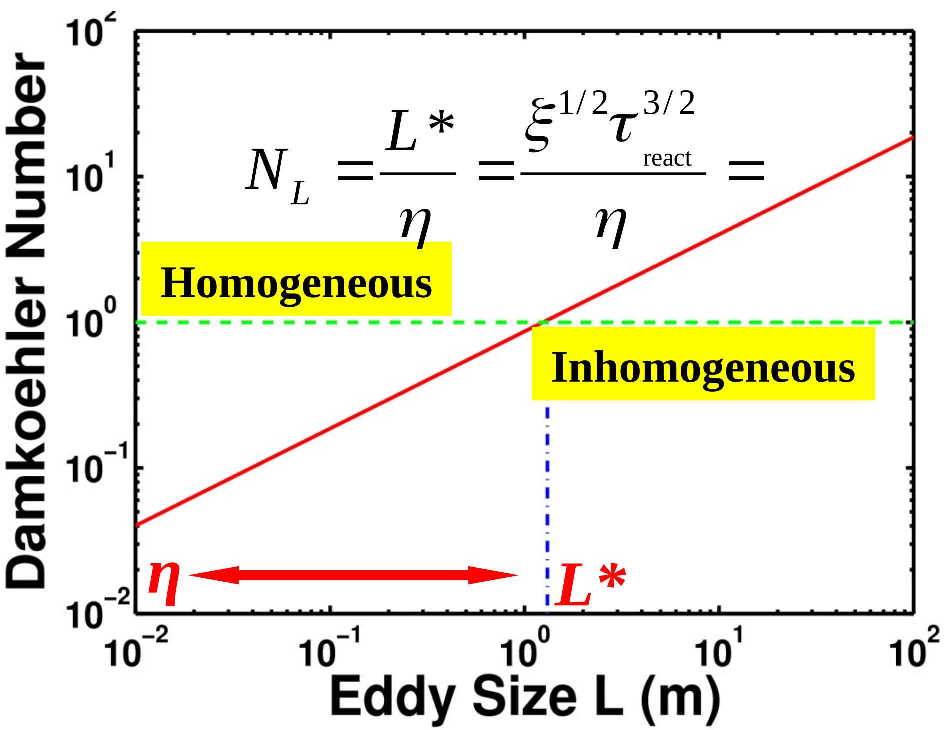


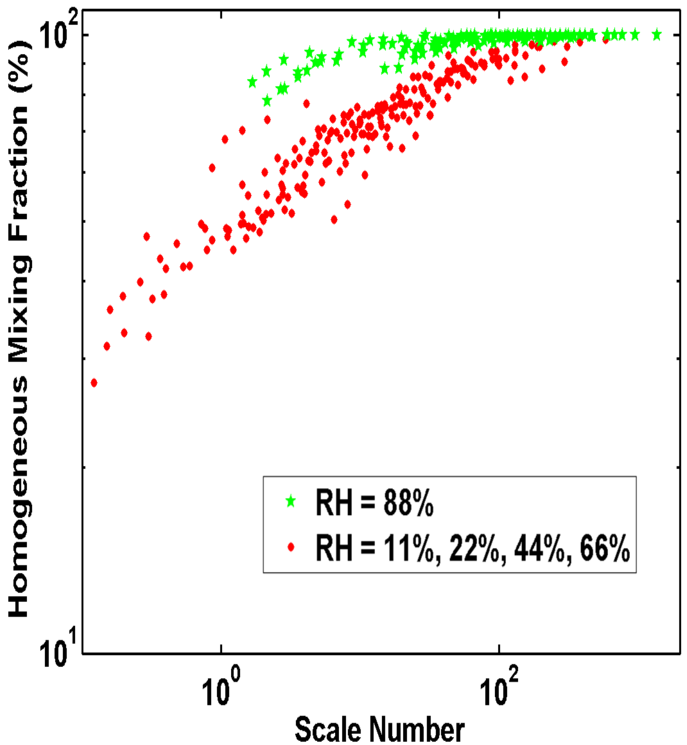
Both mean and standard deviation of ln (λ) decrease with increasing distance from cloud core D.

Ref: Lu et al 2012: Entrainment rate in cumuli: PDF and dependence on distance. Geophys. Res. Lett. (in press)

Homogeneous Mixing Fraction

η: Kolmogorov scale; L* transition scale; N_L transition scale number





Further parameterization of the scale number leads to a much needed parameterization for homogeneous mixing fraction.

Lu et al 2011: Examination of turbulent entrainment-mixing mechanisms using a combined approach. J. Geophys. Res.; 2012: Relationship between homogeneous mixing fraction and transition scale number, Environ. Res. Lett. (to be submitted)

Three Definitions of Homogeneous Mixing Fraction --- Ψ_3

$$\boxed{\psi_3 = \frac{\ln N - \ln N_i}{\ln N_h - \ln N_i} = \frac{\ln r_v^3 - \ln r_{vi}^3}{\ln r_{vh}^3 - \ln r_{vi}^3}}$$

This definition, Ψ_3, turns out to be related to α:

$$\psi_3 = 1 - \alpha$$

where α was defined by Morrison and Grabowski (2008):

$$N = N_0 \left(\frac{q}{q_0}\right)^{\alpha}$$

Two Transition Scale Numbers (2)

τ_{react} **is based on:**

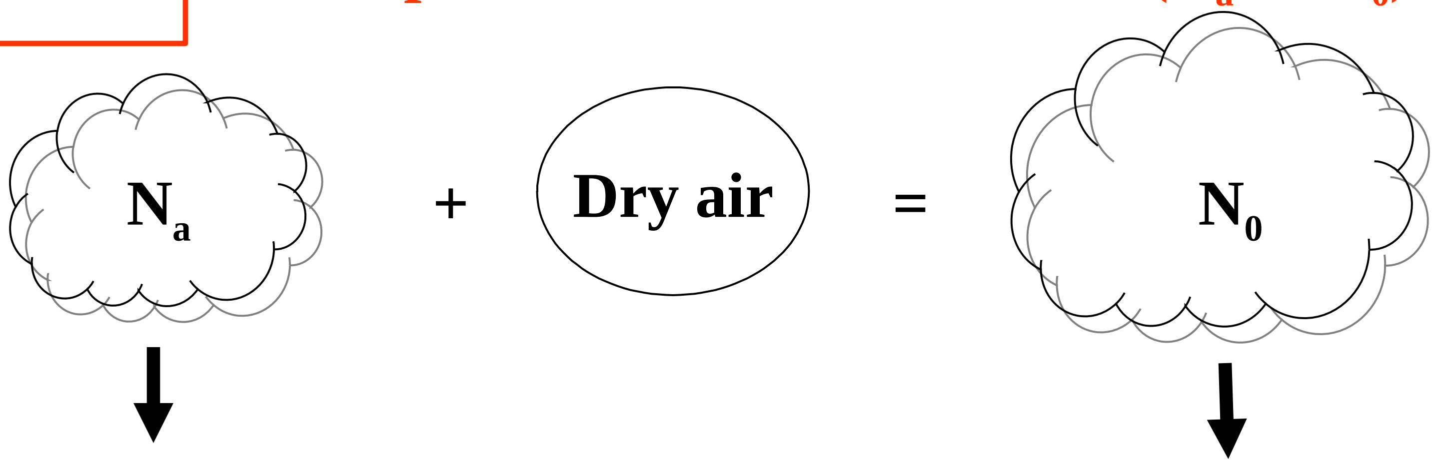
$$\boxed{\begin{cases} \dfrac{dr}{dt} = A\dfrac{s}{r} \\ \dfrac{ds}{dt} = -Brs \end{cases}}$$

r: **droplet radius;**

s: **supersaturation;**

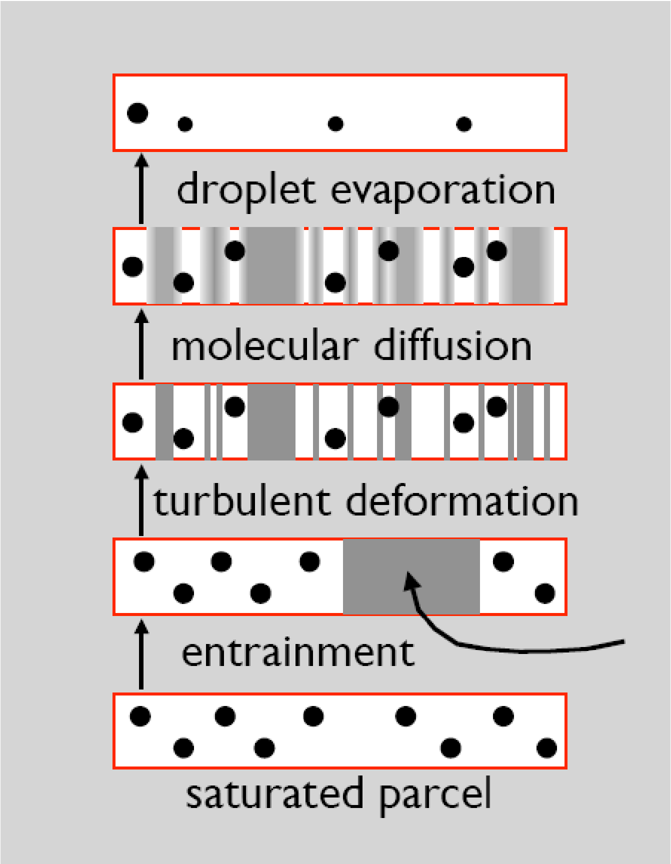
A: **a function of pressure and temperature;**

B: **a function of pressure, temperature and droplet number concentration (N_a or N_0).**



Scale Number N_{La} N_{L0}

Explicit Mixing Parcel Model (EMPM)



Krueger (2008)

Domain size:

20 m× 0.001 m × 0.001 m ;

Adiabatic Number Concentration:

102.7, 205.4, 308.1, 410.8, 513.5 c

Relative humidity:

11%, 22%, 44%, 66%, 88%;

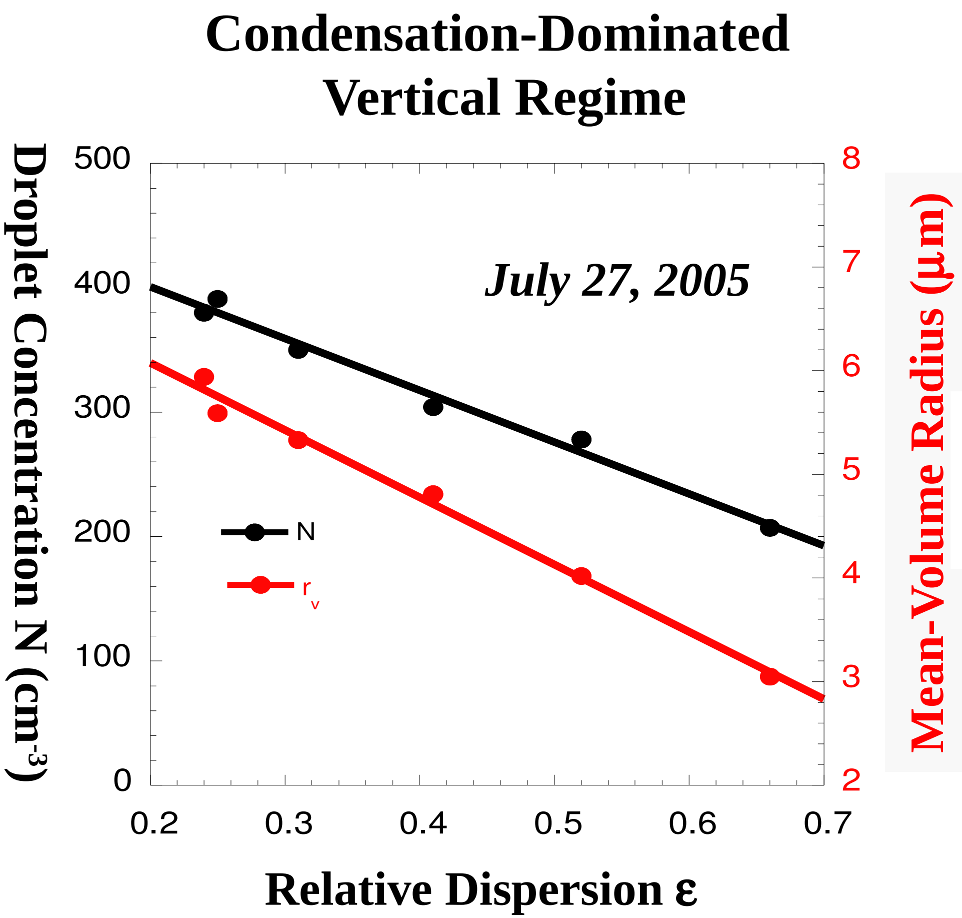
Dissipation rate:

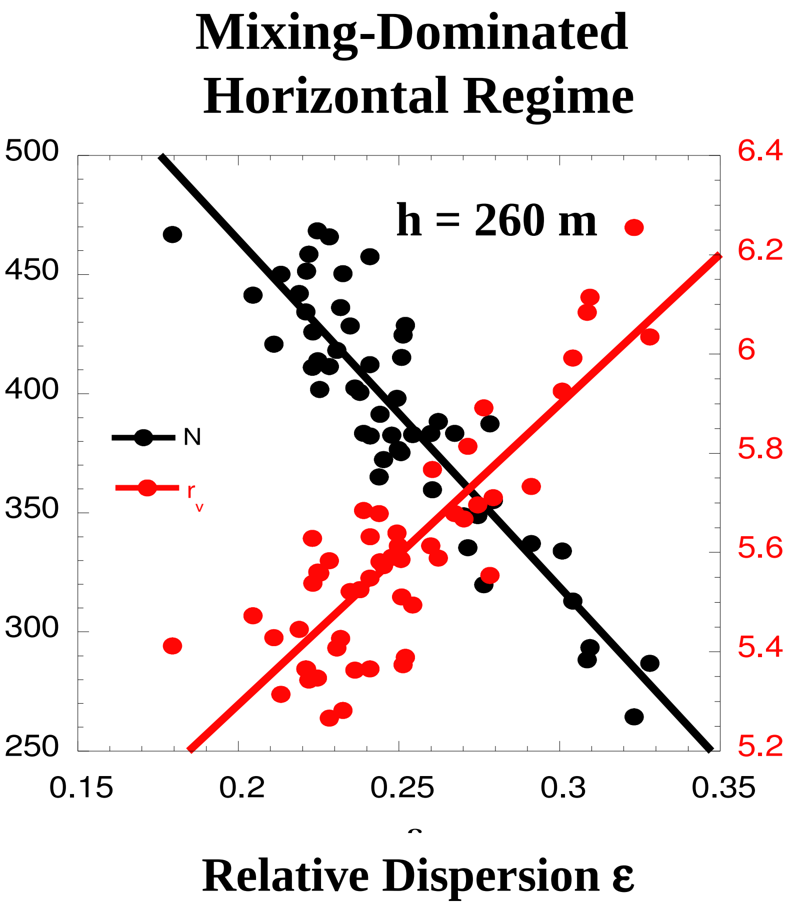
1e-5, 5e-4, 1e-3, 5e-3, 1e-2, 5e-2 m

Mixing fraction of dry air:

0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9.

Entrainment-mixing processes complicate the dispersion effect as well.





Note the opposite relationships of mean-volume radius to relative dispersion in the two figures. The left panel is largely consistent with the adiabatic condensation theory whereas the right one with entrainment-mixing processes.

Brief History and Motivation

Entrainment: environmental air into clouds (e.g., Stommel 1947, Squires 1949)

Parameterization for GCMs (Fractional) entrainment rate (Arakawa and

Cloud Physics: Mixing mechanisms & microphysics (Warner 1969)

Needs for unifying entrainment, mixing and microphysics parameterization, and for understanding scale-dependence.

There are still significant knowledge gaps to fill.

Large Eddy Simulations

- **Cases:** **Cumuli on May 22, 23 and 24, 2009 in RACORO.**
- **Model:** **WRF-FASTER (Reconfigured WRF to better take large scale forcing etc)**
- **Domain Size:** **9.6×9.6 km^2.**
- **Horizontal Resolution:** **75 m (128 points×128 points)**
- **Vertical Resolution:** **~40 m for the 125 levels below 5 km and a sponge layer for 13 grid levels up to 5.5 km.**

(Endo et al., JGR, 2015).

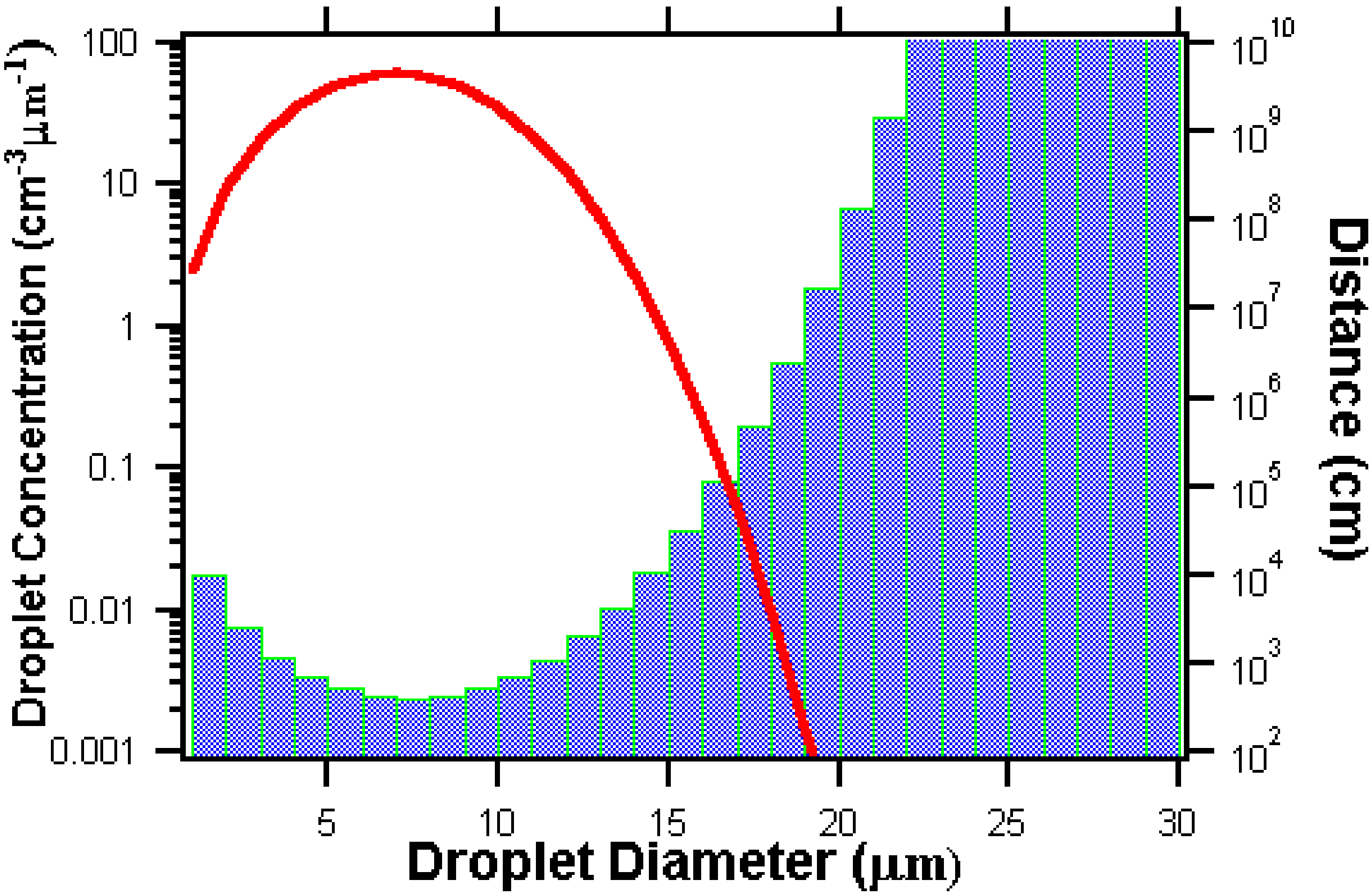
A Brief Summary I

Scale-dependence
(Droplet size distributions depend on the scale over which they are sampled)

Why does the Weibull distribution describe observed size distributions most accurately?

Spectral broadening ?
[Observed n(r) is broader than that predicted by uniform models]

FSSP Sampling Scale



FSSP Sampling

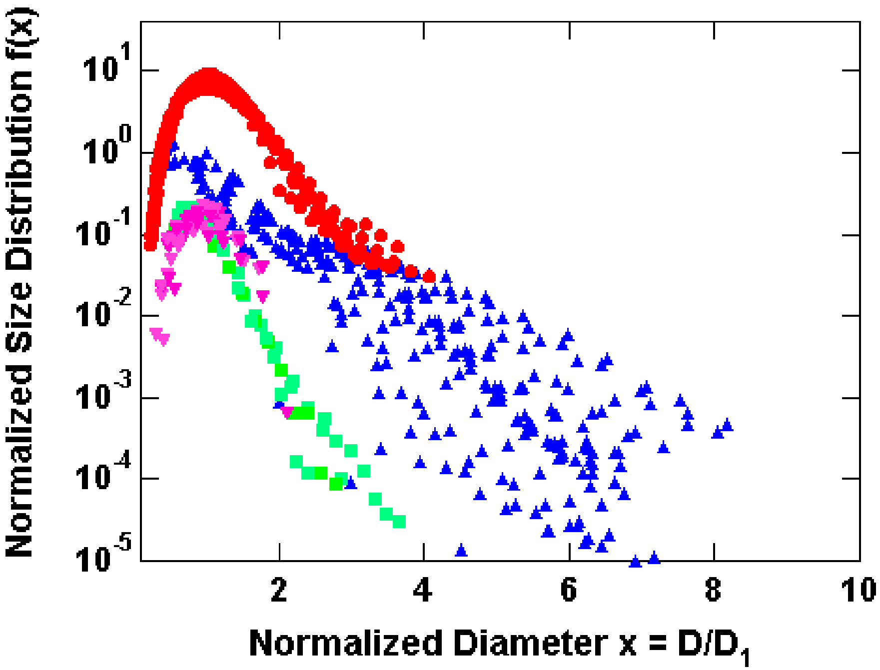
A ~ 0.004 cm²

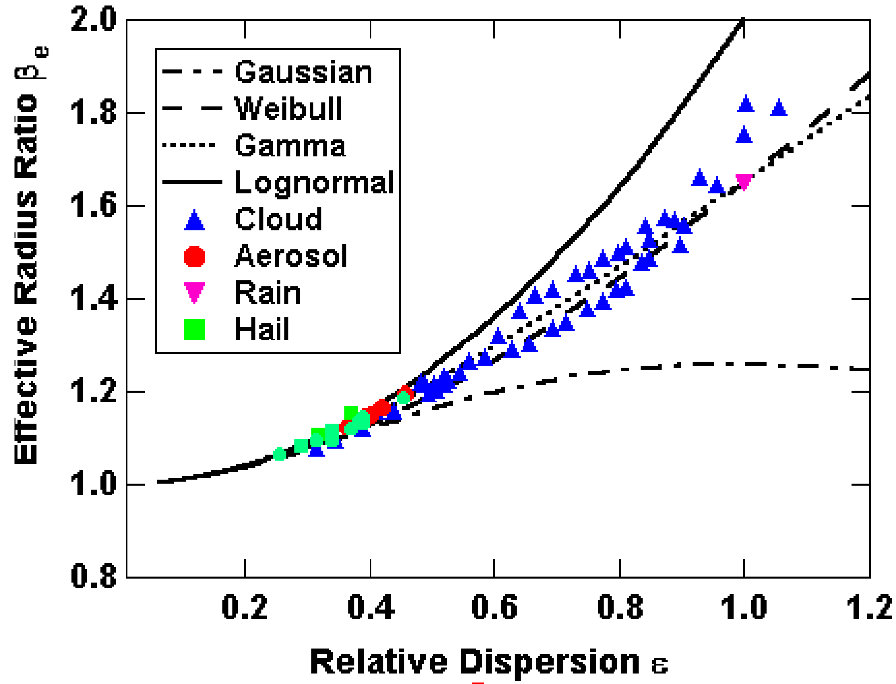
L_c ~ 100/A/n
~ 250/n (m)

Uncertainty: 10%.

The size distribution (red curve) is from Liu et al. (1995, Atmos. Res.).

Two Kinds of Universalities



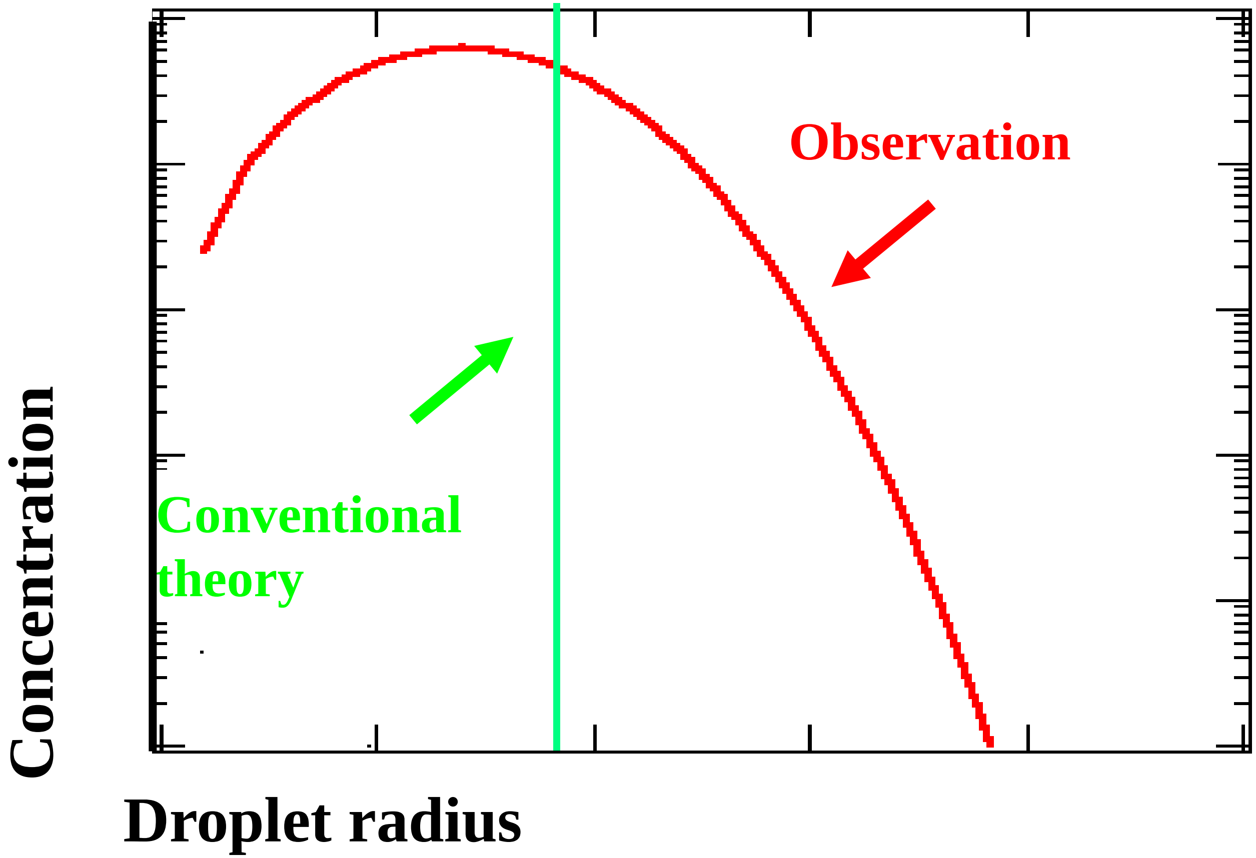


No 1st kind universality

2nd kind universality exists!

The first kind of universality is case-specific; the 2nd universality seems universal for atmospheric particle size distributions.

Spectral broadening is a long-standing anomaly in cloud physics since 1



We have developed a systems theory based on the maximum entropy principle, and applied it to **derive a representation of clouds.**

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

Droplet Population as a System

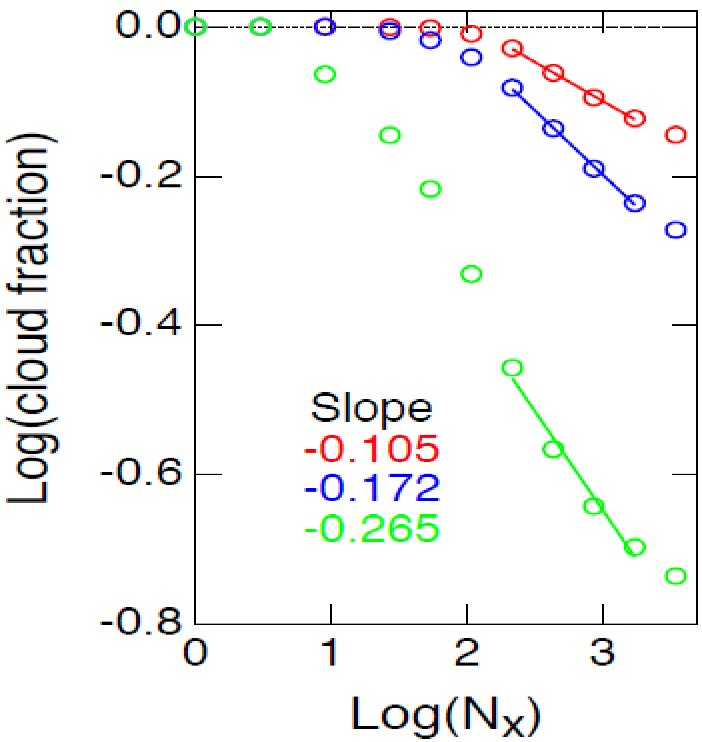
Various fluctuations associated with *turbulence* and aerosols suggest considering droplet population as a system to obtain information on droplet size distributions without knowing details of individual droplets and their interactions.

Molecular system (gas)	**Cloud**
Molecules & Newton’s mechanics for each molecule	**Droplets & equations for each droplet (DNS)**
Boltzmann equation	**Various kinetic equations (e.g., stochastic condensation)**
Maxwell, Boltzmann & Gibbs introduced statistical principles & established statistical mechanics	**Systems theory**
Most probable energy distribution	**Most probable size distribution** **Least probable size distribution**

We developed a systems theory (Liu & Hallett, QJ, 1998; Liu et al., AR, 1995, JAS, 1998, 2002a, b). Today mainly on MPSD based on the maximum entropy principle.

Steve's 5Dec2014 Seminar

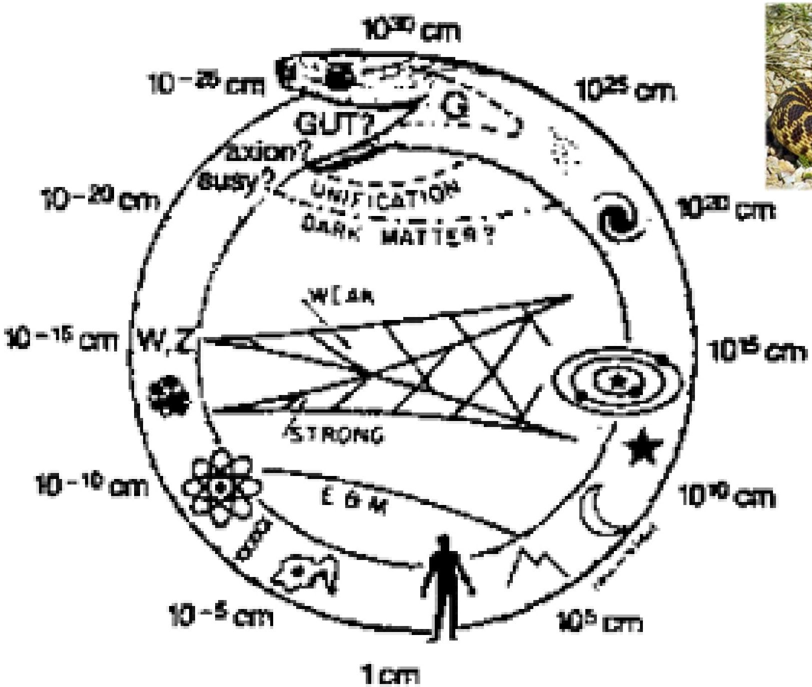
WHAT CAN WE LEARN FROM HIGH RESOLUTION SURFACE-BASED IMAGING OF CLOUDS?



Cloud fraction depends on averaging scale and cloud threshold.

This seminar: As a basic nature of turbulence, scale-dependence is true for cloud microphysics; deeper understanding and parameterization demands high-res obs, modeling, and fresh ideas!

Multiscale Climate Hierarchy in Bigger Picture: Climate Uroboros



Cosmic Uroboros was originated by Dr. Sheldon Glashow and popularized by Dr. Joel Primack.

I am suggesting the concept of Climate Uroboros

Uroboros is a legendary snake swallowing its own tail, representing hope for a unified theory that links the largest and smallest scales.

Turbulent Collection

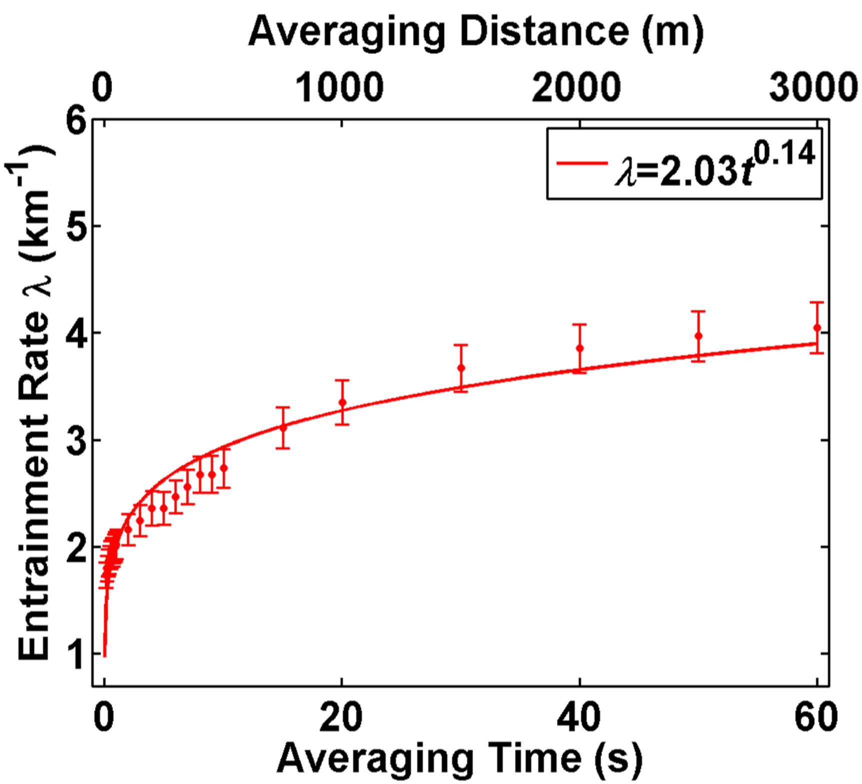
- Turbulent processes affect spatial distribution of cloud droplets and drops (the so-called clustering).

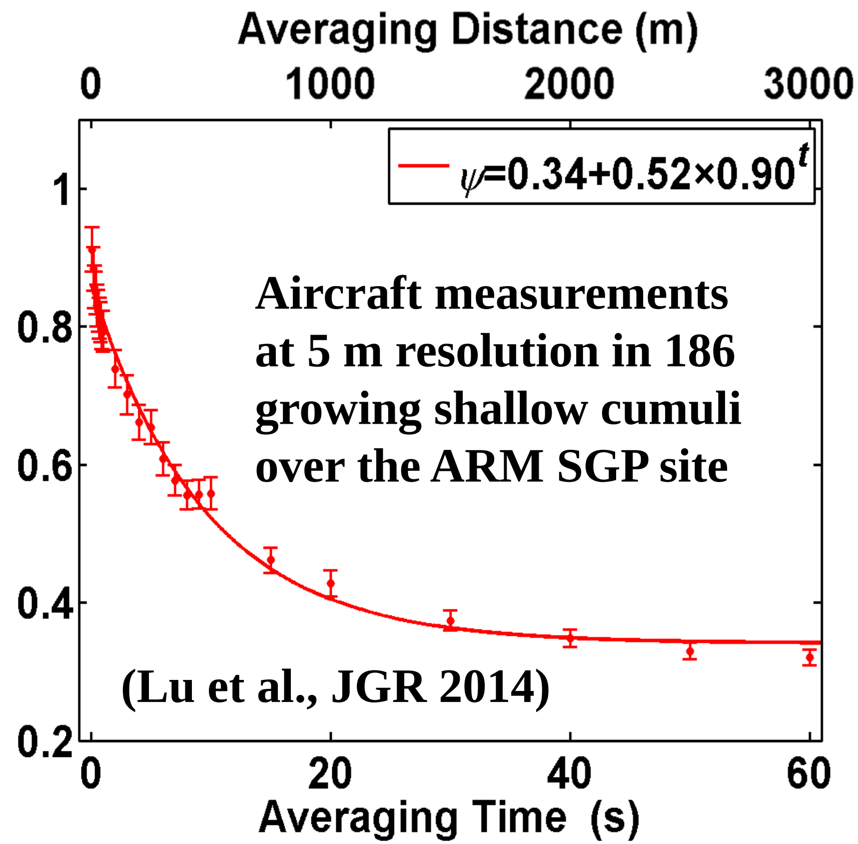
- Turbulent processes affect the collection kernel by altering
  * collection efficiency
  * relative velocities of droplets

- Different turbulent eddies may collide with other, and carry droplets with them.

- Qualitatively speaking, turbulent processes enhance collection process and rain formation, but quantitatively, turbulent effects are poorly understood.

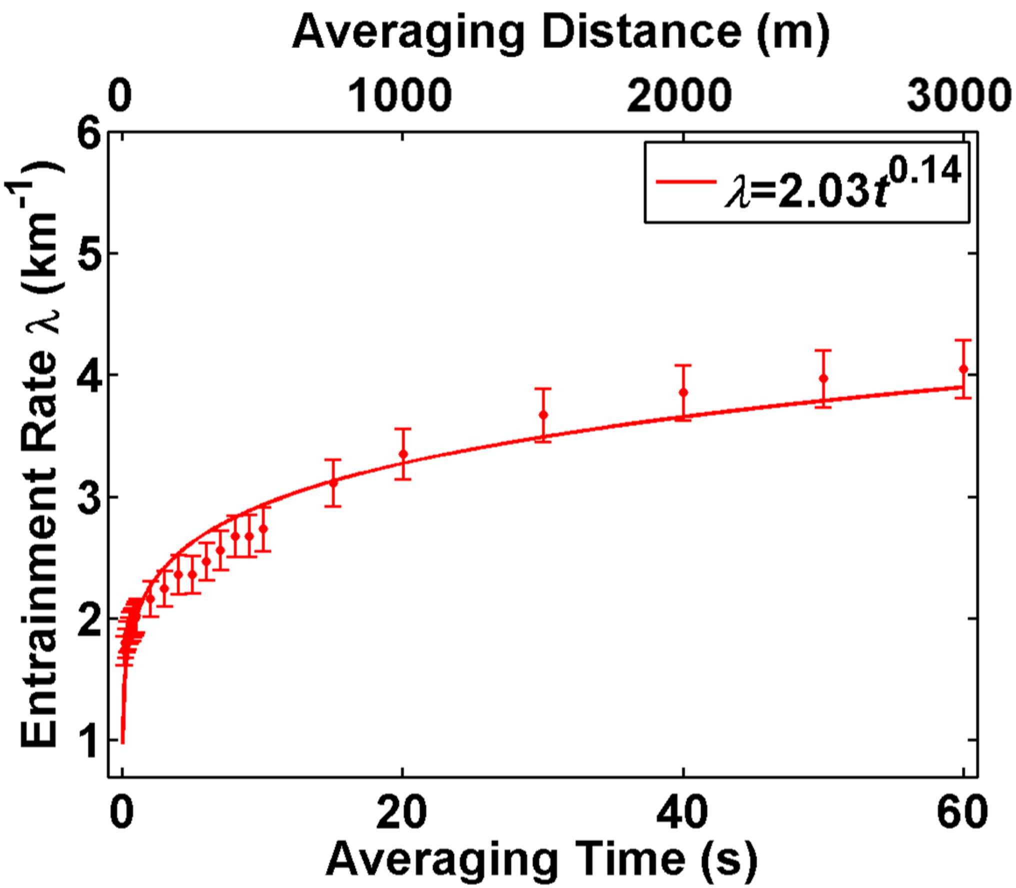
Scale-Dependence of Entrainment Rate





Entrainment rate increases with increasing averaging scales, probably due to higher chance to sample bigger entrained parcels. Mixing mechanism apparently approaches extreme inhomogeneous mixing with increasing averaging scales, mainly due to (1) dilution (Baker 1984) and (2) bigger entrained parcels (Lehmann et al. 2009).

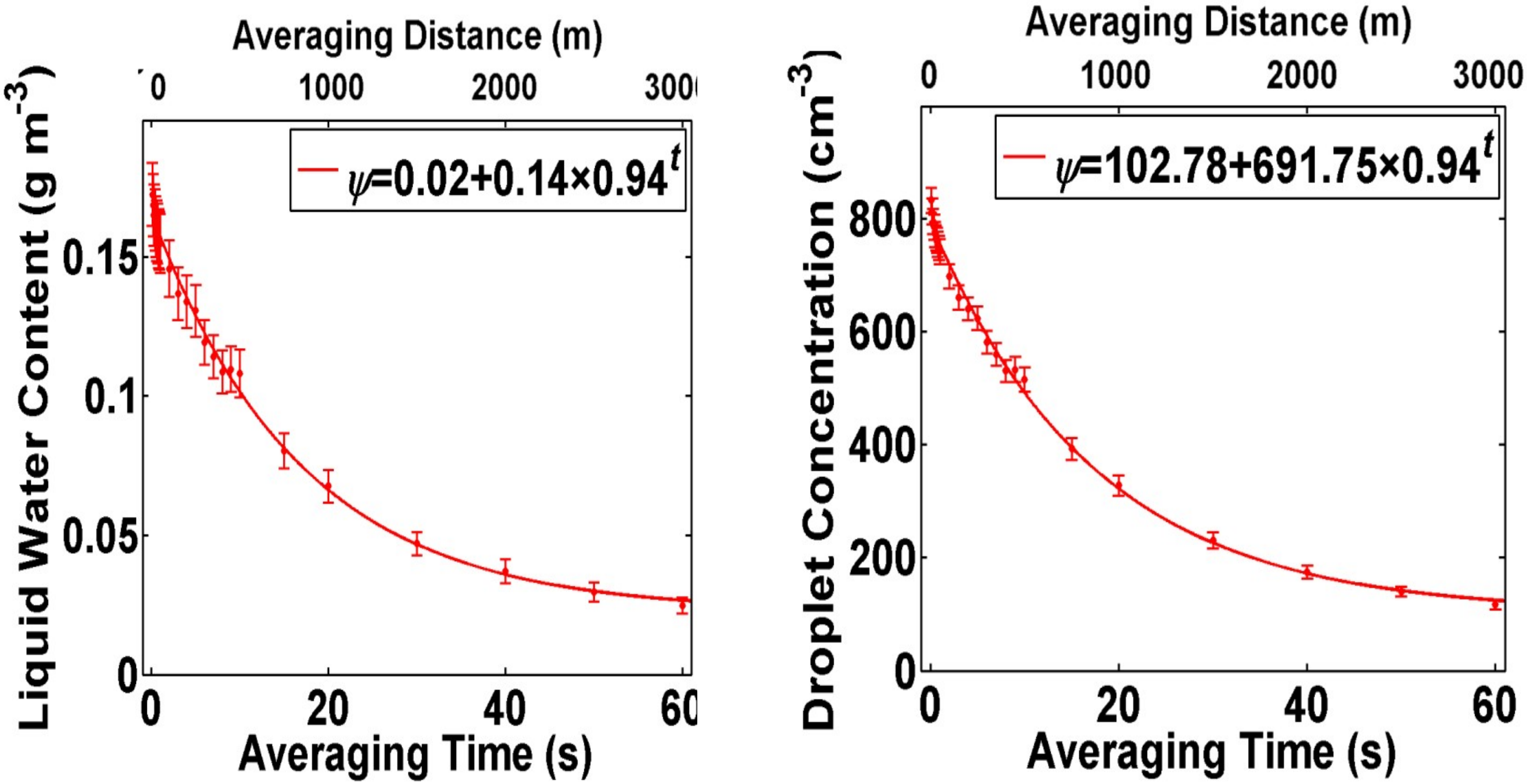
Scale-Dependence of Entrainment Rate



See Lu et al (GRL, 2012) for approach to estimating entrainment rate

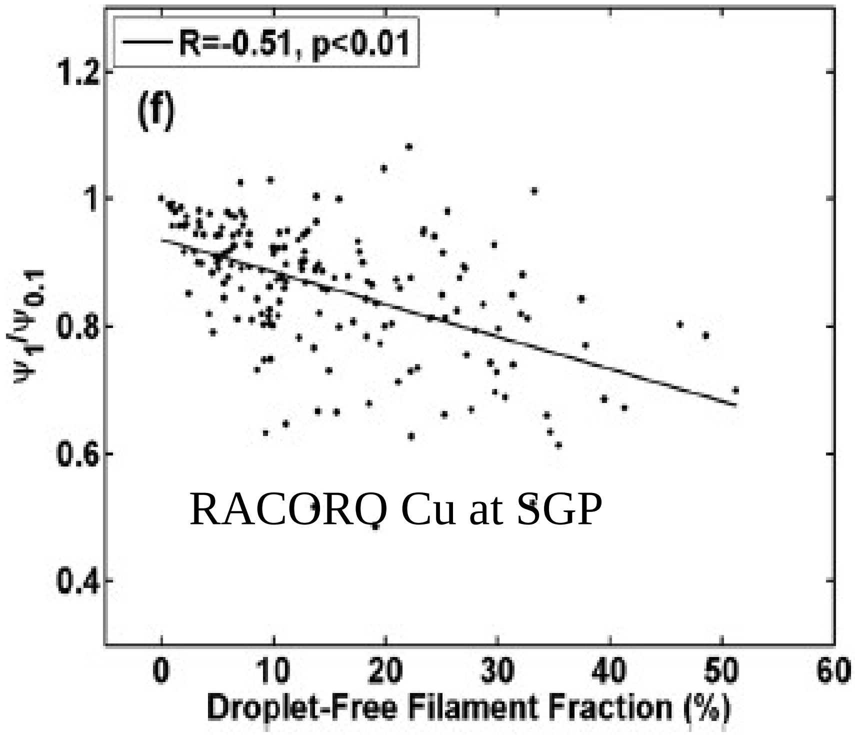
Entrainment rate increases with increasing averaging scales, probably because of increasing chance to sample bigger entrained parcels.

Scale-Dependence of Microphysics



More dilution and evaporation due to entrained dry air leads to decreases of LWC and droplet concentration with increasing averaging scales.

Important Processes from 1mm to 100m



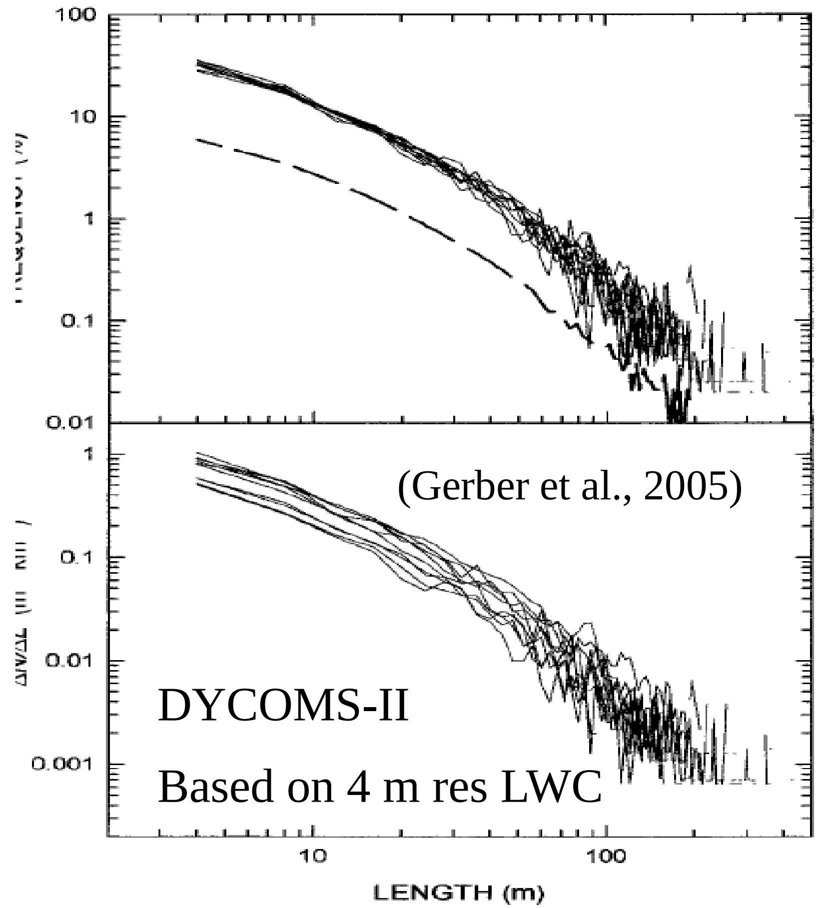
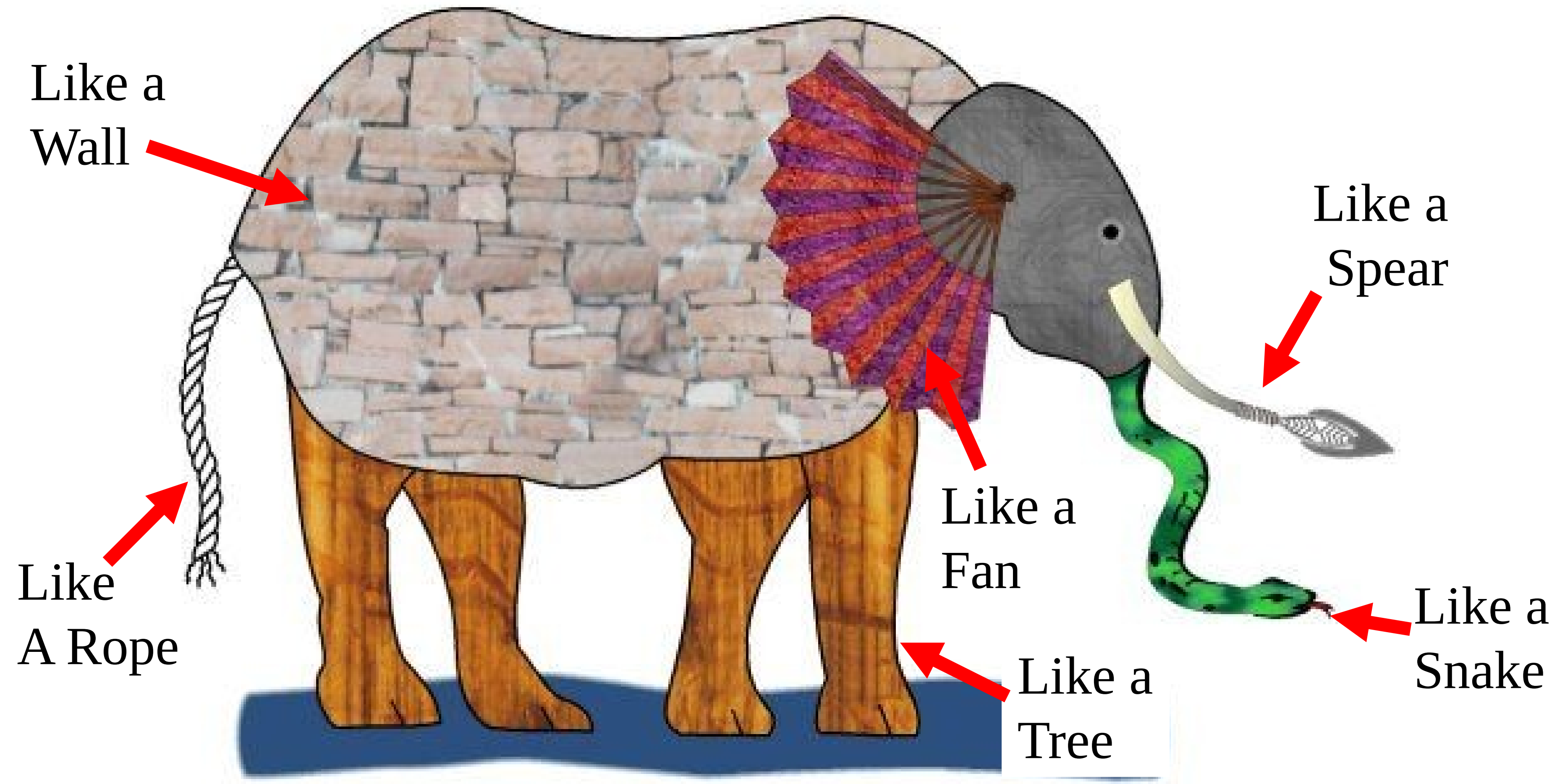


FIG. 5. (top) The frequency distribution of hole lengths for all nine flights measured on average 100 m below cloud top and the best fit dashed curve (shifted for clarity) of lengths derived from a lognormal distribution of widths, and (bottom) the concentration N of holes per kilometer as a function of length for all flights, $\Delta L = 4$ m.

Critical processes occur across scales from ~ 100 m to 1 cm to 1 mm, posing challenges to modeling (sub-LES challenge).

Six Blind Men and the Elephant I



(after John Godfrey Saxe's (1816-1887) version of the Indian legend)

Each was partly right; however, all were wrong about the whole ! → Importance of scale!

Systems Theory on Atmospheric Particle Systems:

Part I: Most Probable Size Distributions

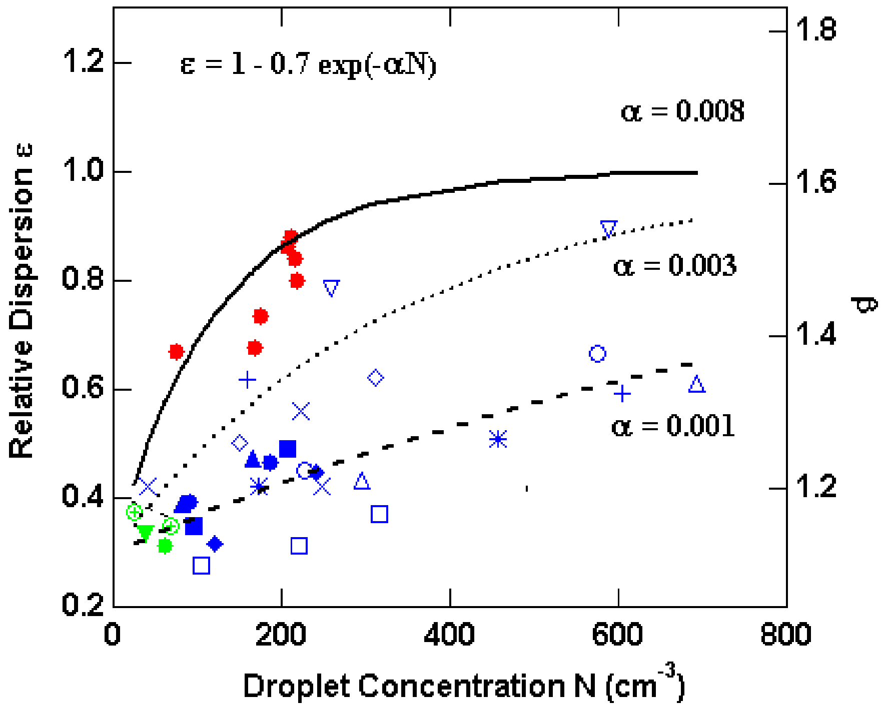
(Liu et al., AR, 1994, 1995; Liu & Hallett, QJ, 1998; JAS, 1998, 2002; Liu et al, 2002)

Part II: On Rain Initiation and Autoconversion

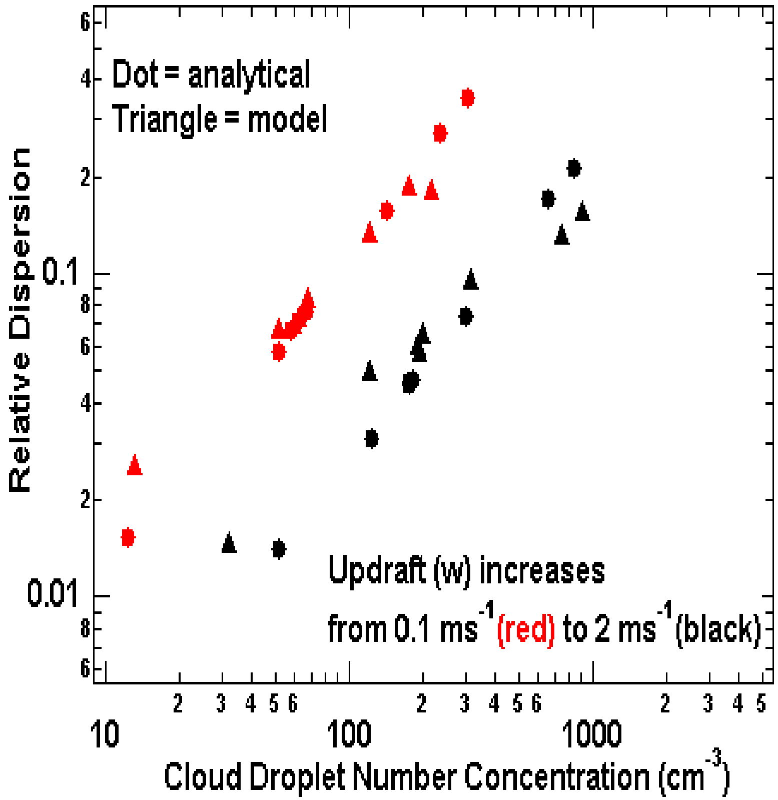
(McGraw and Liu, PRL, 2003, PRE, 2004; Liu et al., GRL, 2004, 2005, 2006, 2007, 2008)

Aerosol-enhanced dispersion causes a warming effect on climate

Decreasing cloud reflectivity



Increasing cloud reflectivity



Enhanced dispersion has a warming effect that offsets the traditional 1st indirect effect by 10-80%, depending on the ε-N relationship (Liu & Daum, Nature 2002; Liu et al. 2006, GRL).

Scales Involved

ce events

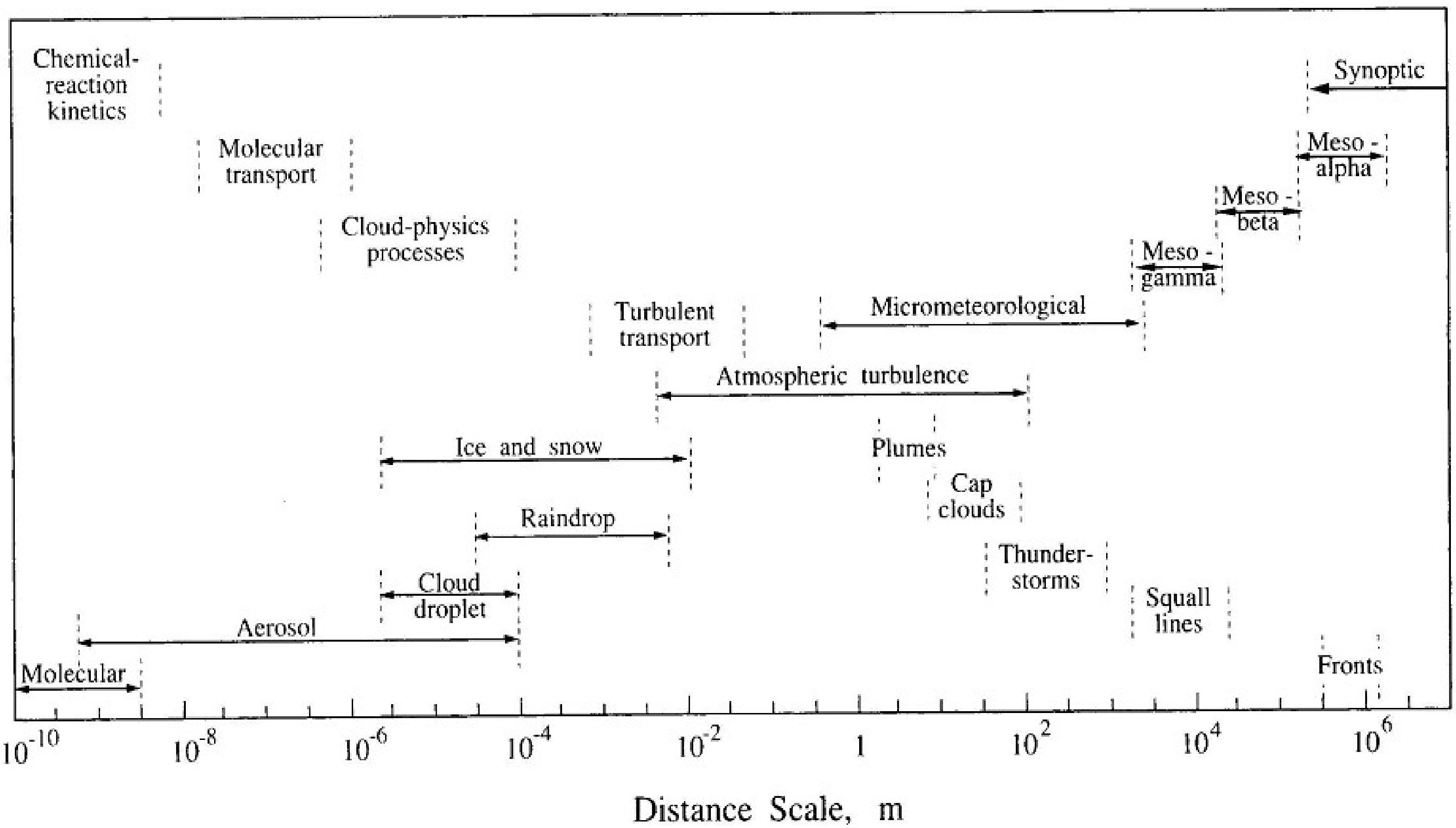


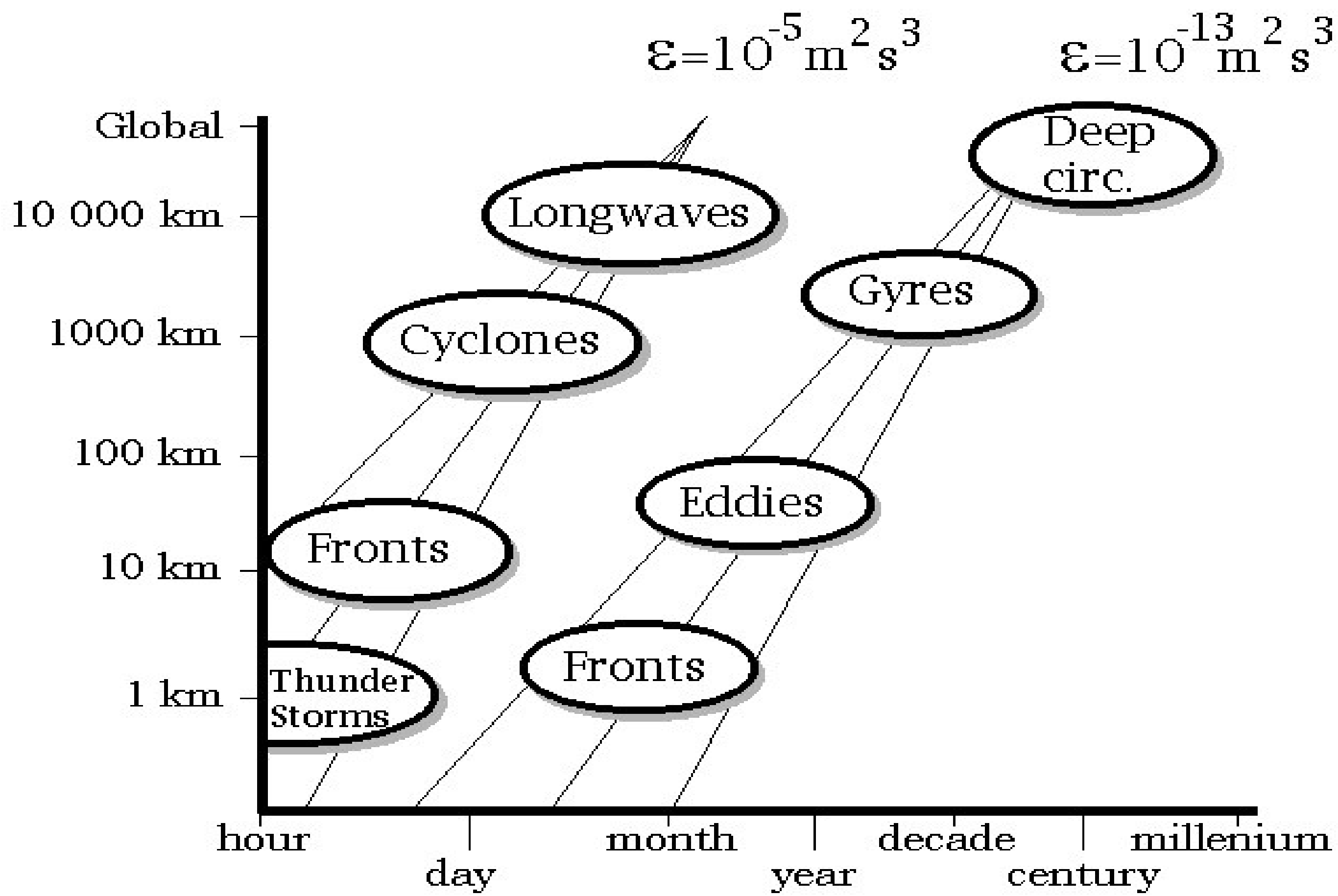
FIGURE 20.2 Lengthscales significant to wet scavenging processes (U.S. NAPAP, 1991)

are

involved in phenomena related to aerosols, clouds, and climate.

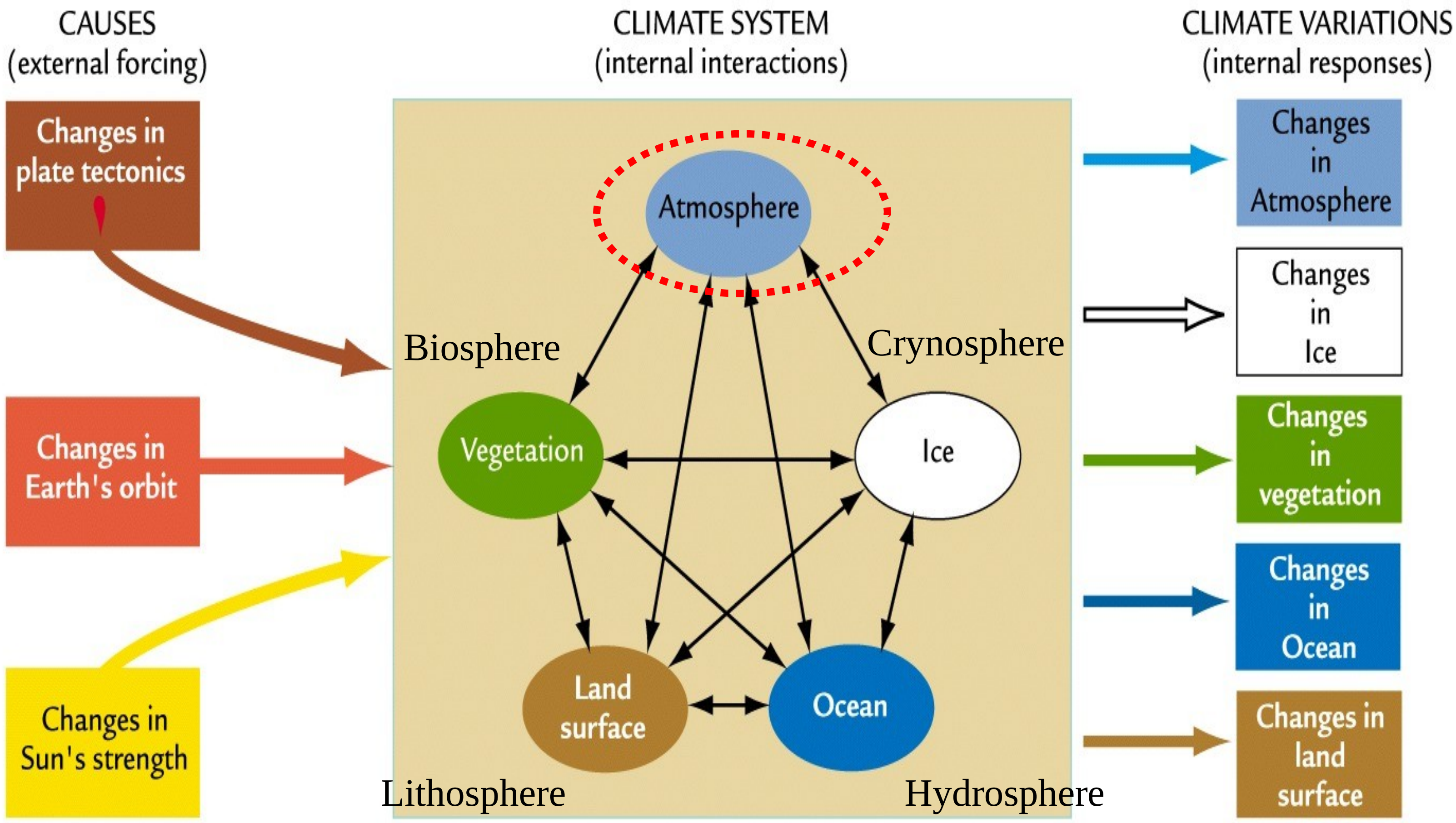
Stommel Diagram for Atmosphere and Oceans

Atmosphere Ocean



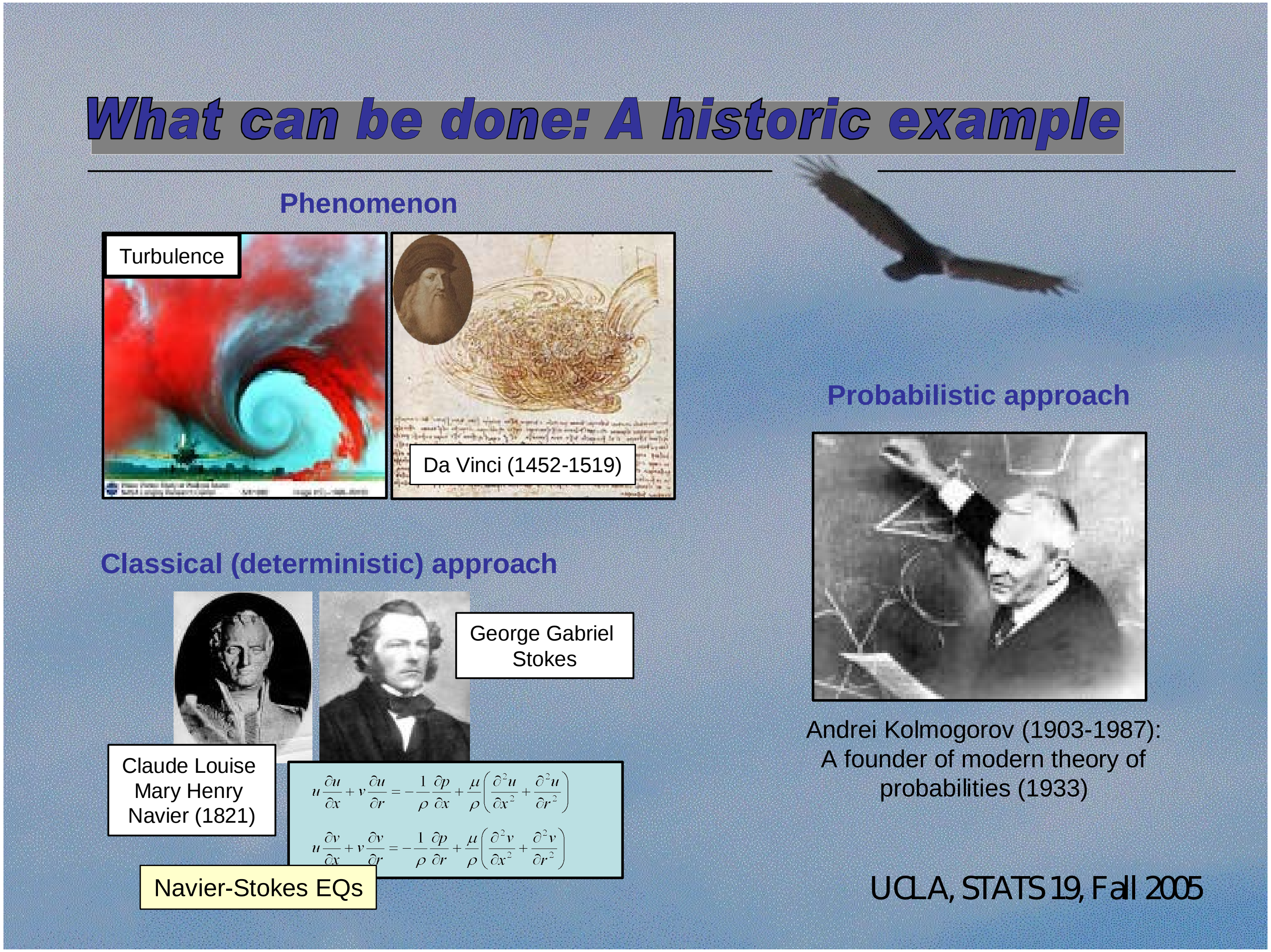
onship

Five Major Climate Components

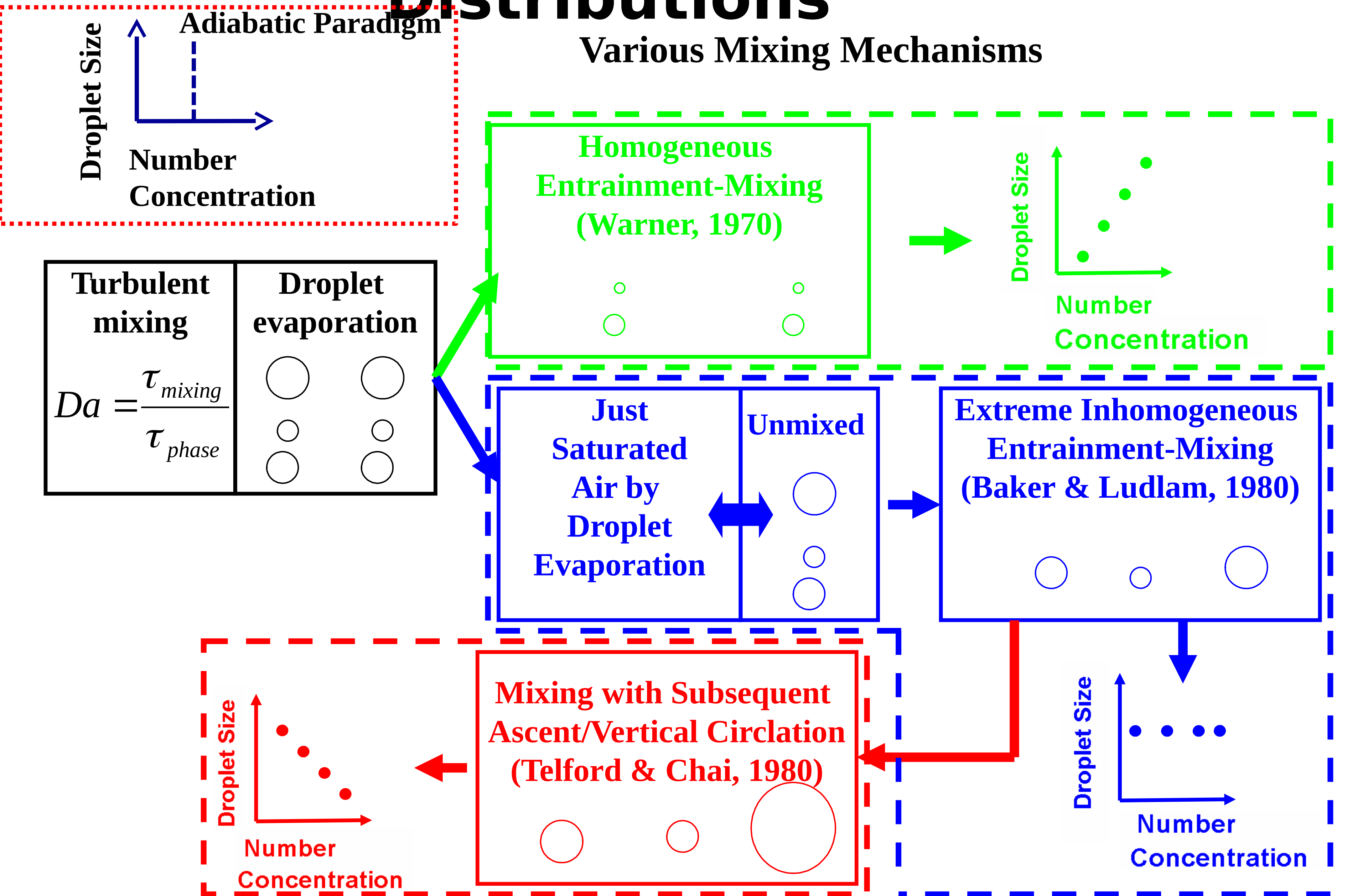


Recall Yin -Yang, and the five elements in traditional Chinese Philosophy and Medicine

The Great Machine of Turbulence

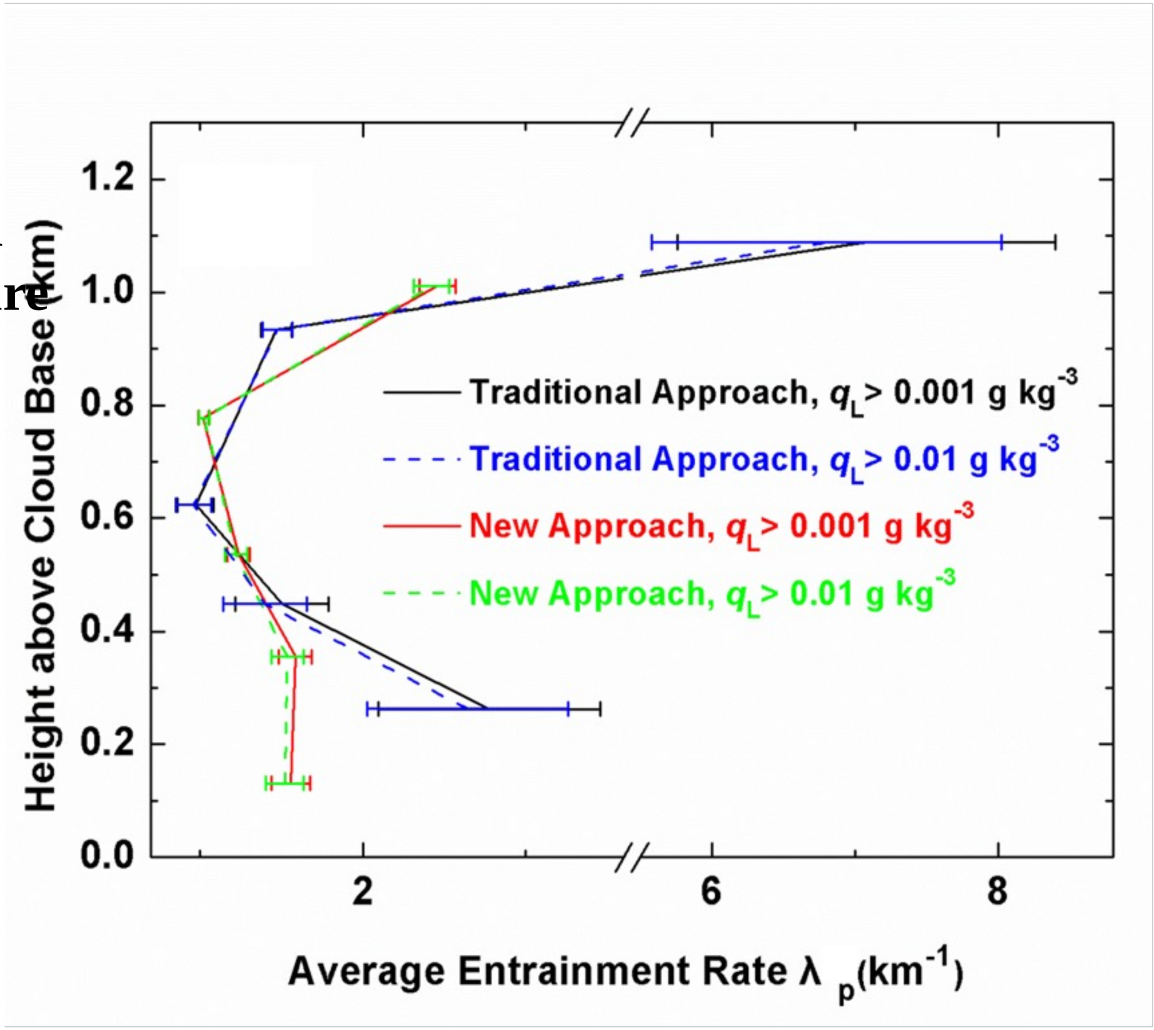


Zoo of Mixing Types & Size Distributions



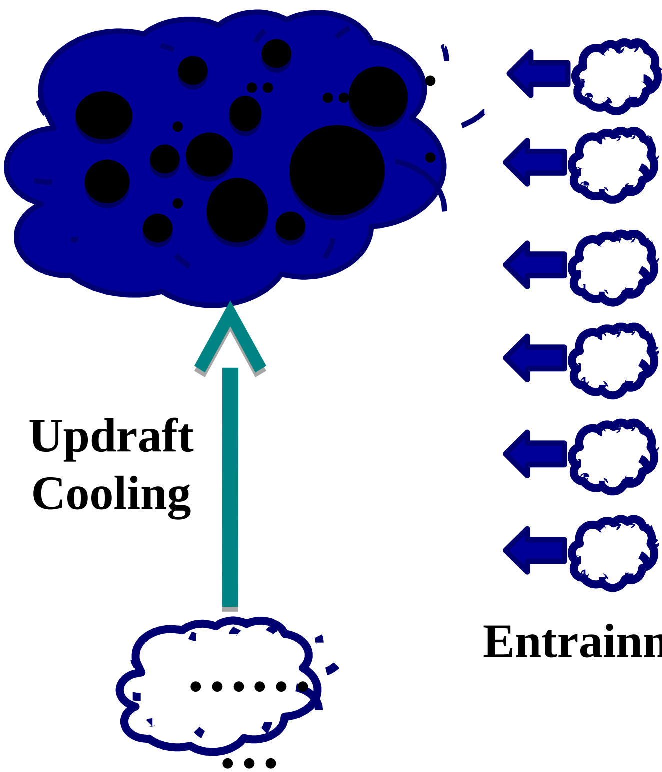
Entrainment Rate: New Approach

- Critical to convection parameterization
- Eliminate need for in-cloud measurements of temperature and water vapor
- Have smaller uncertainty
- Have potential for linking entrainment dynamics to microphysical effects
- Have potential for remote sensing technique (underway)



(Lu et al , GRL, 2012)

New Entraining Cloud Parcel Model



Built on adiabatic version
(Chen et al., GRL, 2016)

- Explicit Microphysics
  - Particle Size
  - Particle Concentration
  - Chemical Composition

Representation of Entrainment

- **Entrainment Rate:**

$$\lambda = \frac{1}{m}\frac{dm}{dt}$$

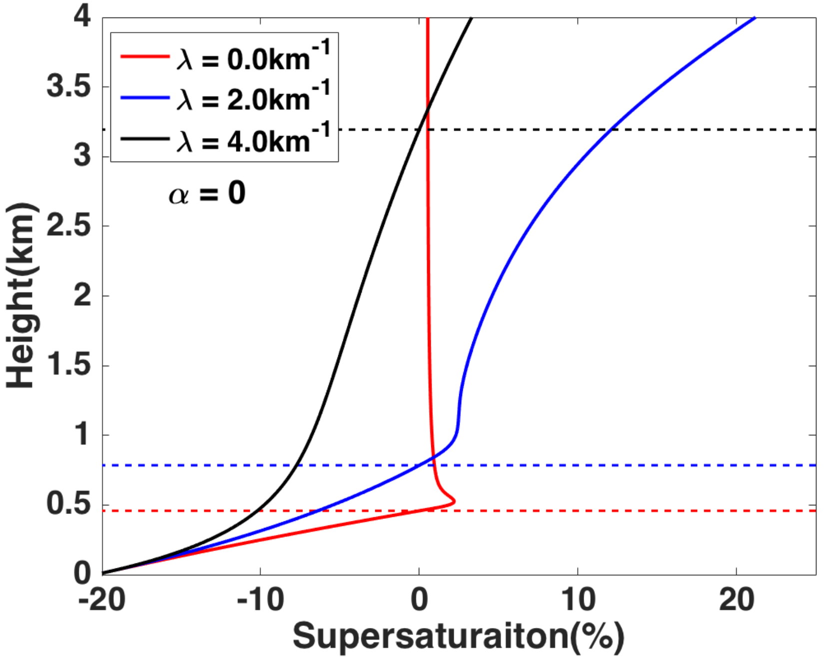
- **Entraining Aerosol Effects:**

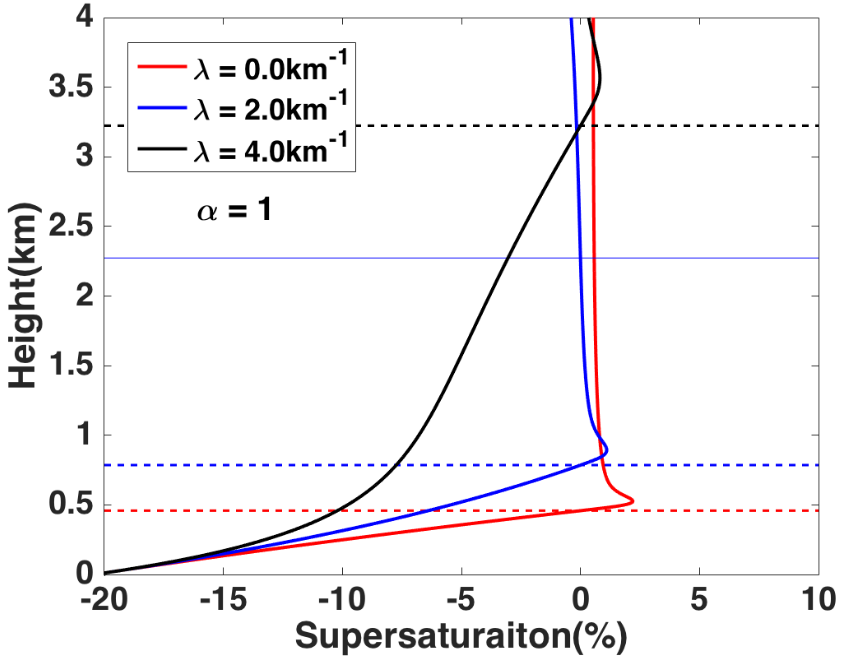
$$\frac{dN_{a,i}}{dt} = -\lambda\, w\, (N_{a,i} - N_{e,i}) = -\lambda\, w\, (1-\alpha) N_{a,i}$$

α=0 : no entrained aerosols

α=1 : entrained aerosols balance the dilut

Effects of Entrainment-Mixing on Supersaturation Profile



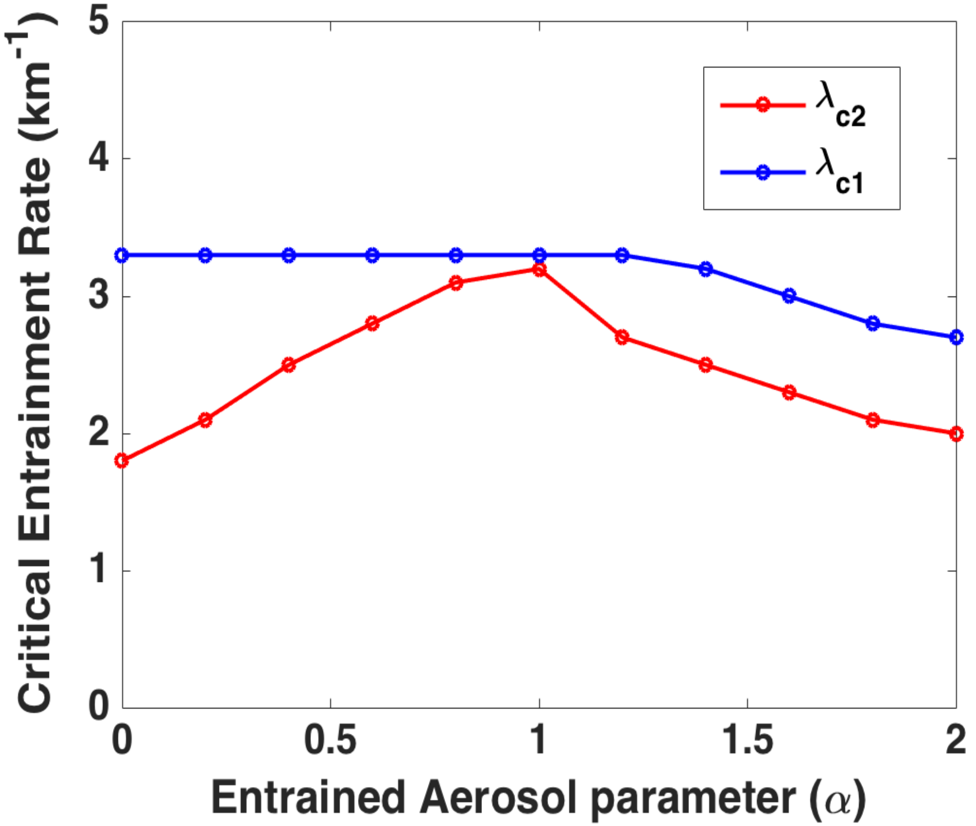


Two Critical Entrainment Rates

- **Barahona and Nenes [2007] defined a critical entrainment rate (λ_{c1}), beyond which droplet can not be activated (essentially no clouds or fog)**

- **We introduce a new critical entrainment rate (λ_{c2}) , beyond which peak supersaturation can r be reached before significant collection or ice processes (auto-conversion threshold < 0.5 and air temperature > 265.15K) (key to ACI parameterization for the altitude of maximum s.**

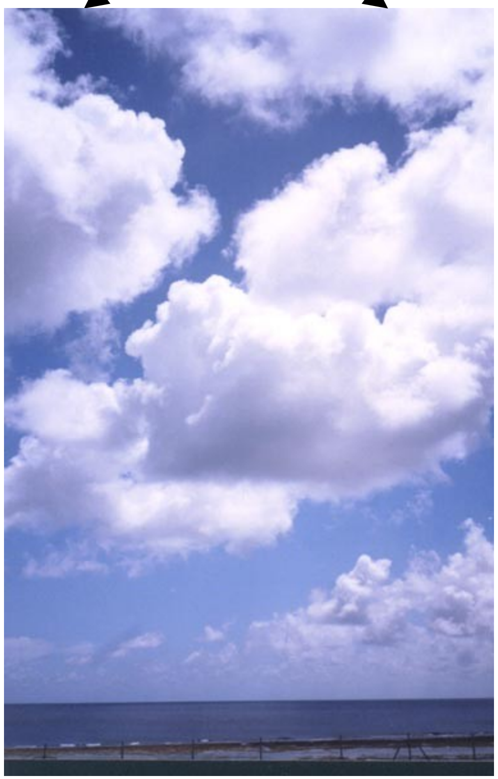
- **The 1st critical entrainment rate always larger than the 2nd.**



Interesting effect of α!

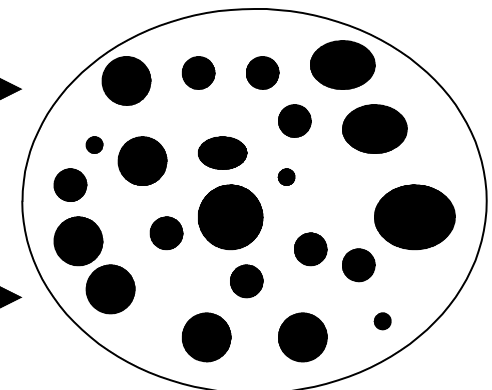
Clouds are systems of water droplets

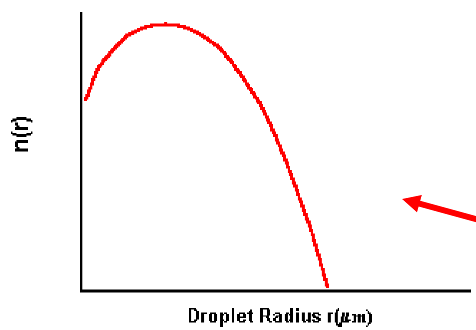
Macroscopic view of clouds
is an optical manifestation of cloud particles



Mean droplet radius
~ 10 micrometer

Microscopic Zoom-in



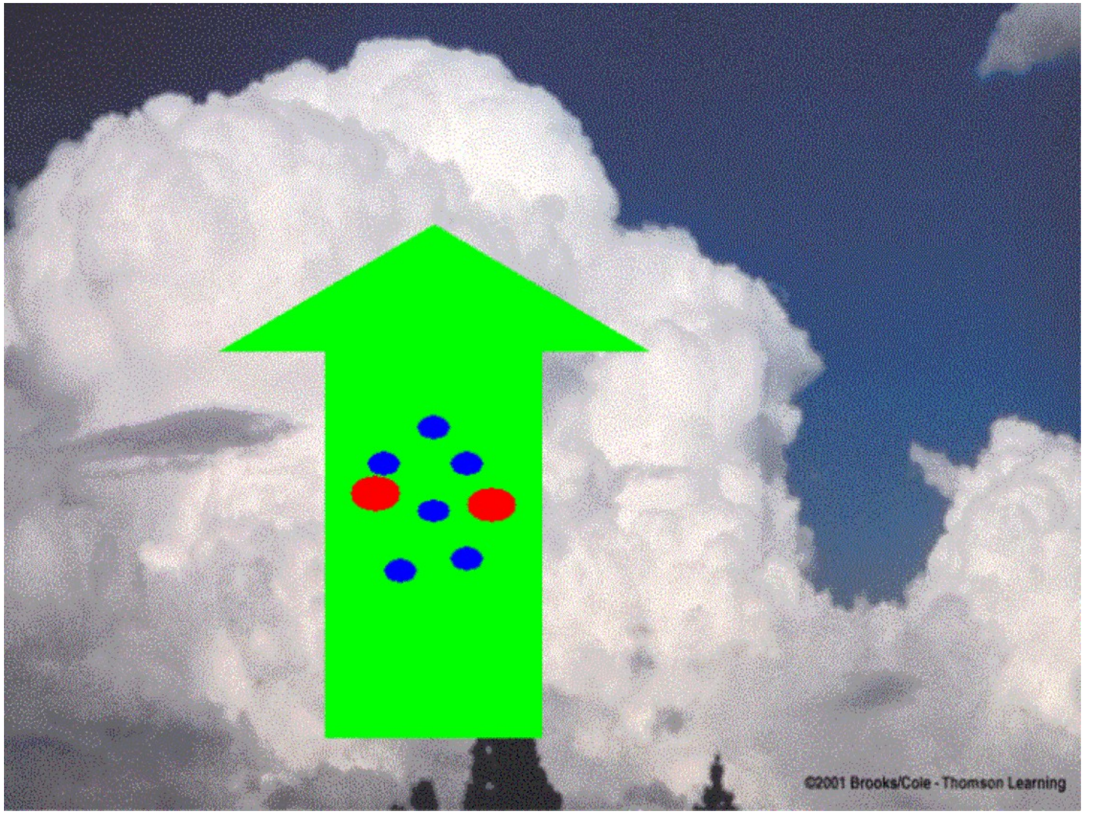


n(r)
($cm^{-3}\mu m^{-1}$)

A central task of cloud physics is surrounding droplet size distribution, n(r). More modes in precipitating clouds ….

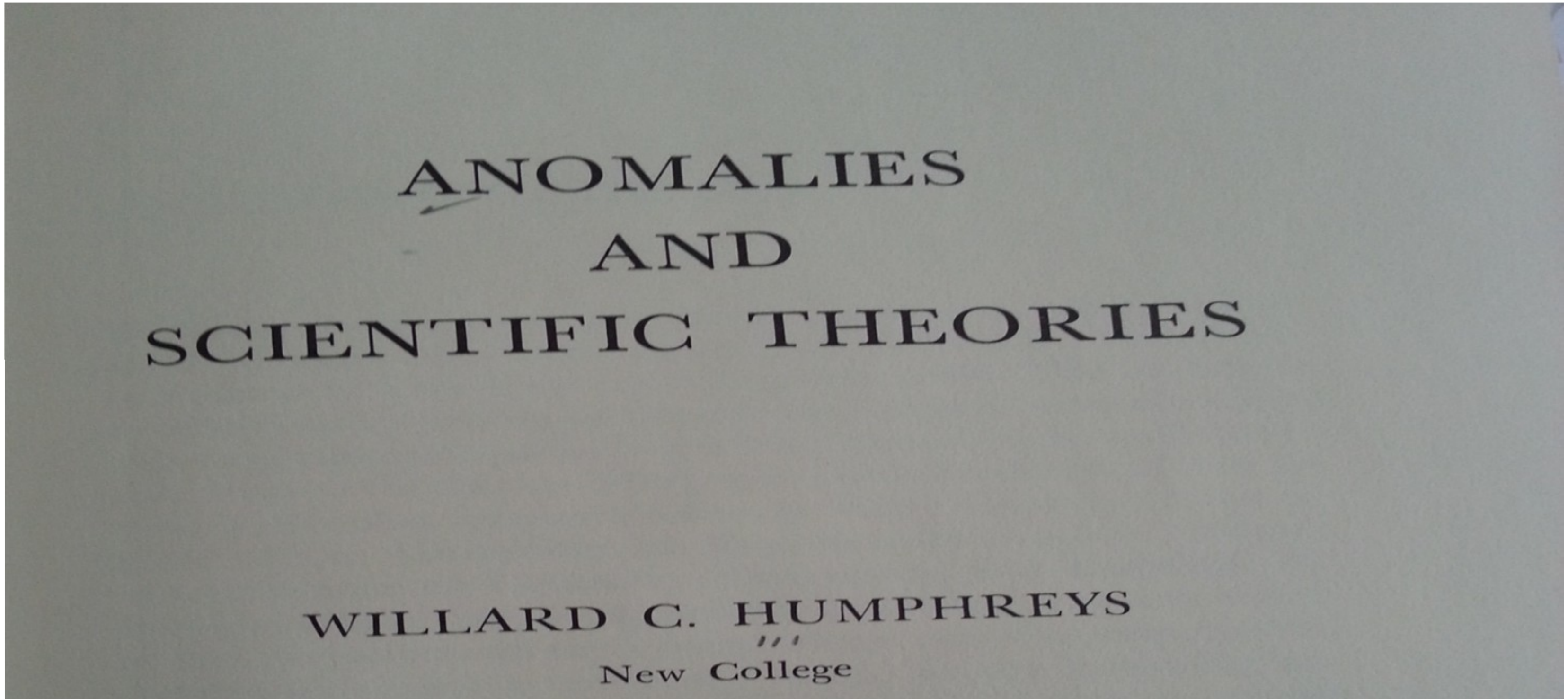
Three ingredients are needed to make clouds

- **Water vapor or supersaturation (thermodynamics)**
- **Updraft that lifts and cools the moist air (dynamics)**
- **Aerosol particles that act as centers onto which water vapor condenses (weather modification; AIE)**



Missing in this simplified picture is turbulence, which, together with related processes, is essential to solve out-standing anomalies in cloud physics !

Scientific Anomaly


ANOMALIES
AND
SCIENTIFIC THEORIES

WILLARD C. HUMPHREYS
New College

"Both the logical structure of scientific theories and their historical evolution are organized around the identification, clarification and explanation of anomalies"

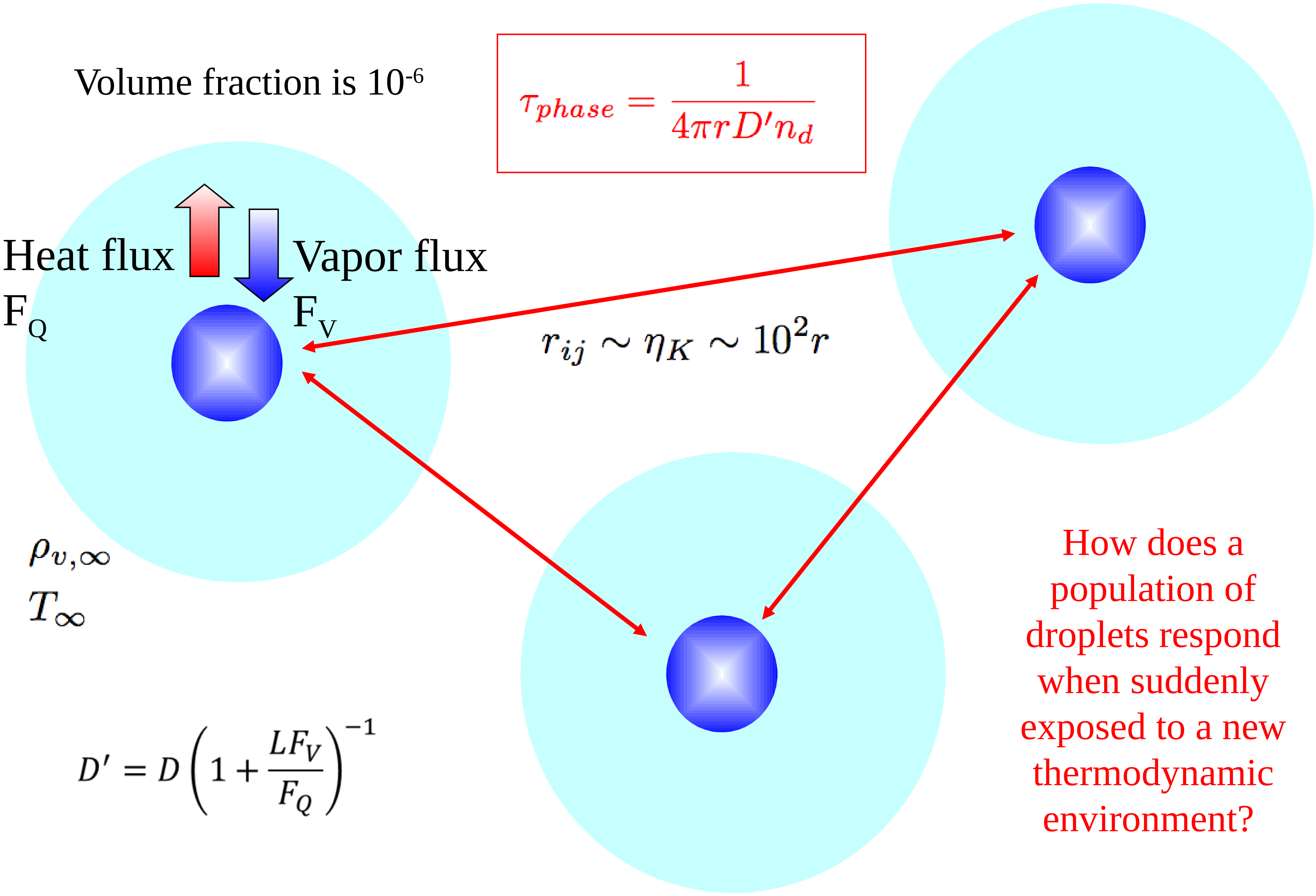
Anomaly: a fact, or event demands explanation -- disagreement between theory and observation >>

-2 outstanding anomalies surrounding (warm) clouds

Time scale for phase relaxation

Volume fraction is 10^{-6}

$$\tau_{phase} = \frac{1}{4\pi r D' n_d}$$



$$D' = D\left(1 + \frac{LF_V}{F_Q}\right)^{-1}$$

How does a population of droplets respond when suddenly exposed to a new thermodynamic environment?

Most Probable Distribution w.r.t. Hamiltonian Variable

Connection to the systems theory
the most probable distribution with respect to x is

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

Under the assumption of conserved liquid water content,
x is the droplet mass. In other words, the most probable
Distribution can be written as

$$\mathbf{n_g = \frac{N}{\alpha}\exp\left(-\frac{g}{\alpha}\right)}$$

Note the difference in the value of α in the two equations

Detailed Balance

Under equilibrium, detailed balance gives:

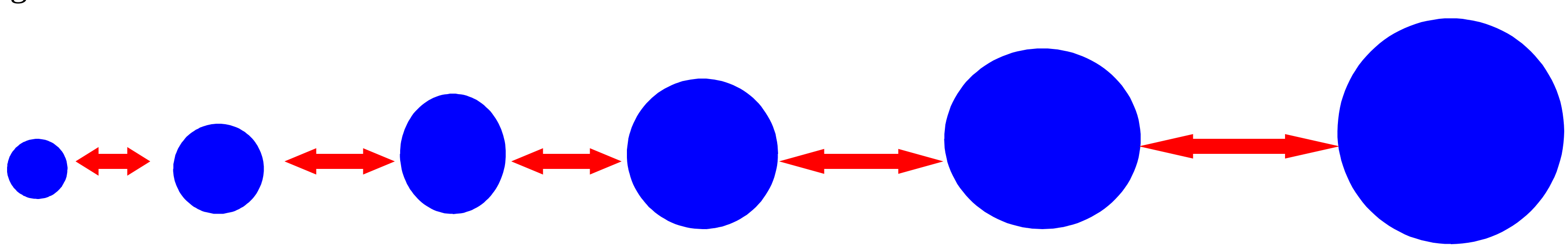
$$\beta_g n_g = \gamma_{g+1} n_{g+1}$$

$$n_g = n_1 \frac{n_2}{n_1}\frac{n_3}{n_2}\cdots\frac{n_g}{n_{g-1}} = n_1 \prod_{i=1}^{g-1} \frac{\beta_i}{\gamma_{i+1}}$$

β_g (s^{-1}) = rate of monomer condensation on the g-drop

γ_g (s^{-1}) = rate of monomer evaporation of the g-drop

n_g (cm^{-3}) = constrained equilibrium concentration of g-drop

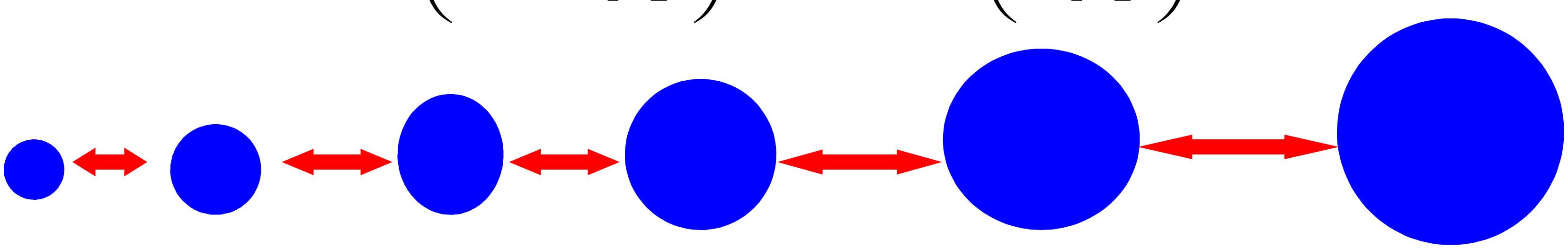


Advantages of Kinetic Potential

The kinetic potential is equivalent to the reduced thermodynamic potential in nucleation theory. However, the kinetic potential is a more general concept in that it is based on rate constants, and well defined even in the absence of equilibrium condition.

Next, we will use the kinetic potential to study the rain initiation.

$$\Phi(g) = -\ln\left(\prod_{i=1}^{g-1}\frac{\beta_i}{\gamma_{i+1}}\right) = -\sum_{i=1}^{g-1}\ln\left(\frac{\beta_i}{\gamma_{i+1}}\right)$$

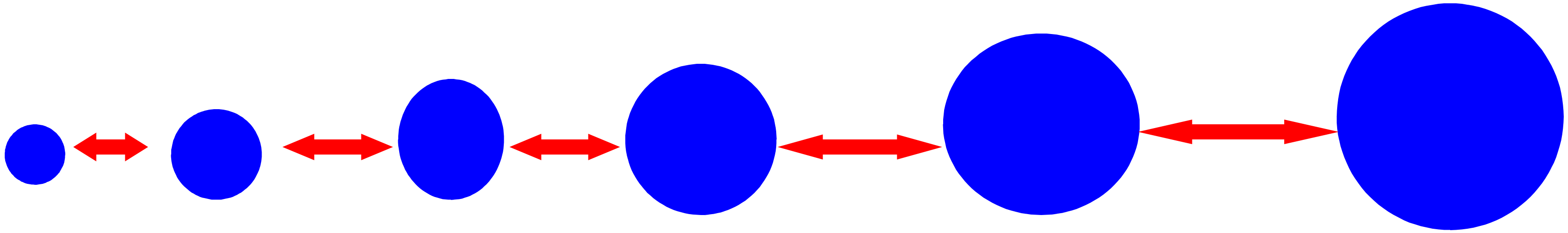


Droplet Growth Rates

The growth of cloud droplets is modeled as a sum of condensation and collection processes:

$$\boldsymbol{\beta}_{\mathbf{g}} = \boldsymbol{\beta}_{\mathbf{g}}^{\mathbf{con}} + \boldsymbol{\beta}_{\mathbf{g}}^{\mathbf{col}}$$

$\boldsymbol{\beta}_{\mathbf{g}}^{\mathbf{con}}$ = Condensation rate; $\boldsymbol{\beta}_{\mathbf{g}}^{\mathbf{col}}$ = Collection growth rate



Long Collection Kernel

The general collection kernel is given by

$$\mathbf{K(R,r) = E\pi\left(R + r\right)^2 \left(V_R - V_r \right)}$$

and its general solution is too complicated to handle.

Long (1978, J. Atmos. Sci.) gave a very accurate approximation:

$$\mathbf{k(R,r) = k_1 R^6} \qquad (10\ \mu\text{m} \le R \le 50\ \mu\text{m})$$

$$\mathbf{k(R,r) = k_2 R^3} \qquad (R > 50\ \mu\text{m})$$

The (gravitational) collection kernel is negligible when R < 10 μm.

Collection Growth Rate

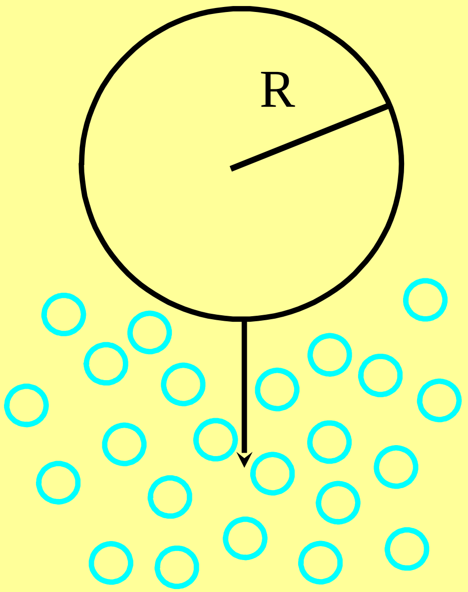
The mass growth rate of the drop is

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

Application of the Long kernel yields the growth rate of the radius R
(10 μm ≤ R ≤ 50 μm):

$$\frac{dm}{dt} = k_1 L m^2$$

$$\beta_g^{col} = \frac{dg}{dt} = k_1 \nu L g^2$$



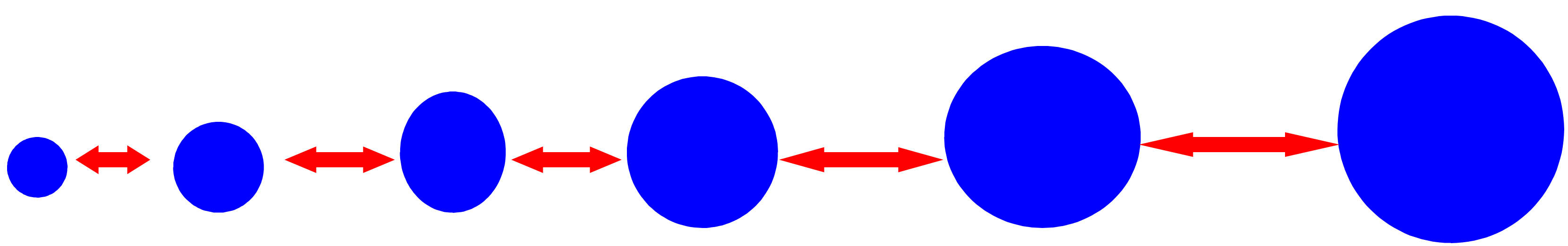
A drop of radius R fall through a polydisperse population of smaller droplets with size distribution n(r).

Relationship between Effective Evaporation Rate and Condensation Rate

Effective evaporation rate is introduced to consider the complex droplet interactions and competition for water vapor such that a typical droplet size distribution is maintained by detailed balance (constrained equilibrium):

$$\frac{n_{g+1}}{n_g} = \frac{\beta_g^{con}}{\gamma_{g+1}} \approx \frac{\beta_g^{con}}{\gamma_g} = \exp\left(-\frac{1}{\alpha}\right)$$

$$\gamma_{g+1} = \exp\left(\frac{1}{\alpha}\right)\beta_g^{con}$$



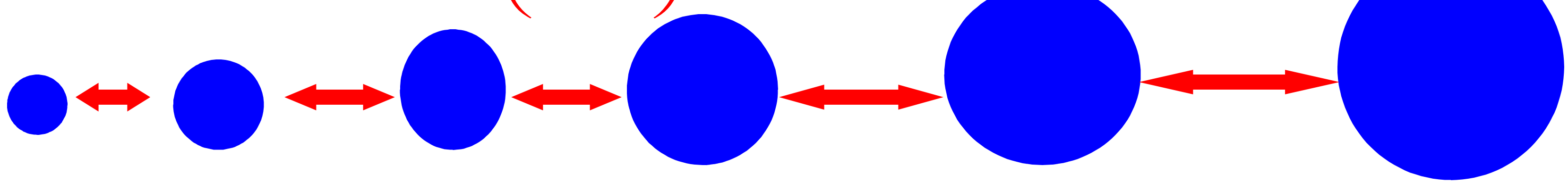
Relating α to L and N

The liquid water content of the droplet system is given by

$$L = v\int g n_g \, dg = v\int \frac{N}{\alpha} g \exp\left(-\frac{g}{\alpha}\right) dg = Nv\alpha$$

$$\alpha = \frac{L}{Nv}$$

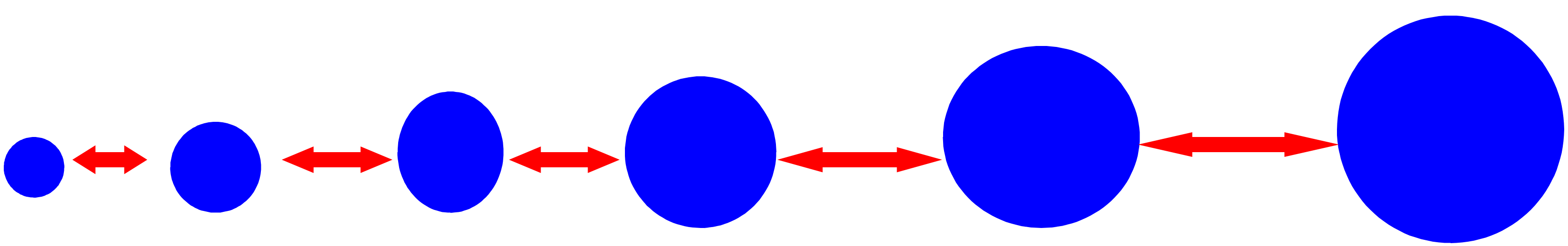
$$\gamma_{g+1} = \exp\left(\frac{vN}{L}\right)\beta_g^{con}$$



Examination of Kinetic Potential

Substituting into kinetic potential equation of the effective evaporation rate and collection growth rate and a typical value of condensation rate, we calculated the kinetic potential as a function of L and N:

$$\Phi(g) = -\sum_{i=1}^{g-1} \ln\left(\frac{\beta_i}{\gamma_{i+1}}\right) = -\sum_{i=1}^{g-1} \ln\left(\frac{\beta_i^{con} + k_1 \nu L i^2}{\exp\left(\frac{\nu N}{L}\right)\beta_i^{con}}\right)$$



Estimation of Condensation Rate Constant

Mean radius of the 6th moment is given

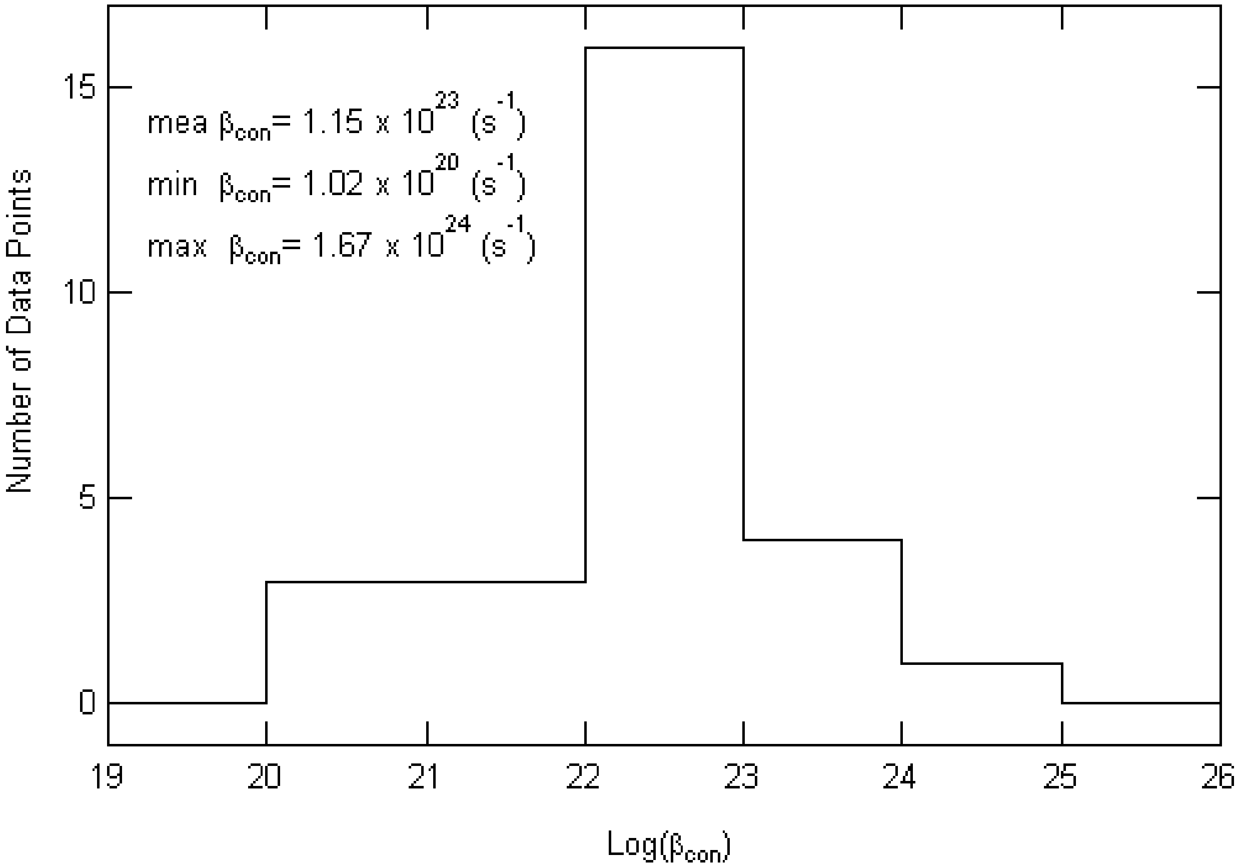
$$\mathbf{r_6 = \left(\frac{3}{4\pi\rho_w}\right)^{1/3} 1.12\left(\frac{L}{N}\right)^{1/3}}$$

According to Liu and Daum (2004, JAS), when $r_6 = r_c$, rain starts:

$$\beta_{con} = \frac{k1.12^6 L_*^4}{(\nu\rho_w)^2 N_*^3}$$

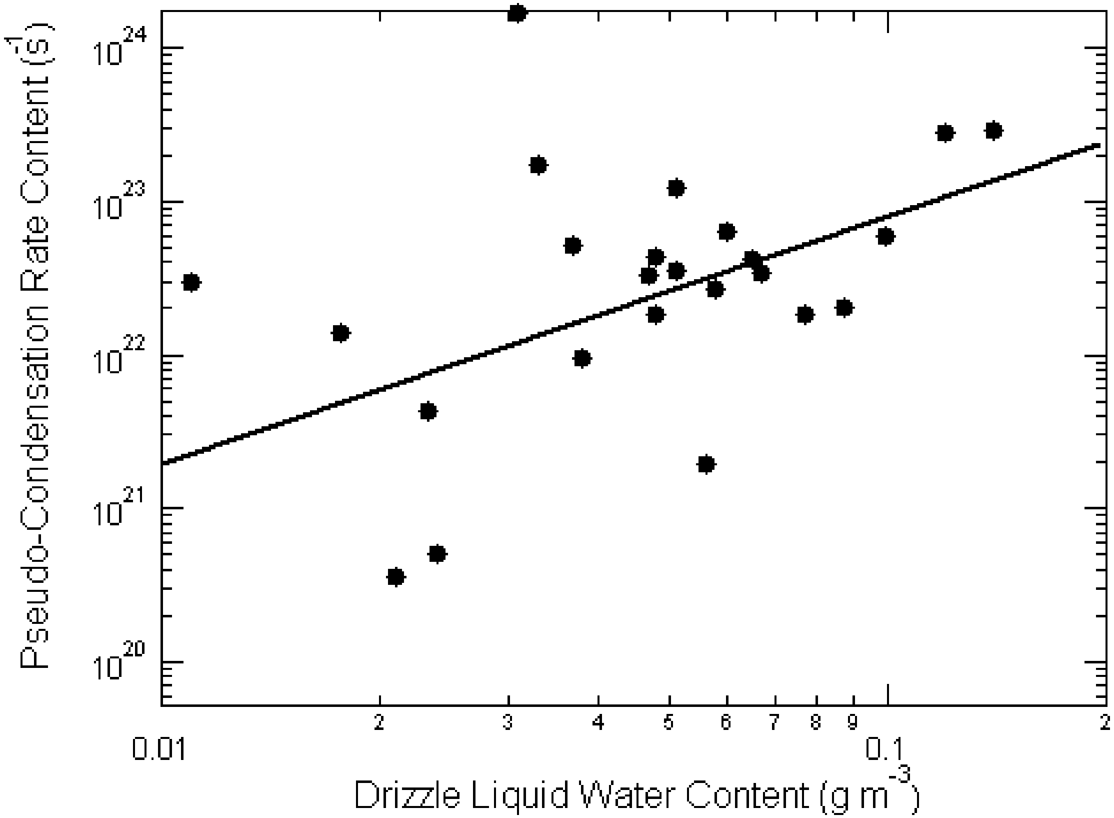
The star denotes that L and N are sampled in drizzling clouds.
(Liu et al. 2003, GRL)

Estimates of Condensation Rate Constant



Liu et al. 2003, GRL

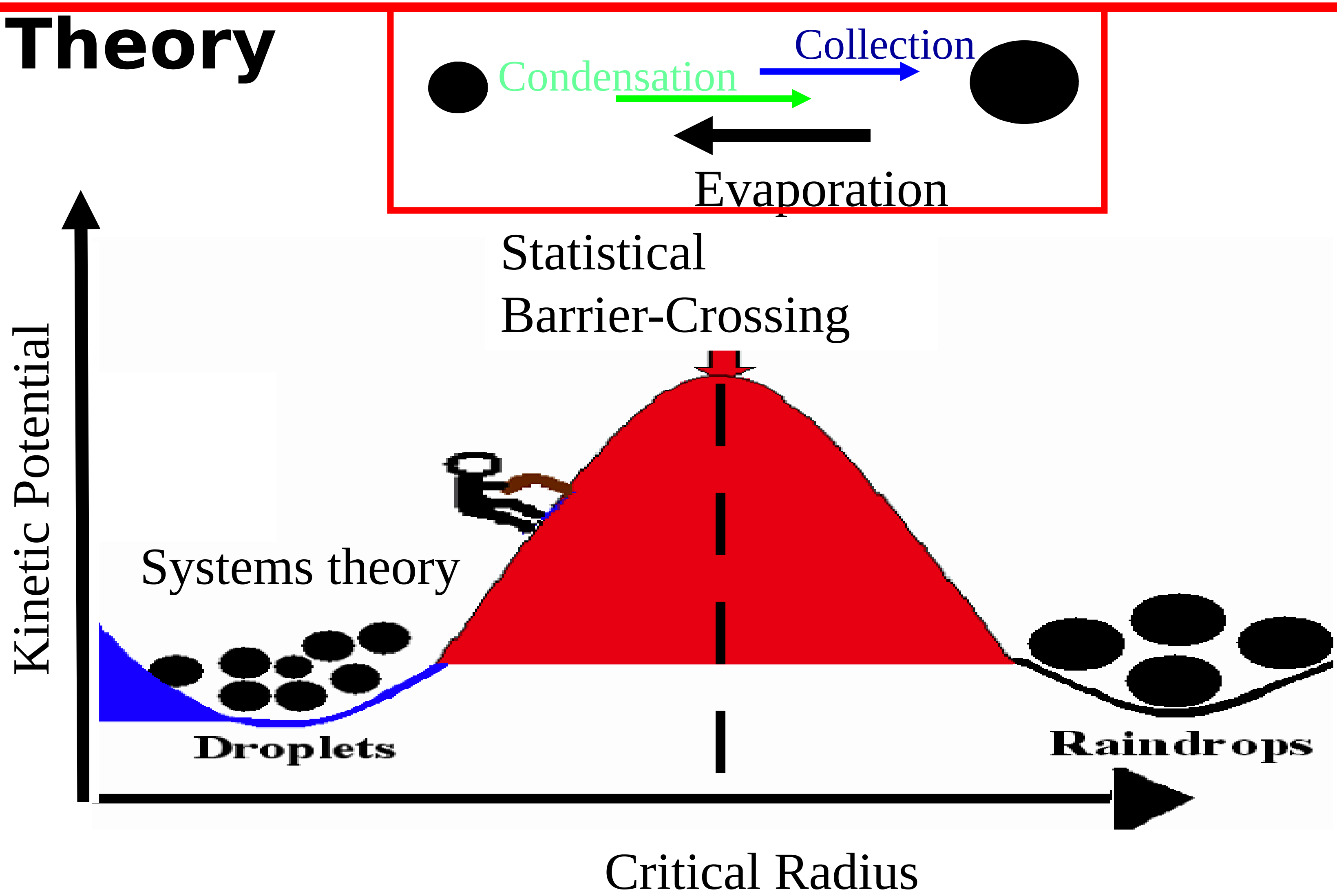
Relationship between Condensate Rate and Drizzle Water Content



Relationship of the pseudo-condensate rate constant to the drizzle water content. The condensation rate constant is chosen at where the fitting line intercepts with the drizzle water content of 0.01 g m^{-3}.

The data are from Yum and Hudson (2002).

Mountain of Life: New Rain Initiation Theory



The new rain initiation theory (kinetic potential theory, KPT) combines statistical barrier crossing with the systems theory for droplet size distributions (McGraw & Liu, Phys. Rev. Lett., 2003; Phys. Rev., 2004), and provides physics for threshold.

Analytical Expressions for Critical Radius

At the critical point, the forward and backward rates are in balance:

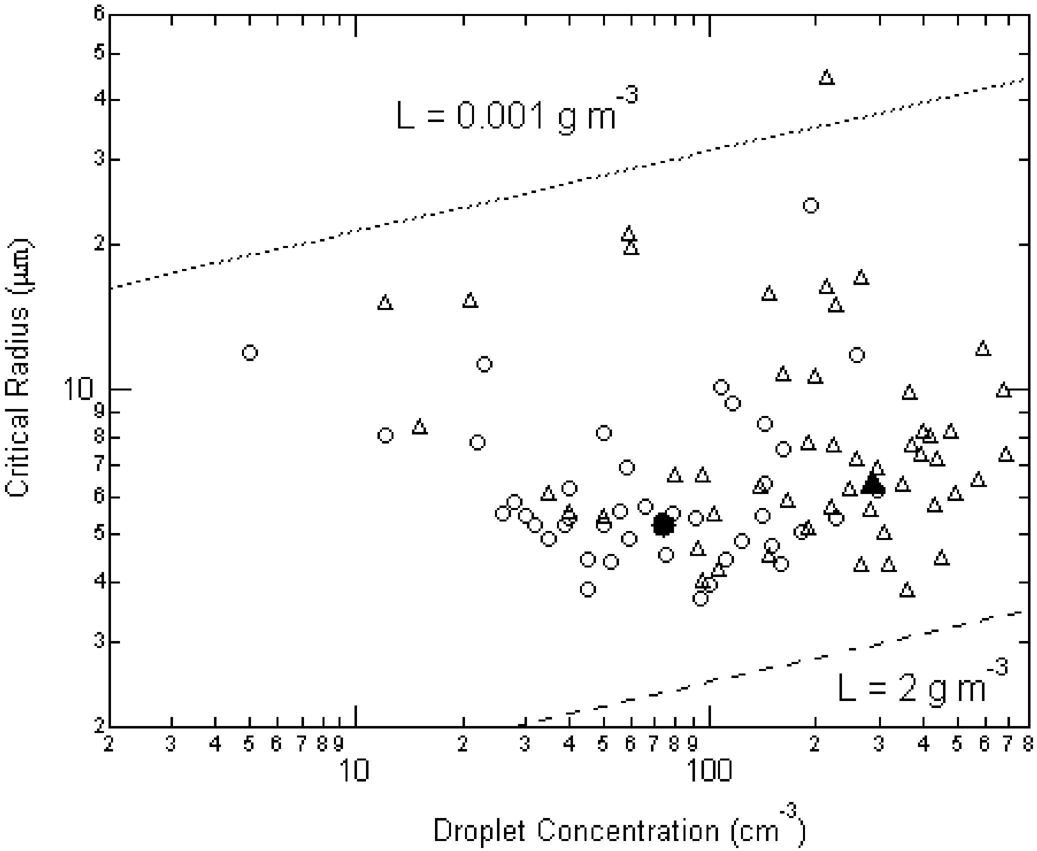
$$\beta^{con} + \beta^{col} = \gamma$$

$$\gamma = \exp\left(\frac{\nu N}{L}\right)\beta^{con}$$

$$\beta_g^{col} = k_1 \nu L g^2$$

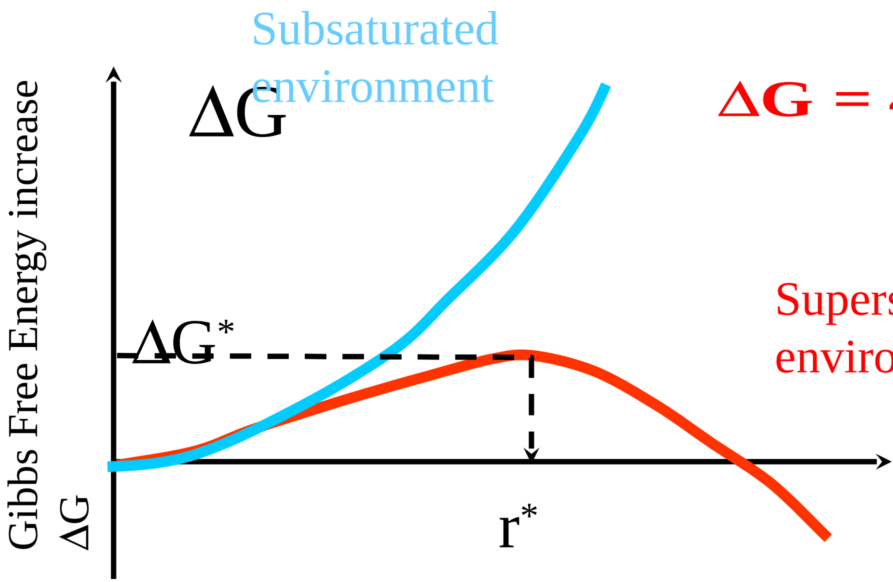
$$r_c = \beta_{con}\left\{\left(\frac{3\nu}{4\pi}\right)^2 \frac{2}{\kappa}\, 0.99\, \frac{N}{L^2}\right\}^{1/6} = N\left(\frac{\;}{L^2}\right)^{1/6}$$

Dependence of Critical Radius on Droplet Concentration and LWC

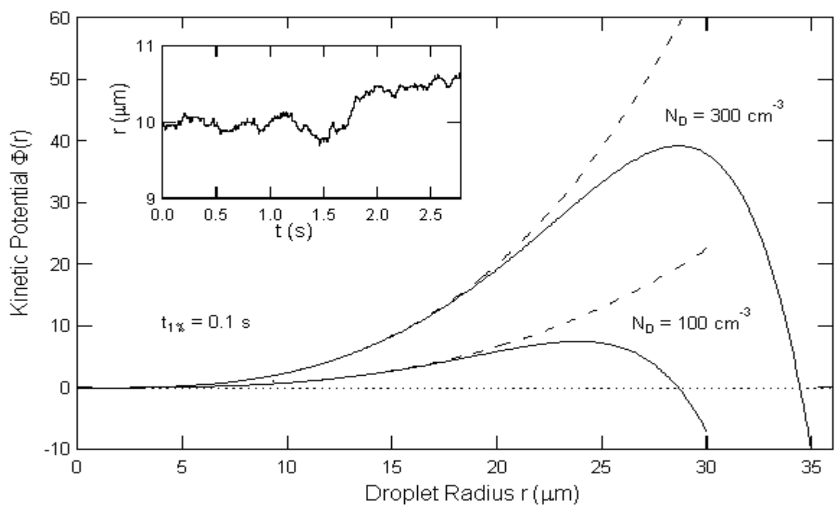


This figure shows that critical radius increases with increasing droplet concentration and decreasing liquid water content. It also shows that on average, continental clouds have a larger critical radius (adapted from Liu et al. 2003, GRL)

Comparison with Nucleation



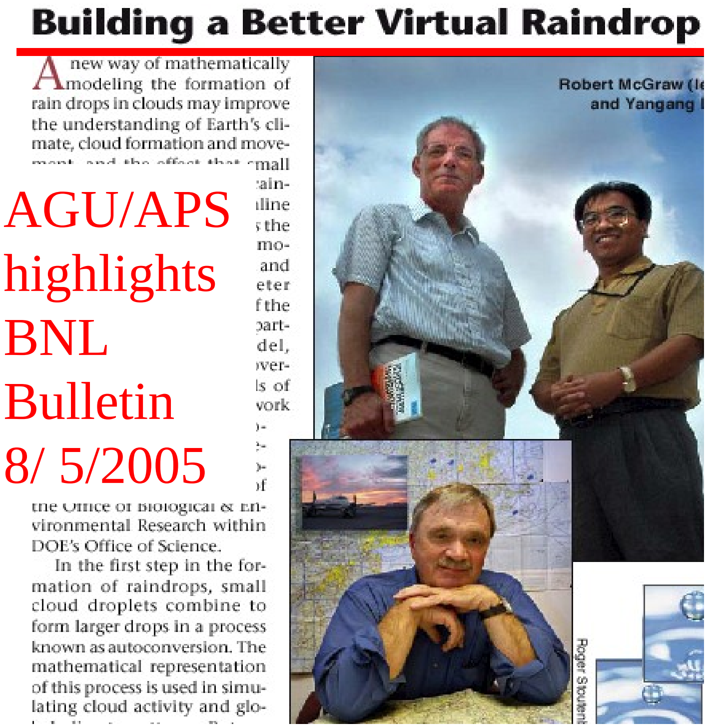
$$\Delta G = 4\pi\sigma r^2 - \frac{4\pi}{3} nkT\ln\left(\frac{e}{e_s}\right) r^3$$



$$\Phi(r) = c_1 r^3 - c_2 r^6$$

Parameters c1 and c2 depend on droplet concentration and liquid water content.

Building A Better Virtual Raindrop



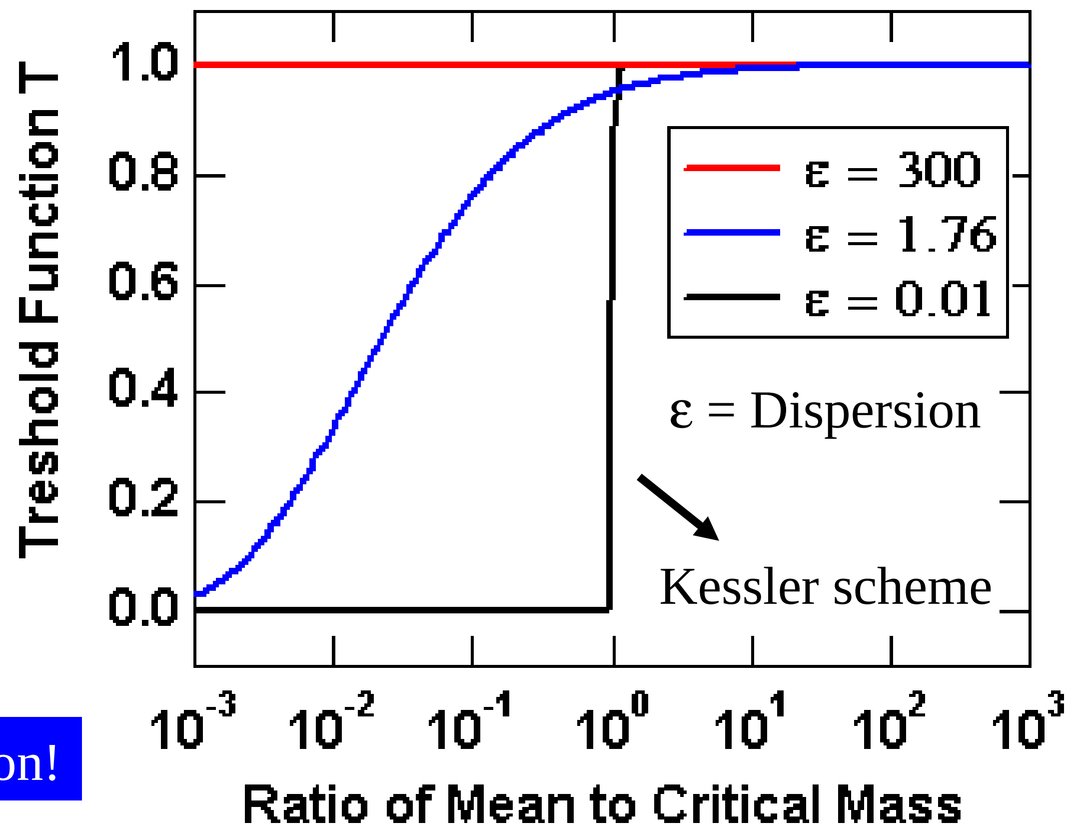
Building a Better Virtual Raindrop

A new way of mathematically modeling the formation of rain drops in clouds may improve the understanding of Earth's climate, cloud formation and move-

the Office of Biological & Environmental Research within DOE's Office of Science.

In the first step in the formation of raindrops, small cloud droplets combine to form larger drops in a process known as autoconversion. The mathematical representation of this process is used in simulating cloud activity and glo-

AGU/APS highlights
BNL Bulletin
8/ 5/2005



Note the importance of dispersion!

Combining this new rain initiation theory with theory for collision and coalescence of cloud drops leads to a suite of theoretical autoconverison parameterizations (Liu & Daum, JAS, 2004; Liu et al., GRL, 2004, 2005, 2006, 2007, 2009).

Mult chy

Big Whorls have little Whorls that

feed on their velocity;

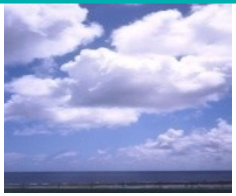
And little whorls have smaller whorls,

and so on to viscosity (e.g., 1 mm)

Footnote in a Numerical Weather Forecasting by Numerical Process by L.F. Richardson 1922

Time Scal

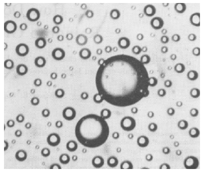
Global



Cloud system

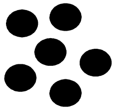
Cloud

Turbulent Eddy



Droplet

Aerosol



Molecule

Space Scale