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

Yangang Liu (lyg@bnl.gov)
Climate and Process Modeling (CMP) Group
Brookhaven National Laboratory (BNL), USA

Thanks: Chunsong Lu (NUIST), Zheng Gao (SBU), Jingyi Chen (PNNL), Satoshi Endo (BNL), Xin Zhou (BNL), Bob McGraw (BNL), Pete Daum (BNL), John Hallett (DRI), Seong Soo Yum (Yonsei U) …

IWCMS, IITM, Pune, India, Aug. 13-19, 2018
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

Aerosol DIRECT effect: Direct reflection of solar radiation back to space

Aerosol INDIRECT effects (AIE,); focus of Today’s talk
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 not represent at all buffering/compensating processes:
  
  • Dispersion effect
  • Non-monotonic dependence (regime dependence)
  • Turbulent Entrainment-mixing processes
  • Process couplings
Dominant View of AIE: **Number Effect**

Twomey (1974, Atmos. Environ): “it is suggested that pollution gives rise to whiter (not darker) clouds -- by increasing the droplet concentrations 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**
“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

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

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

$\varepsilon = 1 - 0.7 \exp(-\alpha N)$

$\alpha = 0.008$

$\alpha = 0.003$

$\alpha = 0.001$

$\varepsilon = \text{Standard Deviation/Mean Radius}$

$Deselecting\text{ cloud reflectivity}$
Wonderful Observations in India

(Kumar et al., ACP, 2016)

• Right: Aircraft measurements during Cloud Aerosol Interaction and Precipitation Enhancement EXperiment (CAIPEEX)

• Left: Ground-based measurements

(Pandithurai et al., JGR 2012)
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. Dispersal may be a reason for overestimating cloud cooling effects by climate models. (Liu et al., ERL, 2008)

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

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

**Warming Dispersion Effect:**
(Lu et al, JGR, 2007; Chen et al, ACP, 2012; Pandithurai et al, JGR, 2012; Kumar et al ACP, 2016)

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

(Kim et al. 2008, JGR)

I = 0.17

Adiabatic clouds

Non-adiabatic clouds
Different entrainment-mixing processes alter cloud properties significantly.

Damkoehler Number

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

A Cloud
LWC = 0.01 g m$^{-3}$/Droplet $\times$ 10 Droplets = 0.1 g m$^{-3}$

Entrainment
LWC decreases from 0.1 g m$^{-3}$ to 0.09 g m$^{-3}$.

Homogeneous
- Decrease the size of each droplet.

Extreme Inhomogeneous
- Decrease the number of droplets.
Observational Examples

March 2000 Cloud IOP at SGP

Adiabatic paradigm
Extreme homogenous

Homogeneous mixing
Leg 2 -- 17 March 2000

Inhomogeneous mixing
with subsequent ascent
Leg 1 -- 18 March 2000

Extreme inhomogeneous mixing
Leg 2 -- 19 March 2000

A measure is needed to cover all!
LES captures the general trend of co-variation of droplet concentration and LWC; but the LES mixing type tends to be more homogeneous than observations (left panel).

(Endo et al JGR, 2014)
Complex entrainment-mixing mechanisms are reduced to one quantity: slope (Andrejczuk et al., 2009), or homogeneous mixing degree (Lu et al., 2013).

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

A measure for all mechanisms:

- $\Psi_1 = 0$ for extreme inhomogeneous
- $\Psi_1 = 1$ for extreme homogeneous

Dynamical Measure: Damköhler Number vs. Transition Scale Number

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

- Transition length $L^*$ is the eddy size of $Da = 1$:
  \[
  \tau_{\text{mixing}} = \tau_{\text{evap}}
  \]
  \[
  \tau_{\text{mix}} \sim \left( L^2 / \xi \right)^{1/3}
  \]
  \[
  L^* = \xi^{1/2} \tau_{\text{evap}}^{3/2}
  \]

- Transition scale number:
  \[
  N_L = \frac{L^*}{\eta} = \frac{\xi^{1/2} \tau_{\text{evap}}^{3/2}}{\eta} = \frac{\xi^{3/4} \tau_{\text{evap}}^{3/2}}{\nu^{3/4}}
  \]

$\eta$: Kolmogorov scale; $\bar{\varepsilon}$: dissipation rate; $\nu$: viscosity

Lehmann et al. (2009)
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

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

Modified from Grabowski and Wang (2013)
Our Particle-Resolved DNS

- 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)

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

Δ x ~ 1cm; Domain ~ 1 m³

Water Vapor Field

Droplets in Motion

Journal of Geophysical Research: Atmospheres

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

Zheng Gao¹, Yangang Liu¹,², Xiaolin Li¹, and Chunsong Lu³
Main DNS Equations

\[ \nabla \cdot \vec{u} = 0 \]
\[ \partial_t \vec{u} + (\vec{u} \cdot \nabla) \vec{u} = -\frac{1}{\rho_0} \nabla p + \nu \nabla^2 \vec{u} + \vec{f} \]

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

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

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

\[ S = \frac{q_v}{q_{vs}} - 1 \]

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

Fluid Dynamics

Microphysics

Droplet Kinetics

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

\[ r(\vec{X}, t) \frac{dr(\vec{X}, t)}{dt} = KS(\vec{X}, t) \]
Six Simulation Scenarios

Case 1

Case 2

Case 3

Two Turbulence Modes: Dissipating & Forced

(Andrejczuk et al., 2004)  (Kumar et al, 2012)  New
Distinct Microphysical Properties for Different Scenarios at Different Times

- Liquid Water Content
- Mean Volume Radius
- Standard Deviation
- Relative Dispersion

Droplet Concentration

Time (S)
First Collapsing: Microphysical Mixing Diagram

Normalized Droplet Concentration

Normalized Mean Droplet Volume
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
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.
Dispersion $\varepsilon$ is the ratio of standard deviation to the mean radius of droplet sizes, which measures the spread of droplet sizes. Dispersion increases from left to right in above figures. The three size distributions have the same L and N.

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

A Cloud
LWC = 0.01 g m\(^{-3}\)/Droplet*10 Droplets = 0.1 g m\(^{-3}\)

Entrainment

LWC decreases from 0.1 g m\(^{-3}\) to 0.09 g m\(^{-3}\).

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

Homogeneous  Extreme Inhomogeneous

Decrease the size of each droplet
Decrease the number of droplets
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

Time Scale

Space Scale

Additional “Macroscopic” Constraints?

Top-Down Approach

Bottom-Up Approach

Molecule

Aerosol

Droplet

Turbulent Eddy

Cloud

Cloud system

Global
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)
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).
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

"Stable" state

Rain Initiation

KPT theory

Work in progress!

Work in progress!
Commonly Used Size Distribution Functions

<table>
<thead>
<tr>
<th>Name</th>
<th>Expression</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weibull</td>
<td>$n(D) = N_0 D^q \exp(-\lambda D^q)$</td>
<td>No, $\lambda$, $q$</td>
</tr>
<tr>
<td>Gamma</td>
<td>$n(D) = N_0 D^\mu \exp(-\lambda D)$</td>
<td>No, $\mu$, $\lambda$</td>
</tr>
<tr>
<td>Lognormal</td>
<td>$n(D) = \frac{N}{\log\sigma\sqrt{2\pi}} \frac{1}{D} \exp\left(\frac{-\log^2(D/D_m)}{2\log^2\sigma}\right)$</td>
<td>$N$, $D_m$, $\sigma$</td>
</tr>
<tr>
<td>Power-law</td>
<td>$n(D) = a D^{-b}$</td>
<td>$a$, $b$</td>
</tr>
<tr>
<td>Exponential</td>
<td>$n(D) = N_0 \exp(-\lambda D)$</td>
<td>No, $\lambda$</td>
</tr>
<tr>
<td>Normal</td>
<td>$n(D) = \frac{N}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(D-D_m)^2}{2\sigma^2}\right)$</td>
<td>$N$, $D_m$, $\sigma$</td>
</tr>
<tr>
<td>Modified gamma</td>
<td>$n(D) = N_0 D^\mu \exp(-\lambda D^q)$</td>
<td>No, $\mu$, $\lambda$, $q$</td>
</tr>
<tr>
<td>Delta function</td>
<td>$n(D) = N \delta(D - D^*)$</td>
<td>$N$, $D^*$</td>
</tr>
</tbody>
</table>

(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

Part II: On Rain Initiation -- Autoconversion

Four Fundamental Sci. Drivers

- Pre-1940s: Scientific Curiosity
- 1940s: Weather Modification
- 1960s: Climate & NWP Modeling
- 1970s: CRM/LES Modeling

Cloud Microphysics
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.

<table>
<thead>
<tr>
<th>Molecular system, Gas</th>
<th>Clouds</th>
</tr>
</thead>
<tbody>
<tr>
<td>Knew Newton’s mechanics for each molecule</td>
<td>Know equations for each droplet</td>
</tr>
<tr>
<td>Kinetics failed to explain observed thermodynamic properties</td>
<td>Models failed to explain observed size distribution</td>
</tr>
<tr>
<td>Maxwell, Boltzmann, Gibbs established statistical mechanics</td>
<td>Establish the systems theory</td>
</tr>
</tbody>
</table>

Most probable distribution
Consider the droplet system constrained by

\[ \int \rho(x)dx = 1 \]  \hspace{1cm} (1)

\[ \int x\rho(x)dx = \frac{X}{N} \]  \hspace{1cm} (2)

\( x = \) Hamiltonian variable, \( X = \) total amount of \( x \) per unit volume, \( n(x) = \) droplet number distribution with respect to \( x \), \( \rho(x) = n(x)/N = \) probability that a droplet of \( x \) occurs.
Droplet spectral entropy is defined as

$$E = - \int \rho(x) \ln(\rho(x)) dx$$  \hspace{1cm} (3)$$

Note the correspondence between the Hamiltonian variable \( x \) and the constraint \( N \int x \rho(x) dx = X \).
Maximizing the spectral entropy subject to the two constraints given by Eqs. (1) and (2) yields the most probable PDF with respect to $x$:

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

(4)

The most probable distribution with respect to $x$ is

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

(5)

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

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

\[
\begin{align*}
 r_e &= \beta \left( \frac{3}{4\pi \rho_w} \right)^{1/3} \left( \frac{L}{N} \right)^{1/3} \\
\beta &= \left( 1 + 2\varepsilon^2 \right)^{2/3} \left( 1 + \varepsilon^2 \right)^{-1/3}
\end{align*}
\]

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.

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.

Aerosol indirect effects constitute the major uncertainty in climate forcing!
Forward GCM AIE estimates suffer from big uncertainty and discrepancy!

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).

(Adapted from Anderson et al., Science, 2003) (Adapted from Quaas et al., ACP, 2006)
Twomey (Number) Effect

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

Cloud Susceptibility:

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

- 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., \( N_{\frac{a}{a}} ^{0.7} N_{\frac{a}{a}} ^{0.7} \) 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:

- $\beta$ has been implicitly assumed to be a constant (only $N$ effect)
- Clouds are adiabatic
Effective radius ratio $\beta$ is an increasing function of relative dispersion.
Further improving $\mu$-parameterization brings the issue to the heart of cloud physics.

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

Uncertainty and Discrepancy

Microphysics Parameterization

- Spectral Broadening
- Rain Initiation
- Cloud Physics
Shallow Cumulus as an Open Multi-Physics System

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

Fundamental difficulties:
- Spectral broadening
- Embryonic Raindrop Formation

\[ \frac{dr}{dt} \sim \frac{1}{r} \]
\[ \frac{dr}{dt} \sim r^4 \]
Autoconversion process is the 1\textsuperscript{st} 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 ($T = \text{Heaviside step function}$)
  * Berry-type ($T = 1$, without threshold function)
  * Sundqvist-type ($T = \text{Exponential-like function}$)

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

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 Parameterizations

- Simulation-based parameterizations are obtained by fitting simulations to a simple function such as a power-law.
- Such a simple function fit distorts either $P_0$ or $T$ (hence $P$) in $P = P_0 T$.

The rate function $P_0$ can be expressed as an analytical function of droplet concentration $N$, liquid water content $L$, and relative dispersion $\varepsilon$ (Liu & Daum 2004; Liu et al. 2006, JAS).
Kessler-Type Autoconversion Parameterizations

Table 1. Kessler-type Autoconversion Parameterizations
\[ P = P_0 H(r_d - r_c) \]

<table>
<thead>
<tr>
<th>Expression</th>
<th>Assumption</th>
<th>Features</th>
</tr>
</thead>
<tbody>
<tr>
<td>Previous</td>
<td>( P = \gamma N^{-1/3} L^{7/3} H(r_3 - r_c) )</td>
<td>Fixed collection efficiency</td>
</tr>
<tr>
<td>New</td>
<td>( P_{LD} = f(\hat{N} \hat{L} \hat{H} r_{-6} r_c) )</td>
<td>Realistic collection efficiency</td>
</tr>
</tbody>
</table>

\( r_3 = 3^{rd} \) moment mean radius; \( r_6 = 6^{th} \) moment mean radius

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

What about the critical radius >> rain initiation theory?
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.


- Kinetic potential peaks at critical radius $r_c$.
- Critical radius & potential barrier both increase with droplet concentration.
- 2nd AIE: Increasing aerosols inhibit rain by enhancing the barrier and critical radius.
Relative dispersion is critical for determining the threshold function

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).

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 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 $< 0.5$. 
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!
The regime equation can be applied to determine global distribution of AIE regimes, which calls for concurrently measuring/representing aerosols and updraft velocity.
Parameterization for Droplet Concentration

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

\[ w = 0.5 \text{ ms}^{-1} \]

- Best in transitional regime but worst in the updraft-limited regime.
- No dispersion parameterization for updraft-limited regime yet.
“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
The regime equation can be applied to determine global distribution of AIE regimes, which calls for concurrently measuring/representing aerosols and updraft velocity.
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 buffering factors/processes that are either poorly represented or not at all (*Steven & Feingold, Nature, 2009*)

Four Related Buffers:
• *Dispersion effect*
• *Regime dependence*
• *Entrainment-mixing processes*
• *Couplings*
Dynamics: Damkoehler Number

- **Damkoehler number:**
  \[ Da = \frac{\tau_{\text{mix}}}{\tau_{\text{react}}} \]

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

- **\( \tau_{\text{react}} \):** the time needed for droplets to evaporate in the entrained dry air or the entrained dry air to saturate (Lehmann et al. 2009):
  \[
  \frac{dr_m}{dt} = -B \times s \\
  \frac{ds}{dt} = -B \times s \\
  r_m: \text{mean radius} \\
  s: \text{supersaturation} \]
Parameterization for Mixing Mechanisms

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

- Cloud fraction (Morrison 2M)
- Cloud fraction (SBM)
- Liquid water content (Morrison 2M)
- Liquid water content (SBM)
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.

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

(Liu et al., ERL, 2008)
Conflicting Results since 2002

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?
Dispersion effect exhibits stronger regime dependence & works to “buffer” number effect!
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?
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.)
Consider an ensemble of drops near the region of embryonic raindrops exchange water vapor molecules with surrounding environment at dynamic equilibrium (detailed balance):

\[ A_g + A_1 = A_{g+1} \]

\( A_g \) = a drop of size \( g \); \( A_1 \) = a monomer
Under equilibrium, $n_g$ 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 $n_g$ expressions yields the kinetic potential

$$\Phi(g) = -\ln \left( \prod_{i=1}^{g-1} \frac{\beta_i}{\gamma_i} \right) = \frac{w(g)}{kT}$$
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 $x = ar^b$

• 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
Big system vs. small system

Molecular system, Gas

- Knew Newton’s mechanics for each molecule
- Kinetics failed to explain observed thermodynamic properties
- Maxwell, Boltzmann, Gibbs introduced statistical principles & established statistical mechanics
- Most probable distribution

Clouds

- Know equations
- For each droplet
- Uniform models failed to explain observed size distributions
- Establish the systems theory

Most probable distribution
Least probable distribution

Difference of Droplet System with Molecular System
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
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).
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

$$n_{\text{min}}(r) = N \delta(r - r_0)$$
Observed droplet size distribution corresponds to 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!
- Fluctuations increases from level 1 to 3.
- Saturation scale $L_s$ is defined as the averaging scale beyond which distributions do not change.
- Distributions are scale-dependent and ill-defined if averaging scale $< L_s$.
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.
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.

<table>
<thead>
<tr>
<th>Molecular system, Gas</th>
<th>Clouds</th>
</tr>
</thead>
<tbody>
<tr>
<td>Knew Newton’s mechanics for each molecule</td>
<td>Know equations</td>
</tr>
<tr>
<td>Kinetics difficult to explain thermodynamic properties</td>
<td>For each droplet</td>
</tr>
<tr>
<td>Statistical mechanics; Phase Transition; Boltzmann equation</td>
<td>Mainstream models difficult to explain size distributions</td>
</tr>
<tr>
<td></td>
<td>Entropy principle; KPT; Fokker-Planck Equation</td>
</tr>
</tbody>
</table>
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

(Baumgarder et al, 1993)
Continental clouds have more droplets, smaller size ranges, and less likely rain. Marine clouds feature less droplets, larger size ranges, and more likely rain.

**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 is 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 for assuming extreme inhomogenous or homogenous mixing;
- Work best for models with 2-moment schemes;
- Testing with SCM and CRM/LES in FASTER;
- Integrating with entrainment rate.

A new parameterization that unifies entrainment rate and mixing effects on cloud microphysics is on the horizon.
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 $\ln(\lambda)$ decrease with increasing distance from cloud core $D$.

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

Three Definitions of Homogeneous Mixing Fraction --- $\Psi_3$

\[
\Psi_3 = \frac{\ln N - \ln N_i}{\ln N_h - \ln N_i} = \frac{\ln r_v^3 - \ln r_{vi}^3}{\ln r_{vh}^3 - \ln r_{vi}^3}
\]

This definition, $\Psi_3$, turns out to be related to $\alpha$:

\[\psi_3 = 1 - \alpha\]

where $\alpha$ was defined by Morrison and Grabowski (2008):

\[N = N_0 \left(\frac{q}{q_0}\right)^\alpha\]
Two Transition Scale Numbers (2)

$\tau_{\text{react}}$ is based on:

\[
\begin{align*}
\frac{dr}{dt} &= A \frac{s}{r}, \\
\frac{ds}{dt} &= -Brs
\end{align*}
\]

$r$: droplet radius;
$s$: supersaturation;
$A$: a function of pressure and temperature;
$B$: a function of pressure, temperature and droplet number concentration ($N_a$ or $N_0$).

$N_a + \text{Dry air} = N_0$

Scale Number $N_{La}$

$N_{L0}$
Explicit Mixing Parcel Model (EMPM)

Domain size:
20 m × 0.001 m × 0.001 m;

Adiabatic Number Concentration:
102.7, 205.4, 308.1, 410.8, 513.5 cm⁻³;

Relative humidity:
11%, 22%, 44%, 66%, 88%;

Dissipation rate:
1e⁻⁵, 5e⁻⁴, 1e⁻³, 5e⁻³, 1e⁻², 5e⁻² m²s⁻³;

Mixing fraction of dry air:
0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9.

Krueger (2008)
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.
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 km².

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).
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]
The size distribution (red curve) is from Liu et al. (1995, Atmos. Res.).
Two Kinds of Universalities

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.

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.

<table>
<thead>
<tr>
<th>Molecular system (gas)</th>
<th>Cloud</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molecules &amp; Newton’s mechanics for each molecule</td>
<td>Droplets &amp; equations for each droplet (DNS)</td>
</tr>
<tr>
<td>Boltzmann equation</td>
<td>Various kinetic equations (e.g., stochastic condensation)</td>
</tr>
<tr>
<td>Maxwell, Boltzmann &amp; Gibbs introduced statistical principles &amp; established statistical mechanics</td>
<td>Systems theory</td>
</tr>
<tr>
<td>Most probable energy distribution</td>
<td>Most probable size distribution</td>
</tr>
<tr>
<td></td>
<td>Least probable size distribution</td>
</tr>
</tbody>
</table>

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

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

I am suggesting the concept of Climate Uroboros.
• 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).
Entrainment rate increases with increasing averaging scales, probably because of increasing chance to sample bigger entrained parcels.
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

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

(RACORQ Cu at SGP)

(DYCOMS-II
Based on 4 m res LWC

(Gerber et al., 2005)
Six Blind Men and the Elephant I

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

(after John Godfrey Saxe’s (1816-1887) version of the Indian legend)
Systems Theory on Atmospheric Particle Systems:

Part I: Most Probable Size Distributions

Part II: On Rain Initiation and Autoconversion
Aerosol-enhanced dispersion causes a warming effect on climate.

Enhanced dispersion has a warming effect that offsets the traditional 1st indirect effect by 10-80%, depending on the $\epsilon$-$N$ relationship (Liu & Daum, Nature 2002; Liu et al. 2006, GRL).
Processes of more than 16 orders of magnitude of length scales are involved in phenomena related to aerosols, clouds, and climate.
Stommel Diagram for Atmosphere and Oceans

Atmosphere

- Longwaves
- Cyclones
- Eddies
- Fronts
- Thunderstorms

Ocean

- Deep circ.
- Gyres

Scaling relationship:

\[ T \sim \varepsilon^{-1/3} L^{2/3} \]

where \( \varepsilon \) is the eddy viscosity:

- Atmosphere: \( \varepsilon = 10^{-5} \text{ m}^2 \text{ s}^{-3} \)
- Ocean: \( \varepsilon = 10^{-13} \text{ m}^2 \text{ s}^{-3} \)
Recall Yin - Yang, and the five elements in traditional Chinese Philosophy and Medicine
The Great Machine of Turbulence

What can be done: A historic example

Phenomenon

Turbulence
Da Vinci (1452-1519)

Classical (deterministic) approach

Claude Louise Mary Henry Navier (1821)
George Gabriel Stokes

Navier-Stokes EQs

Probabilistic approach

Andrei Kolmogorov (1903-1987):
A founder of modern theory of probabilities (1933)

UCLA, STATS 19, Fall 2005
Zoo of Mixing Types & Size Distributions

Various Mixing Mechanisms

Homogeneous Entrainment-Mixing (Warner, 1970)

Just Saturated Air by Droplet Evaporation

Unmixed

Extreme Inhomogeneous Entrainment-Mixing (Baker & Ludlam, 1980)

Mixing with Subsequent Ascent/Vertical Circulation (Telford & Chai, 1980)

Turbulent mixing

Droplet evaporation

\[ Da = \frac{\tau_{\text{mixing}}}{\tau_{\text{phase}}} \]

Adiabatic Paradigm

Number Concentration

Droplet Size

Number Concentration

Droplet Size

Droplet Size

Data

Number Concentration

Droplet Size

Number Concentration

Droplet Size
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

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

Representation of Entrainment
- Entrainment Rate:

\[ \lambda = \frac{1}{m} \frac{dm}{dt} \]

- Entraining Aerosol Effects:

\[ \frac{dN_{a,i}}{dt} = -\lambda w \left( N_{a,i} - N_{e,i} \right) \]

\[ = -\lambda w \left(1 - \alpha \right) N_{a,i} \]

\( \alpha = 0 \) : no entrained aerosols
\( \alpha = 1 \) : entrained aerosols balance the dilution

Built on adiabatic version (Chen et al., GRL, 2016)
Effects of Entrainment-Mixing on Supersaturation Profile
Barahona and Nenes [2007] defined a critical entrainment rate ($\lambda_{c1}$), beyond which droplet can not be activated (essentially no clouds or fog).

We introduce a new critical entrainment rate ($\lambda_{c2}$), beyond which peak supersaturation can not 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 $\alpha$!
Clouds are systems of water droplets

Macroscopic view of clouds is an optical manifestation of cloud particles

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 outstanding anomalies in cloud physics!
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 >> 3 outstanding anomalies surrounding (warm) clouds
Time scale for phase relaxation

Volume fraction is $10^{-6}$

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

Heat flux $F_Q$

Vapor flux $F_V$

$\rho_{v,\infty}$

$T_\infty$

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

$\eta_K \sim 10^2 r$

How does a population of droplets respond when suddenly exposed to a new thermodynamic environment?
Connection to the systems theory
the most probable distribution with respect to \( x \) is

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

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

\[
n_g = \frac{N}{\alpha} \exp \left( -\frac{g}{\alpha} \right)
\]

Note the difference in the value of \( \alpha \) in the two equations.
Under equilibrium, detailed balance gives:

\[ \beta_g n_g = \gamma_{g+1} n_{g+1} \]

\[ n_g = n_1 \frac{n_2}{n_1} \frac{n_3}{n_2} \ldots \frac{n_g}{n_{g-1}} = n_1 \prod_{i=1}^{g-1} \frac{i}{i+1} \]

\[ \beta_g \text{ (s}^{-1}) = \text{rate of monomer condensation on the g-drop} \]
\[ \gamma_g \text{ (s}^{-1}) = \text{rate of monomer evaporation of the g-drop} \]
\[ n_g \text{ (cm}^3) = \text{constrained equilibrium concentration of g-drop} \]
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.

\[
\Phi(g) = -\ln\left(\prod_{i=1}^{g-1} \frac{\beta_i}{Y_{i+1}}\right) = -\sum_{i=1}^{g-1} \ln\left(\frac{\beta_i}{Y_{i+1}}\right)
\]
The growth of cloud droplets is modeled as a sum of condensation and collection processes:

\[ \beta_g = \beta_{g_{\text{con}}} + \beta_{g_{\text{col}}} \]

\[ \beta_{g_{\text{con}}} \] = Condensation rate; \[ \beta_{g_{\text{col}}} \] = Collection growth rate
The general collection kernel is given by

\[ K(R, r) = E \pi (R + r)^2 (V_R - V_r) \]

and its general solution is too complicated to handle.

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

\[ k(R, r) = k_1 R^6 \quad (10 \, \mu m \leq R \leq 50 \, \mu m) \]

\[ k(R, r) = k_2 R^3 \quad (R > 50 \, \mu m) \]

The (gravitational) collection kernel is negligible when \( R < 10 \, \mu m \).
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 $R$

$$(10 \, \mu m \leq R \leq 50 \, \mu m):$$

$$\frac{dm}{dt} = k_1 Lm^2$$

$$\beta_{col}^g = \frac{dg}{dt} = k_1 vLg^2$$

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

\[
\frac{n_{g+1}}{n_y} = \frac{\beta^{	ext{con}}_g}{\gamma_{g+1}} \approx \frac{\gamma^{	ext{con}}_g}{\gamma} = \exp \left( - \frac{1}{\alpha} \right)
\]

\[
\gamma_{g+1} = \exp \left( \frac{1}{\alpha} \right) \beta^{	ext{con}}_g
\]
Relating $\alpha$ to $L$ and $N$

The liquid water content of the droplet system is given by

$$L = \nu \int g n \, dg = \nu \int \frac{N}{g} \exp \left( \frac{g}{\alpha} \right) - \left( \frac{g}{\alpha} \right) = N \nu \alpha$$

$$\alpha = \frac{L}{N \nu}$$

$$\gamma_{g+1} = \exp \left( \frac{\nu N}{L} \beta^\text{con}_g \right)$$
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:

\[
\Phi(g) = - \sum_{i=1}^{g-1} \ln \left( \frac{\beta_i}{Y_{i+1}} \right) = - \sum_{i=1}^{g-1} \ln \left( \frac{\beta_i^{con} + k_1 vL i^2}{\exp \left( \frac{v N}{L} \frac{\beta_i^{con}}{} \right)} \right)
\]
Estimation of Condensation Rate Constant

Mean radius of the 6th moment is given

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

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

\[
 \beta_{\text{con}} = \frac{k \cdot 1.12^6 L_*^4}{\left( \nu \rho_w \right)^2 N_*^3}
\]

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

$\beta_{\text{con}}$: Estimated Condensation Rate Constant

- **mea**: $1.15 \times 10^{23} \, \text{s}^{-1}$
- **min**: $1.02 \times 10^{20} \, \text{s}^{-1}$
- **max**: $1.67 \times 10^{24} \, \text{s}^{-1}$
The relationship between condensate rate and drizzle water content is illustrated in the graph. The pseudo-condensate rate constant is chosen at the point where the fitting line intercepts with the drizzle water content of 0.01 g m\(^{-3}\). The data are from Yum and Hudson (2002).
At the critical point, the forward and backward rates are in balance:

\[ \beta_{\text{con}} + \beta_{\text{col}} = \gamma \]

\[ \gamma = \exp \left( \frac{vN}{L} \right) \beta_{\text{con}} \]

\[ \beta_{\text{col}} = k_1 vLg^2 \]

\[ r_c = \beta \left( \left( \frac{3v}{4\pi} \right)^2 \frac{2}{\kappa} \frac{N_{0.99}}{L^2} \right)^{1/6} = N \left( \frac{L^2}{\frac{1}{1/6}} \right) \]
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

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

Parameters \( c_1 \) and \( c_2 \) depend on droplet concentration and liquid water content.

Note the importance of dispersion!
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 « Numerical Weather Forecasting by Numerical Process by L.F. Richardson 1922