

A satellite image of a tropical cyclone, showing a well-defined eye and spiral cloud bands over a dark ocean surface. The image is the background of the slide.

**EPFL**

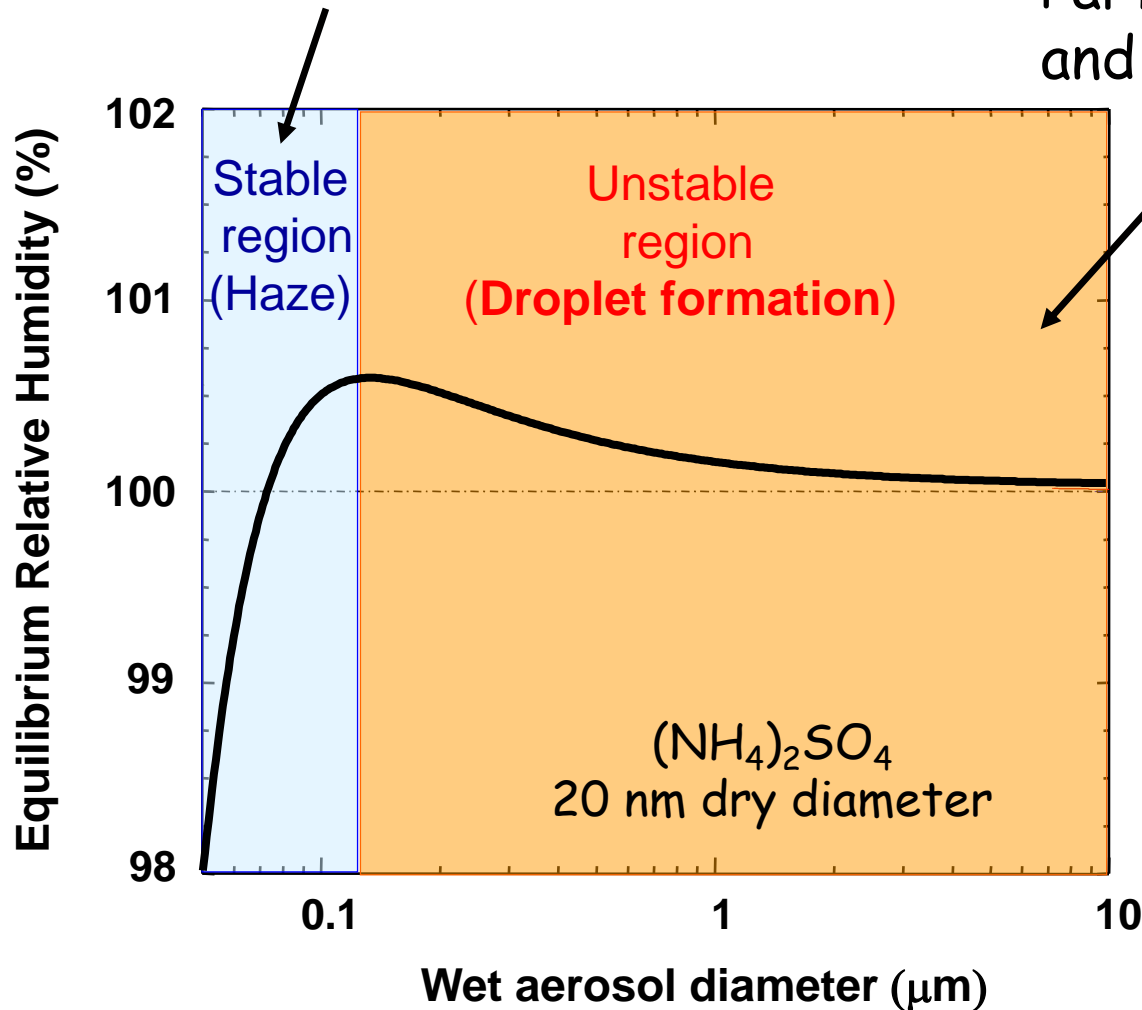
# **Aerosols and Cloud microphysics**

## **Lecture 5 - ENV 407**

# When does an aerosol particle act as a CCN ?

Ambient RH less than  $S_c$  ->  
*stable equilibrium.*

Ambient RH above  $S_c$  ->  
unstable equilibrium.  
Particles act as CCN  
and make droplets



**Köhler theory:**

$$s_c = \left( \frac{4A^3}{27B} \right)^{1/2}$$

$$s_c \sim d_{dry}^{-3/2}, \epsilon_{soluble}^{-1/2}$$

Size is more  
important than  
composition

# Issue: aerosols are complex

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## Primary emissions

Automobiles, industry, domestic, vegetation, forest fires, seasalt, ...

## Secondary transformations

Oxidation of precursors (by  $O_3$ ,  $H_2O_2$ ,  $OH$ ,  $NO_3$ , etc.) generates organic compounds.

Reaction of volatile bases ( $NH_3$ ) with acids, dust and seasalt form salts like  $(NH_4)_2SO_4$ .



# Parameterizing "characteristic" CCN activity...

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Peters and Kreidenweis (2007) expressed the Kohler theory parameter  $B$  in terms of a "hygroscopicity parameter",  $\kappa$

$$s_c = \left( \frac{4A^3}{27B} \right)^{1/2} = \left( \frac{4A^3}{27\kappa d^3} \right)^{1/2} \longrightarrow s_c = \left( \frac{4A^3}{27\kappa} \right)^{1/2} d^{-3/2}$$

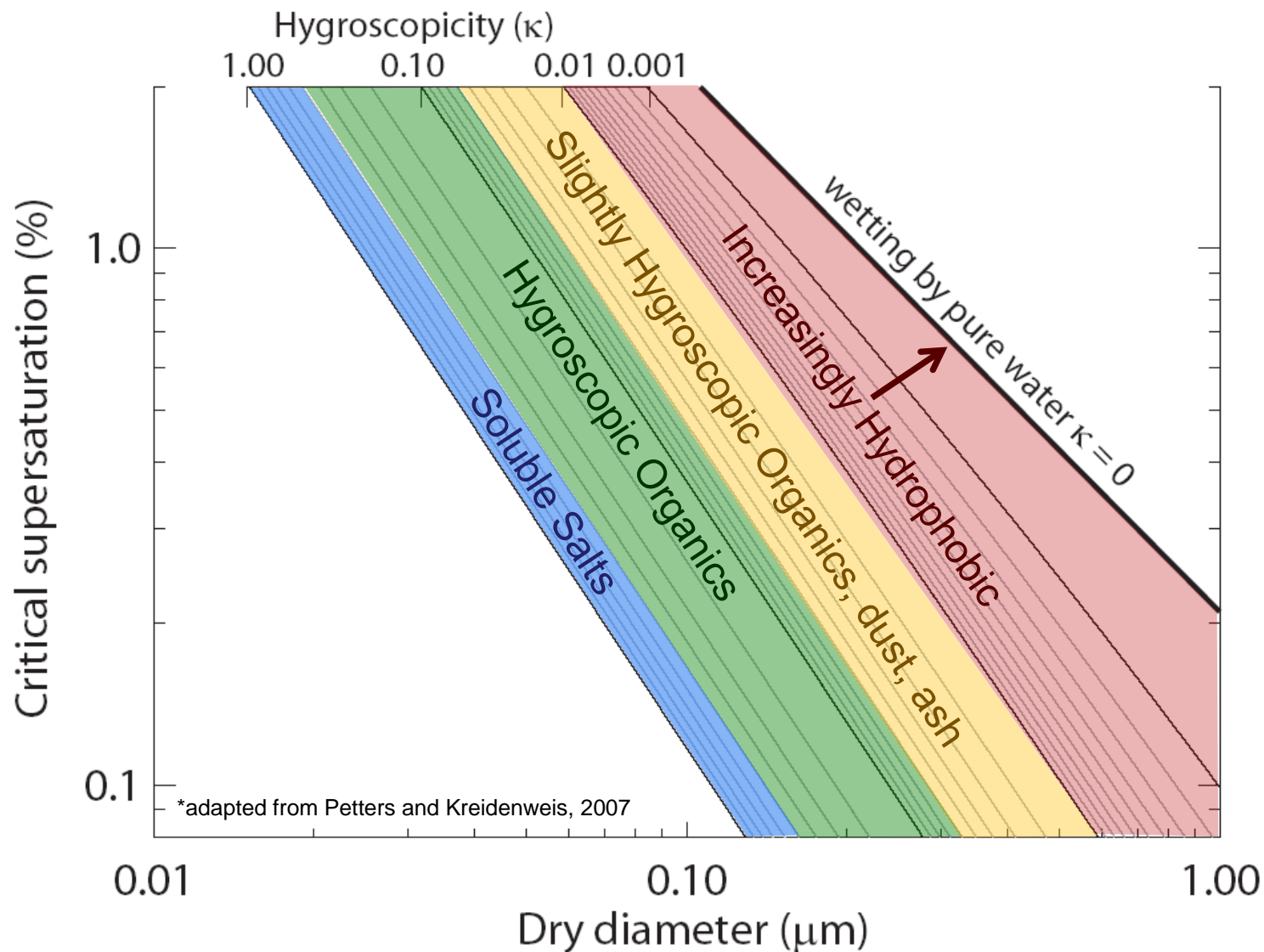
$\kappa \sim 1$  for seasalt,  $\sim 0.6$  for  $(\text{NH}_4)_2\text{SO}_4$ ,  $\sim 0-0.3$  for organics

$\kappa$  rarely exceeds 1 in atmospheric aerosol

Simple way to think of  $\kappa$ : the "equivalent" volume fraction of seasalt in the aerosol (the rest being insoluble).

$\kappa \sim 0.6 \Rightarrow$  particle behaves as 60% seasalt, 40% insoluble

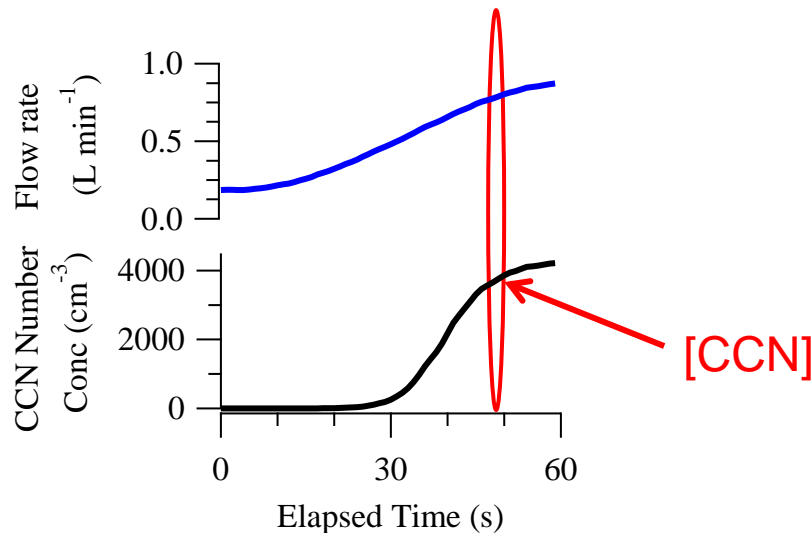
# Hygroscopicity Space



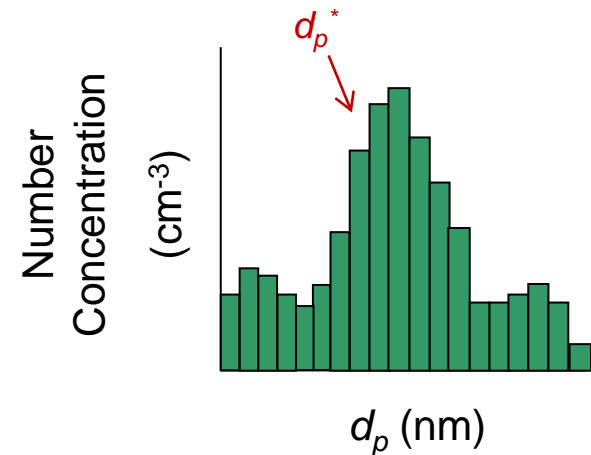
- $\kappa$  is used to parameterize the activation of particles in the atmosphere

# Getting $\kappa$ from CCN Measurements

1. Measure CCN concentration,  $[CCN]$ , at a given  $s^*$



2. Find where backwards integrated size distribution =  $[CCN]$  to obtain the critical diameter,  $d_p^*$



3. Calculate  $\kappa$

$$\kappa \approx \frac{4A^3}{27d_p^3s^{*2}} = \frac{M_w\rho_s}{\rho_wM_s}v\varepsilon_s$$

# Measuring CCN: Basic Principles

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**Operation:** Expose particle sample to a known water vapor supersaturation, and measure those that become droplets.

**Desired range:** 0.01% - 1.0% supersaturation ( $S$ )

**Main challenges:**

Generating a highly controlled level of supersaturation

- *Fluctuations in supersaturation are always a problem*
- *This is more difficult for low supersaturations*

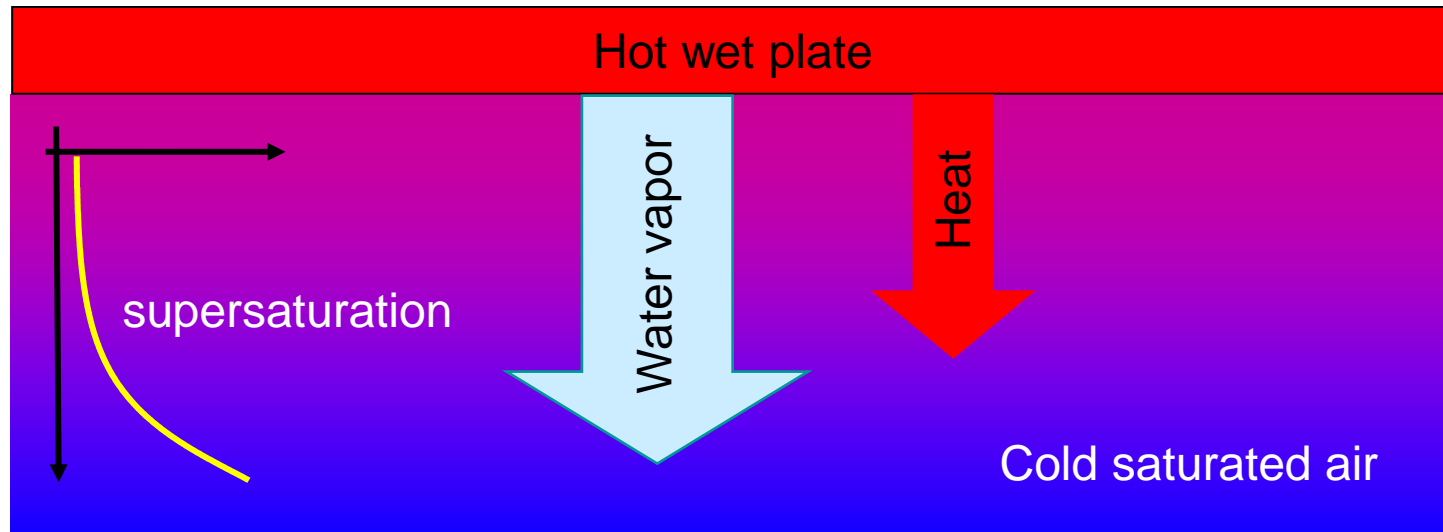
Droplet detection

- *Need to differentiate from particles that do not become droplets*
- *Generally more difficult for low supersaturations*

# CCN Instruments: Generating Supersaturation

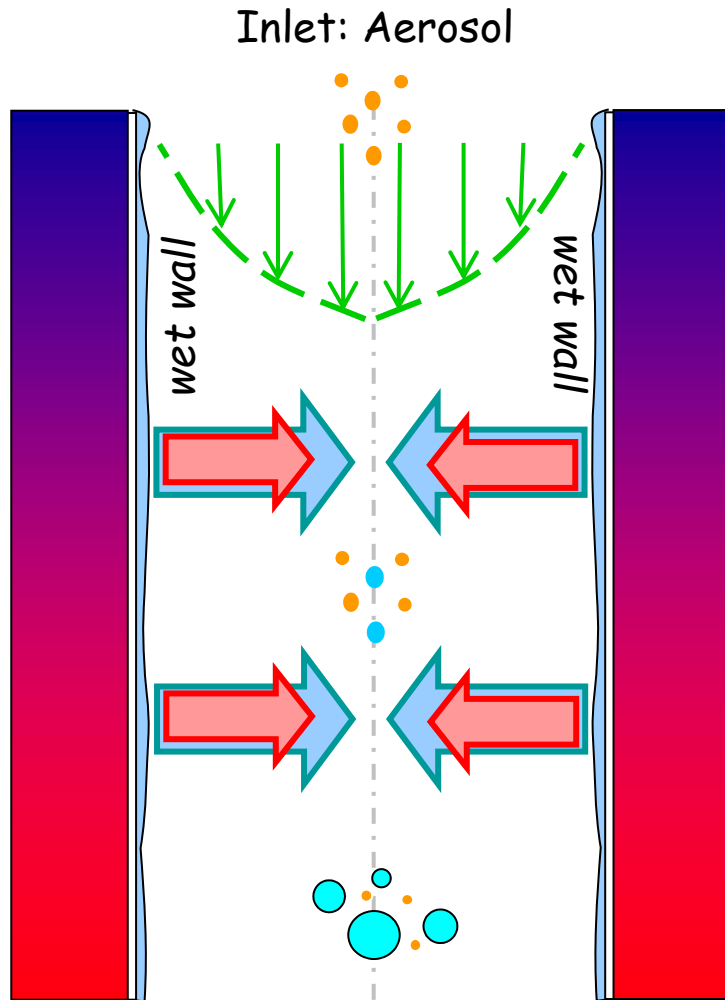
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**Relative Diffusion Instruments:** Exploiting the difference in diffusivity between heat and water vapor.



- Generate a condition where saturated water vapor diffuses into a (saturated) colder region.
- *Supersaturation* develops in the "cold" region (why? Hint:  $H_2O$  molar mass 18, air molar mass  $\sim 29$ ).

# Continuous-Flow Streamwise Thermal Gradient Chamber



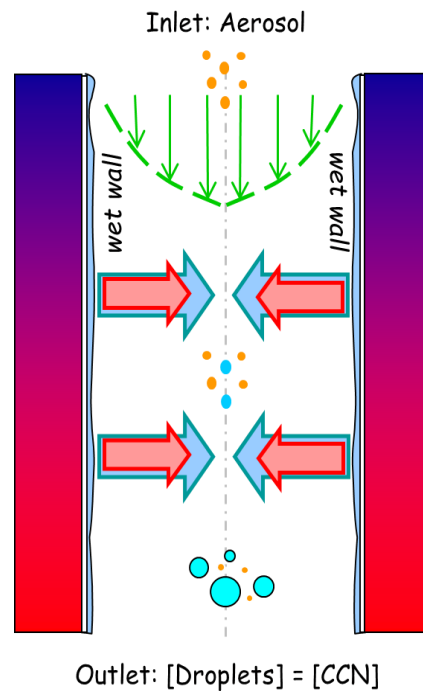
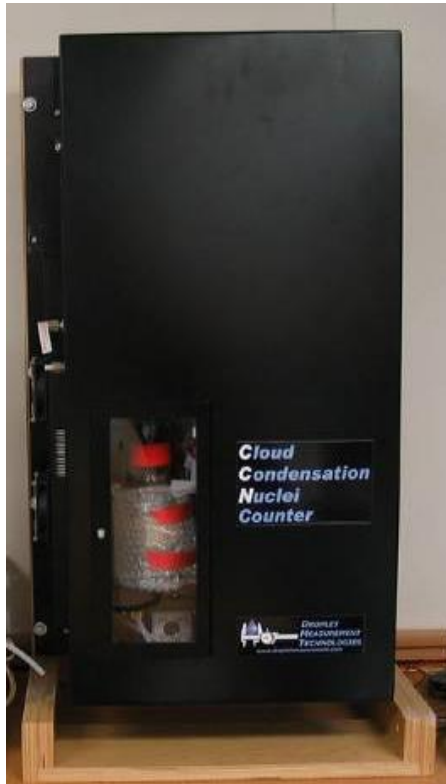
- Standard CCN measurement (>120 instruments in operation).
- Metal cylinder with wetted walls
- Streamwise Temp. Gradient
- Water diffuses faster than heat
- Supersaturation,  $S$ , generated at the centerline =  $f$  (Flowrate, Pressure, and Temp. Gradient)
- Operated as a *spectrometer* using Scanning Flow CCN Analysis (Moore and Nenes, 2009)

Roberts and Nenes (2005), US Patent 7,656,510

Lance et al., (2006), Lathem and Nenes (2011),  
Raatikainen et al. (2012, 2013)

# In-situ (and remote sensing data) key for studying the aerosol-CCN (microphysical) link

## The CFSTGC or "DMT CCN Counter"



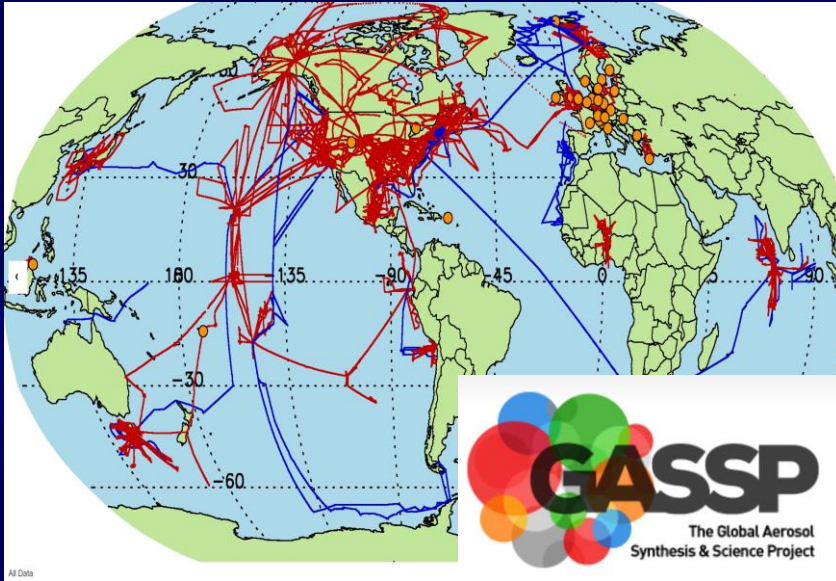
- Metal cylinder with wetted walls
- Streamwise Temp. Gradient
- Water diffuses faster than heat to centerline of flowtube
- Supersaturation,  $S$ , generated at the centerline =  $f$  (Flowrate, Pressure, and Temp. Gradient)

Roberts and Nenes (2005), US Patent 7,656,510

Used to test theory and develop a climatology over 15 years...

# The community has obtained a CCN climatology over the 15 years

Some locations sampled ...



## Measured:

CCN, Aerosol concentrations and size distributions, and aerosol chemistry

Cloud hydrometeor distributions (liquid/ice) and dynamics.

## Environments:

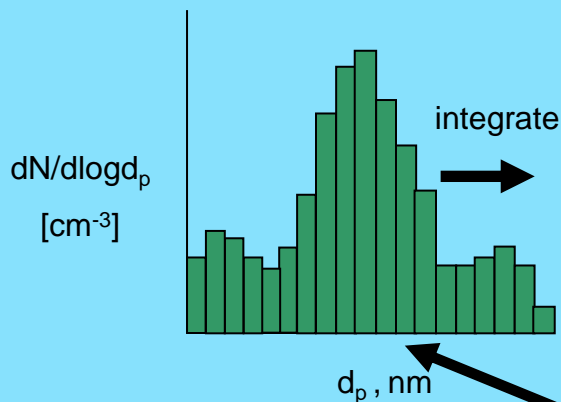
Arctic, urban pollution, biomass burning, marine aerosol, hurricanes, oil spills, the tropics....



# Testing CCN activation theory: CCN "Closure" studies

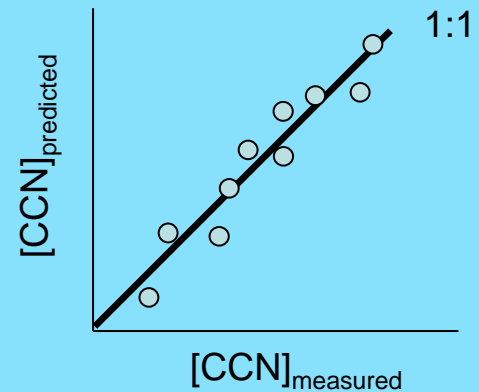
Compare measurements of CCN to predictions using Köhler activation theory and  $\kappa$  description

Aerosol Size Distribution



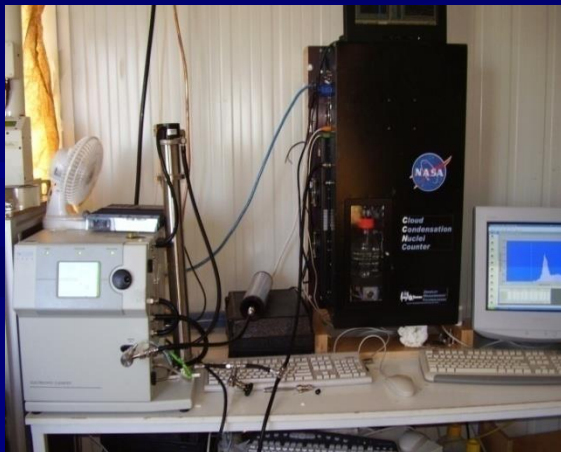
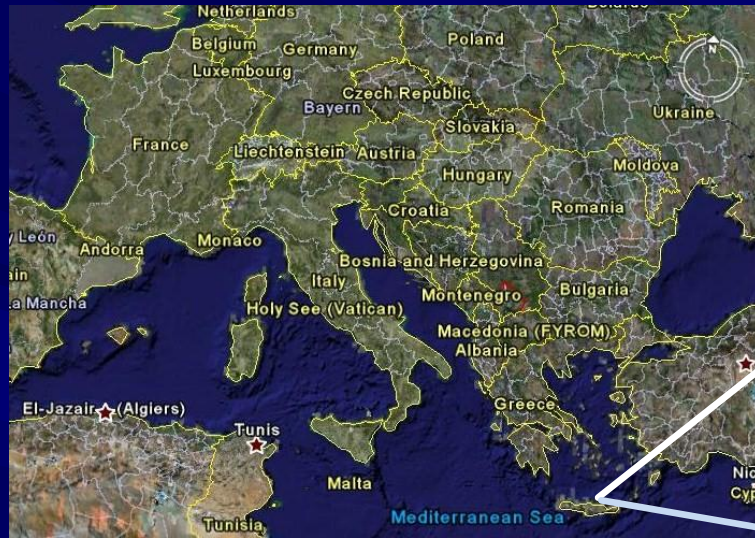
$[\text{CCN}]_{\text{predicted}}$

CCN Closure



Use theory to predict the particles that can act as CCN based on measured chemical composition and CCN instrument supersaturation.

# (The first) Finokalia Aerosol Measurement Campaign (FAME-07) - Summer 2007



**DMT CCN counter**  
*Supersaturation  
range: 0.2-1.0%*

**TSI 3080 SMPS**  
*Size: 20-460 nm*

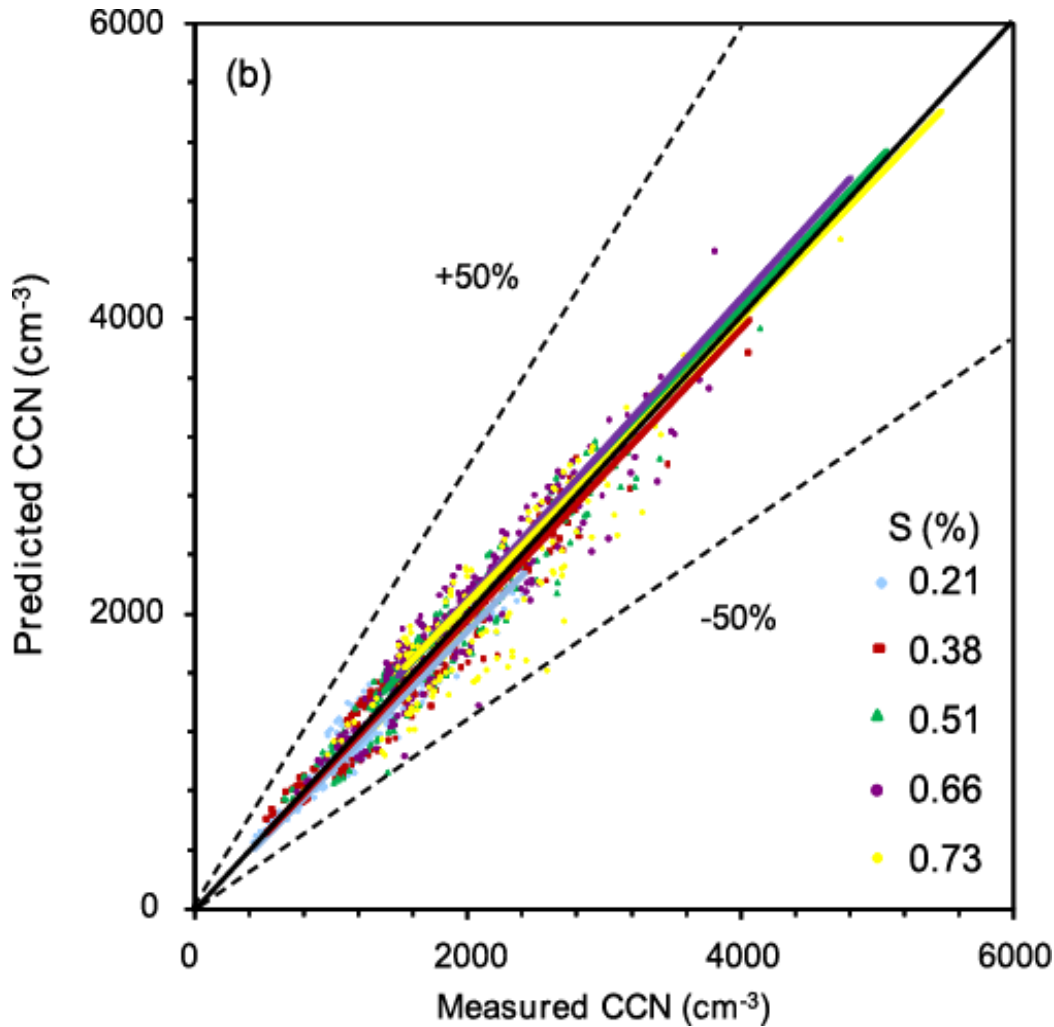
**Low-vol impactor**  
*Ionic composition  
measured via IC*

*WSOC/EC/OC also  
measured*



(Bougiatioti et al., ACP, 2009)

# Finokalia Aerosol Measurement Campaign (FAME-07) - CCN closure



Organics: very oxidized (MO-OOA)  $\kappa \sim 0.15$

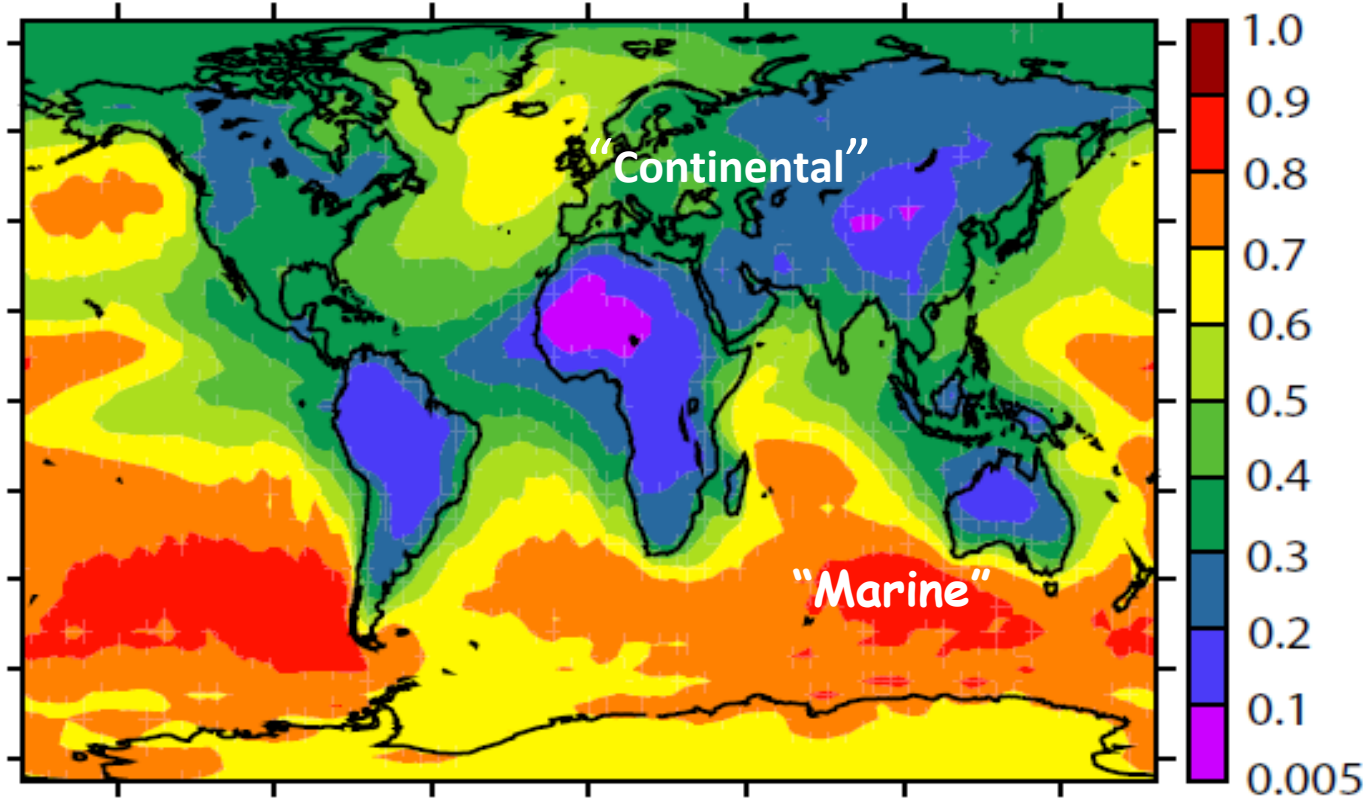
Excellent CCN closure (error  $\sim 2\%$ ).

Köhler (CCN) theory *really* works.

Simple treatments of hygroscopicity in general work "well enough" for CCN/CDNC predictions

(Bougiatioti et al., ACP, 2009)

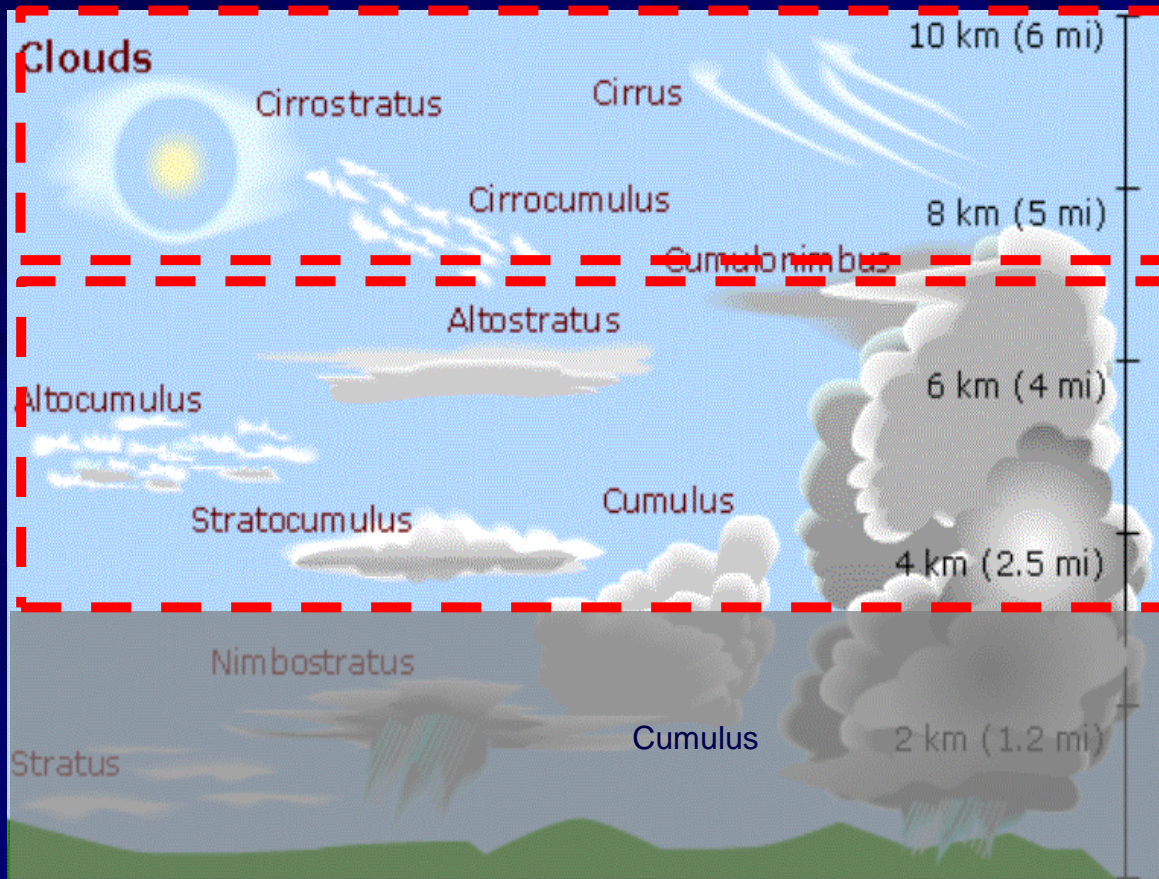
# Global "average" distribution for $\kappa$



Pringle et al., ACP, (2010)

**Fig. 2.** Annual mean distribution of  $\kappa$  at the altitude of the planetary boundary layer.

# Focus on "Slush" and Ice clouds now



- **Ice (cold) clouds:**  
Made of ice crystals at  $T < 235$  K.
- **Mixed Phase clouds:**  
Mixture of liquid droplets and ice for  $T$  between 235 and 273K
- **Liquid (warm) clouds:**  
Made of liquid droplets at  $T > 273$  K

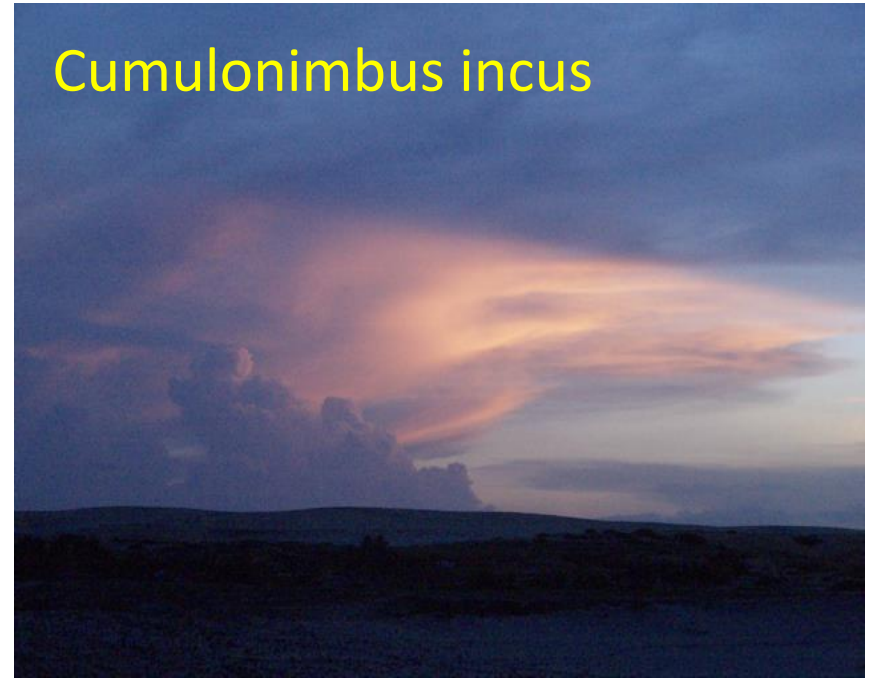
Cloud particles are not created directly from the vapor phase but from **suspended aerosol particles**

# Pictures of ice clouds

Tropical Cumulonimbus



Cumulonimbus incus



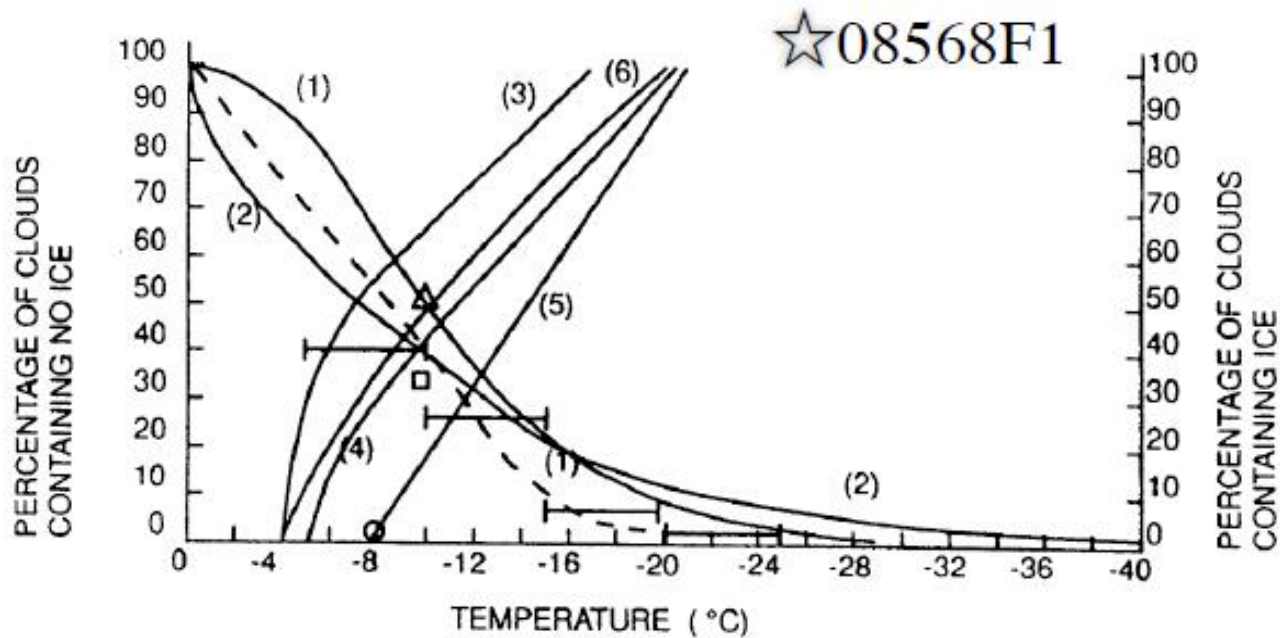
Cirrostratus



Subvisible Cirrus



# When do atmospheric clouds freeze?



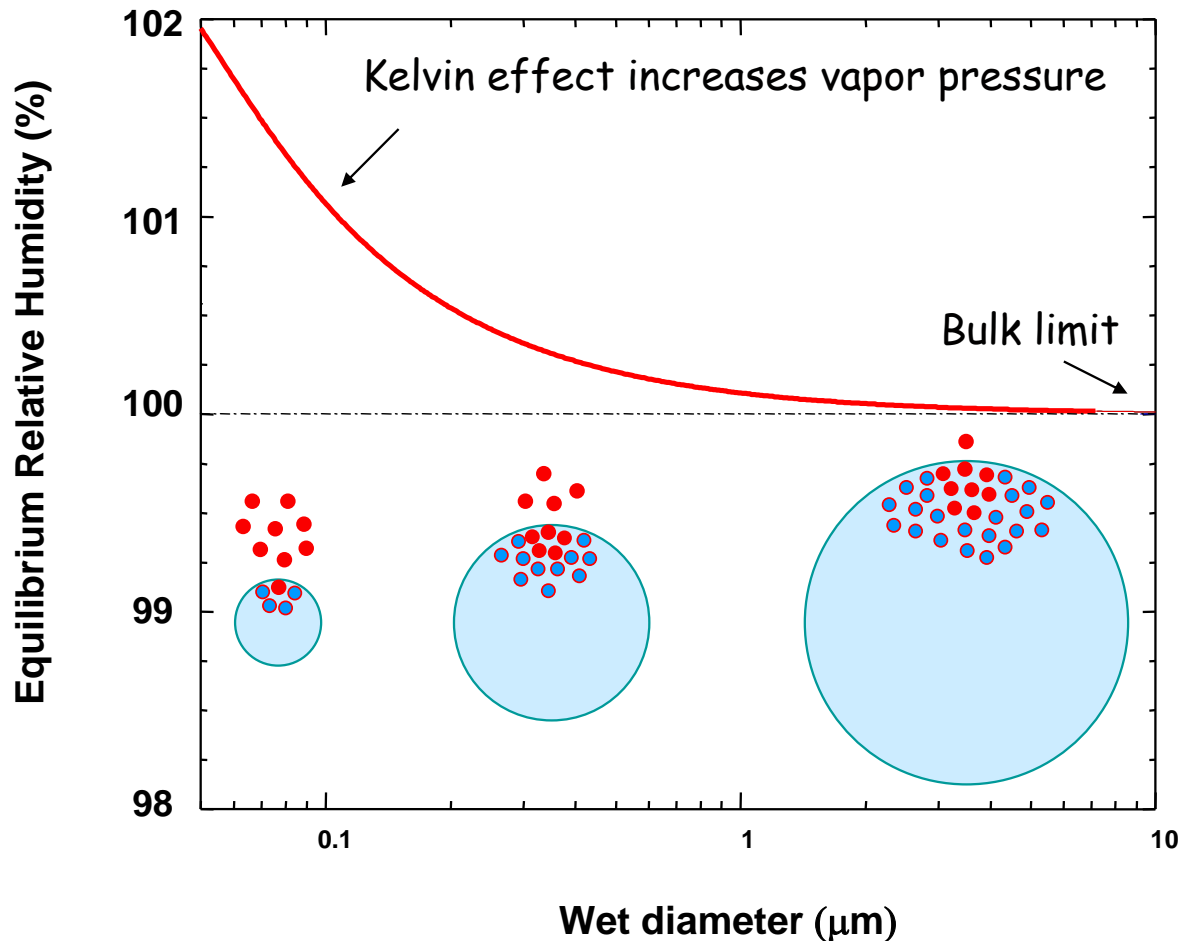
- At the nominal freezing point of water nearly all cloud contain no ice
- Below -4 C or so, clouds start containing ice
- Below -20 C or so all clouds contain some ice
- Below -40 C or so all clouds are ice clouds

**Surprise!** It takes a *lot* of cooling to freeze

# Understanding why it takes a lot of cooling to freeze

Impact of curvature:

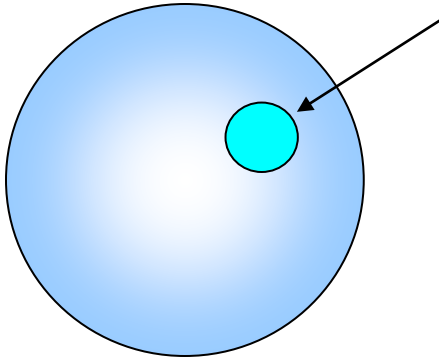
$$P = P^* \exp\left(\frac{2\sigma v_l}{RTR_p}\right) \quad \text{Kelvin Equation}$$



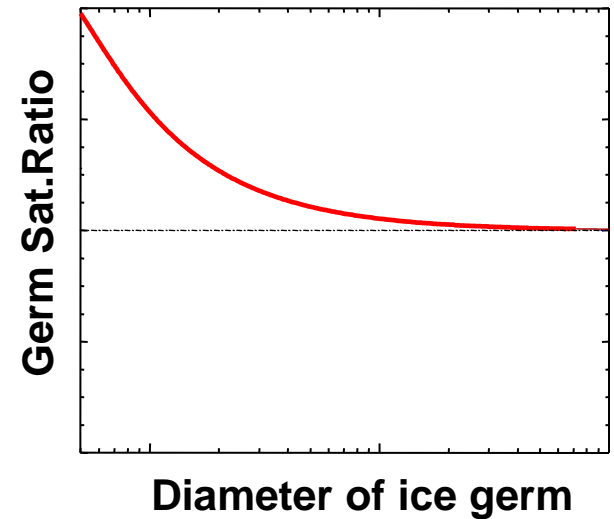
# Curvature effect: the basis of nucleation

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**Ice Germs:** generated in supercooled droplets

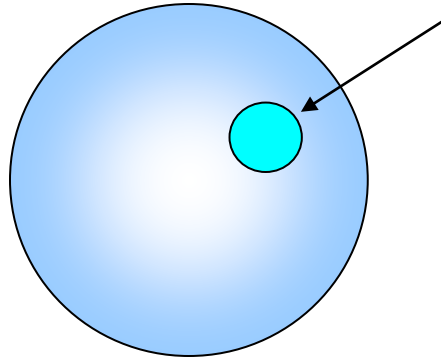


If you are cool enough - the ice germ will be around long enough to grow into a stable crystal.

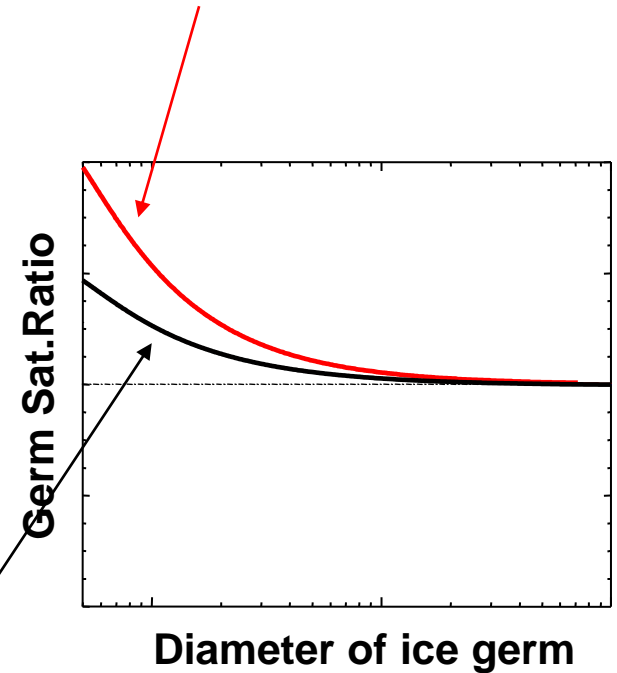


# Curvature effect: the basis of nucleation

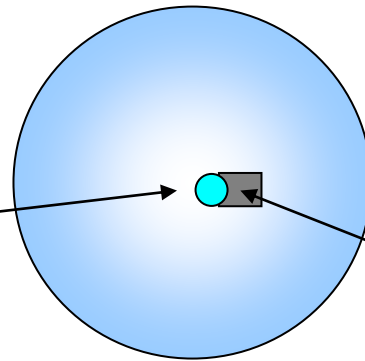
**Ice Germs:** generated in supercooled droplets



If you are cool enough - the ice germ will be around long enough to grow into a stable crystal.



**Ice Germs:** generated on insoluble particles inside of supercooled droplets or suspended in air

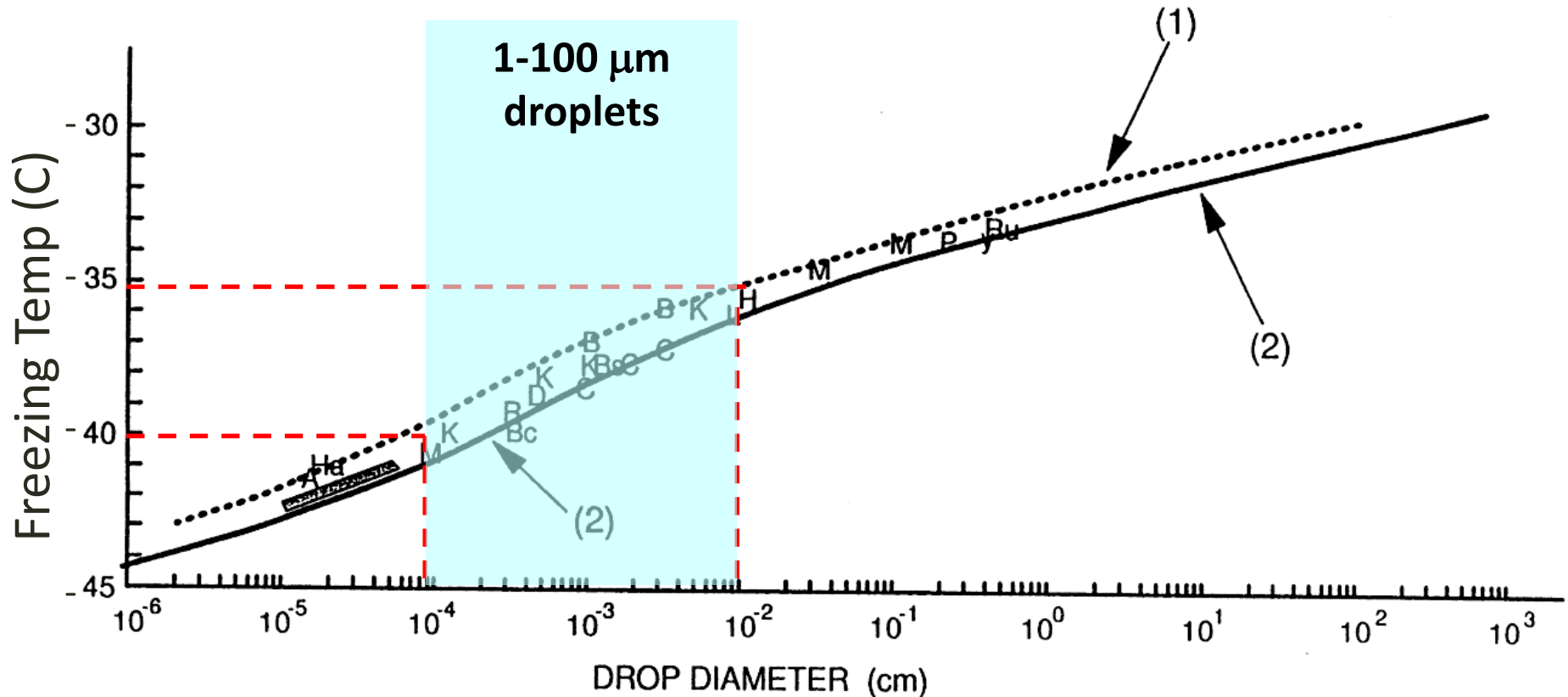


Insoluble particle



(why do insolubles help?)

# Maximum supercooling of extrapure water

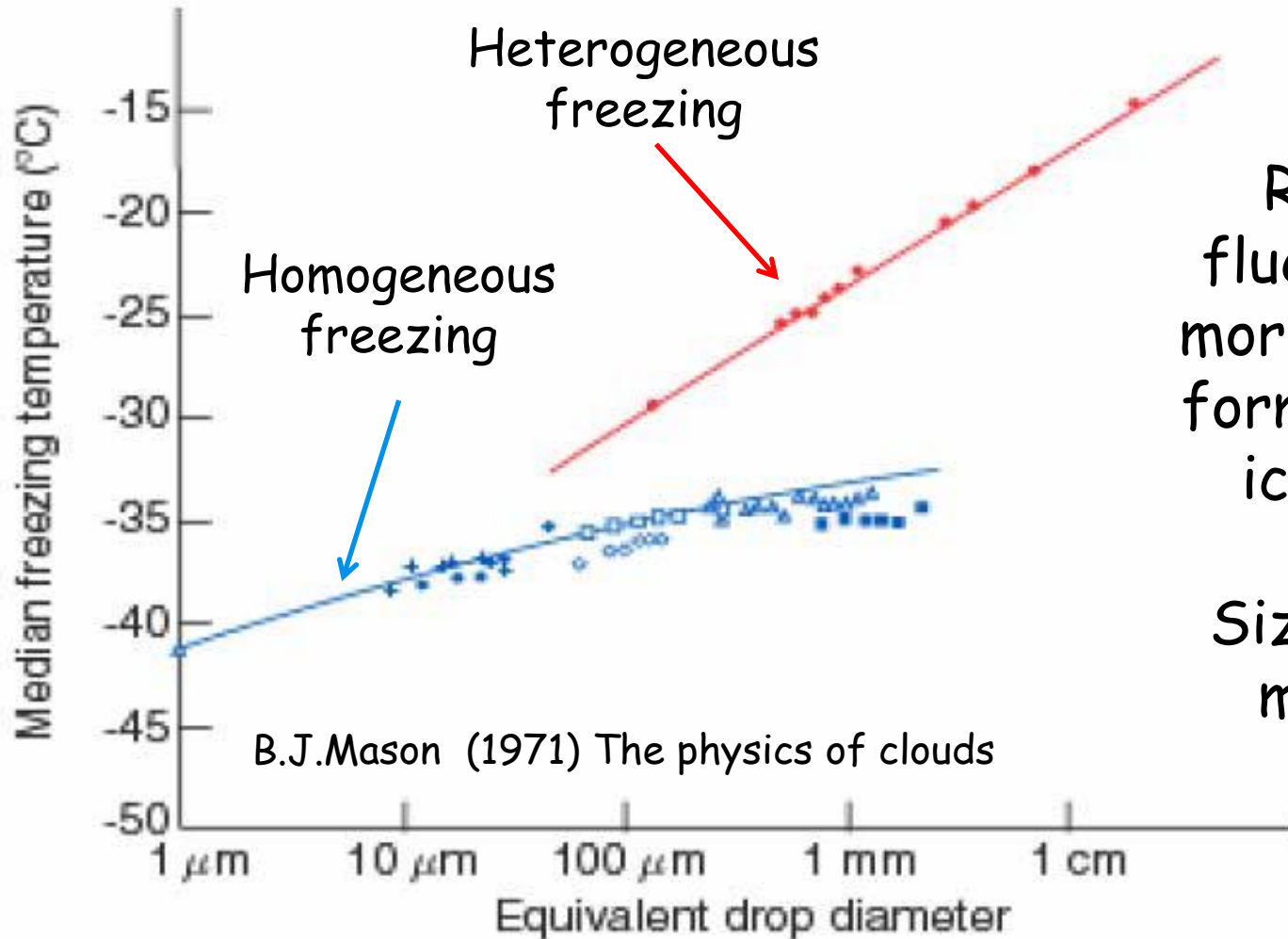


**Result:** Pure water cloud droplets in the atmosphere effectively start freezing at around  $-35^{\circ}\text{C}$

**Problem:** Why do ice cubes form in our freezer? Are the Olympian gods “bending the rules” so we can enjoy ούζο, φραππέ or your favorite drink

# Median freezing T of water samples

Not really... Heterogeneous Freezing Greatly Facilitates Ice Formation



Homogeneous freezing

Heterogeneous freezing

Random fluctuations more likely to form a stable ice germ:

Size always matters

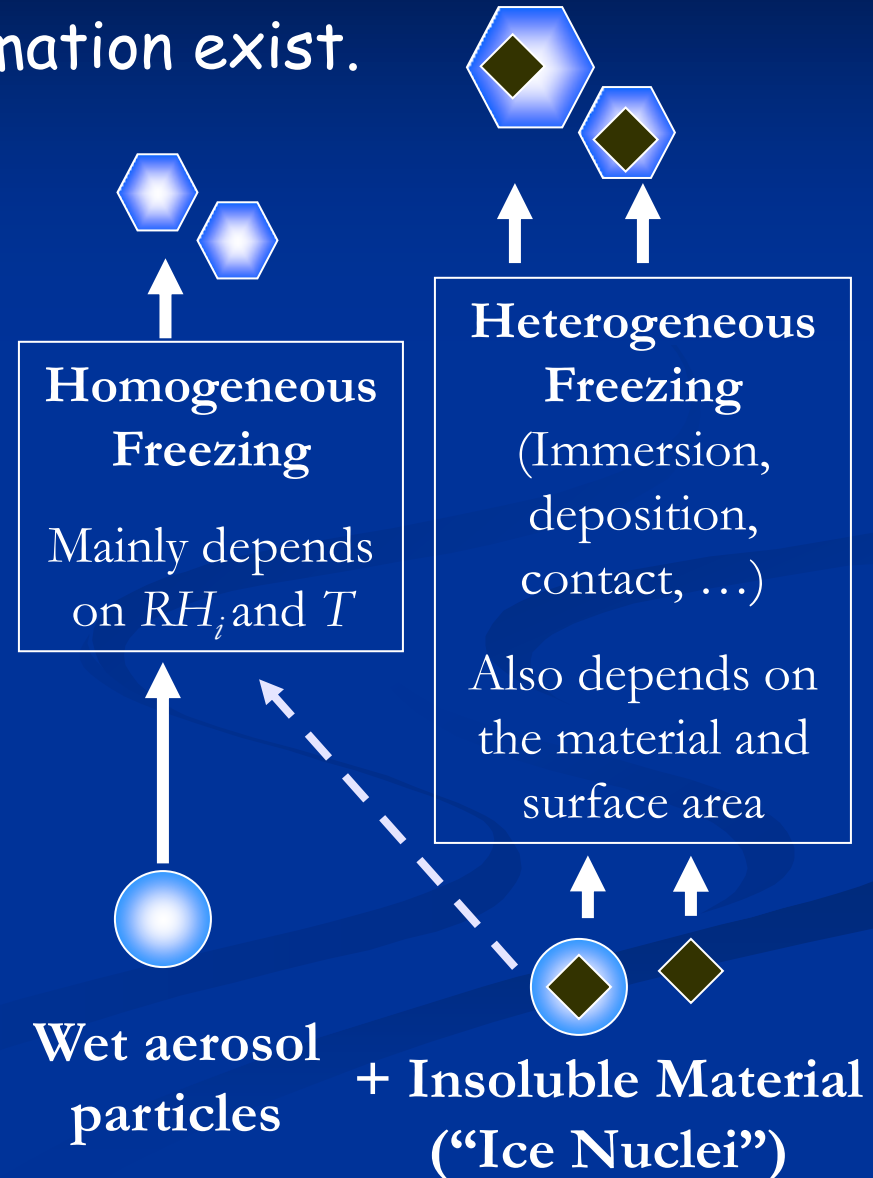
B.J.Mason (1971) The physics of clouds

# How do (ice water) clouds form?

Ice crystals also form on preexisting particles.  
Multiple mechanisms for ice formation exist.



<http://www.alanbauer.com>



# Heterogeneous ice nuclei: freezing modes

**Homogeneous nucleation**



**Deposition nucleation**



**Immersion freezing**



**Condensation freezing**



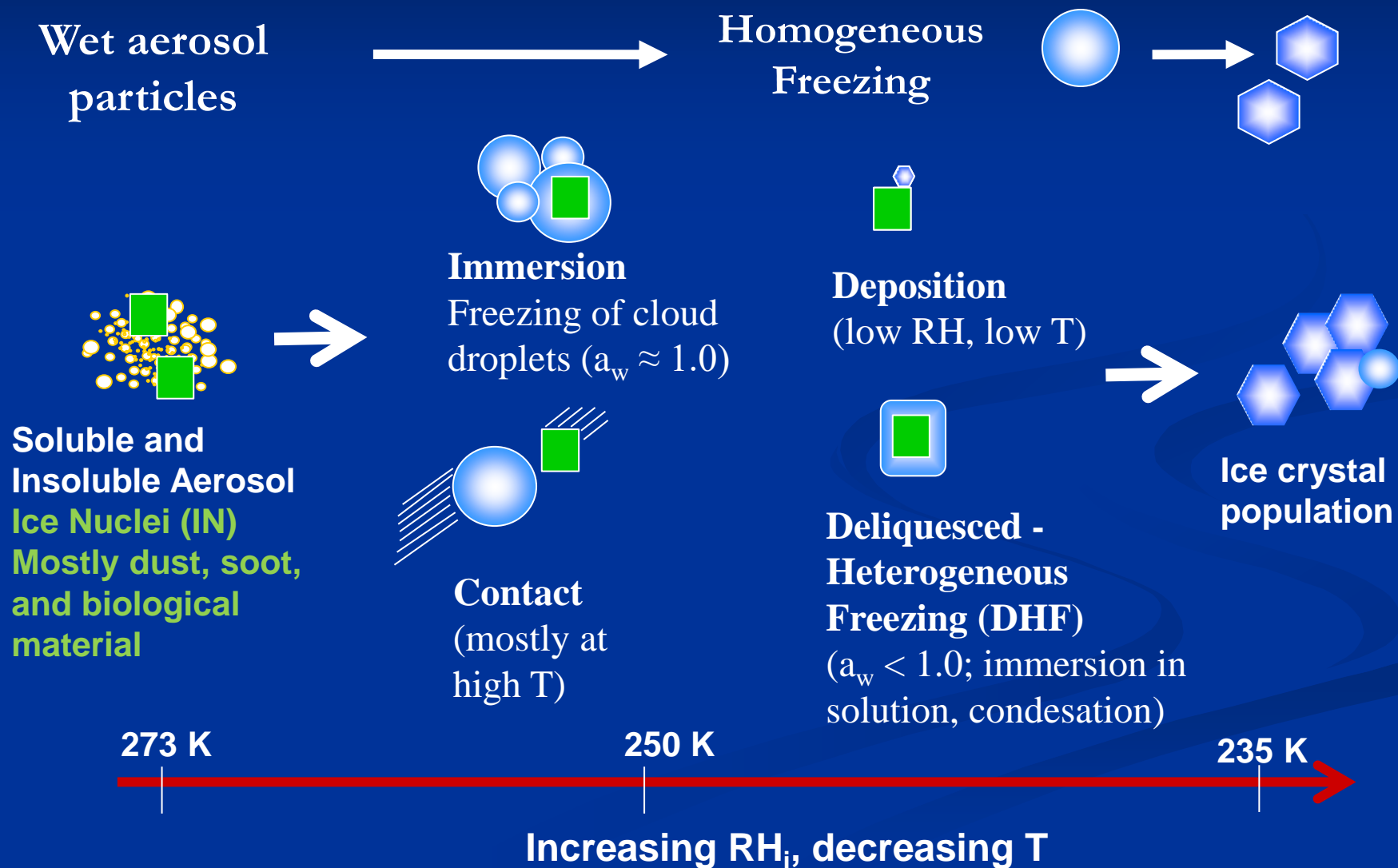
**Contact freezing**



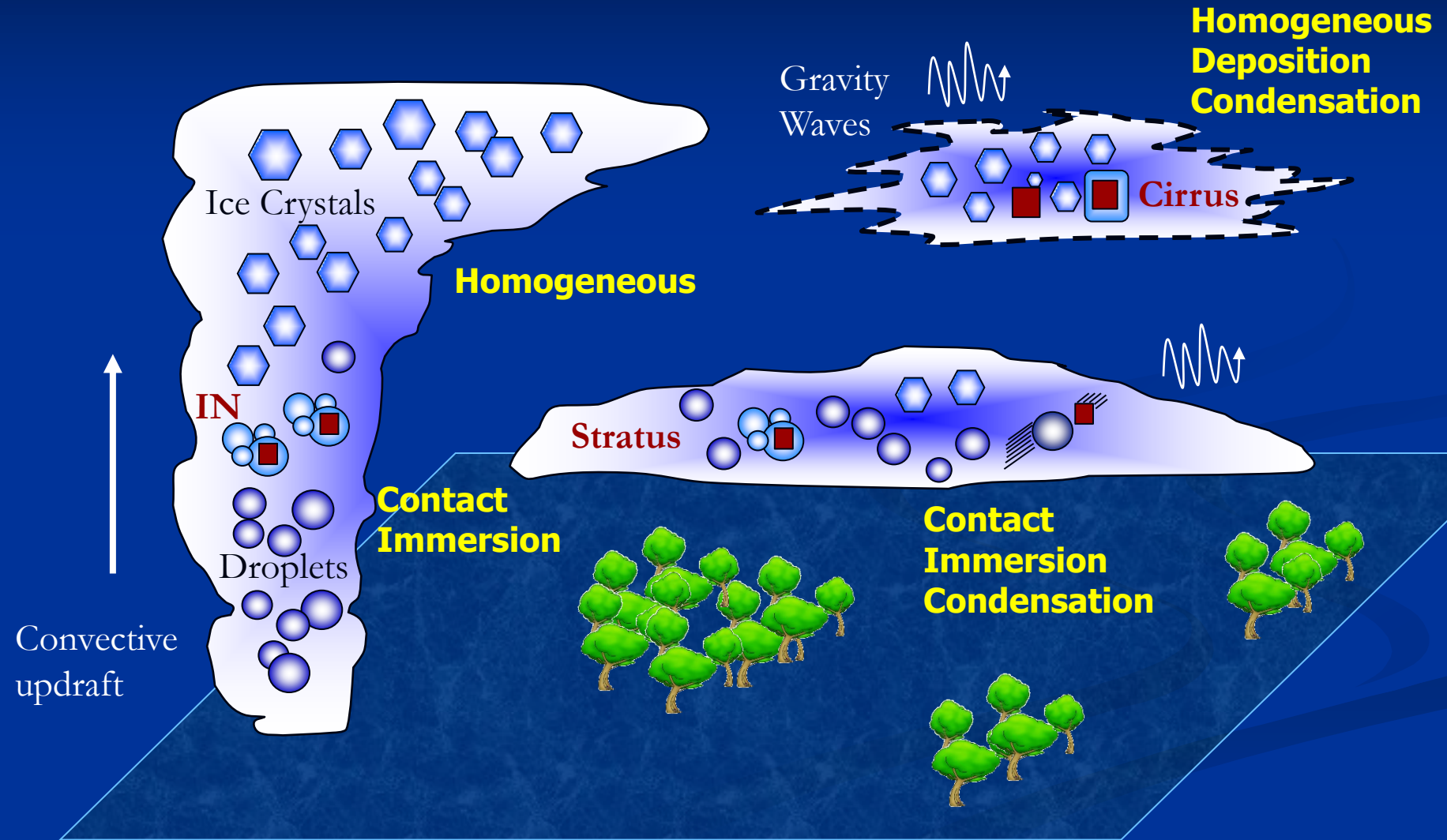
 = heterogeneous ice nucleus (e.g. mineral dust)

# Heterogeneous ice nuclei: freezing modes

Multiple mechanisms for ice formation can be active.



# Ice formation "modes" depends on cloud conditions (T, RH) and IN



# IN vs CCN: implications for clouds

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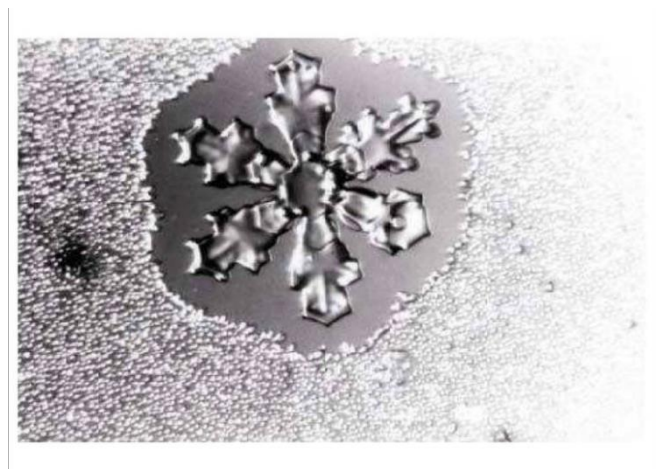
IN are far less abundant than CCN. (1 in a million!)

CCN: 100-1000  $\text{cm}^{-3}$  vs IN: 0.001-0.01  $\text{cm}^{-3}$

Hence, in an ice cloud, cloud water is typically distributed on ***orders of magnitude fewer*** cloud particles than in a liquid cloud.

Consequently, the ice crystals are much larger than cloud droplets and therefore much more likely to fall out as precipitation.

**Most precipitation on the planet is initiated from the ice phase.**



# Heterogeneous ice nuclei: requirements

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**Insolubility:** A rigid substrate is needed for the ice "germ" formation.

**Size:** Larger particles are better IN (more active sites for forming a germ).

**Chemical bond/Crystallography:** A similar bond as the ice crystal lattice is beneficial. Geometry of aerosol (surface steps/imperfections) is important.

**Coatings:** worsens the IN activity, because it depresses the water activity of the aerosol, and may deactivate the ice-forming sites on the particle.

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Table 2. Activation Temperatures  $T_a$  and Median Freezing Temperatures  $T_m$  Determined From Laboratory Experiments<sup>a</sup>

| Particle Type   | Immersion Freezing                                       |  | Contact Freezing                |   |
|-----------------|--|--|---------------------------------|---|
|                 | $T_a$  | $T_m$  | $T_a$                           | $T_m$   |
| Bacteria        | $-4^\circ\text{C}$ , <sup>b</sup> $a = 250 \mu\text{m}$  | $-7^\circ\text{C}$ , <sup>b</sup> $a = 250 \mu\text{m}$  | $-3^\circ\text{C}$ <sup>b</sup> | $-4.5^\circ\text{C}$ <sup>b</sup>               |
| Leaf litter     | $-5^\circ\text{C}$ , <sup>c</sup> $a = 350 \mu\text{m}$  | $-9^\circ\text{C}$ , <sup>c</sup> $a = 350 \mu\text{m}$  | $-5^\circ\text{C}$ <sup>c</sup> | $-10^\circ\text{C}$ <sup>c</sup>                |
| Pollen          | $-9^\circ\text{C}$ , <sup>d</sup> $a = 250 \mu\text{m}$  | $-14^\circ\text{C}$ , <sup>d</sup> $a = 250 \mu\text{m}$   | $-5^\circ\text{C}$ <sup>d</sup> | $-10^\circ\text{C}$ <sup>d</sup>                |
| Montmorillonite | $-12^\circ\text{C}$ , <sup>e</sup> $a = 350 \mu\text{m}$ | $-19^\circ\text{C}$ , <sup>e</sup> $a = 350 \mu\text{m}$ , and $-24^\circ\text{C}$ , <sup>f</sup> $a = 50 \mu\text{m}$   | $-3^\circ\text{C}$ <sup>e</sup> | $-8^\circ\text{C}$ <sup>e</sup>                 |
| Kaolinite       | $-14^\circ\text{C}$ , <sup>e</sup> $a = 350 \mu\text{m}$ | $-23^\circ\text{C}$ , <sup>e</sup> $a = 350 \mu\text{m}$ , and $-32.5^\circ\text{C}$ , <sup>f</sup> $a = 50 \mu\text{m}$ | $-5^\circ\text{C}$ <sup>e</sup> | $-12^\circ\text{C}$ <sup>e</sup>                |
| Soot            | $-18^\circ\text{C}$ , <sup>g</sup> $a = 350 \mu\text{m}$ | $-28^\circ\text{C}$ , <sup>g</sup> $a = 350 \mu\text{m}$   |                                 | $-18^\circ\text{C}$ <sup>h</sup> (extrapolated) |

<sup>a</sup>Immersion freezing temperatures are for defined drop radii  $a$ , and contact freezing temperatures are for arbitrary drop sizes.

<sup>b</sup>Levin and Yankofsky [1983].

<sup>c</sup>Diehl et al. [2001b].

<sup>d</sup>Diehl et al. [2002].

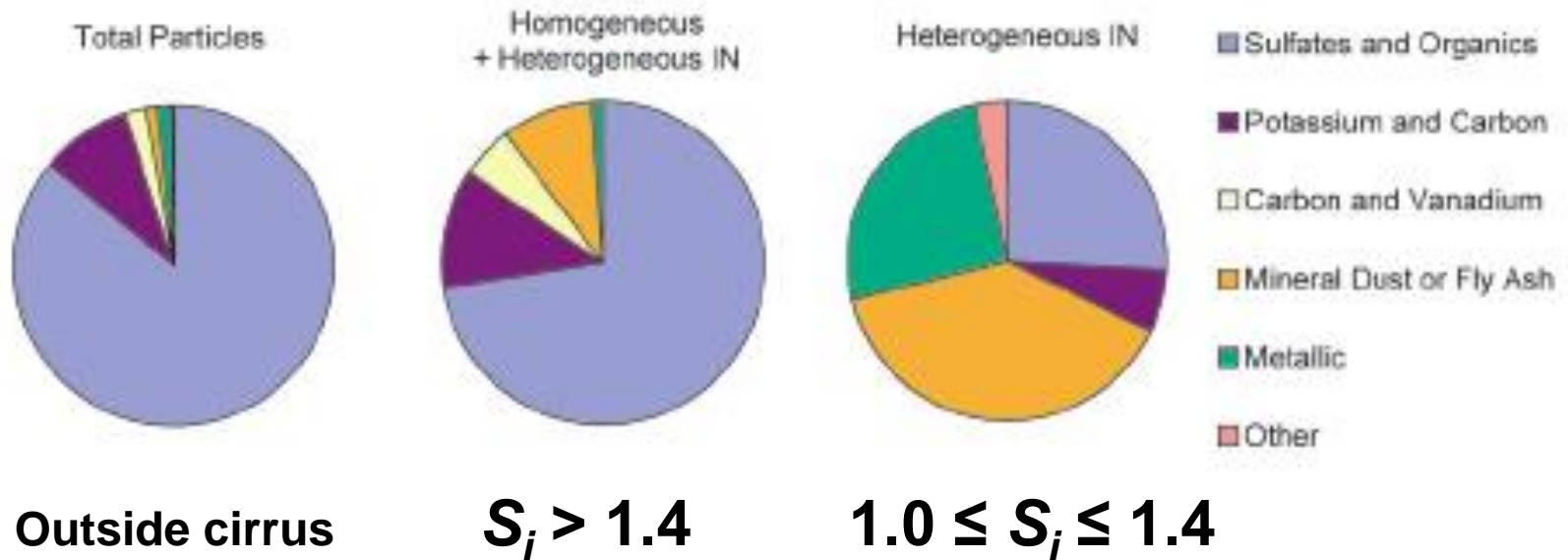
<sup>e</sup>Pitter and Pruppacher [1973].

<sup>f</sup>Hoffer [1961].

<sup>g</sup>Diehl and Mitra [1998].

<sup>h</sup>Gorbunov et al. [2001].

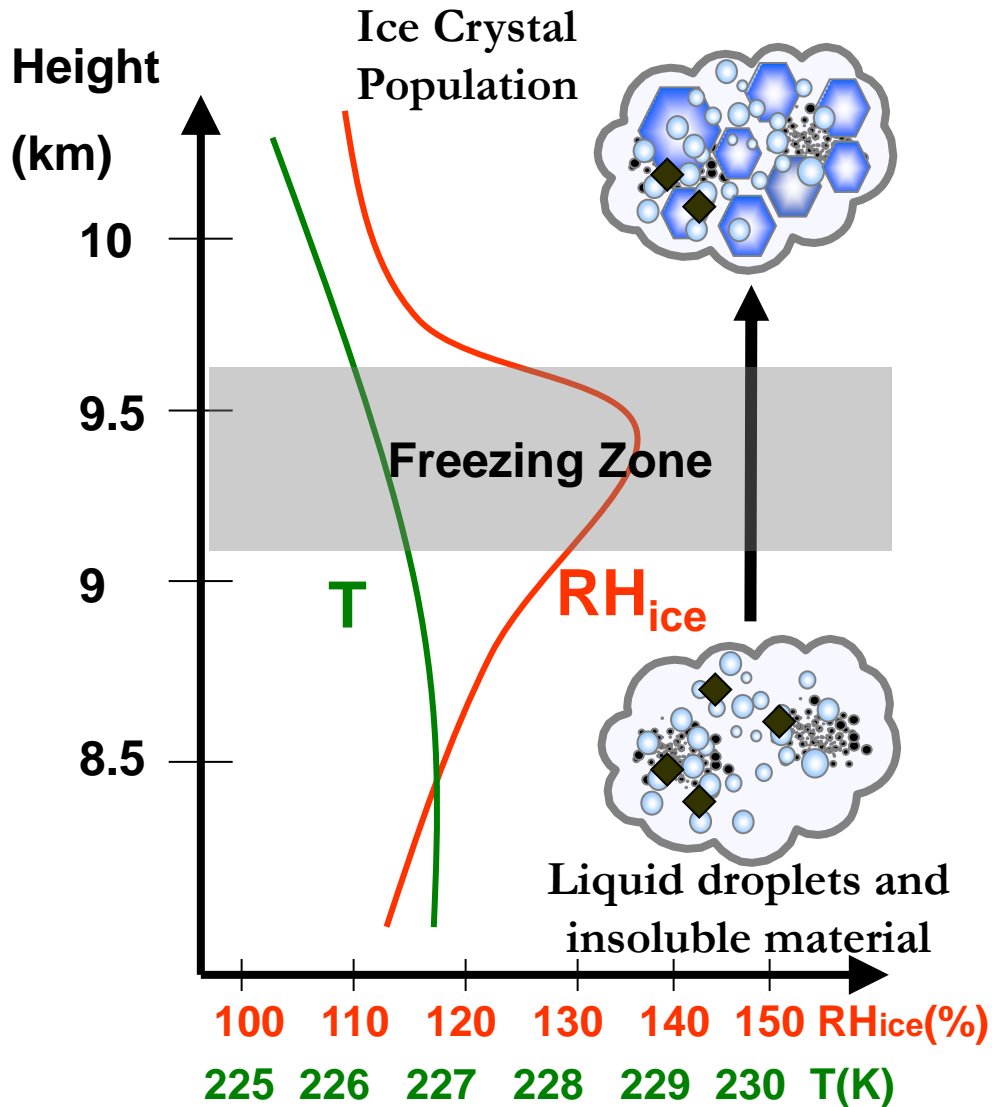
# What do evaporated ice crystals from ambient clouds tell us?



Evaporated ice crystals: Enriched in K, C, dust, metallic  
(especially for IN-dominated clouds!!!)

Composition of evaporated crystals from IN very different from the “total aerosol”! – IN are *rare* (1 in a million!)

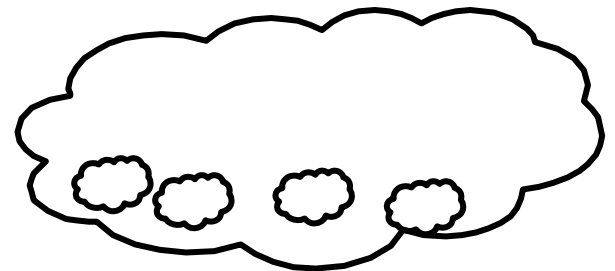
# Ice cloud formation: The essence



Conceptual steps are:

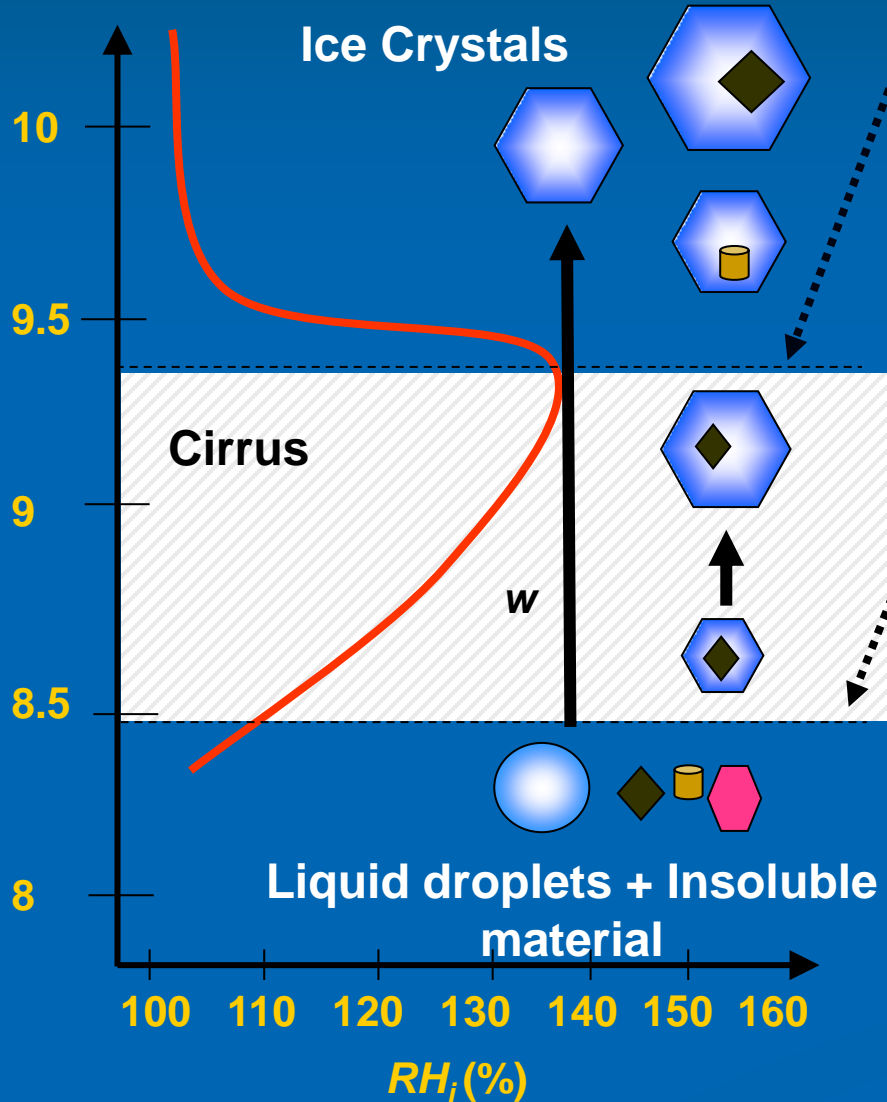
- Air parcel cools
- Eventually exceeds frost point
- Water vapor is supersaturated
- Ice starts forming.
- Condensation of water on crystals becomes intense.
- $S$  reaches a maximum
- No more crystals form

Cloud Layer



**There is a distribution of formation conditions in every cloud**

# Application: Cirrus (Pure Ice) Clouds



Homogeneous freezing of droplets

Crystal growth, fresh IN continue to freeze and deplete vapor

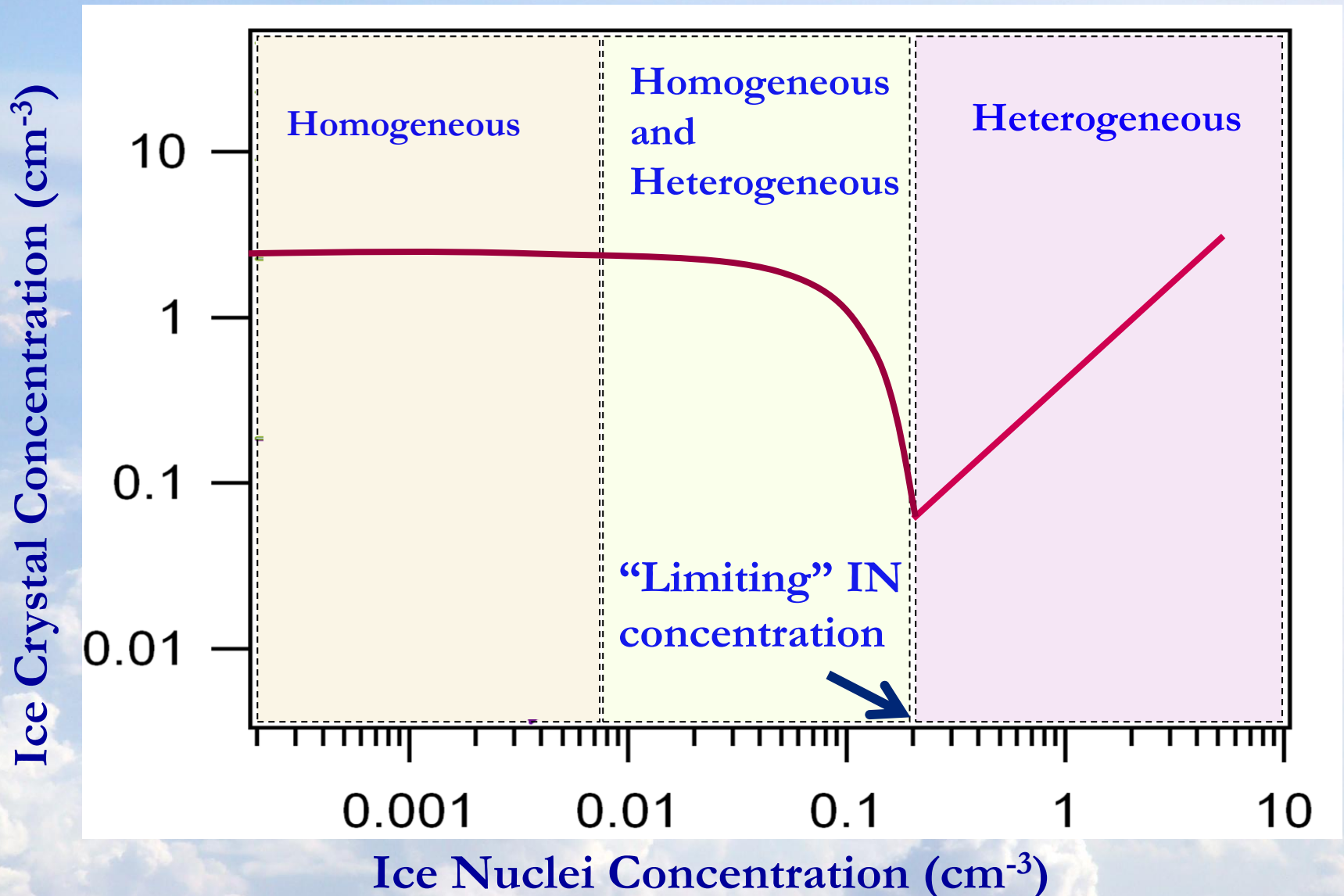
Heterogeneous IN freezing begin forming ice, droplets form

Expansion cooling and supersaturation development

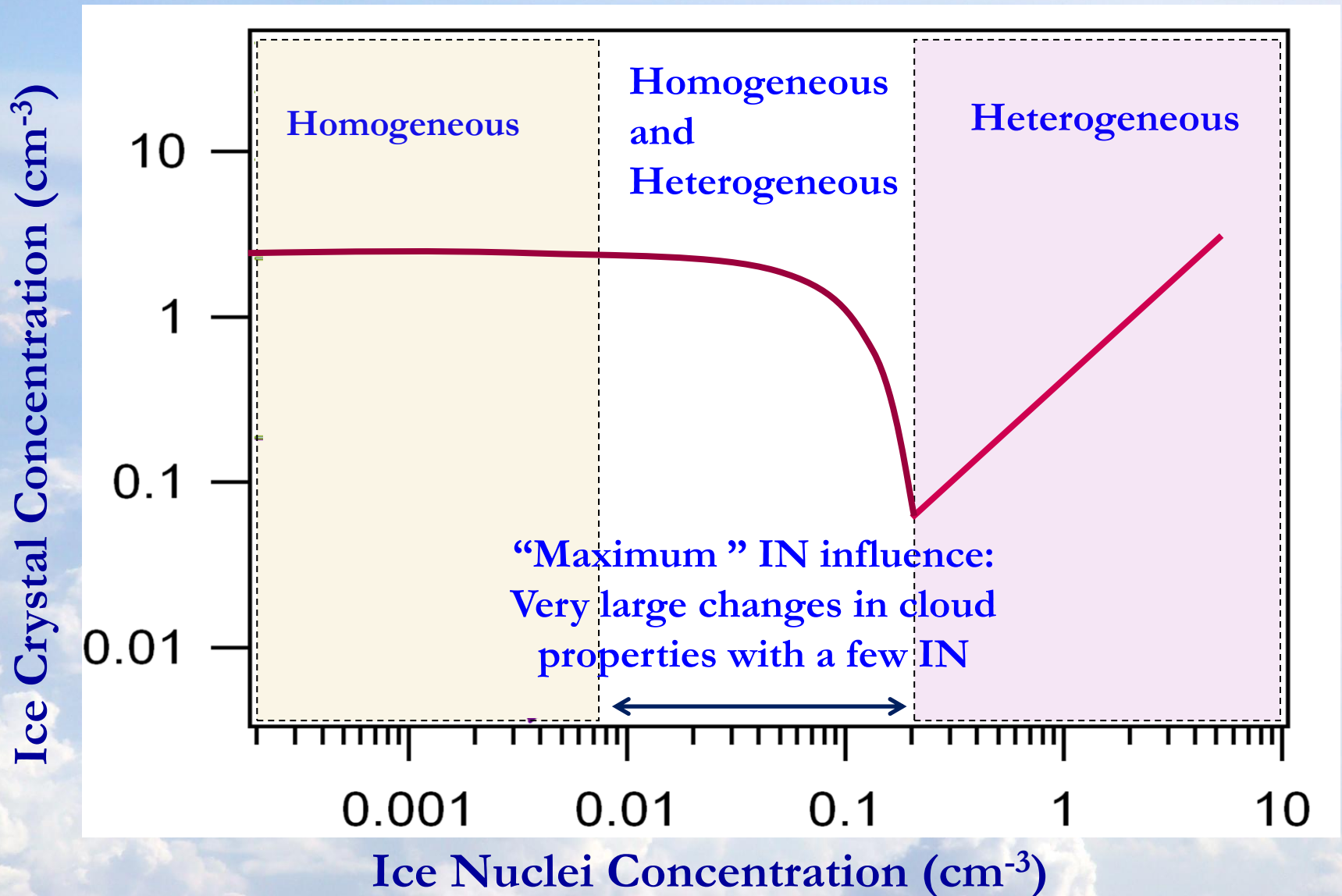
Soluble and insoluble aerosol initial distribution

Conceptual steps:

# Source of strong nonlinearity: IN effects on Ice Crystal Concentration



# Source of strong nonlinearity: IN effects on Ice Crystal Concentration



# Measuring INP: Basic Principles

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**Operation:** Expose particle sample to a known water vapor supersaturation, and measure those that become ice crystals. Depending on the freezing mechanism, also may need "preactivation" into cloud droplets.

**Desired range:** 1% - 50% supersaturation ( $S$ )

**Main challenges:**

Technique to invoke nucleation

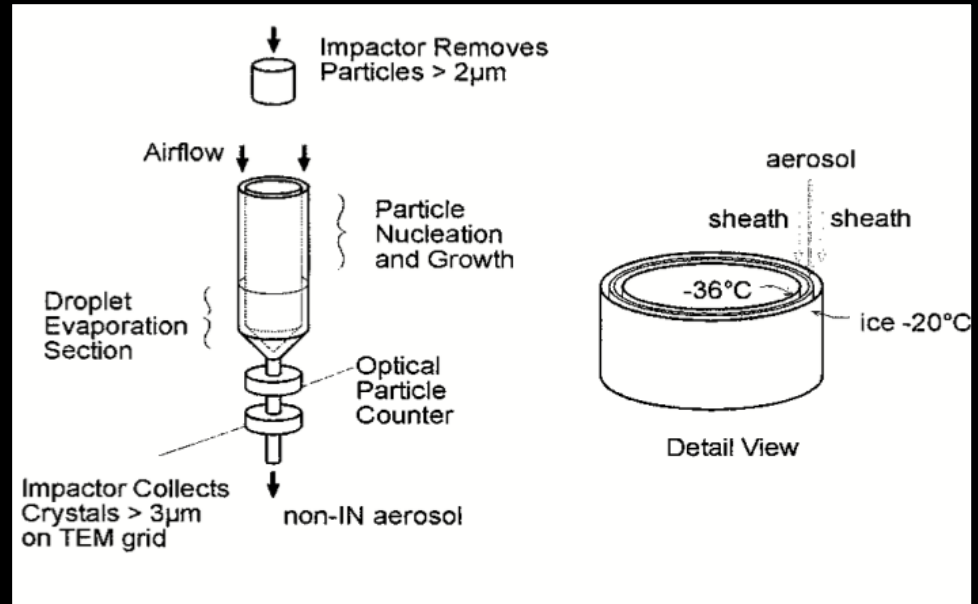
Low counting statistics (important for warmer  $T$ ).

Ice crystal detection

- Need to differentiate from particles that do not become ice crystals.
- Generally more difficult when you have dust/large non-ice particles (dust, ash).

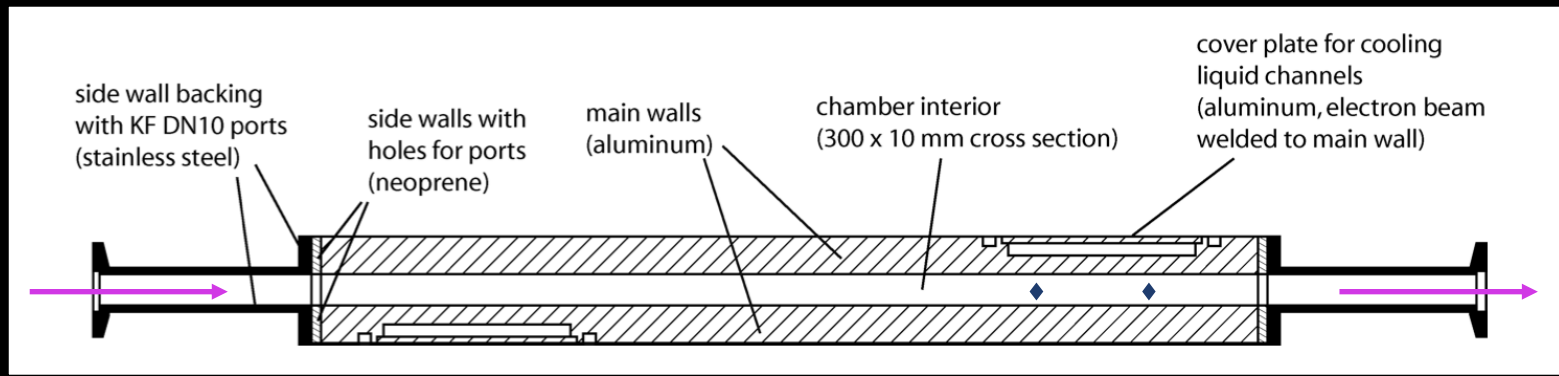
# CFDC (CONTINUOUS FLOW DIFFUSION CHAMBER)

- Developed by David C. Rogers
- The chamber consists of two ice-coated concentric cylinders at different temperatures



- In the last third of the chamber the outer cylinder are not ice-coated
- Analyze deposition and immersion modes

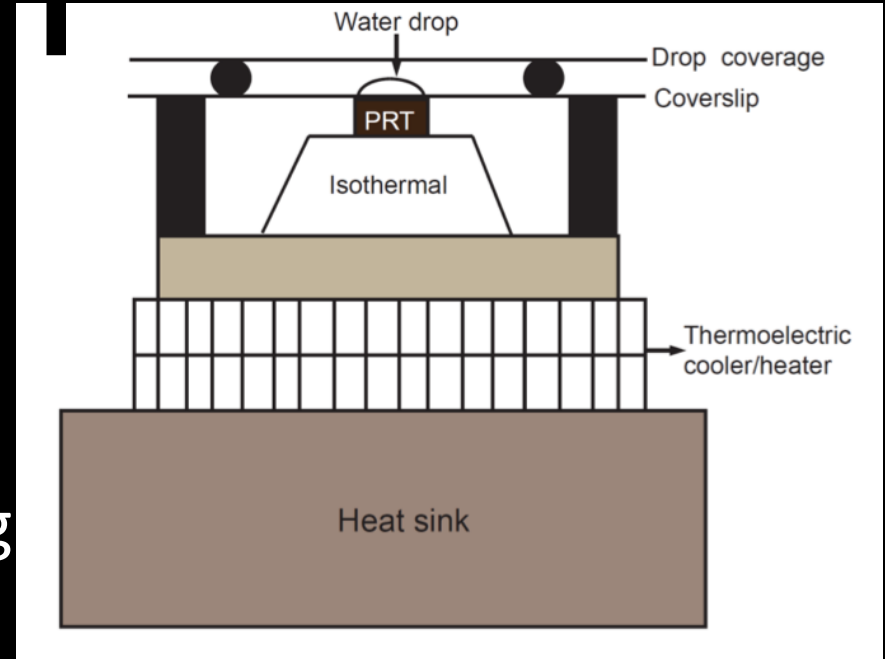
# ZINC (ZURICH ICE NUCLEATION CHAMBER)



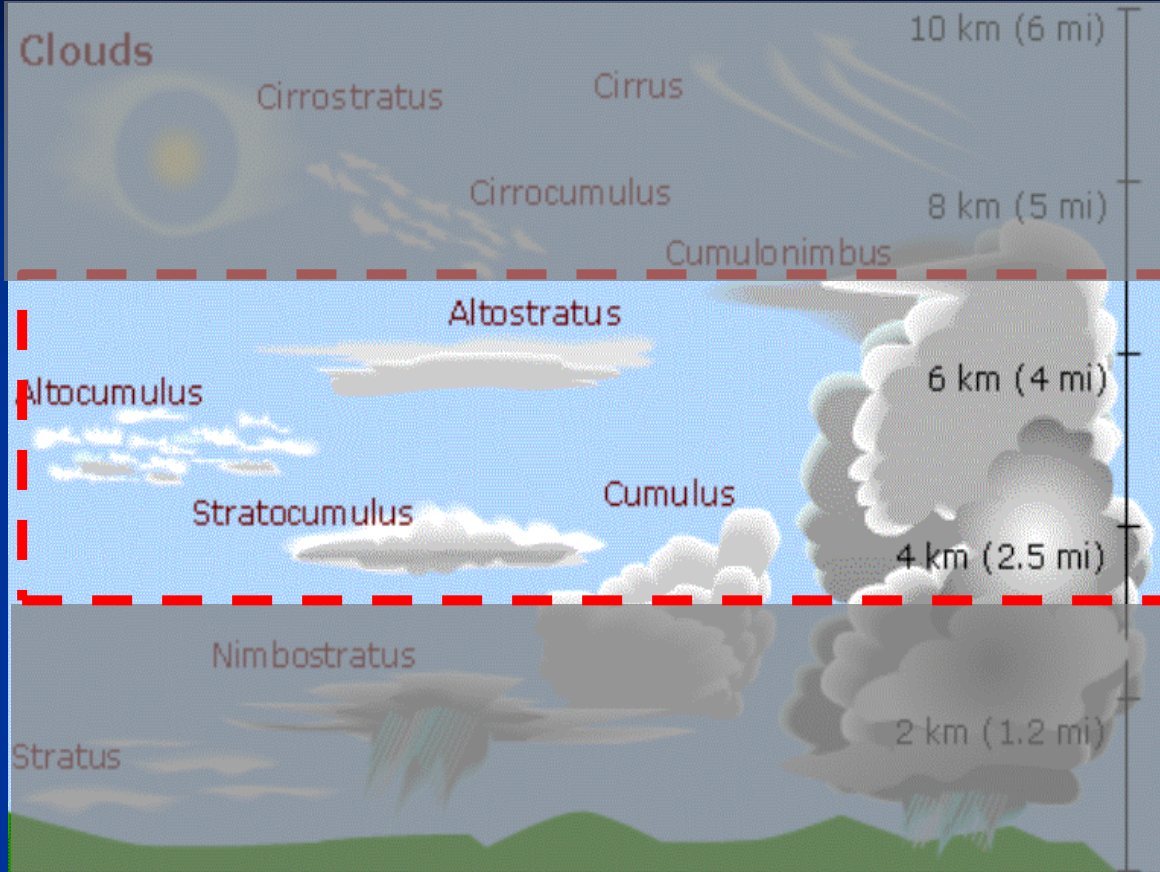
- CFDC type of chamber, which use a flat parallel plate design instead of a concentric cylinder geometry
- Can reach 236 K with ice supersaturations of up to 50%
- Analyze condensation and deposition mode
- Portable version (PINC) also available commercially.

# COLD PLATE TECHNIQUE (CONTACT FREEZING)

- Consist of a metallic surface coated with a thin layer of hydrophobic material
- Two different ways to perform a contact freezing experiment:
  - ✓ Static cold plate
  - ✓ Dynamic cold plate
- Many studies use variations of this method.



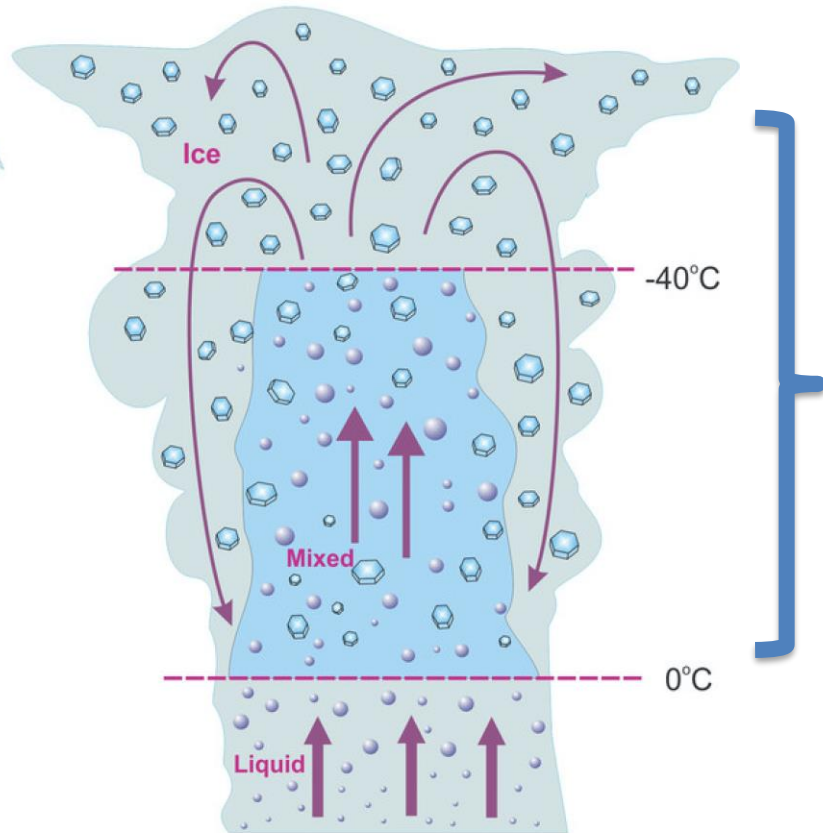
# Mixed phase clouds: "final frontier"



- **Ice (cold) clouds:**  
Ice crystals,  $T < 235$  K.  
Warm climate
- **Mixed Phase clouds:**  
Liquid droplets & ice,  
 $235\text{K} < T < 273\text{K}$   
Warm/cool climate
- **Liquid (warm) clouds:**  
Liquid droplets  
 $T > 273$  K  
Cool climate

Cloud particles are not created directly from the vapor phase but from **suspended aerosol particles**

# Mixed-Phase clouds control precipitation on a regional and global scale



Precipitation at mid- and high-latitudes mostly generated from the mixed- and ice- cloud phase

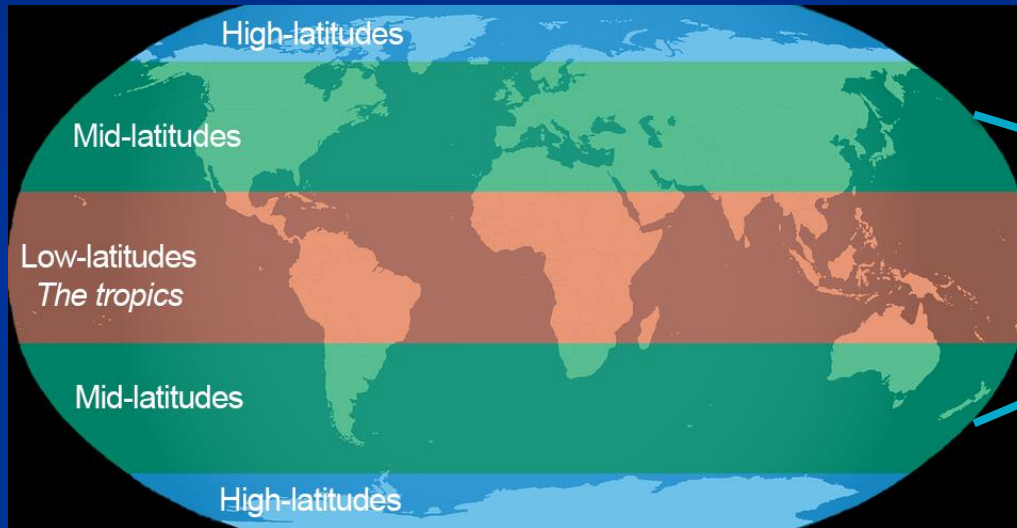
*Mulmenstadt et al. 2015*



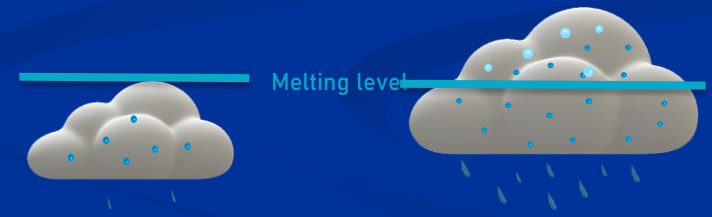
Precipitation extremes have huge impacts on economy and society at large.



# Liquid+ice (“mixed-phase”) clouds Are very important for climate



30-50% of precipitation occurs from the ice phase

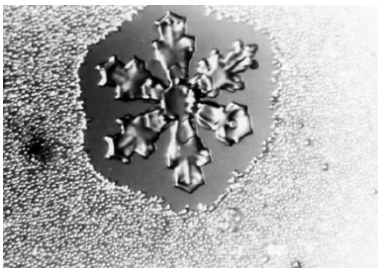
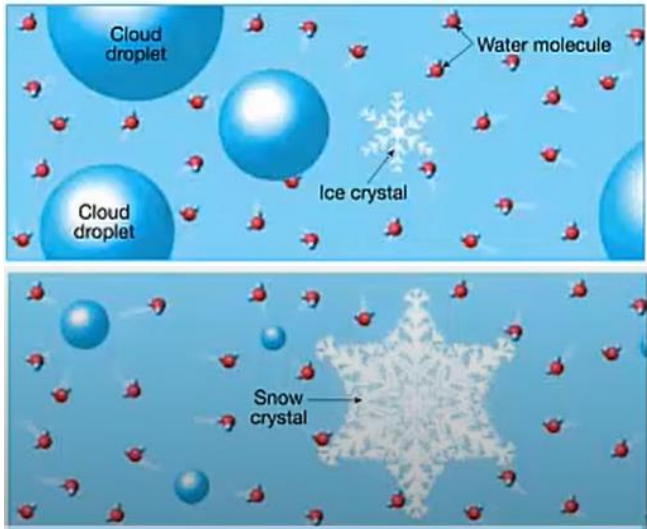


*Field and Heymsfield, 2015*  
*Mülmenstädt et al. 2015*

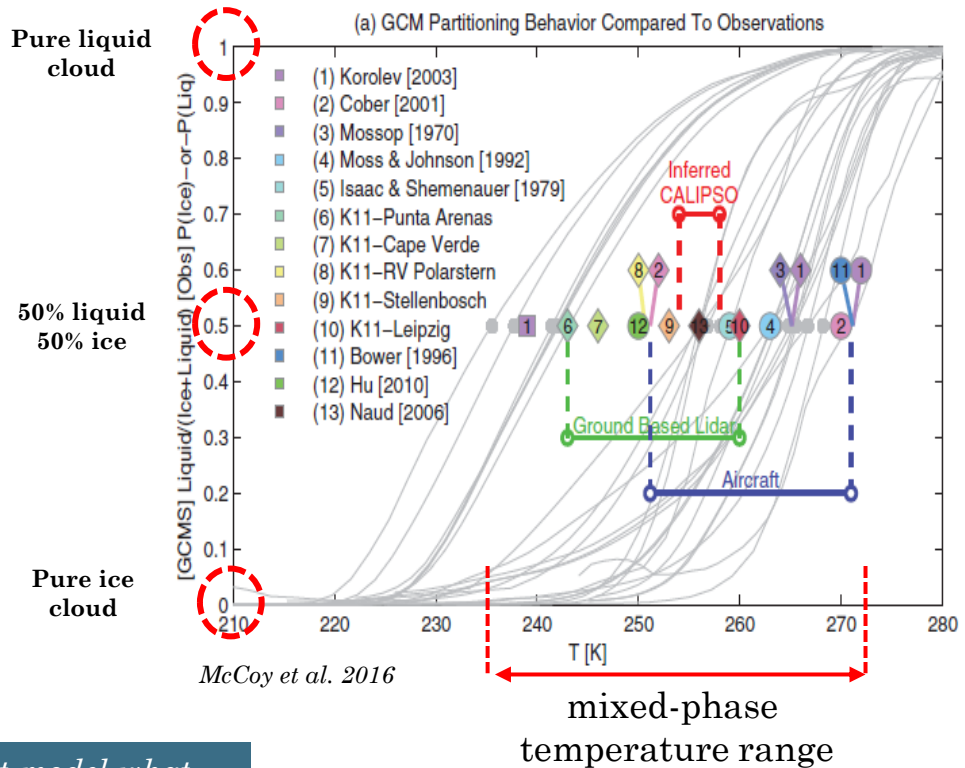
*“...much of what is rain, when it arrives at the surface of the Earth, might have been snow, when it began its descent . . .”*

# Challenges of representing MPCs within modeling frameworks

- ✓ Important to predict the **amount** and **distribution** of ice and liquid (liquid-ice phase partitioning) in MPCs
- ✓ Models tend to convert water to the ice phase too aggressively



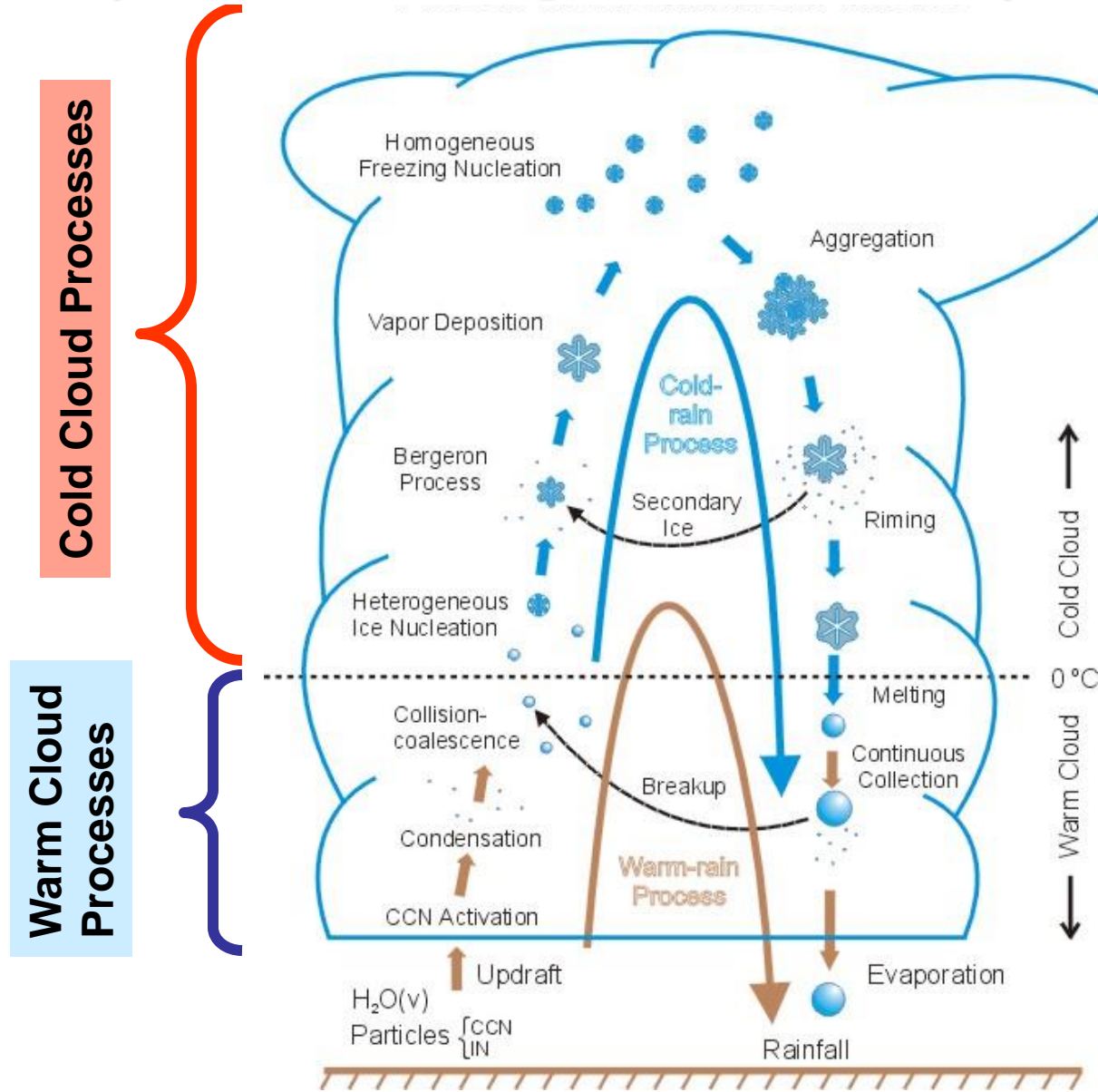
*Wegener-Bergeron-Findeisen process (WBF)*



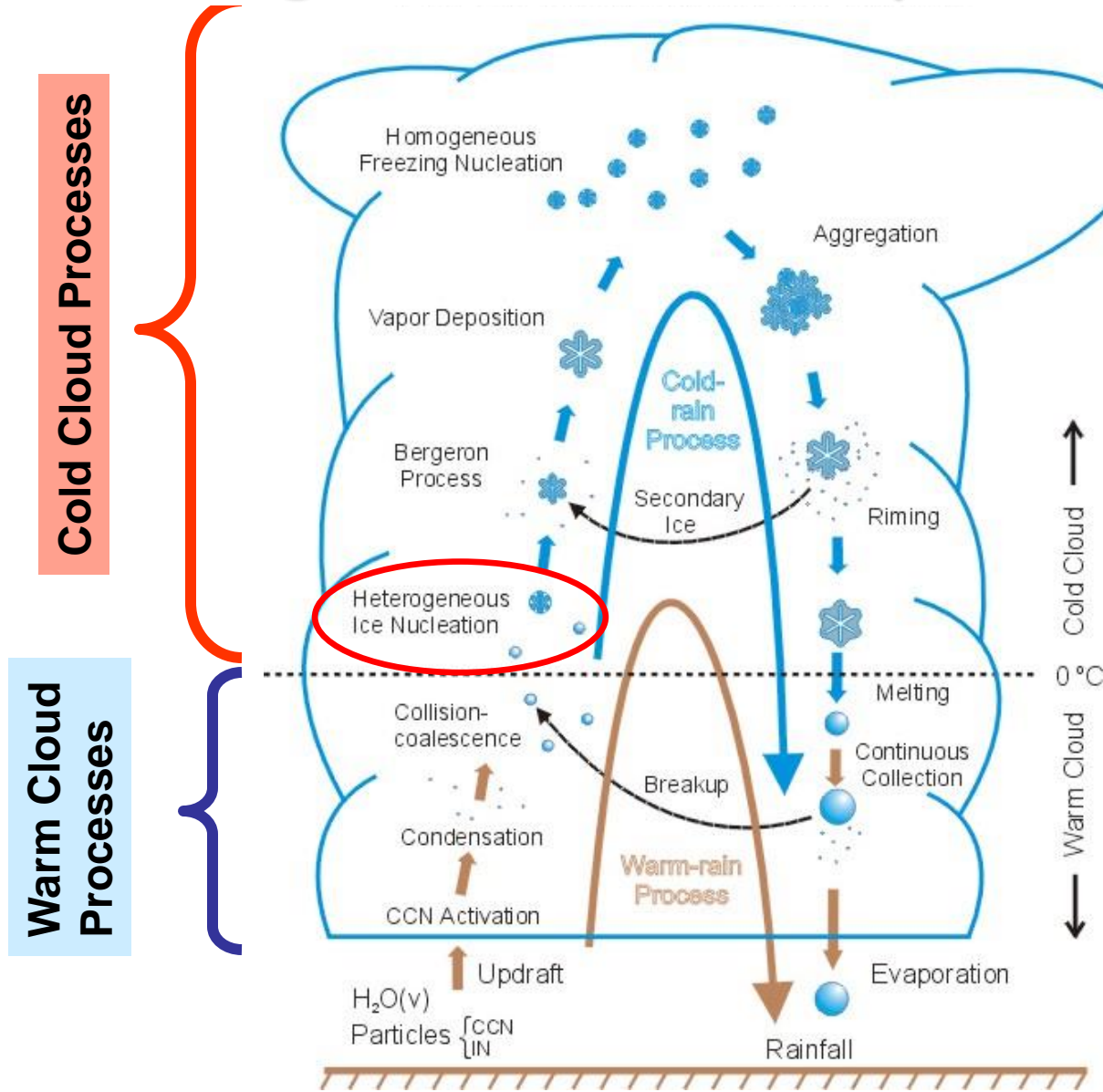
McCoy et al. 2016

*“One cannot model what one does not understand”*

# Ice production/growth interplay in clouds



# The ice grows at the expense of drops



# Primary ice production mechanisms

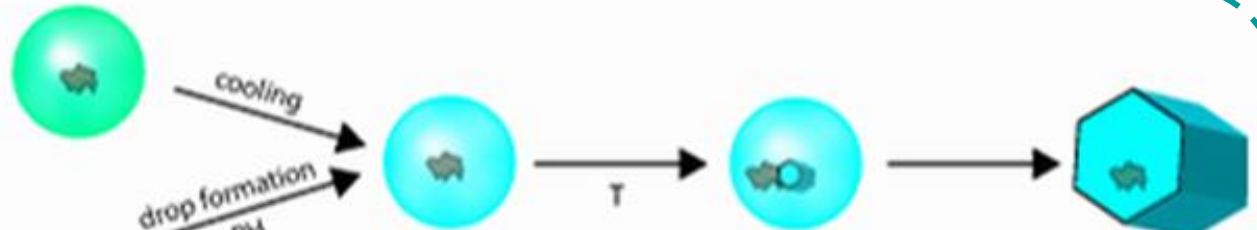
Homogeneous nucleation



Deposition nucleation



Immersion freezing




Condensation freezing

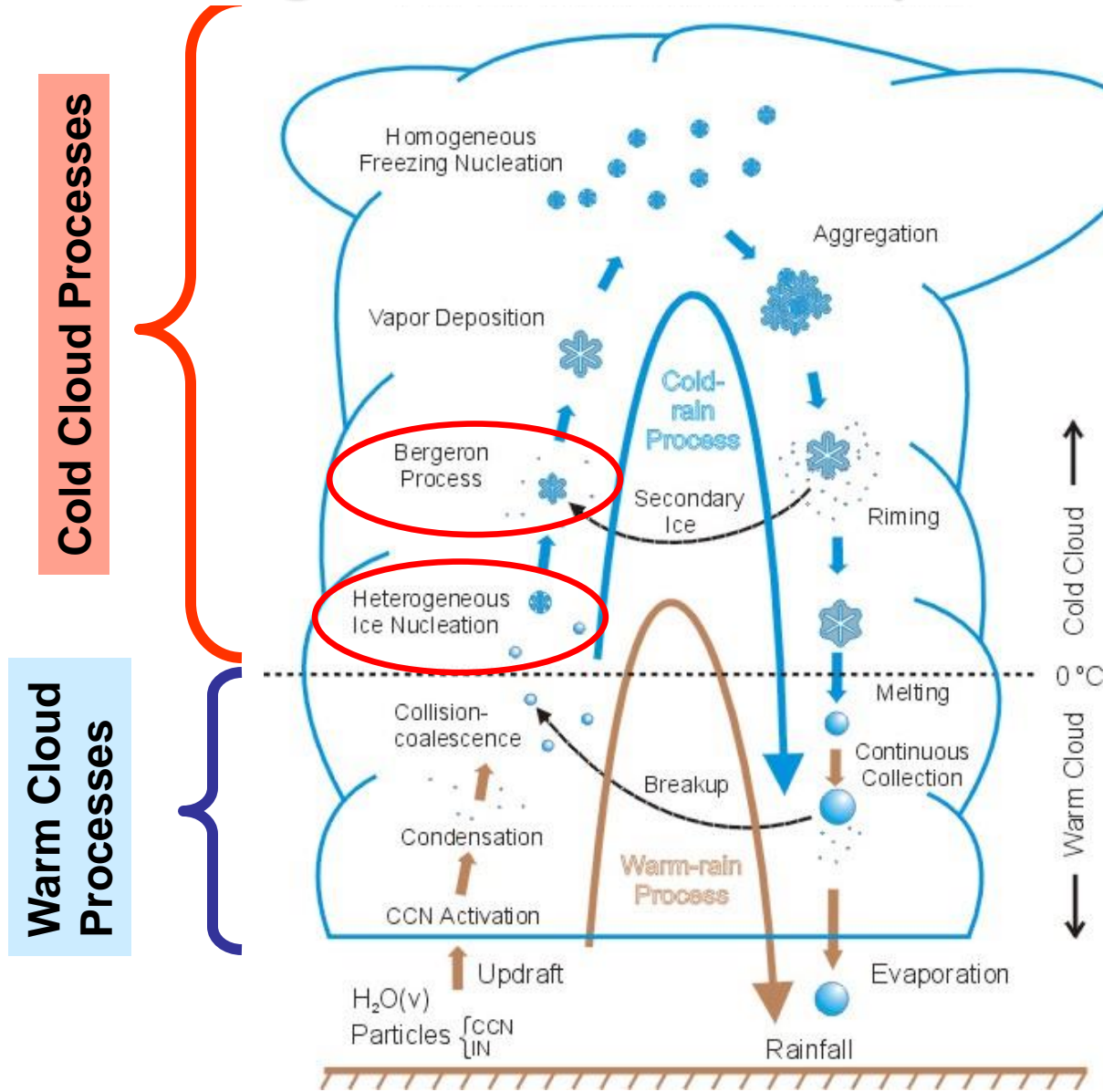


Contact freezing



 = heterogeneous ice nucleus (e.g. mineral dust)

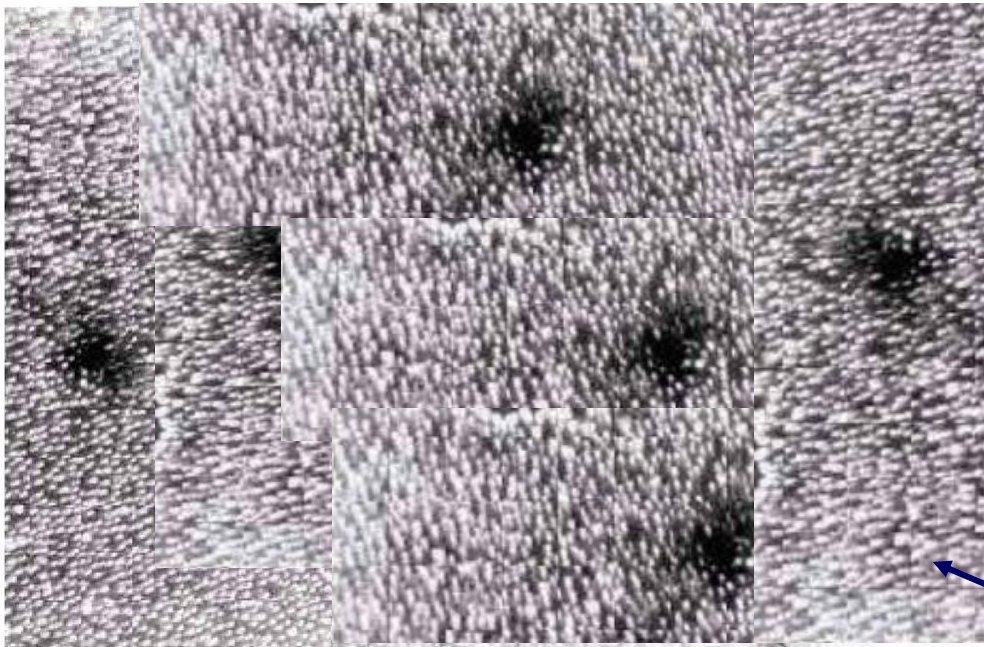
# The ice grows at the expense of drops



# Bergeron-Findesien process

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Critical for the microphysical evolution of mixed-phase clouds

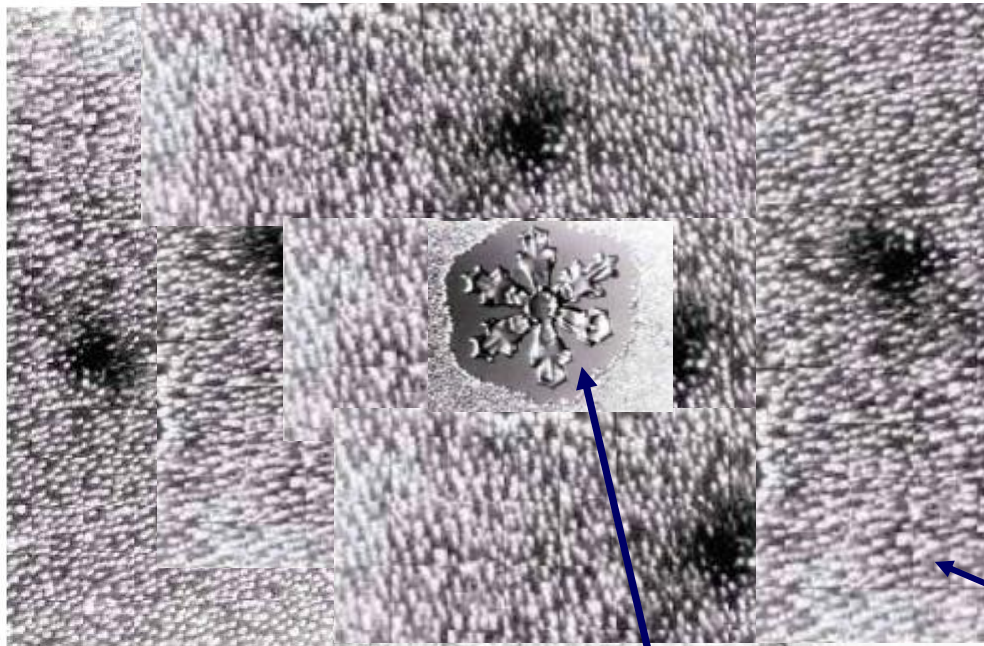


→ Droplets on a plate

# Bergeron-Findesien process

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Critical for the microphysical evolution of mixed-phase clouds



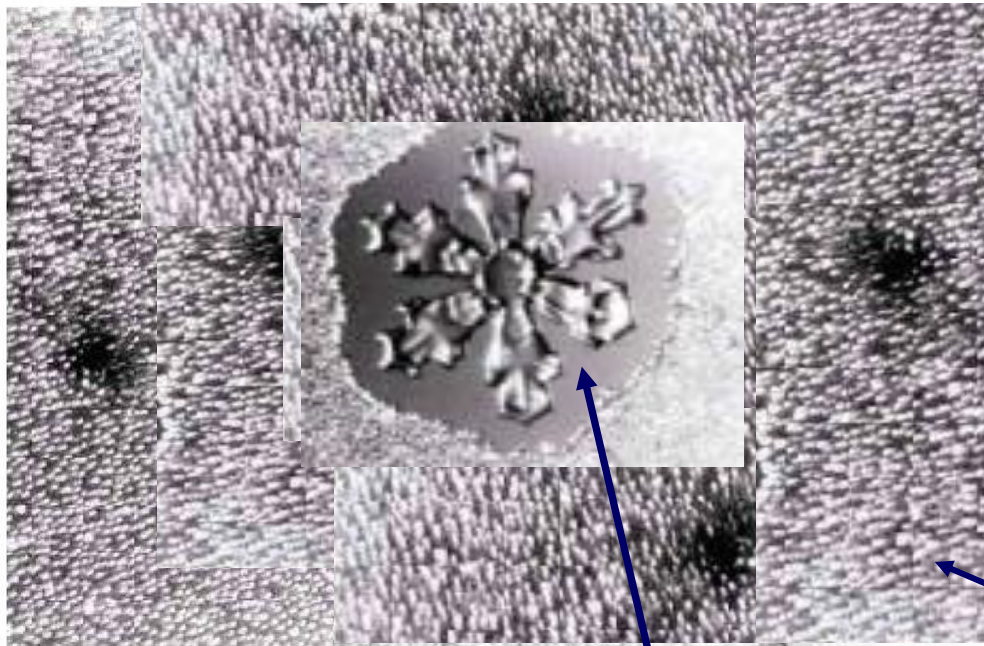
Droplets on a plate

Crystal forms

# Bergeron-Findeisen process

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Critical for the microphysical evolution of mixed-phase clouds  
Vapor pressure over liquid water is HIGHER than over ice, so mass transfers between them.



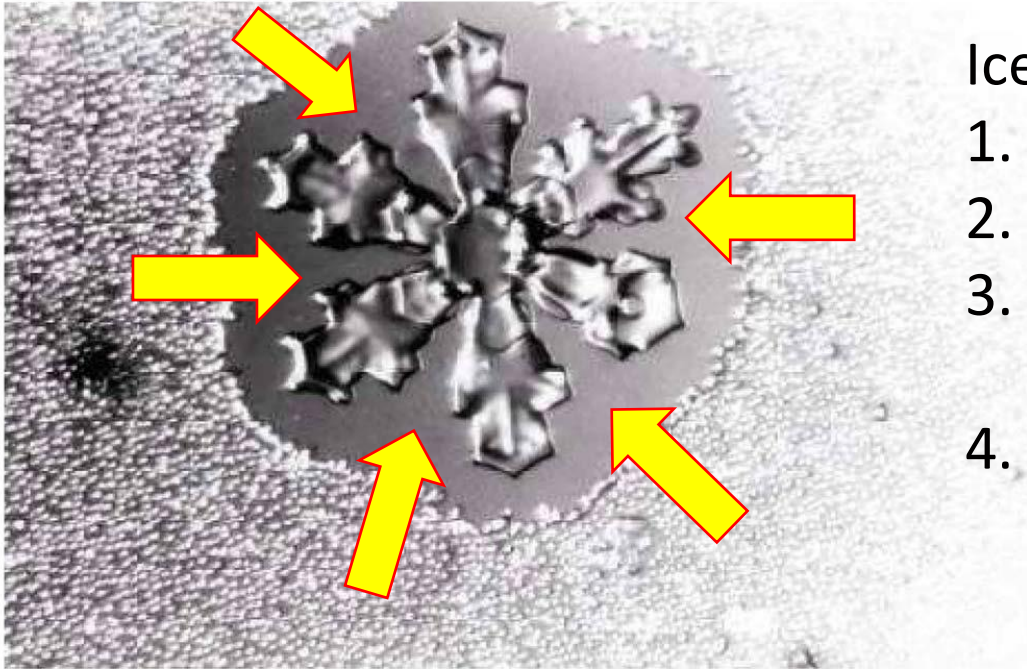
Droplets on a plate

Crystal grows

# Bergeron-Findesien process

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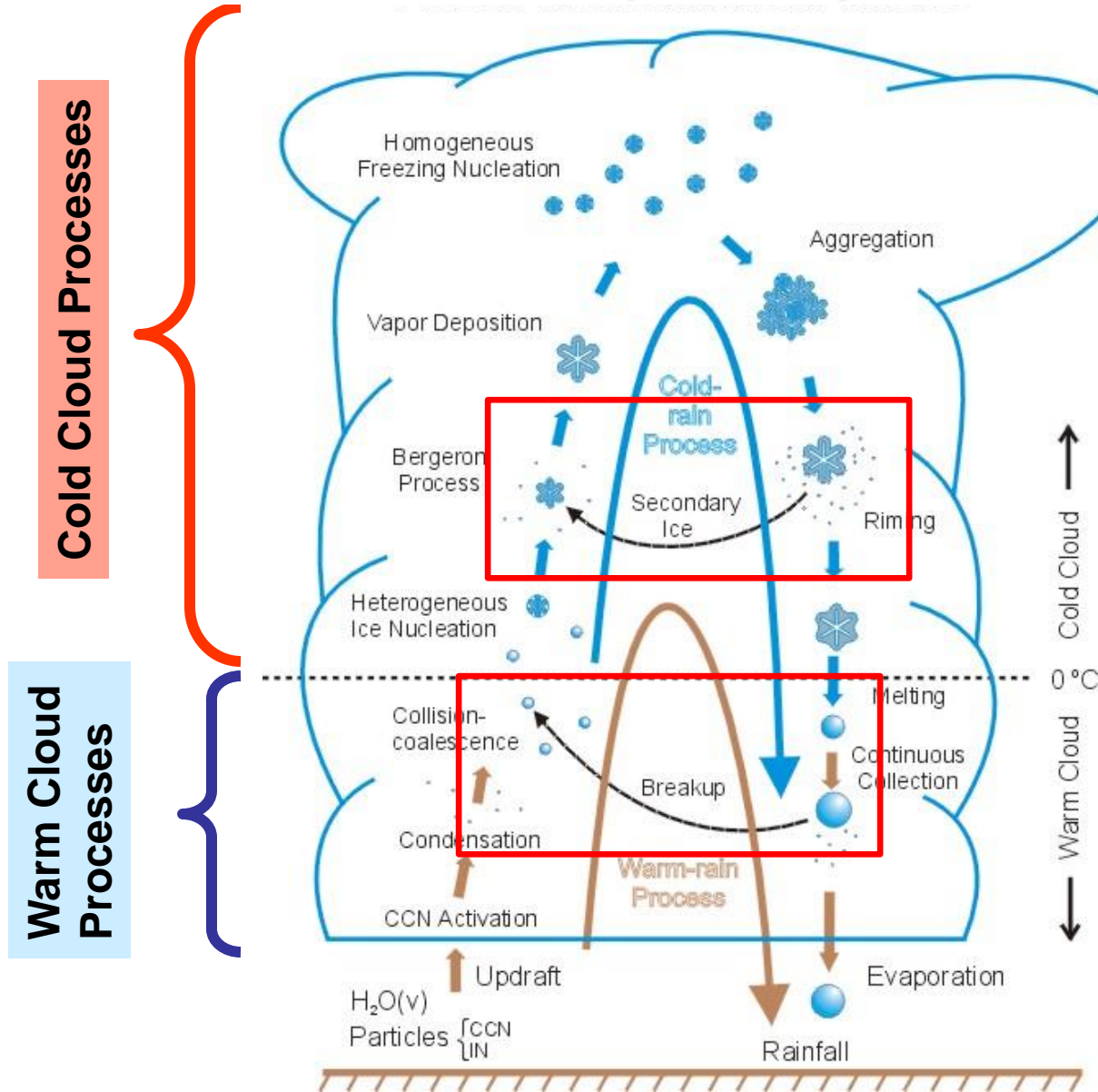


Ice crystals near droplets cause:

1. Liquid drops to evaporate
2. Vapor deposits on ice
3. Process continues until liquid water completely gone.
4. General particle size increases considerably from BF process because  $IN \ll CCN$

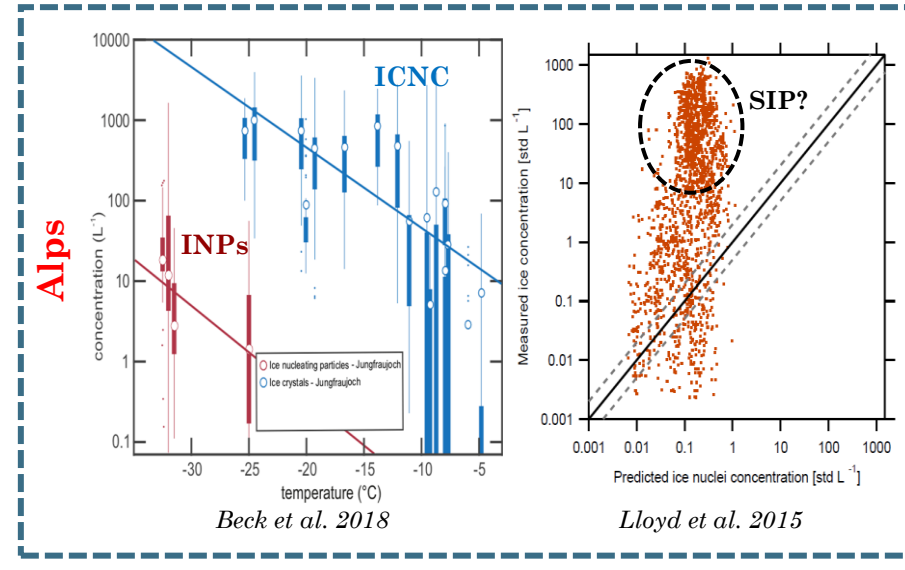
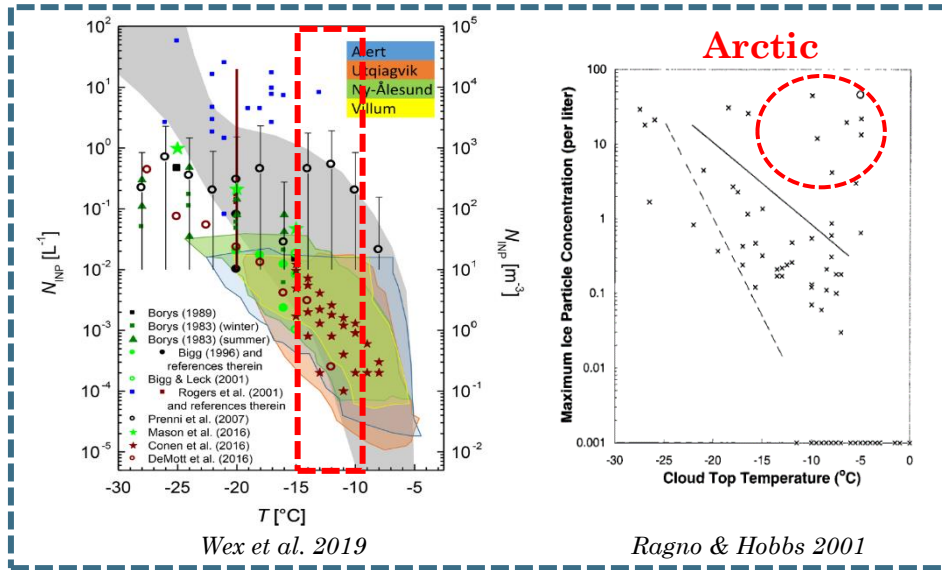
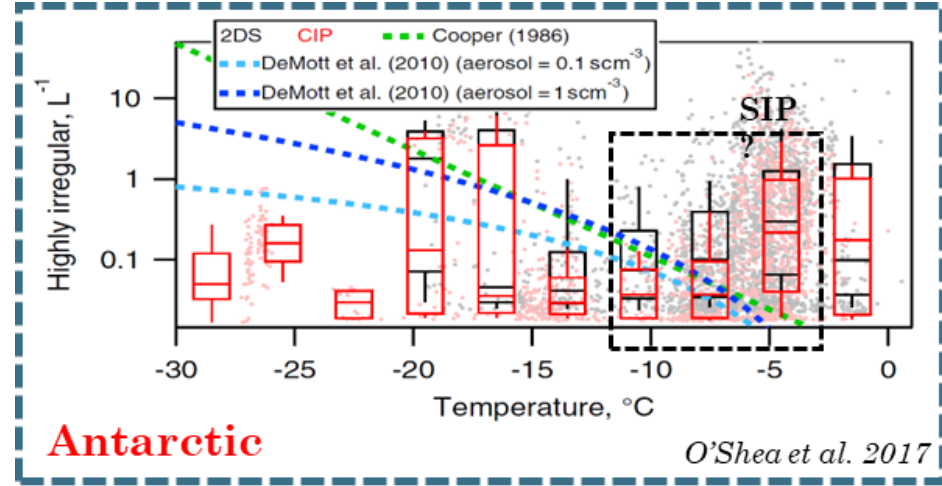
Latent heat release important for dynamical evolution of clouds.  
Shifts in particle size affects radiation and precipitation of cloud.

# Where secondary ice production "lies"



# Measured Ice Crystal Number is much higher than pre-cloud INPs!

- ✓ Ice Nucleating Particles (INPs) are few in remote polar regions - compared to the ice crystal (ICNCs)
- ✓ Alpine (orographic) clouds have the same behavior.
- ✓ Secondary Ice Production (SIP) processes must be invoked to explain the large difference between INPs and ICNCs

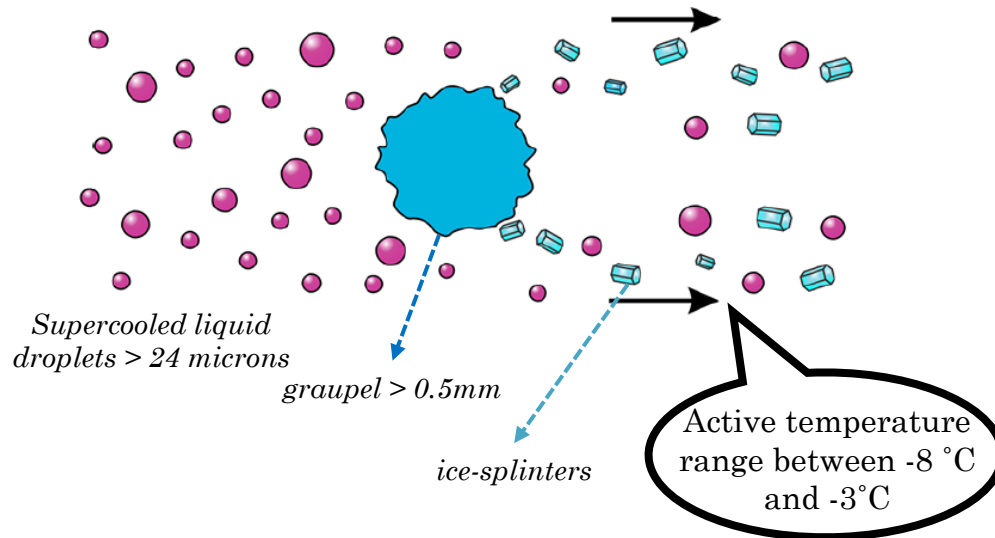


# Measured Ice Crystal Number is much higher than pre-cloud INPs!

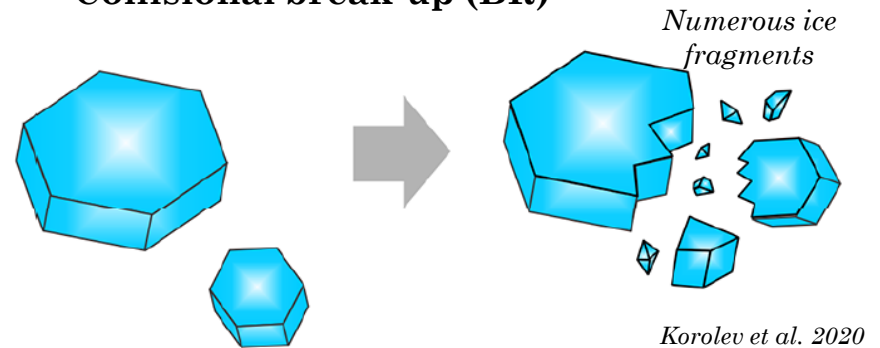
## The cause of this cloud-ice paradox → Secondary Ice Production (SIP)\*

\* SIP = multiplication of primary ice crystals through “other processes” not involving INPs

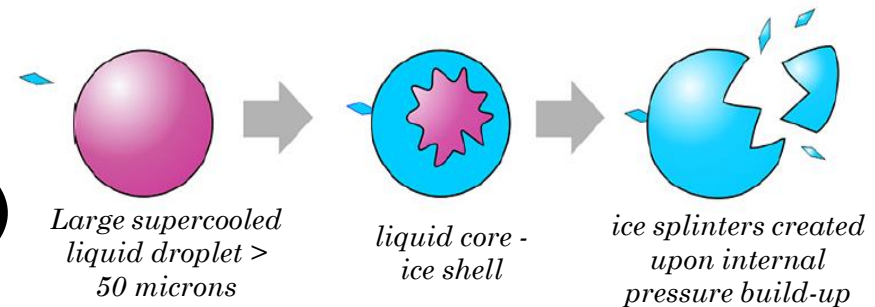
### ✓ Rime Splintering (RS) or the Hallett-Mossop process (H-M)



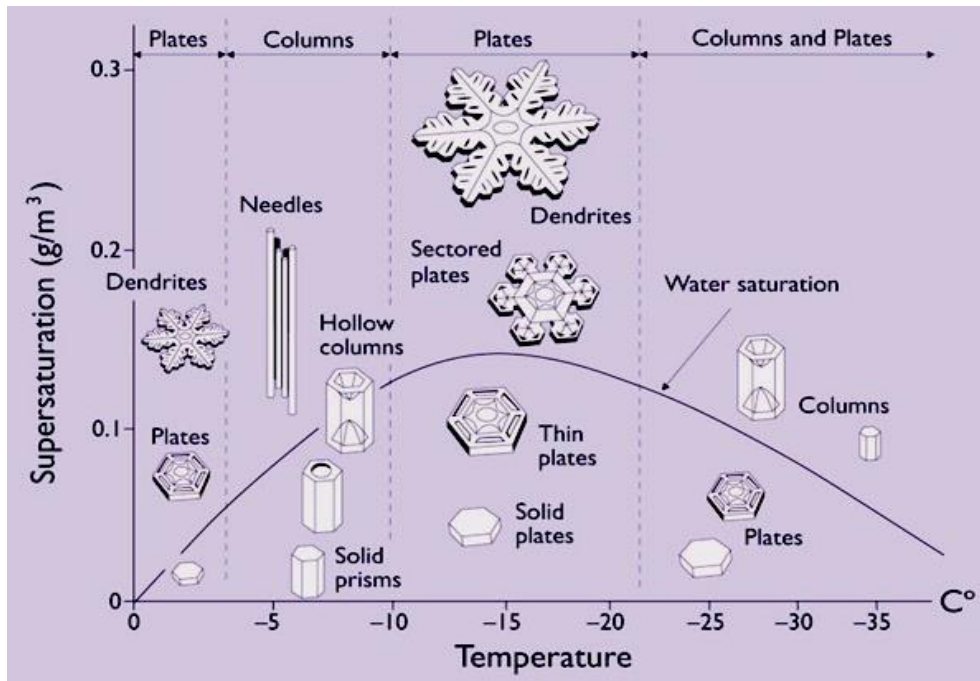
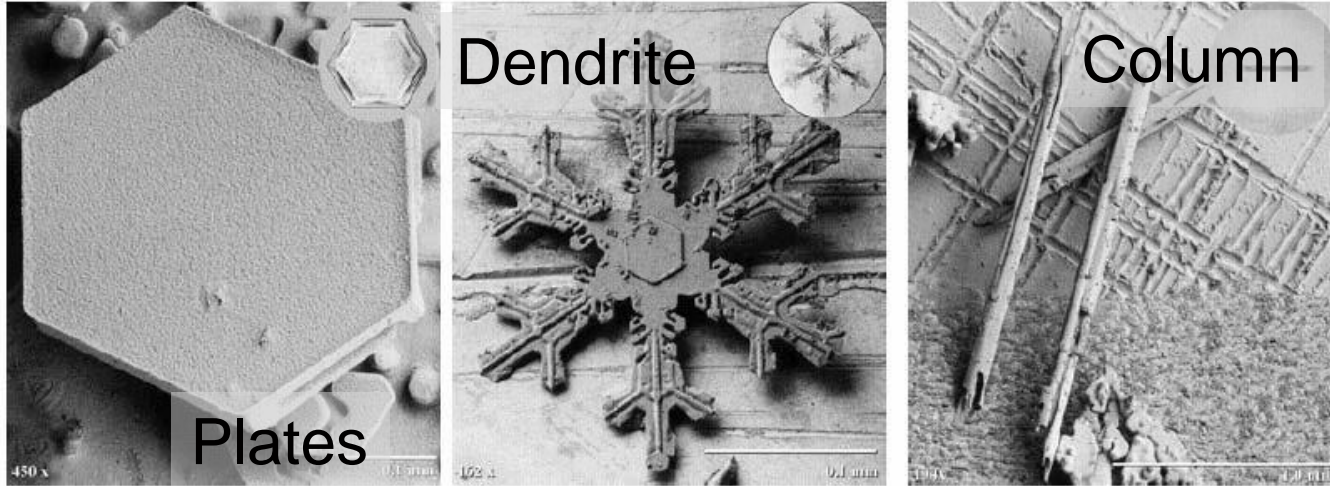
### ✓ Collisional break-up (BR)



### ✓ Droplet Shattering (DS) during freezing



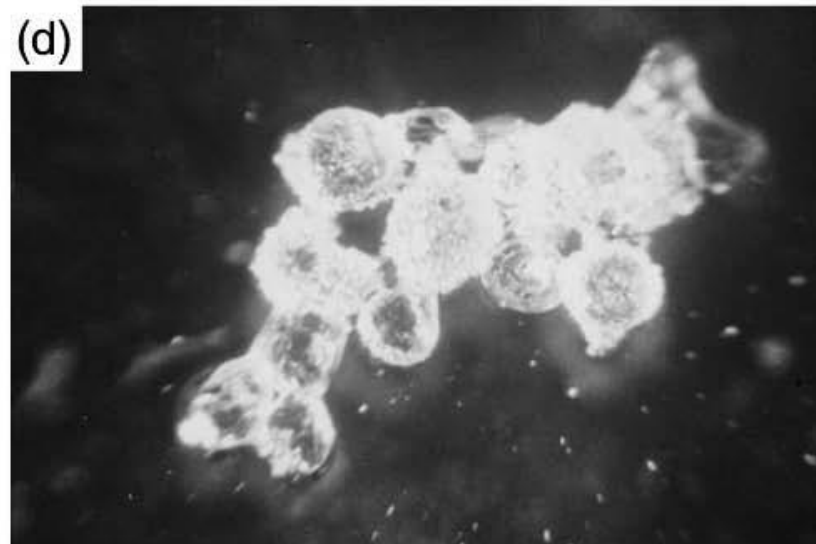
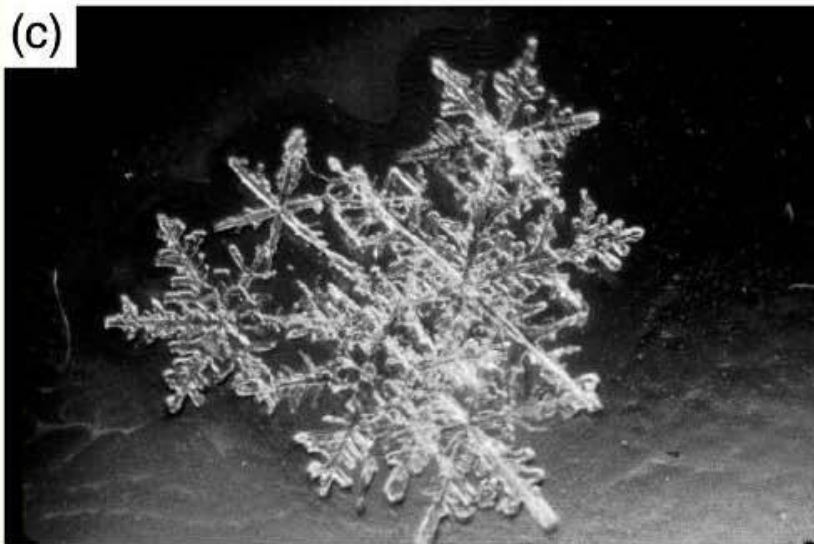
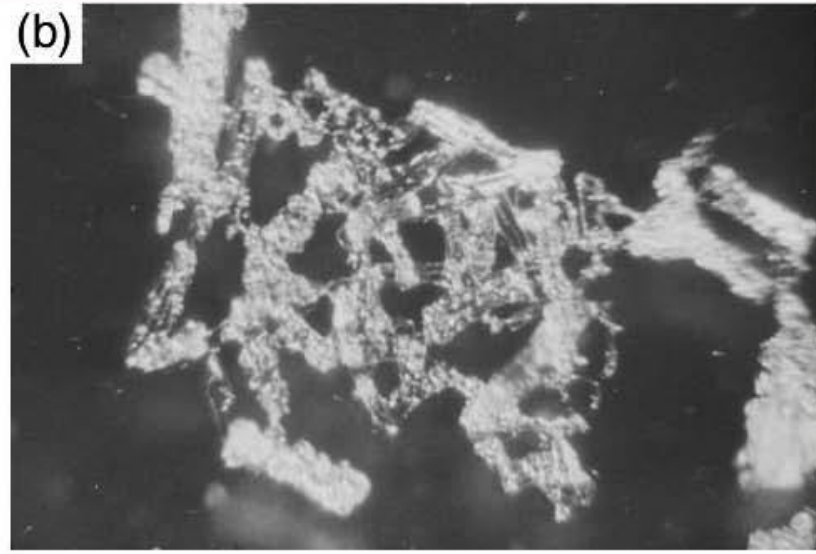
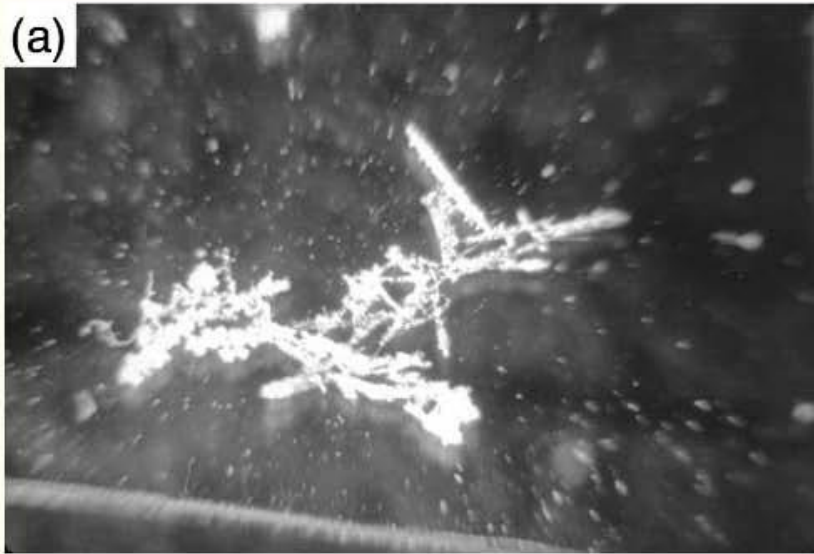
# "Pristine" ice crystals - before riming



Primary ice production

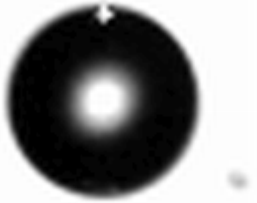
Growth via water vapor deposition only.

# "Pristine" ice crystals - after riming



# Other mechanisms – Droplet Explosions

-12 ms



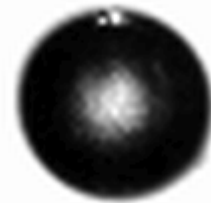
0 ms



1.4 ms



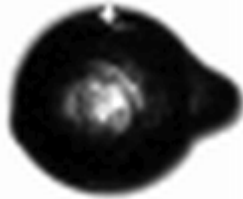
10.7 ms



699.8 ms



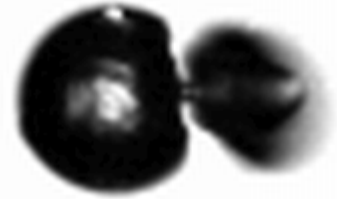
867.07 ms



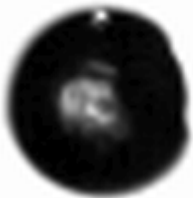
867.10 ms



867.14 ms



867.23 ms



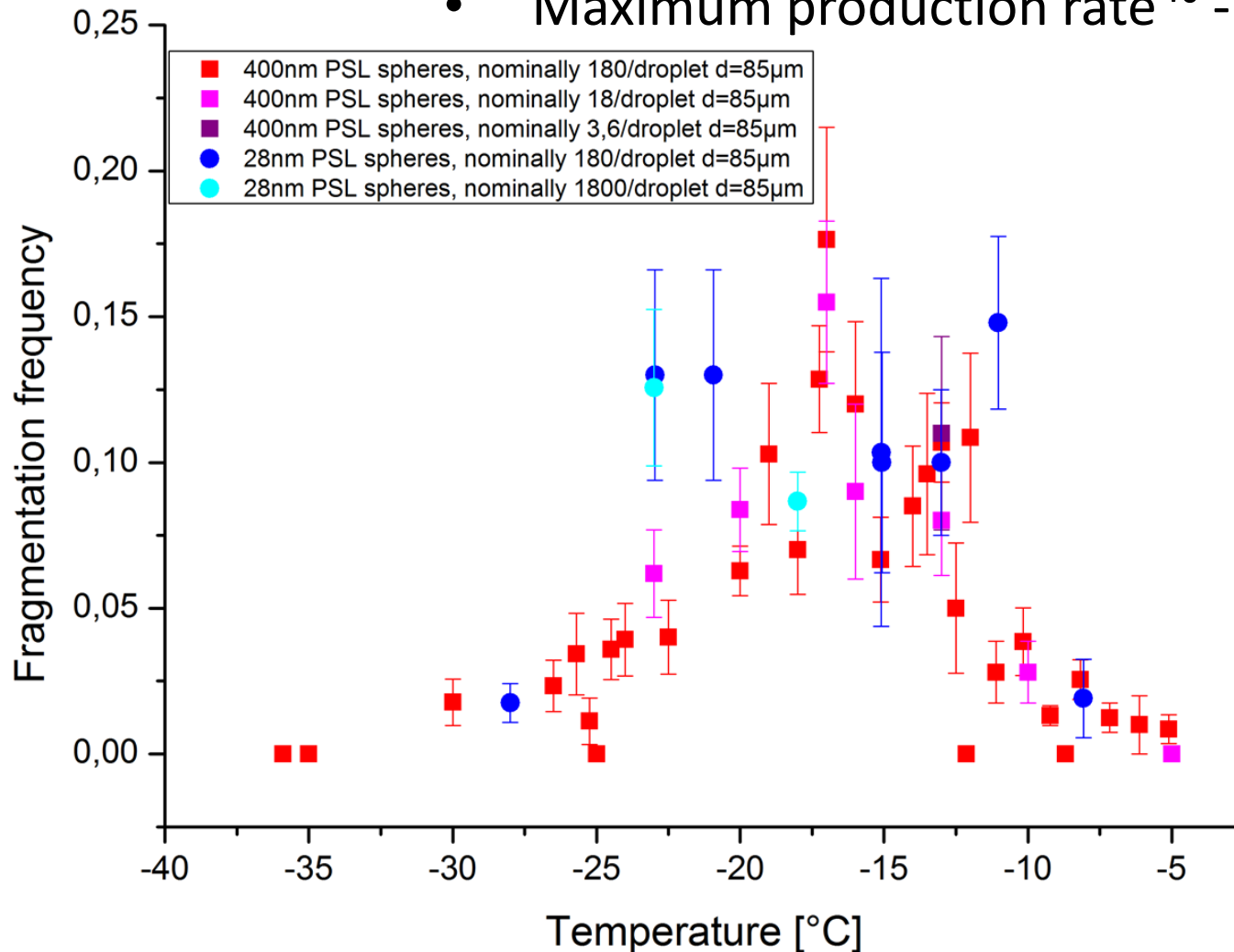
867.41 ms



*From Leisner et al.*

# Other mechanisms – Droplet Explosions (Leisner, Kieselev et al.)

- Maximum production rate  $\sim -16^{\circ}\text{C}$



# Ice-ice collisions

- Two ice spheres were brought into controlled collision
- Count the fragments produced during collision under different T, riming condition, size,

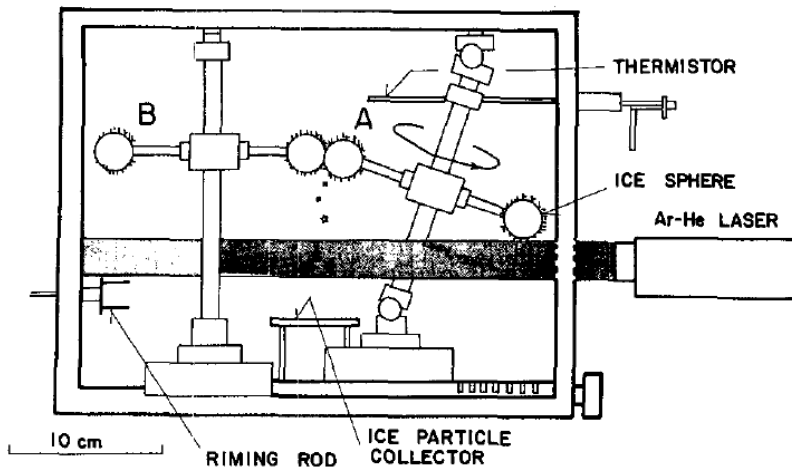


FIG. 1. Experimental apparatus: (A) ice spheres (right) were set up to rotate at a tangential speed of  $4 \text{ m s}^{-1}$ , while (B) ice spheres (left) were stationary. The ejected ice particles were collected on a plate at the bottom of the chamber. A thermistor for temperature measurement was installed at the top. Cloud droplets were supplied through the tube from the center of the right wall. During the actual experiment, all parts except ice spheres exposed to the cloud droplets were cleaned by ethylene glycol for frost prevention.

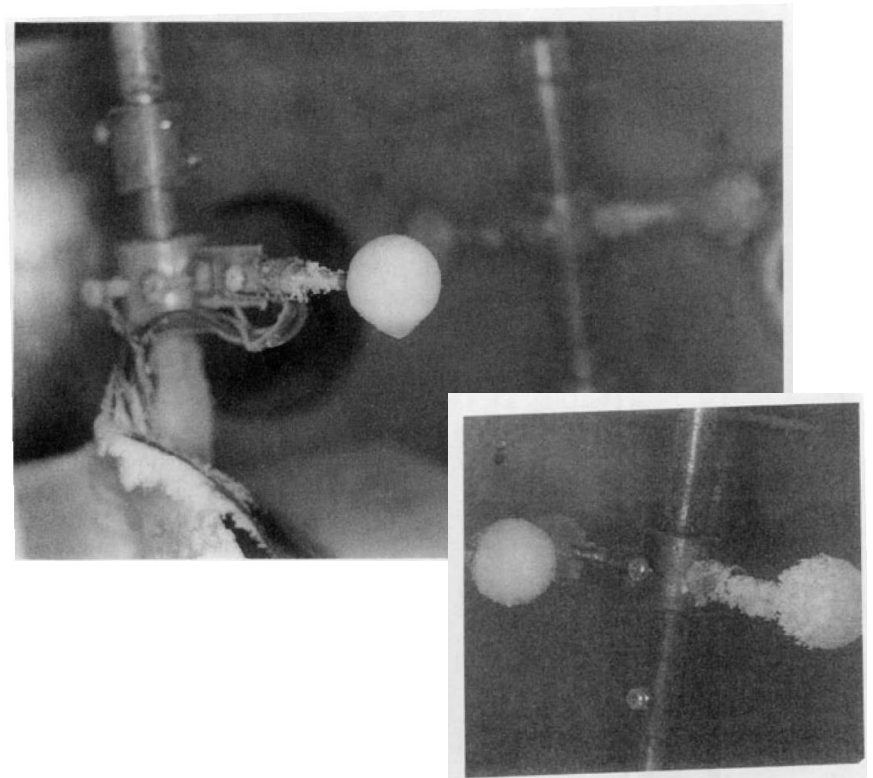


FIG. 2. Ice spheres in the chamber. Ice particle collecting plate is seen at the bottom of left picture. (Left) Stationary ice sphere after depositional growth. (Right) Rotating rimed ice sphere (far right).

**Takahashi et al. (1995)**

# Ice-ice collisions

- Larger number of fragments produced vs. other mechanisms
- Maximum production rate  $\sim -16^{\circ}\text{C}$

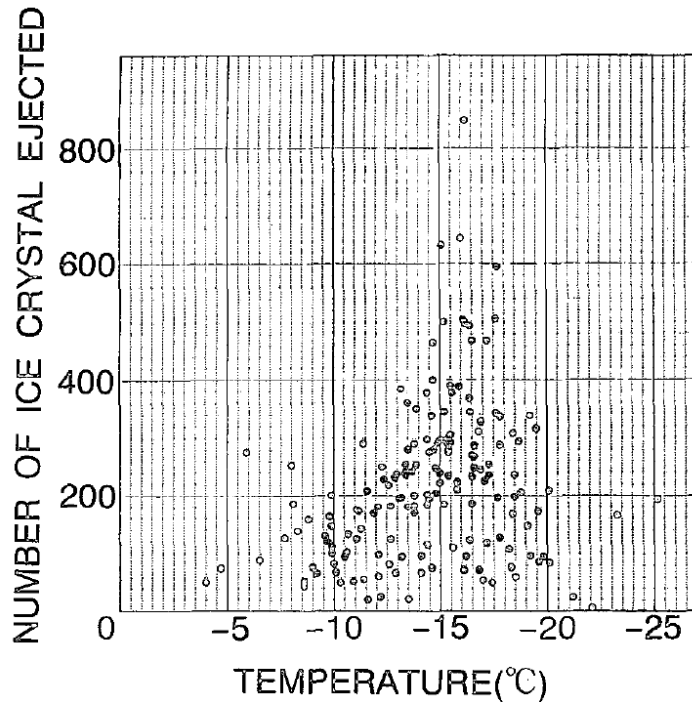


FIG. 4. Total number of ejected ice particles as a function of chamber temperature. The number of ice crystals collected by the main plate was simply multiplied by 4. The total number is therefore approximate. Cases of various collision forces were all included. Collision force was on the order of 500 dyn.

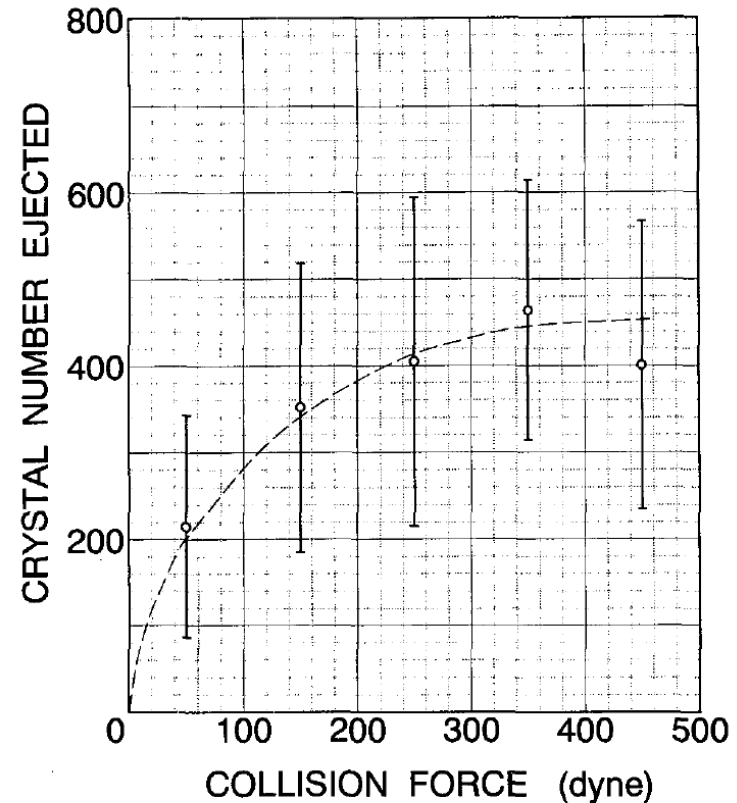
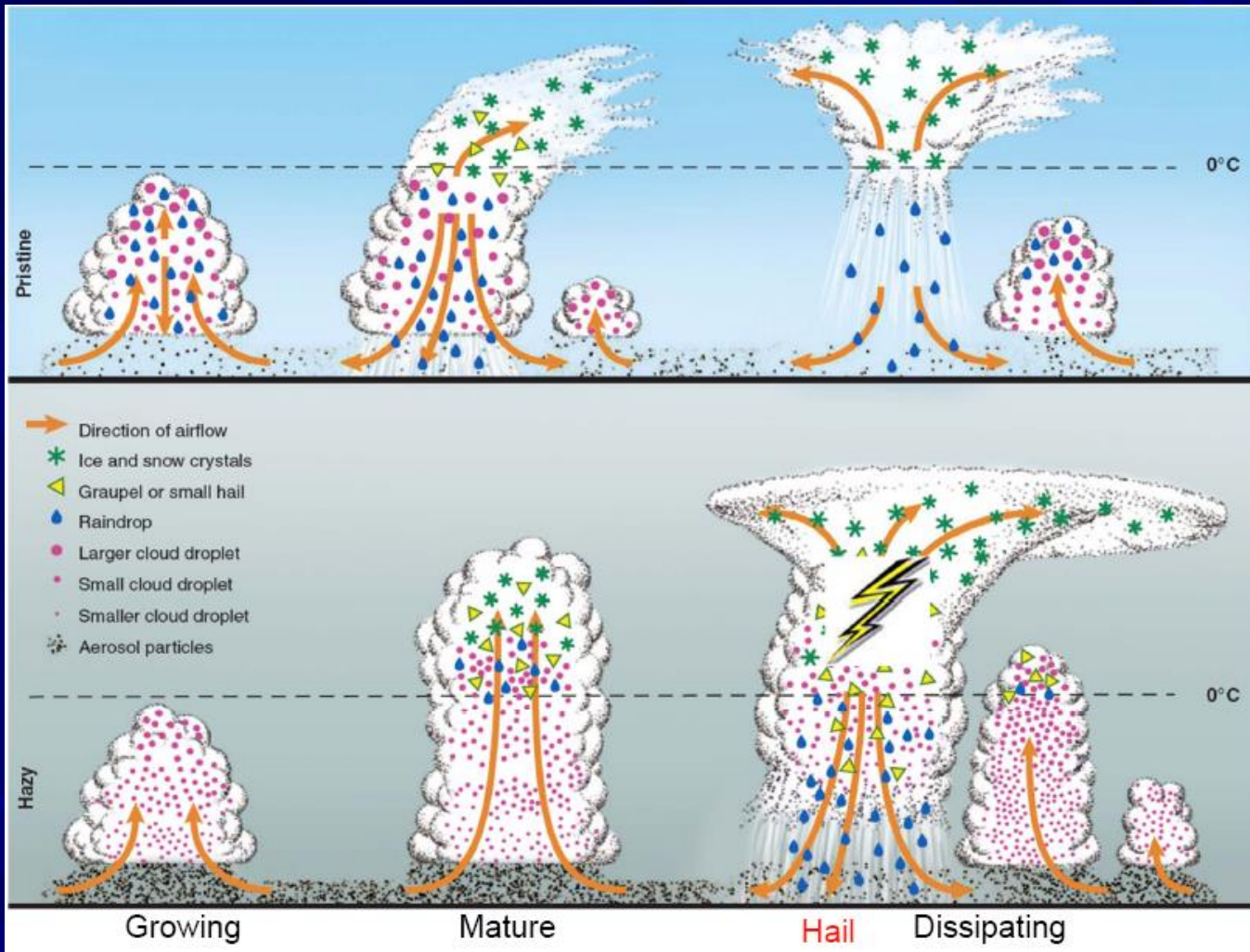


FIG. 5. Total ejected ice particle numbers vs collision force. Thick marks represent standard deviations of the measurements, and the thin dashed line is the smoothed value. Data was grouped in 100-dyn range for analysis.

# Aerosol-Precipitation Feedbacks



Aerosols reduce drizzle.

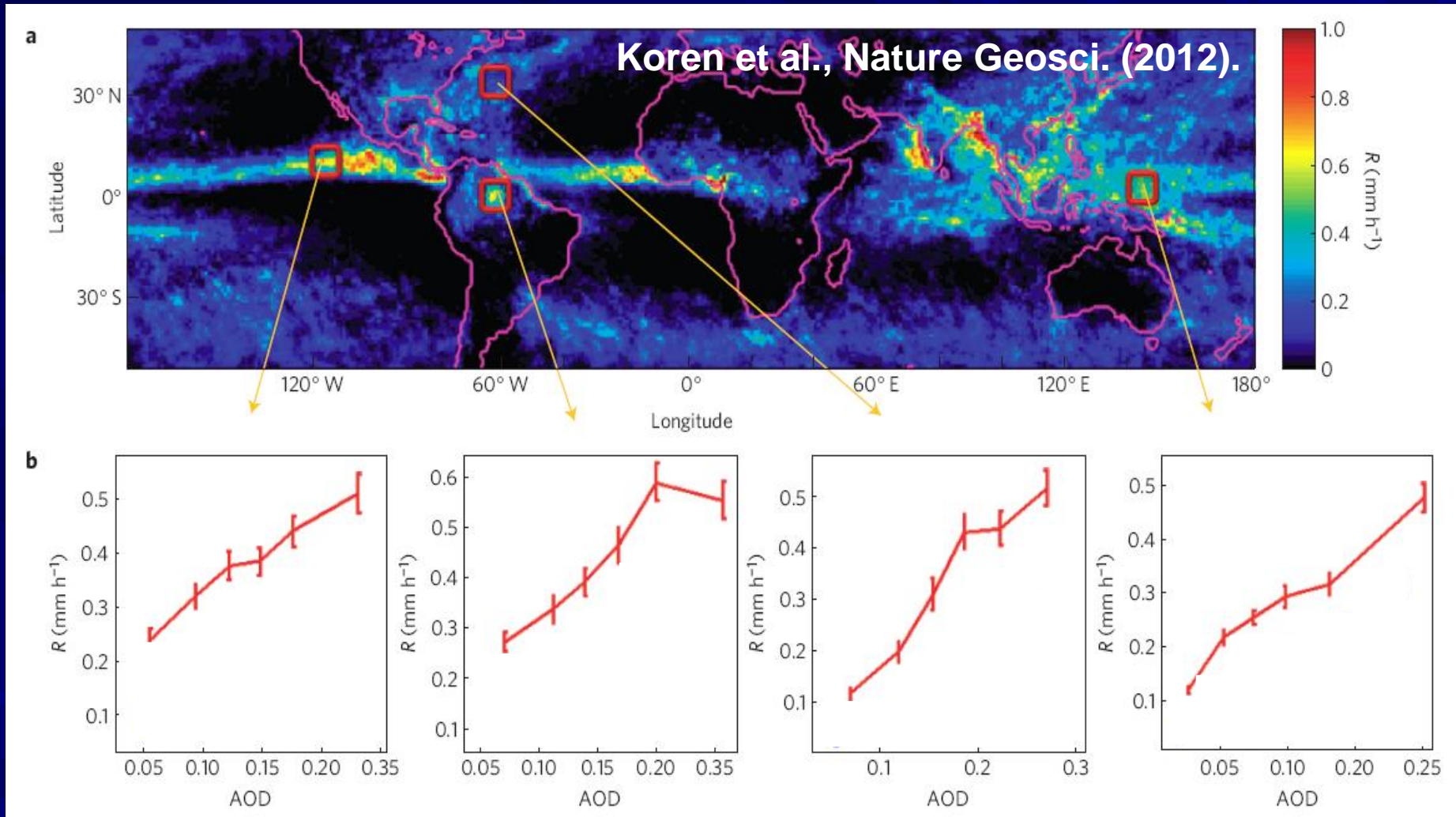
More water reaches the freezing level.

More latent heat is released during freezing.

Convective invigoration.

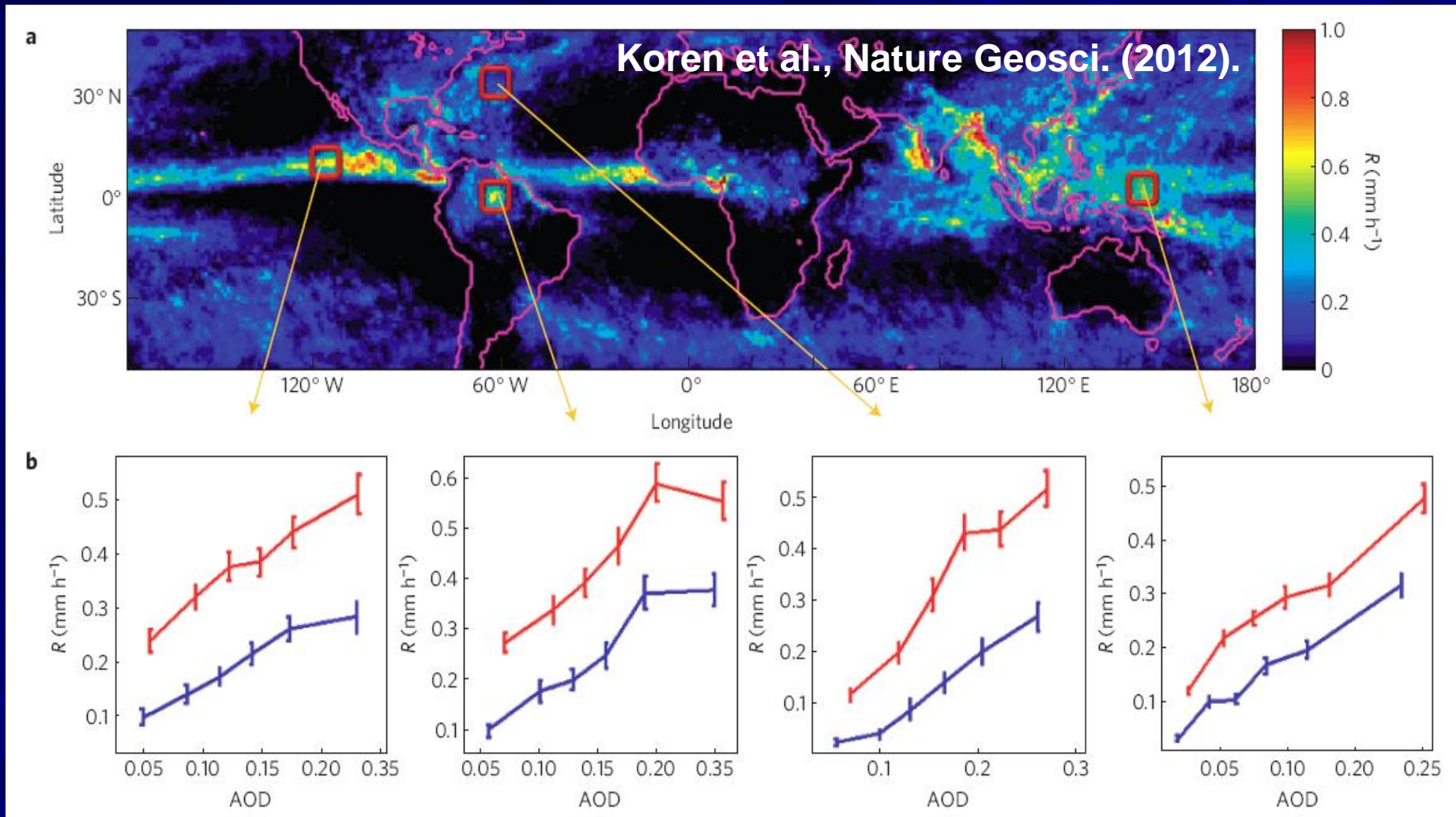
Dynamical feedbacks from aerosol effects can change cloud structure/precipitation patterns (cloud feedback?).

# Convective invigoration is everywhere



“Invigoration of clouds and the intensification of rain rates is a preferred response to an increase in aerosol concentration.”

# Aerosol-Precipitation interactions



**“Invigoration of clouds and the intensification of rain rates is a preferred response to an increase in aerosol concentration.”**

# Some take-home messages

- Aerosols provide the nuclei for all cloud hydrometeor formation, and modulations thereof have significant impacts on radiation and the hydrological cycle.
- The theory for droplet formation is well established and can be well parameterized for models; instruments and methods constrain the aerosol-drop link very well.
- Ice formation is much less understood - but progress is rapid on IN parameterization development and ice production relationships that consider both homogeneous and heterogeneous freezing concurrently.
- Vertical velocity is far less constrained or tested (compared to aerosols) in global models. There is a strong need for that quantity.
- Description of mixed-phase clouds in large-scale models is still at its infancy - but crucial for precipitation/climate. Next few years promises a lot of progress.