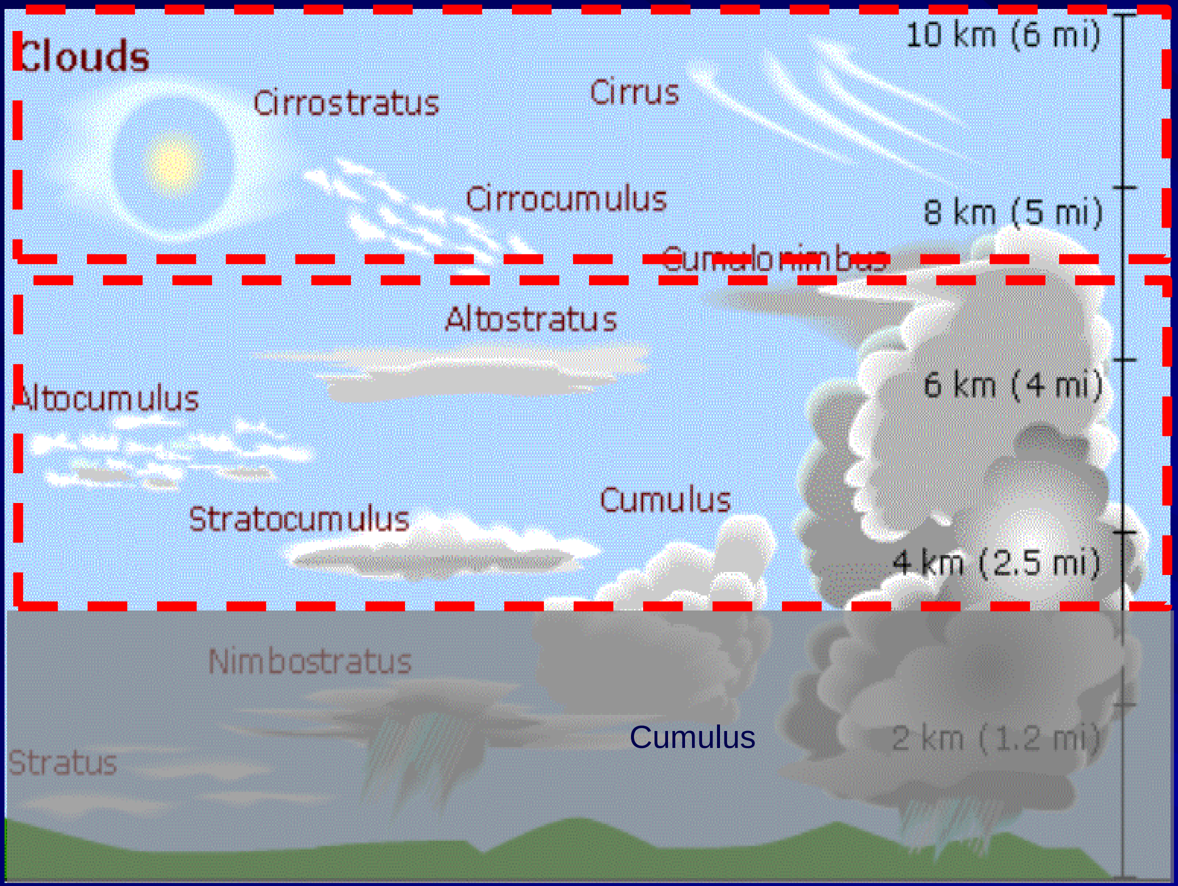
EPFL

Lecture 3 – Cold Cloud Processes

ENV 320

Focus on "Slush" and Ice clouds now

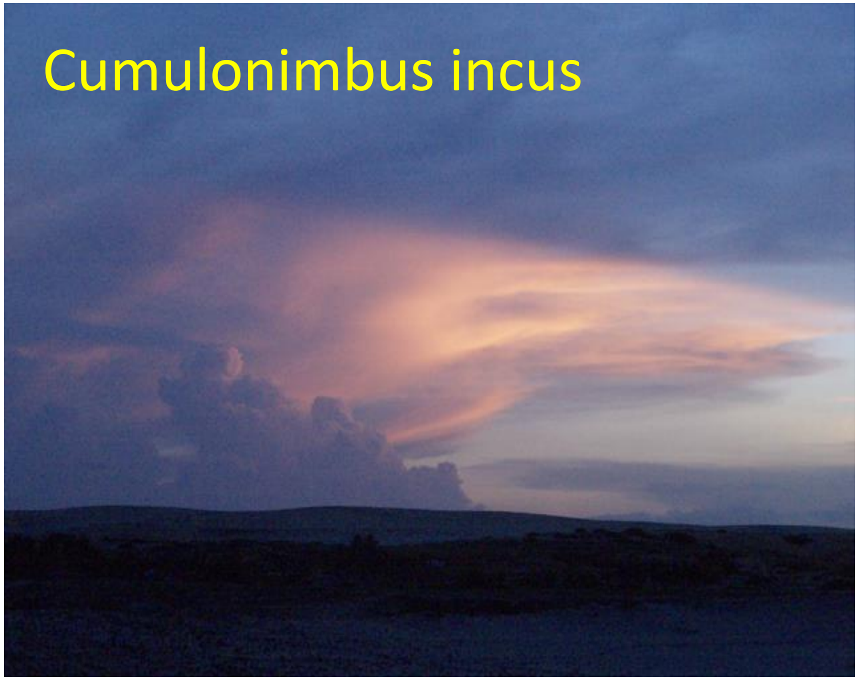


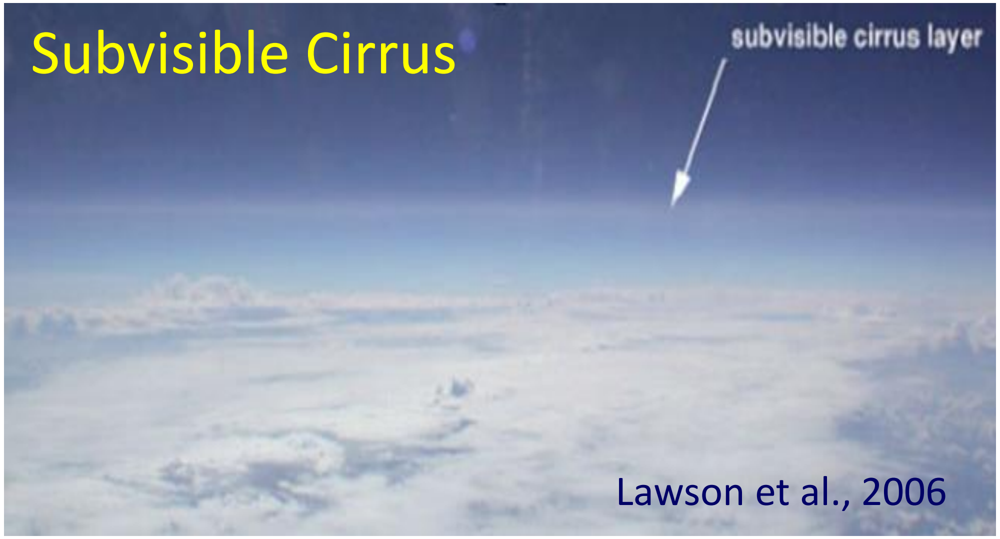
- **Ice (cold) clouds**: Made of ice crystals at *T* < 235 K.
- **Mixed Phase clouds**: Mixture of liquid droplets and ice for *T* between 235 and 273K
- **Liquid (warm) clouds**: Made of liquid droplets at *T* >273 K

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

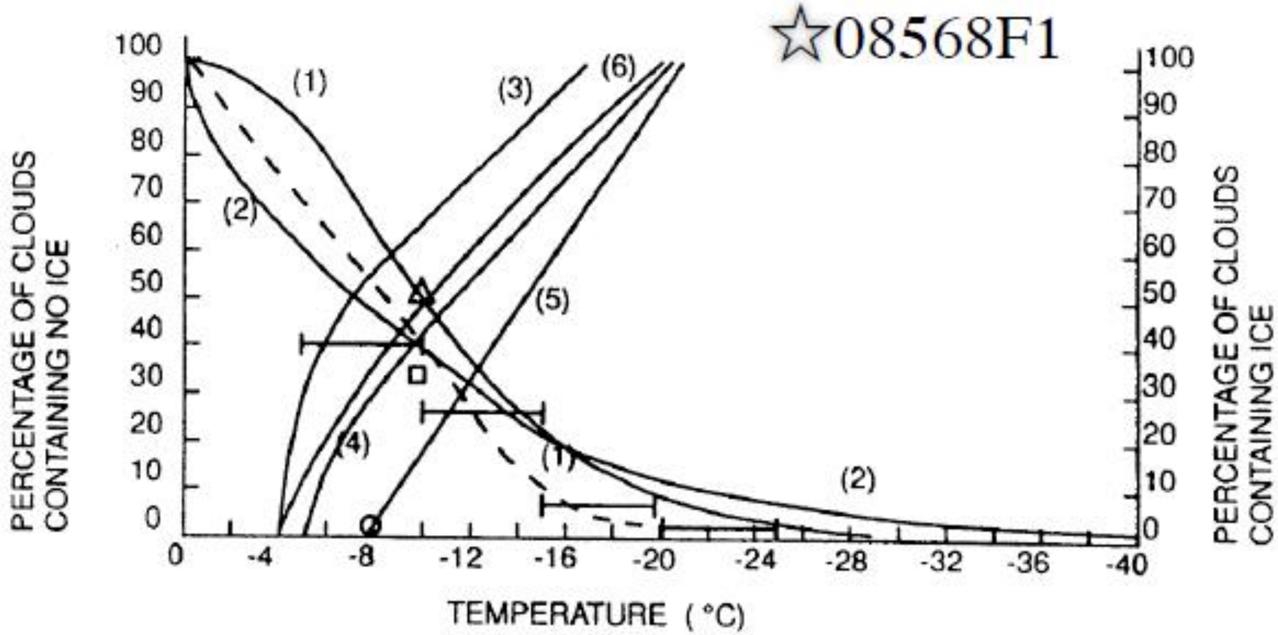
http://www.ucar.edu/news

Pictures of ice clouds





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

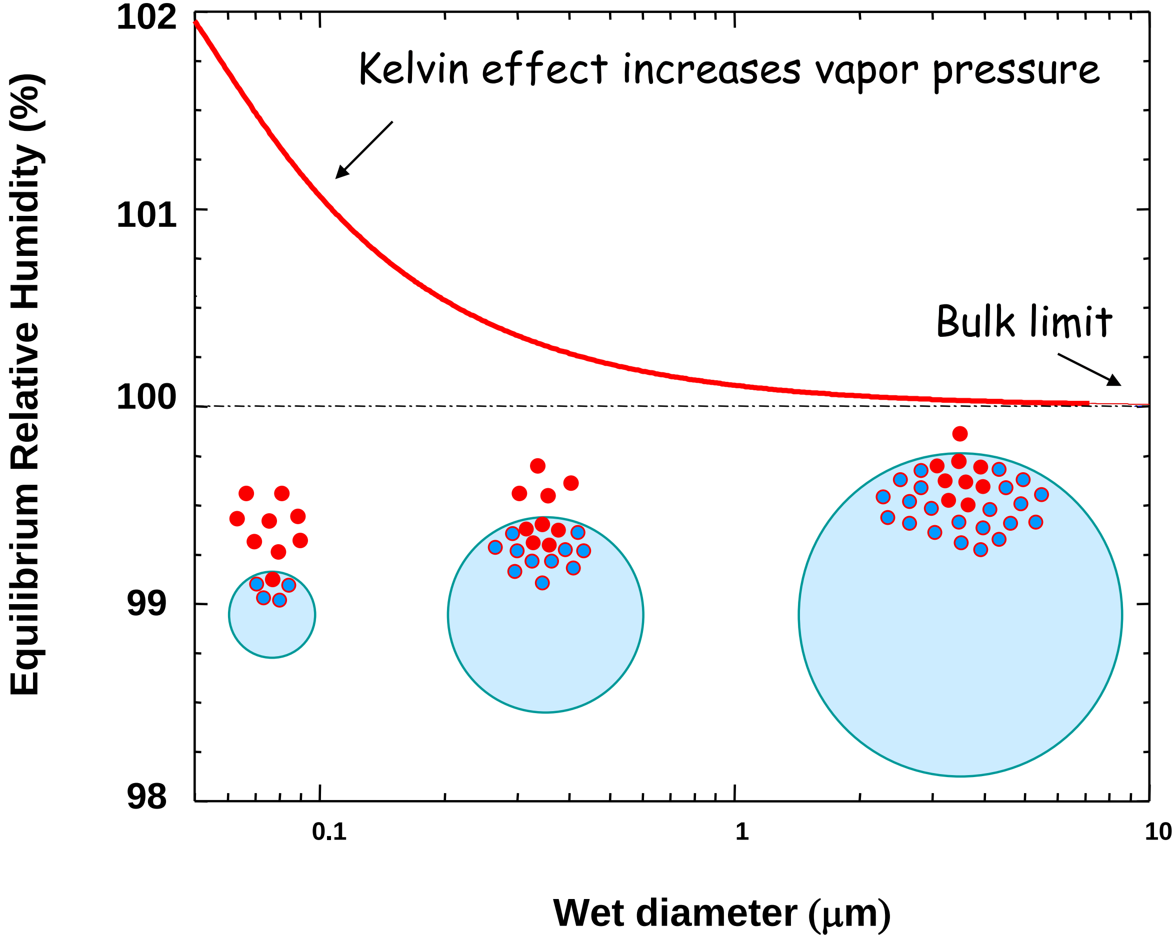
Surprise! It takes *a lot* of cooling to freeze

Understanding why it takes a lot of cooling to freeze

Impact of curvature:

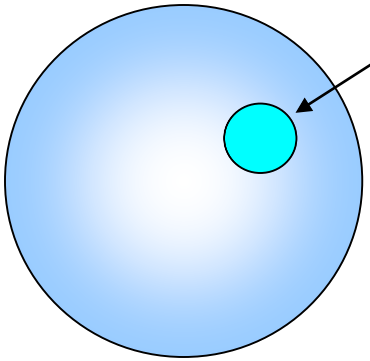
$$P = P^{*} \exp\left(\frac{2\sigma v_l}{RTR_p}\right)$$

Kelvin Equation

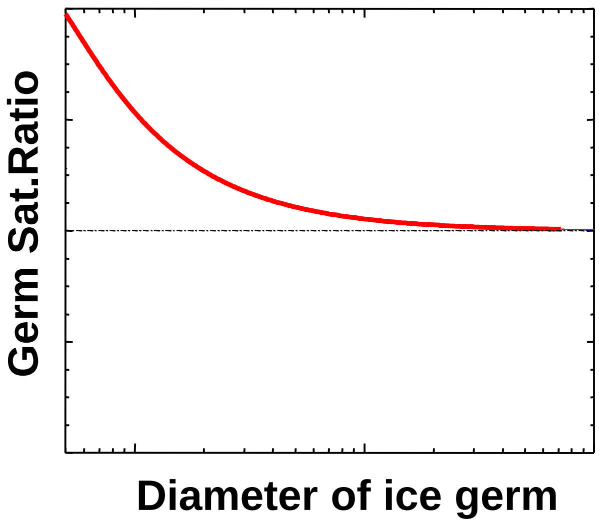


Curvature effect: the basis of nucleation

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.



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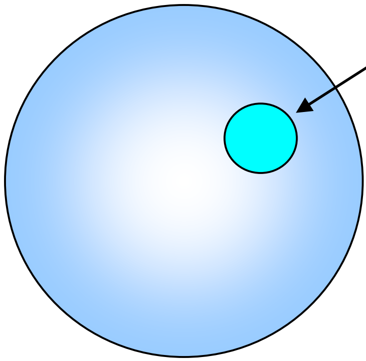

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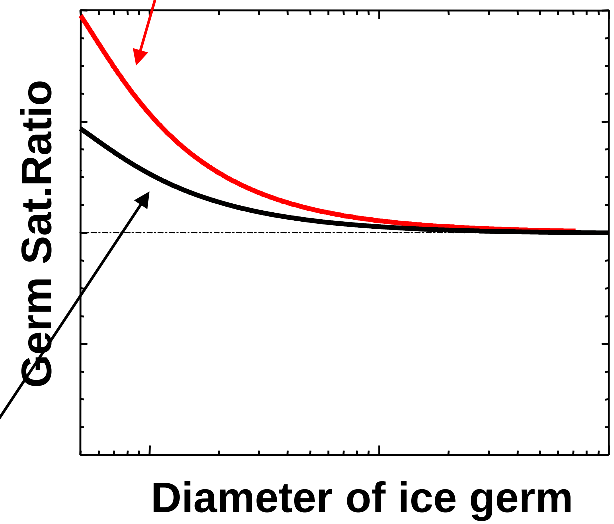
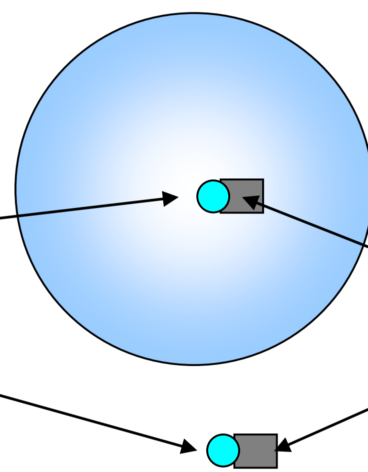




Ice Germs: generated on insoluble particles inside of superscooled droplets or suspended in air

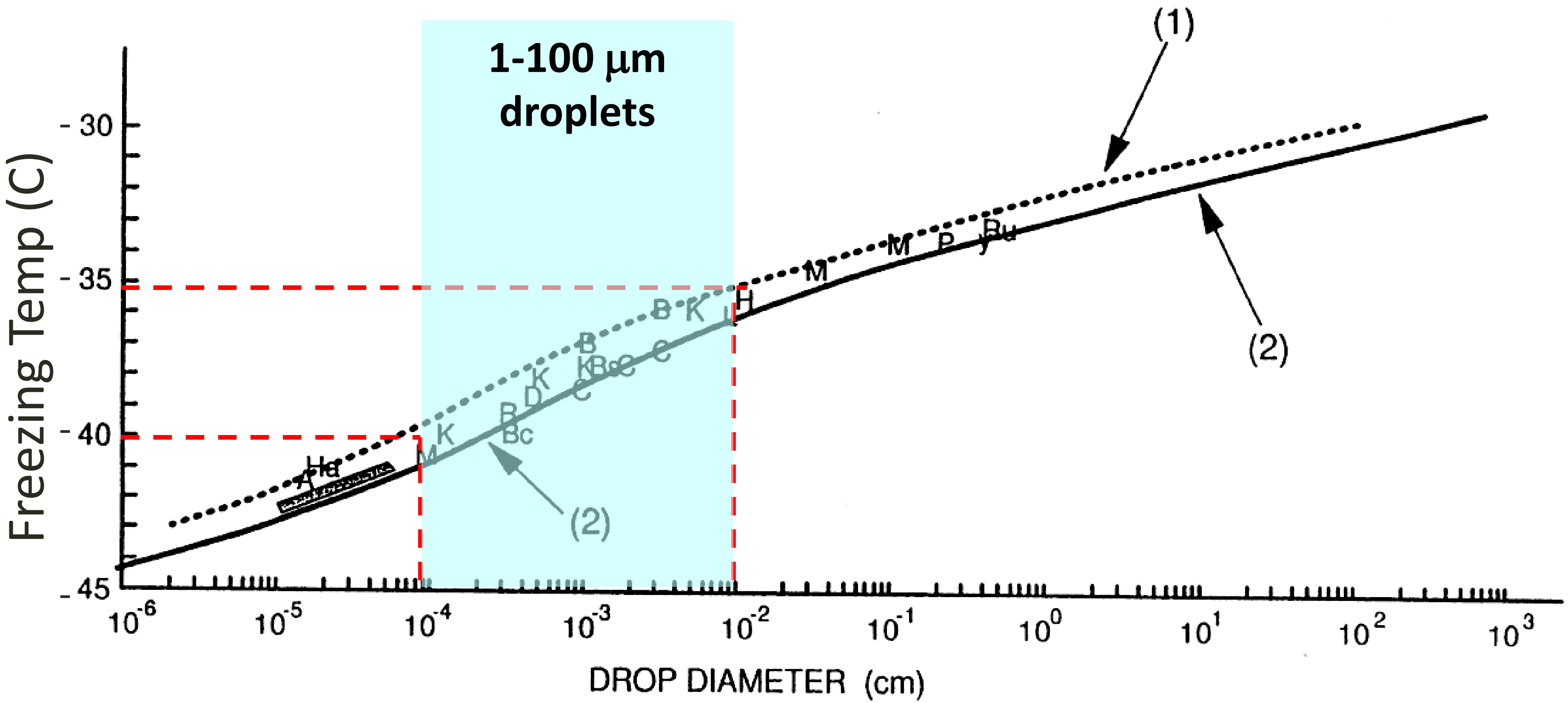
Insoluble particle

(why do insolubles help?)

So .. How does water freeze in the end?

- Equilibrium thermodynamics of the phases of water does not explain the initiation of the ice phase
- The formation of the ice phase from the liquid phase does not begin in a continuous process
- It takes place spontaneously as a result of temperature and density fluctuations in the original phase when a critical supersaturation of the original phase is reached
- This spontaneous process is called **nucleation**

Maximum supercooling of extrapure water

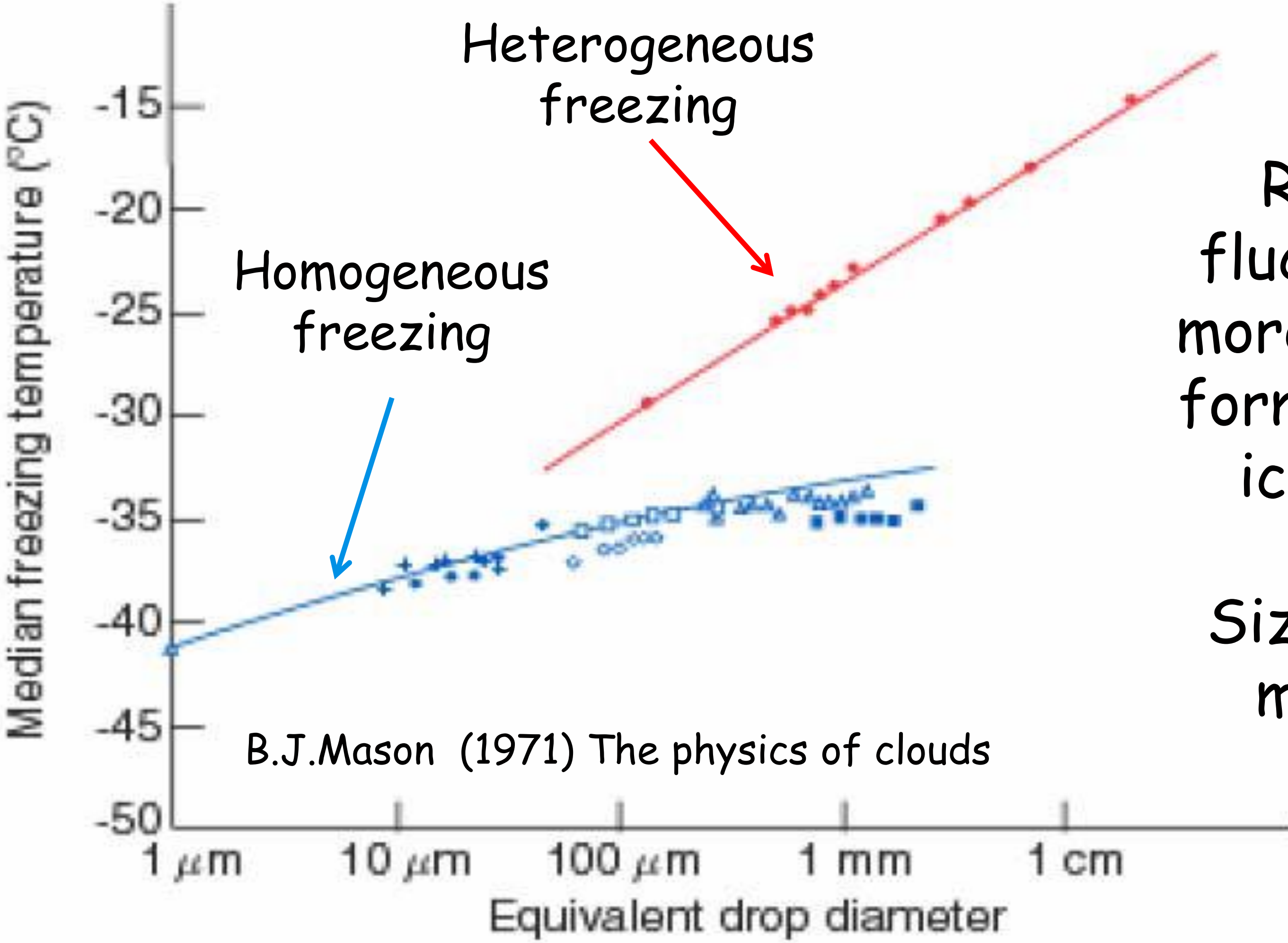


Result: Pure water cloud droplets in the atmosphere effectively start freezing at around -35^{o}C

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

Median freezing T of water samples

Not really…. Heterogeneous Freezing Greatly Facilitates Ice Formation

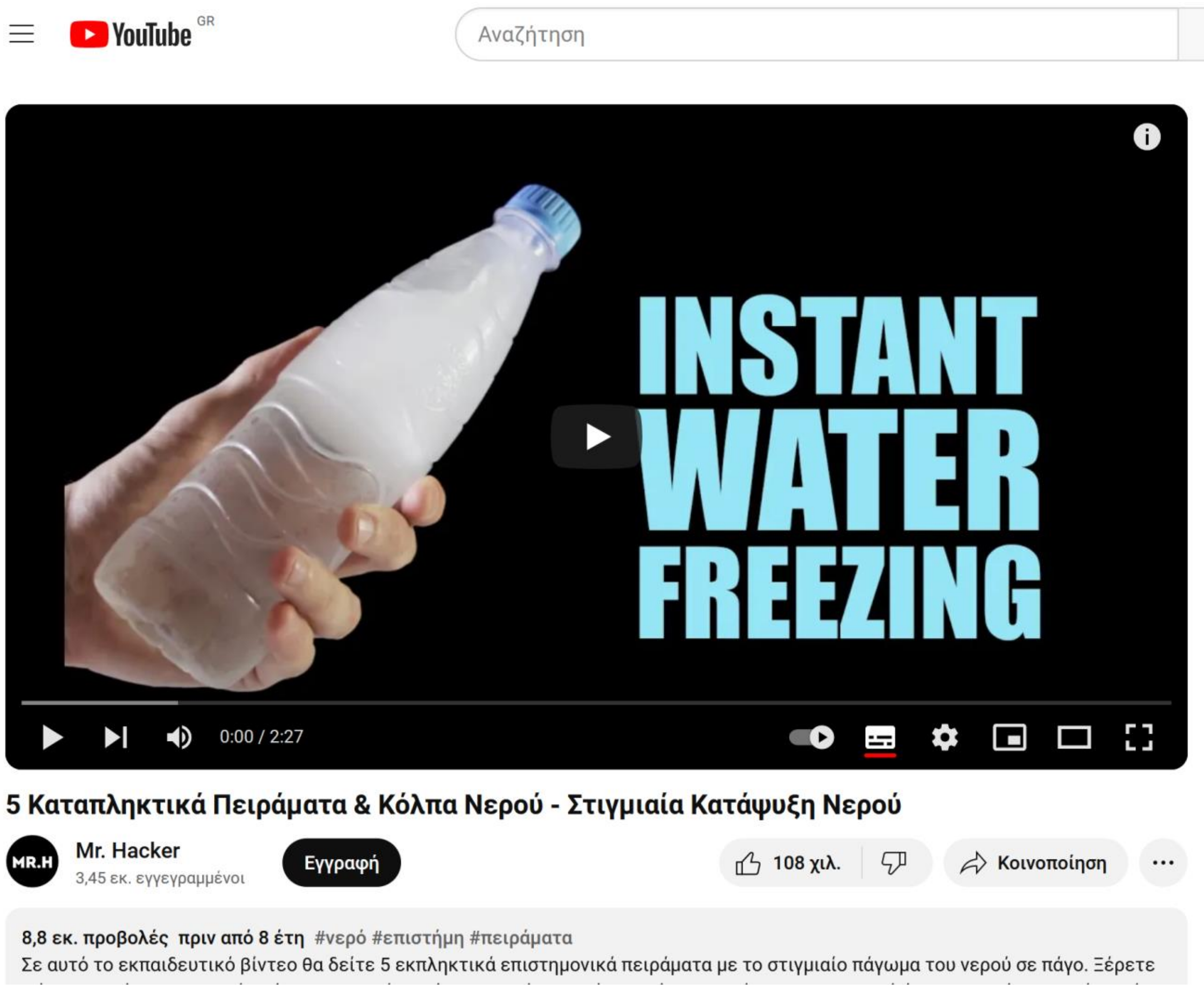


B.J.Mason (1971) The physics of clouds

Random fluctuations more likely to form a stable ice germ:

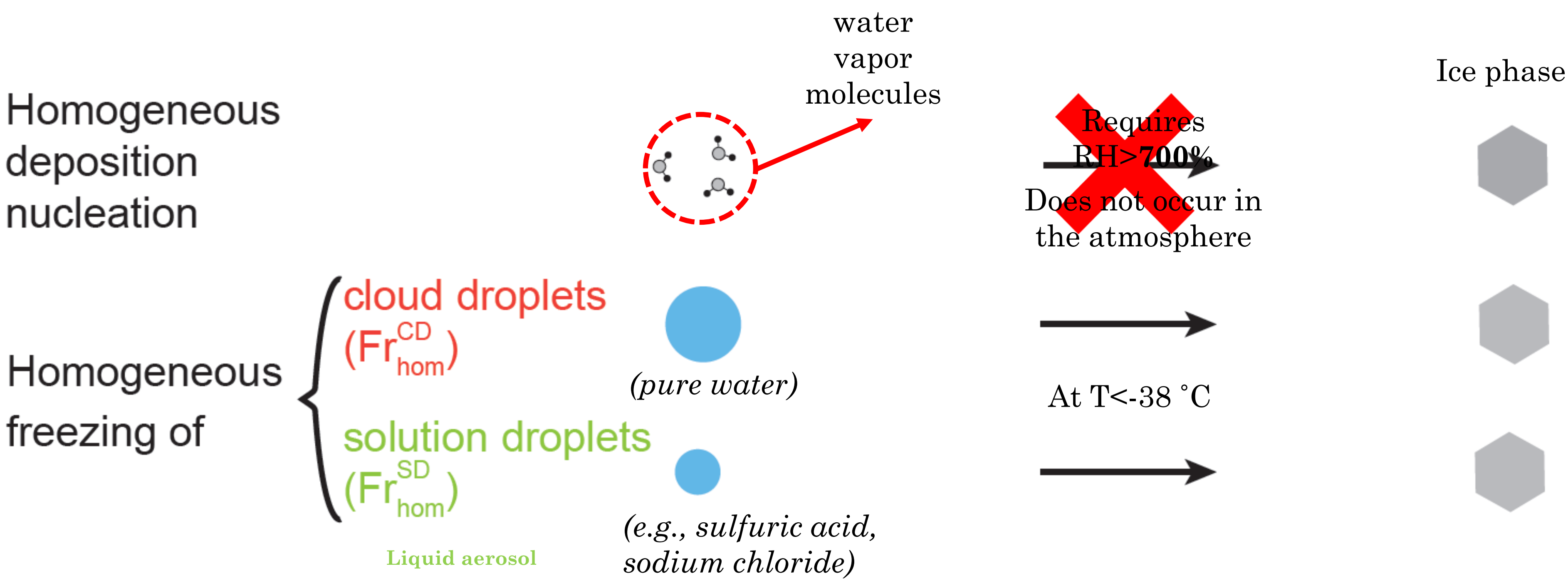
Size always matters

The need to supercool and the role of perturbations in ice formation



https://www.youtube.com/watch?v=kEHdyiBMgAg

Homogeneous nucleation modes

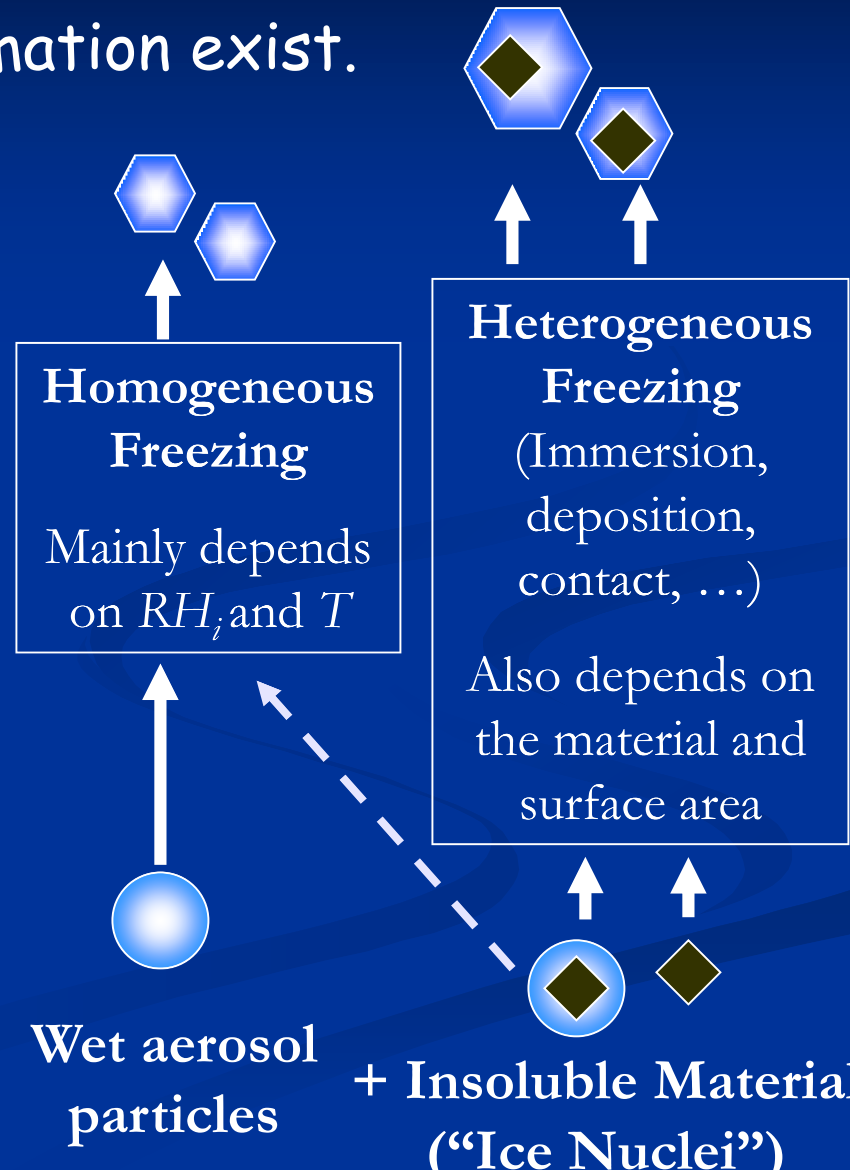


Unlike the formation of liquid droplets, which does not occur homogeneously in the atmosphere, the ***homogeneous nucleation of ice crystals from the liquid phase (homogeneous freezing) occurs in the atmosphere*** *at temperatures below ∽ -38 °C*

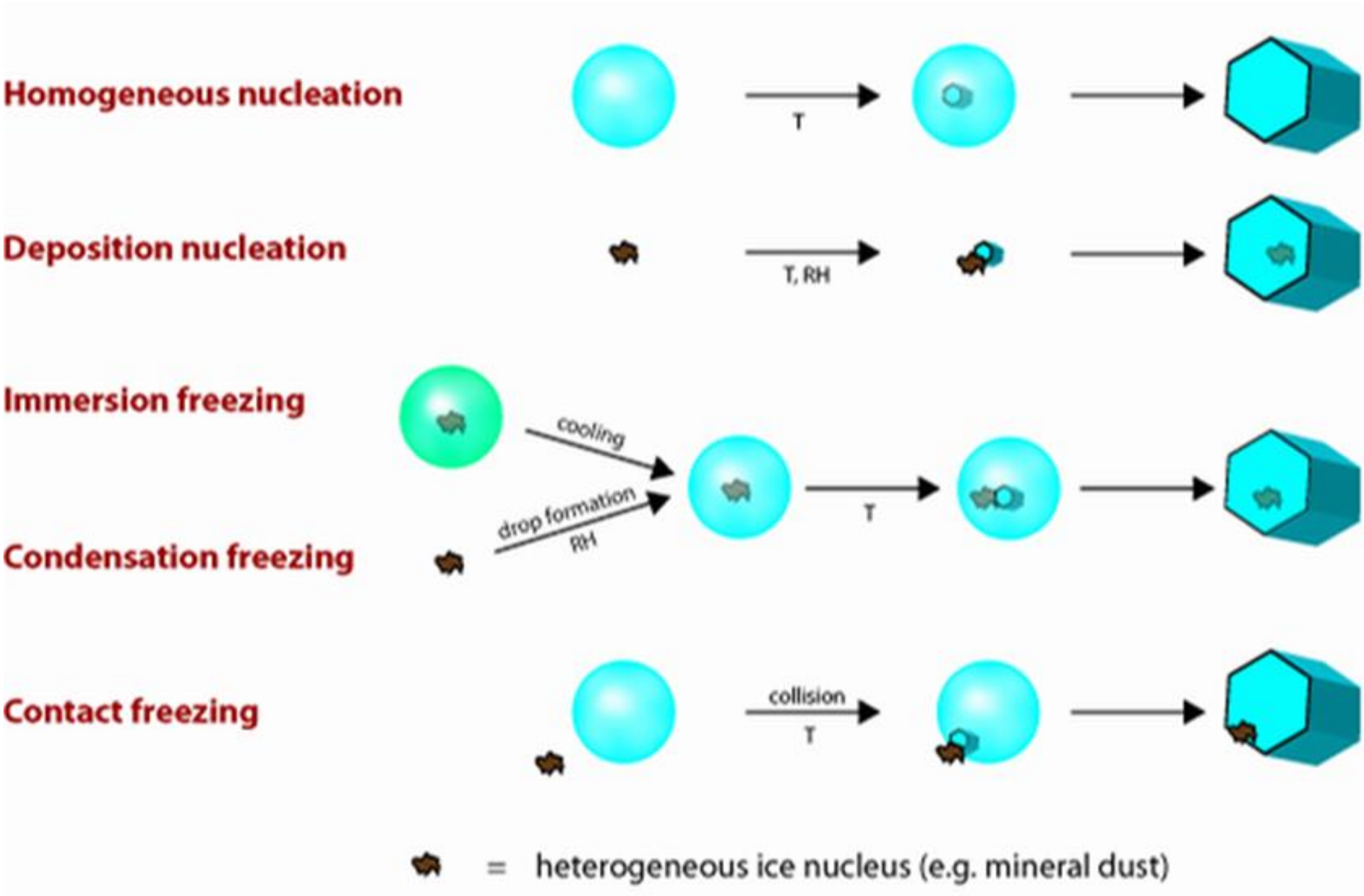
How do (ice water) clouds form?

Ice crystals also form on preexisting particles.

Multiple mechanisms for *ice formation exist.*

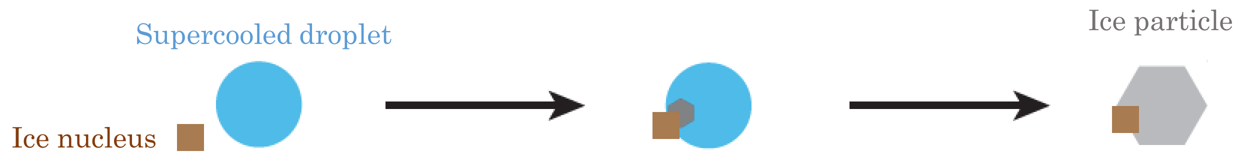


Heterogeneous ice nuclei: freezing modes



Four main pathways for heterogeneous ice nucleation:

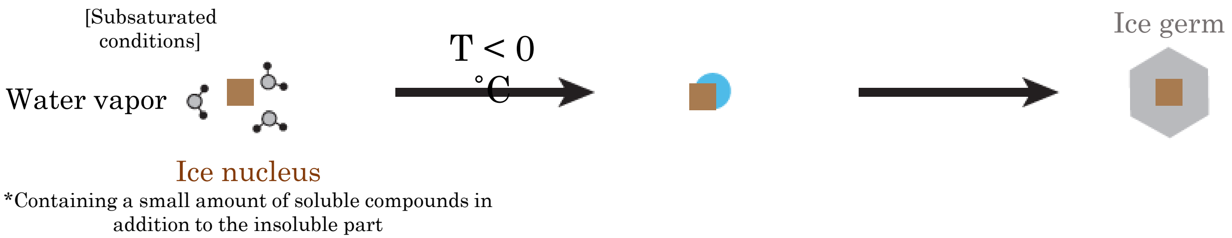
Contact freezing (Fr^{CD}_{cnt})



- ✓ Freezing starts when suitable particle (contact nucleus) comes into contact with super-cooled droplet
- ✓ Dependent on:
  1. temperature
  2. Drop volume
  3. Type of ice nucleus
- ✓ Contact freezing is limited by the collision rate, which depends on:
  - collision efficiency between drops and aerosol particles
  - the number density of aerosol particles available for collisions

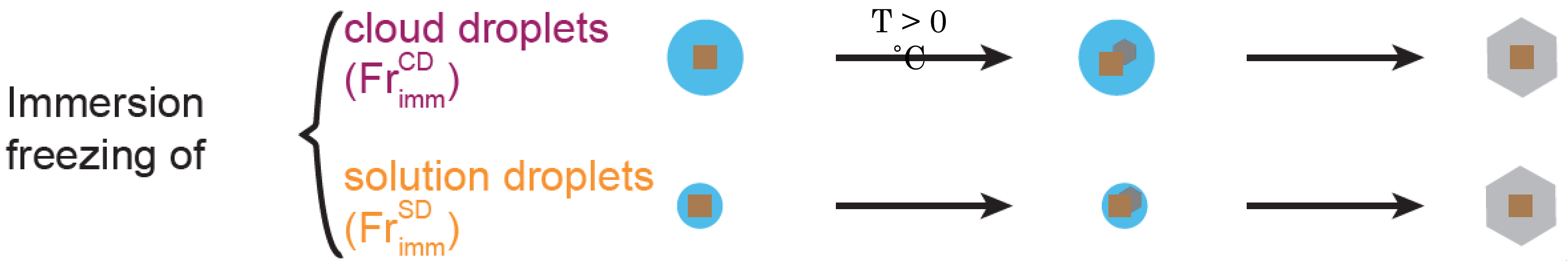
Four main pathways for heterogeneous ice nucleation:

Condensation freezing (Fr^{CD}_{cnd})



- ✓ Condensation of water vapor on ice nuclei (condensation freezing nucleus), followed by freezing of the drop
- ✓ **Requirements**:
  - dry ice nucleus which is suited as condensation nucleus
  - super-saturation with respect to water
  - critical temperature to initiate ice nucleation

Four main pathways for heterogeneous ice nucleation:



✓ A suitable particle (immersion nucleus) is already immersed in the droplet at higher temperatures and causes freezing upon cooling

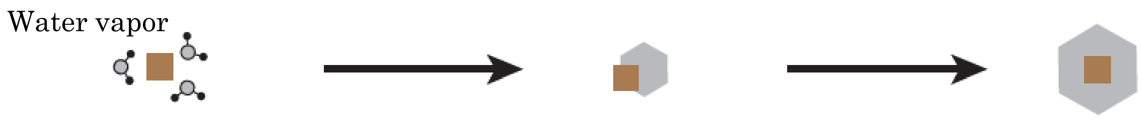
✓ Immersion freezing is the most the dominant nucleation mode in most mixed-phase clouds (Whale, 2018).

✓ Dependent on:

1. temperature
2. Drop volume
3. Type of ice nuclei

Four main pathways for heterogeneous ice nucleation:

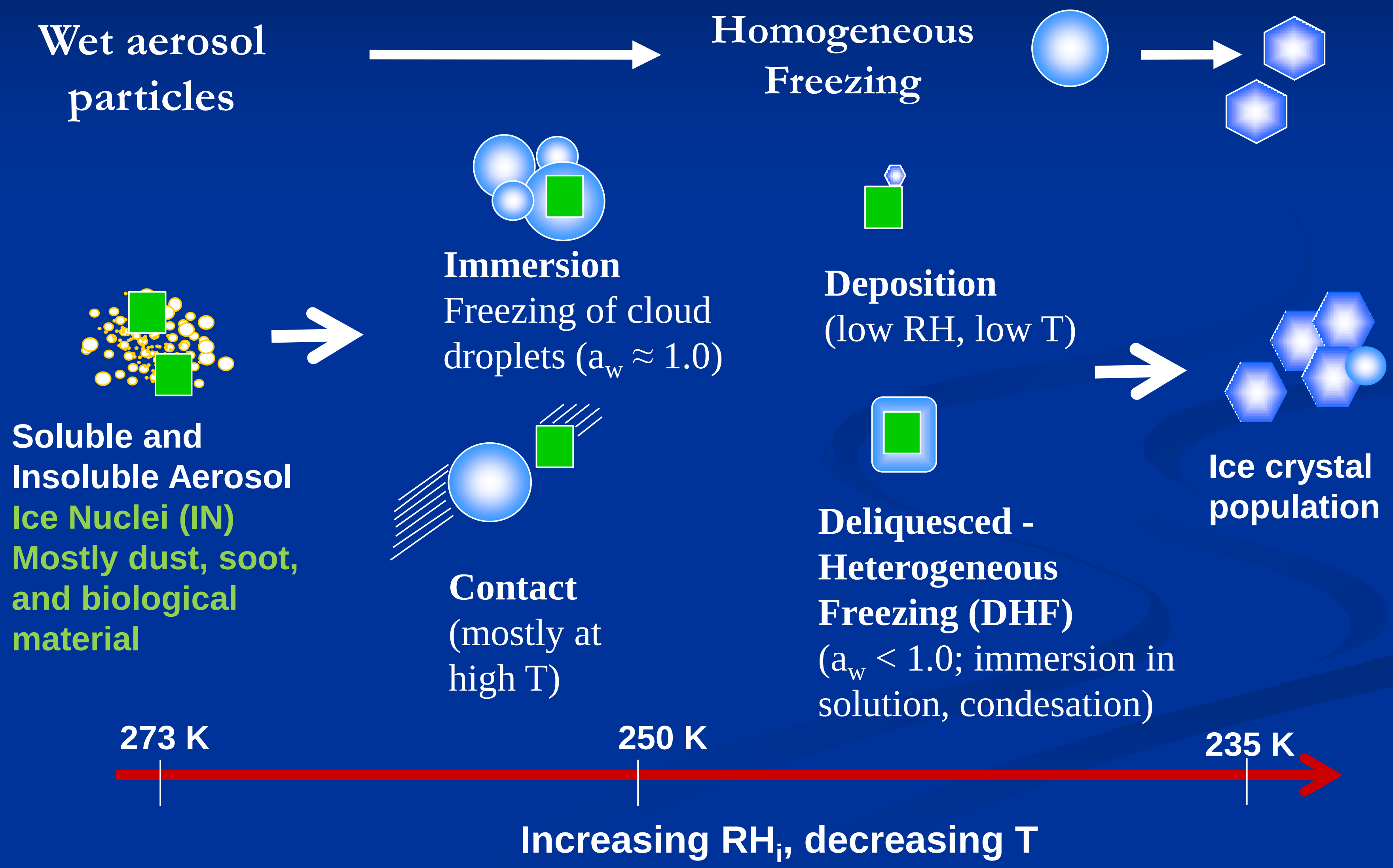
Deposition nucleation



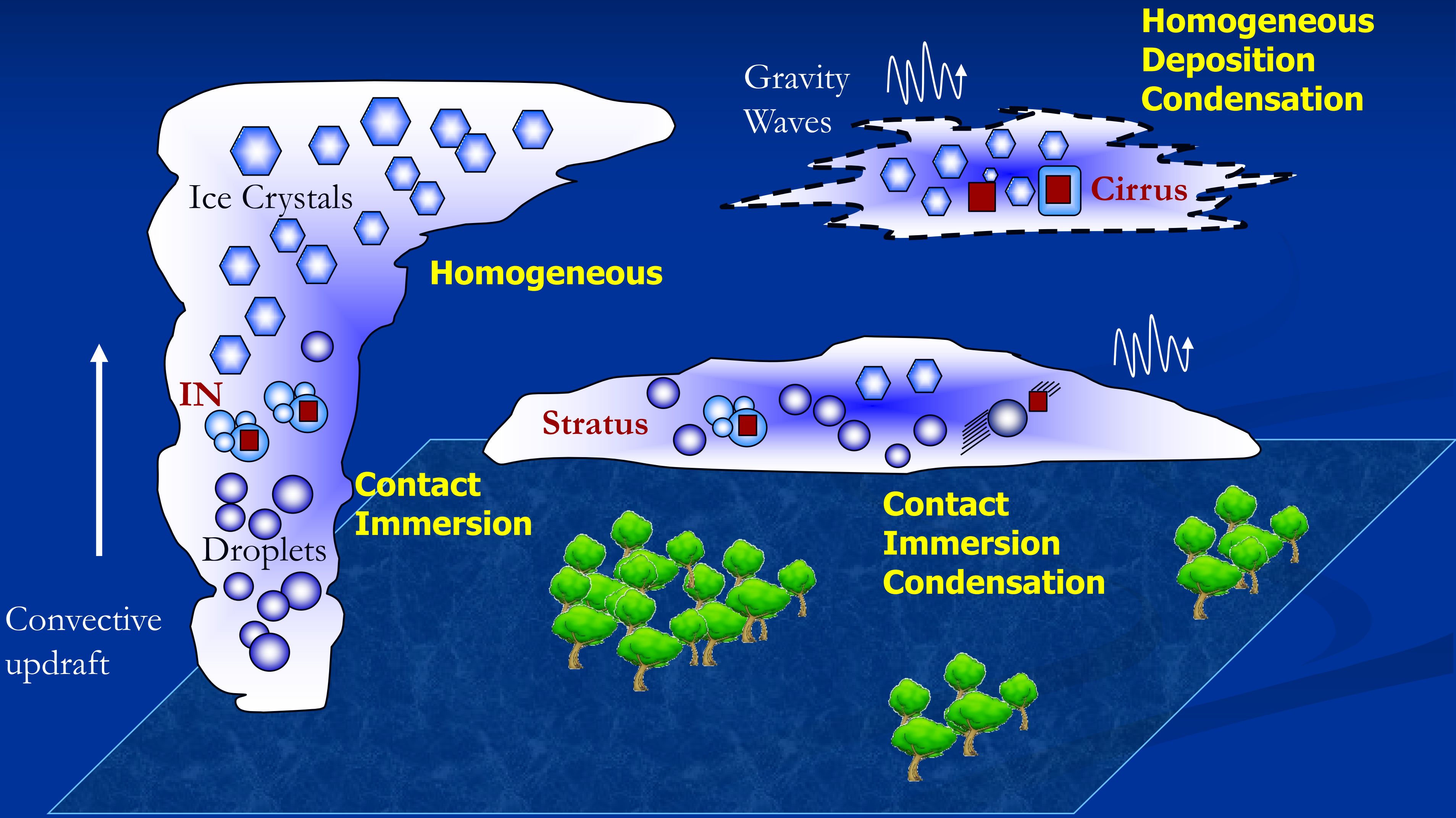
- ✓ Some particles (deposition nuclei) serve as centers where ice forms directly from the vapor phase
- ✓ **Requirements:**
  - dry ice nucleus
  - super-saturation with respect to ice
  - critical temperature to initiate ice nucleation
- ✓ **Result:** ice crystals in various habits, depending on:
  1. temperature
  2. supersaturation

Heterogeneous ice nuclei: freezing modes

Multiple mechanisms for ice formation can be active.



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

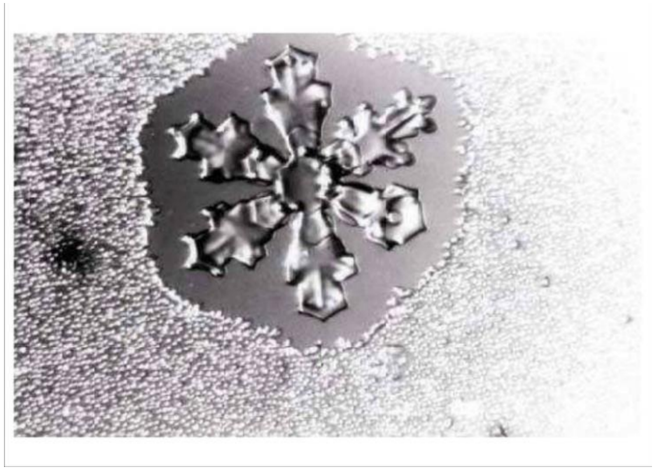


IN vs CCN: implications for clouds

IN are far less abundant than CCN. (1 in a million!)
CCN: 100-1000 cm^{-3} vs IN: 0.001-0.01 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

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/Chrystallography: 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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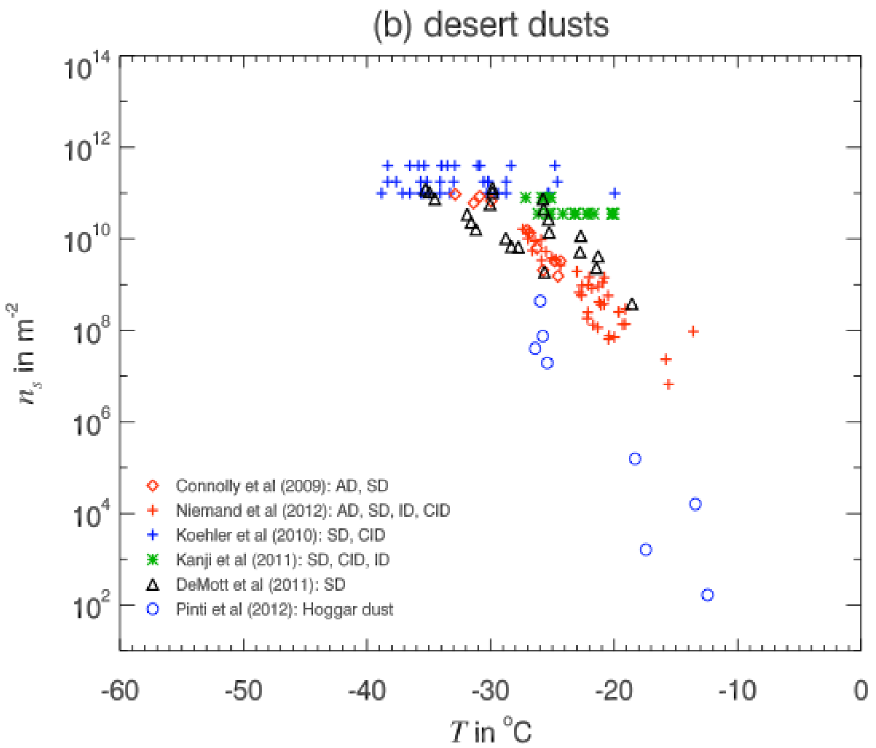
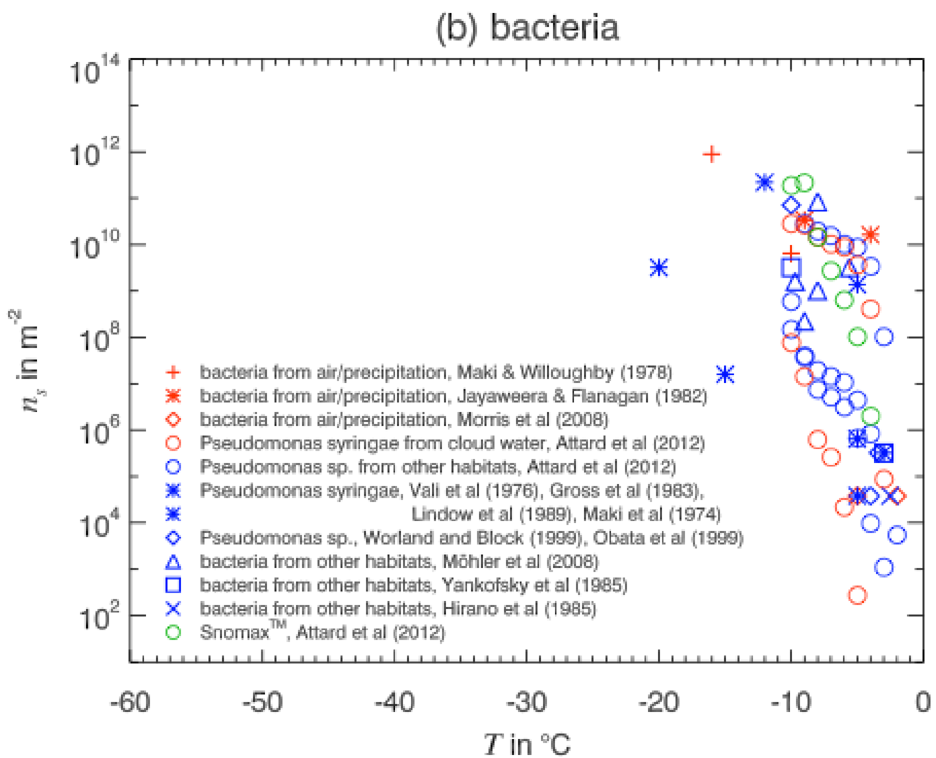
Table 2. Activation Temperatures T_a and Median Freezing Temperatures T_m Determined From Laboratory Experiments[a]

	Immersion Freezing		Contact Freezing	
Particle Type	T_a	T_m	T_a	T_m
Bacteria	−4°C,[b] a = 250 μm	−7°C,[b] a = 250 μm	−3°C[h]	−4.5°C[h]
Leaf litter	−5°C,[c] a = 350 μm	−9°C,[c] a = 350 μm	−5°C[c]	−10°C[c]
Pollen	−9°C,[d] a = 250 μm	−14°C,[d] a = 250 μm	−5°C[d]	−10°C[d]
Montmorillonite	−12°C,[e] a = 350 μm	−19°C,[e] a = 350 μm, and −24°C,[f] a = 50 μm	−3°C[e]	−8°C[e]
Kaolinite	−14°C,[e] a = 350 μm	−23°C,[e] a = 350 μm, and −32.5°C,[f] a = 50 μm	−5°C[e]	−12°C[e]
Soot	−18°C,[g] a = 350 μm	−28°C,[g] a = 350 μm		−18°C[h] (extrapolated)

[a]Immersion freezing temperatures are for defined drop radii a, and contact freezing temperatures are for arbitrary drop sizes.
[b]*Levin and Yankofsky* [1983].
[c]*Diehl et al.* [2001b].
[d]*Diehl et al.* [2002].
[e]*Pitter and Pruppacher* [1973].
[f]*Hoffer* [1961].
[g]*Diehl and Mitra* [1998].
[h]*Gorbunov et al.* [2001].

Heterogeneous ice nuclei: particle types

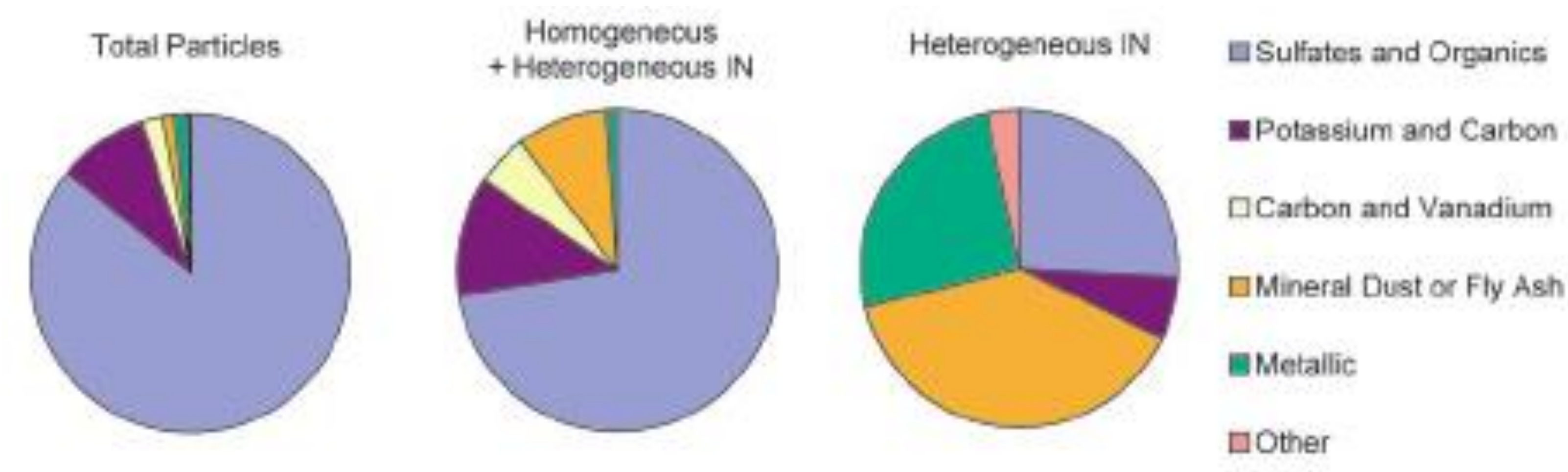




Dust, biological particles, soot, glassy aerosol all can contribute to the population of IN in the atmosphere.

The species contributing to ice formation varies considerably over space and time.

What do evaporated ice crystals from ambient clouds tell us?

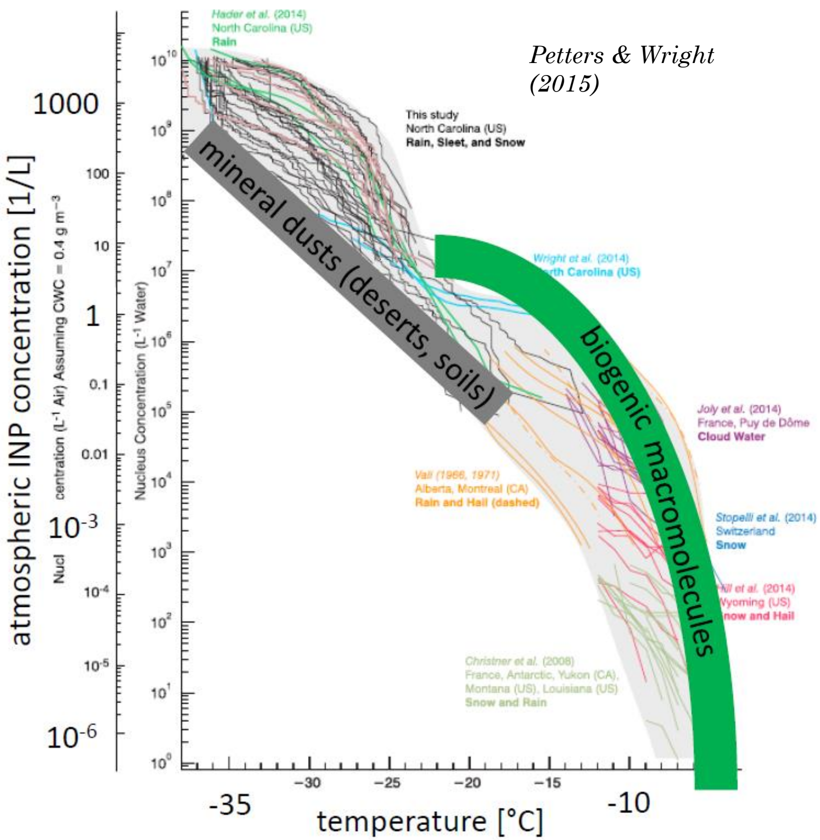


Outside cirrus $\quad$ $S_i > 1.4$ $\quad$ $1.0 \le S_i \le 1.4$

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

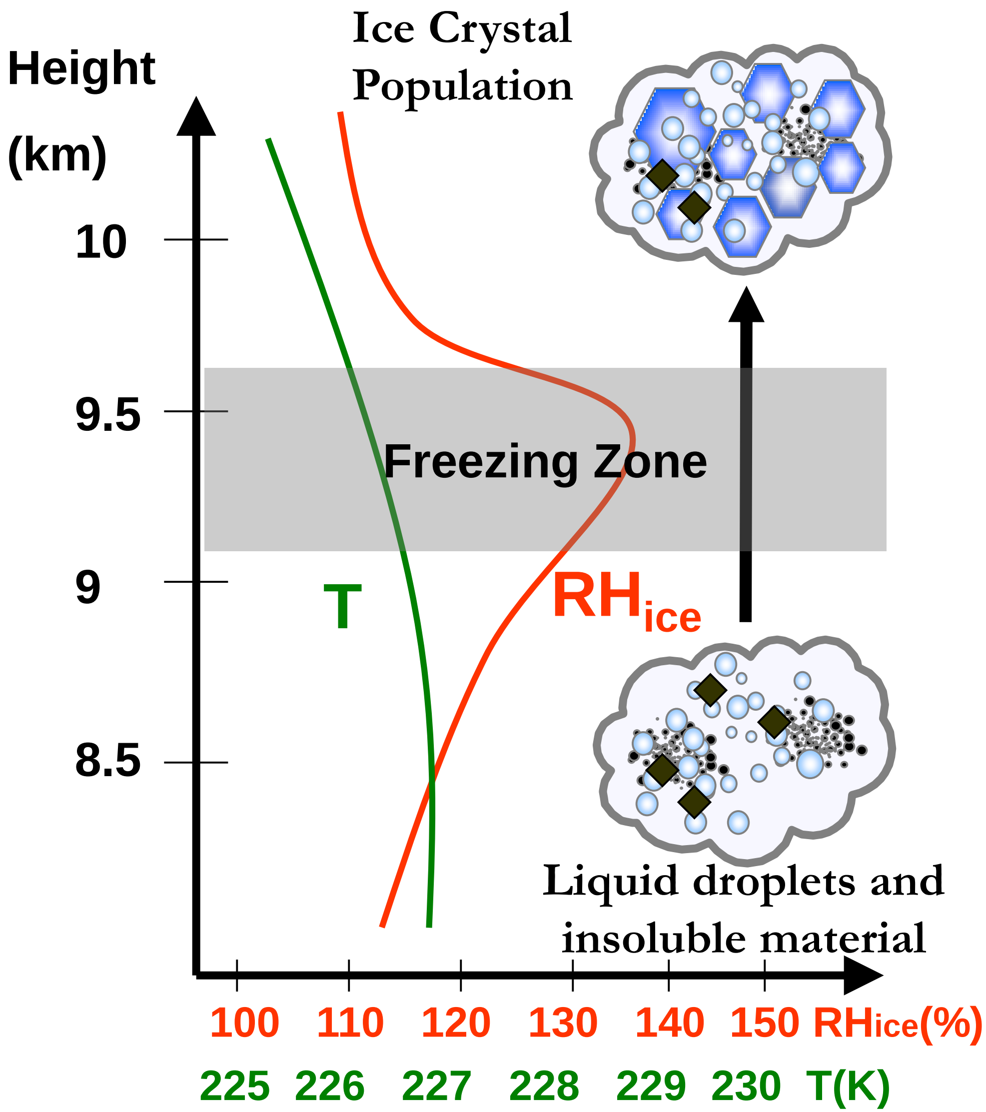
Composition of evaporated crystals from IN very different from the “total aerosol”! – IN are *rare* (1 in a million!)

Observed concentrations of INPs around the globe



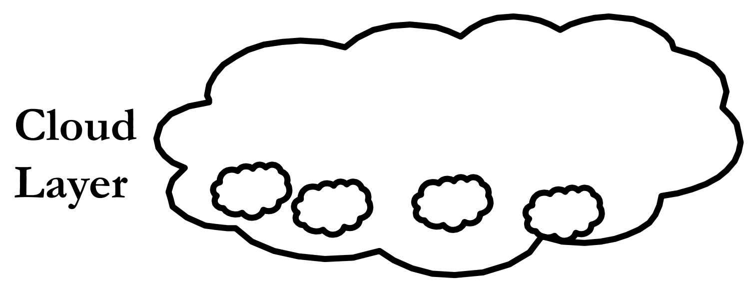
- ✓ Ice nuclei are **rare** compared to CCN (one in **10^5** to **10^6** aerosol particles at T> −38 ˚C)
- ✓ In the atmosphere, mainly two main INP types contribute:
  mineral dust particles and **biological particles** (e.g., pollen or bacteria)
- ✓ Microorganisms have macromolecules causing the ice activity (proteins or polysaccharides)
  -They are very ice active, but VERY rare
  -Heat can destroy bio INP (proteins)
- ✓ Mineral dust particles have ice active sites
  -They are ice active at lower temperatures but are more abundant, however, still rare
  -**K-feldspar** is the most ice active mineral dust

Ice cloud formation: The essence



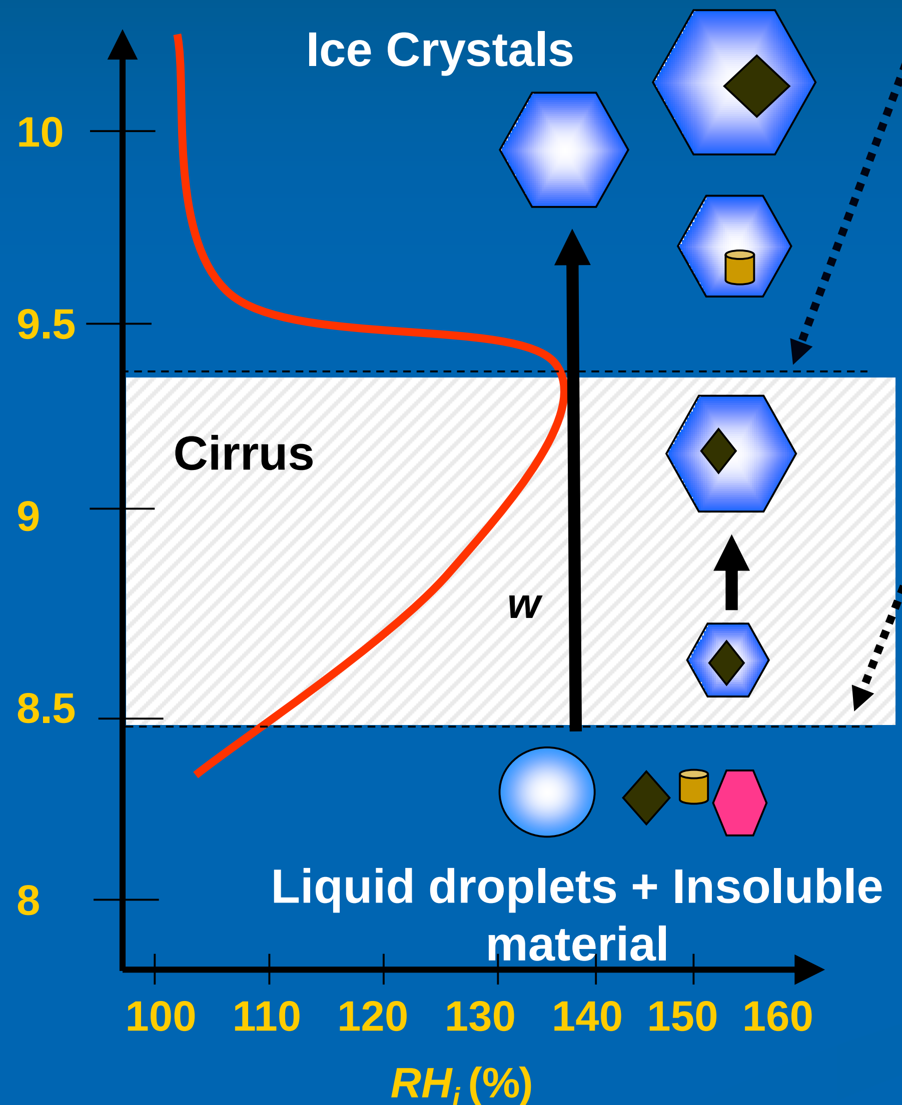
Conceptual steps are:

- 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



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

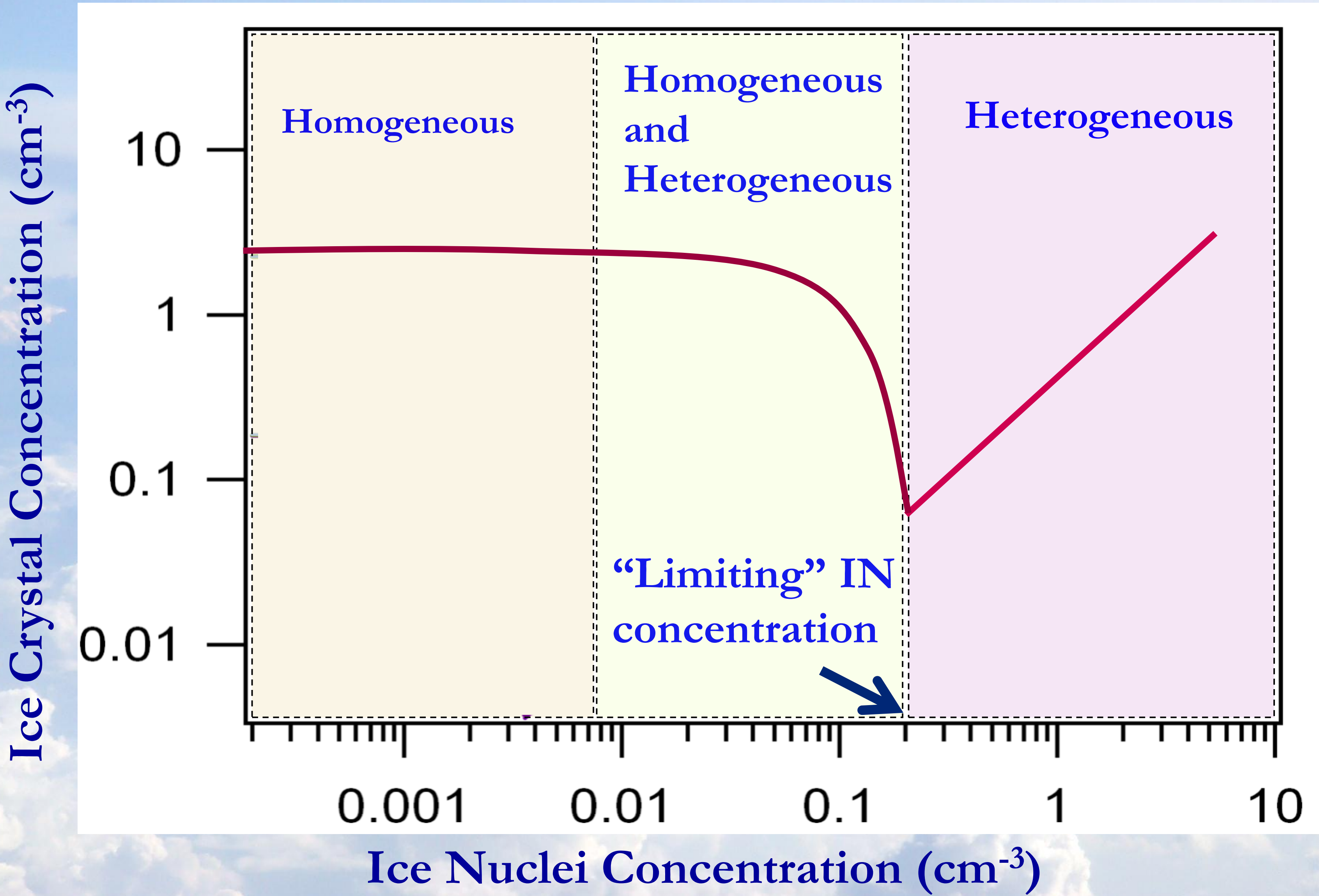
Heterogeneous IN freezing begin forming ice, droplets form

Expansion cooling and supersaturation development

Soluble and insoluble aerosol initial distribution

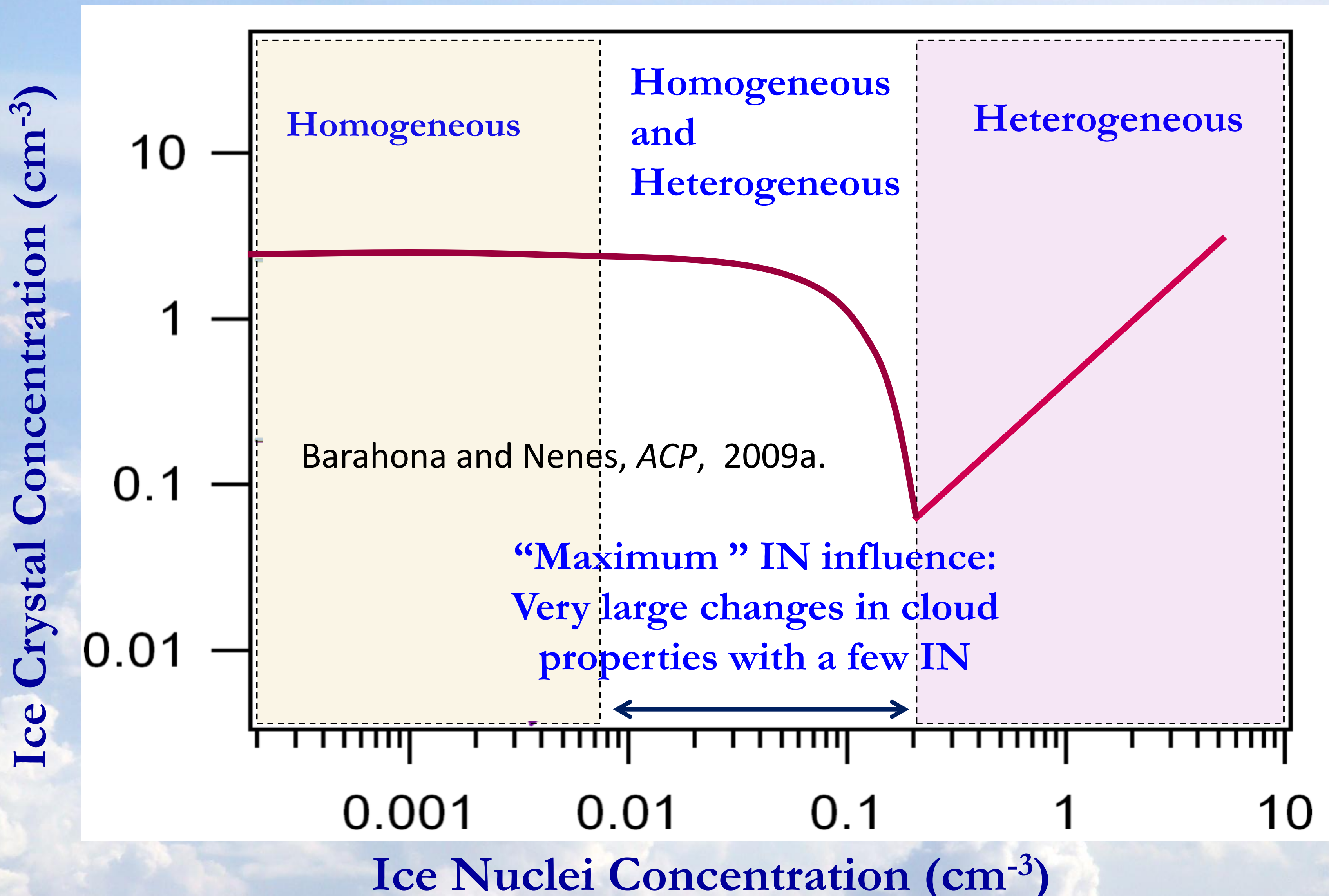
Conceptual steps:

Source of strong nonlinearity:
IN effects on Ice Crystal Concentration



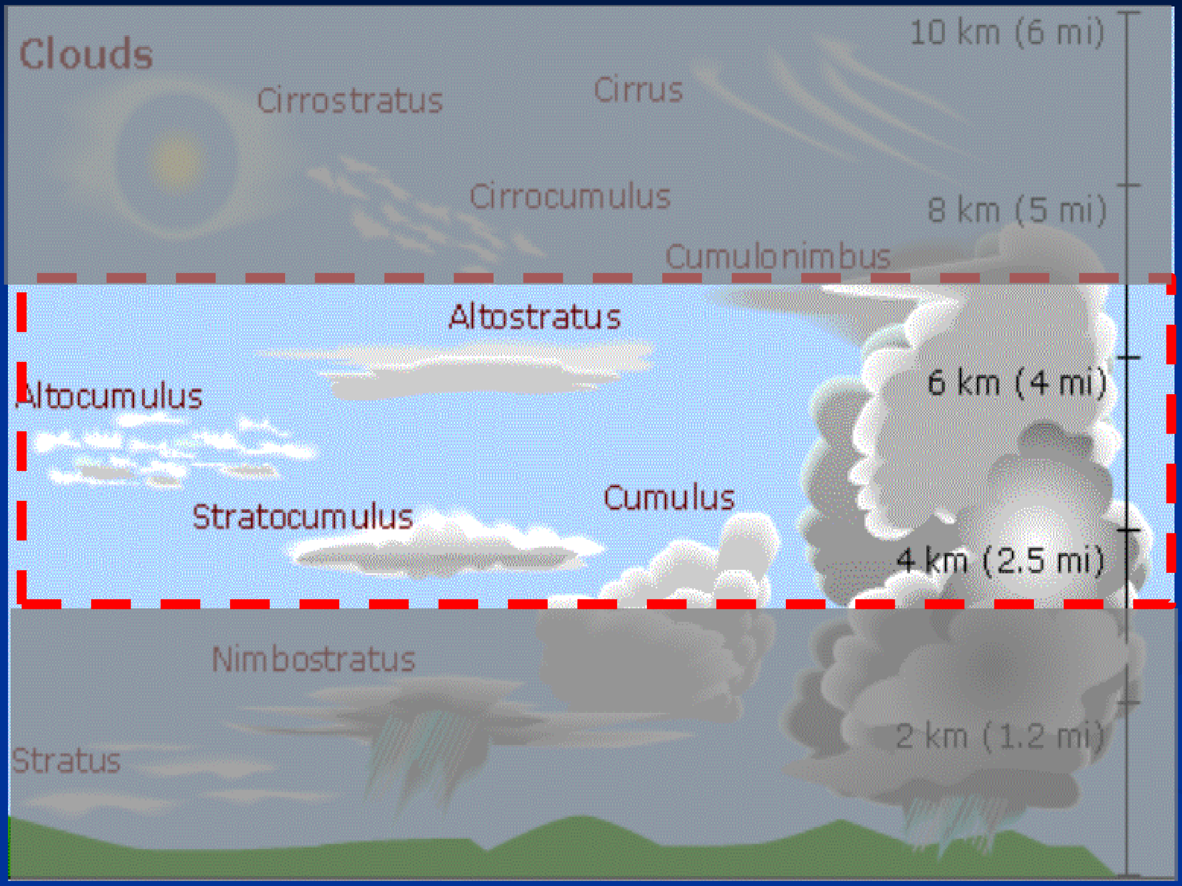
Barahona and Nenes, *ACP*, 2009a.

Source of strong nonlinearity:
IN effects on Ice Crystal Concentration



Thinning effect of moderate INPs on cirrus is the basis of one "geoengineering" approach

Mixed phase clouds: "final frontier"

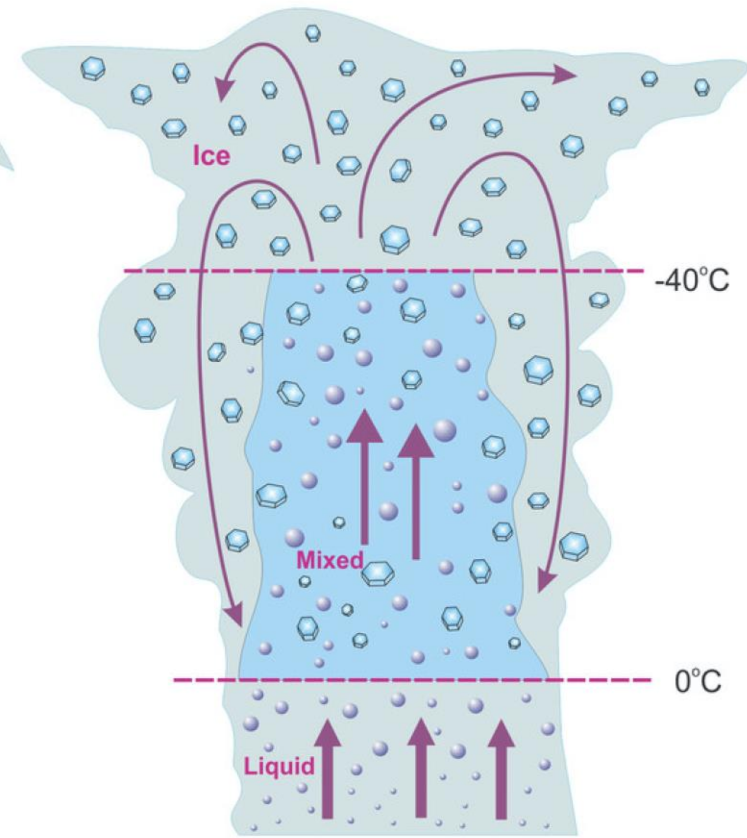


- **Ice (cold) clouds**: Ice crystals, T<235 K. Warm climate
- **Mixed Phase clouds**: Liquid droplets & ice, 235K < T < 273K Warm/cool climate
- **Liquid (warm) clouds**: Liquid droplets T >273 K Cool climate

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

http://www.ucar.edu/news

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

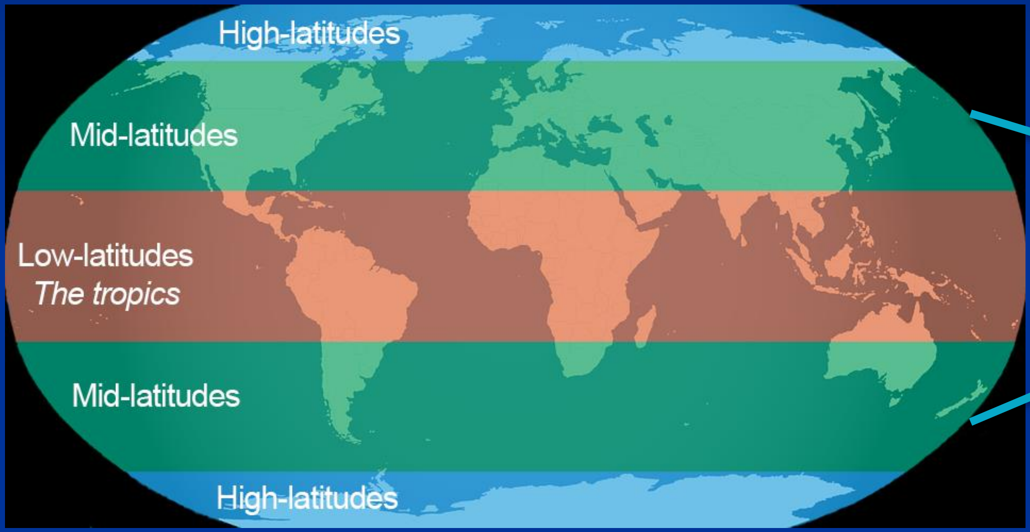


Precipitation at mid- and high-latitudes mostly generated from the mixed- and ice- cloud phase

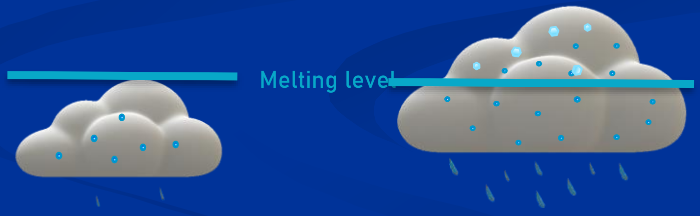
Mulmenstadt et al . 2015

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

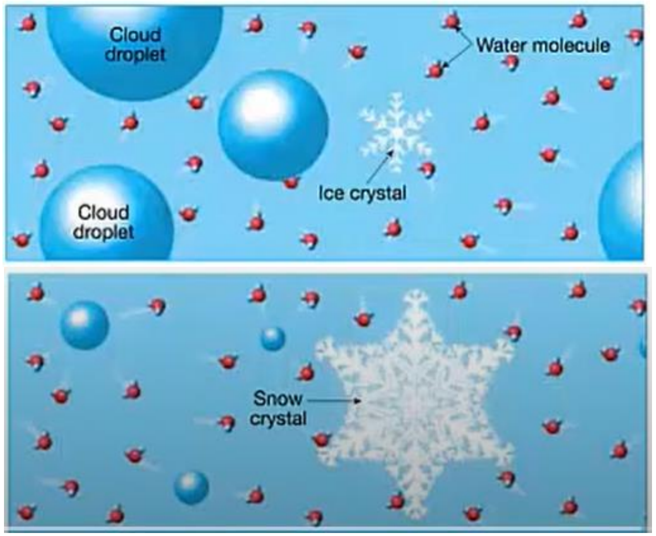


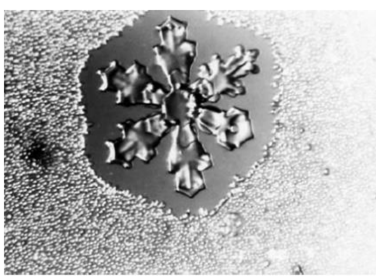
Field and Heymsfield, 2015
Mülmenstädt et al. 2015

"…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



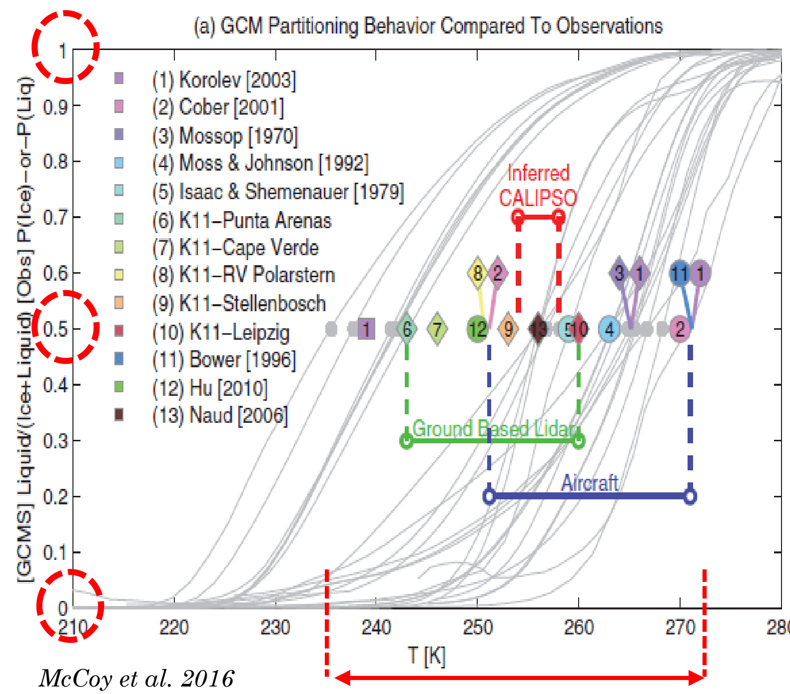


Wegener-Bergeron-Findeisen process (WBF)

Pure liquid cloud

50% liquid 50% ice

Pure ice cloud



McCoy et al. 2016

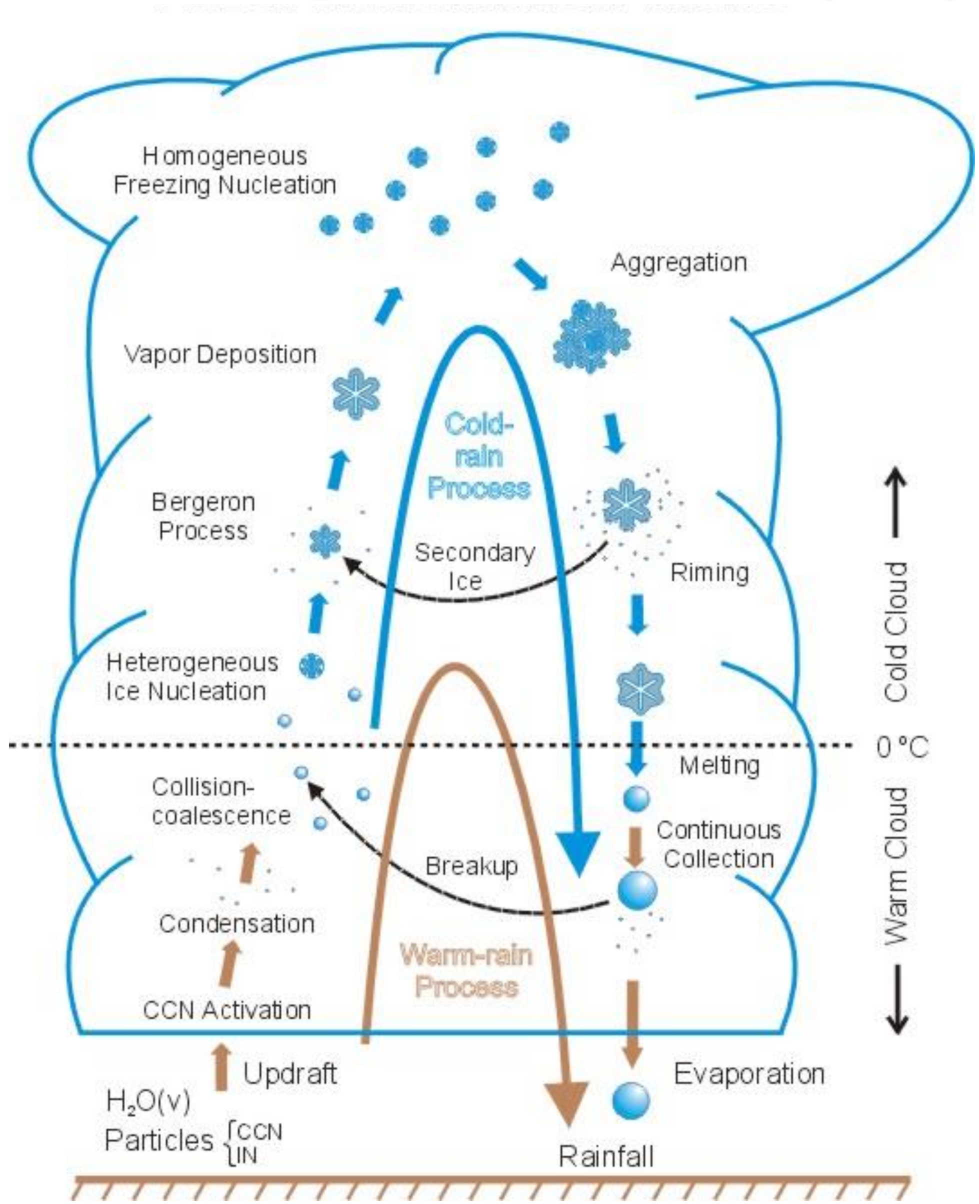
mixed-phase temperature range

> *"One cannot model what one does not understand"*

Ice production/growth interplay in clouds

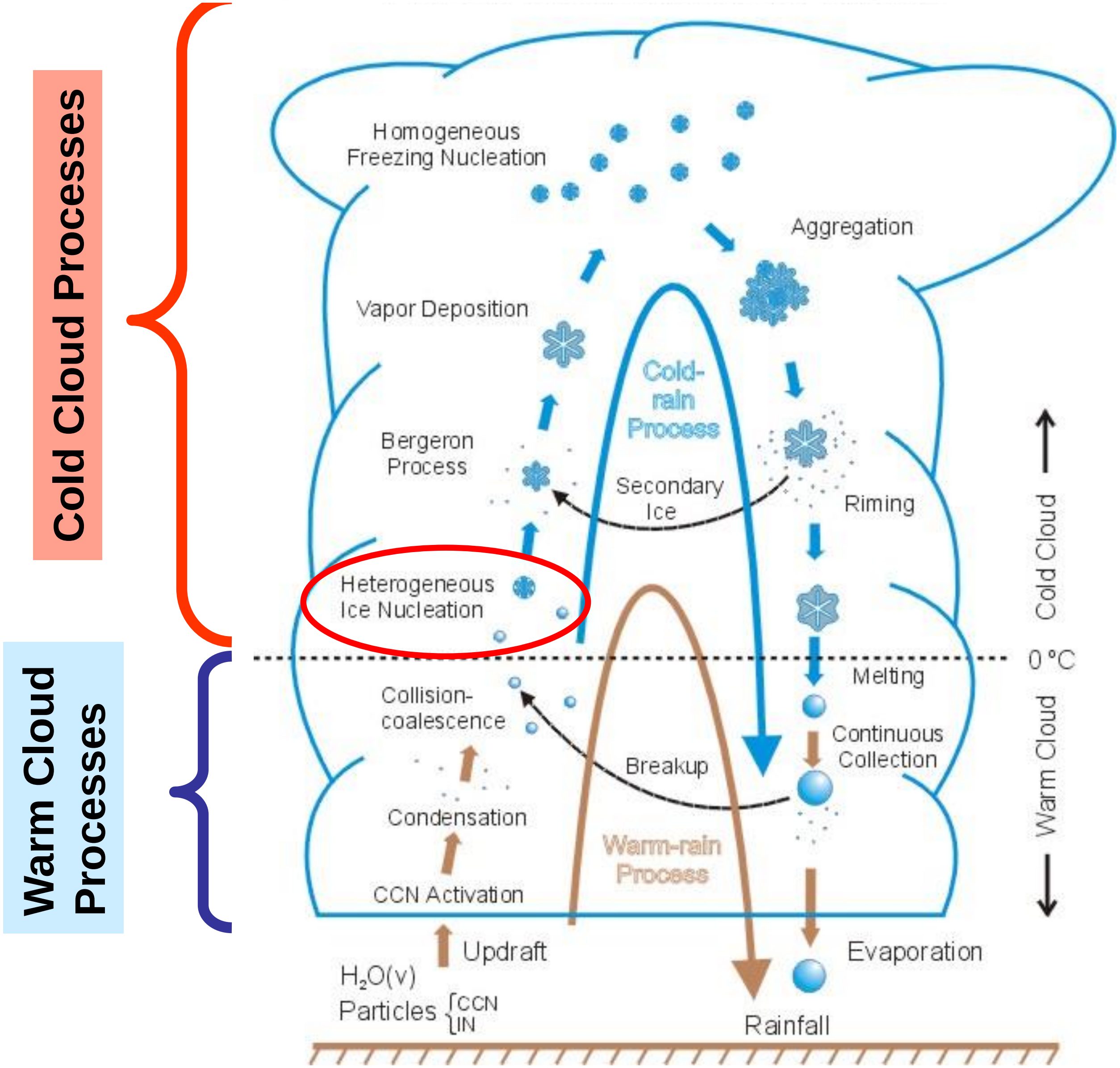
Cold Cloud Processes

Warm Cloud Processes



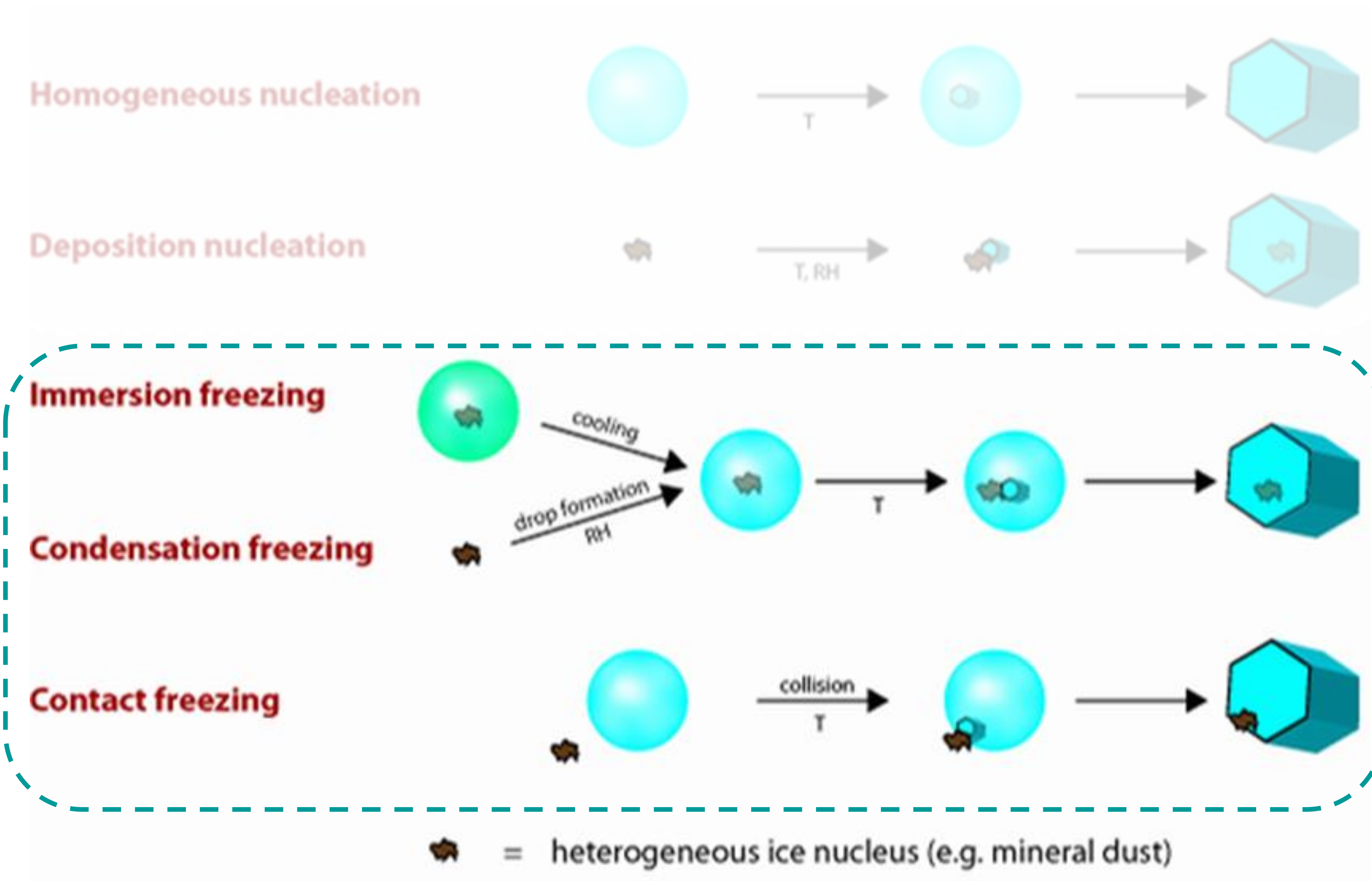
Platnick

The ice grows at the expense of drops



Platnick

Primary ice production mechanisms



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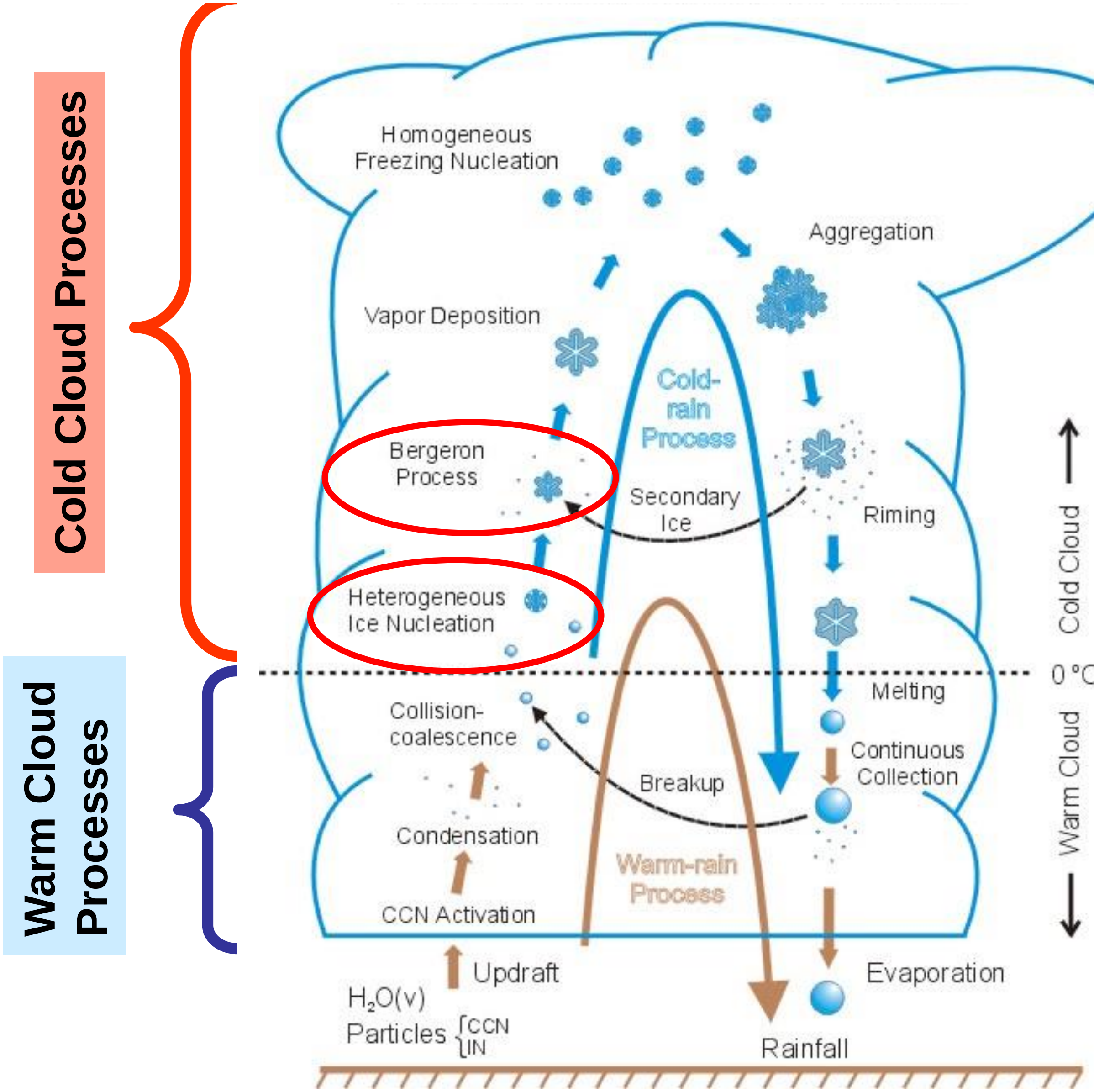
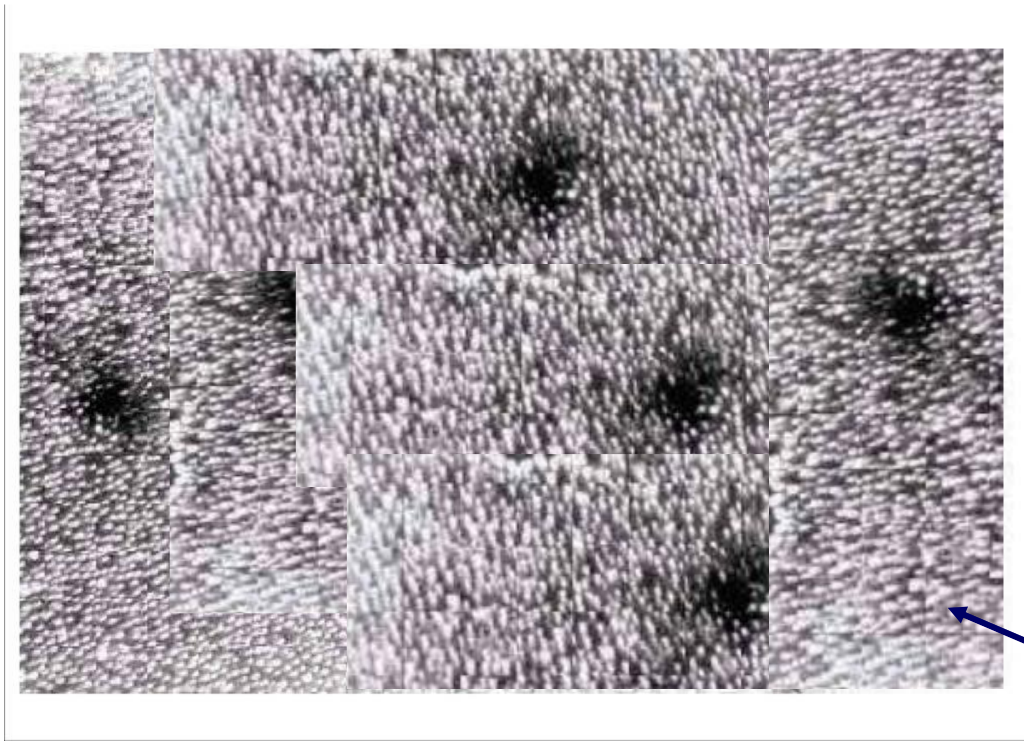


Platnick

Bergeron-Findesien process

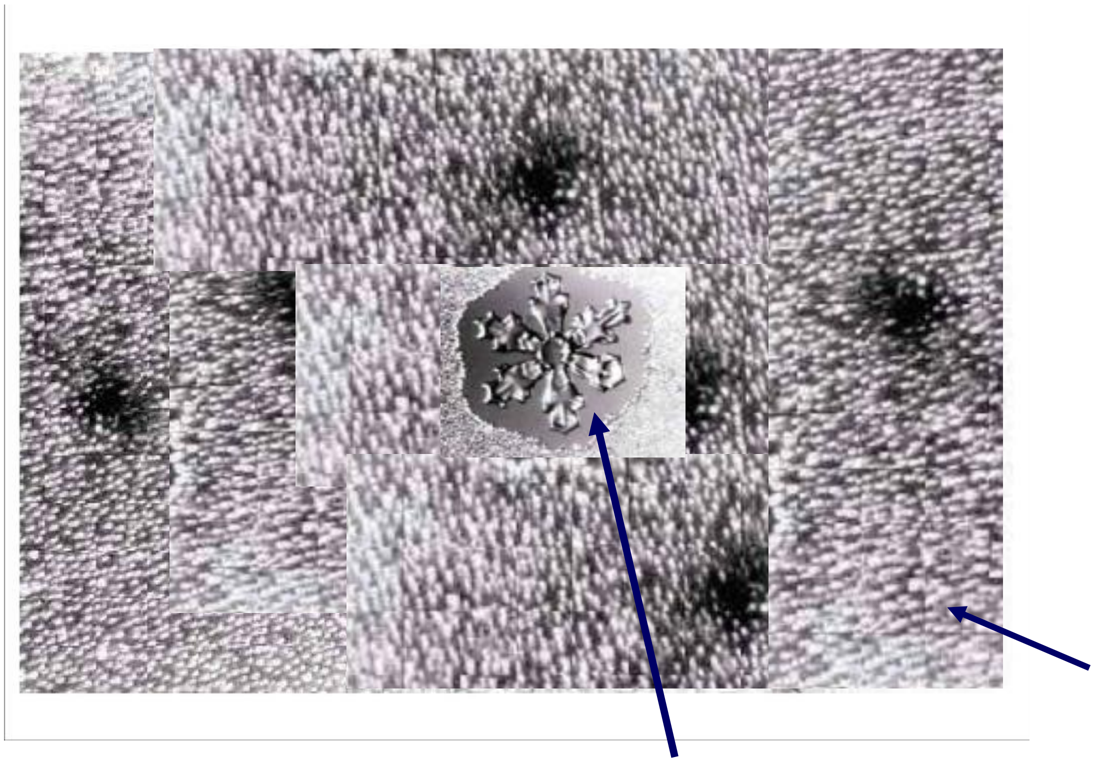
Critical for the microphysical evolution of mixed-phase clouds



Droplets on a plate

Bergeron-Findesien process

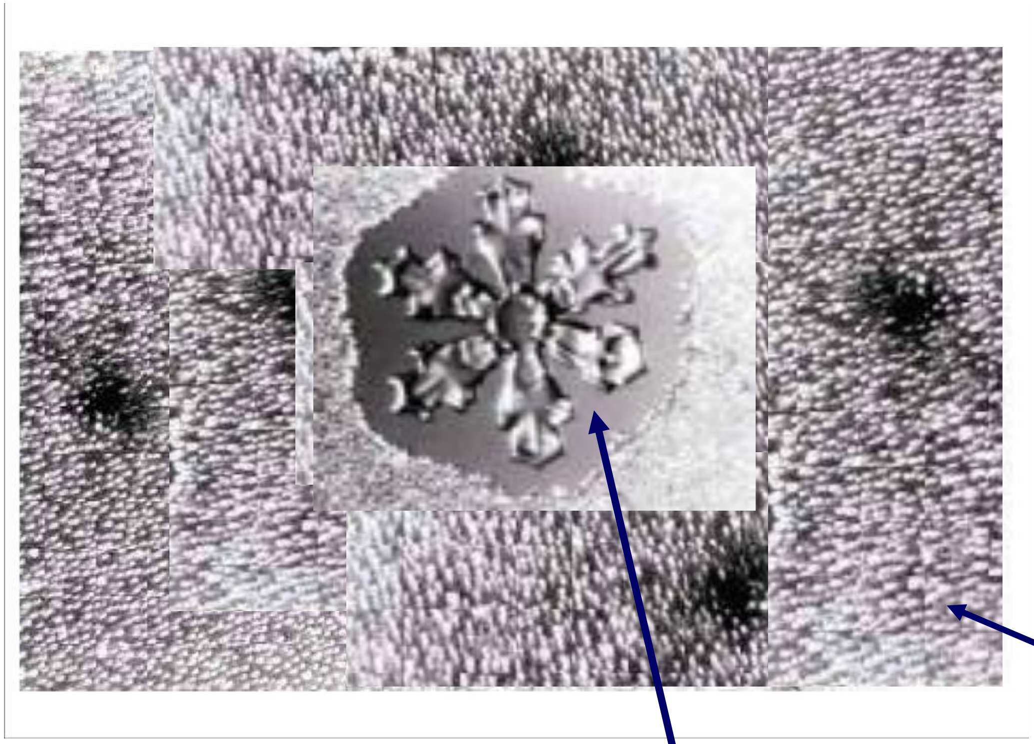
Critical for the microphysical evolution of mixed-phase clouds



Bergeron-Findesien process

Critical for the microphysical evolution of mixed-phase clouds

Vapor pressure over liquid water is HIGHER than over ice, so mass transfers between them.



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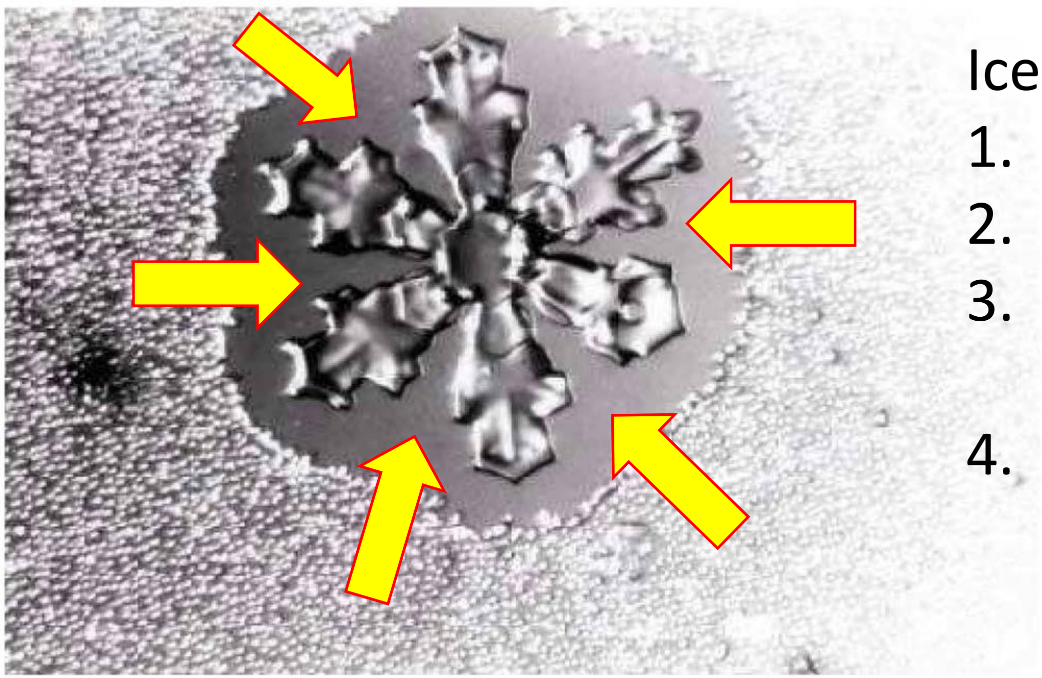


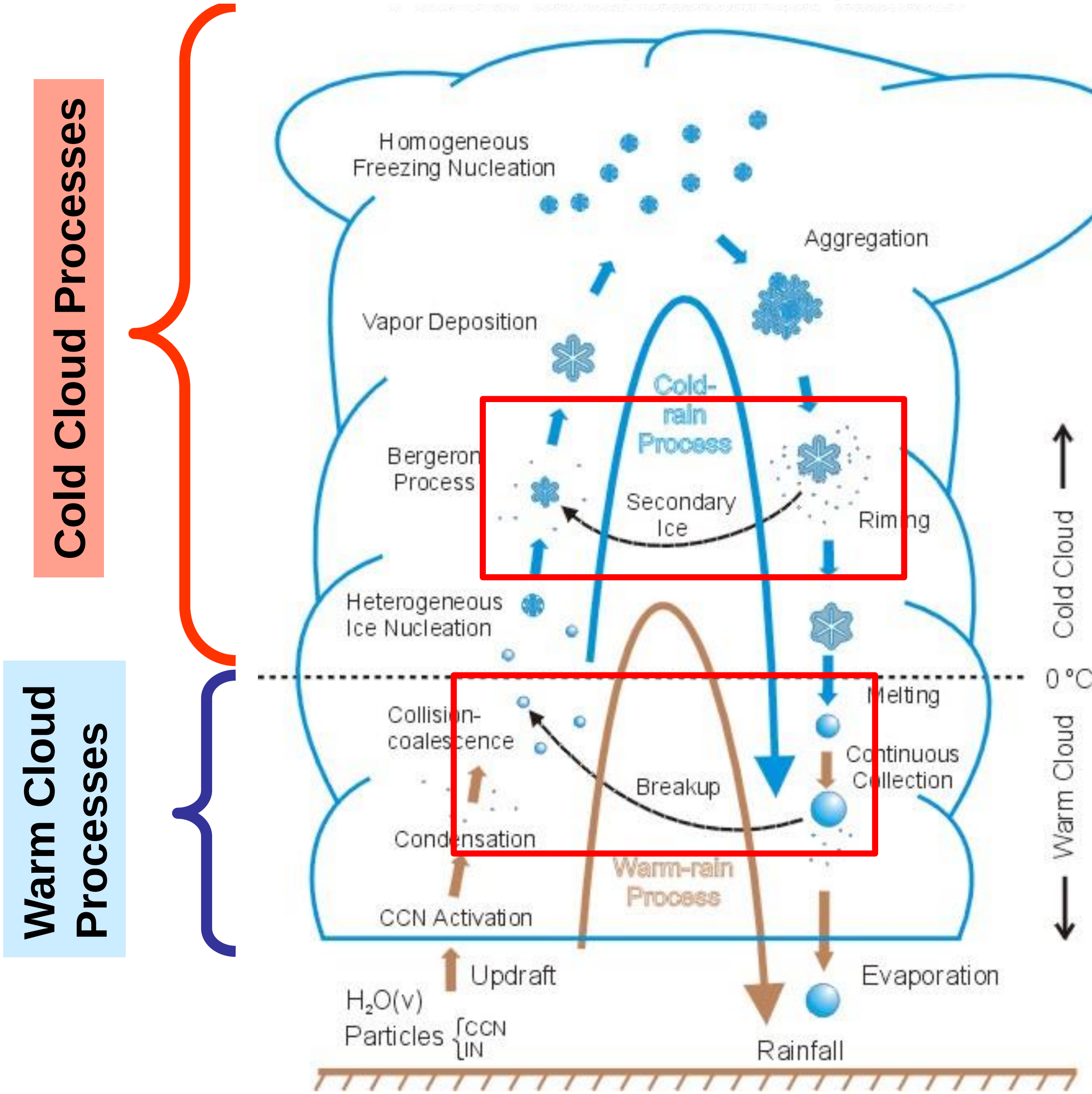
Ice crystals near droplets cause:

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 << CCN

Latent heat release important for dynamical evolution of clouds.

Shifts in particle size affects radiation and precipitation of cloud.

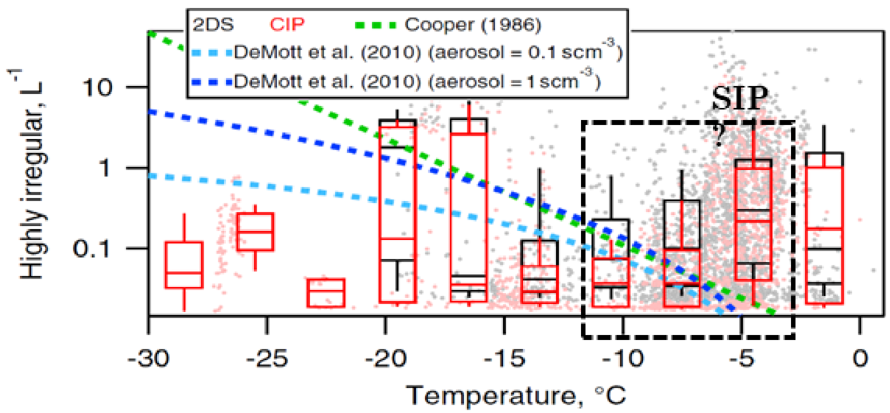
Where secondary ice production "lies"



Platnick

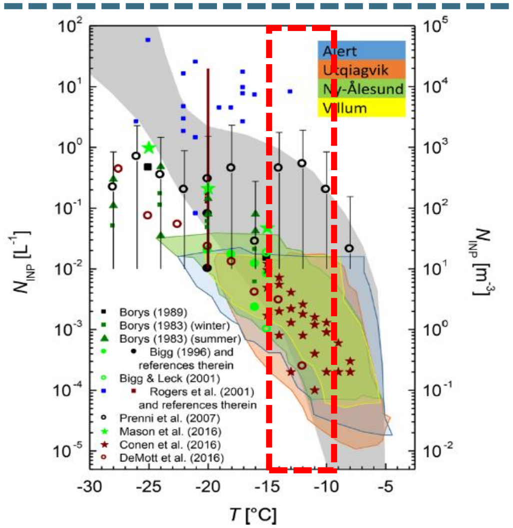
Measured Ice Crystal Number is much higher than pre-cloud INPs!

- ✓ 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

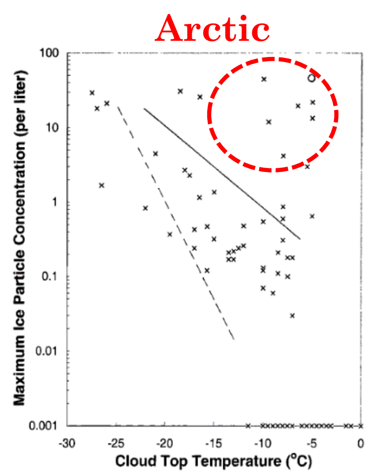


Antarctic

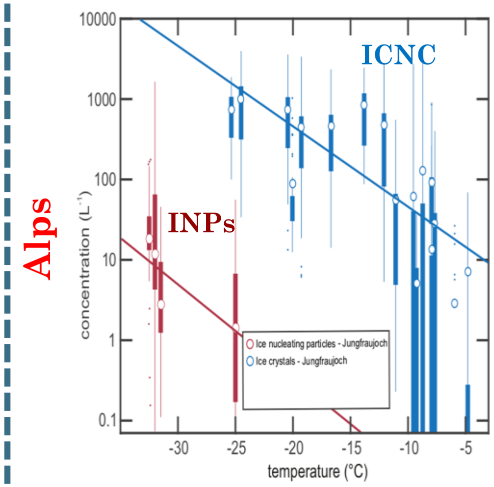
O'Shea et al. 2017



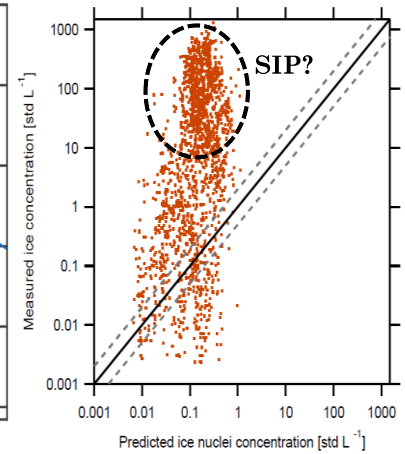
Wex et al. 2019



Ragno & Hobbs 2001



Beck et al. 2018



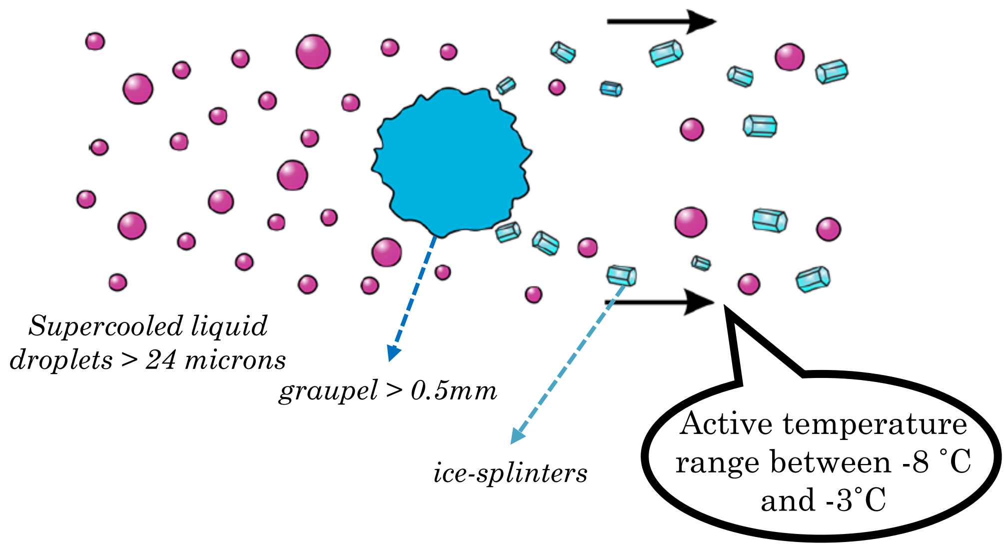
Lloyd et al. 2015

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

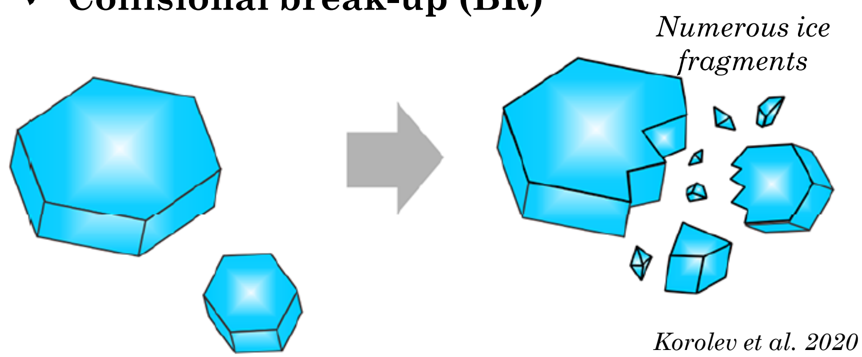
The cause of this cloud-ice paradox → Secondary Ice Production (SIP)*

* SIP = multiplication of primary ice crystals through "other processes" not involving INPs

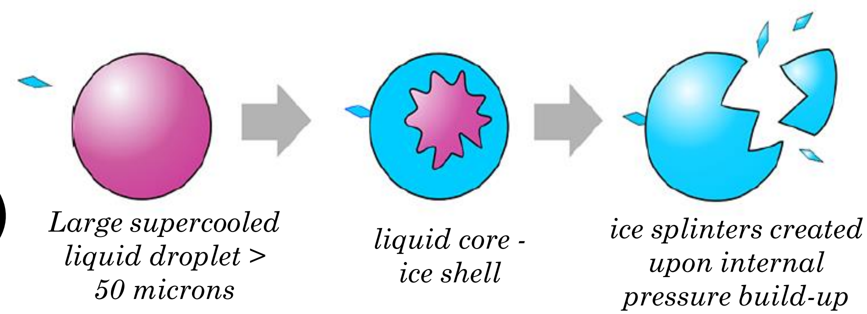
✓ **Rime Splintering (RS) or the Hallett-Mossop process (H-M)**



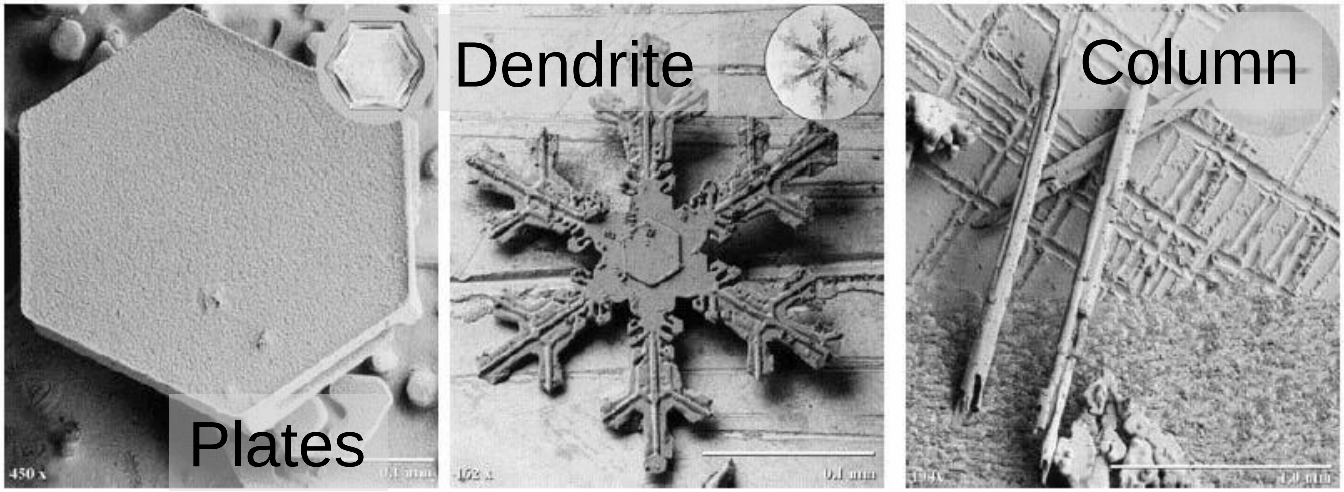
✓ **Collisional break-up (BR)**

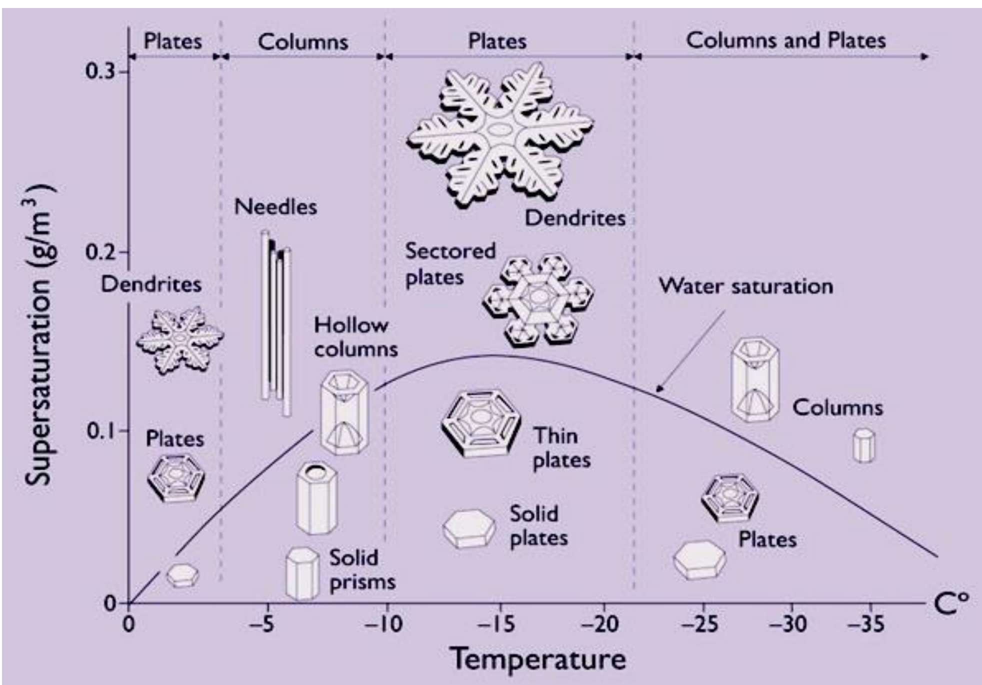


✓ **Droplet Shattering (DS) during freezing**



"Pristine" ice crystals – before riming

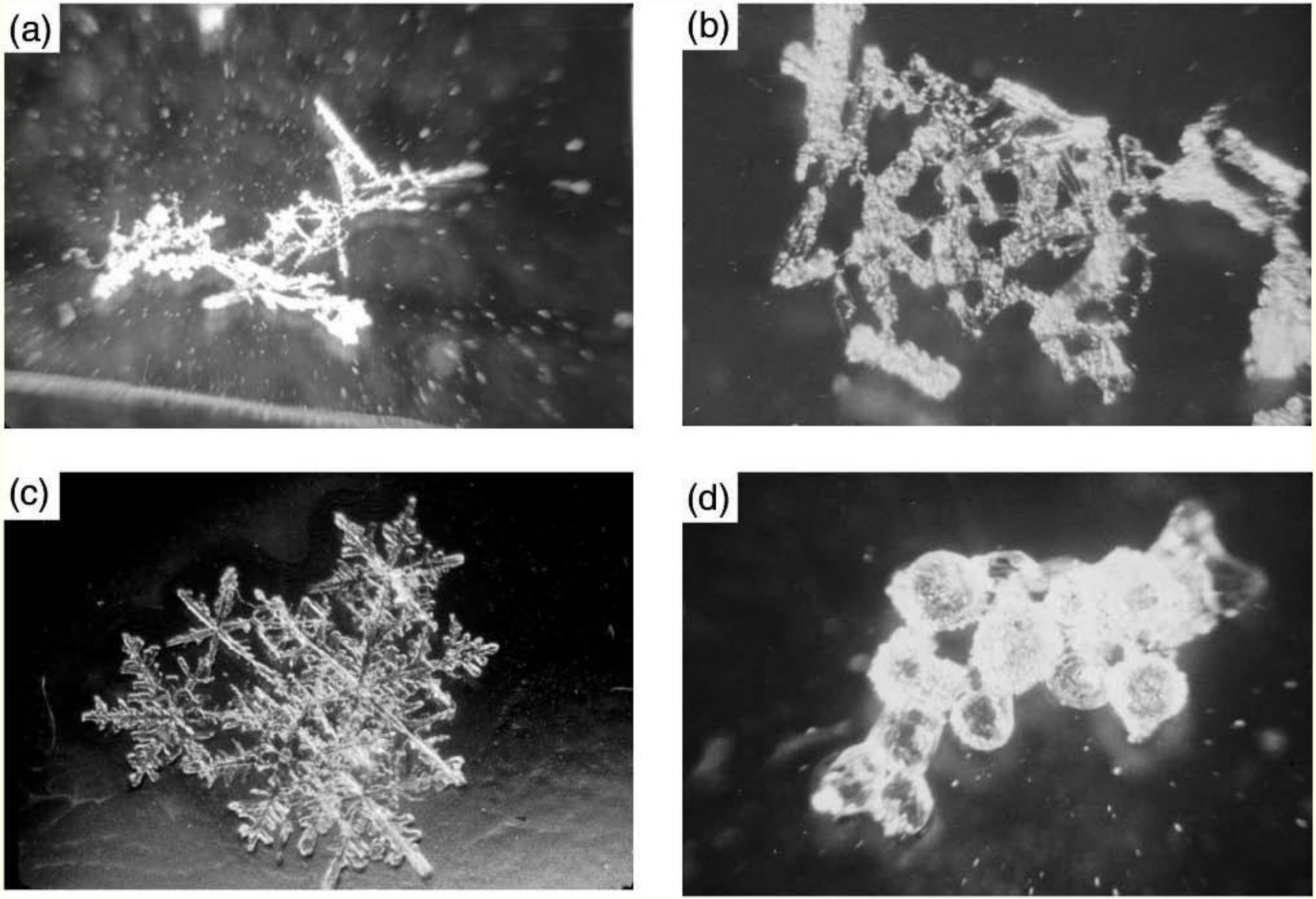




Primary ice production

Growth via water vapor deposition only.

"Pristine" ice crystals – after riming



Pioneering work of Hallett & Mossop

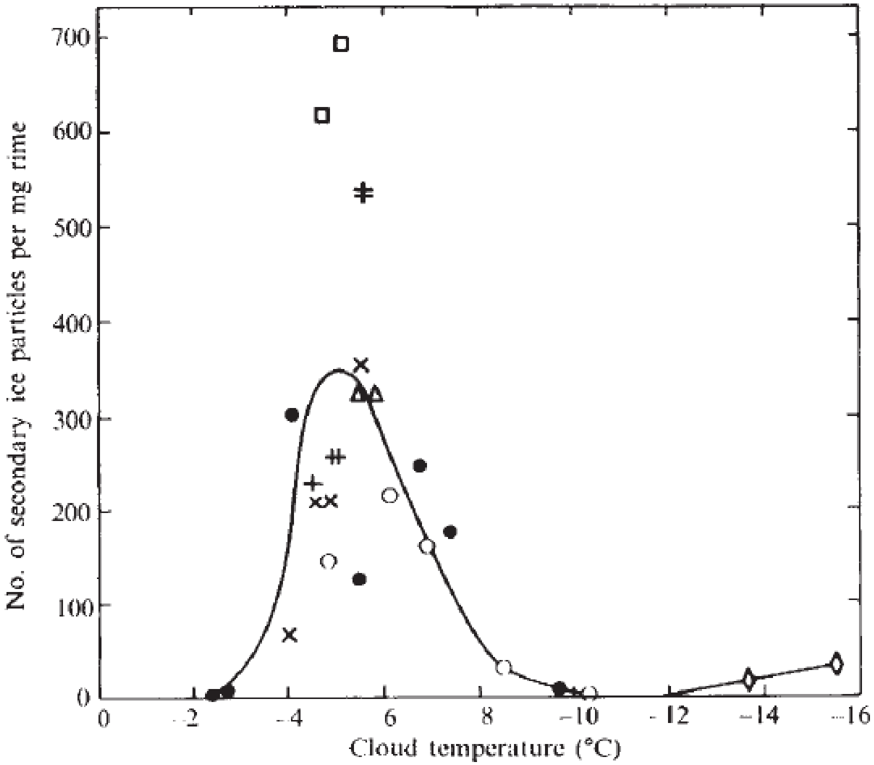


Fig. 2 Production of secondary ice particles by riming as a function of temperature at a target velocity of 2.7 m s^{-1}. Different symbols indicate different days. The curve was drawn by averaging the points over narrow temperature intervals.

Hallett and Mossop 1974

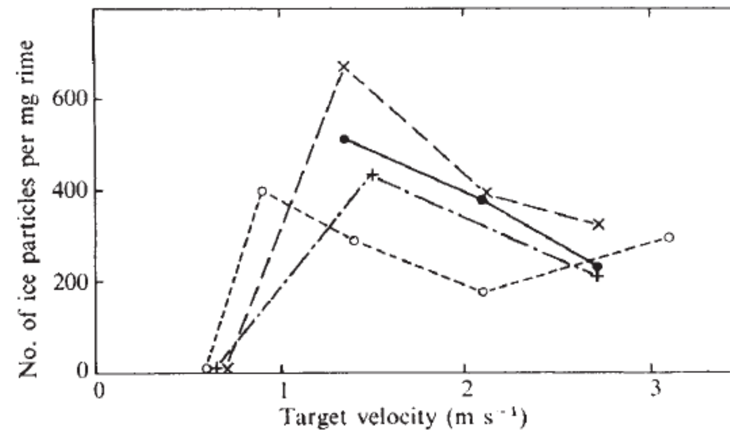
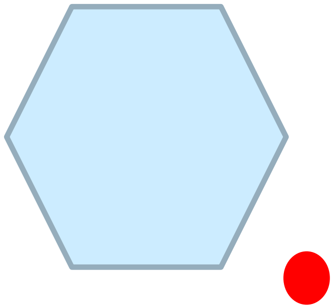


Fig. 3 Production of secondary ice particles by riming as a function of target velocity at a temperature of about −5° C. ●, November 5, 1973; —4.5° C. ×, November 9, 1973; —5.5° C. ○, December 3, 1973; —5.7° C. +, December 5, 1973; —5.2° C.

- Mossop and Hallett (1974): production of splinters requires presence of droplets >25 μm diameter.
- One ice crystal can generate up to 300 times its concentration in secondary ice crystals (explosive growth of ice) – rapid glaciation.

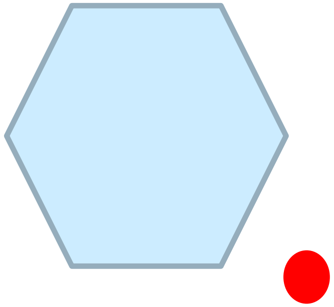
Physical Mechanism for splinter production



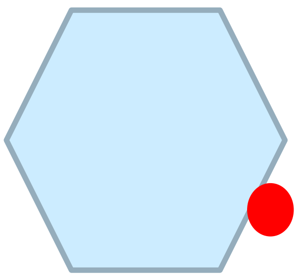
Rimer approaches liquid drop with diameter >25μm

Choularton et al. 1978

Physical Mechanism for splinter production



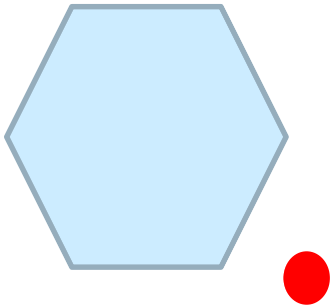
Rimer approaches liquid
drop with diameter >25μm



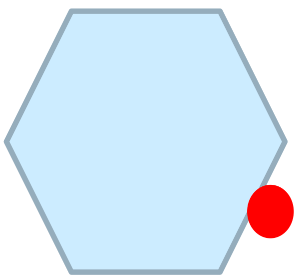
Rimer contacts liquid
drop and they stick

Choularton et al. 1978

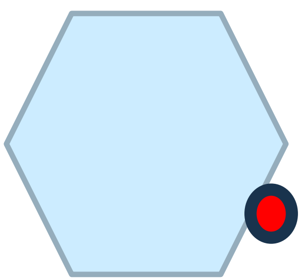
Physical Mechanism for splinter production



Rimer approaches liquid drop with diameter >25μm



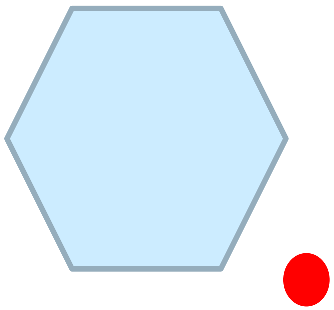
Rimer contacts liquid drop and they stick



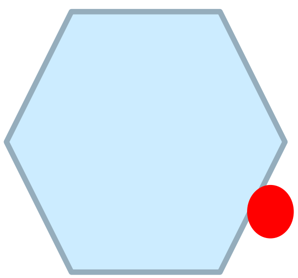
Ice shell forms

Choularton et al. 1978

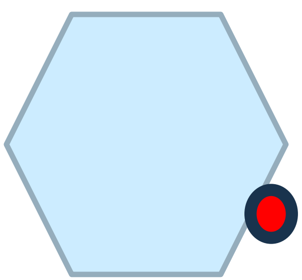
Physical Mechanism for splinter production



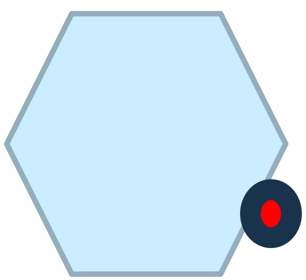
Rimer approaches liquid drop with diameter >25μm



Rimer contacts liquid drop and they stick



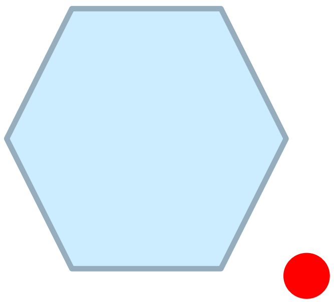
Ice shell forms



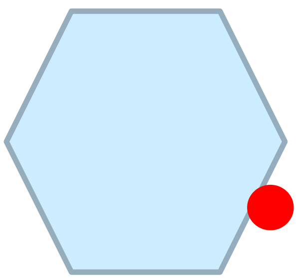
Ice shell **thickens** and builds internal pressure

Choularton et al. 1978

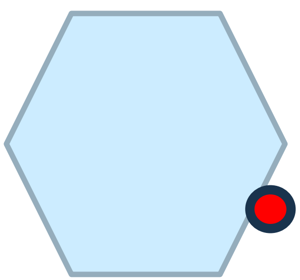
Physical Mechanism for splinter production



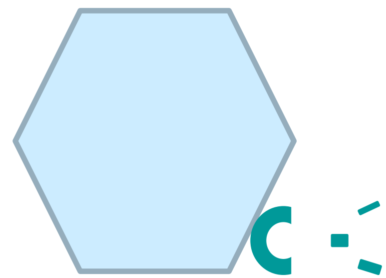
Rimer approaches liquid drop with diameter > 25μm



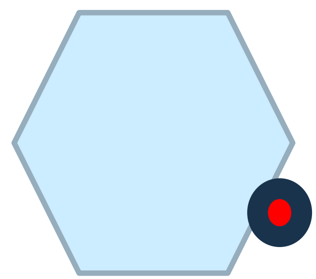
Rimer contacts liquid drop and they stick



Ice shell forms



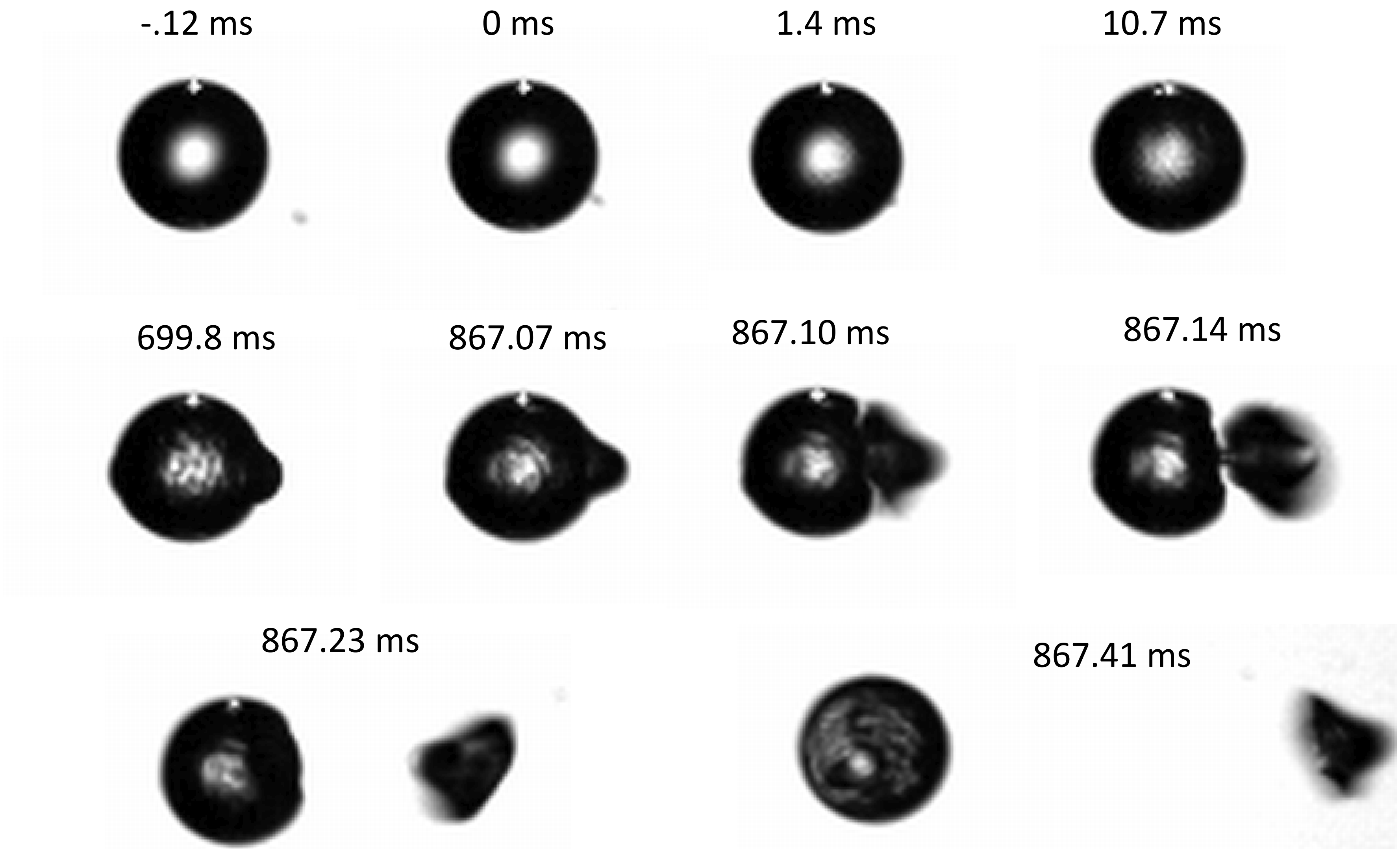
Pressure relieved by cracking shell and ejected splinters



Ice shell **thickens** and builds internal pressure

Choularton et al. 1978

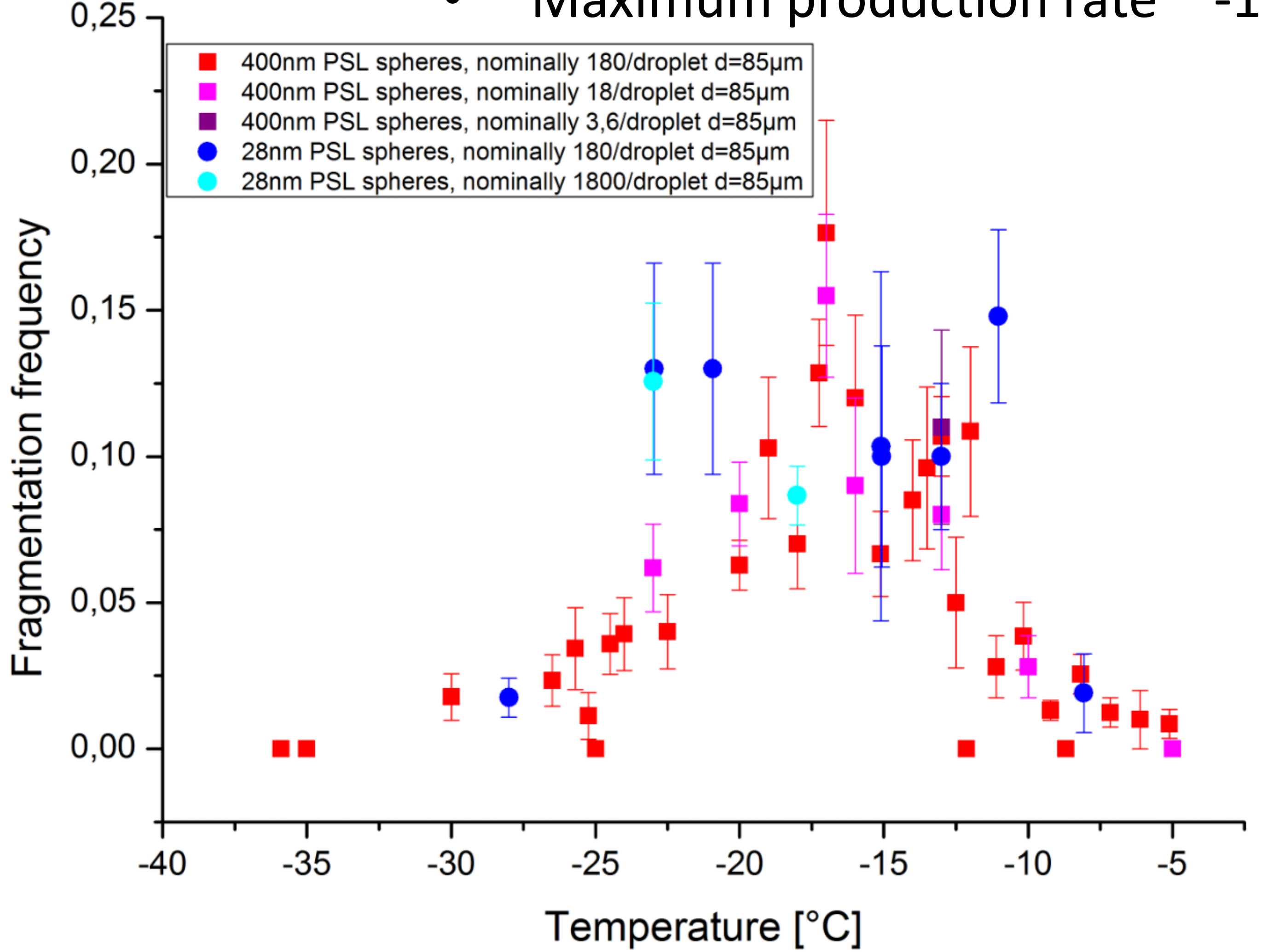
Other mechanisms – Droplet Explosions



From Leisner et al.

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

- Maximum production rate ~ -16^{o}C



Ice-ice collisions

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

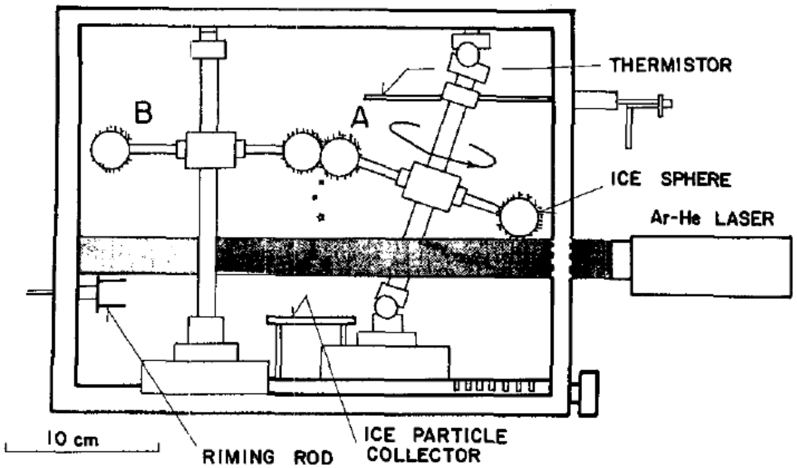
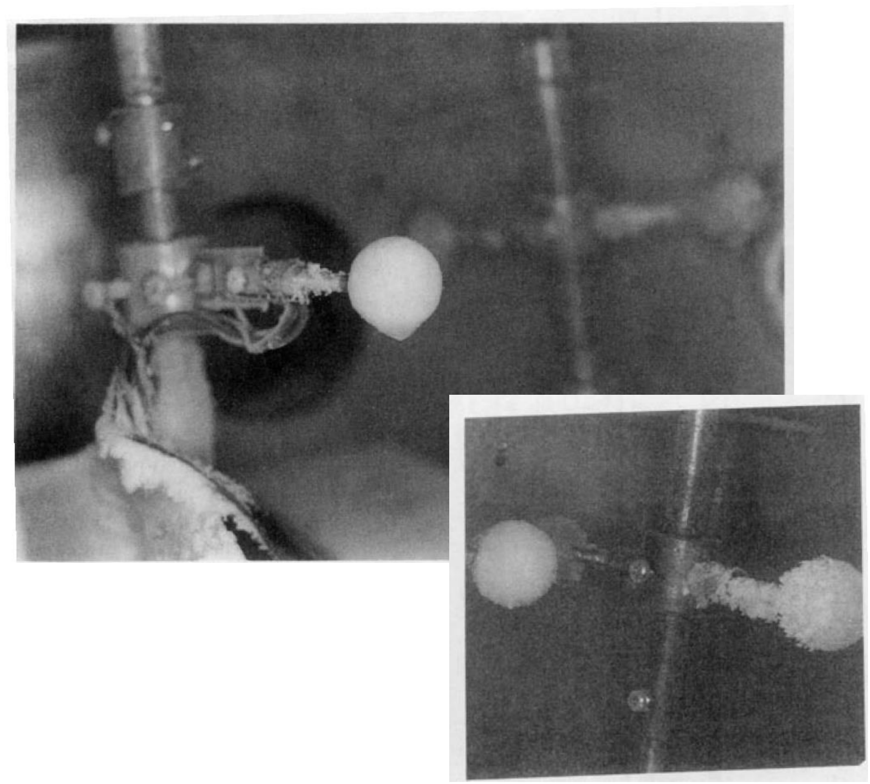


FIG. 1. Experimental apparatus: (A) ice spheres (right) were set up to rotate at a tangential speed of 4 m s^{-1}, while (B) ice spheres (left) were stationary. The ejected ice particles were collected on a plate at the bottom of the chamber. A thermistor for temperature measurement was installed at 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.



Takahashi et al. (1995)

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).

Ice-ice collisions

- Larger number of fragments produced *vs*. other mechanisms
- Maximum production rate ~ -16°C

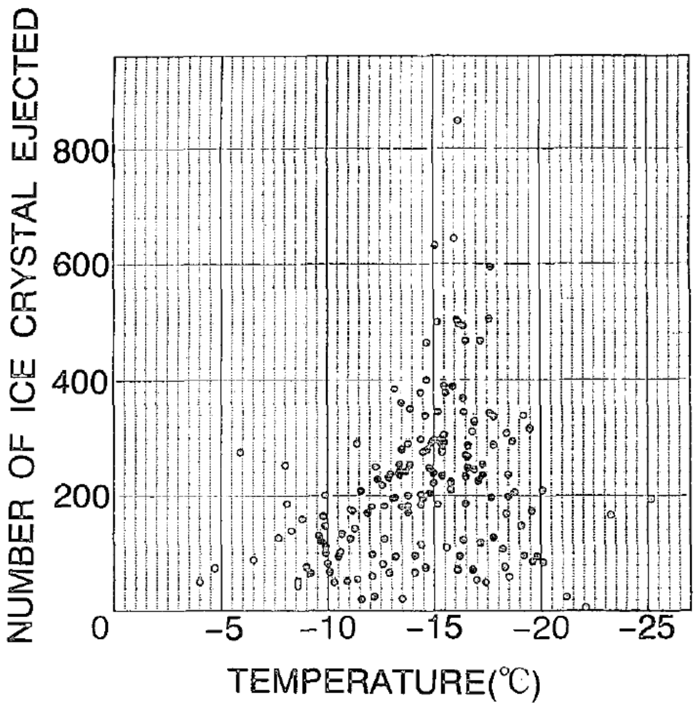


FIG. 4. Total number of ejected ice particles as a function of chamber temperature. The number of ice crystals collected by the main plate was simply multiplied by 4. The total number is therefore approximate. Cases of various collision forces were all included. Collision force was on the order of 500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

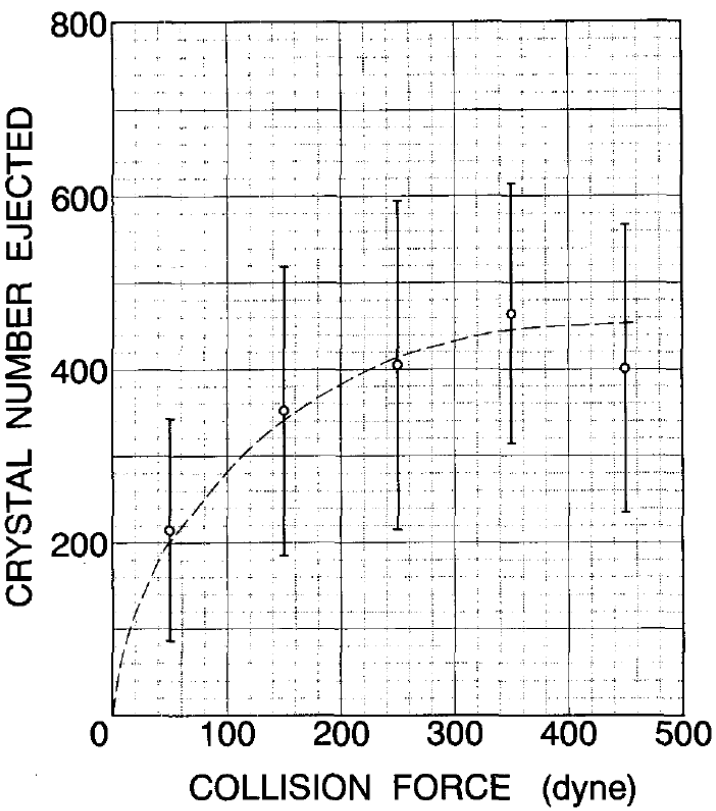
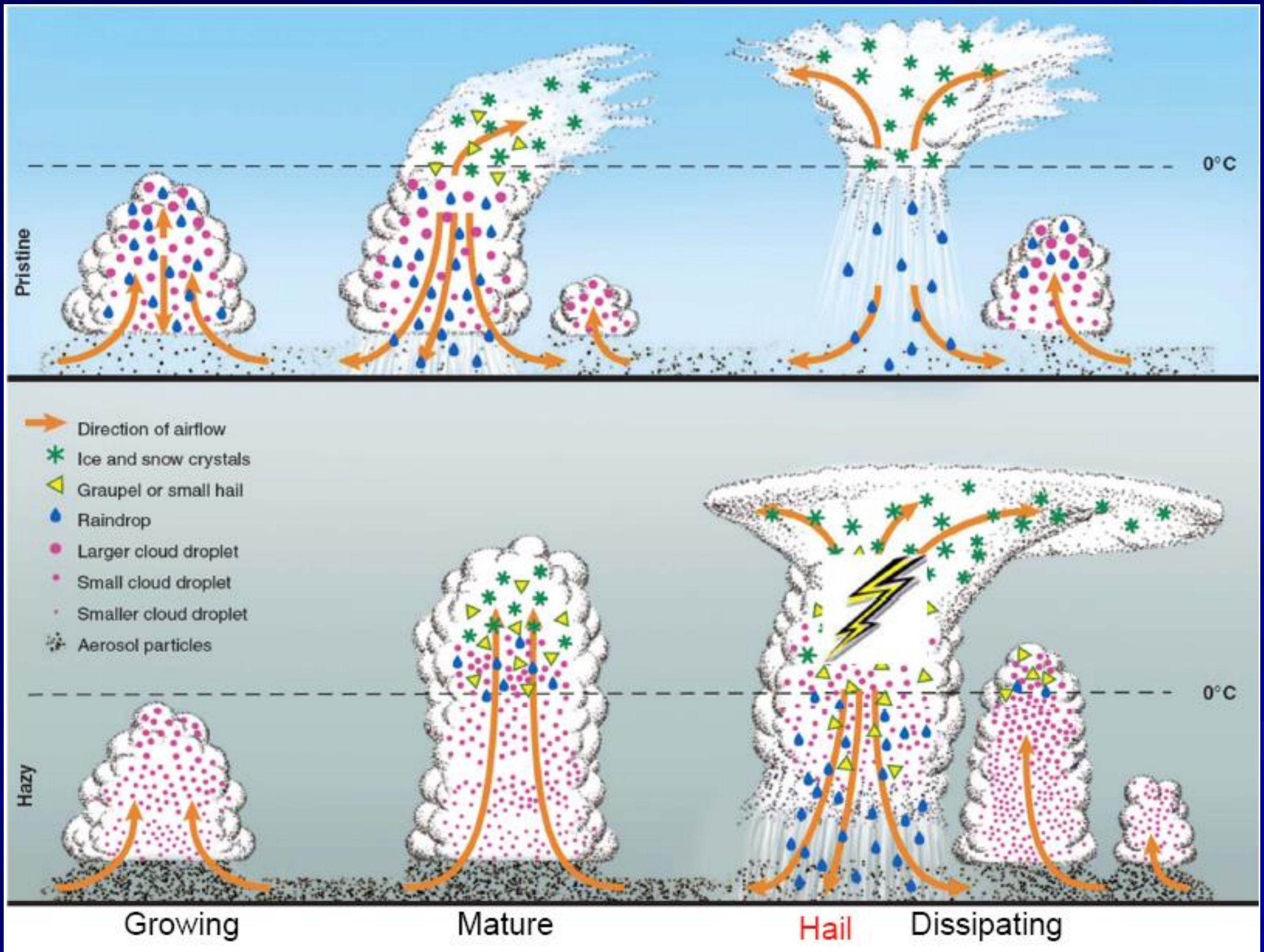


FIG. 5. Total ejected ice particle numbers vs collision force. Thick marks represent standard deviations of the measurements, and the thin dashed line is the smoothed value. Data was grouped in 100-dyn range for analysis.

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?).

Some take-home messages

- Aerosols provide the nuclei for all cloud hydrometer 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.

Acknowledgments

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C·STACC
Center for the Study of Air Quality & Climate Change

EPFL

Thank you!!
Ευχαριστώ!!!

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NASA

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SCHWEIZERISCHER NATIONALFONDS
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