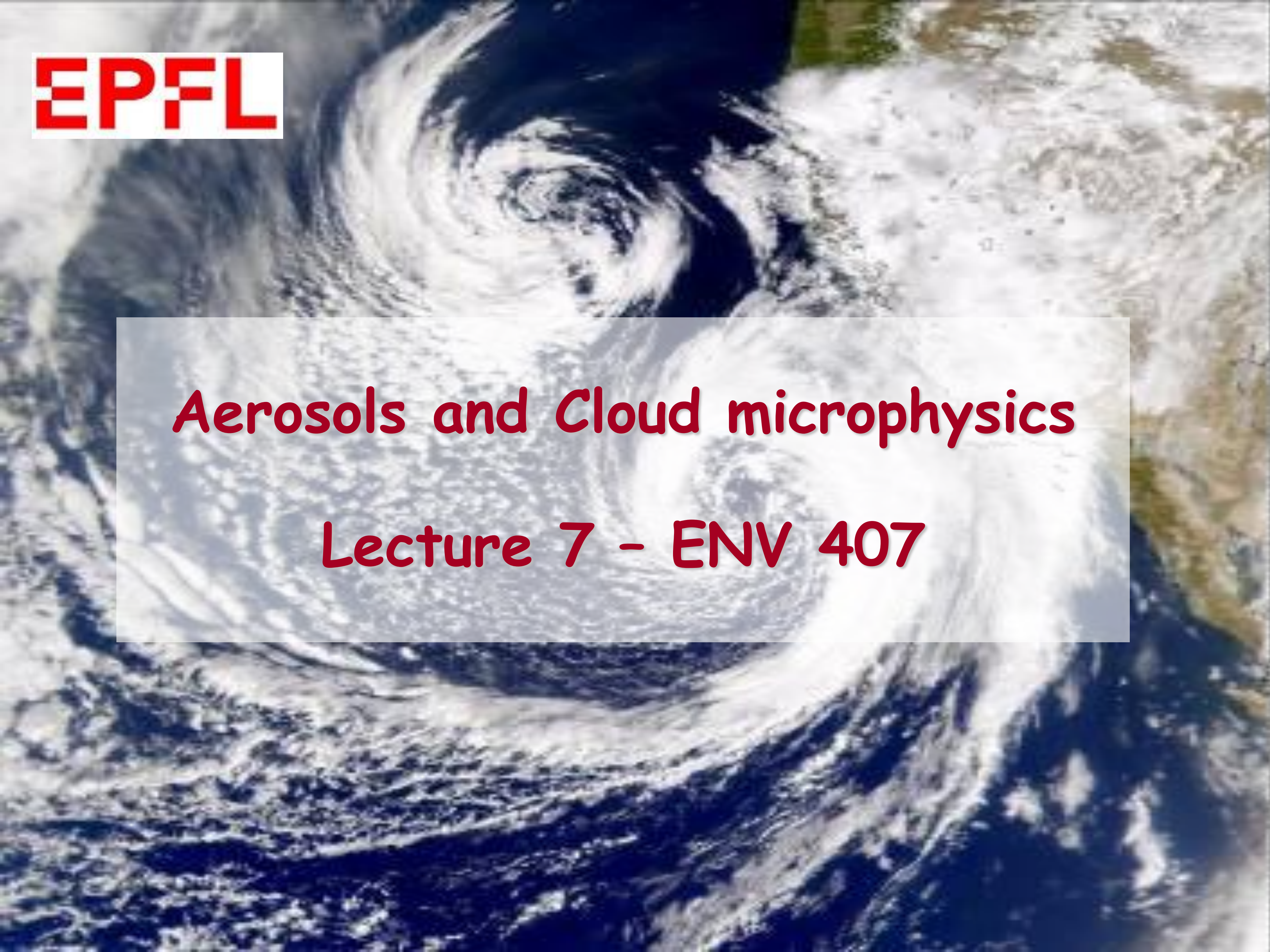
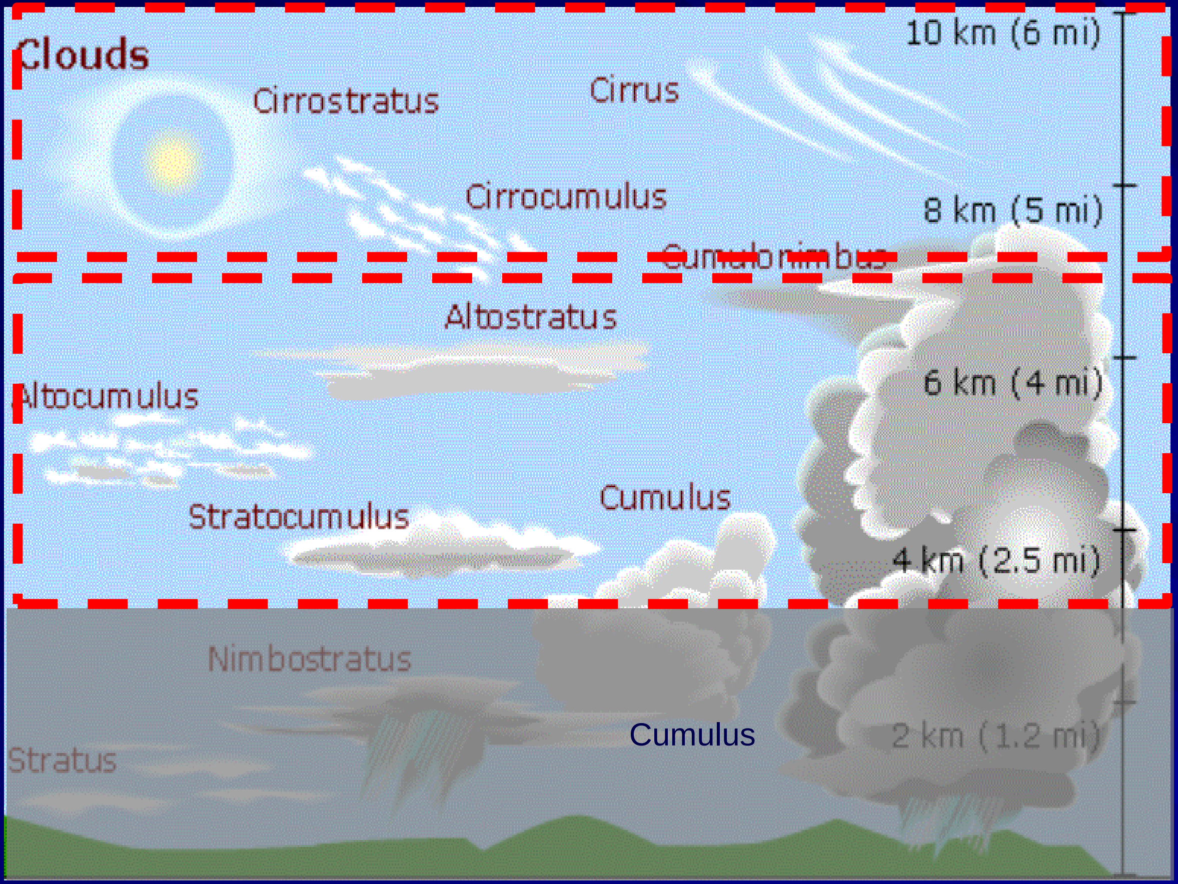


EPFL

Aerosols and Cloud microphysics

Lecture 7 – ENV 407

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

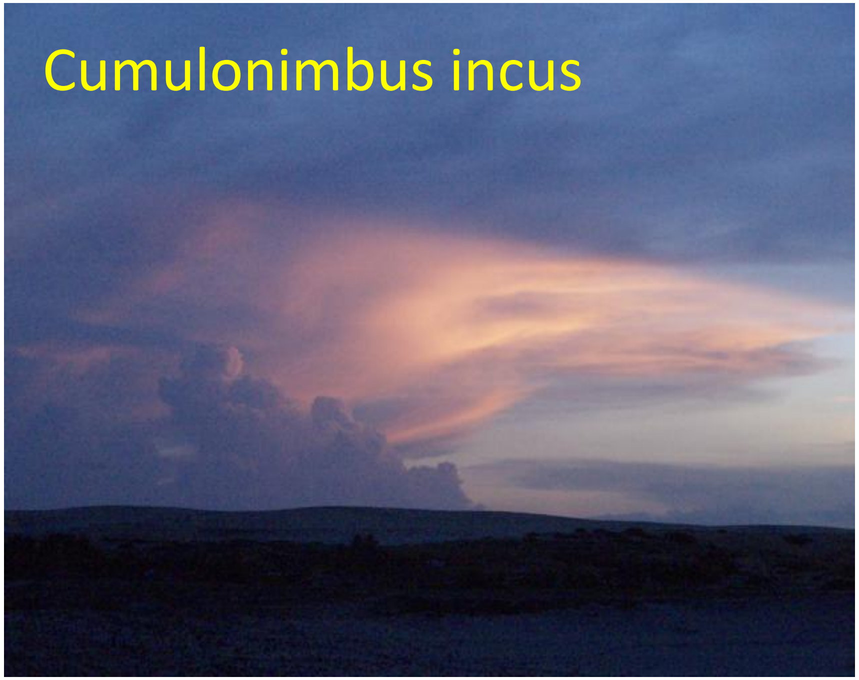
Cloud particles are not created directly from the vapor phase but from **suspended aerosol particles**

http://www.ucar.edu/news

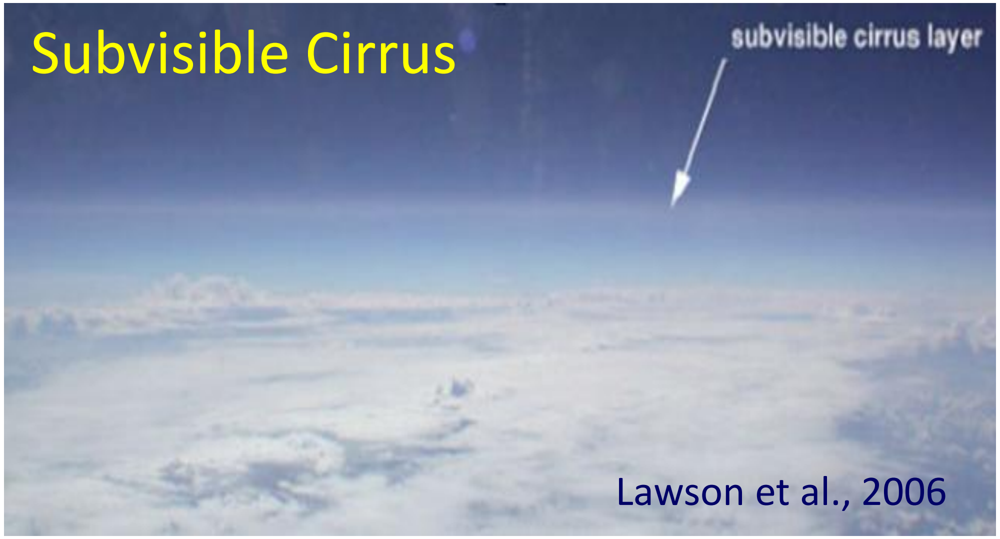
Pictures of ice clouds


Tropical Cumulonimbus

www.solarviews.com

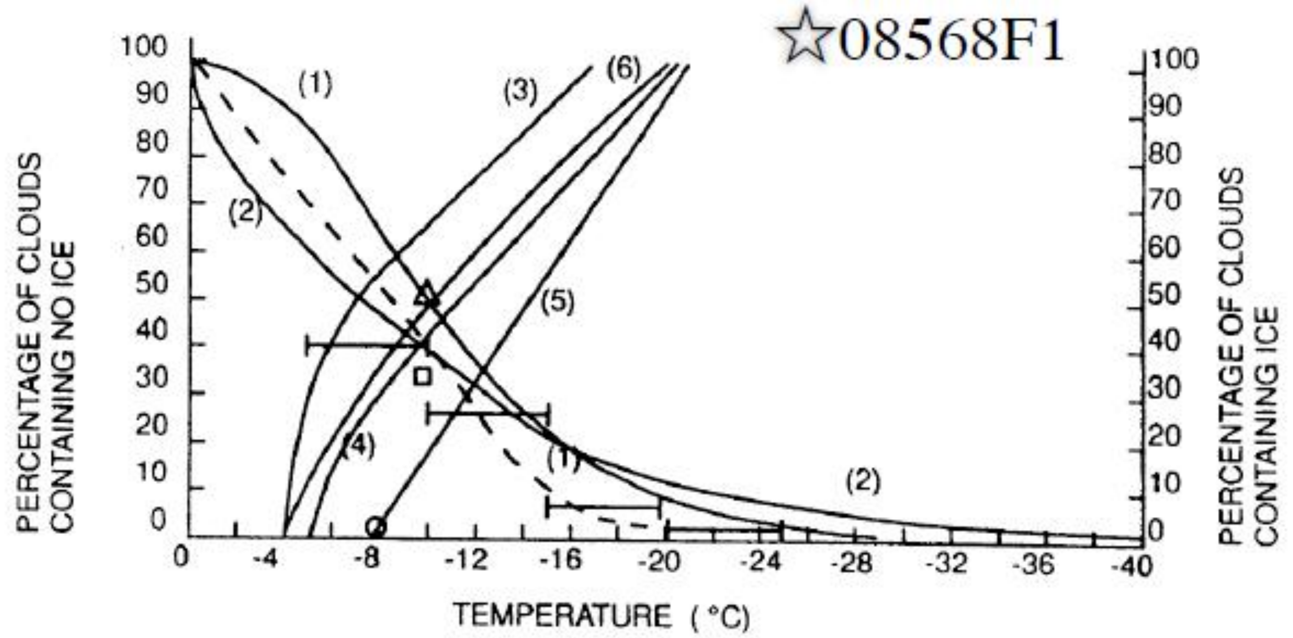

Cumulonimbus incus


Cirrostratus


Subvisible Cirrus

Lawson et al., 2006

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

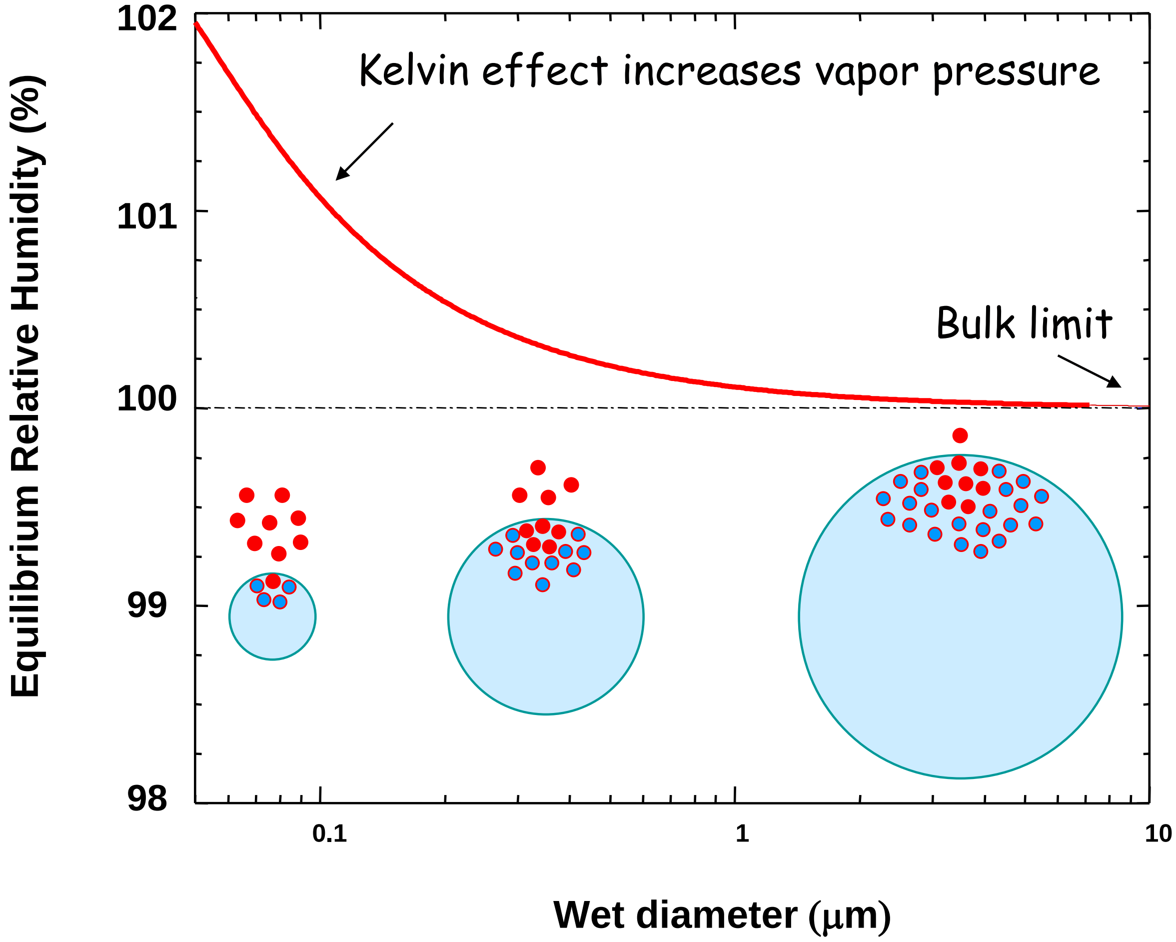
Surprise! It takes *a lot* of cooling to freeze

Understanding why it takes a lot of cooling to freeze

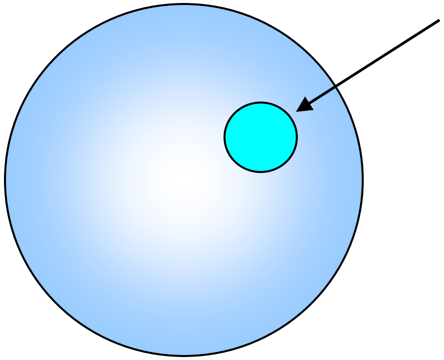
Impact of curvature:

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

Kelvin Equation

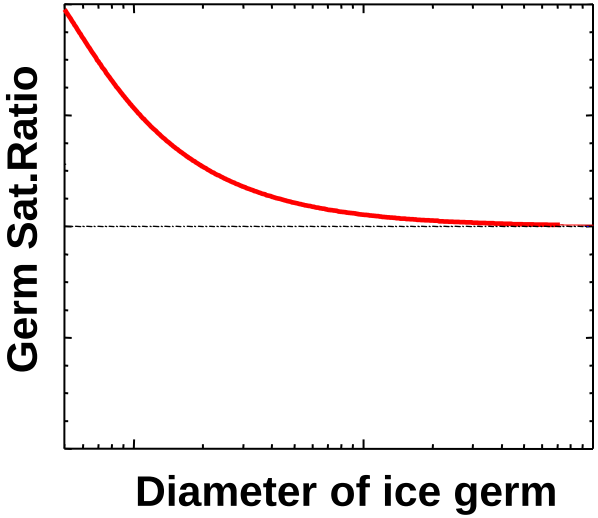


Curvature effect: the basis of nucleation



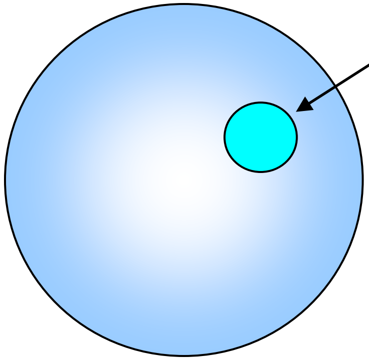
Ice Germs: generated in supercooled droplets

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

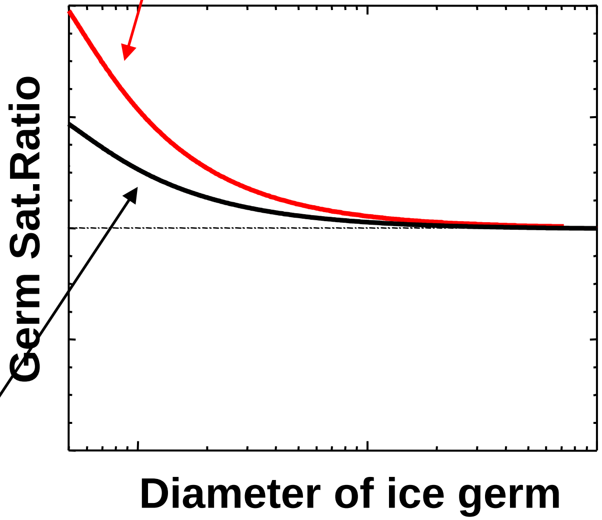


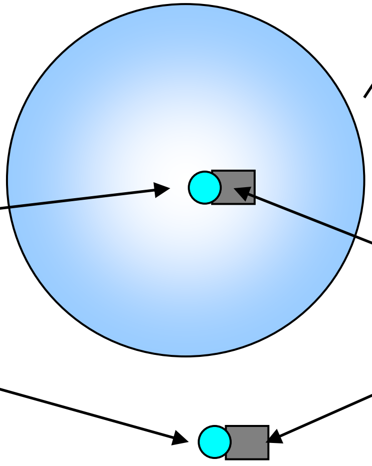
Curvature effect: the basis of nucleation

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 superscooled droplets or suspended in air

Insoluble particle

(why do insolubles help?)

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

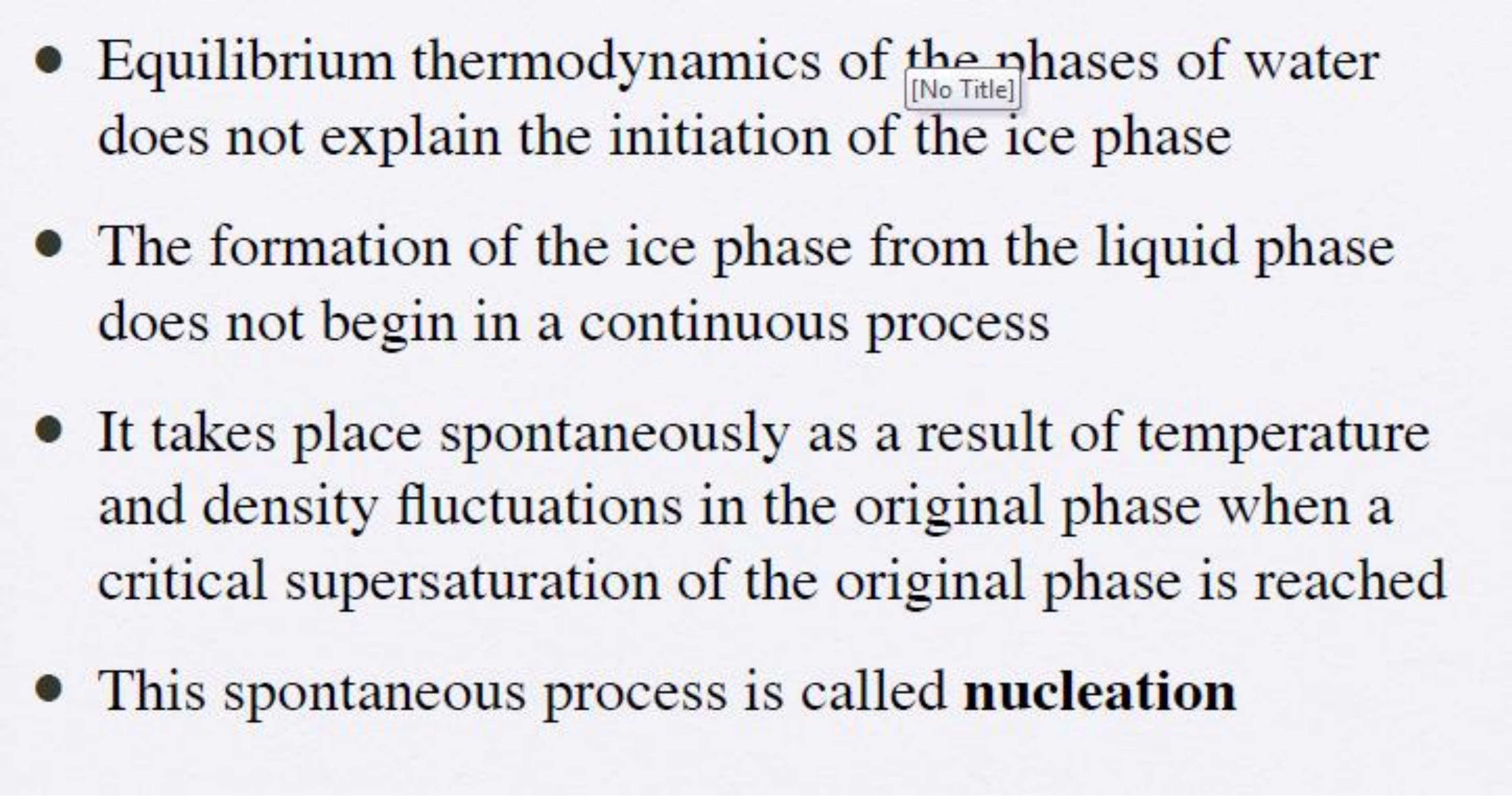
5 Καταπληκτικά Πειράματα & Κόλπα Νερού - Στιγμιαία Κατάψυξη Νερού

Mr. Hacker
3,45 εκ. εγγεγραμμένοι

8,8 εκ. προβολές πριν από 8 έτη #νερό #επιστήμη #πειράματα
Σε αυτό το εκπαιδευτικό βίντεο θα δείτε 5 εκπληκτικά επιστημονικά πειράματα με το στιγμιαίο πάγωμα του νερού σε πάγο. Ξέρετε

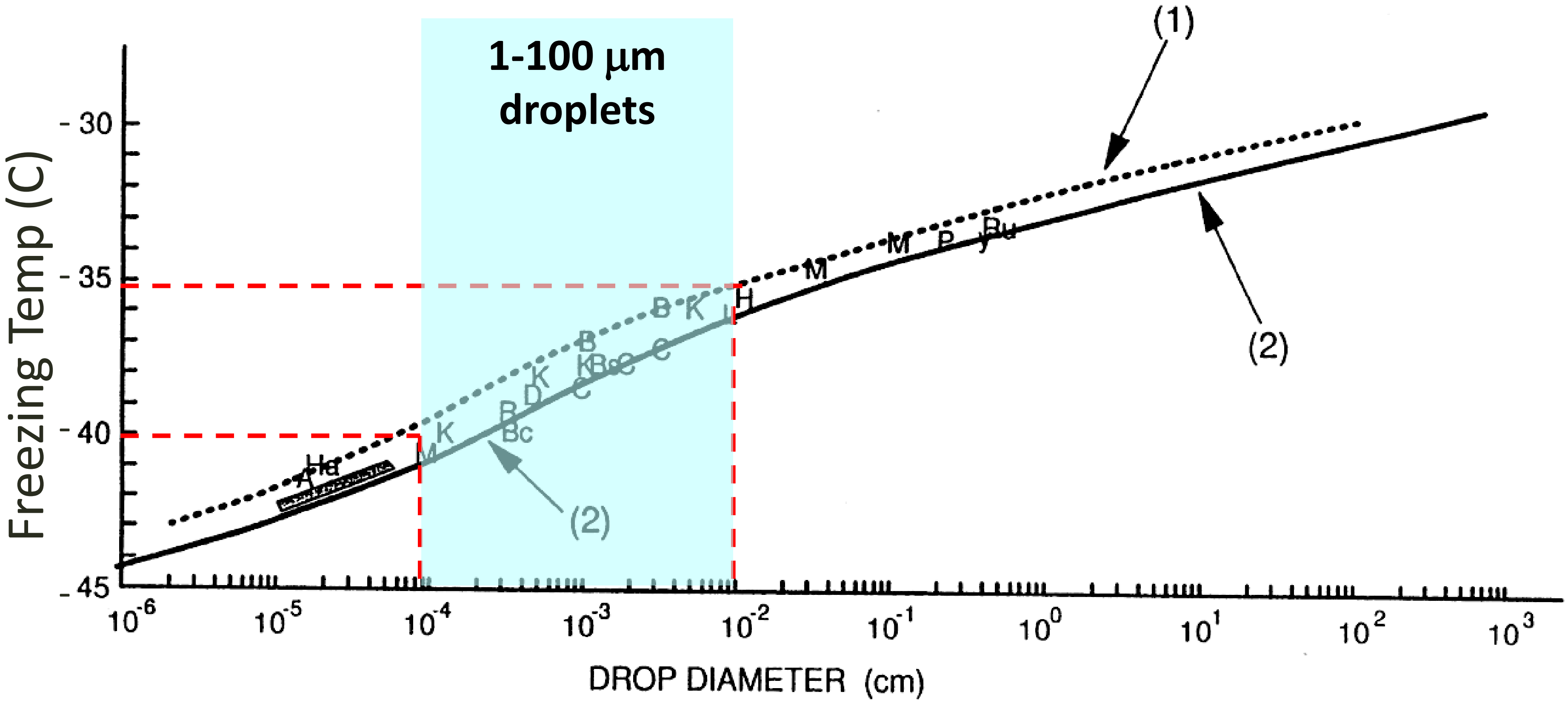
https://www.youtube.com/watch?v=kEHdyiBMgAg

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

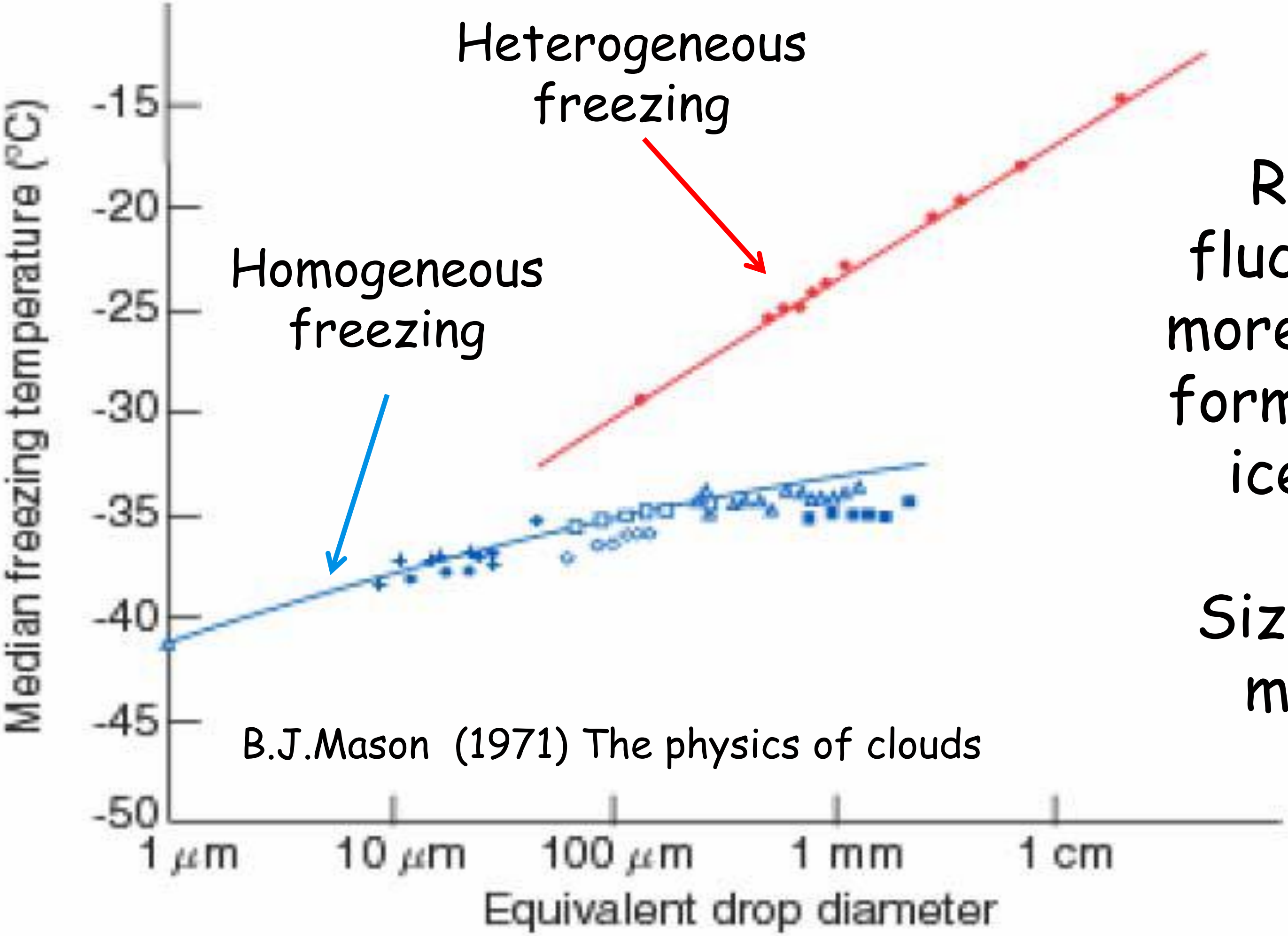


Result: Pure water cloud droplets in the atmosphere effectively start freezing at around -35^{o}C

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

Median freezing T of water samples

Not really.... Heterogeneous Freezing Greatly Facilitates Ice Formation



Heterogeneous freezing

Homogeneous freezing

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

Random fluctuations more likely to form a stable ice germ:

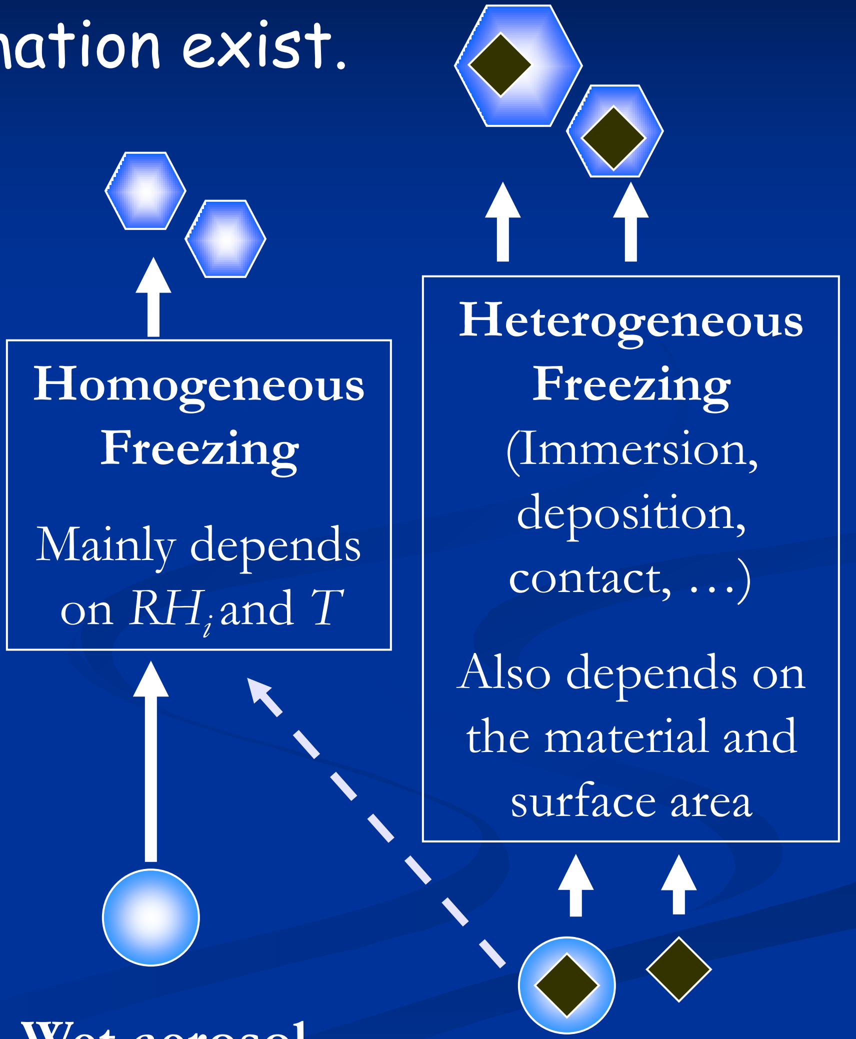
Size always matters

How do (ice water) clouds form?

Ice crystals also form on preexisting particles.

Multiple mechanisms for ice formation exist.

http://www.alanbauer.com



Homogeneous Freezing

Mainly depends on RH_i and T

Heterogeneous Freezing

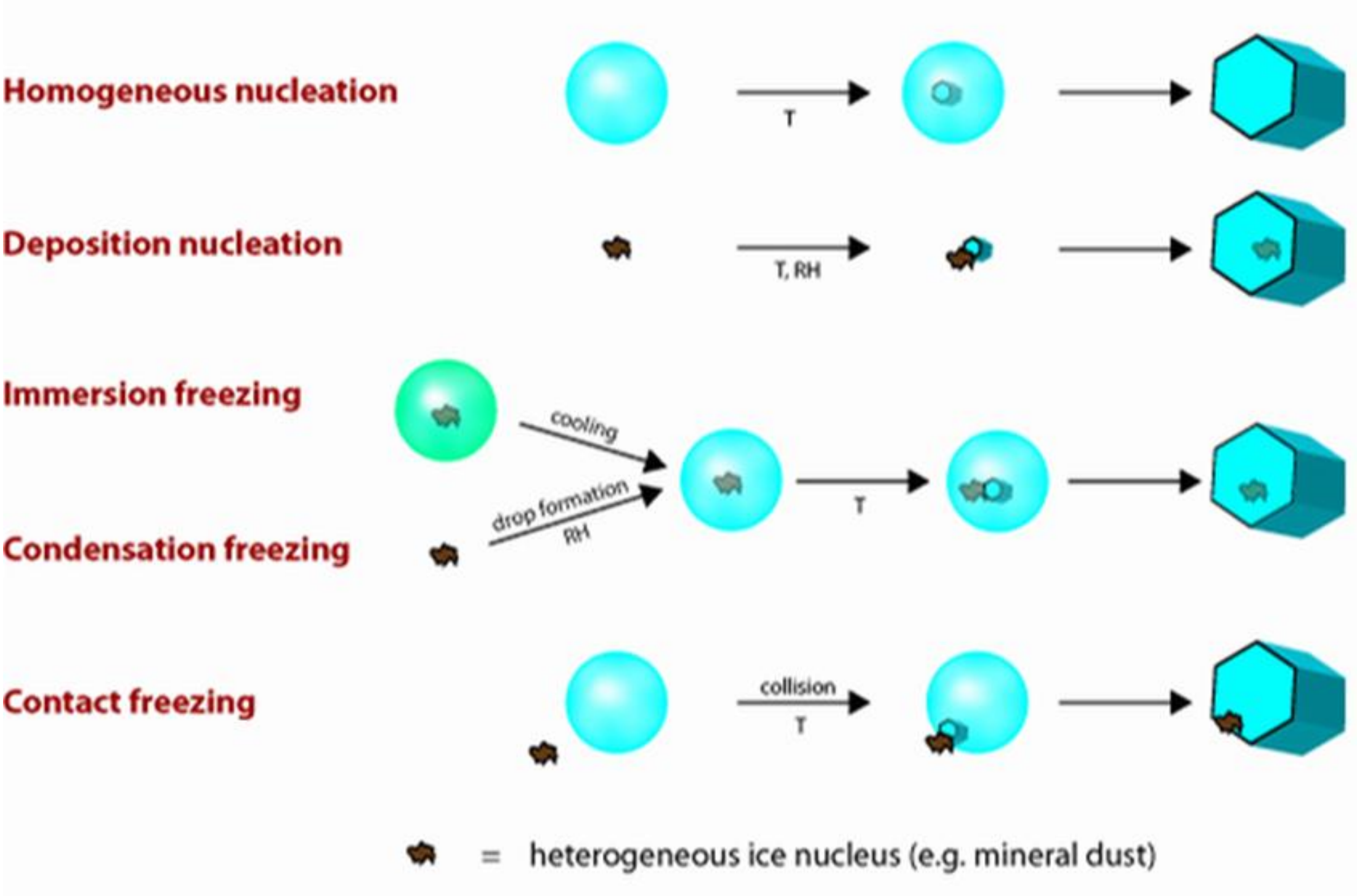
(Immersion, deposition, contact, …)

Also depends on the material and surface area

Wet aerosol particles

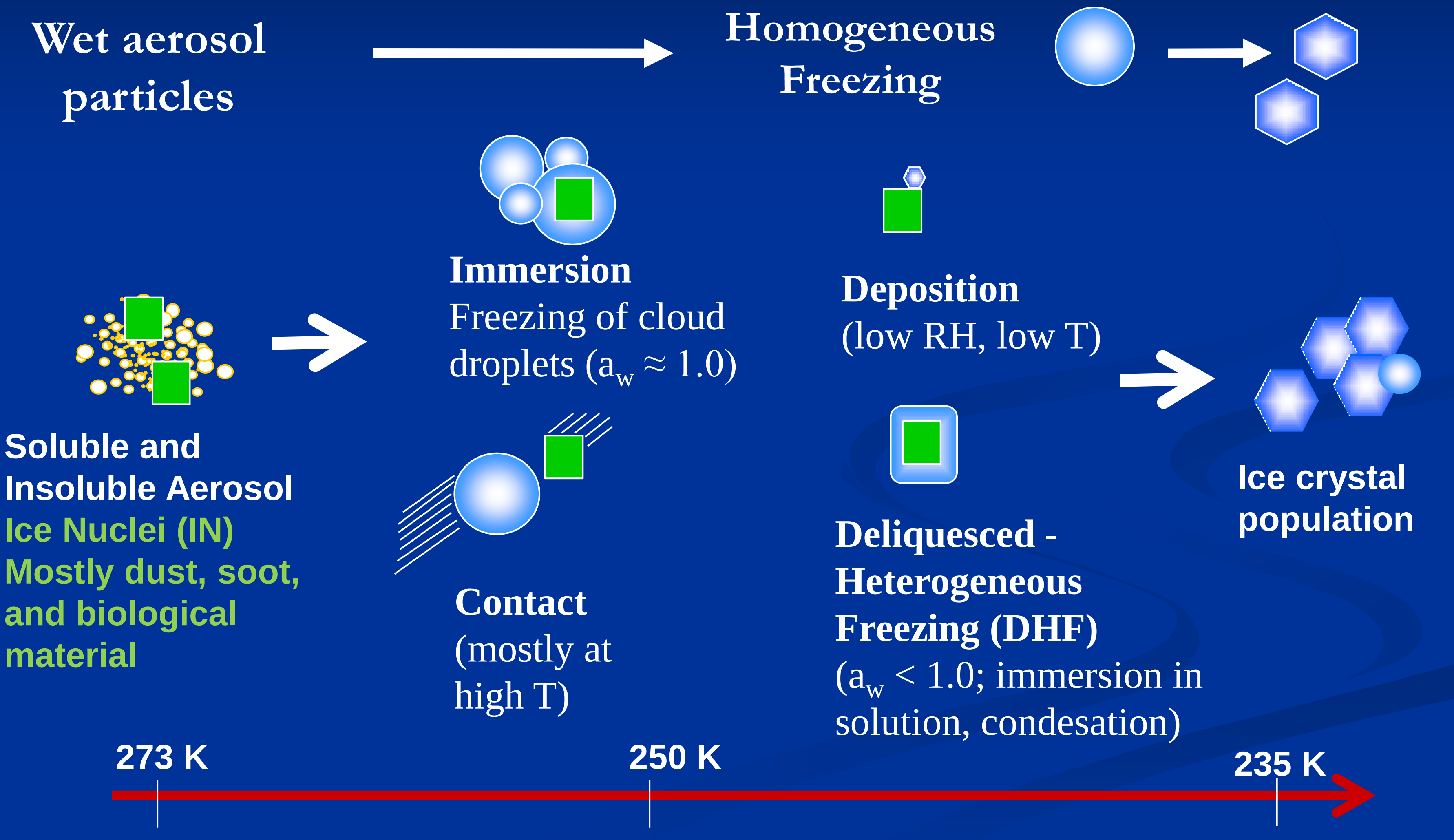
+ Insoluble Material ("Ice Nuclei")

Heterogeneous ice nuclei: freezing modes



Heterogeneous ice nuclei: freezing modes

Multiple mechanisms for ice formation can be active.



Wet aerosol particles

Homogeneous Freezing

Soluble and Insoluble Aerosol

Ice Nuclei (IN)
Mostly dust, soot, and biological material

Immersion
Freezing of cloud droplets ($a_w \approx 1.0$)

Contact
(mostly at high T)

Deposition
(low RH, low T)

Deliquesced - Heterogeneous Freezing (DHF)
($a_w < 1.0$; immersion in solution, condesation)

Ice crystal population

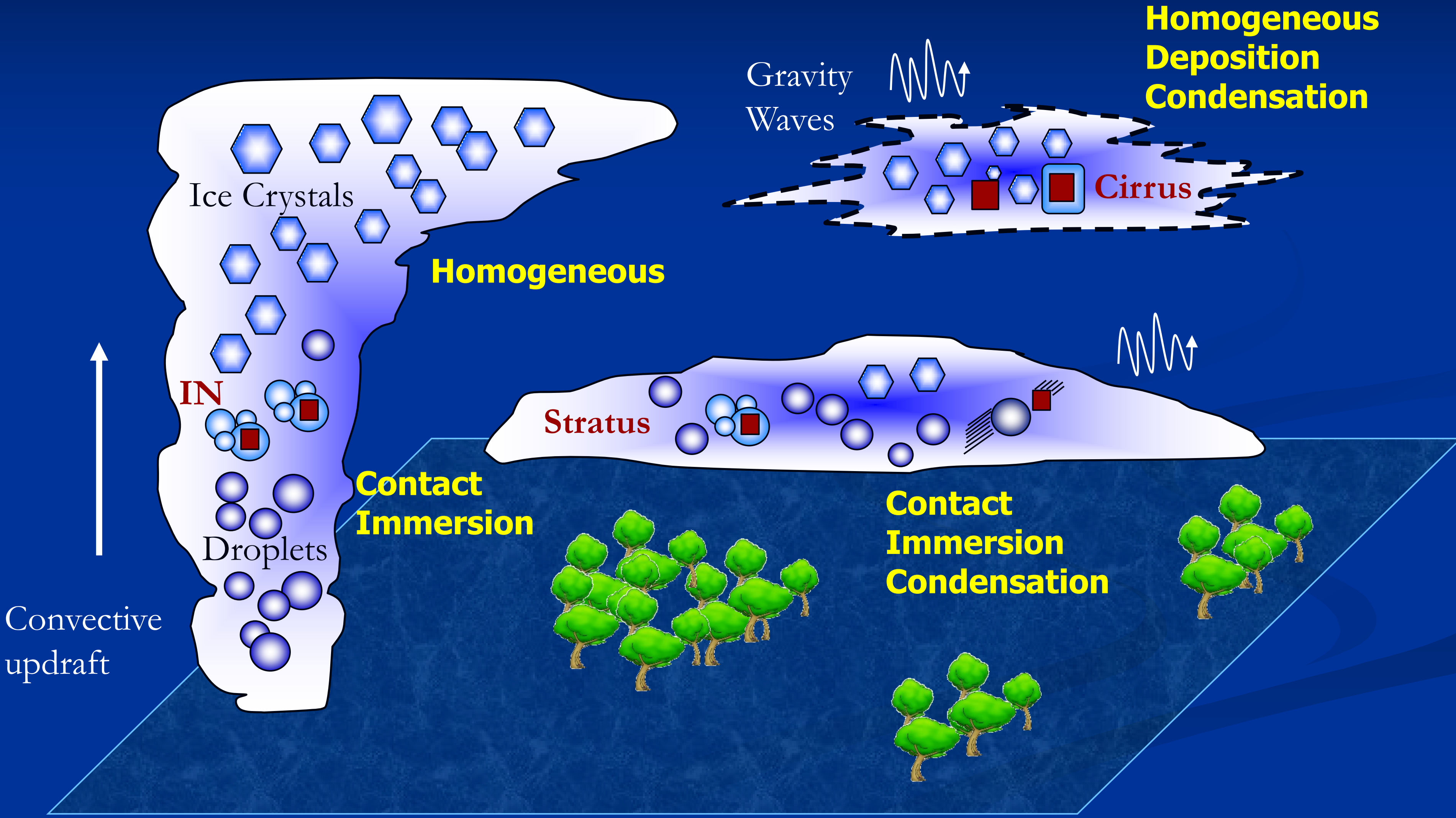
273 K

250 K

235 K

Increasing RH_i, decreasing T

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

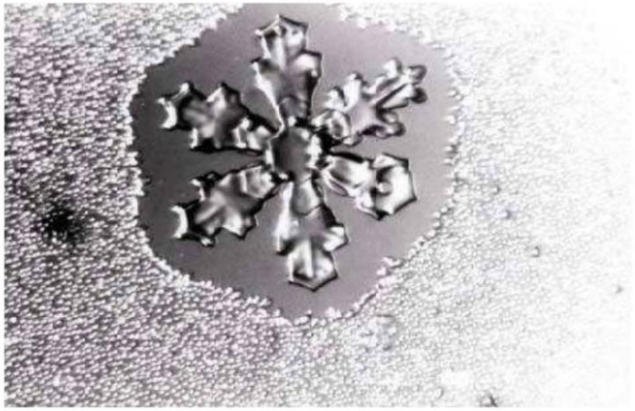


IN vs CCN: implications for clouds

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.

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.

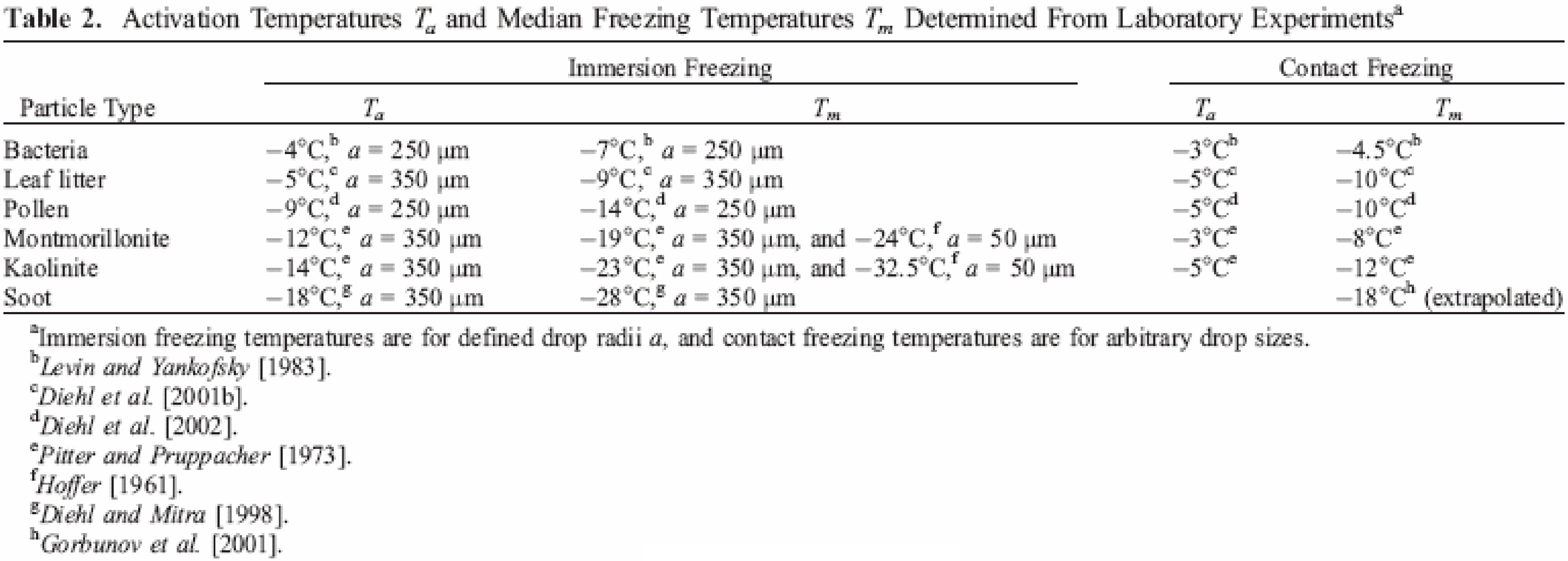


Table 2. Activation Temperatures T_a and Median Freezing Temperatures T_m Determined From Laboratory Experiments[a]

	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[b]	−4.5°C[b]
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].

Measuring INP: Basic Principles

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 clout droplets.

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

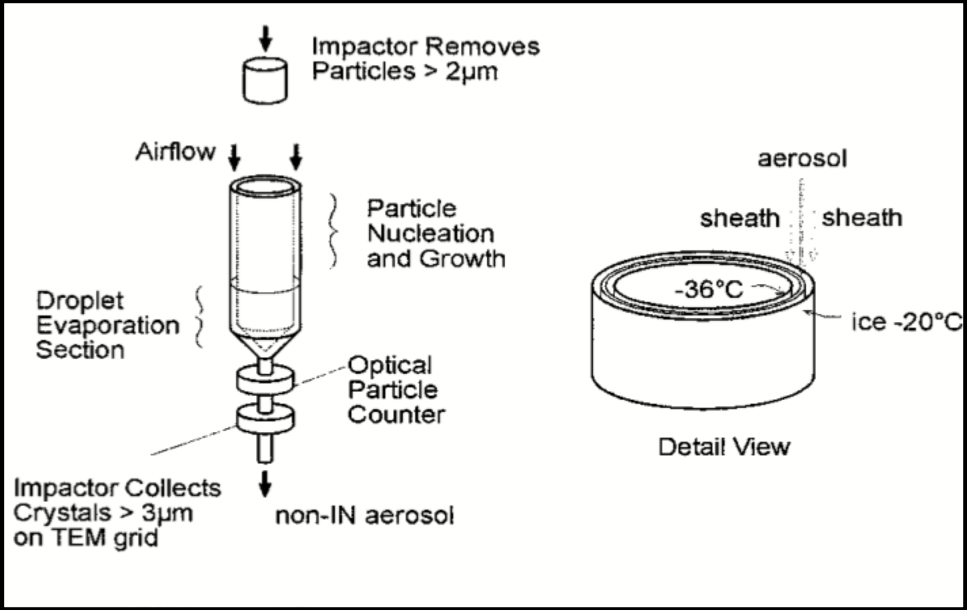
Main challenges:
Technique to invoke nucleation
Low counting statistics (important for warmer T).
Ice crystal detection

- 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

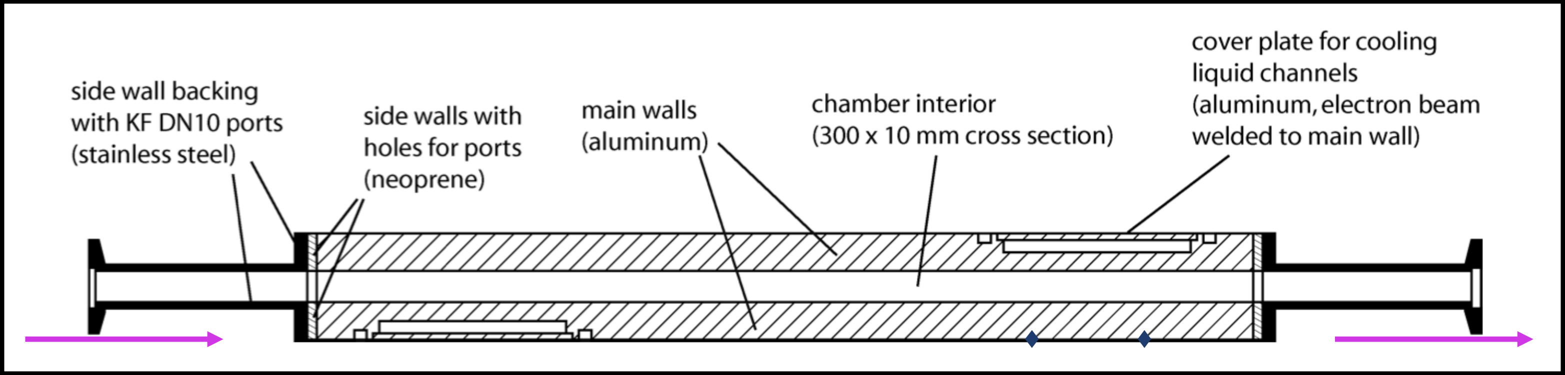
- The chamber consist of two ice- coated concentric cylinders at different temperatures



- In the last third of the chamber the outer cylinder are not ice-coated

- Analyze deposition and immersion modes

ZINC (ZURICH ICE NUCLEATION CHAMBER)



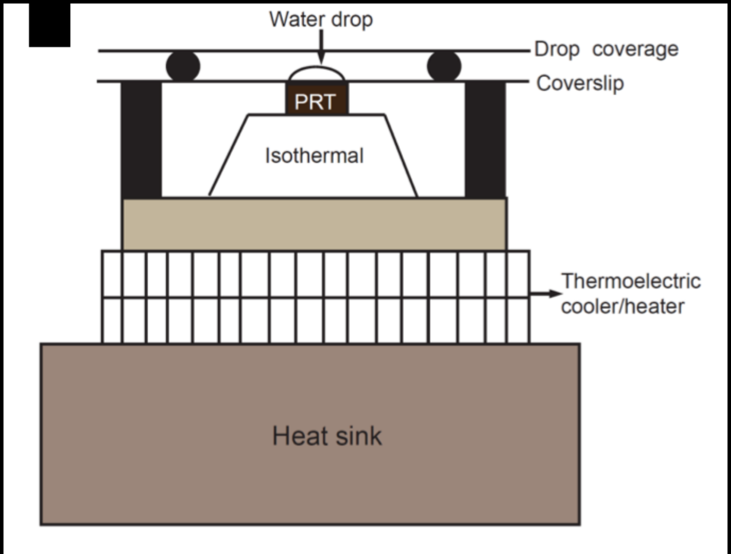
- CFDC type of chamber, which use a flat parallel plate design instead of a concentric cylinder geometry
- Can reach 236 K with ice supersaturations of up to 50%
- Analyze condensation and deposition mode
- Portable version (PINC) also available commercially.

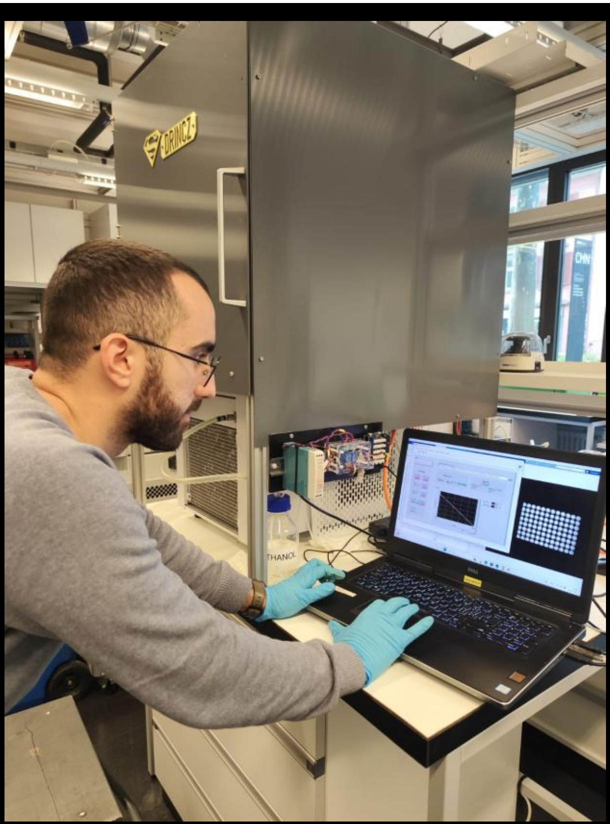
COLD PLATE TECHNIQUE (IMEERSION FREEZING)

- 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

- LAPI's INP mode of measurements

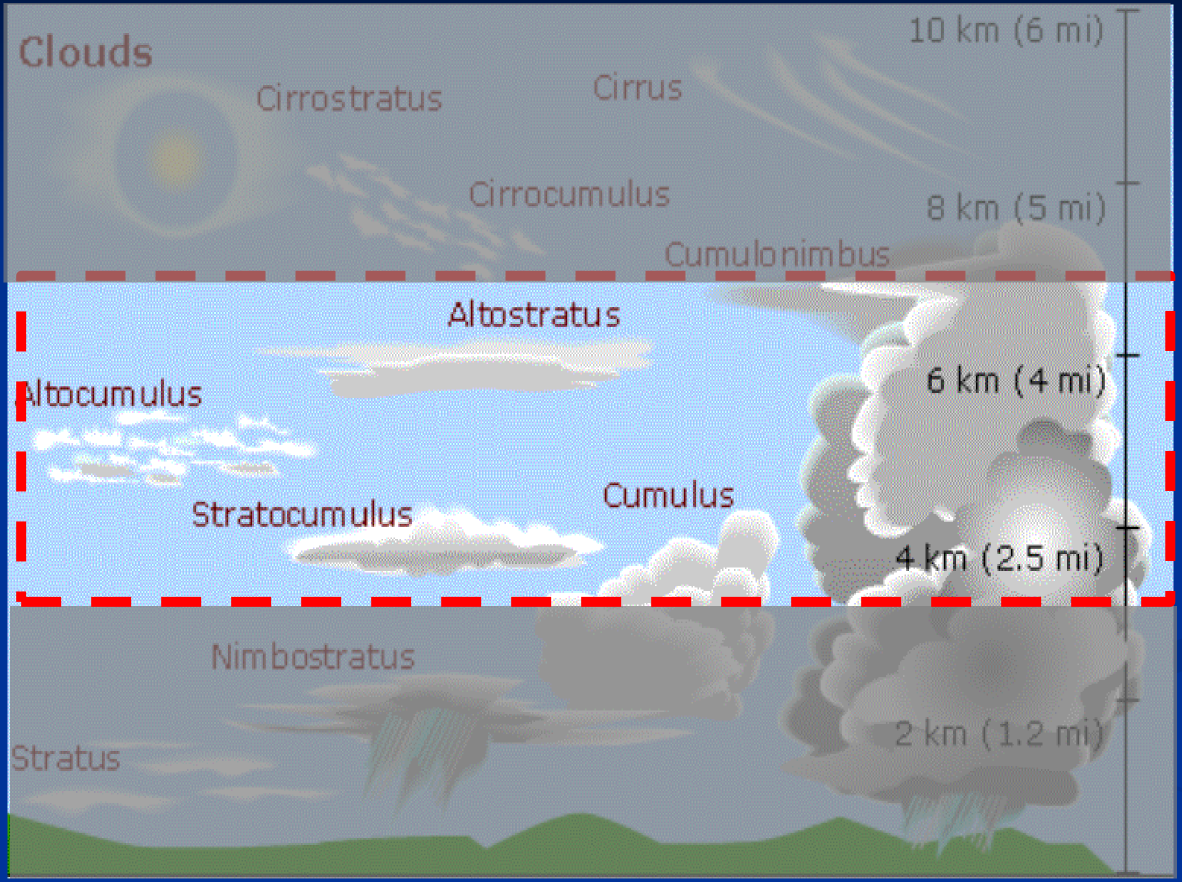




PORTABLE ICE NUCLEATION CHAMBER (IMMERSION/DEPOSITION FREEZING)

- An expansion chamber inside generates supersaturation
- We can create a water droplet first around particles (immersion mode freezing) or directly deposite water vapor (deposition mode).
- Operates from -10 to -60C
- LAPI's INP mode of measurements

Mixed phase clouds: "final frontier"

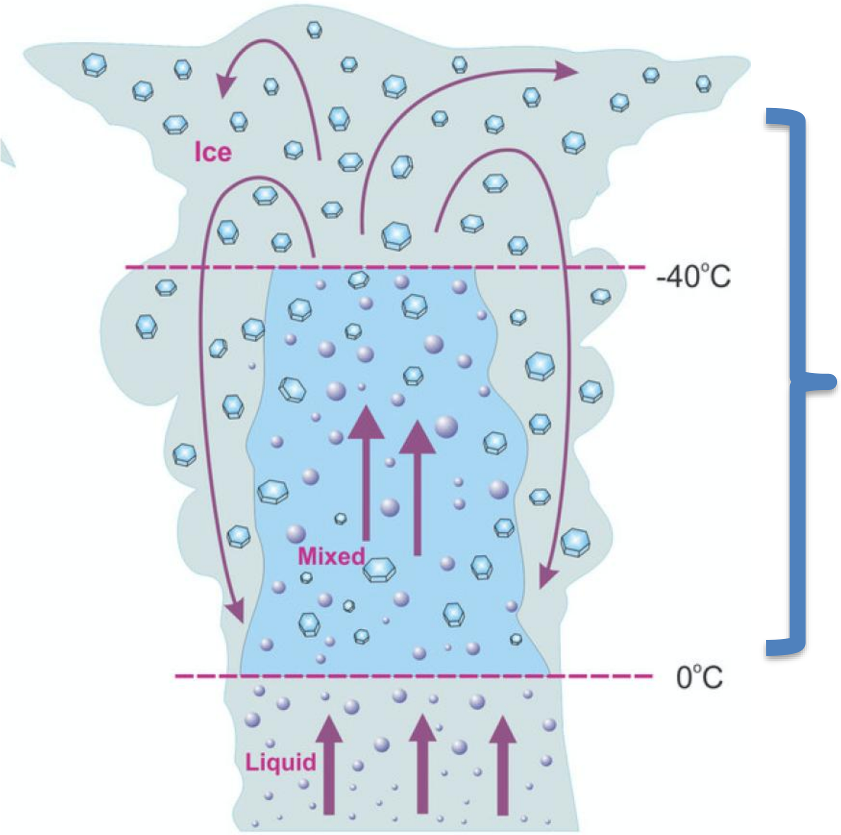


- **Ice (cold) clouds**: Ice crystals, T<235 K. Warm climate
- **Mixed Phase clouds**: Liquid droplets & ice, 235K < T < 273K Warm/cool climate
- **Liquid (warm) clouds**: Liquid droplets T >273 K Cool climate

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

The international journal of science / 13 January 2022

nature

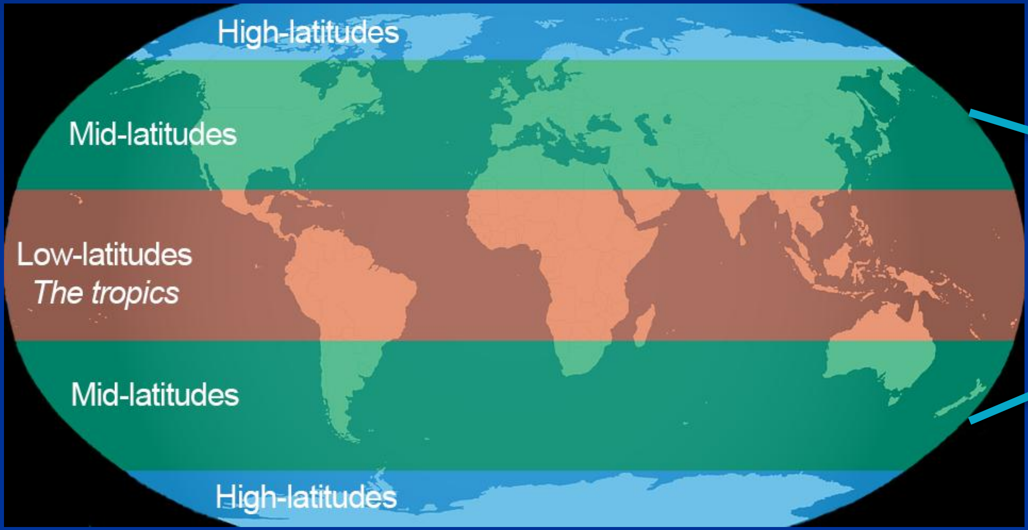
RAIN STOPS GAIN

Excessive wet weather reduces economic growth rate

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

Greek “Snowmageddon”, 28 Jan, 2022

Liquid+ice ("mixed-phase") clouds Are very important for climate



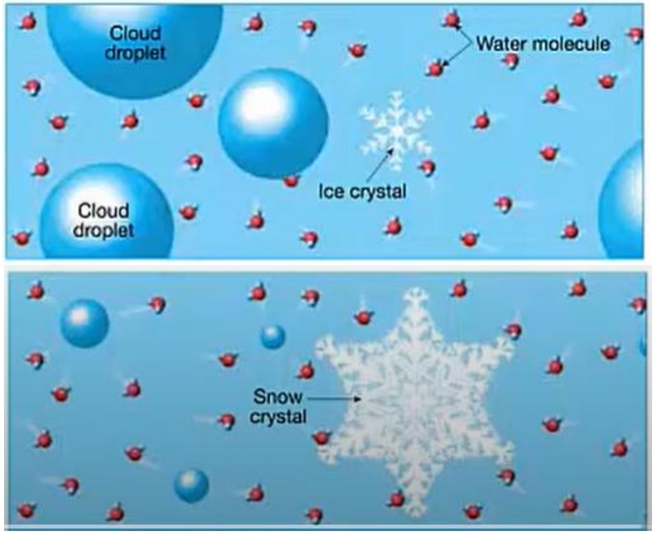
Field and Heymsfield, 2015
Mülmenstädt et al. 2015

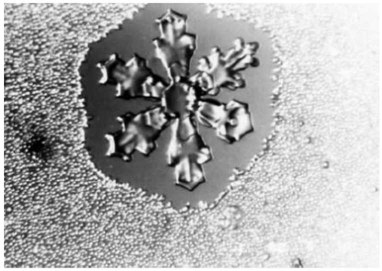
30-50% of precipitation occurs from the ice phase

> *"…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



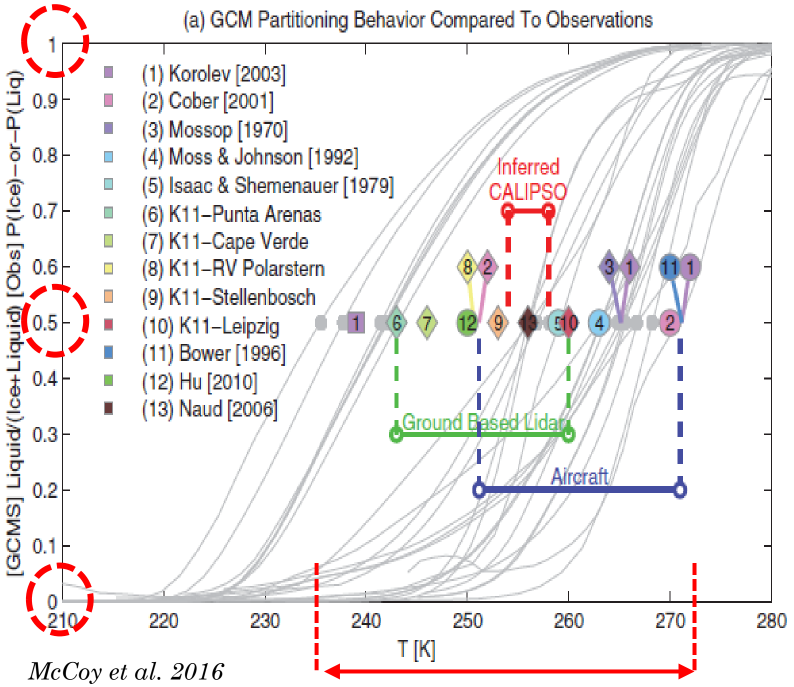


Wegener-Bergeron-Findeisen process (WBF)

Pure liquid cloud

50% liquid 50% ice

Pure ice cloud



McCoy et al. 2016

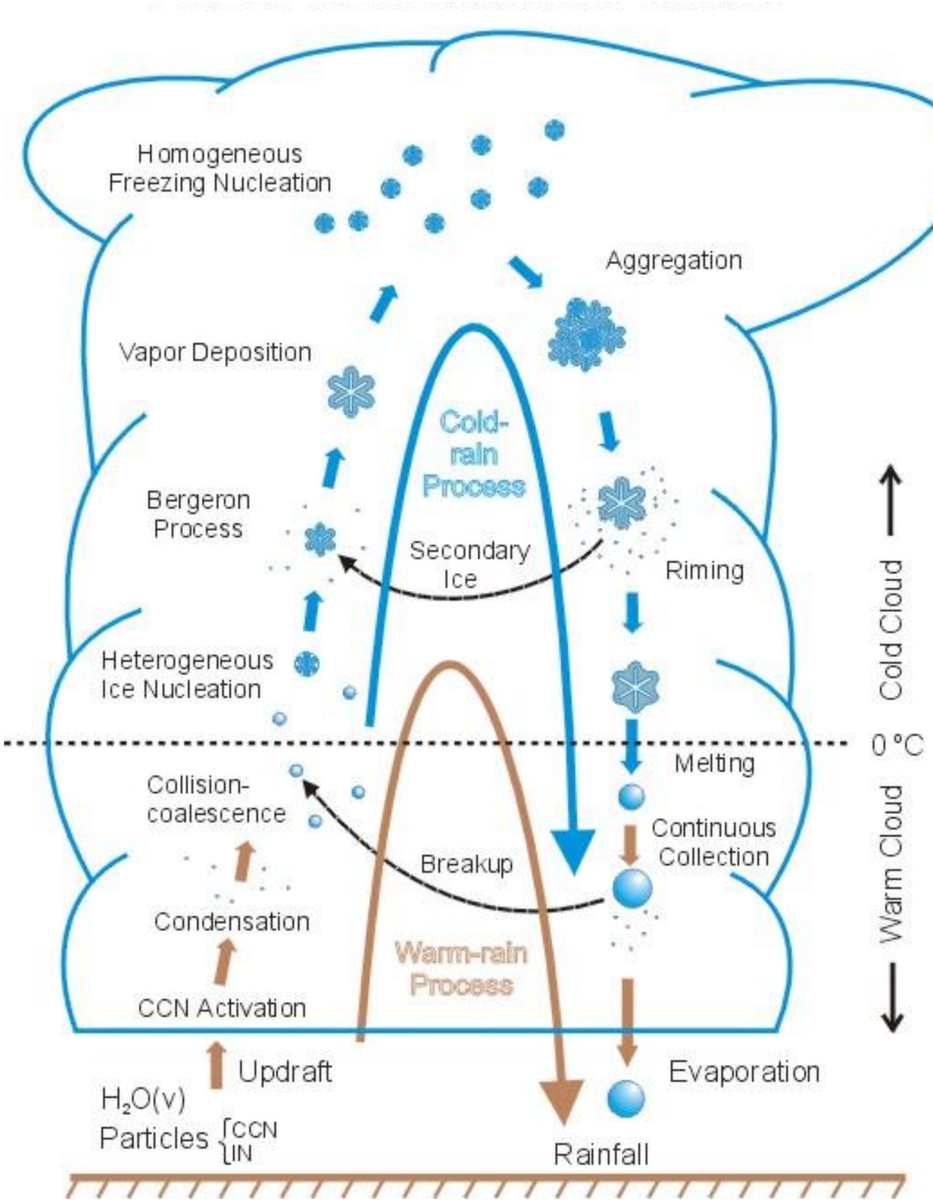
mixed-phase temperature range

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

Ice production/growth interplay in clouds

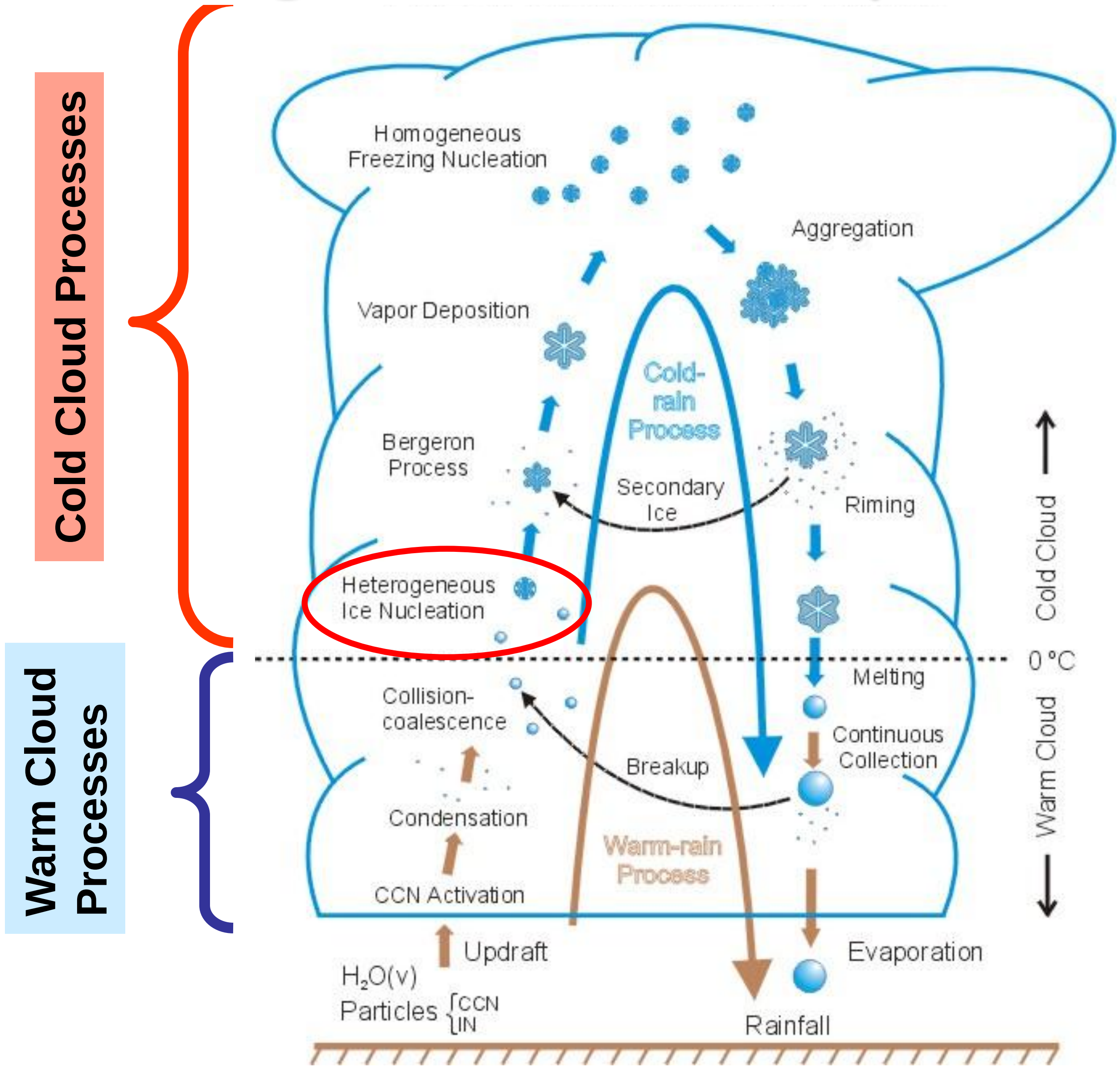
Cold Cloud Processes

Warm Cloud Processes



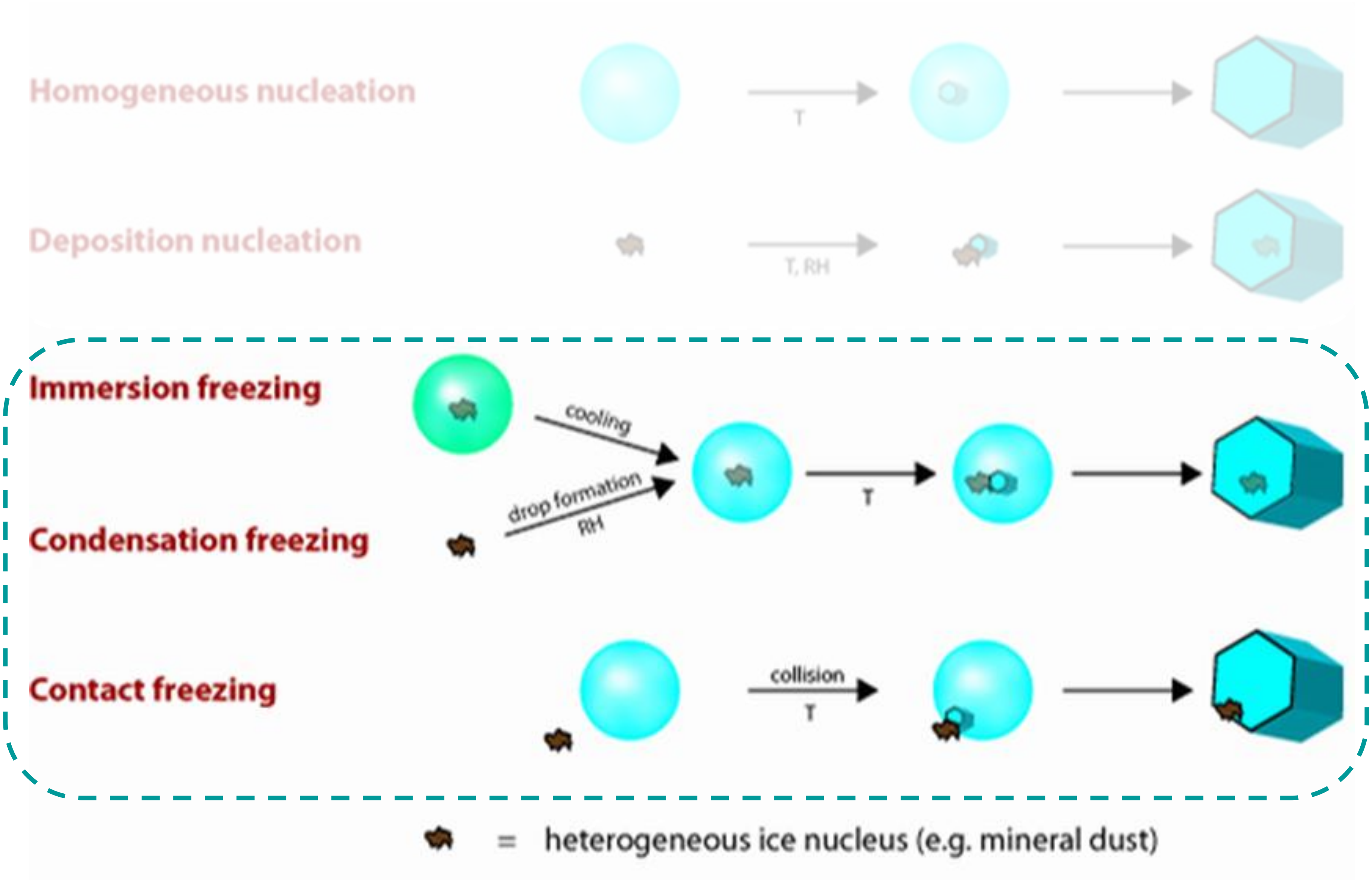
Platnick

The ice grows at the expense of drops

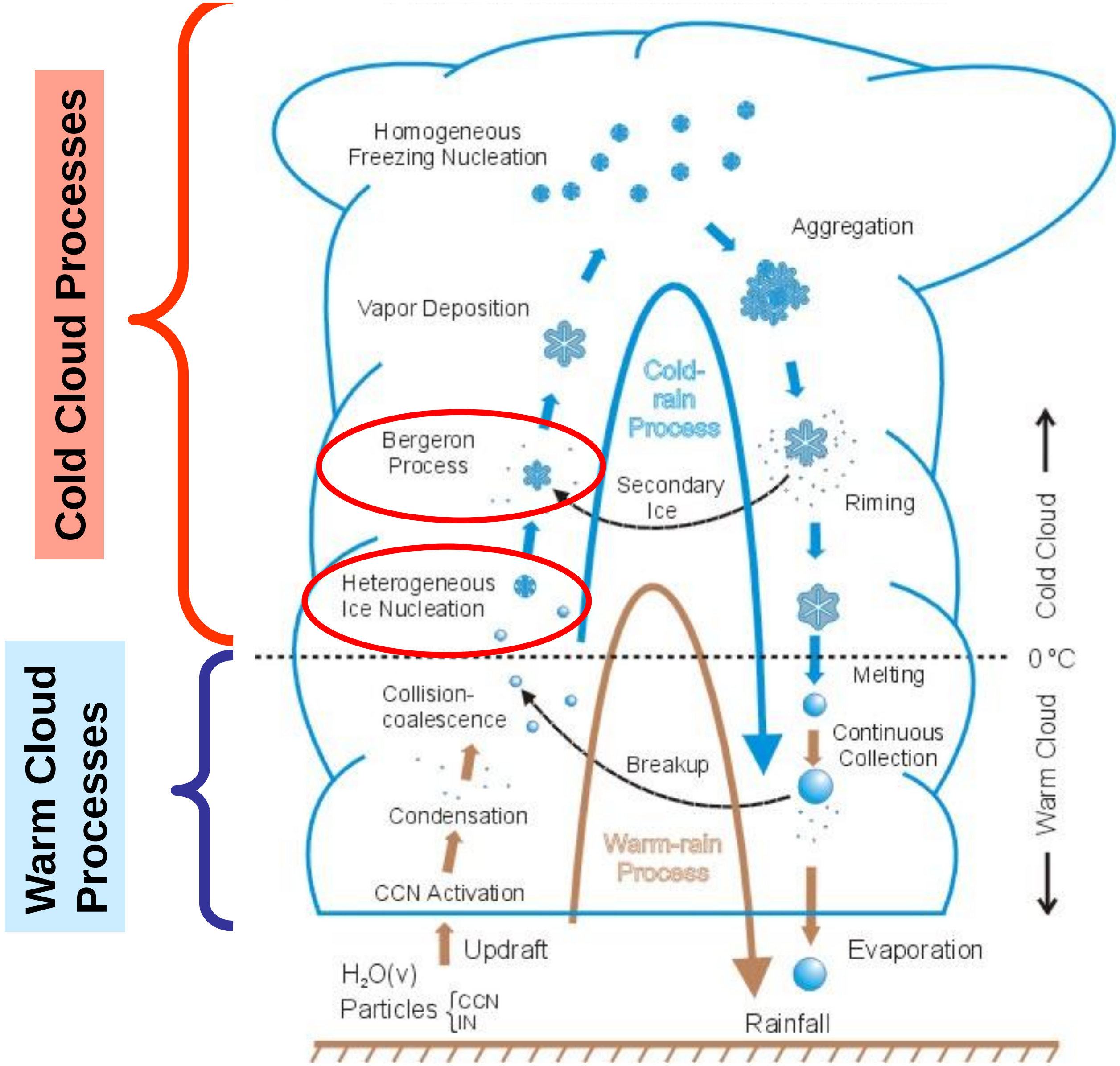


Platnick

Primary ice production mechanisms



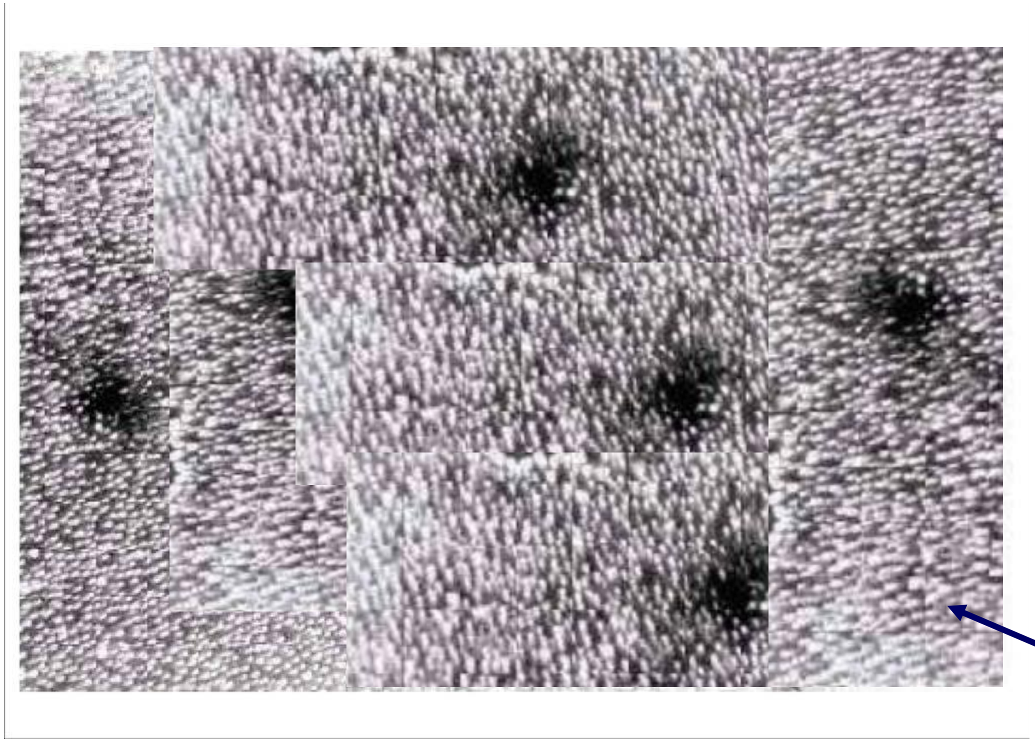
The ice grows at the expense of drops



Platnick

Bergeron-Findesien process

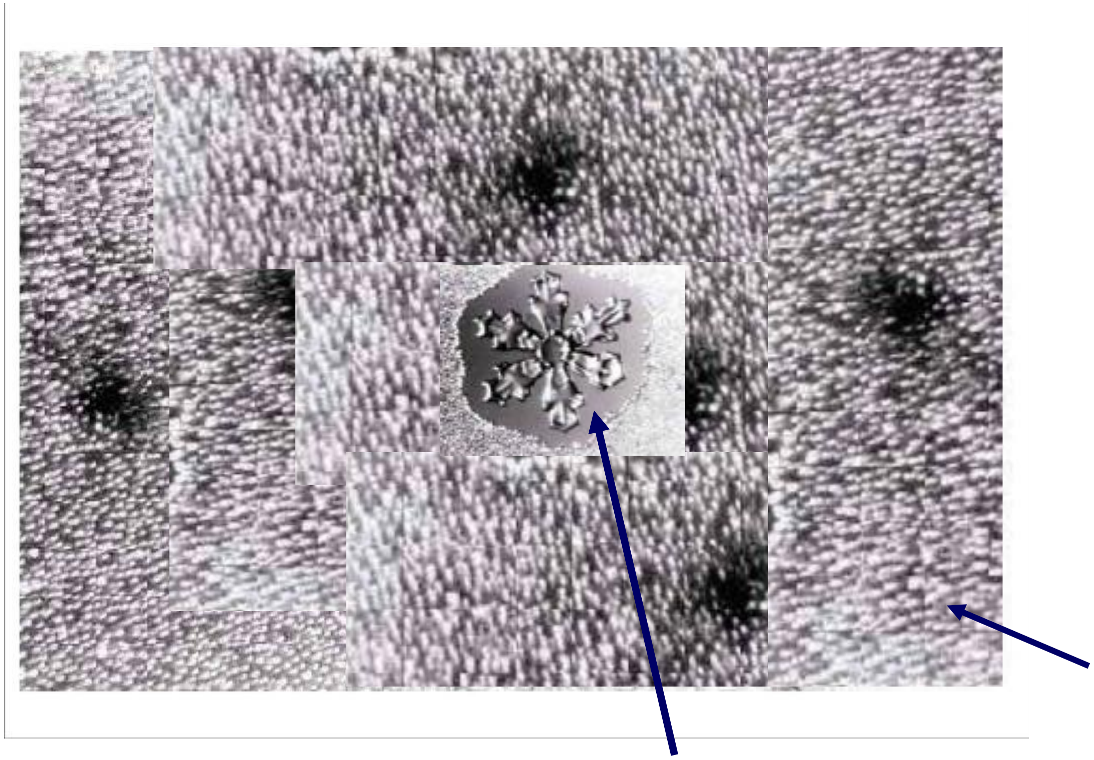
Critical for the microphysical evolution of mixed-phase clouds



Droplets on a plate

Bergeron-Findesien process

Critical for the microphysical evolution of mixed-phase clouds



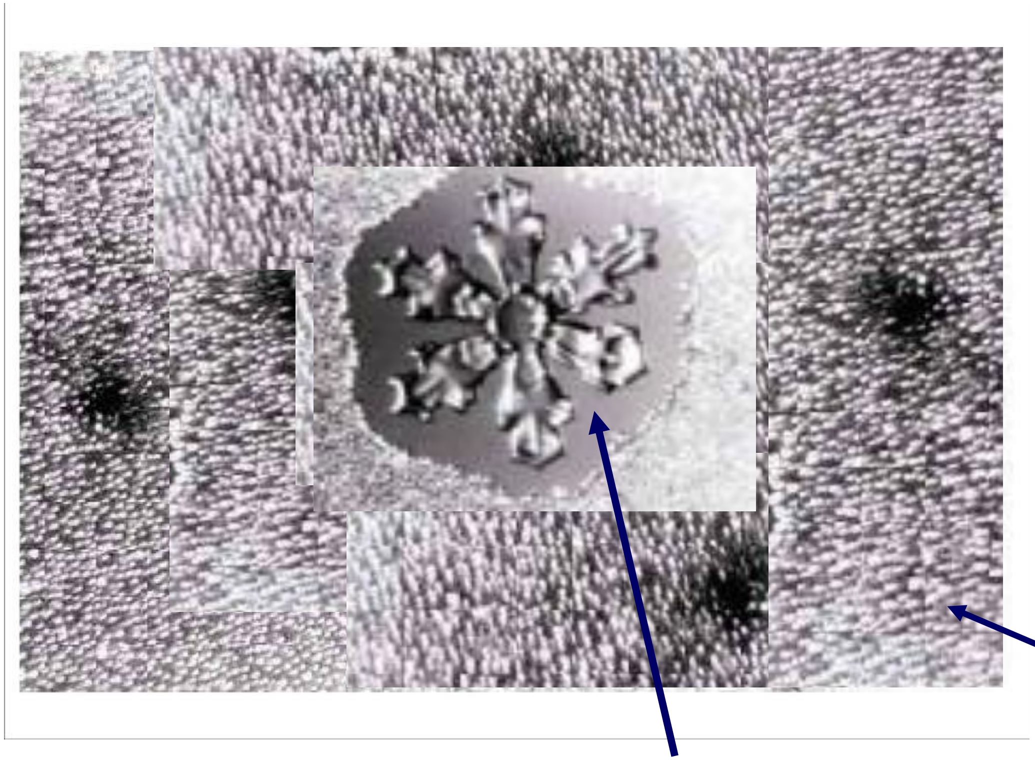
Droplets on a plate

Crystal forms

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.



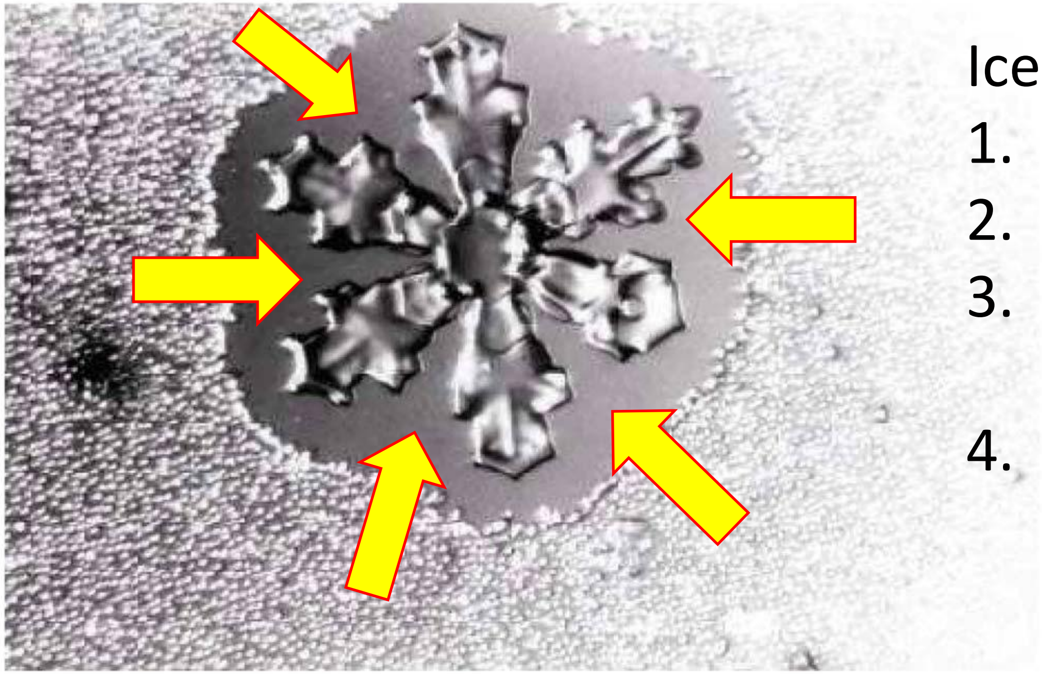
Droplets on a plate

Crystal grows

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.



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

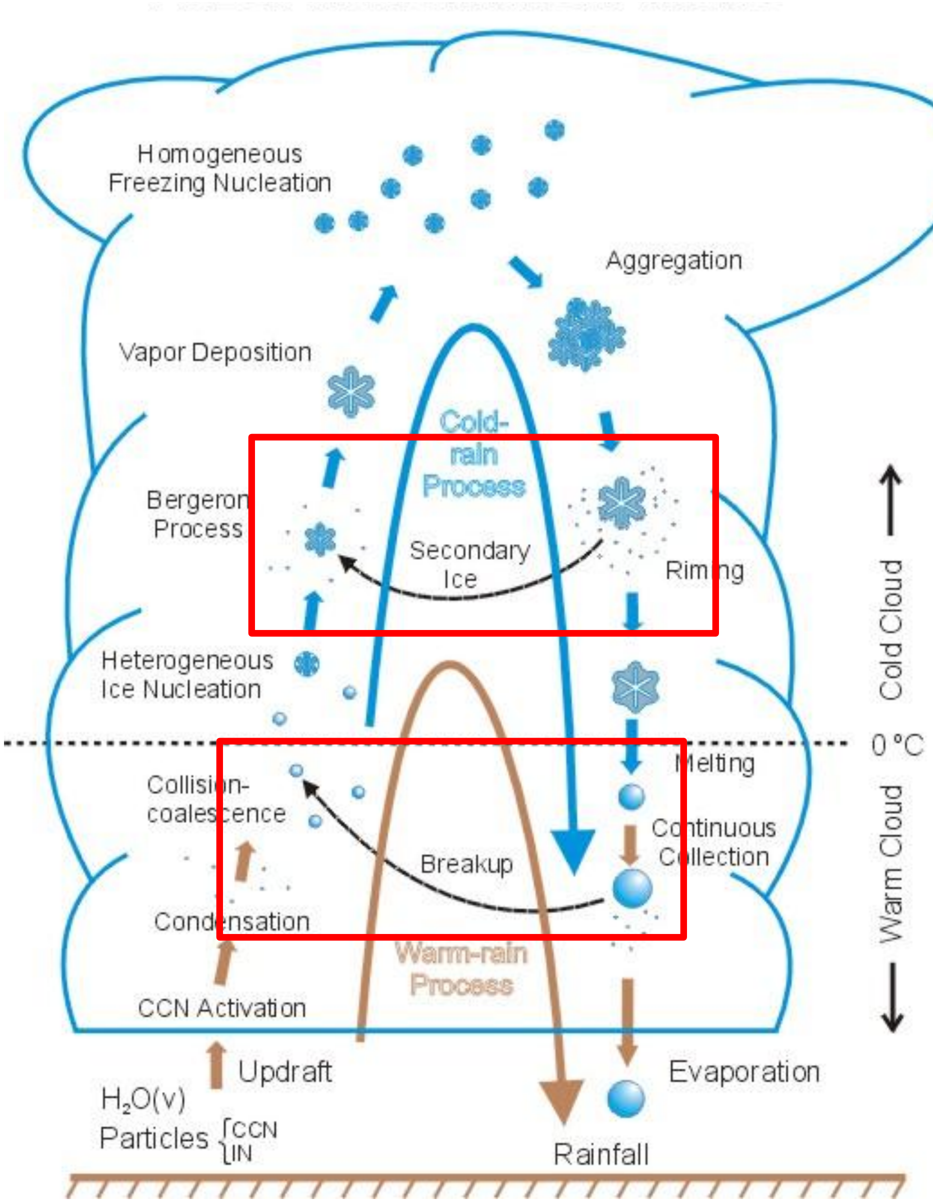
Latent heat release important for dynamical evolution of clouds.

Shifts in particle size affects radiation and precipitation of cloud.

Where secondary ice production “lies”

Cold Cloud Processes

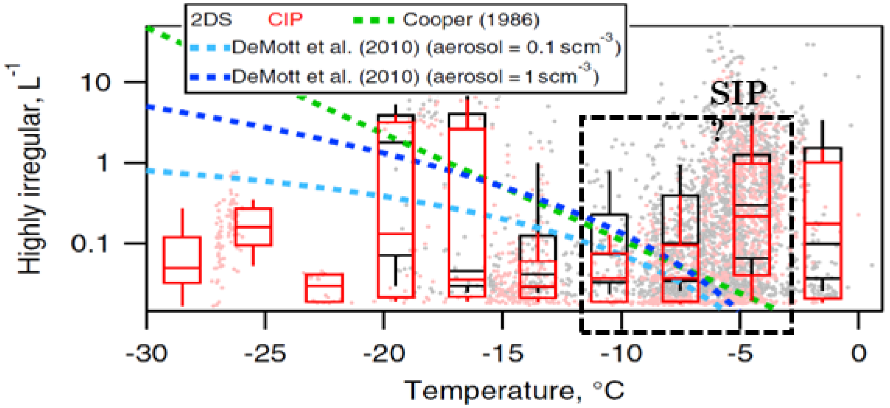
Warm Cloud Processes



Platnick

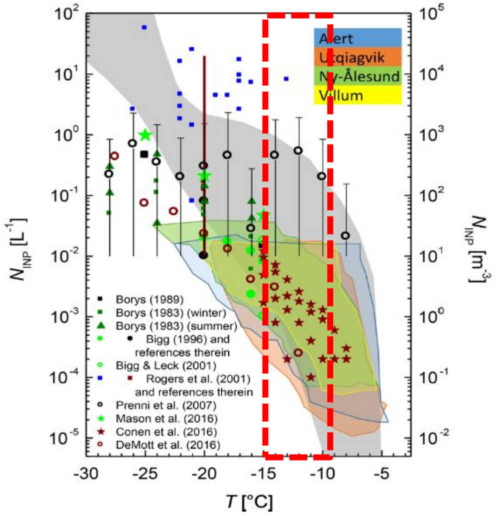
Measured Ice Crystal Number is much higher than pre-cloud INPs!

- ✓ 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



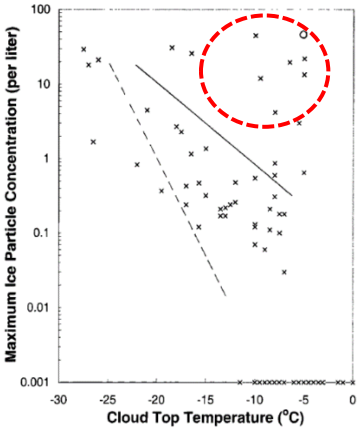
Antarctic

O'Shea et al. 2017



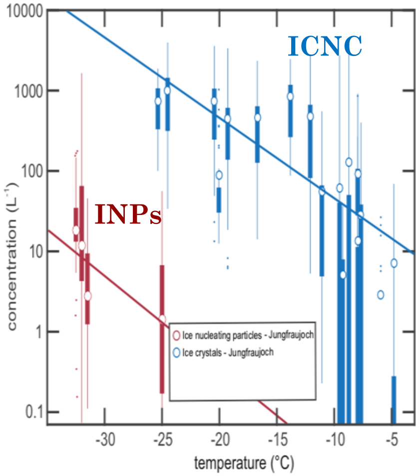
Wex et al. 2019

Arctic

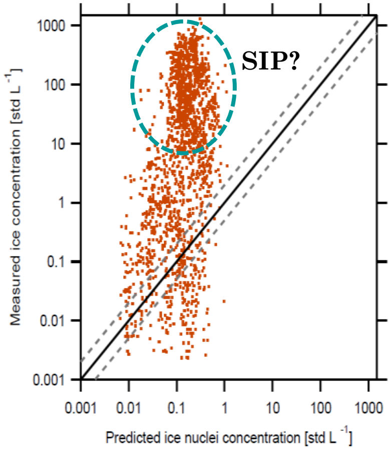


Ragno & Hobbs 2001

Alps



Beck et al. 2018



Lloyd et al. 2015

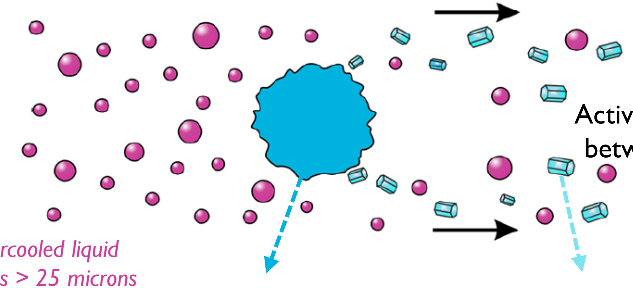
Courtesy: P.Georgakaki

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

I. Rime Splintering or the Hallett-Mossop process (HM)



Supercooled liquid droplets > 25 microns

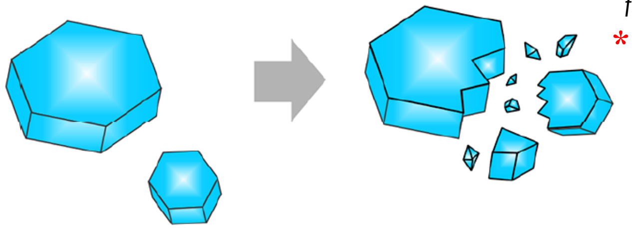
Large ice particle > 0.5mm

ejected ice-splinters

Korolev et al., 2020

Active temperature range between -8 ˚C and -3˚C

II. Ice-ice collisional break-up (BR)



Numerous ice fragments

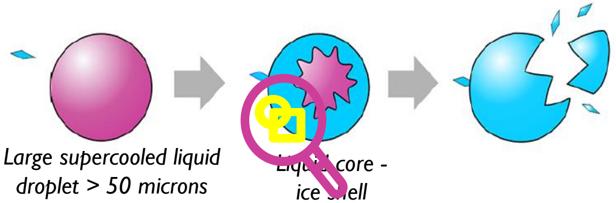
* Max ice splinter production rate at ∽ **-15 ˚C**

IV. Sublimational break-up (SUBBR)

Dendritic snow (-10<T<-20 ˚C) or rimed particles

SIP particles separate from parent ice particle when ice bridge sublimates (Relative humidity w.r.t ice < 100%)

III. Droplet Shattering during freezing (DS)

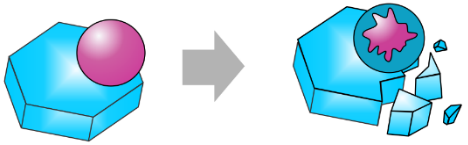


Large supercooled liquid droplet > 50 microns

Liquid core - ice shell

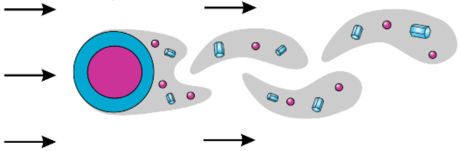
Ice splinters created upon internal pressure build-up

V. Ice fragmentation during thermal shock



Fragmentation caused due to latent heat release from the freezing droplet

VI. Activation of INPs in transient supersaturation

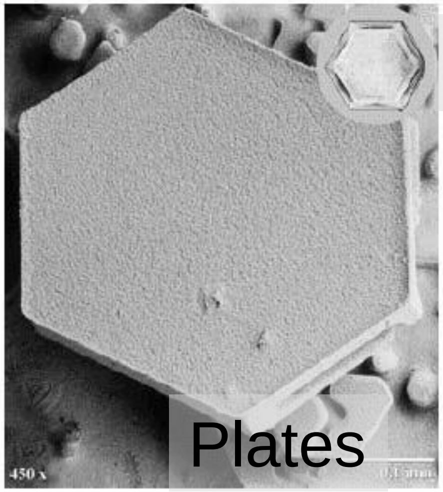


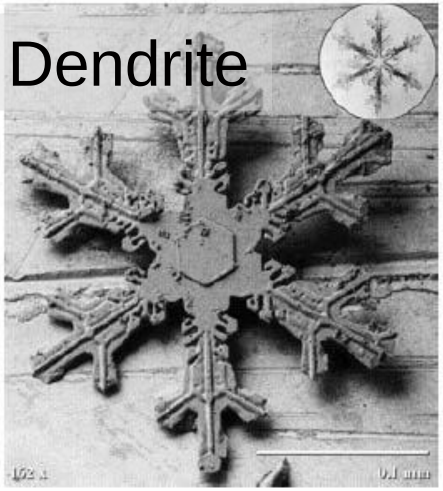
Frozen hydrometeor falling within the cloud

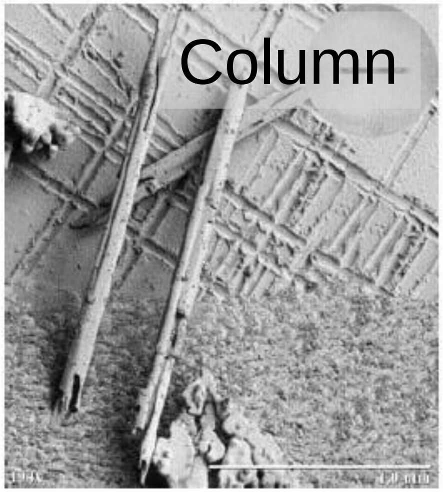
Regions of high transient supersaturation

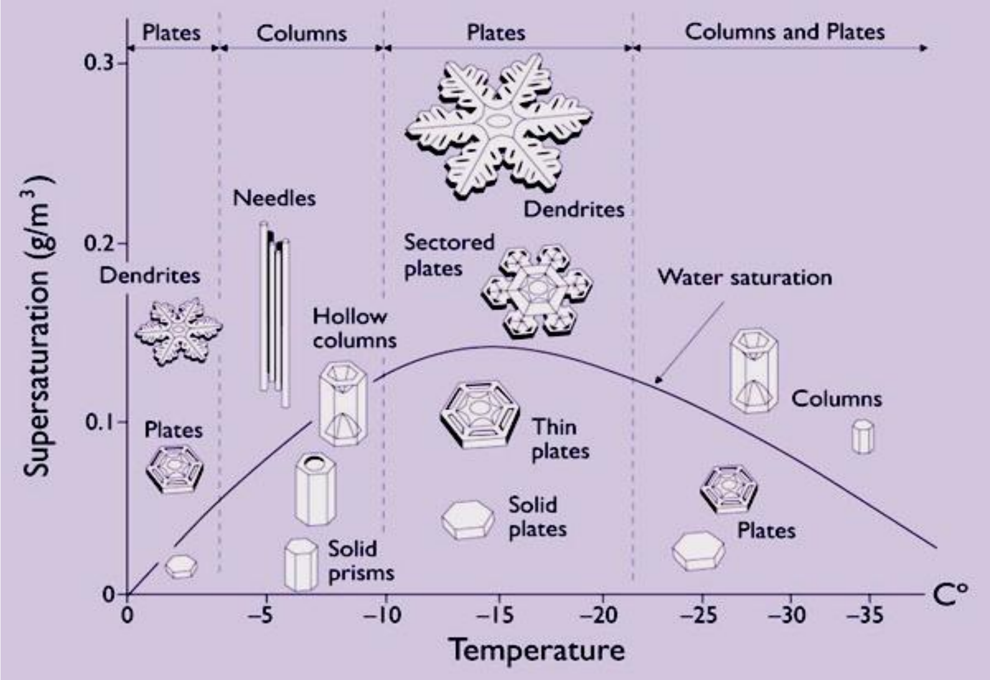
Courtesy: P.Georgakaki

“Pristine” ice crystals – before riming





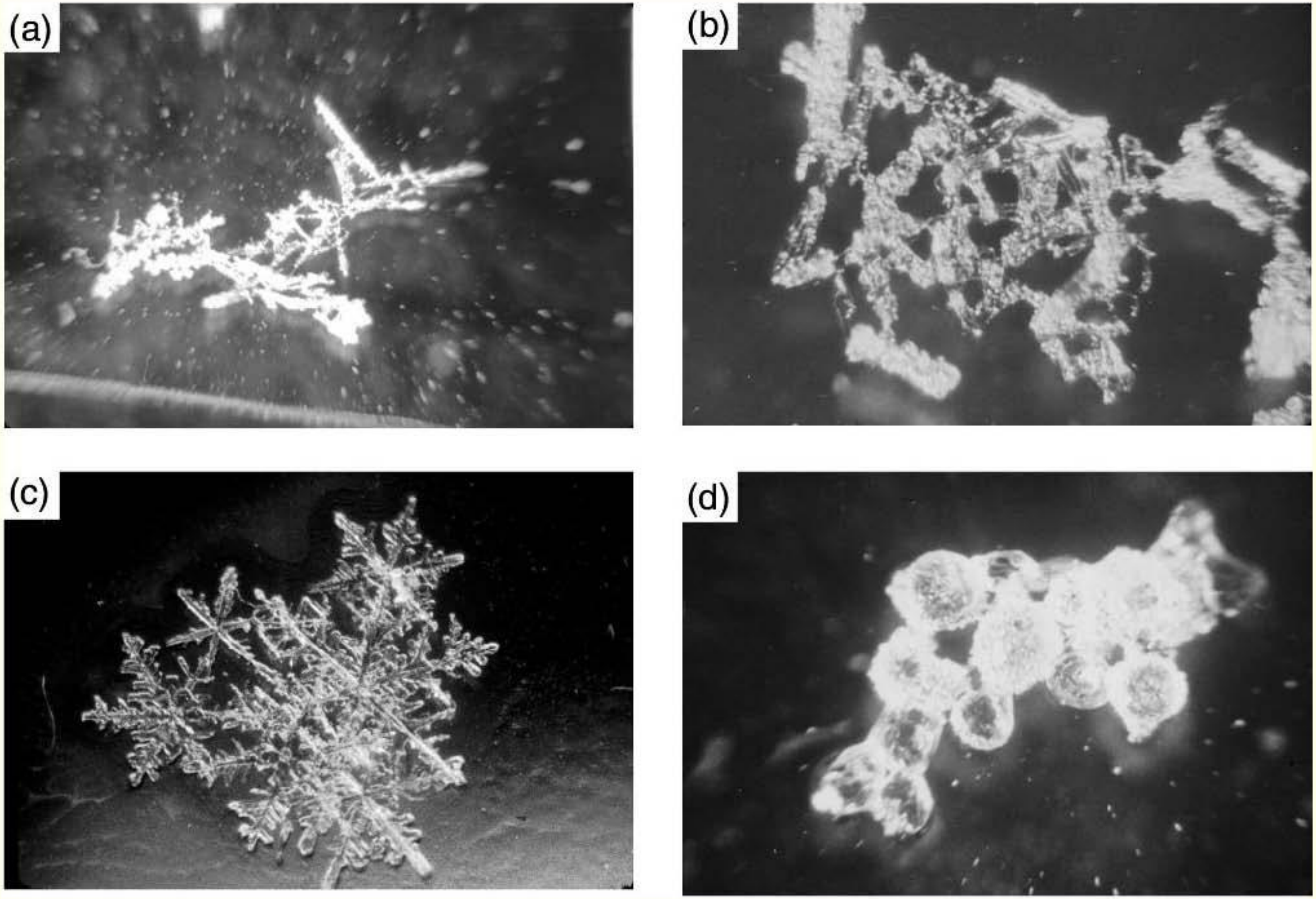




Primary ice production

Growth via water vapor deposition only.

“Pristine” ice crystals – after riming



Pioneering work of Hallett & Mossop

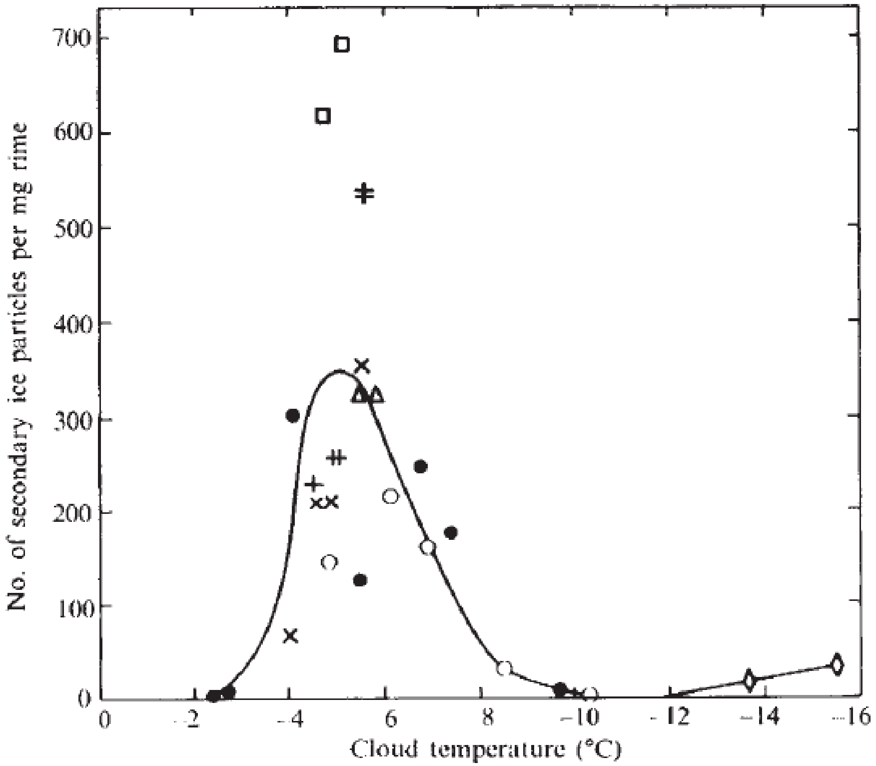


FIG. 2 Production of secondary ice particles by riming as a function of temperature at a target velocity of 2.7 m s^{-1}. Different symbols indicate different days. The curve was drawn by averaging the points over narrow temperature intervals.

Hallett and Mossop 1974

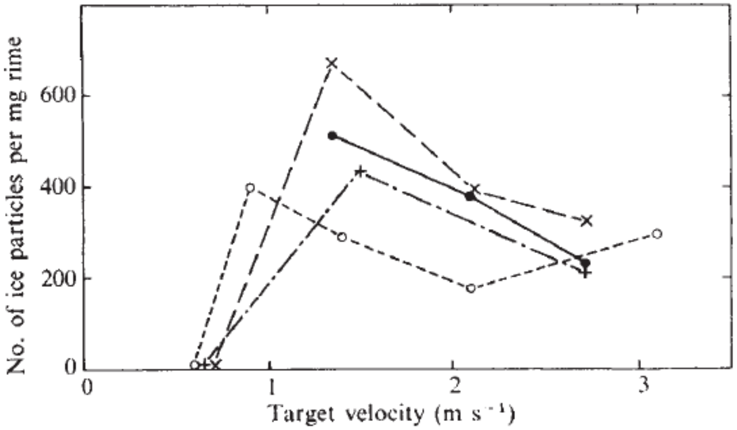
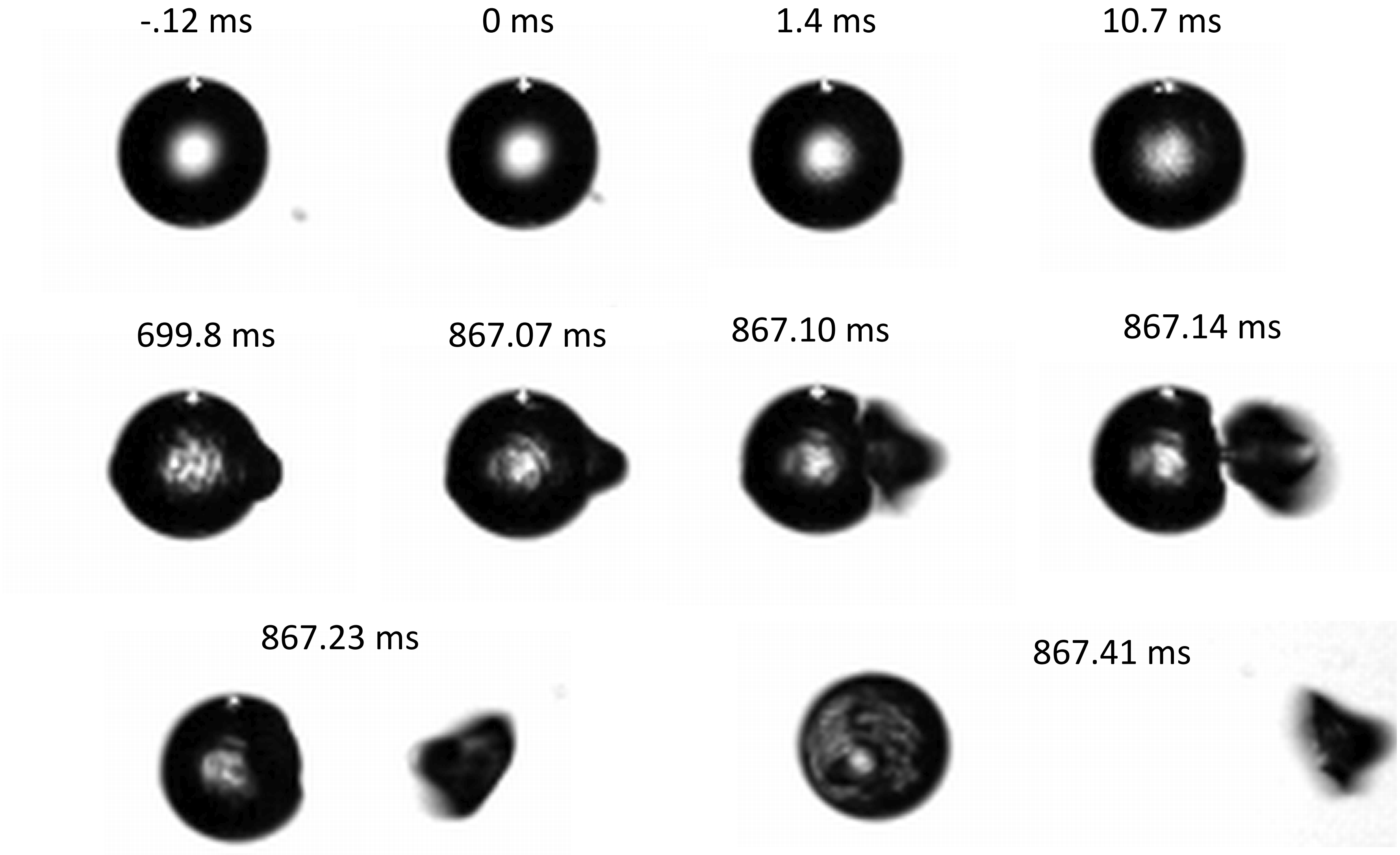


FIG. 3 Production of secondary ice particles by riming as a function of target velocity at a temperature of about −5° C. ●, November 5, 1973; −4.5° C. ×, November 9, 1973; −5.5° C. ○, December 3, 1973; −5.7° C. +, December 5, 1973; −5.2° C.

- 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.

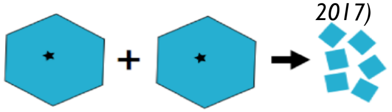
Other mechanisms – Droplet Explosions



From Leisner et al.

Including secondary ice production in models

Collisional break-up *(Phillips et al., 2017)*



Number of fragments per collision

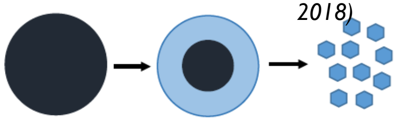
asperity–fragility coefficient Collisional kinetic energy

$$F_{BR} = \boldsymbol{a}\boldsymbol{A}\left(1 - exp\left\{-\left[\frac{\boldsymbol{C}\ \boldsymbol{K_0}}{\boldsymbol{a}\boldsymbol{A}}\right]^{\gamma}\right\}\right) \quad , \quad \boldsymbol{a} = \pi \mathrm{D}^2$$

Surface area

number density of breakable asperities

Droplet shattering *(Phillips et al., 2018)*



Both tiny and big splinters generated

$$F_{DS} = \boldsymbol{\Xi}(\mathrm{D})\boldsymbol{\Omega}(\mathrm{T})\left[\frac{\zeta\eta^2}{(\mathrm{T}-\mathrm{T0})^2} + \beta\mathrm{T}\right]$$

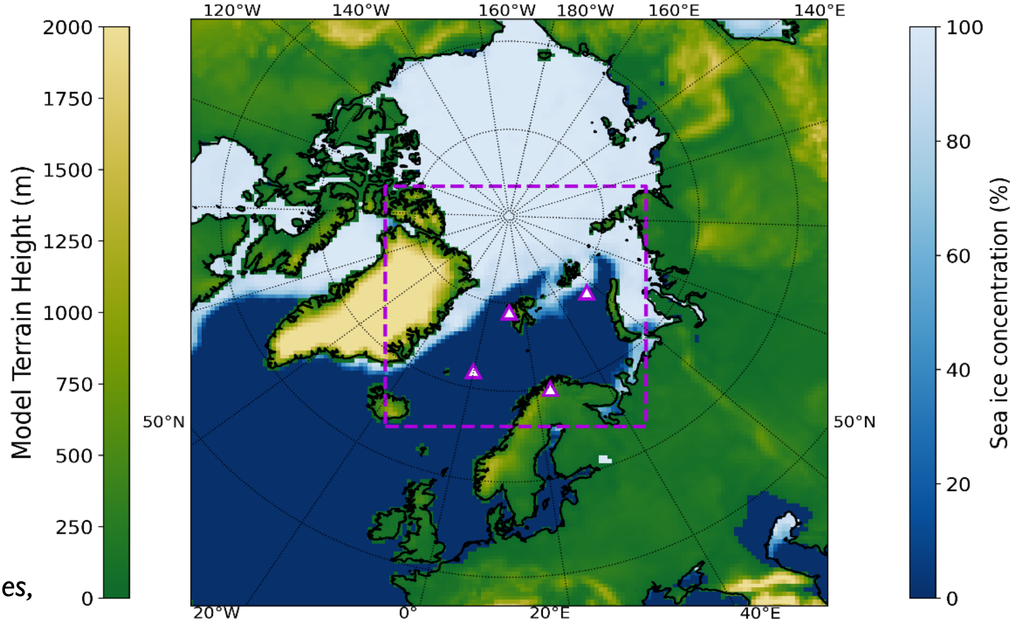
Shattering probability

Freezing probability

- ✓ Complex parameterizations
- ✓ Models need to consider multiple liquid/ice species and interactions among them

Mesoscale model simulations

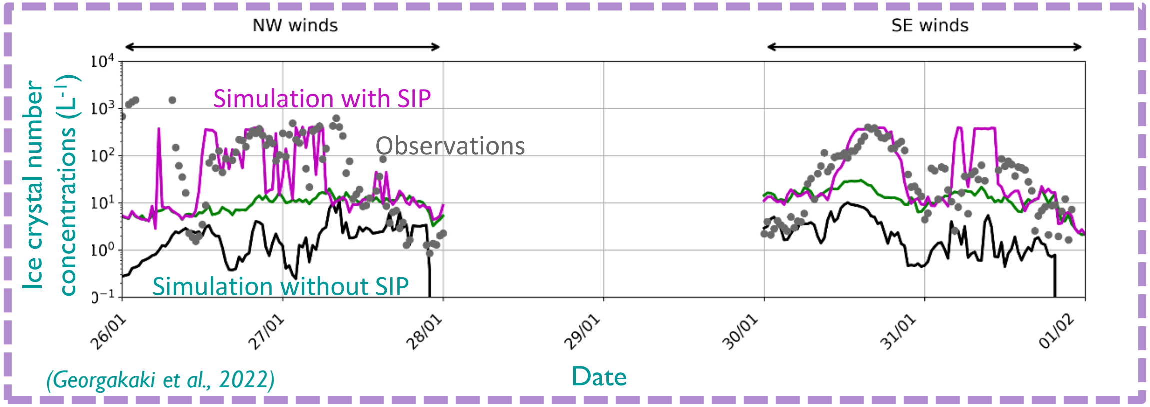
2-year (**2016-2017**) regional climate simulations over the **pan-Arctic region** with the updated version of WRF with detailed microphysics



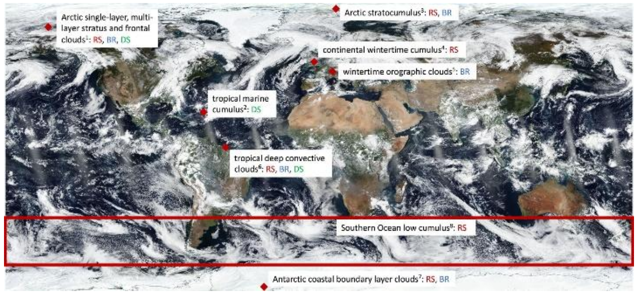
(e.g., Georgakaki & Nenes, 2024)

Secondary ice production mechanisms: our research points its always important!

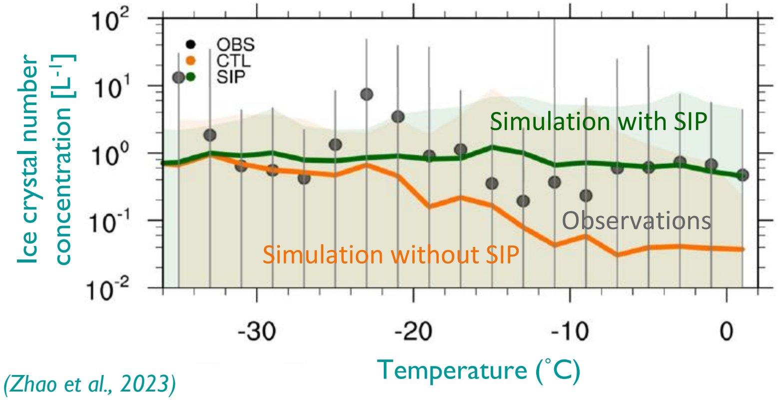
Orographic clouds



(Georgakaki et al., 2022)



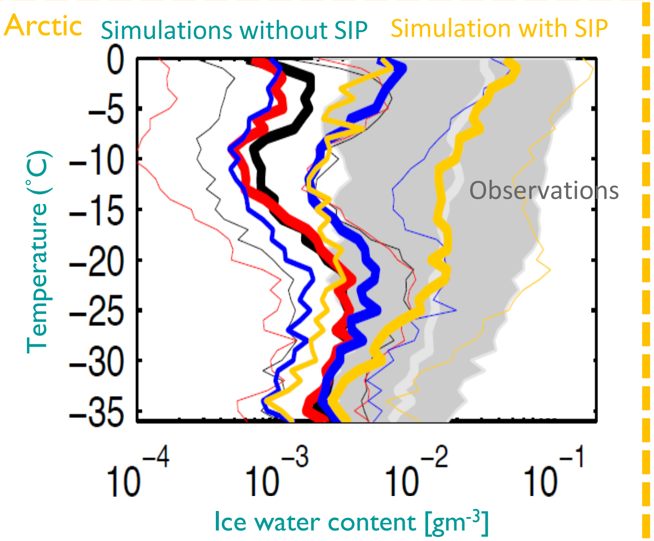
Southern Ocean



(Zhao et al., 2023)

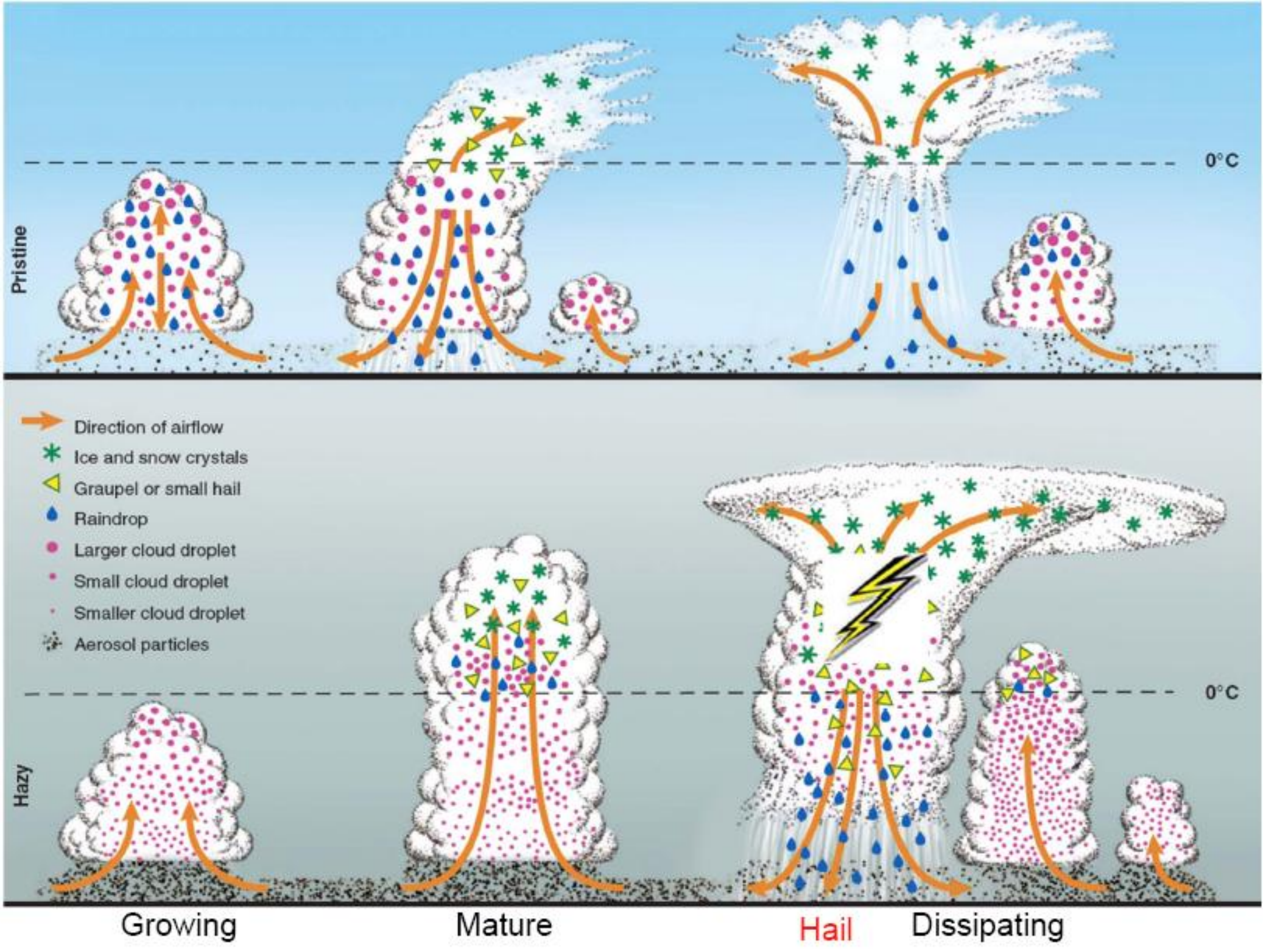
All the sources of ice production (primary and secondary) needed in models to reproduce observations

Arctic



(Sotiropoulou et al., 2022)

Aerosol-Precipitation Feedbacks



Aerosols reduce drizzle.

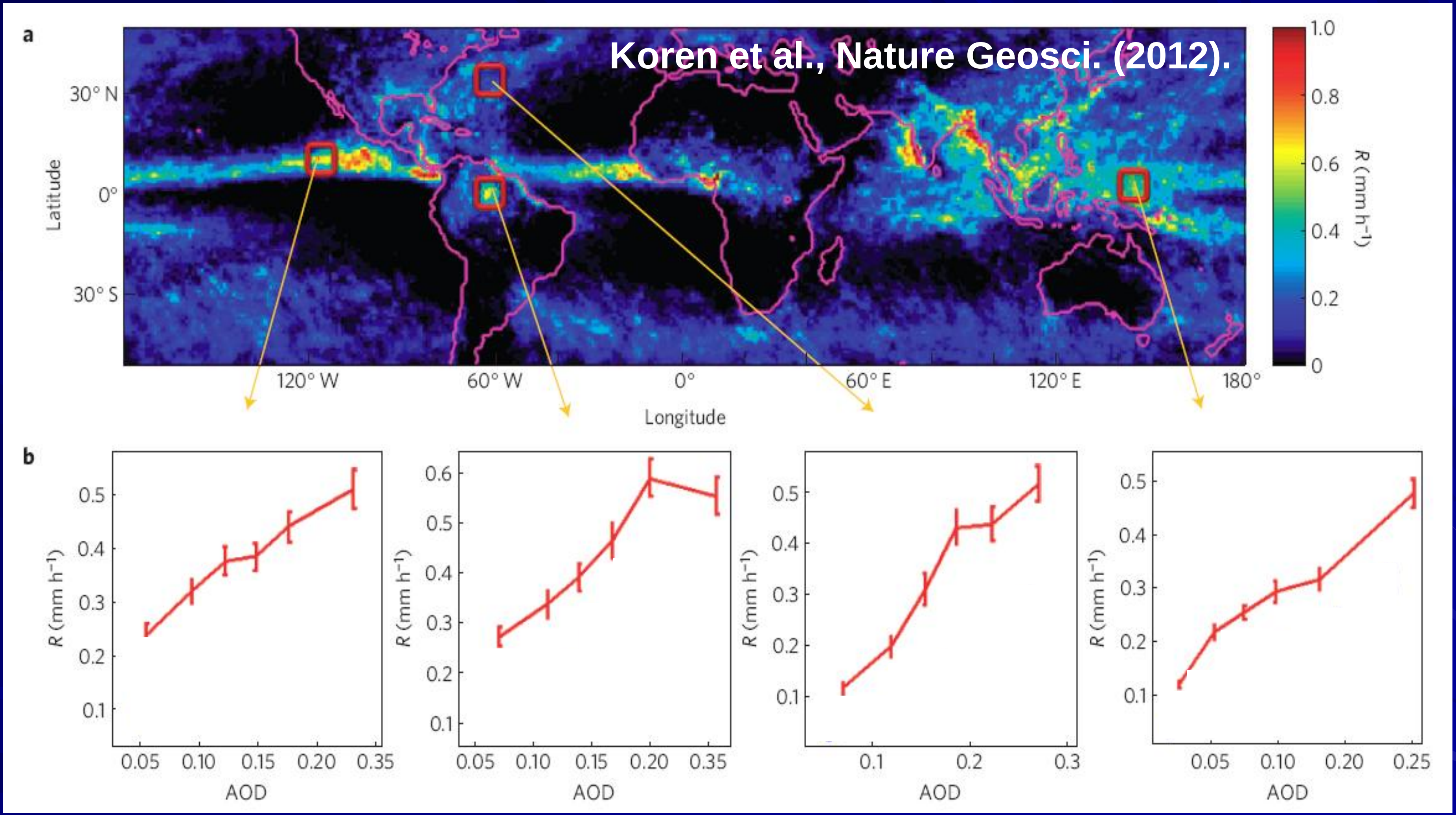
More water reaches the freezing level.

More latent heat is released during freezing.

Convective invigoration.

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

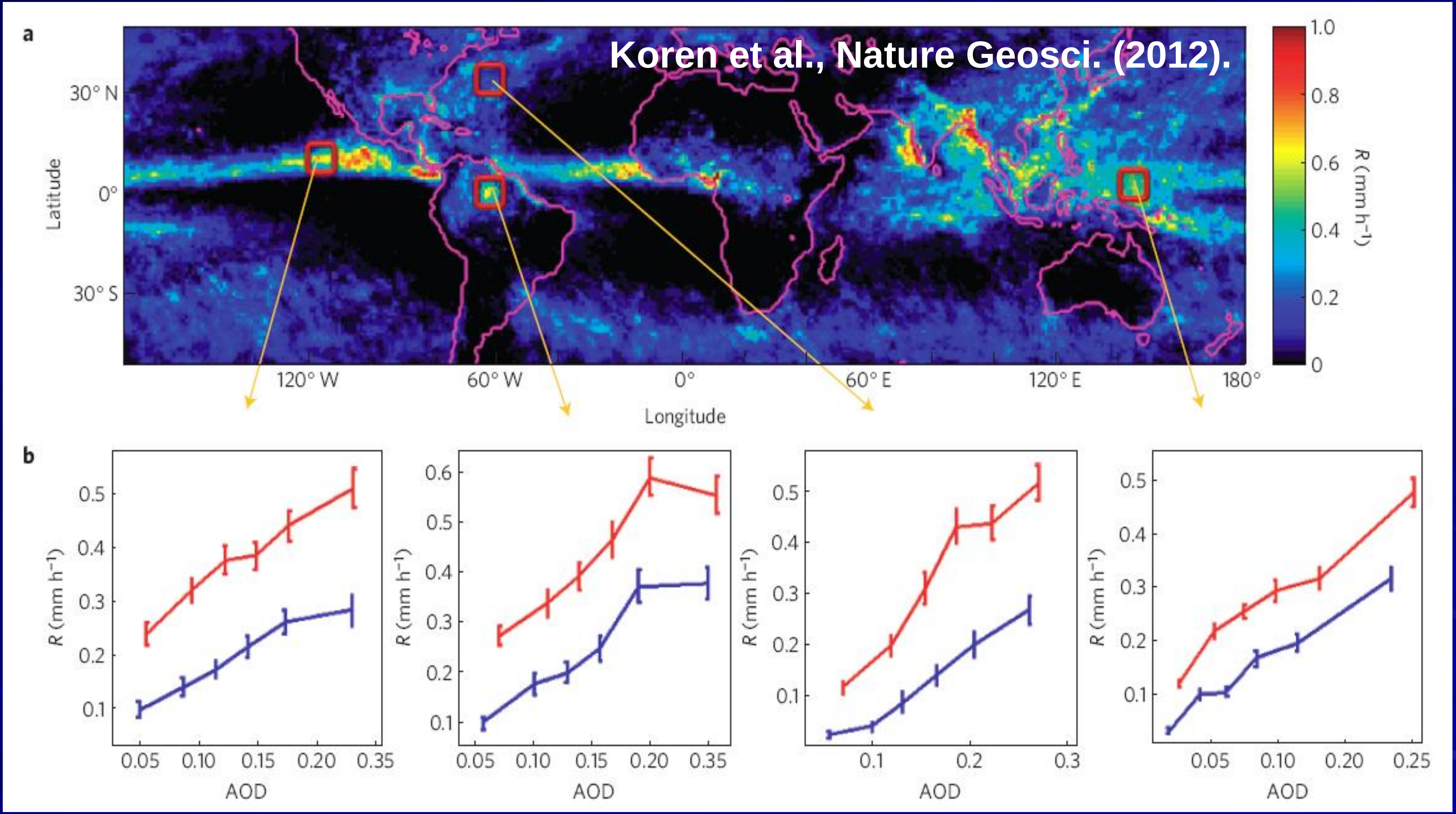
Convective invigoration is everywhere



Koren et al., Nature Geosci. (2012).

“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.”

Ongoing research: CleanCloud Mediterranean (CHOPIN) campaign

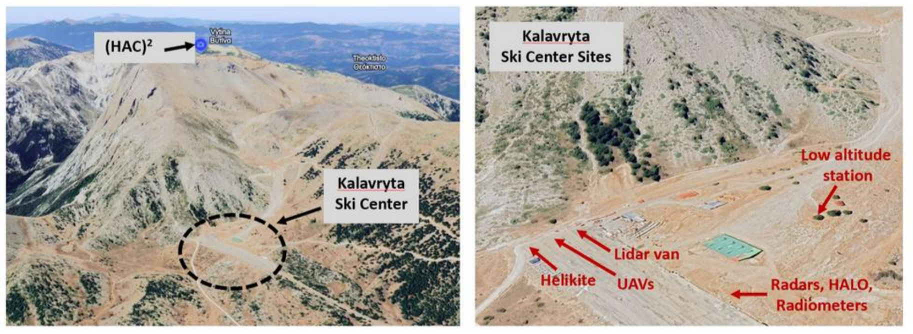

http://go.epfl.ch/chopin-campaign

Oct.24 – Jun.25

Where mythology, aerosols, clouds and climate meet

chopin

Cleancloud Helmos OrograPhic sIte experimeNt



DEMOKRITOS

ITE

EPFL

AARHUS UNIVERSITY

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

The Cyprus Institute

FINNISH METEOROLOGICAL INSTITUTE

ACADEMY OF ATHENS

nilu

CNR ISAC

Stockholm University

LapUp Laboratory of Atmospheric Physics University of Patras

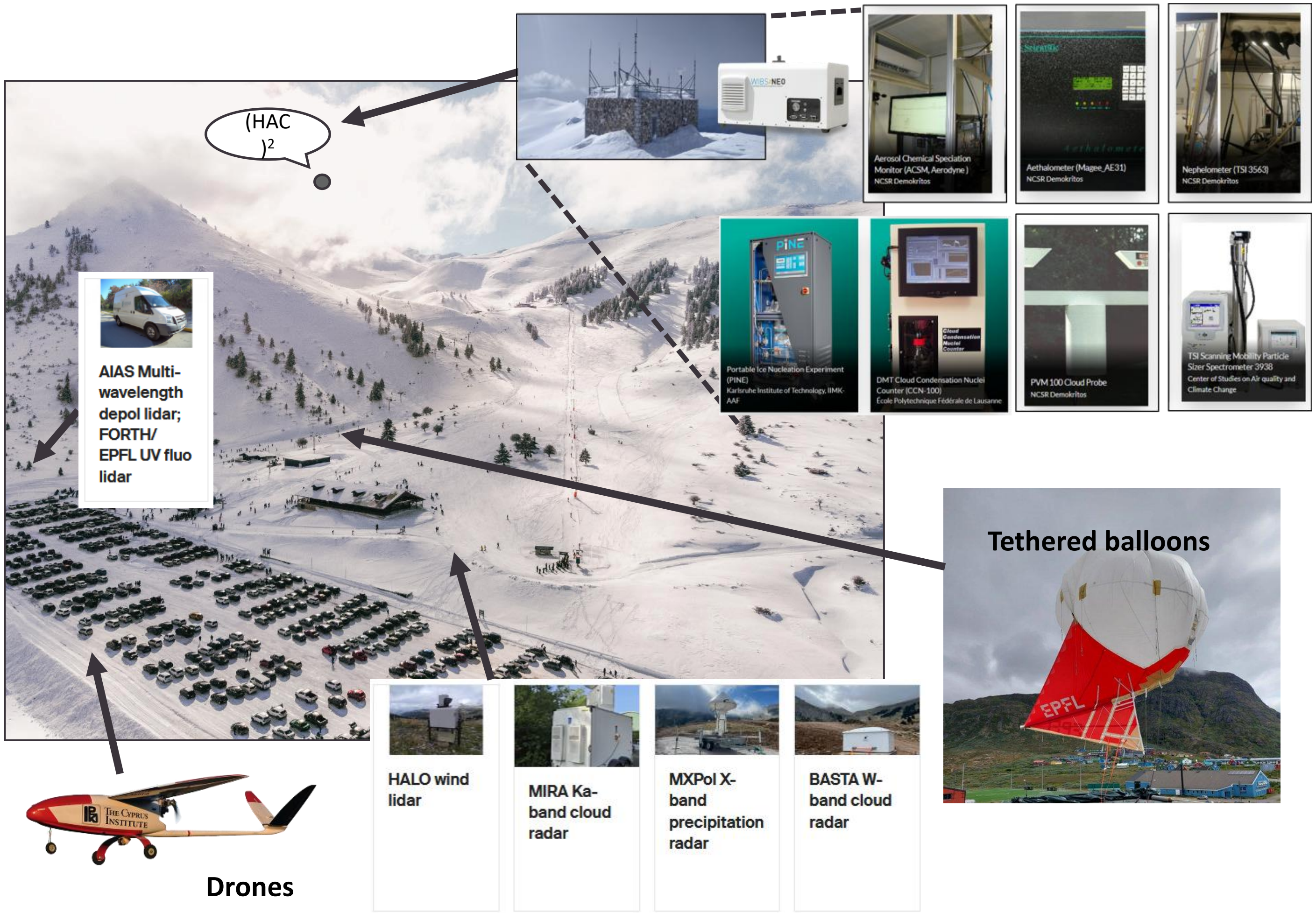
Federal Office of Meteorology and Climatology MeteoSwiss

Funded by the European Union

Activities and sites

chopin

Cleancloud Helmos
OrograPhic sIte experimeNt



Aerosol Chemical Speciation Monitor (ACSM, Aerodyne)
NCSR Demokritos

Aethalometer (Magee_AE31)
NCSR Demokritos

Nephelometer (TSI 3563)
NCSR Demokritos

Portable Ice Nucleation Experiment (PINE)
Karlsruhe Institute of Technology, IIMK-AAF

DMT Cloud Condensation Nuclei Counter (CCN-100)
École Polytechnique Fédérale de Lausanne

PVM 100 Cloud Probe
NCSR Demokritos

TSI Scanning Mobility Particle Sizer Spectrometer 3938
Center of Studies on Air quality and Climate Change

AIAS Multi-wavelength depol lidar; FORTH/ EPFL UV fluo lidar

Tethered balloons

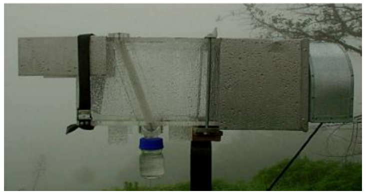
HALO wind lidar

MIRA Ka-band cloud radar

MXPol X-band precipitation radar

BASTA W-band cloud radar

Drones



Cloudwater collector

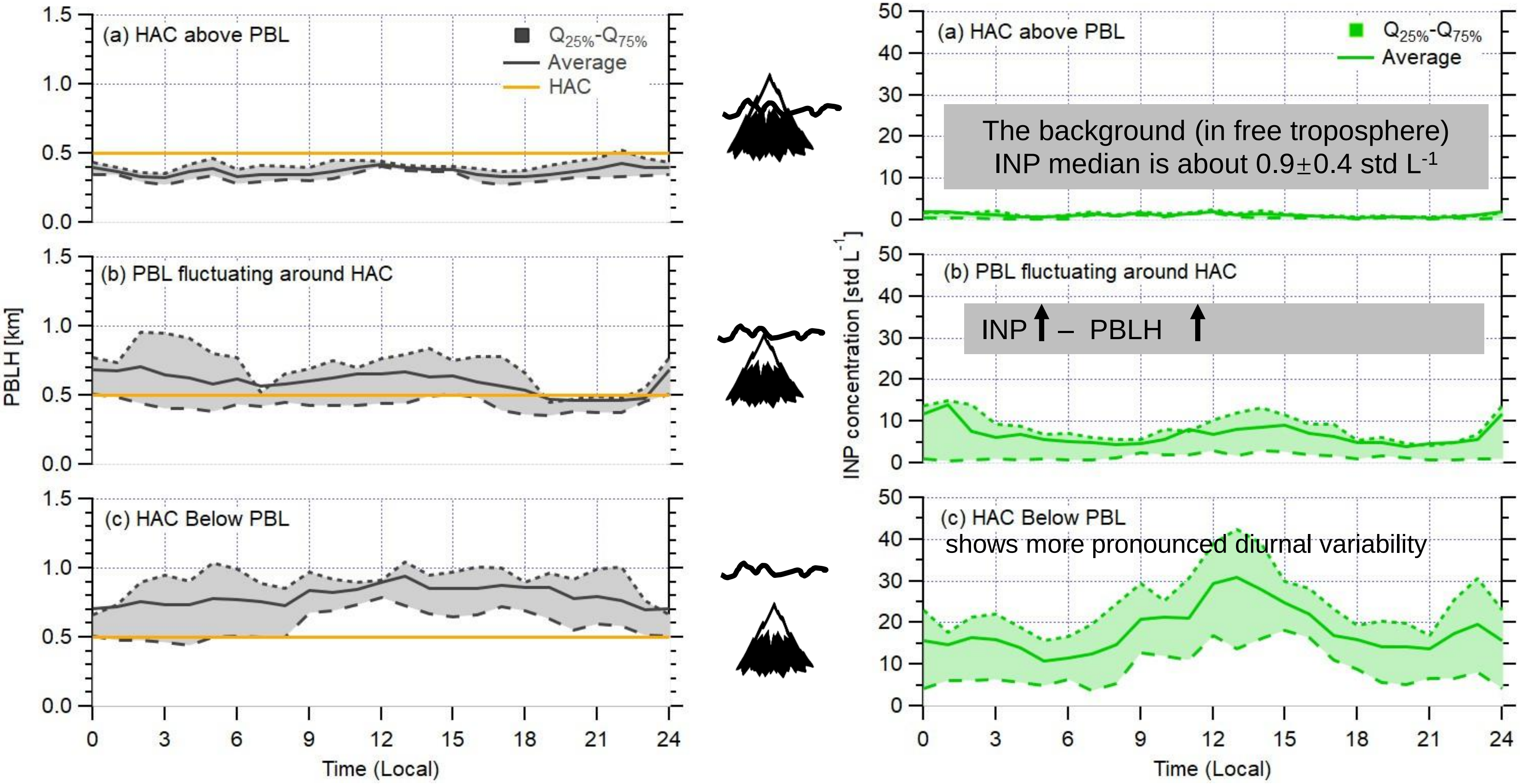
Amazing set of in-situ and remote sensing observation platforms

Funded by the European Union

One team – One Dream

Funded by the European Union

CHOPIN Results: (INP diurnal cycle, FT/PBL)



The background (in free troposphere) INP median is about 0.9±0.4 std L^{-1}

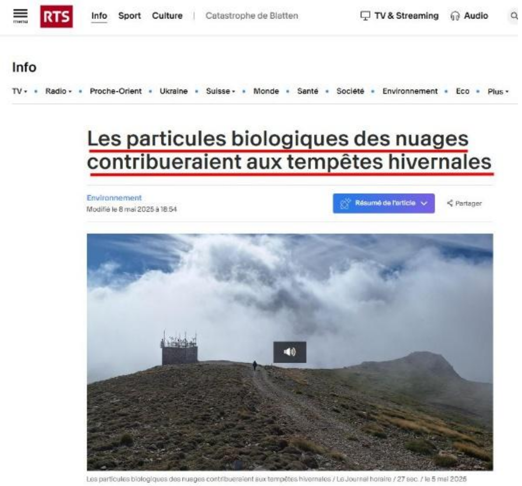
INP ↑ – PBLH ↑

shows more pronounced diurnal variability

(Gao et al., npj Climate and Atmospheric Sciences, 2025)

CHOPIN Results (bioaerosol-driven INP cycle)

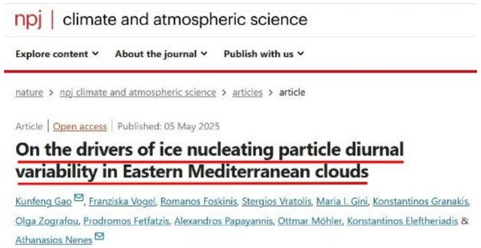
chopin
Cleancloud Helmos
OrograPhic site experimeNt



Les particules biologiques des nuages contribueraient aux tempêtes hivernales

Environnement
Modifié le 8 mai 2025 à 18:54

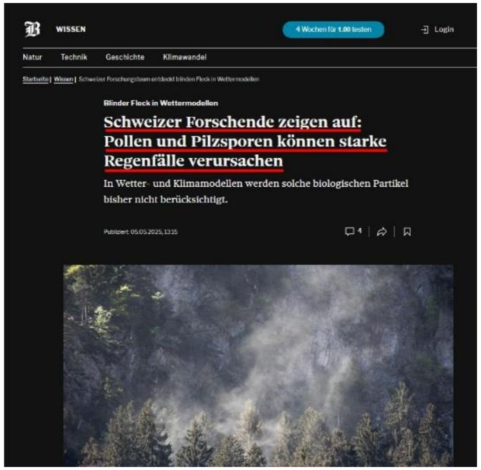
Les particules biologiques des nuages contribueraient aux tempêtes hivernales / Le Journal horaire / 27 sec. / le 5 mai 2025



Article | Open access | Published: 05 May 2025

On the drivers of ice nucleating particle diurnal variability in Eastern Mediterranean clouds

Kunfeng Gao, Franziska Vogel, Romanos Foskinis, Stergios Vratolis, Maria I. Gini, Konstantinos Granakis, Olga Zografou, Prodromos Fetfatzis, Alexandros Papayannis, Ottmar Möhler, Konstantinos Eleftheriadis & Athanasios Nenes



Blinder Fleck in Wettermodellen

Schweizer Forschende zeigen auf: Pollen und Pilzsporen können starke Regenfälle verursachen

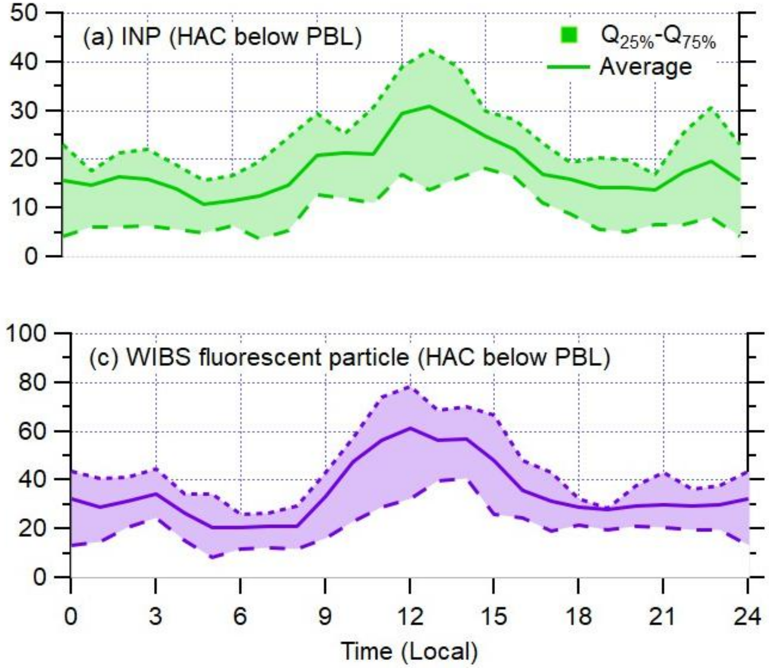
In Wetter- und Klimamodellen werden solche biologischen Partikel bisher nicht berücksichtigt.

Publiziert 05.05.2025, 13:15

Skyborne specks of life may influence rainfall patterns

A buildup of small particles of bacteria, pollen and spores may lead to rainier weather

Bacteria, pollen, spores and other biological particles released by ecosystems may have a profound influence on rainfall patterns.
ZPAGISTOCK/GETTY IMAGES



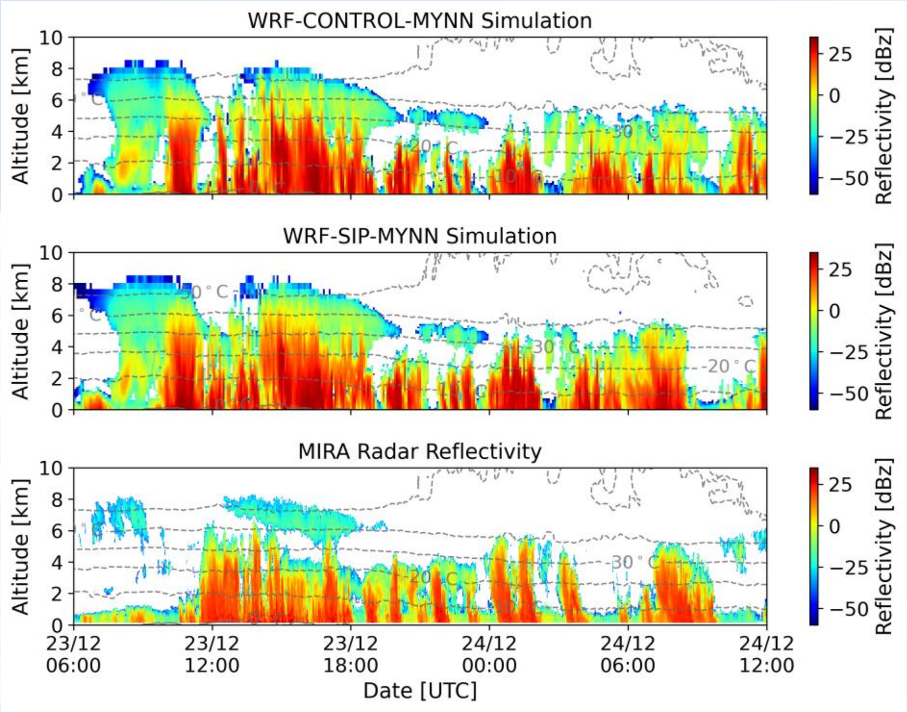
- **Contribution of biological particles key driver! Biology forces cloud formation and precipitation**

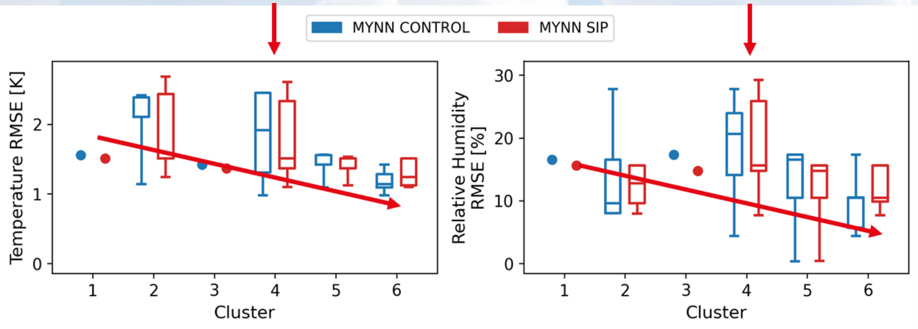
(Gao et al., npj Climate and Atmospheric Sciences, 2025)

CHOPIN Results (model improved)

WRF/EPFL with enhanced cloud mixed-phase microphysics (SIP)

- Compared cloud simulations to radar products from MIRA (35 GHz) during the whole CHOPIN
- Boundary layer captured reasonably well
- SIP parameterizations lead to better predictive fields in medium reflectivity multi-layer clouds





(Georgakaki et al., 2024; Haniotis et al., in prep)

Funded by the European Union

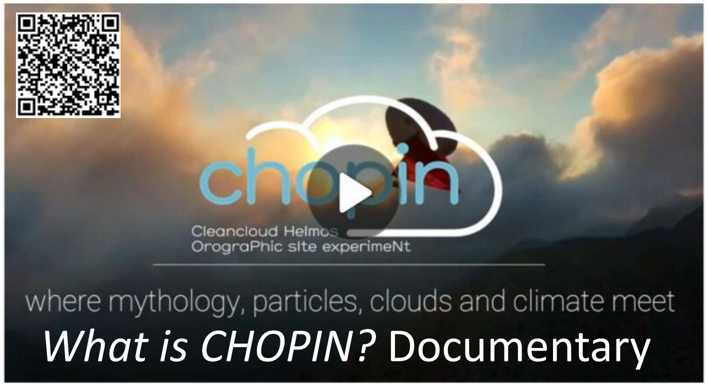
chopin
Cleancloud Helmos
OrograPhic sIte experimeNt

Moments

CleanCloud

Funded by
the European Union

CHOPIN media @ CleanCloud Channel

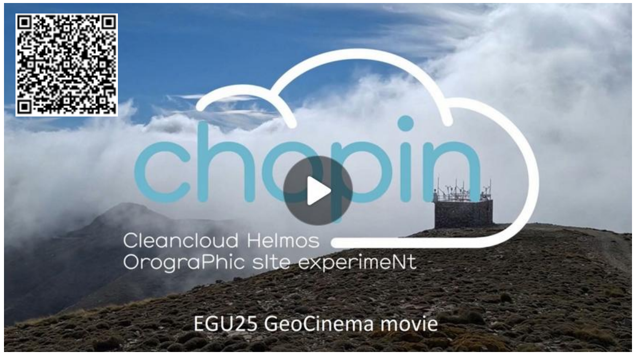


What is CHOPIN? Documentary

What is CHOPIN? Springtime

Early morning moments

Winter beauty at Mt.Helmos



EGU25 GeoCinema movie

Scientific Photography award by Nature
CHOPIN *Fog Chaser* by Lionel Favre

https://mediaspace.epfl.ch/channel/CleanCloud

Funded by
the European Union

Studies of CCN & INPs of fresh and aged seasalt and dust – in the lab

Facilities and locations

AURA (Aarhus CCC)

- Marine aerosols
- Lab & in-situ

FORTH ASC (FORTH/CSTACC)

- Marine & Aged dust
- Lab & in-situ air

CleanCloud *chamber cam**P**aign for study**I**ng the cloud-relev**A**nt properties of **N**atural aeros**O**l*: Patras and Aarhus, or

CleanCloud PIANO@Aarhus
June-July 2025

CleanCloud PIANO@Patras
July-August & Sep-Nov 2025

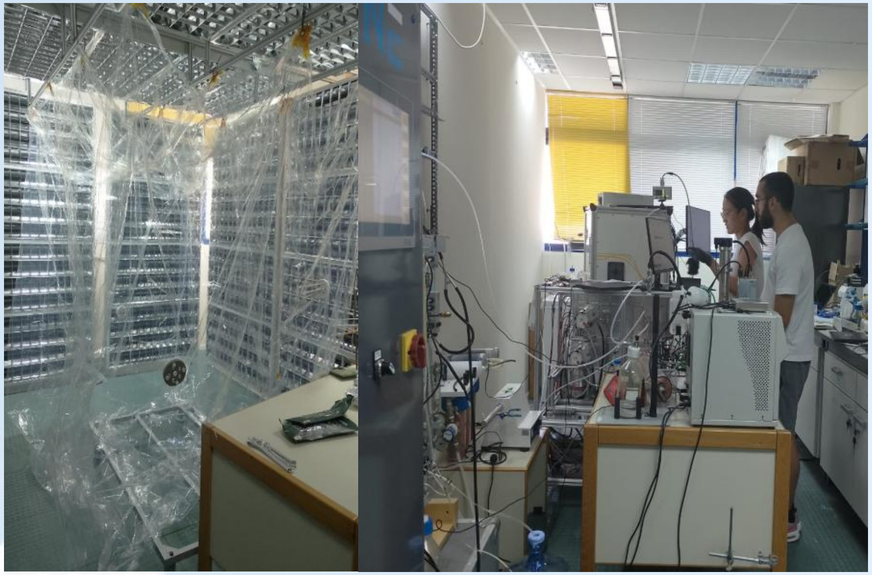
Funded by the European Union

The CleanCloud PIANO teams

Funded by the European Union

The CleanCloud PIANO Facilities

@Aarhus



@Patras

Funded by
the European Union

PIANO Videos and outreach

Videos on the CleanCloud Channel

https://mediaspace.epfl.ch/media/CleanCloud

Funded by the European Union

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.