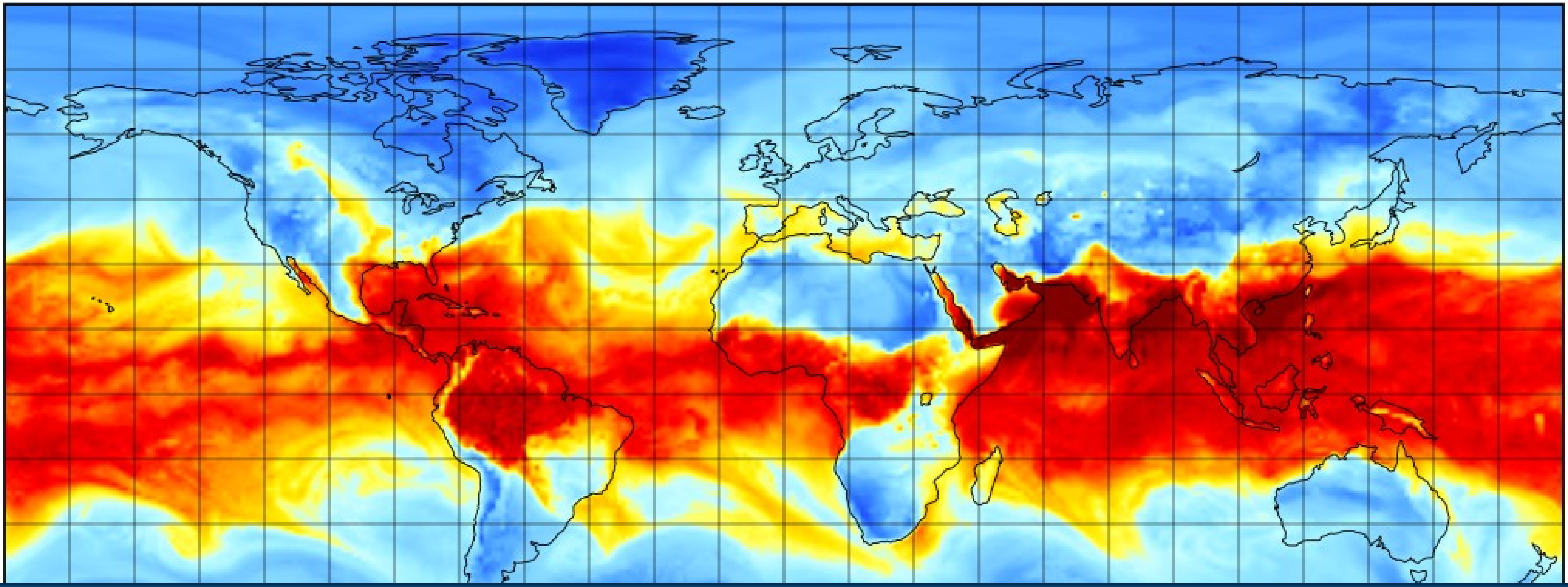


WATER IN THE ATMOSPHERE AND THE ROLE FOR CLIMATE

Part 3: Cloud formation (water and ice clouds)

WS 22/23 I CHRISTIAN ROLF

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

JÜLICH Forschungszentrum

TOPICS

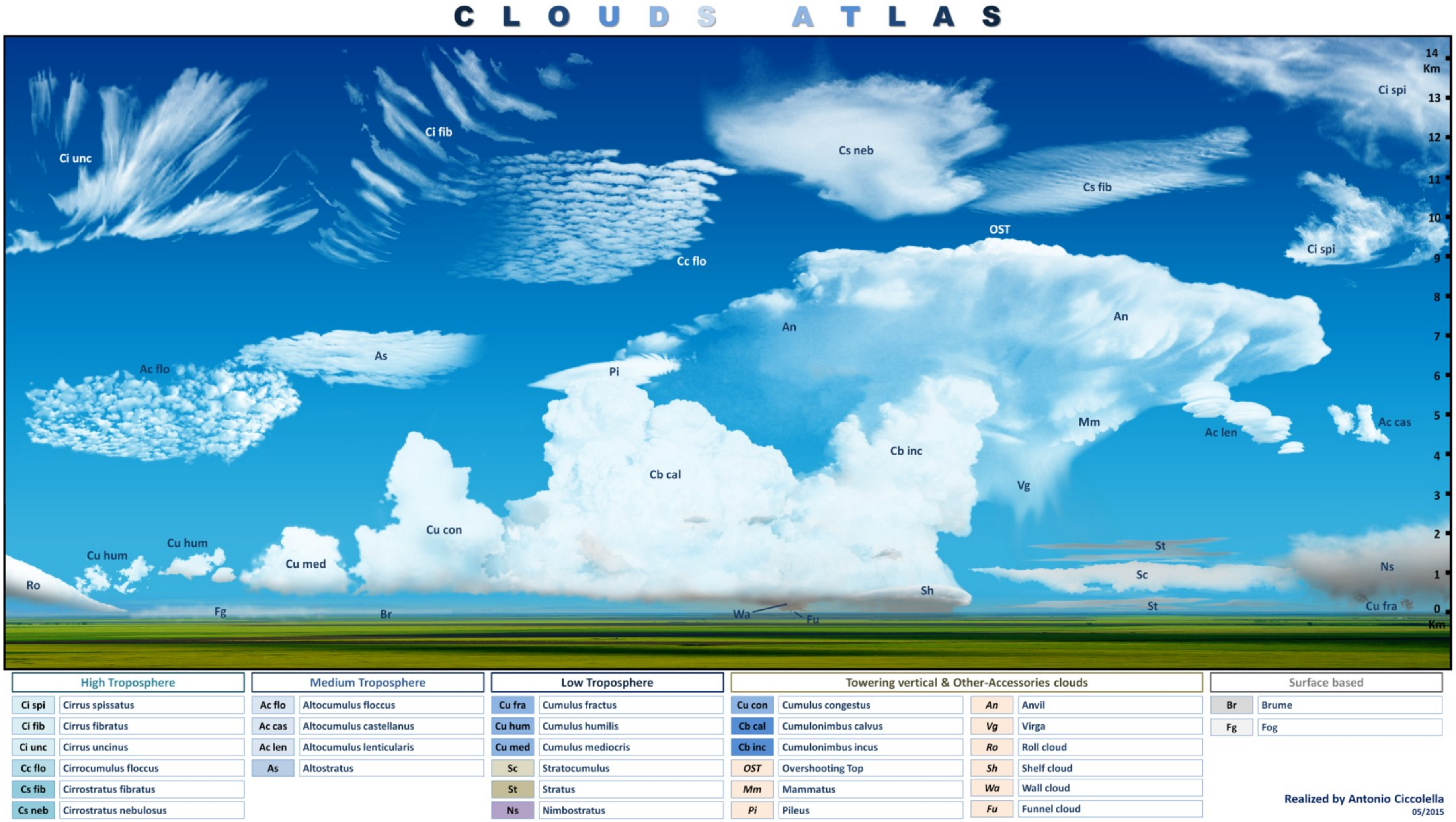
1. Introduction into units and definitions
2. Water vapor distribution in the atmosphere
3. Cloud formation (water and ice clouds)
4. Water cycle
5. Water and climate feedback
6. Measurement of water in the atmosphere

SUBTOPICS

3. Cloud formation (water and ice clouds)

- **Cloud types**
- Cloud properties and saturation vapor pressure
- *Warm clouds*
  - Raoult’s law, Kelvin effect, Koehler curve
  - Cloud condensation nuclei (CCN)
  - Cloud droplet growth and other processes
- *Ice clouds / Ice cloud life cycle*
  - Freezing process: Homogeneous nucleation
  - Freezing process: Heterogeneous nucleation
  - Ice crystal growth, shapes and sedimentation
- *Mixed phase clouds*
  - Bergeron-Findeisen Process

CLOUD TYPES



High Troposphere	
Ci spi	Cirrus spissatus
Ci fib	Cirrus fibratus
Ci unc	Cirrus uncinus
Cc flo	Cirrocumulus floccus
Cs fib	Cirrostratus fibratus
Cs neb	Cirrostratus nebulosus

Medium Troposphere	
Ac flo	Altocumulus floccus
Ac cas	Altocumulus castellanus
Ac len	Altocumulus lenticularis
As	Altostratus

Low Troposphere	
Cu fra	Cumulus fractus
Cu hum	Cumulus humilis
Cu med	Cumulus mediocris
Sc	Stratocumulus
St	Stratus
Ns	Nimbostratus

Towering vertical & Other-Accessories clouds			
Cu con	Cumulus congestus	*An*	Anvil
Cb cal	Cumulonimbus calvus	*Vg*	Virga
Cb inc	Cumulonimbus incus	*Ro*	Roll cloud
OST	Overshooting Top	*Sh*	Shelf cloud
Mm	Mammatus	*Wa*	Wall cloud
Pi	Pileus	*Fu*	Funnel cloud

Surface based	
Br	Brume
Fg	Fog

Realized by Antonio Ciccolella
05/2015

Clouds Atlas von Antonio Ciccolella, CC BY-SA 4.0

CLOUD TYPES

1. **Warm clouds (T > 0°C)**
   - only water droplets
   - drop and rain formation

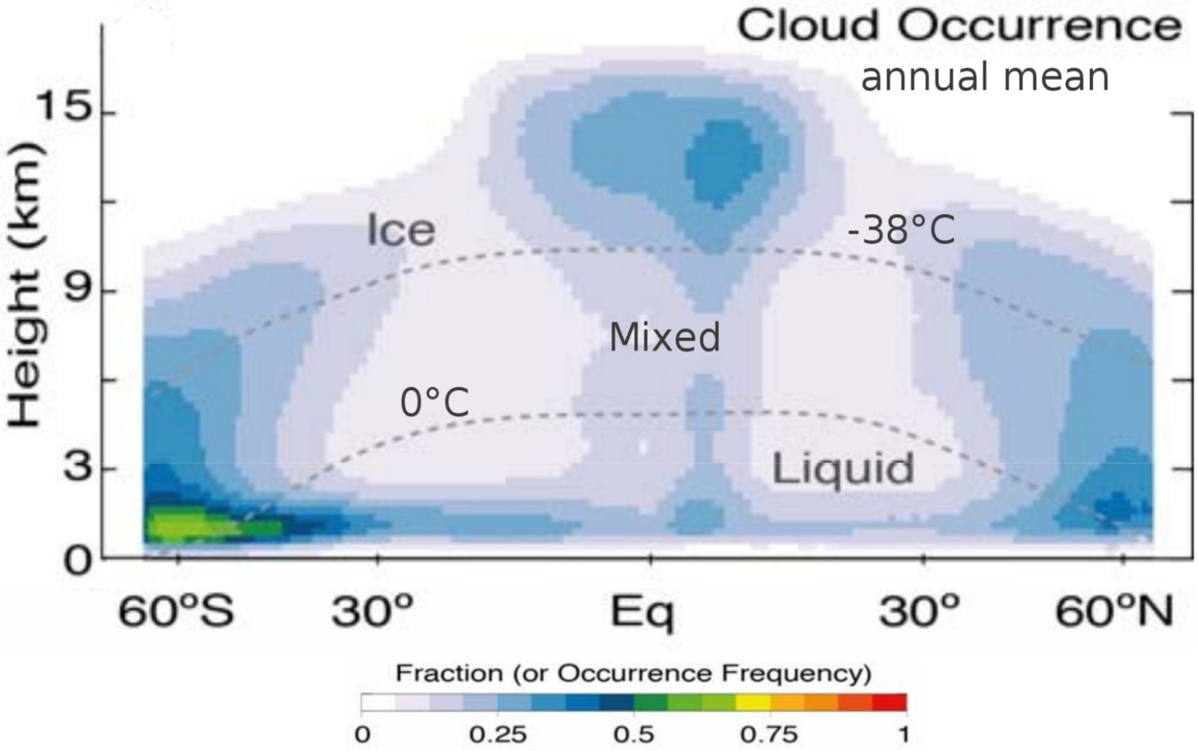
2. **Cold clouds (T < 0°C)**
   - Ice formation

2.1 Mixed Phase Clouds

- water drops and ice crystals
- generation of precipitation

2.2 Ice Clouds

- only ice crystals
- aerosol impact on ice clouds



IPCC 2013

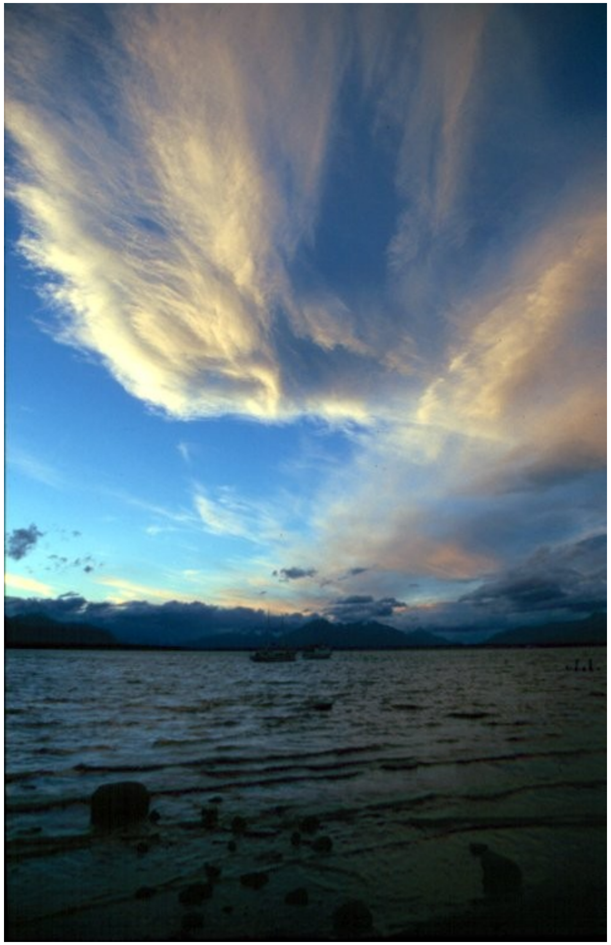
from CloudSat/CALIPSO 2B-GEOPROF-LIDAR dataset for 2006-2011; Mace et al., 2009

Chapter 7: Clouds and Aerosols (Boucher et al., 2013)

WARM CLOUDS (LOW LEVEL)

Pictures are taken from
Karlsruher Wolkenatlas
http://www.wolkenatlas.de

ICE CLOUDS, CIRRUS (HIGH LEVEL)



Pictures are taken from
Karlsruher Wolkenatlas
http://www.wolkenatlas.de

ICE CLOUDS, CONTRAILS (HIGH LEVEL)

Cirrus clouds:

- Ice crystals
  (no water droplets)
- Temperature range:
  -38 to -90°C
- Altitude range:
  5 - 18km

Contrails are anthropogenic induced cirrus clouds

Pictures are taken from
Karlsruher Wolkenatlas
http://www.wolkenatlas.de

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CLOUD PROPERTIES

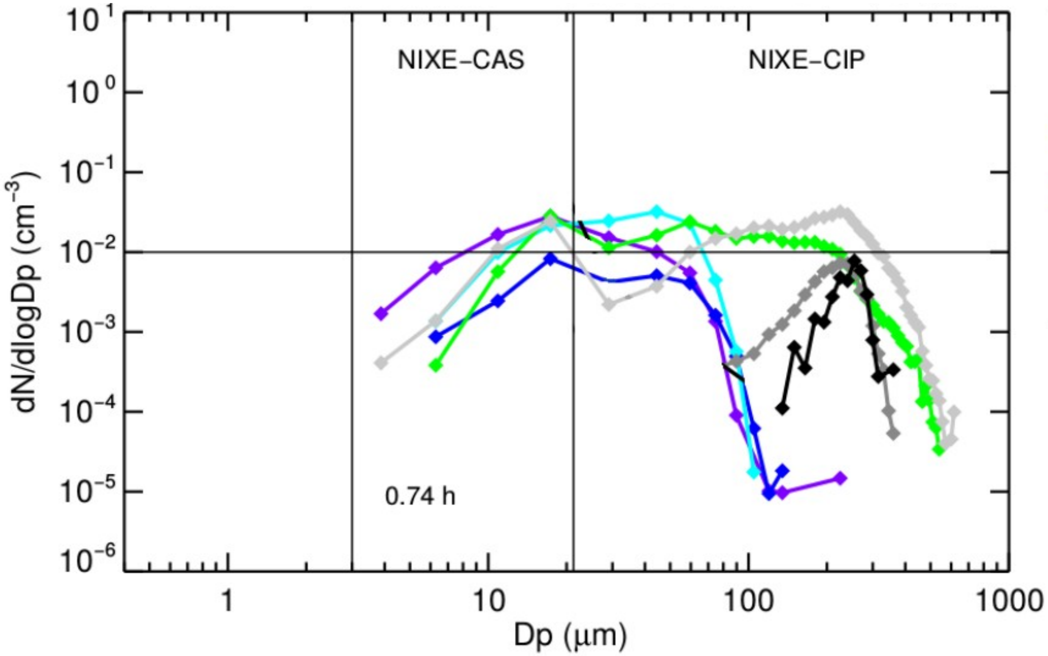
- Particle size distribution N(Dp)

Number concentration N
Size distribution Dp

- Liquid/ Ice water content

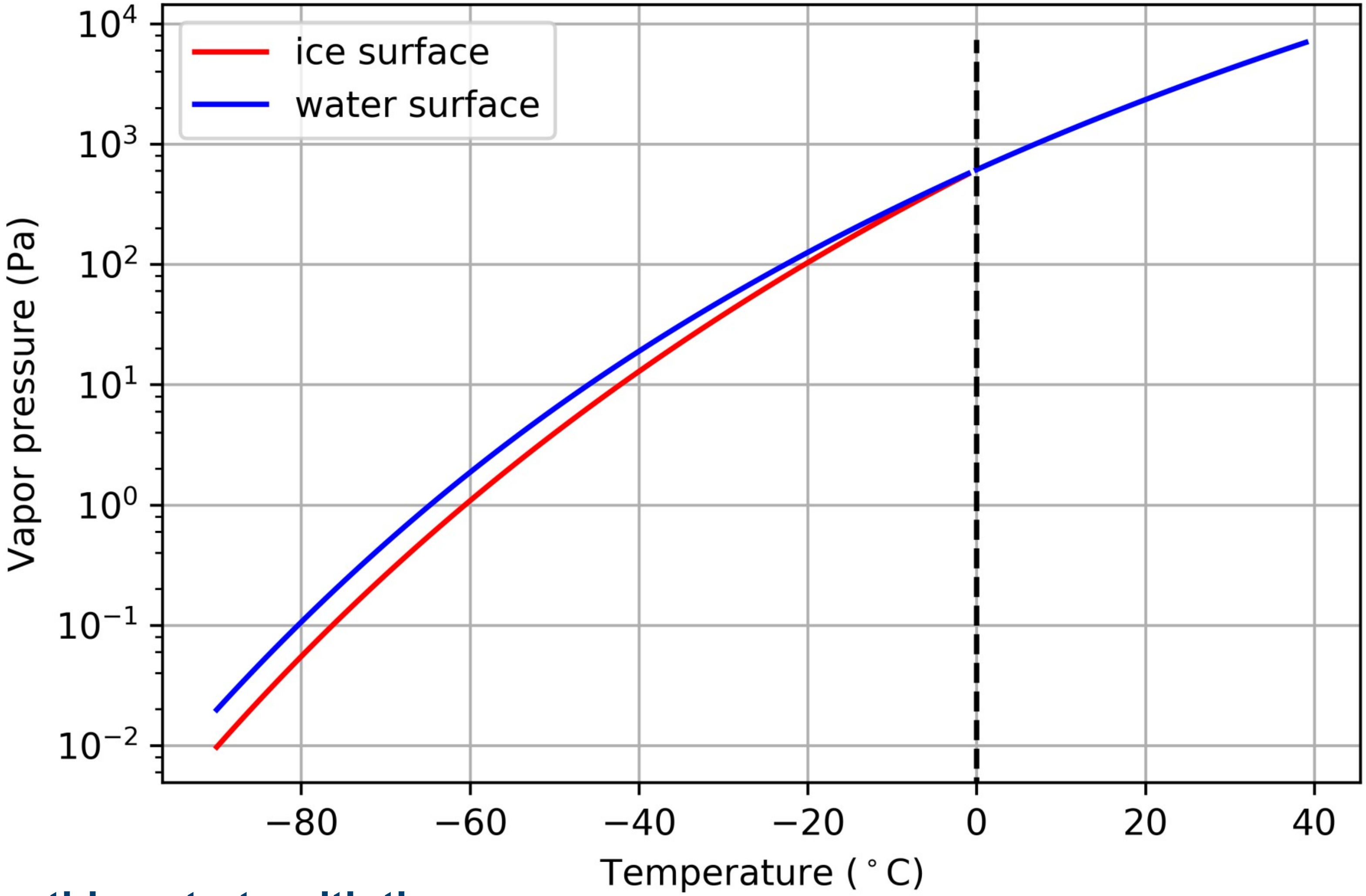
$$LWC/IWC = \rho \int_{Dp_{min}}^{Dp_{max}} N(Dp)\, dDp$$

- Relative humidity wrt.
  Ice (RH_i) and water (RH_w)



➢ All quantities are inhomogeneous distributed in the atmosphere (time and space)

Saturation vapor pressure



Everything starts with the saturation vapor pressure difference between liquid and ice surface

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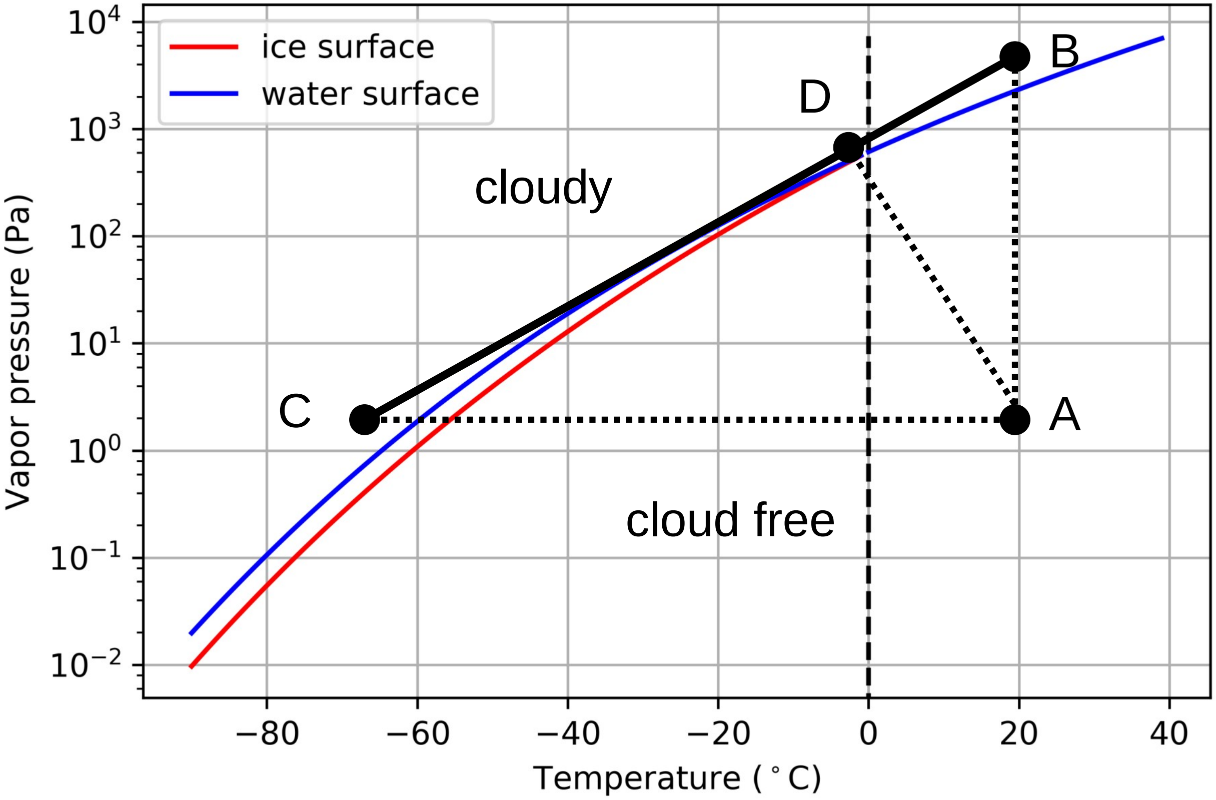
How to saturate the air ?



< e_s: Sub-saturation

= e_s: Saturation

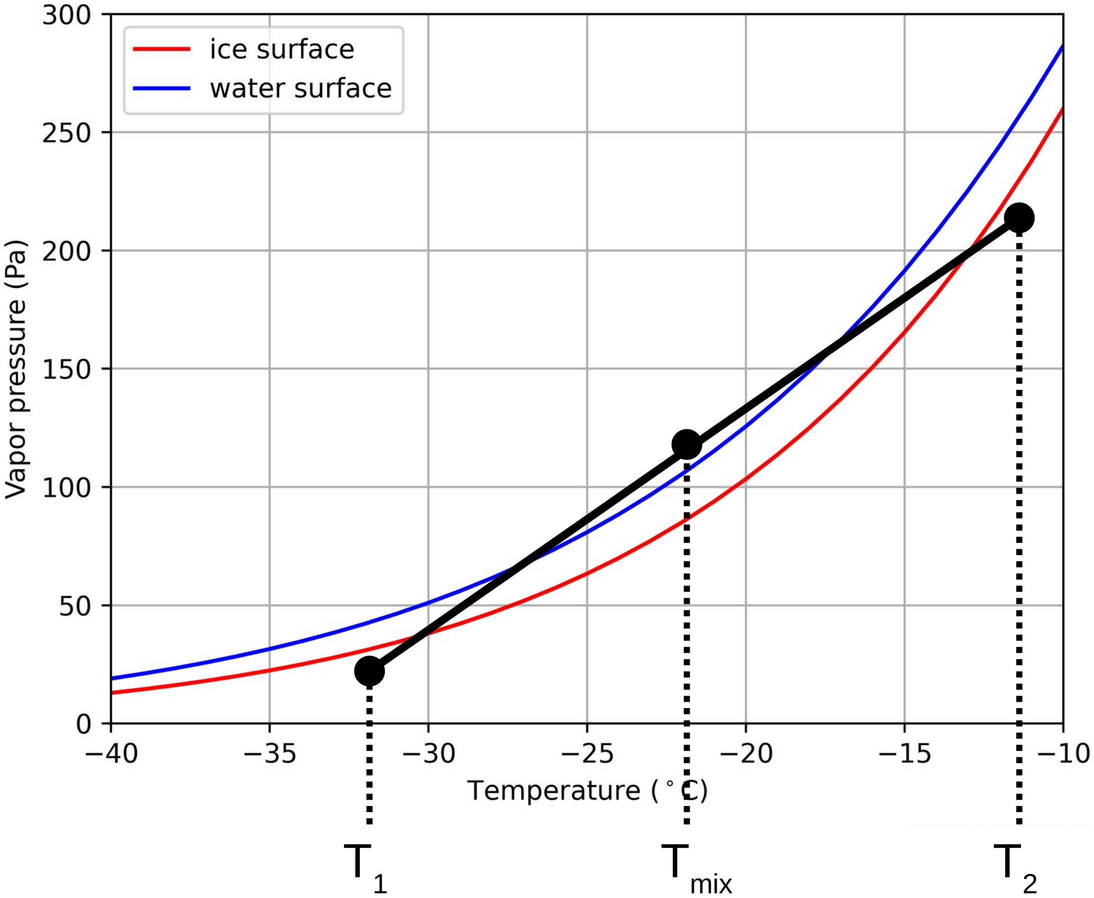
> e_s: Supersaturation

→Condensation

Three ways:

- Increase (inject more) water vapor to the air (A → B).
- Reduce the temperature of the air (A → C).
- Mix cold air with warm, moist air (A → D).

How to saturate the air ?



Under certain conditions, the mixing of two air masses which are not saturated can lead to condensation, for example in air craft contrails.

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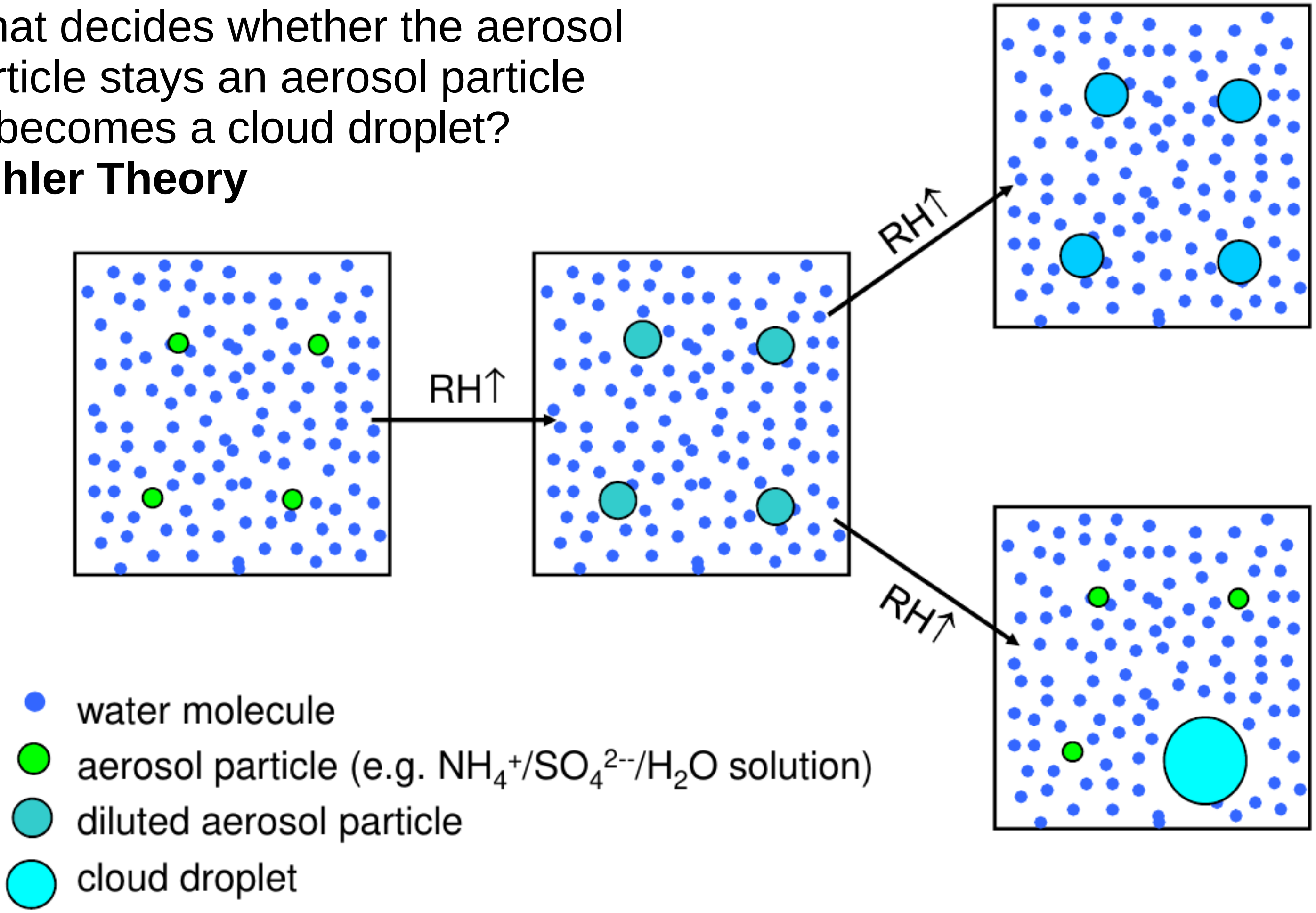
WARM CLOUDS IN THE ATMOSPHERE

Cloud droplet formation
A fierce competition without which precipitation would be massively impeded!

What decides whether the aerosol particle stays an aerosol particle or becomes a cloud droplet?
Köhler Theory



CLOUD DROPLET FORMATION

- **Raoult's law (1870)**
  small droplets have higher solute concentrations (salts, acids) and this reduces the H2O vapor pressure
  → *advantage for small droplets*

- **Kelvin effect (1879)**
  small droplets have a higher H2O vapor pressure (curvature effect)
  → *disadvantage for small droplets*

- **Köhler equation (1921)**
  balance between Kelvin and Raoult terms
  → *quantitative understanding*

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RAOULT‘S LAW

Vapor pressure of liquid mixtures A and B:

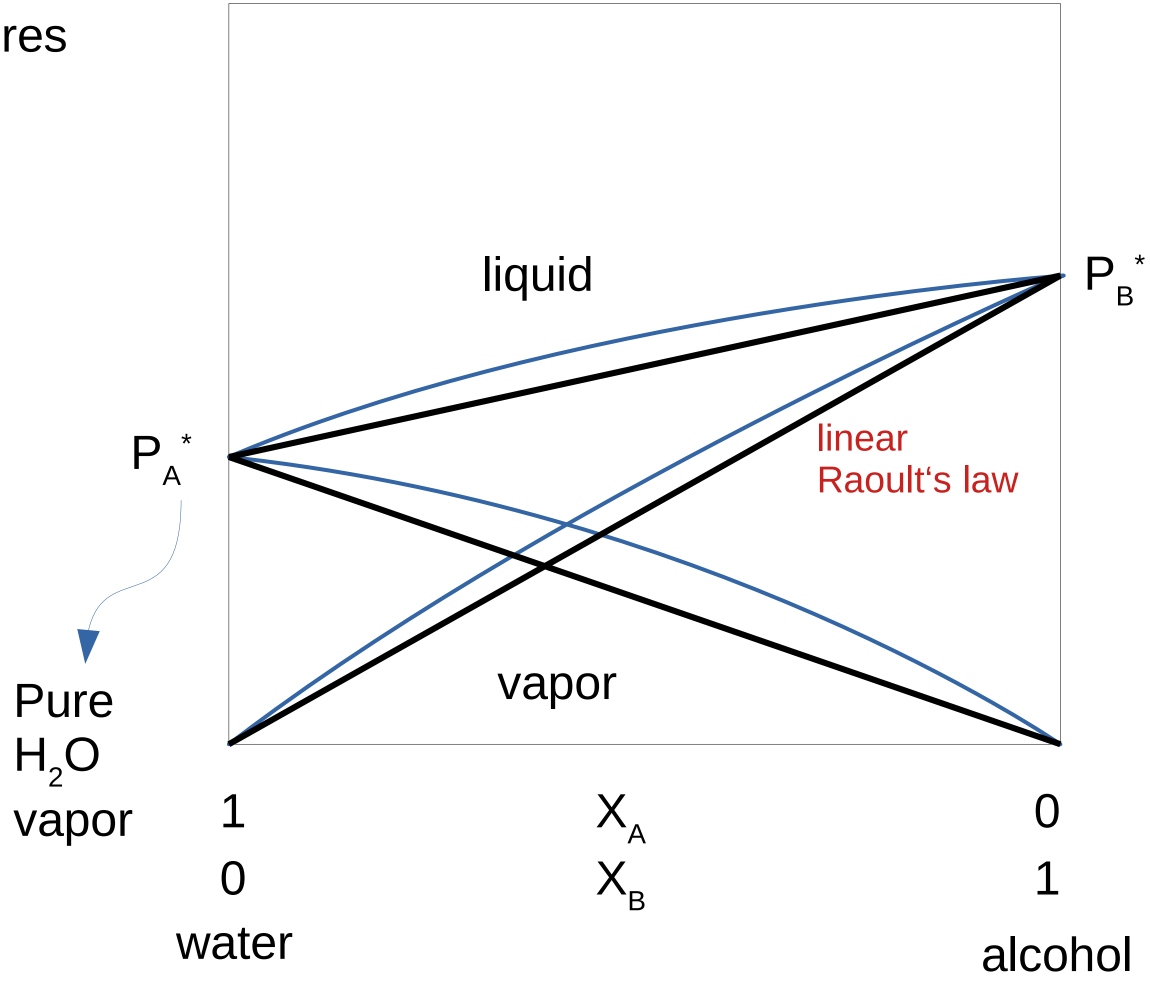
$$p_A = X_A p_A^*, \; p_B = X_B p_B^*$$

$$P = p_A + p_B$$

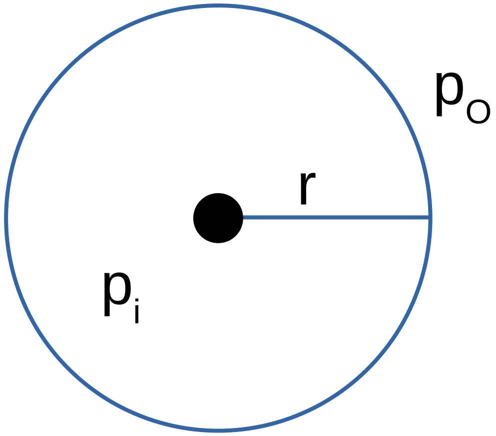
P = total pressure
$P_{A,B}$ = partial pressure
$P_{A,B}^*$ = pressure of pure substance
$X_{A,B}$ = mole fraction
$n_{A,B}$ = mole number

$$X_{A,B} = \frac{n_{A,B}}{\sum n_i}$$

For water droplets: $X_w \propto 1 - \frac{r_d^3}{r^3 - r_d^3}$



KELVIN EFFECT



Work required to increase the surface area A of the liquid-vapor interface:
dW = σdA
σ = surface tension
A = 4πr^2 = surface area

$$dW = \sigma\, dA \wedge dW = A\, dp$$

$$4\pi r^2 (p_i - p_o) = A\, dp = \sigma\, d(4\pi r^2) = 8\pi r \sigma$$

$$p_i = p_o \underbrace{+2\frac{\sigma}{r}}_{\text{Important for } r < 50\ \mu\text{m (5-10\% effect)}}$$

Laplace equation (valid for bubble or droplet)

Since the surface tension tends to decrease the surface area, the pressure P$_i$ inside the spherical drop is greater than the pressure P$_o$ of the surrounding. The greater the surface tension, the greater is the pressure difference dp.

KELVIN EFFECT (1)

What is the influence of the higher pressure inside a curved surface on the vapor pressure of a droplet?

KELVIN EFFECT (2)

KELVIN EFFECT

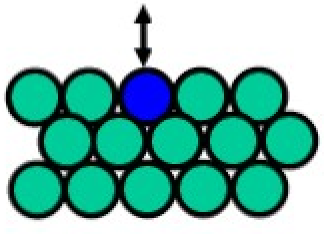
$$\ln\left(\frac{p(r)}{p(\infty)}\right)=\frac{V_{m,liquid}\,\Delta p}{RT}$$

$p(\infty)$ = vapor pressure over flat surface,
r = particle radius, R = gas constant,
Vm,liquid = liquid molar volume, e.g. of H_2O,
σ = surface tension

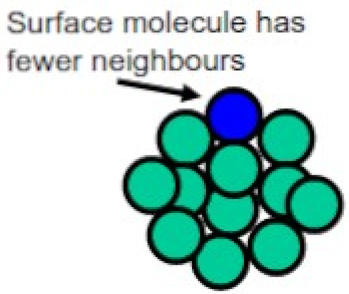
For curved surfaces: $\Delta p = 2\sigma / r$

$$p(r)=p(\infty)\,e^{2\sigma V_{m,liquid}/(rRT)}$$

Kelvin Equation



Planar surface: Equilibrium when ($e=e_s$) and number of molecules impinging on surface equals rate of evaporation.



Curved surface: saturation vapor pressure increases with smaller drop size since surface molecules have fewer binding neighbours

effect proportional to 1/r:
Kelvin effect

COMBINATION OF RAOULT AND KELVIN (1)

Kelvin eq.:

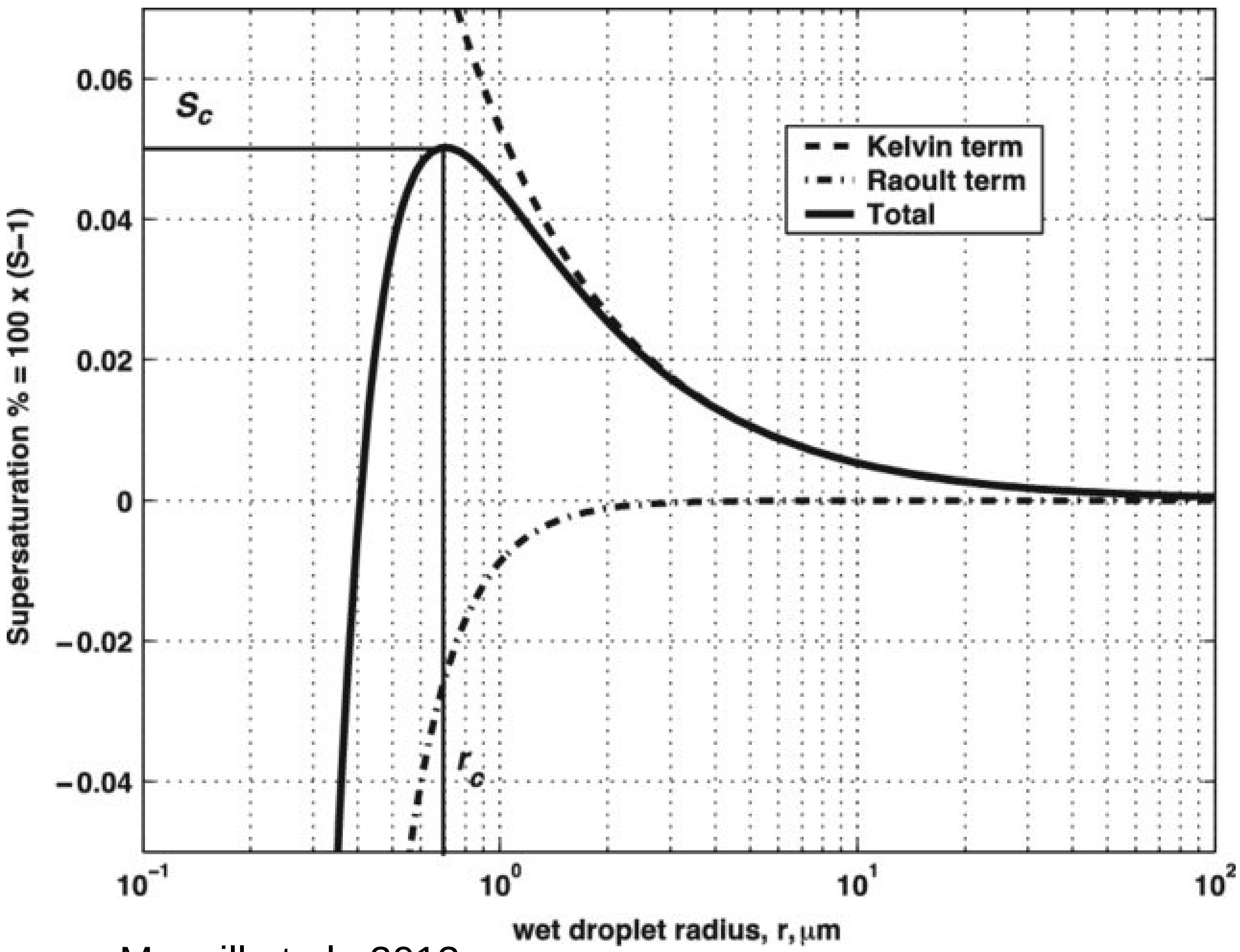
$$p(r) = p(\infty)\,\mathrm{e}^{2\sigma V_{m,liquid}/(rRT)}$$

$p_w(\infty)$ = H_2O vapor pressure over liquid water,
σ = surface tension of solution [kg / s^2],
r = particle radius [m],
$V_{m.liquid}$ = liquid molar volume of H_2O [m^3 / mol],
n_s = molar density of solute [1/mol]
R = Gas constant [kg m^2 / s^2/ K / mol]

COMBINATION OF RAOULT AND KELVIN (2)

KÖHLER THEORY

$$S_w - 1 = \frac{p_w(r_p)}{p_w(\infty)} \approx \exp\left(\frac{2\sigma V_{m,liquid}}{r_p RT} - \frac{3 n_s V_{m,liquid}}{4\pi r_p^3}\right)$$



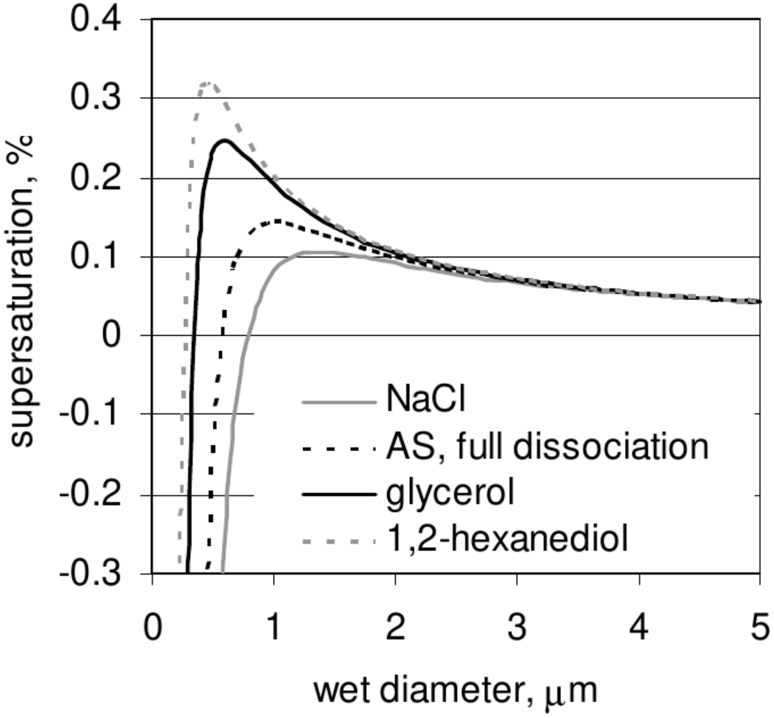
Mcneill et al., 2013

Köhler curve for a NaCl particle with a dry diameter of 50 nm.

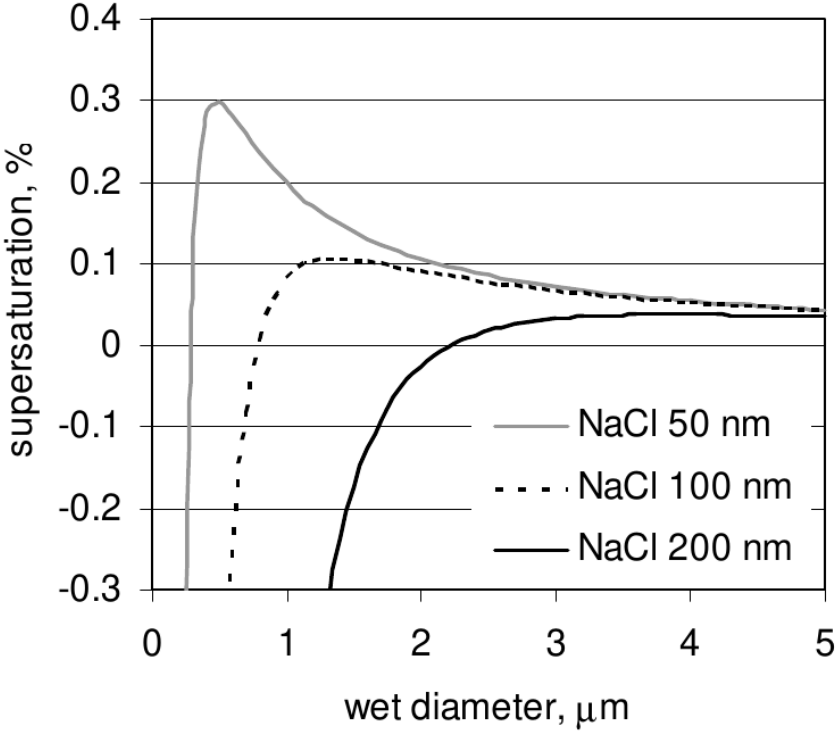
r_c: critical radius for further growth

KÖHLER CURVES

Köhler curves for particles (with dry diameters of 100 nm) of different compositions



Köhler curves for NaCl particles of different dry diameters



The Köhler curve describes the equilibrium vapor pressure of a droplet with a specified dry diameter as it takes up or loses water.

- **Dependence on cooling rate dS/dt**

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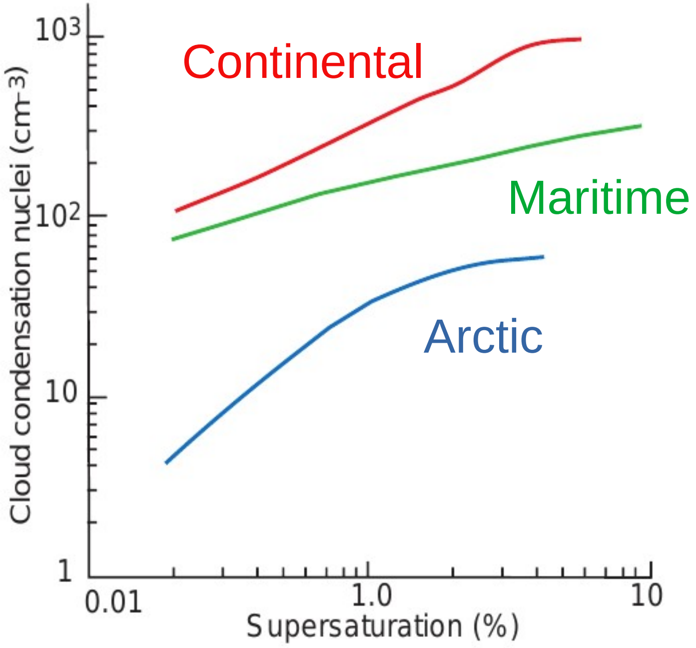
CLOUD CONDENSATION NUCLEI (CCN)

Random condensation to achieve critical radius is unlikely. The presence of condensation nuclei (CCN) is necessary for condensation process. This lowers the surface tension ($\sigma_{H2O} = 8 \cdot 10^{-2}$ N/m) and therefore the required saturation level.

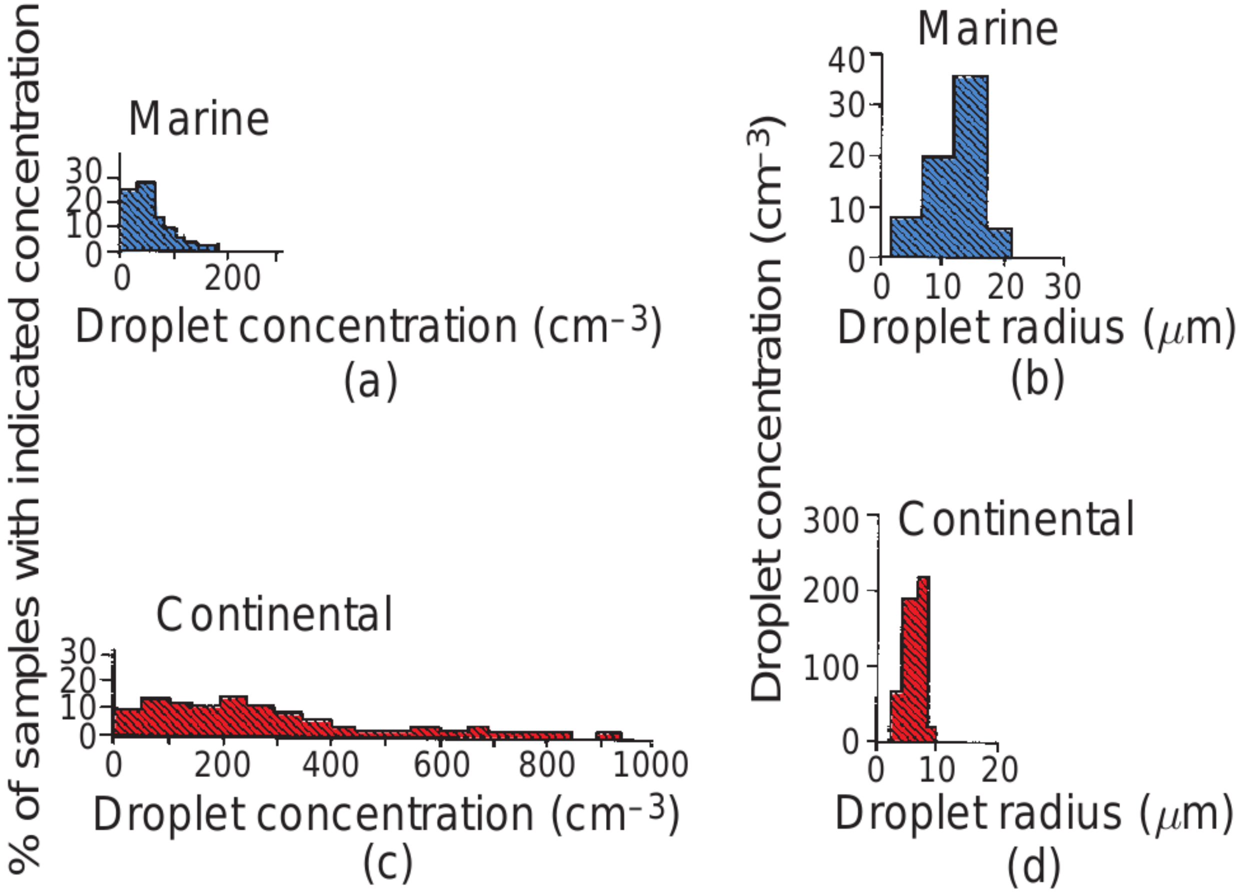
Super-saturation level in natural clouds S≈0.1%!
That is mostly insufficient for cloud condensation!

Optimum and most effective condensation nuclei are hygroscopic aerosols (e.g. sea salt, sodium chlorate, ammonium sulfate etc), which lower the relative humidity necessary for providing the critical droplet size. Sea salt provides condensation conditions at relative humidity of <100%, sulphuric and nitric acid particles provide already condensation at a low relative humidity of 75%.



Wallace & Hobbs

CCN CONDITIONS



Wallace & Hobbs

CCN concentration have impact on the resulting particle size distribution

CCN EFFECT ON CLOUD DROP NUMBER

Wallace & Hobbs

Ship tracks (white lines) in marine stratus clouds over the Atlantic Ocean as viewed from the NASA Aqua satellite on January 27, 2003.

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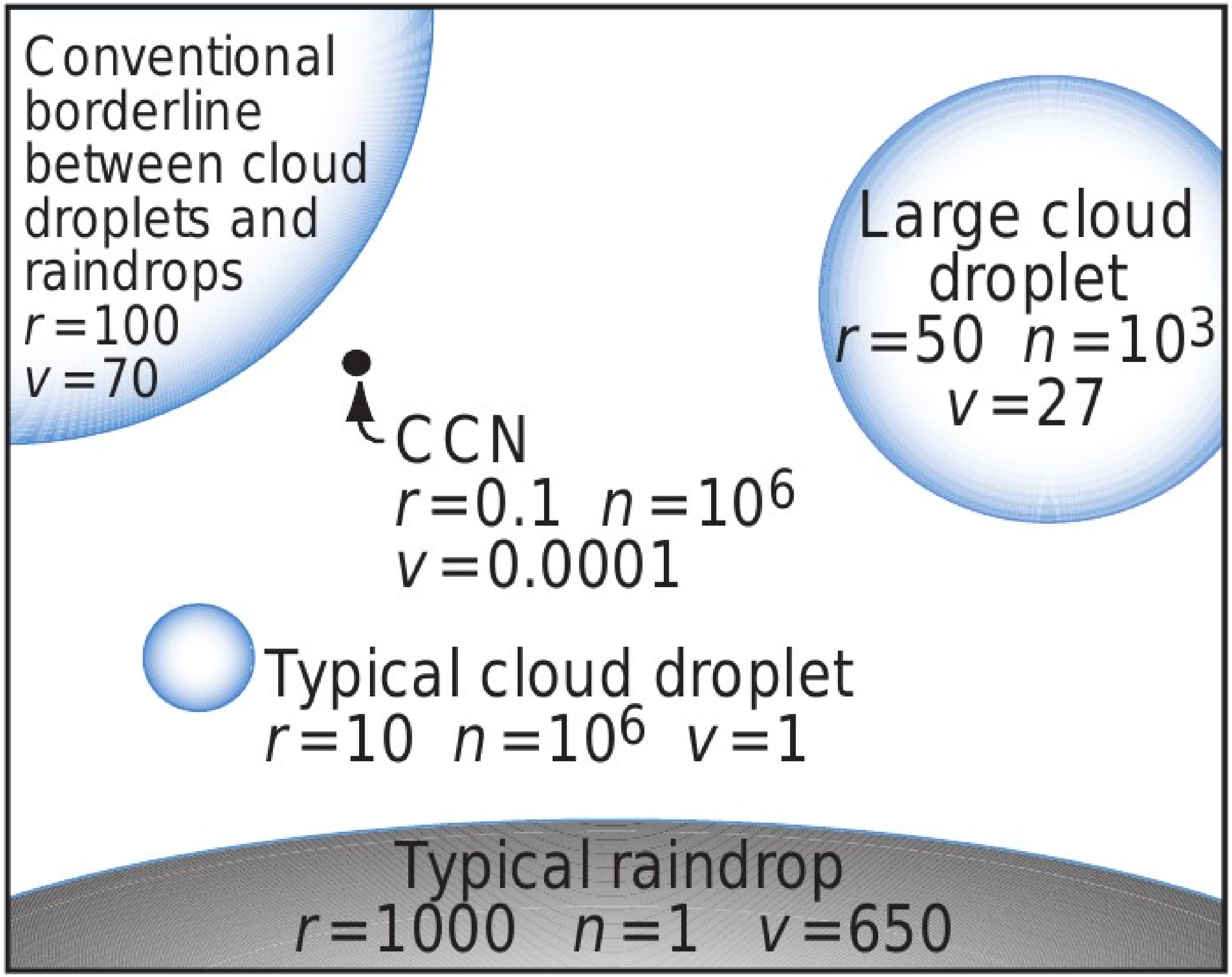
REL. SIZES OF CLOUD DROPLETS AND RAINDROPS



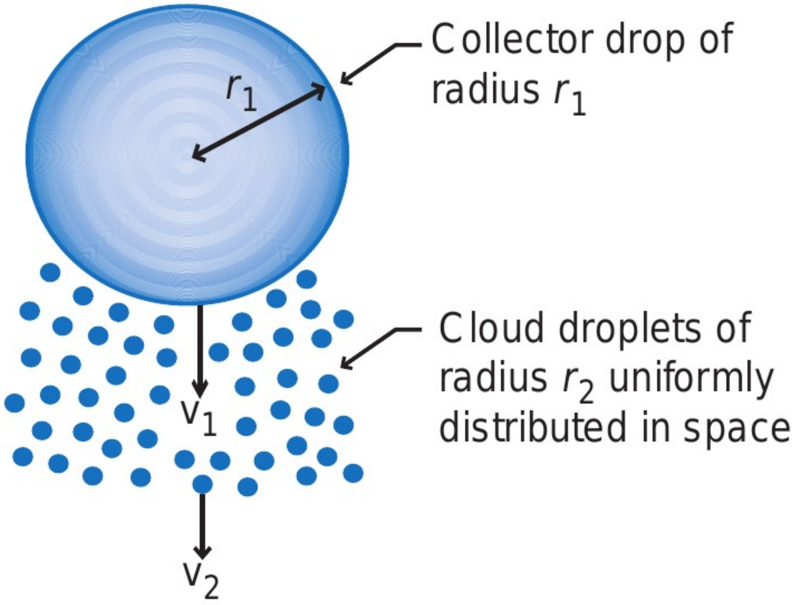
- r is the radius in micrometers
- n is the number per liter of air
- v is the terminal fall speed in cm/s

The circumferences of the circles are drawn approximately to scale, but the black dot representing a typical CCN is 25 times larger than it should be relative to the other circles.

Wallace & Hobbs

GROWTH OF DROPLETS

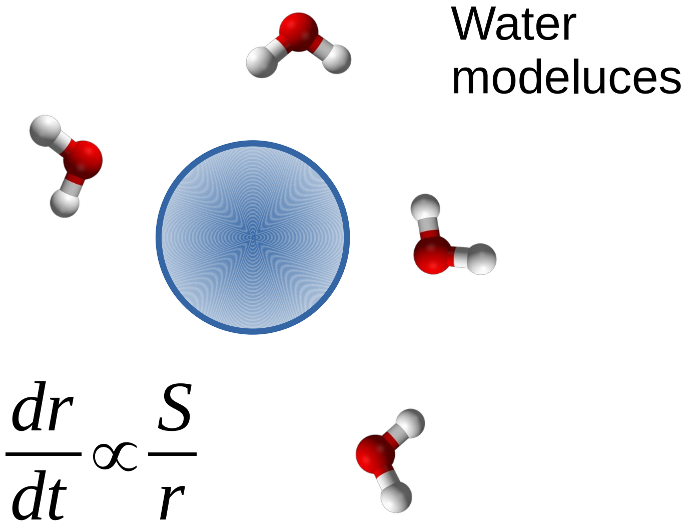
Collision of droplets



$$\frac{dr_1}{dt} \propto (v_1 - v_2)\, LWC_2\, E$$

E: Collection efficiency $\propto$ r

Diffusion of water vapor

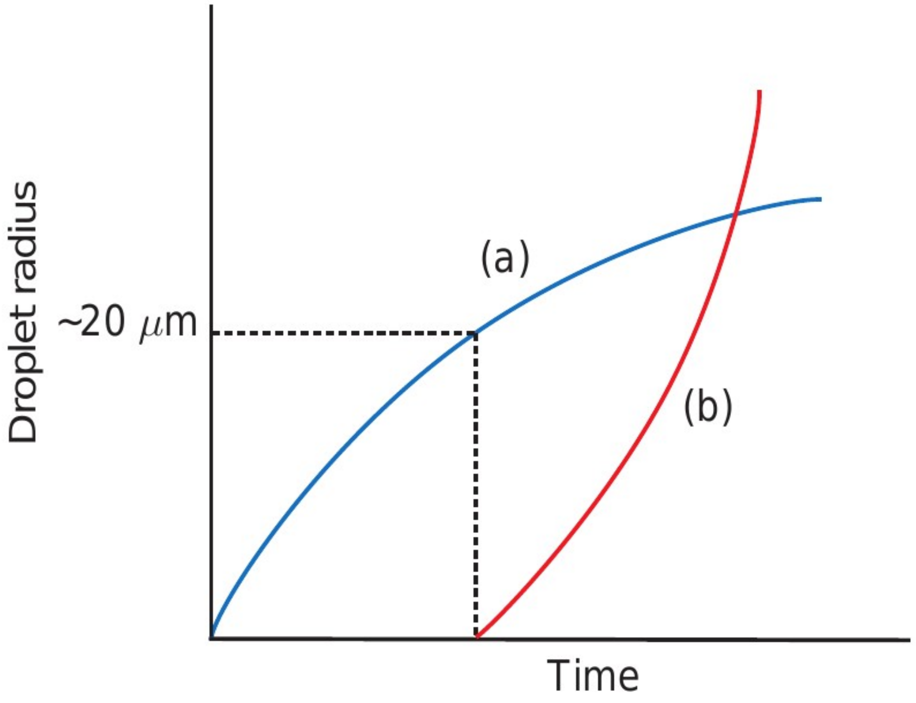


$$\frac{dr}{dt} \propto \frac{S}{r}$$

S: Supersaturation $(e-e_s)/e_s$

Change of radius depends on radius itsself and partial pressure of water vapor

GROWTH OF DROPLETS (COLLISION VS. DIFFUSION)

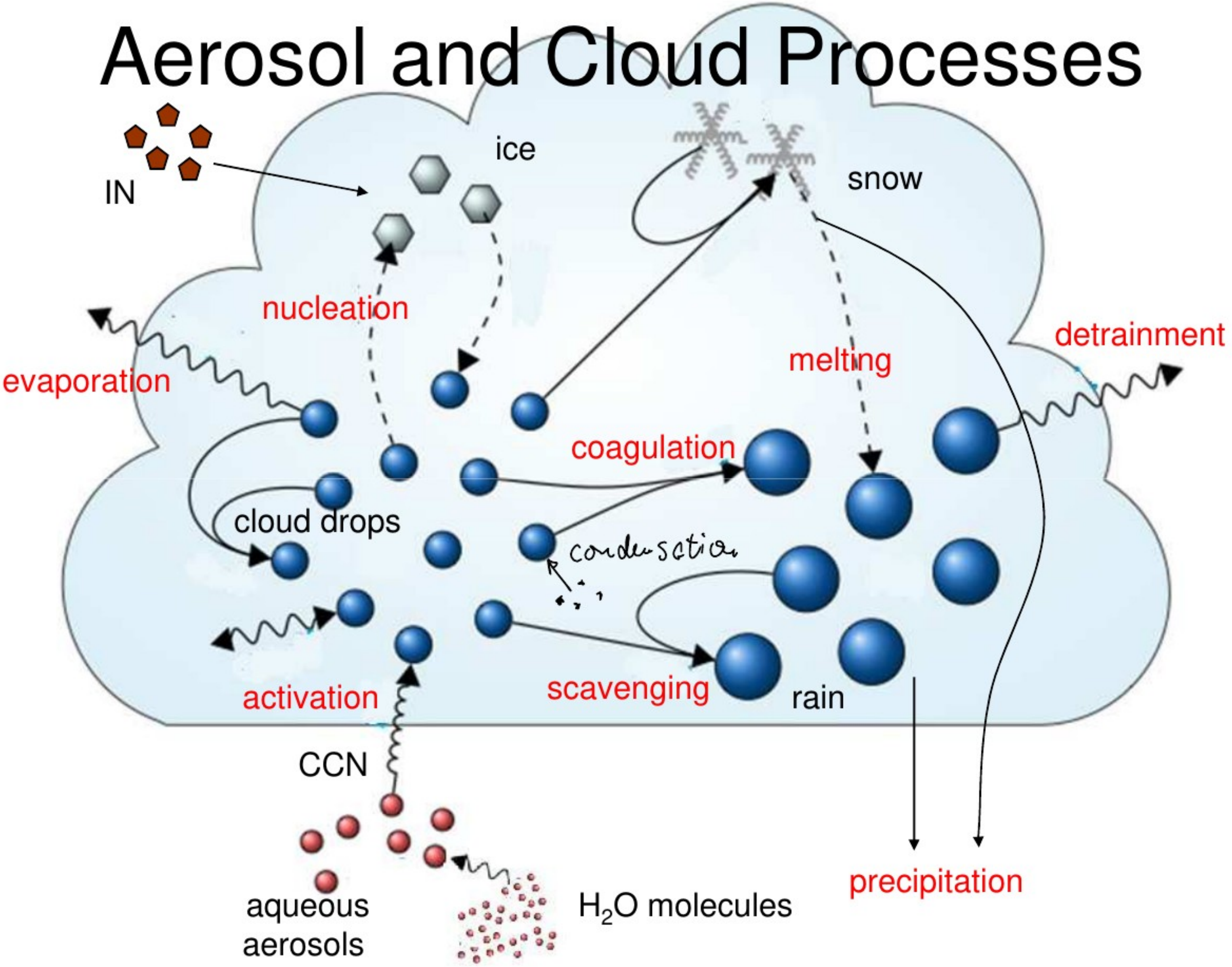


- Collision depends on weight & fall speed
- Diffusion depends on supersuration surface
- Dommination process depends on the environmental conditions but also on the droplet size

Fig. 6.15 Schematic curves of droplet growth (a) by condensation from the vapor phase (blue curve) and (b) by collection of droplets (red curve).

Wallace & Hobbs

Aerosol and Cloud Processes



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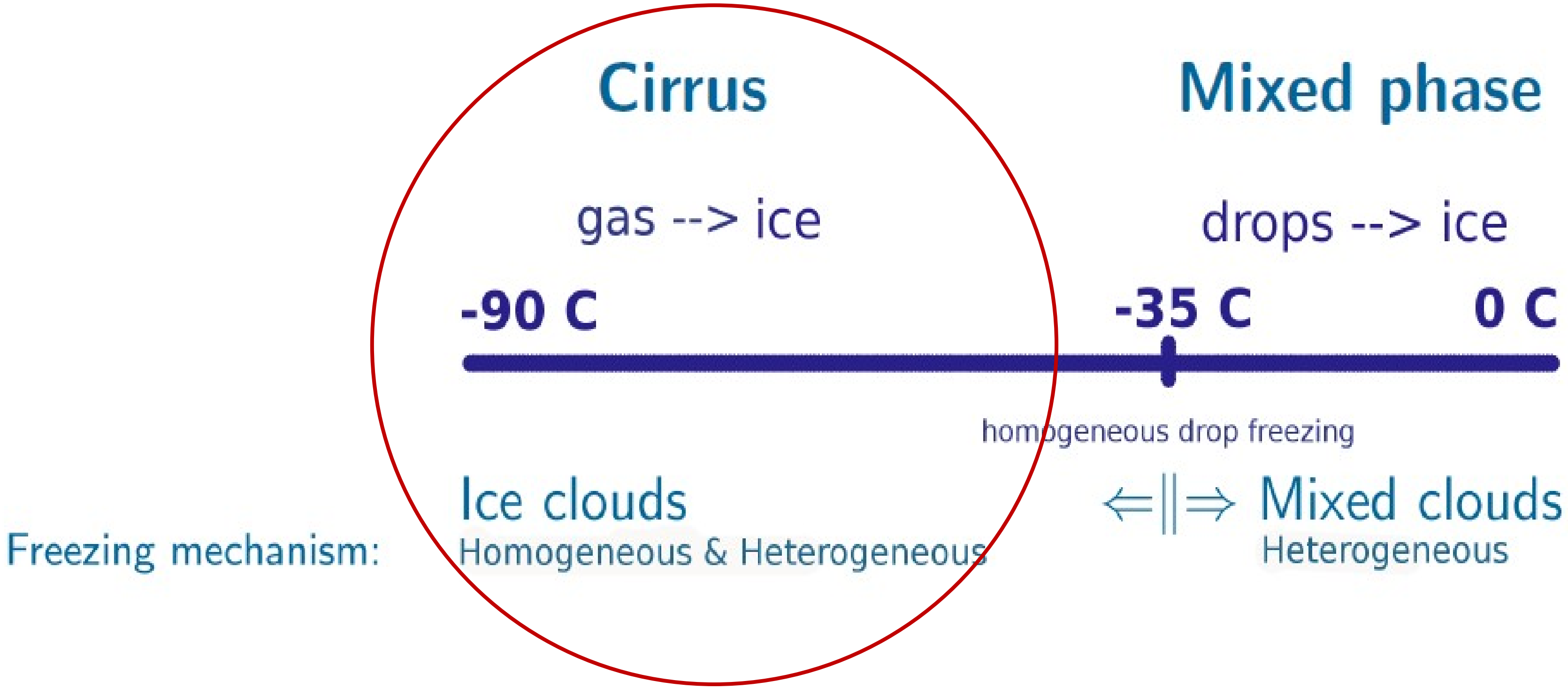
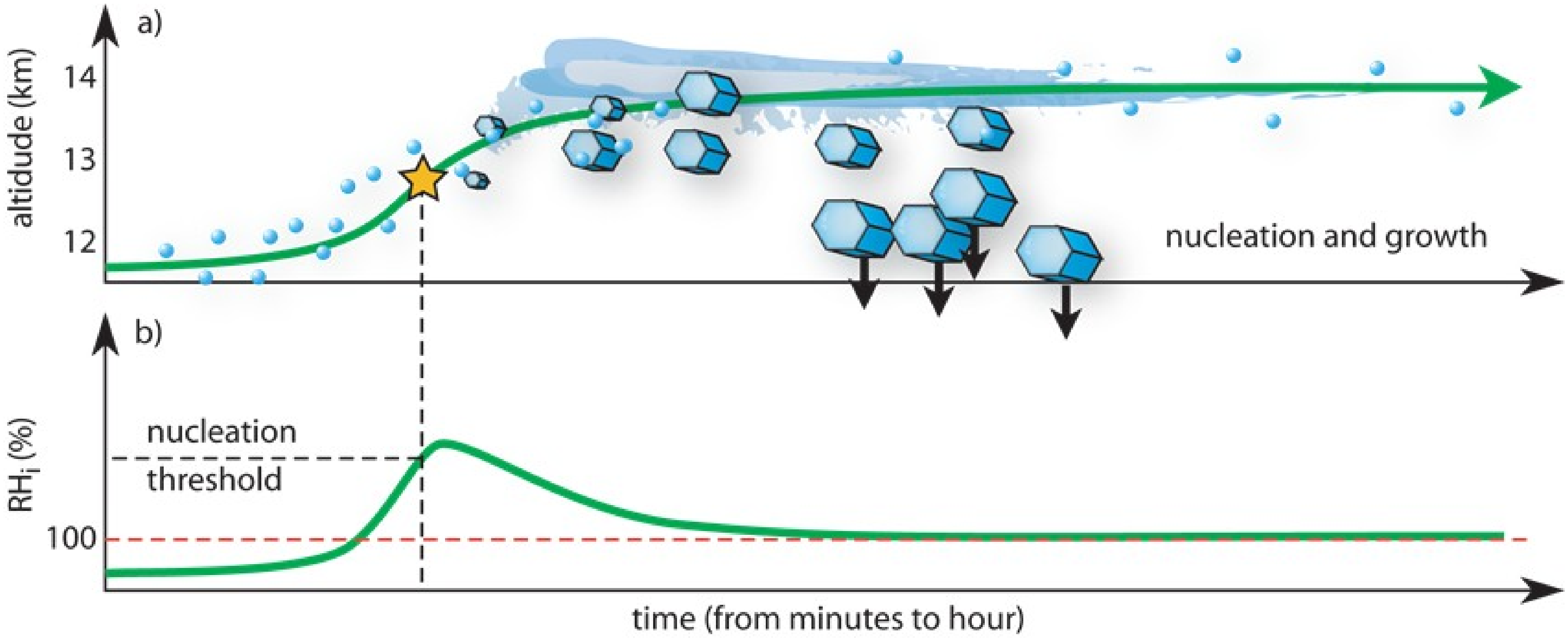
COLD CLOUDS IN THE ATMOSPHERE



Ice cloud life cycle



Adapted from Peter et al., 2005

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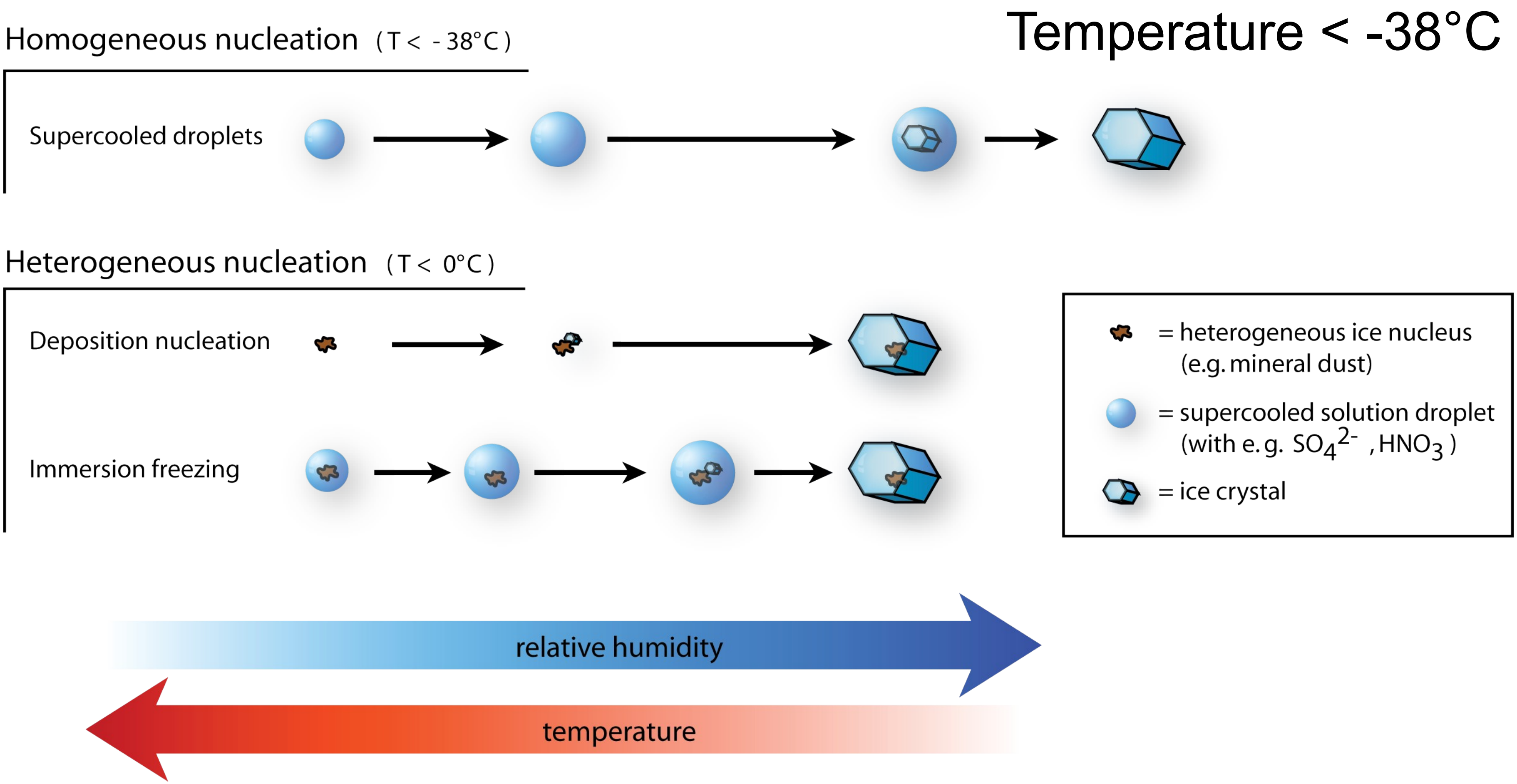
FREEZING MECHANISM OF CIRRUS CLOUDS



Temperature < -38°C

Homogeneous nucleation (T < -38°C)

Supercooled droplets

Heterogeneous nucleation (T < 0°C)

Deposition nucleation

Immersion freezing

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

= supercooled solution droplet (with e.g. SO_4^{2-}, HNO_3)

= ice crystal

relative humidity

temperature

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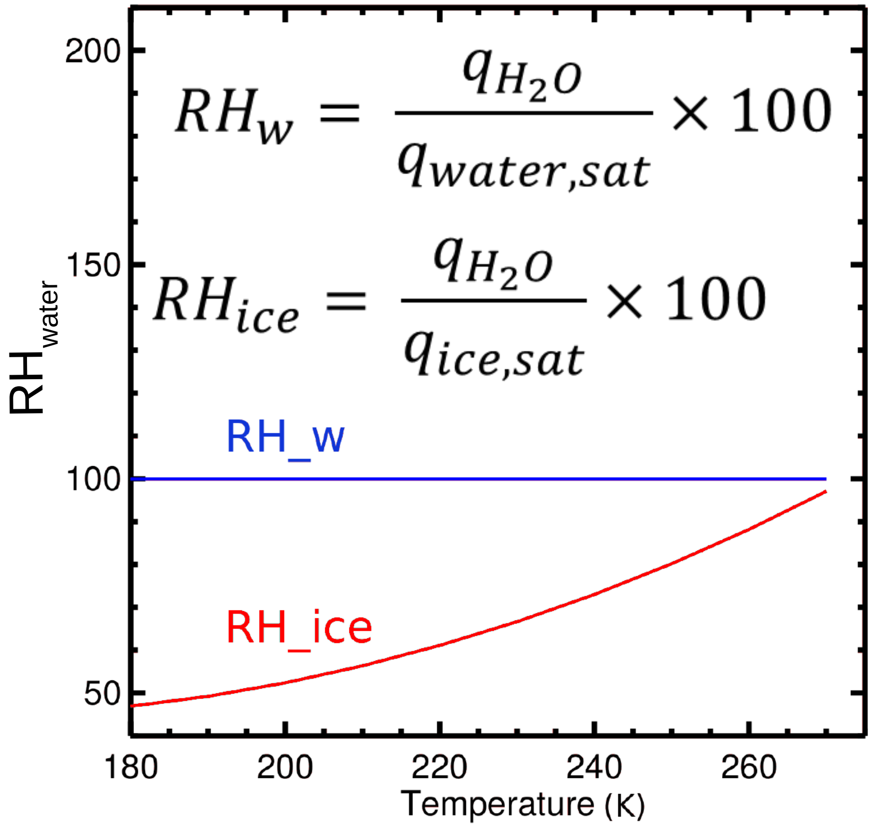
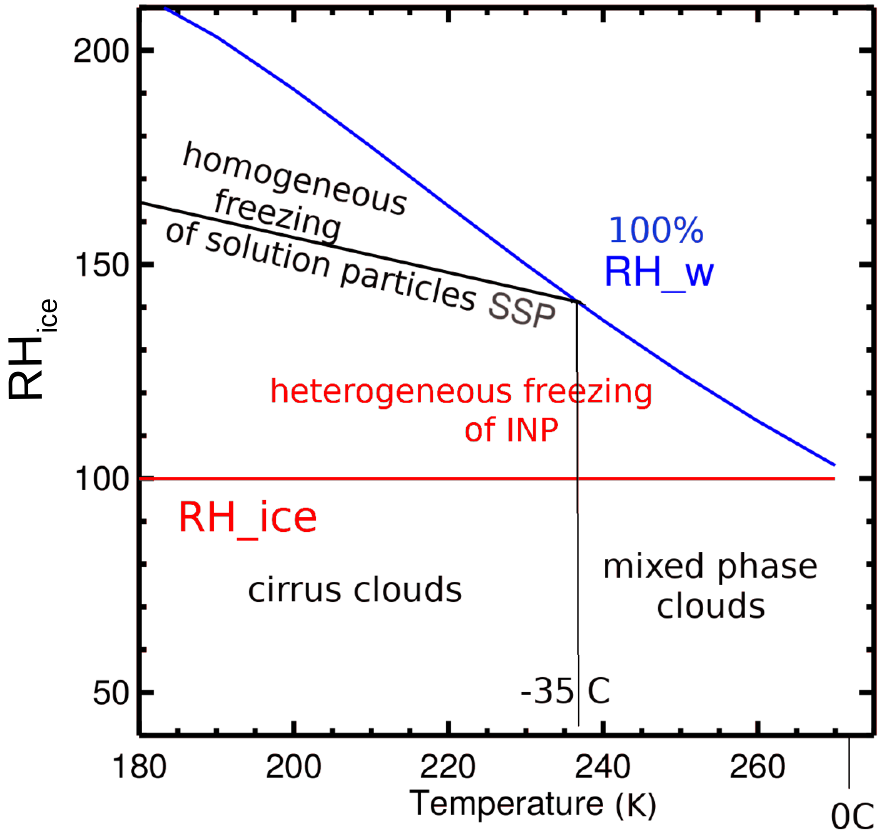
Relative humidity at cold conditions



$$RH_w = \frac{q_{H_2O}}{q_{water,sat}} \times 100$$

$$RH_{ice} = \frac{q_{H_2O}}{q_{ice,sat}} \times 100$$



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HOMOGENEOUS ICE NUCLEATION RATE

- Pure homogeneous nucleation of water molecules occurs below -38°C

  ➔ Freezing of super-cooled solution particles (SSP)

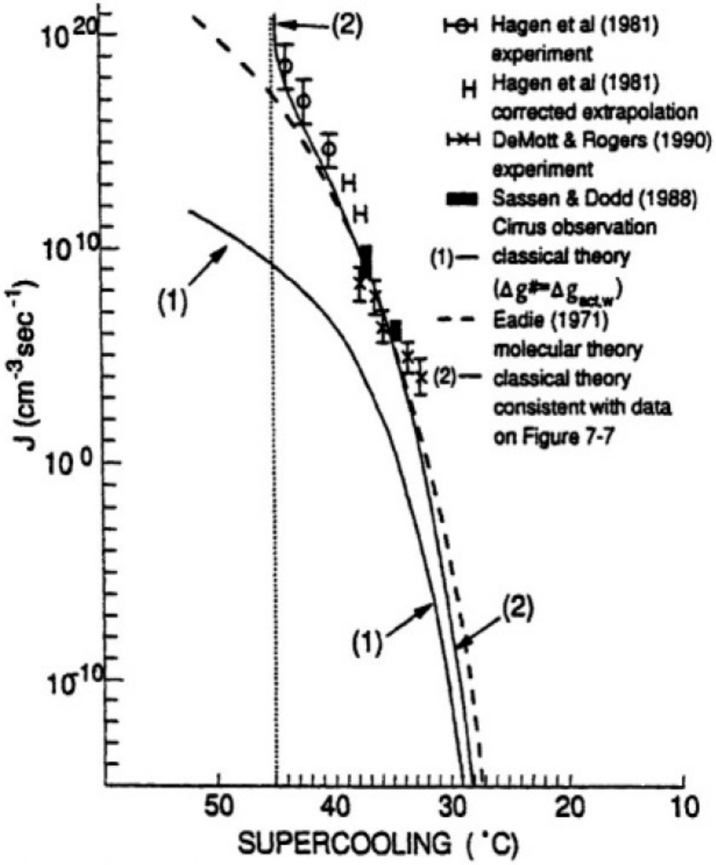
- Freezing of particle of volume V is a stochastic process

- Probability solution particle freezes within a timespan Δt:

$$P = 1 - \exp(-J \times V \times \Delta t)$$

J is the (homogeneous) nucleation rate

HOMOGENEOUS ICE NUCLEATION RATE



Pruppacher & Klett 1997

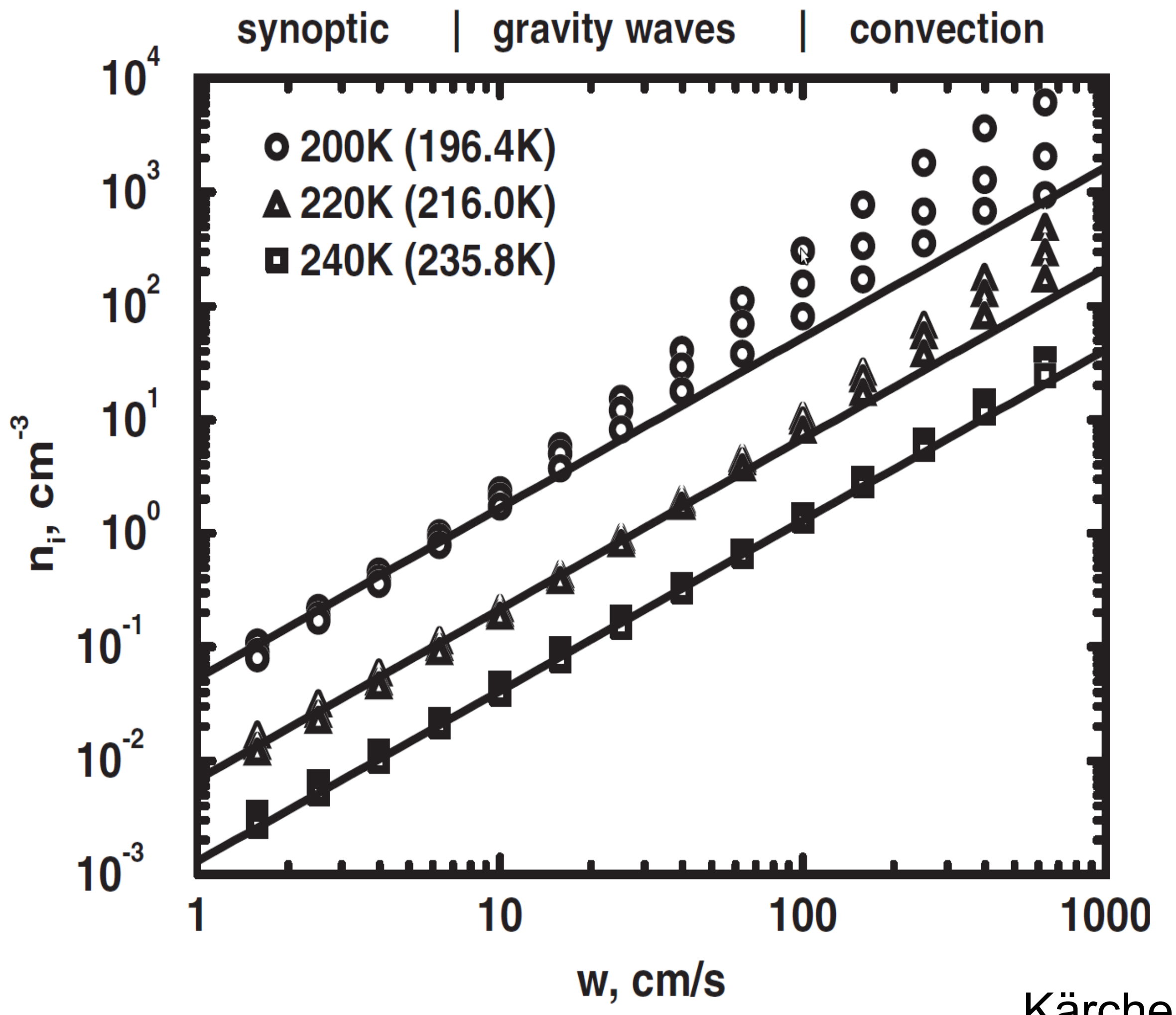
N_c: Number of water molecules
ρ: density of ice (i), liquid (l)
k: Boltzmann constant
T: Temperature
σ: Surface tension

$$J = 2N_c \left(\frac{\rho_l kT}{\rho_i h}\right) \left(\frac{\sigma_{i/l}}{kT}\right)^{1/2} \exp\left(-\frac{\Delta F_{act} + \Delta F_g}{kT}\right)$$

Activation energy

Germ formation energy (work against surface tension)

HOMOGENEOUS ICE NUCLEATION RATE



Kärcher and Lohmann (2002)

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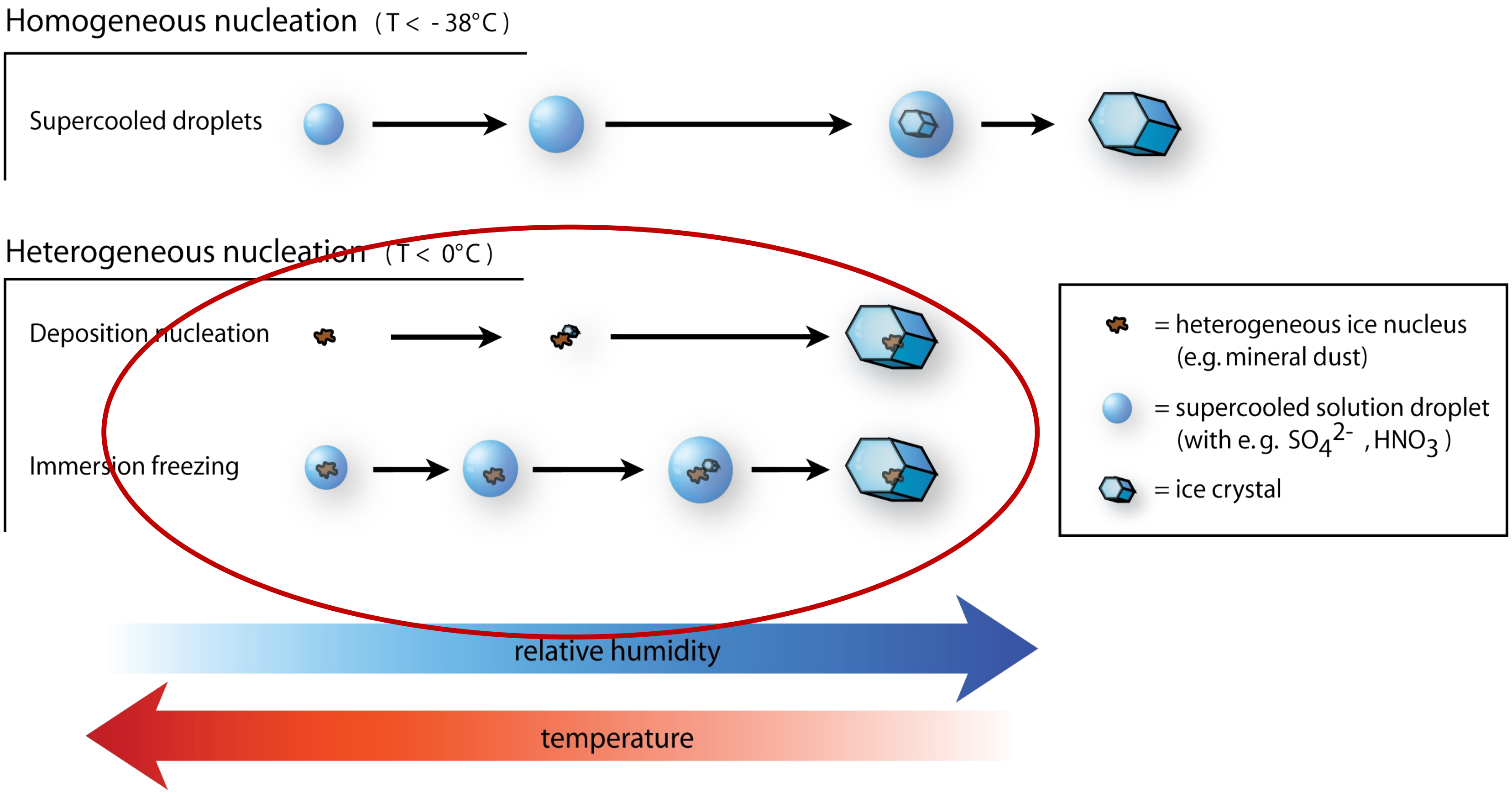
HETEROGENEOUS FREEZING



Homogeneous nucleation (T < -38°C)

Supercooled droplets

Heterogeneous nucleation (T < 0°C)

Deposition nucleation

Immersion freezing

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

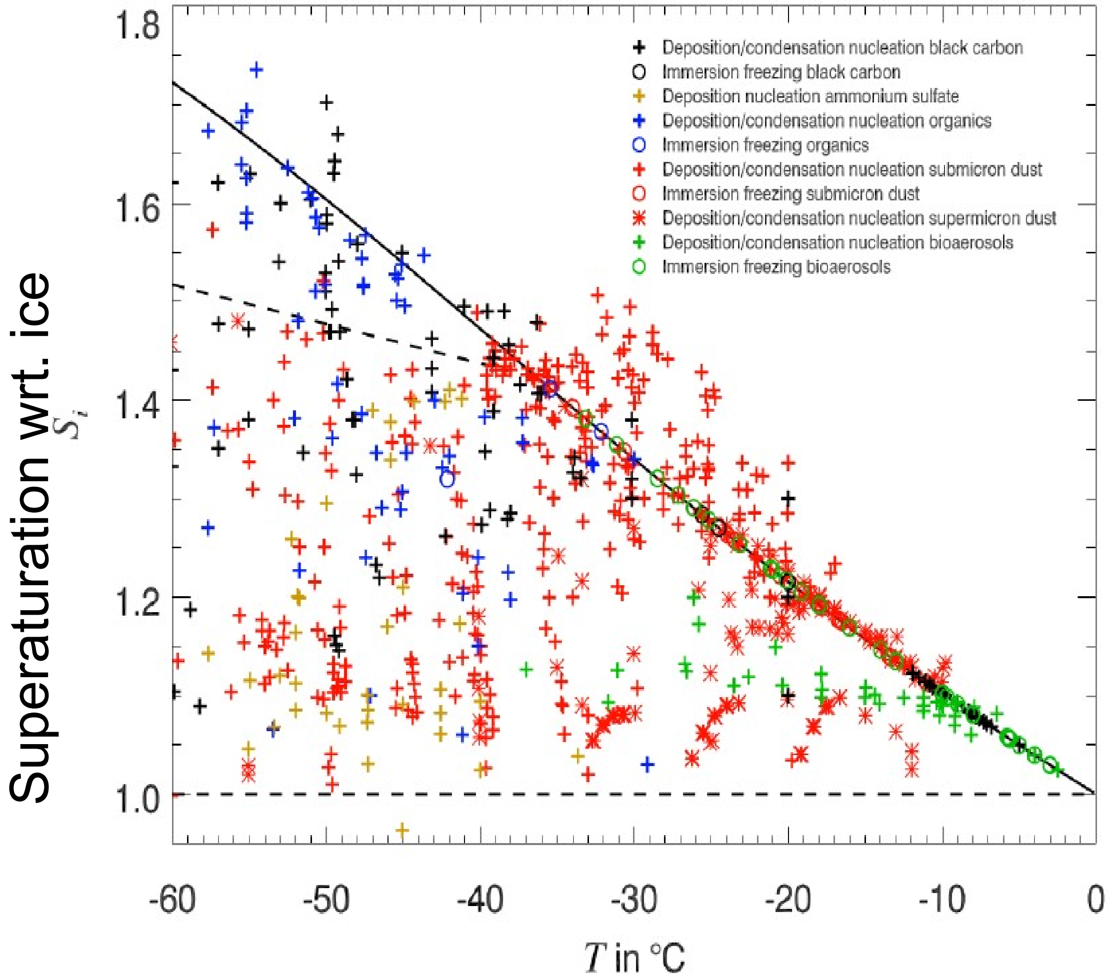
= supercooled solution droplet (with e.g. SO_4^{2-}, HNO_3)

= ice crystal

relative humidity

temperature

HETEROGENEOUS FREEZING



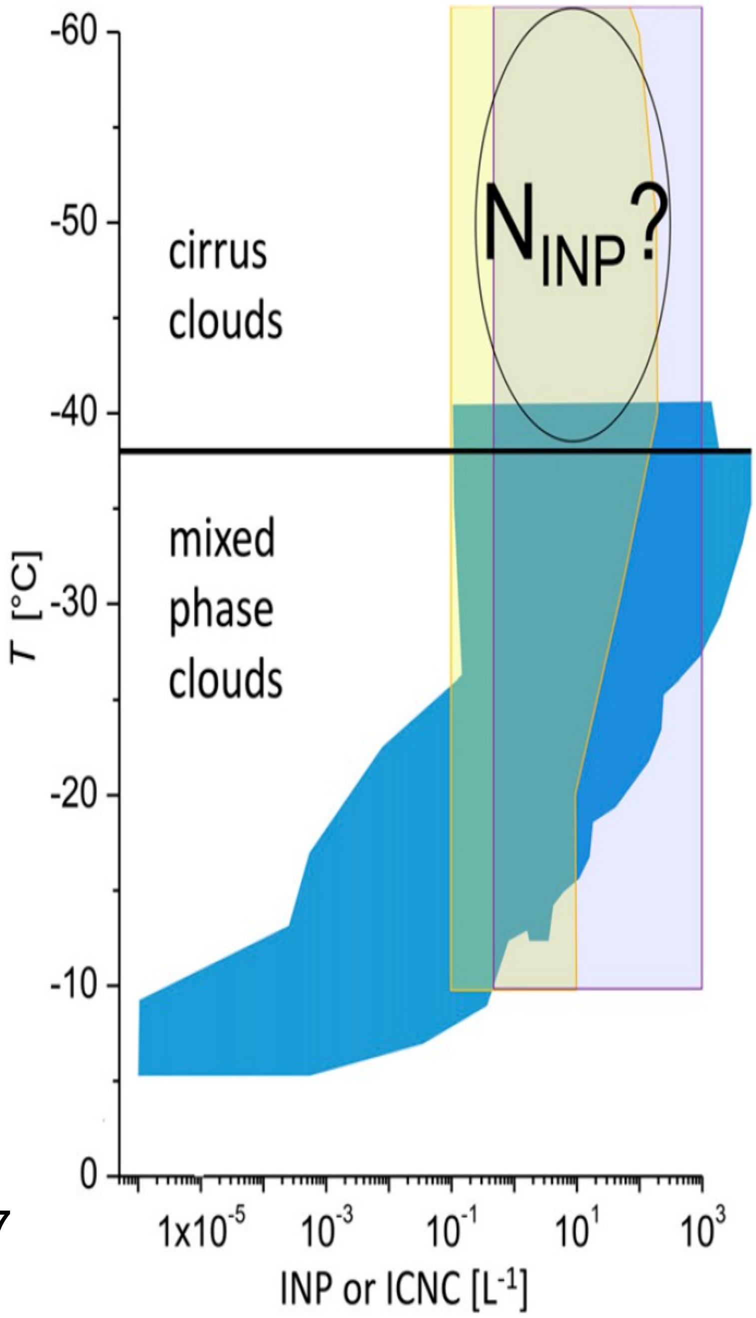
Heterogeneous freezing:

- INP chemistry and freezing activity are a focus of ice cloud research
- Heterogeneous freezing occurs at lower RH_{ice} than homogeneous

Hoose and Möhler, ACP (2012)

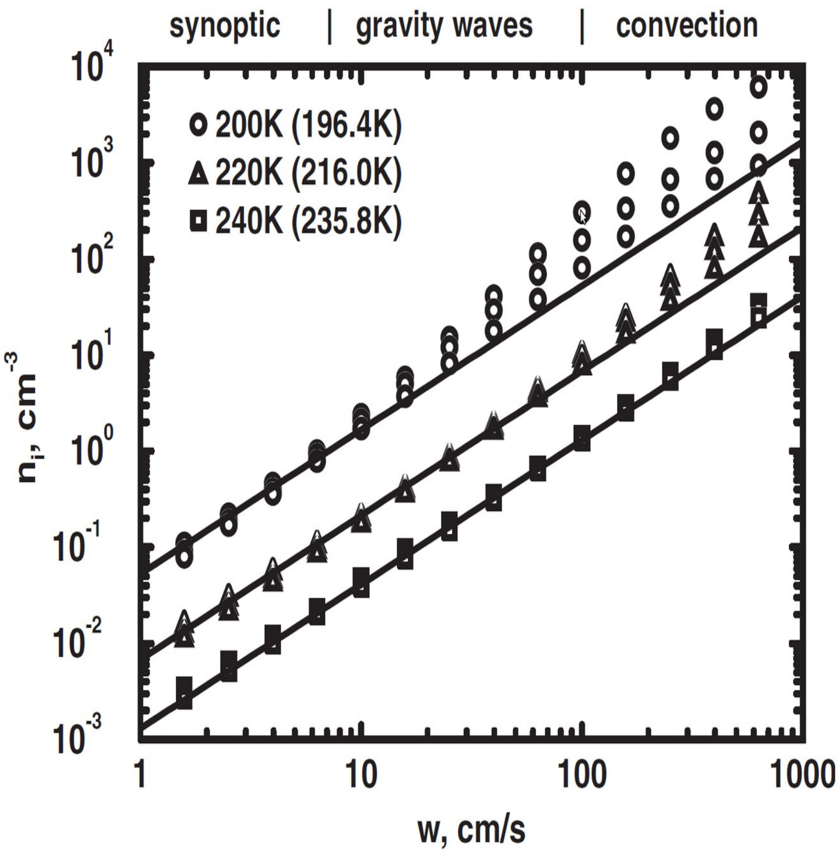
OBSERVATION OF INP

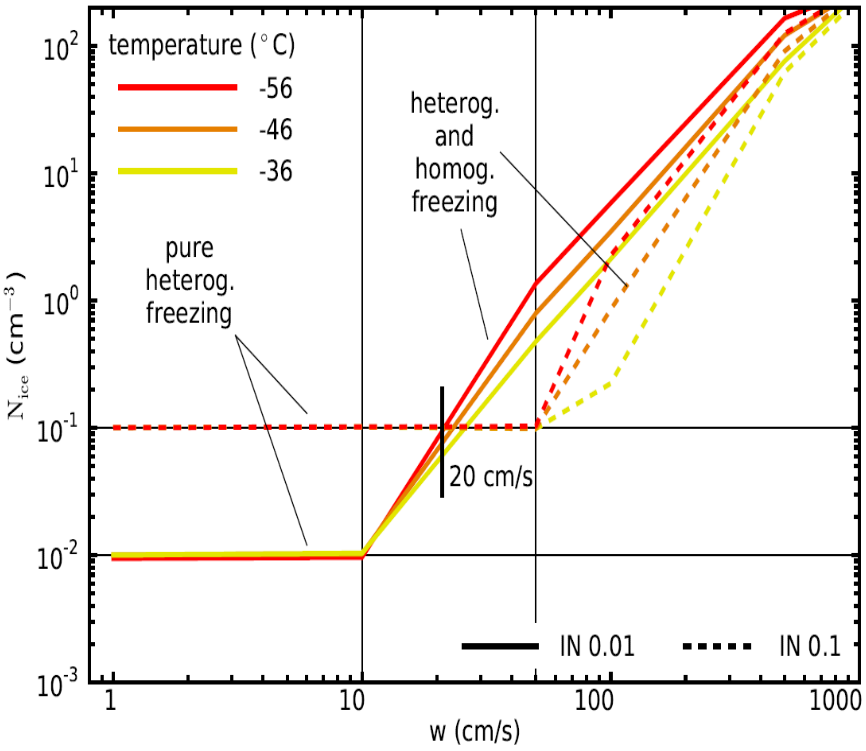
- Heterogeneous ice crystals numbers are a focus of ice cloud research
  (ice nucleating particles = INP)



Kanji et al., AMS, 2017

IMPACT ON ICE CRYSTAL NUMBER





Homogeneous freezing:

nearly no impact

Heterogeneous freezing:

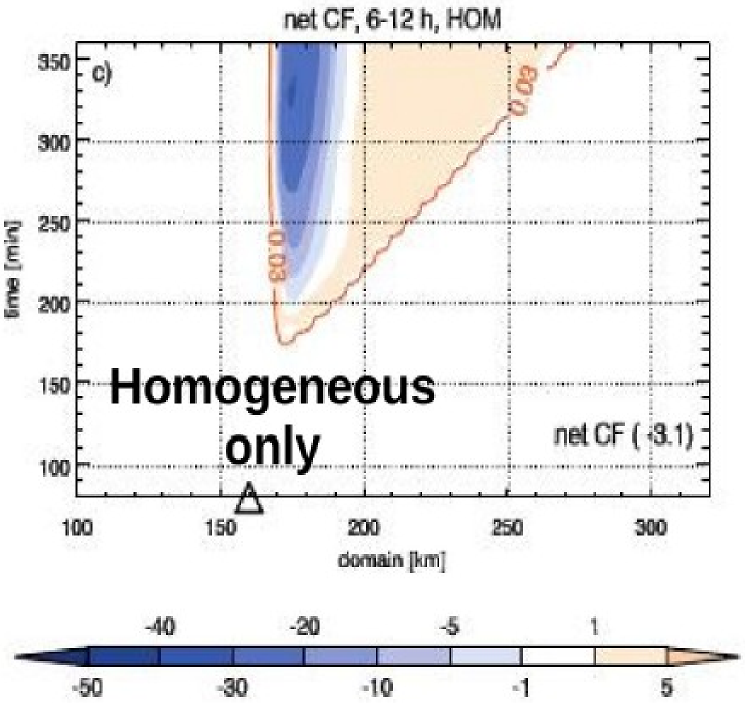
impact (i.e. mineral dust vulcanic ash, aircraft soot)

of aerosol number and composition on ice crystal number

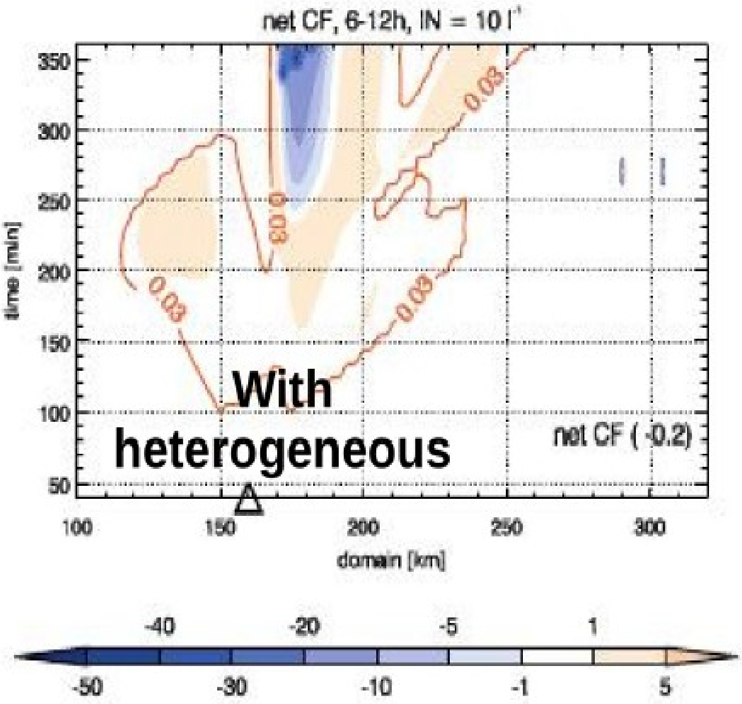
Kärcher and Lohmann (2002), JGR and Rolf et al. (2012)

HOMOGENEOUS VS. HETEROGENEOUS

Effect of the freezing mechanism on net cirrus forcing



Homogeneous only



With heterogeneous

Joos et al. (2014)

SUBTOPICS

3. Cloud formation (water and ice clouds)

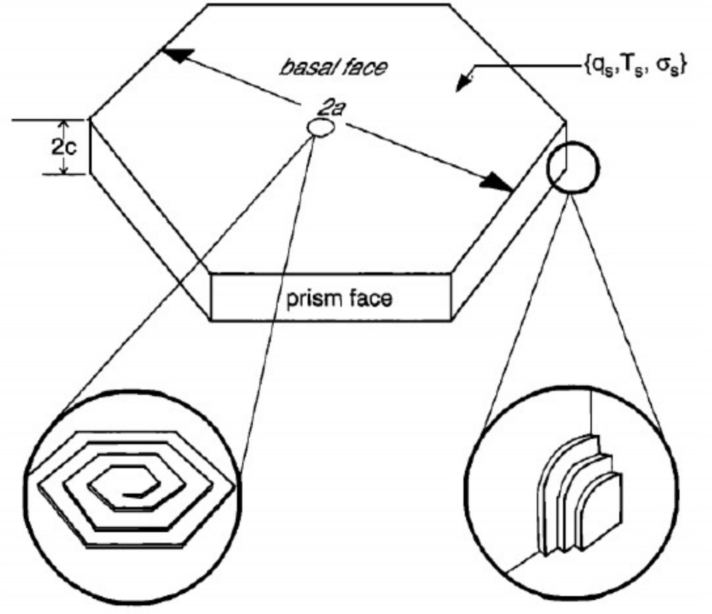
- Cloud types
- Cloud properties and saturation vapor pressure
- *Warm clouds*
  - Raoult’s law, Kelvin effect, Koehler curve
  - Cloud condensation nuclei (CCN)
  - Cloud droplet growth and other processes
- *Ice clouds / Ice cloud life cycle*
  - Freezing process: Homogeneous nucleation
  - Freezing process: Heterogeneous nucleation
  - **Ice crystal growth, shapes and sedimentation**
- *Mixed phase clouds*
  - Bergeron-Findeisen Process

ICE CRYSTAL GROWTH

Classical diffusion of water molecules

$$\frac{dm}{dt} \simeq \frac{4\pi D\beta\kappa\phi M_w}{R^*T}(p_{\infty,w} - p_{\mathrm{sat},i})$$

R*: Universal gas constant
T: temperature
D: diffusion coeff.
β: Mass deposition coeff.
k: capacity factor (shape dependent)
Φ: ventilation factor
M_w: Molar weight of water
p_w: partial pressure of water vapor
p_i: saturation partial pressure wrt. Ice

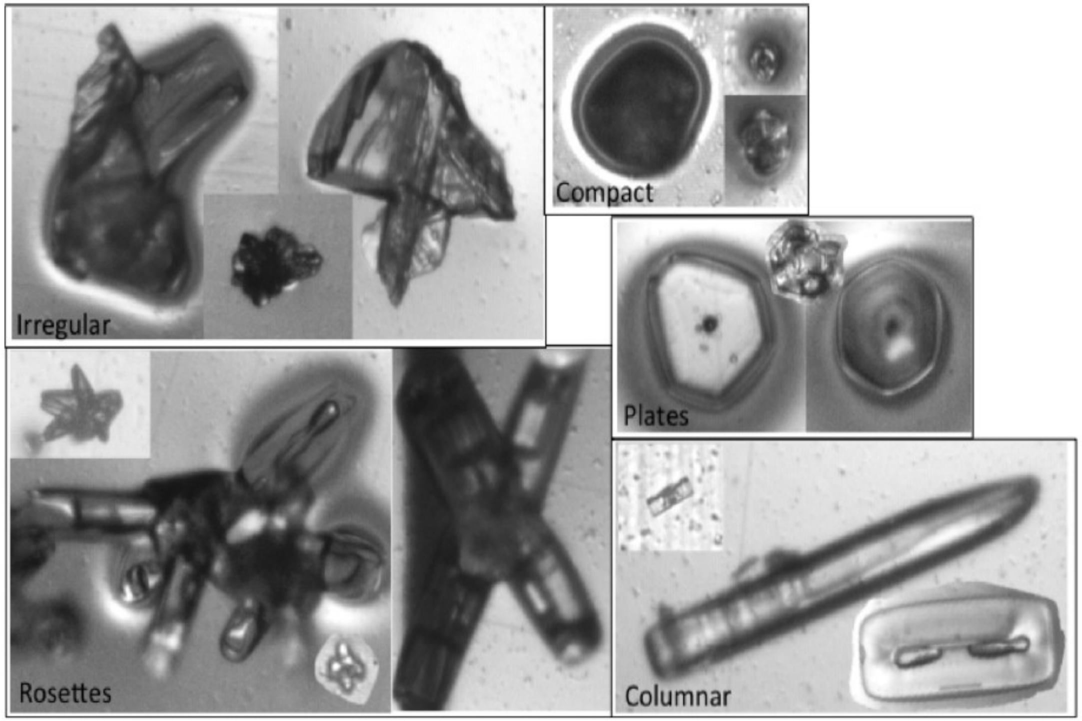


screw (spiral) dislocation

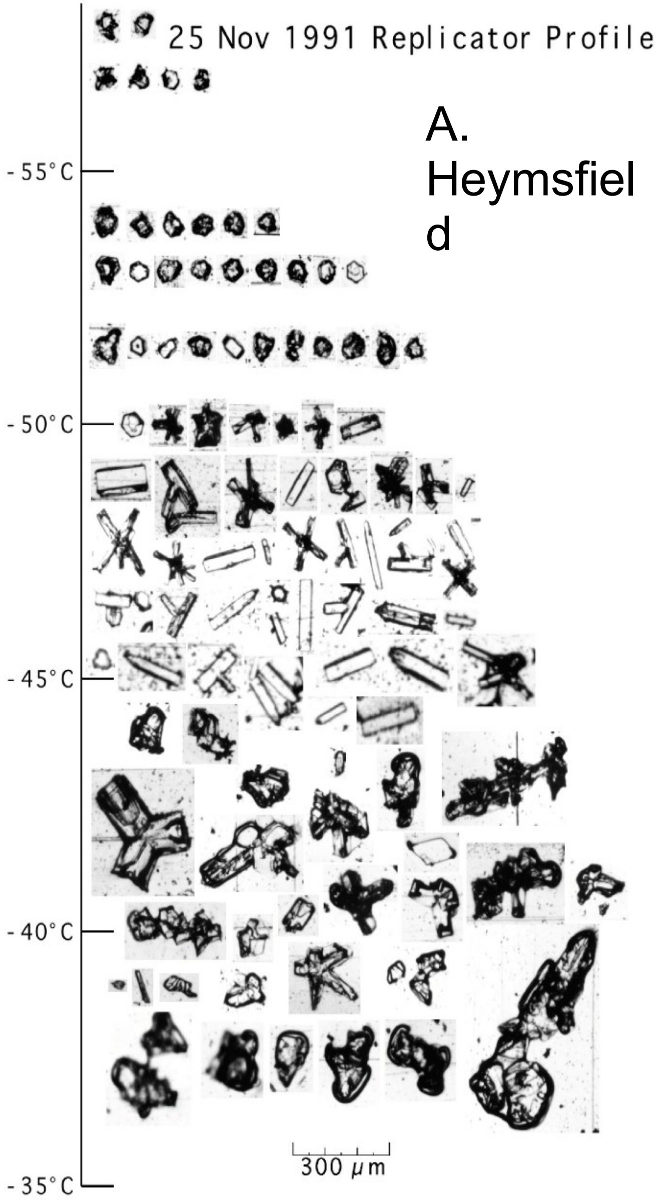
two-dimensional nucleation

Manly depend on partial pressure difference
➔ Sub-saturated → negative → decreasing mass
➔ Supersaturated → positive → increasing mass

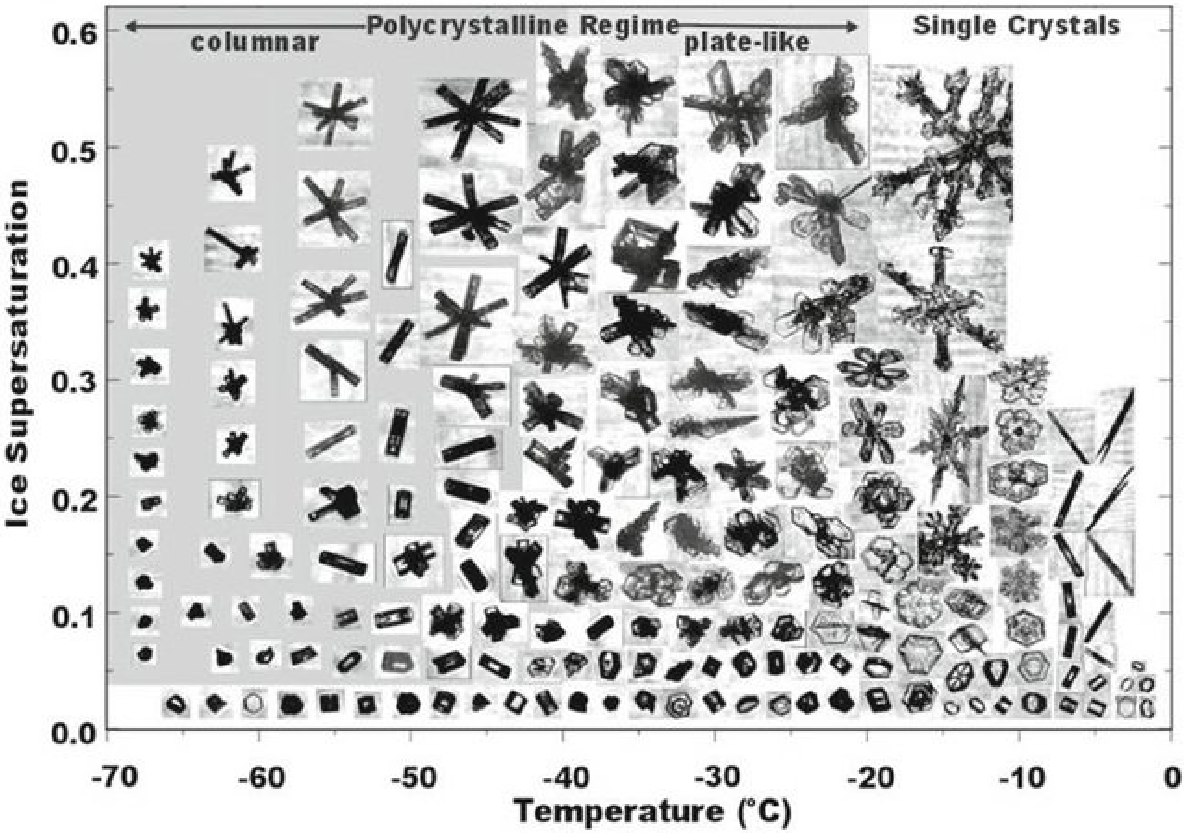
ICE PARTICLE SHAPES



Wolf et al., ACPD 2018

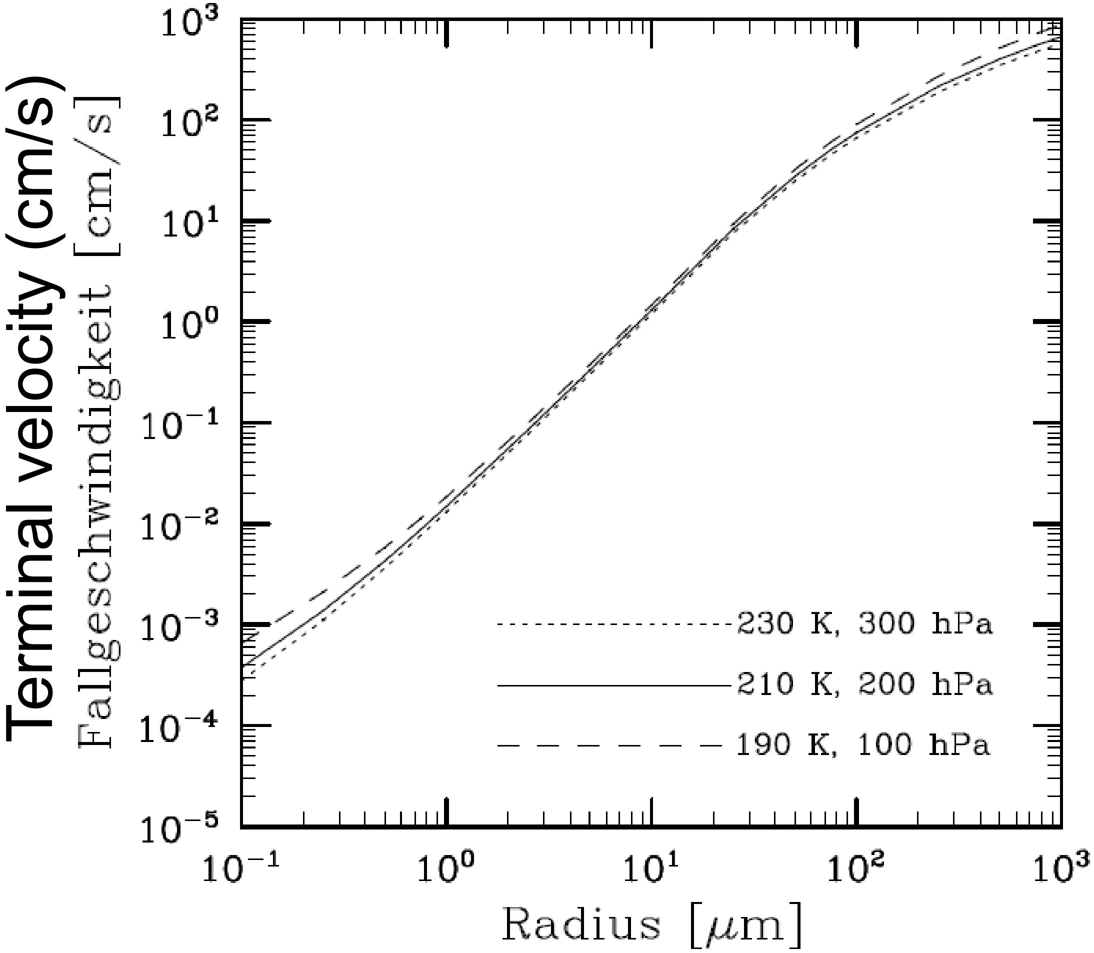


A. Heymsfield



Diedenhoven 2018
Remote Sensing of
Crystal Shapes in Ice
Clouds

SEDIMENTATION OF ICE CRYSTALS



Haag, 2003

SUBTOPICS

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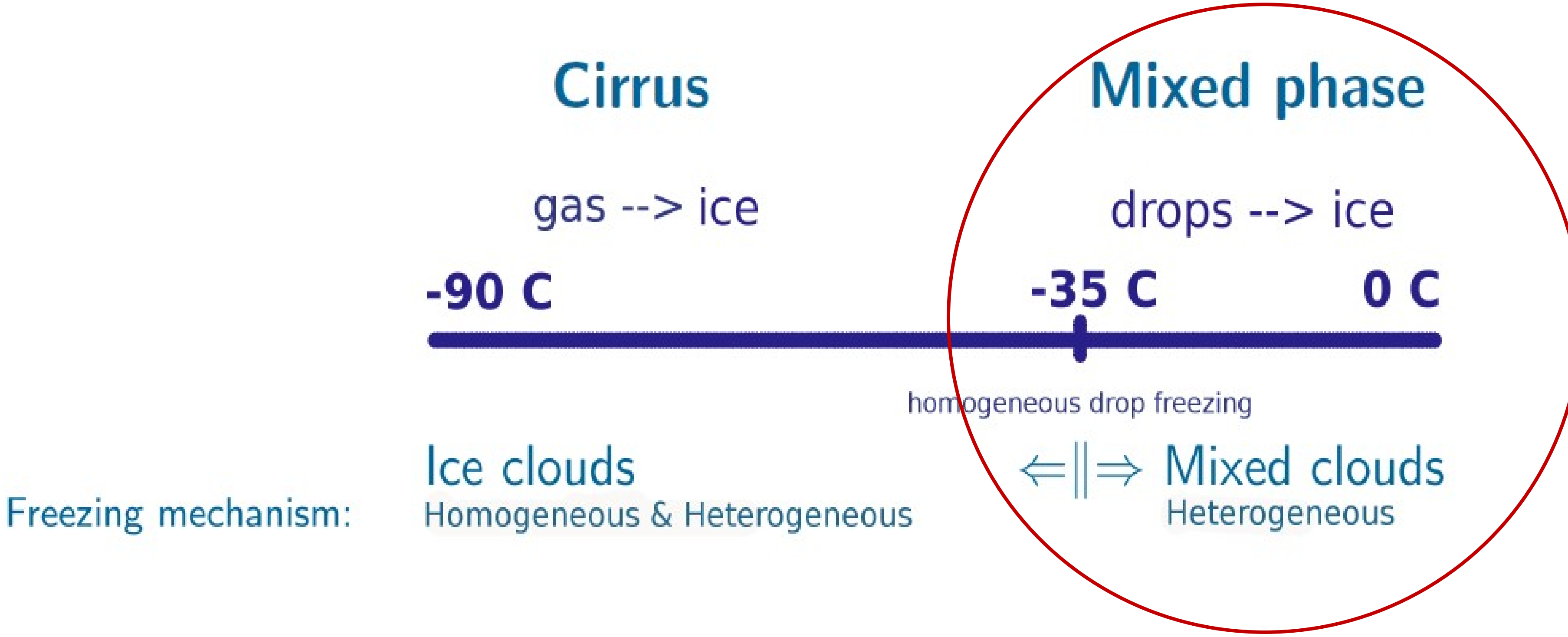
MIXED PHASE CLOUDS



Cirrus

gas --> ice

Mixed phase

drops --> ice

-90 C

-35 C

0 C

homogeneous drop freezing

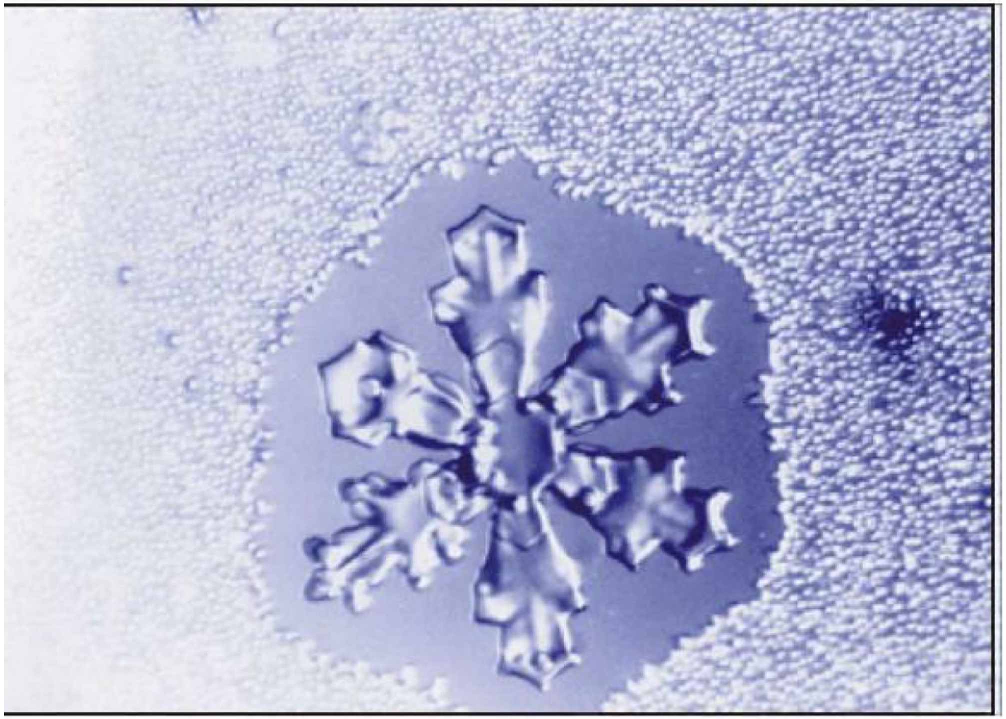
Freezing mechanism:

Ice clouds
Homogeneous & Heterogeneous

⇐‖⇒ Mixed clouds
Heterogeneous

MIXED PHASE CLOUDS

Thunderstrom



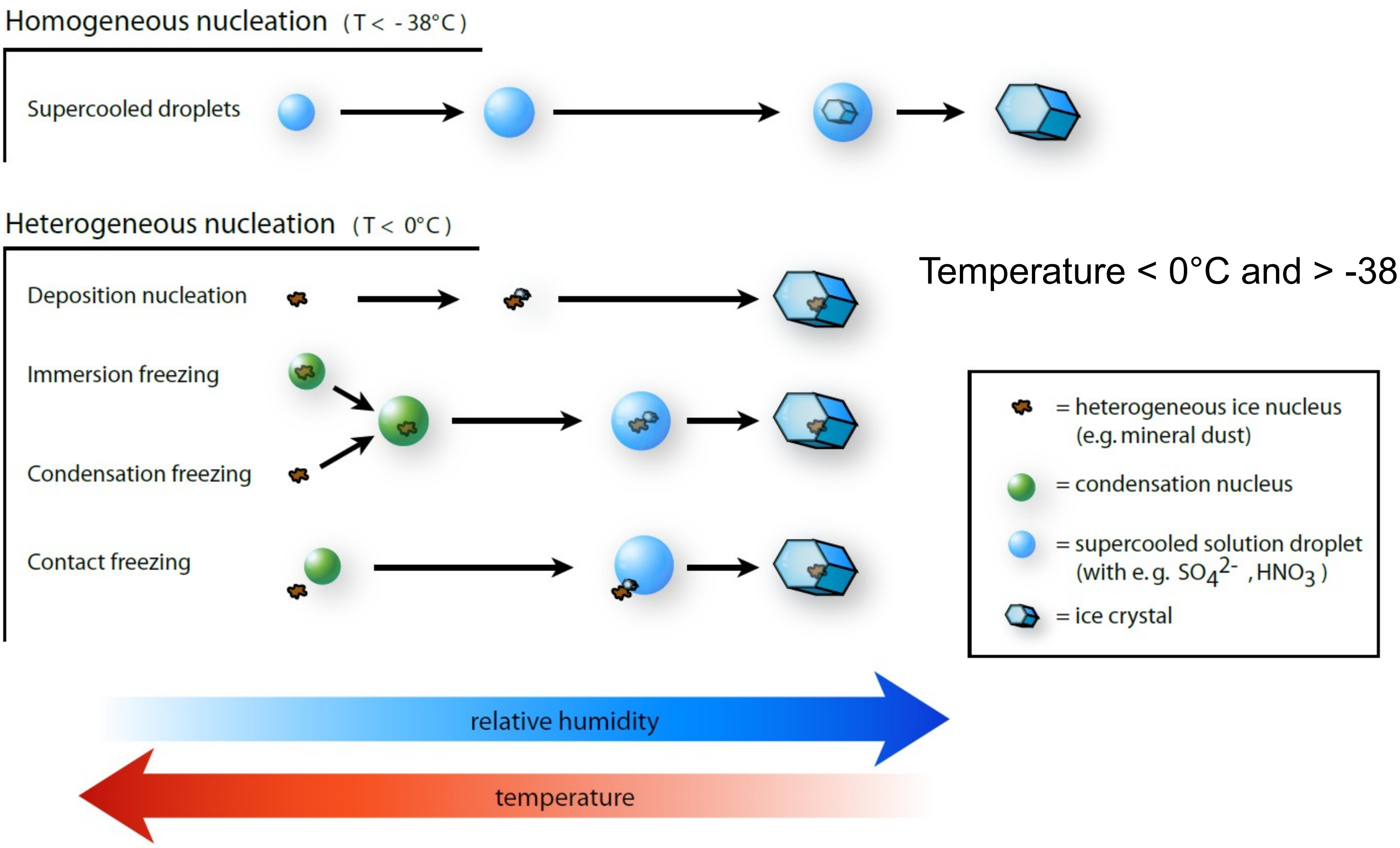
EFFECT OF PHASE DIFFERENCE

R.L. Pitter

Coexistence of drops and ice crystals

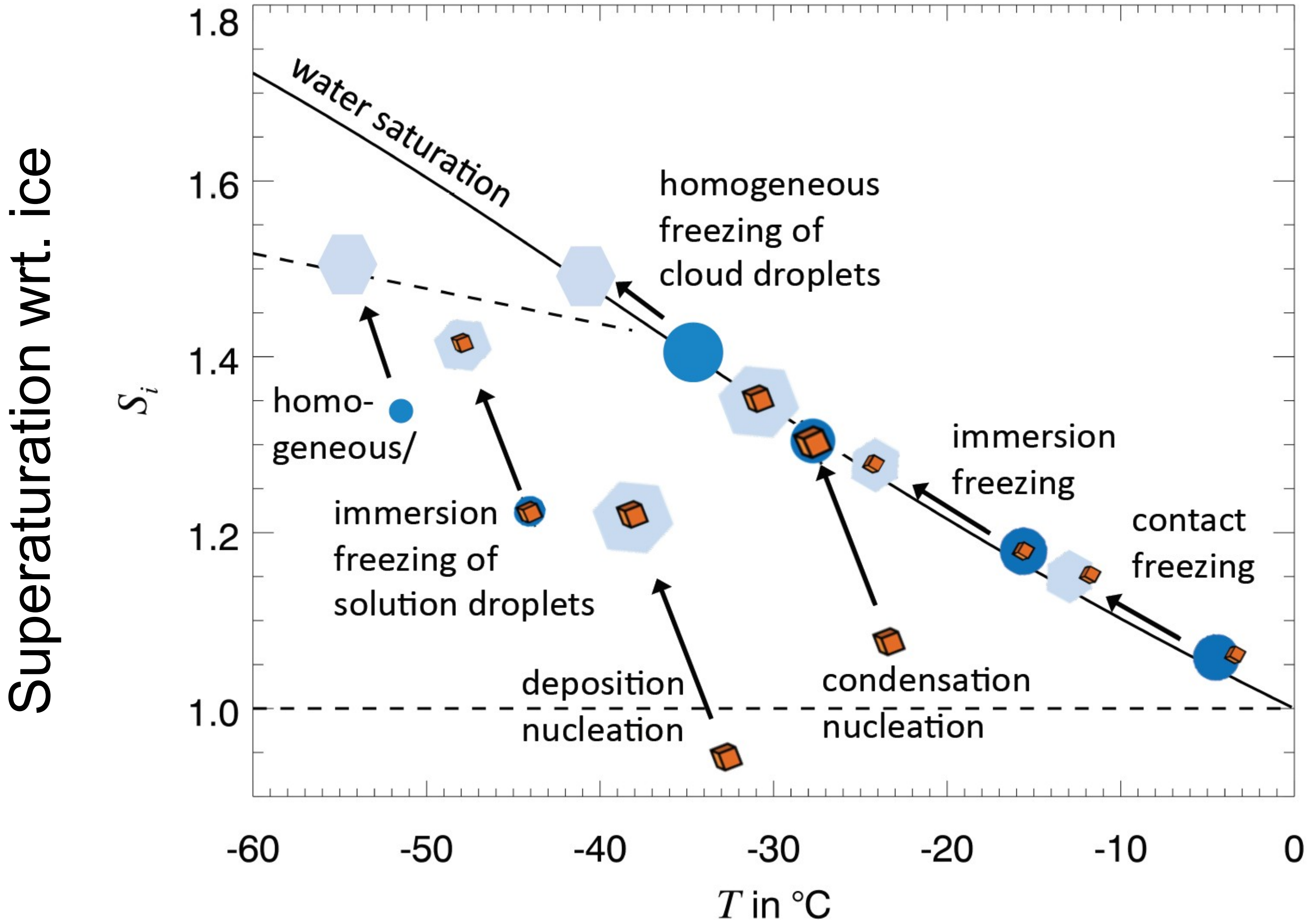
Pictures are taken from
Karlsruher Wolkenatlas
http://www.wolkenatlas.de

MIXED PHASE CLOUD FORMATION



Temperature < 0°C and > -38°C

MIXED PHASE CLOUDS



Hoose & Möhler, ACP; 2012

SUBTOPICS

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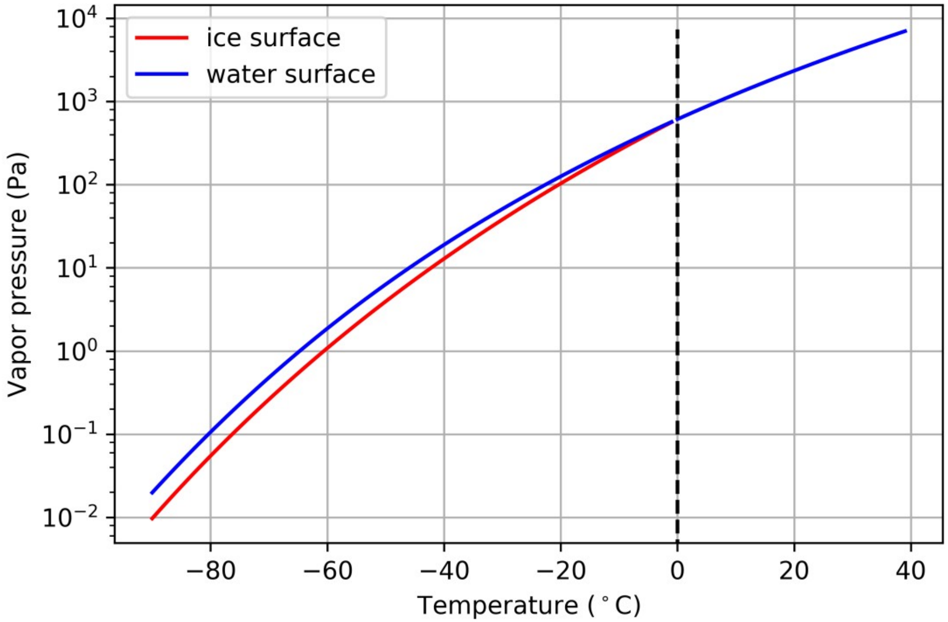
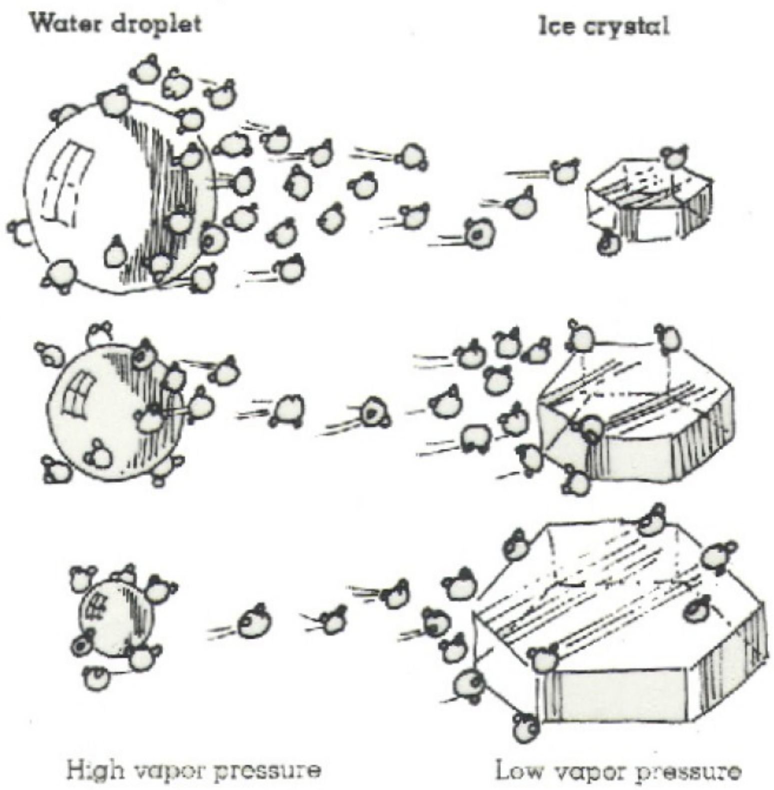
GLACIATION OF MIXED PHASE CLOUDS

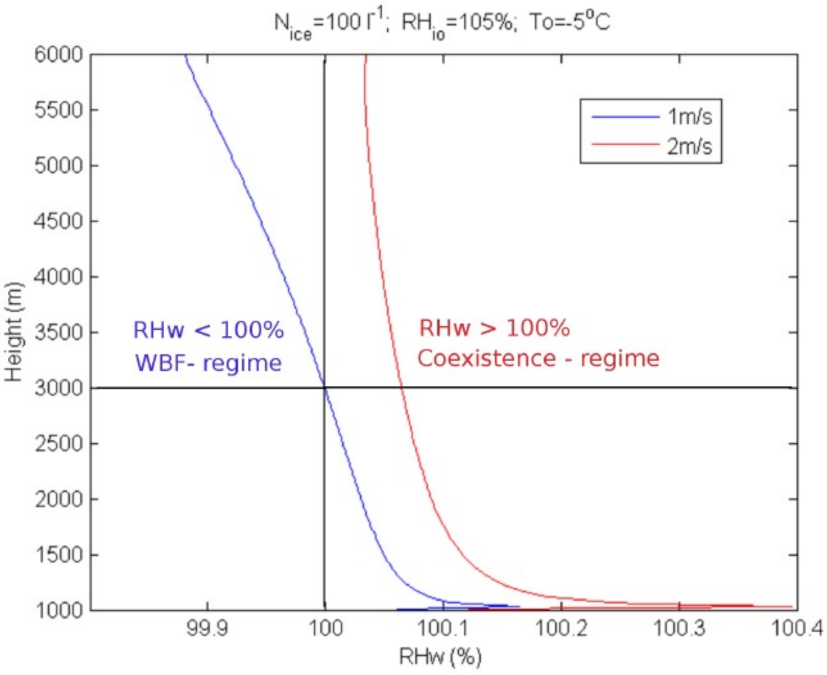
Bergeron - Findeisen Process

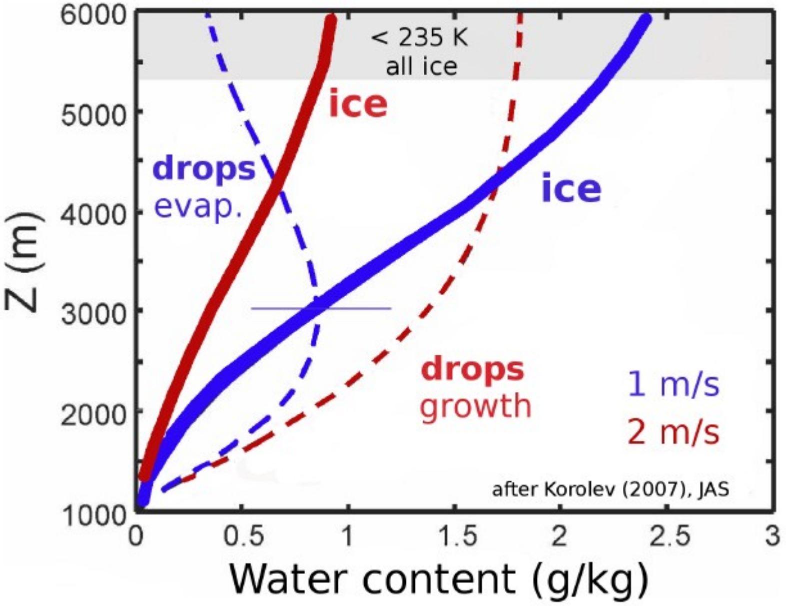




- $RH_w < 100\%$, $RH_{ice} > 100\%$
  - ➢glaciation increases
  - ➢rain formation

Bergeron - Findeisen Process





updraft 1 m/s: $RH_w < 100\%$ ➡ complete glaciation (Berg-Find)

updraft 2 m/s: $RH_w > 100\%$ ➡ drop - ice coexistence