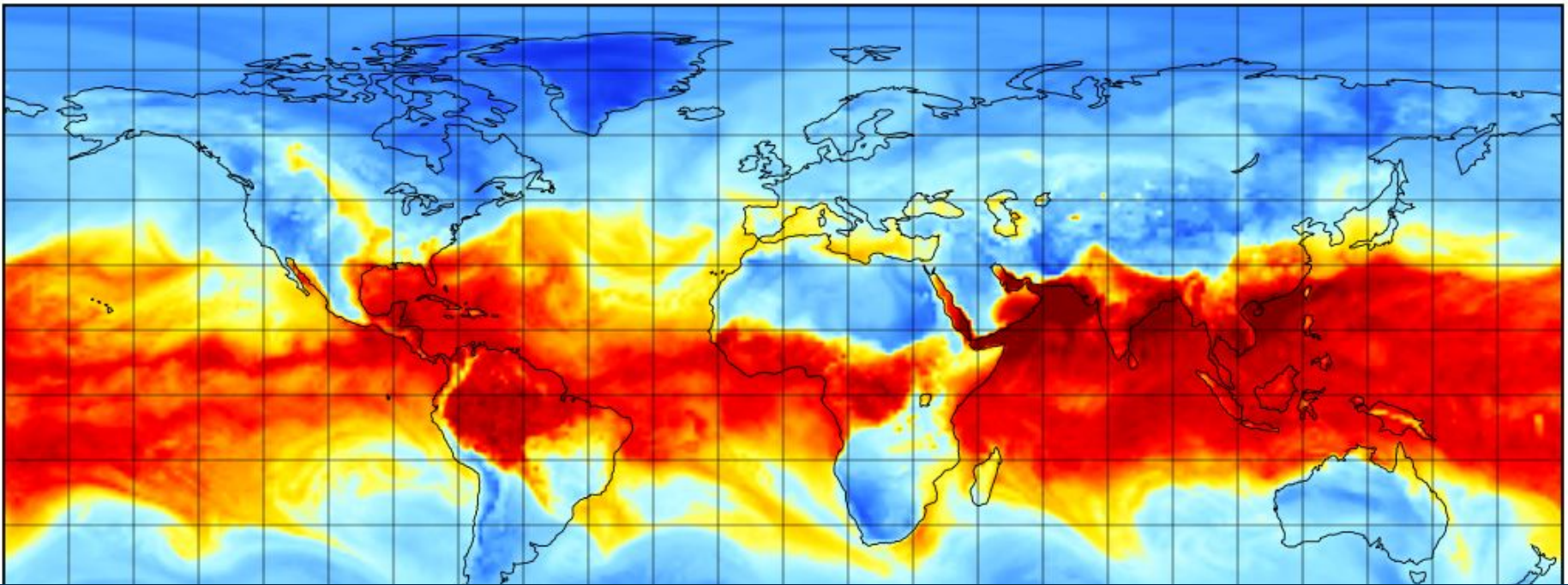




WATER IN THE ATMOSPHERE AND THE ROLE FOR CLIMATE

Part 3: Cloud formation (water and ice clouds)

WS 25/26 | CHRISTIAN ROLF

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

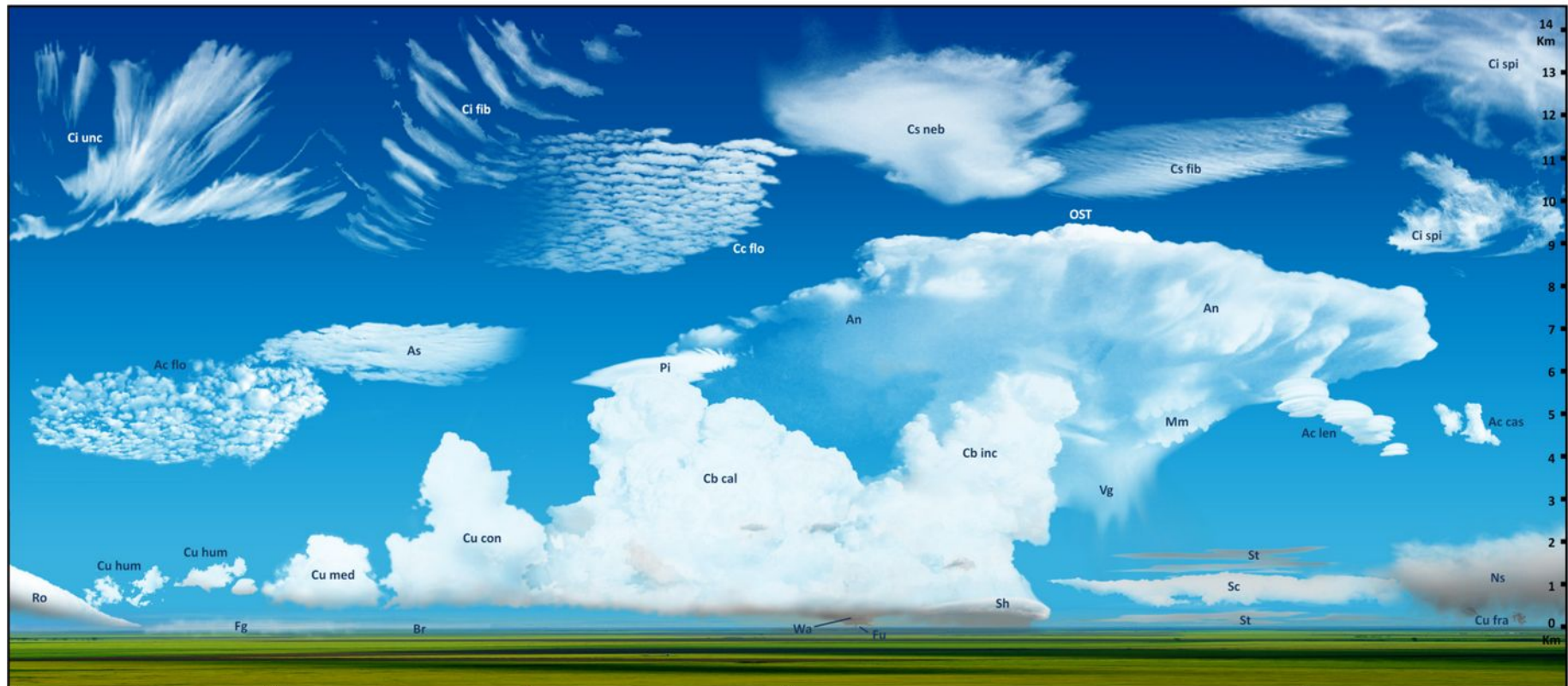
SUBTOPICS

3. Cloud formation (water and ice clouds)

- **Cloud types**
- Cloud properties and saturation vapor pressure
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- *Mixed phase clouds*
 - Bergeron-Findeisen Process

CLOUD TYPES

CLOUDS ATLAS



High Troposphere		Medium Troposphere		Low Troposphere		Towering vertical & Other-Accessories clouds		Surface based	
Ci spi	Cirrus spissatus	Ac flo	Alto cumulus floccus	Cu fra	Cumulus fractus	Cu con	Cumulus congestus	An	Anvil
Ci fib	Cirrus fibratus	Ac cas	Alto cumulus castellanus	Cu hum	Cumulus humilis	Cb cal	Cumulonimbus calvus	Vg	Virga
Ci unc	Cirrus uncinus	Ac len	Alto cumulus lenticularis	Cu med	Cumulus mediocris	Cb inc	Cumulonimbus incus	Ro	Roll cloud
Cc flo	Cirrocumulus floccus	As	Altostratus	Sc	Stratocumulus	OST	Overshooting Top	Sh	Shelf cloud
Cs fib	Cirrostratus fibratus			St	Stratus	Mm	Mammatus	Wa	Wall cloud
Cs neb	Cirrostratus nebulosus			Ns	Nimbostratus	Pi	Pileus	Fu	Funnel cloud

Realized by Antonio Ciccolella
05/2015

Clouds Atlas von Antonio Ciccolella, [CC BY-SA 4.0](#)

CLOUD TYPES

1. Warm clouds ($T > 0^{\circ}\text{C}$)

- only water droplets
- drop and rain formation

2. Cold clouds ($T < 0^{\circ}\text{C}$)

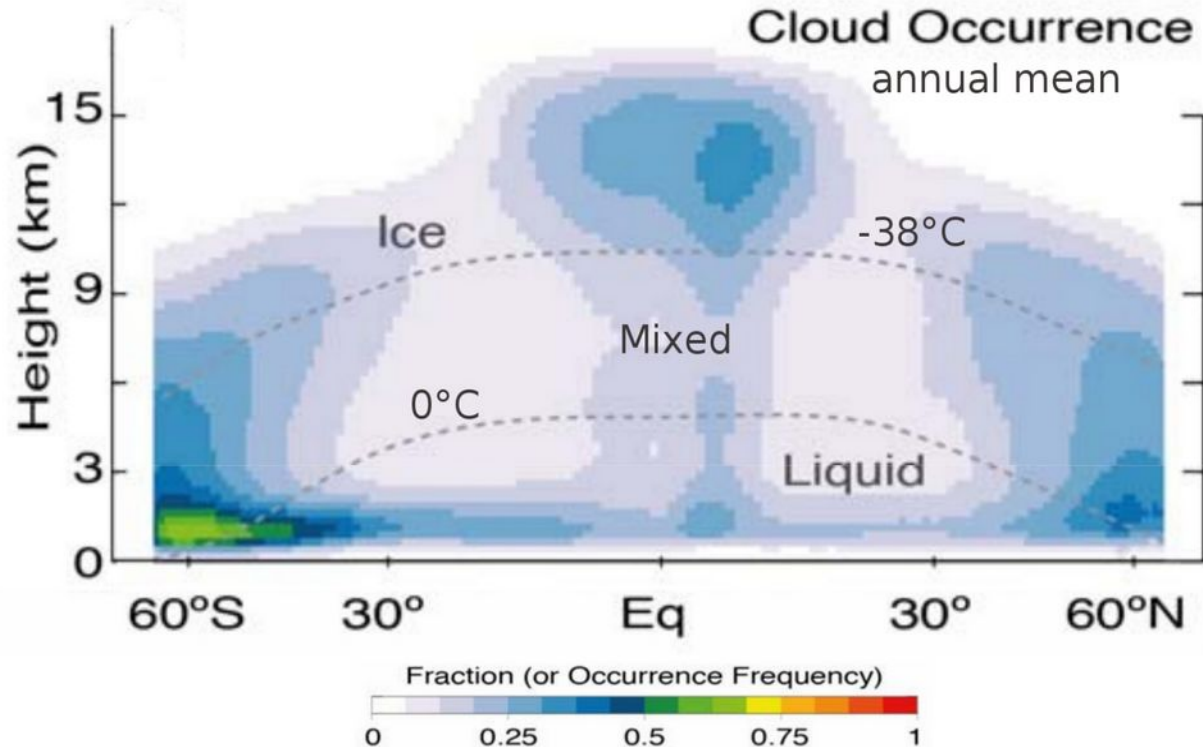
- Ice formation

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)

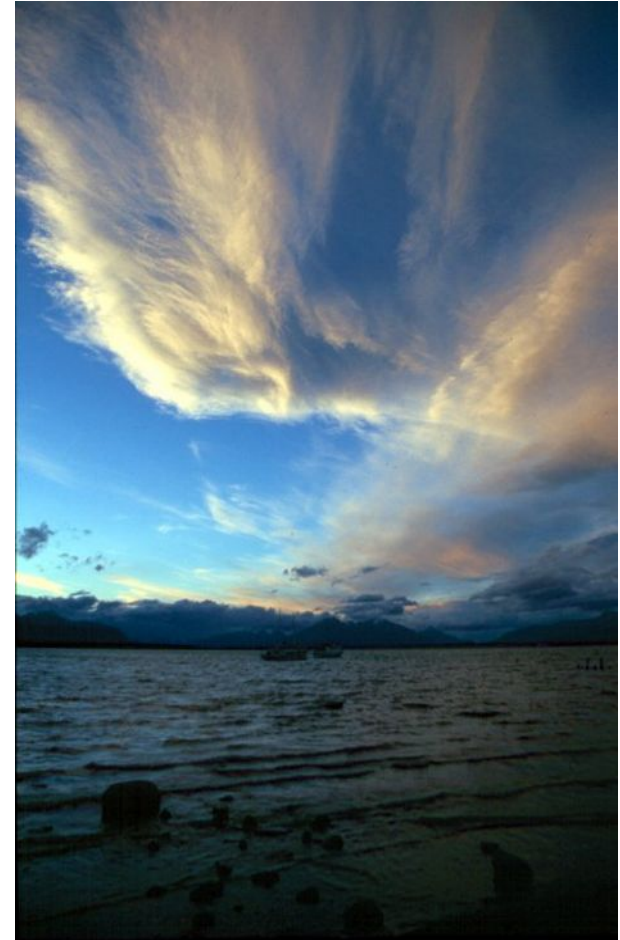


Pictures are taken from
Karlsruher Wolkenatlas
<http://www.wolkenatlas.de>

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>

JOHANNES GUTENBERG
UNIVERSITÄT MAINZ



JÜLICH
Forschungszentrum

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CLOUD PROPERTIES

- Particle size distribution $N(D_p)$

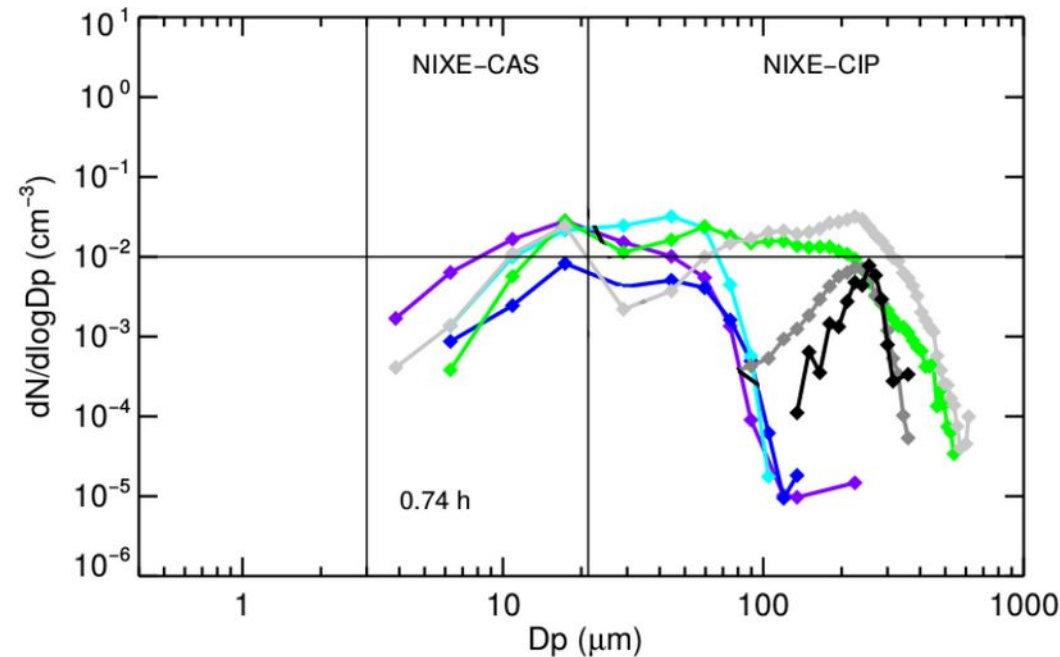
Number concentration N
Size distribution D_p

- Liquid/ Ice water content

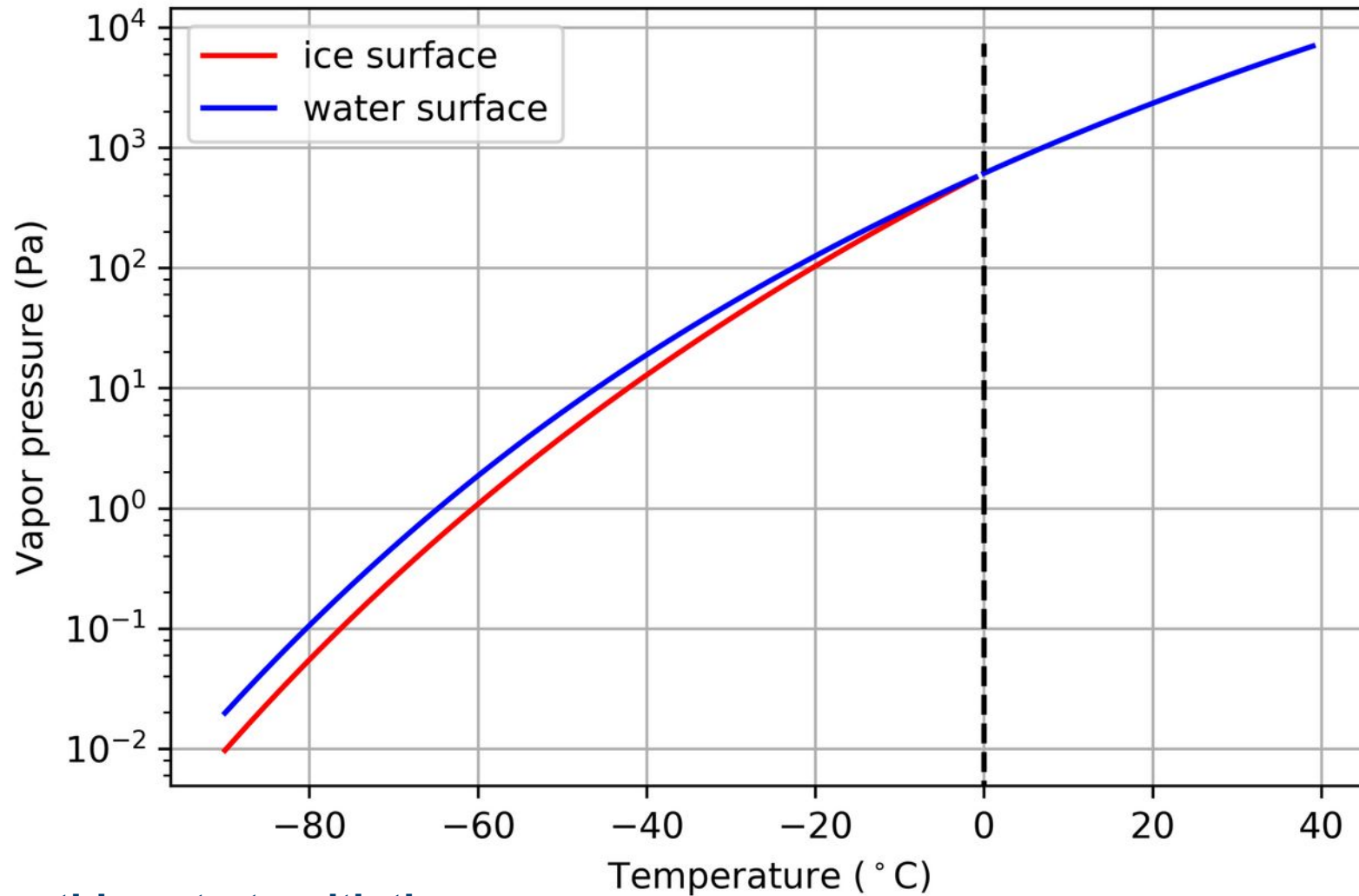
$$LWC / IWC = \rho \int_{D_{p_{min}}}^{D_{p_{max}}} N(D_p) dD_p$$

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

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

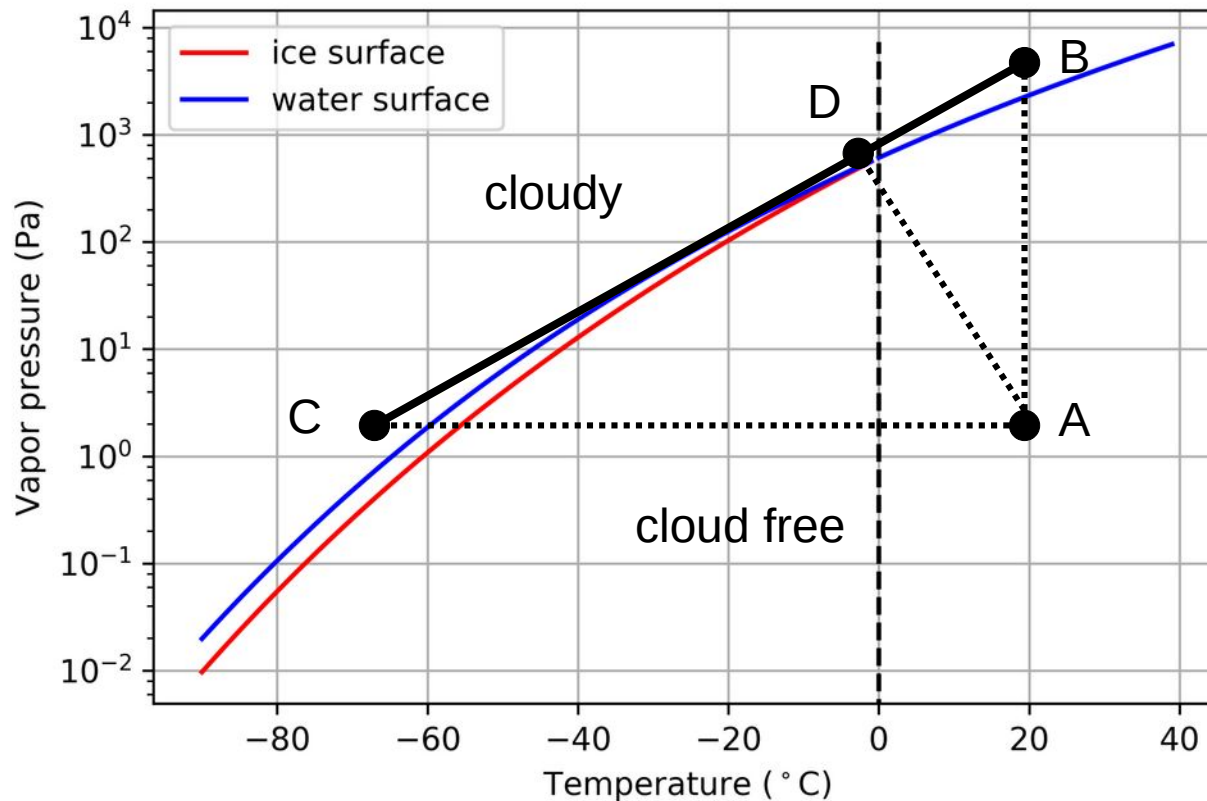


Saturation vapor pressure



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

How to saturate the air ?

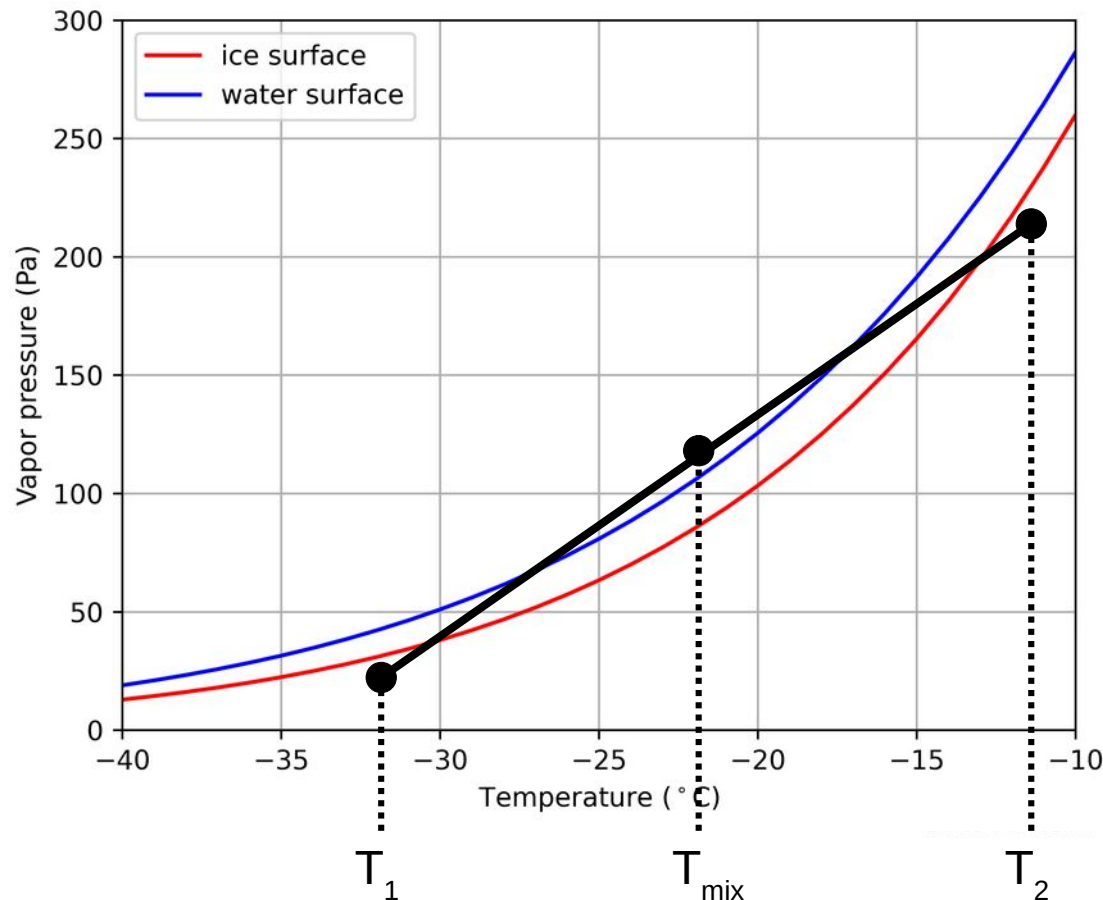


- e_s : Sub-saturation
- e_s : Saturation
- e_s : Supersaturation
→ Condensation

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.

SUBTOPICS

3. Cloud formation (water and ice clouds)

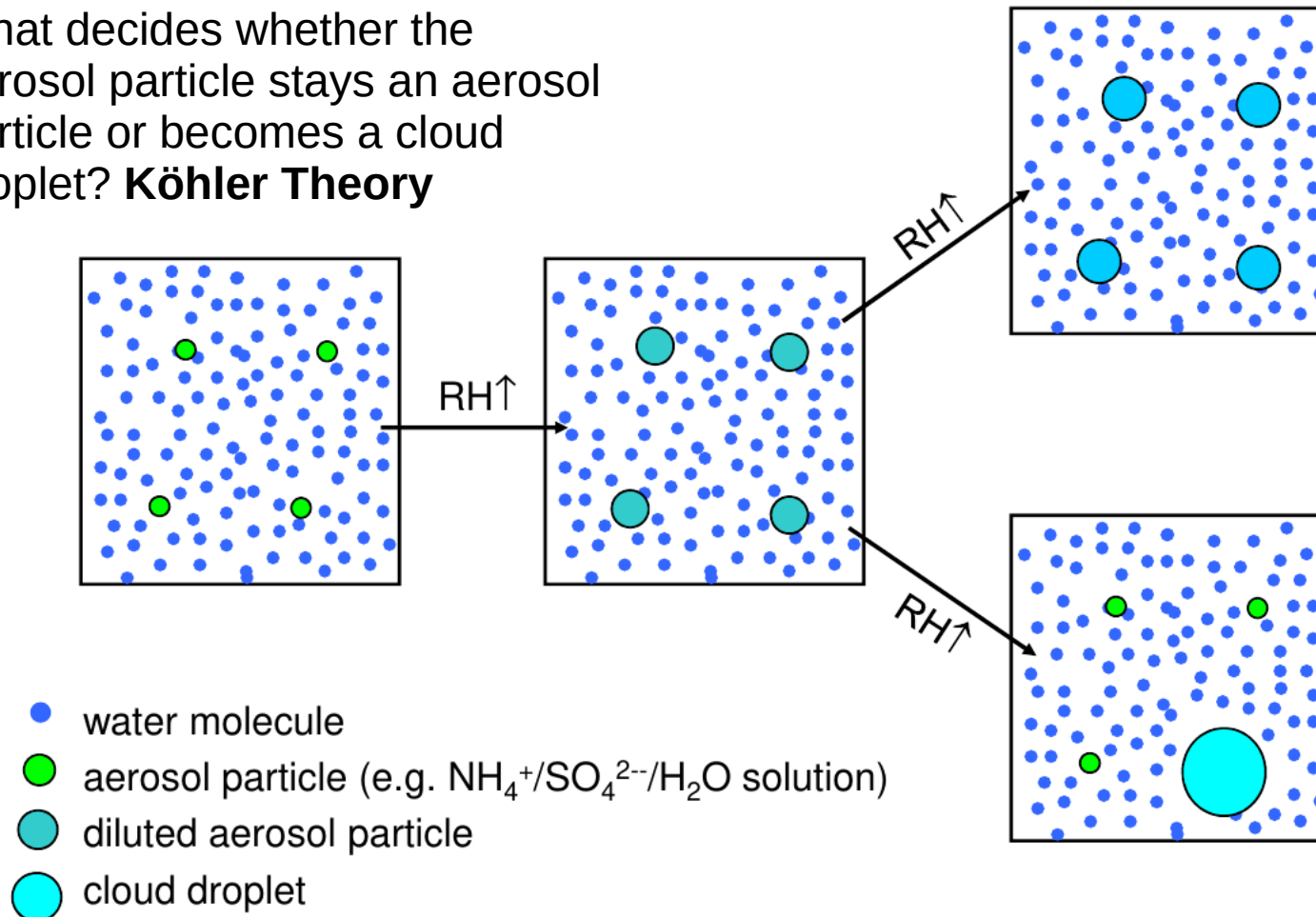
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WARM CLOUDS IN THE ATMOSPHERE

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

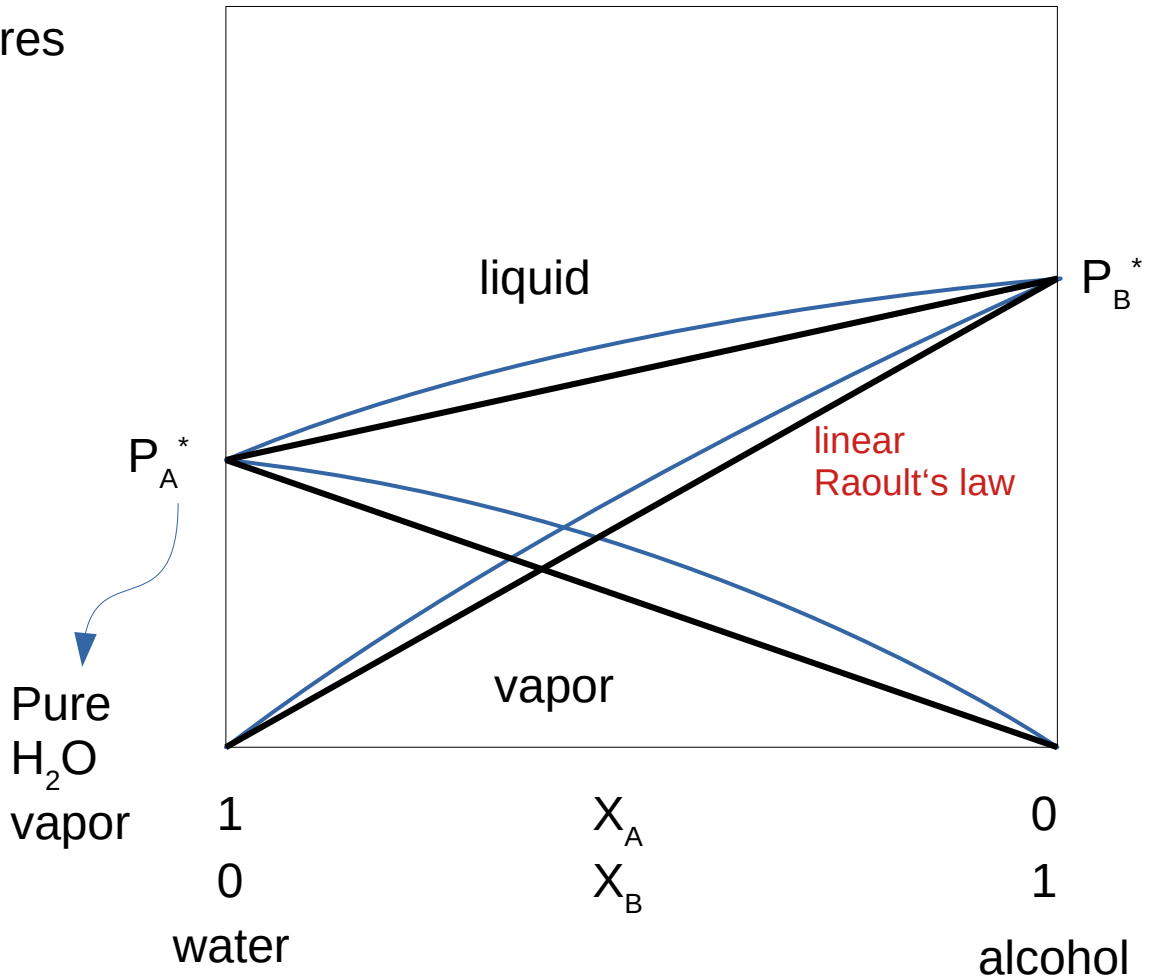
$X_{A,B}$ = mole fraction

$n_{A,B}$ = mole number

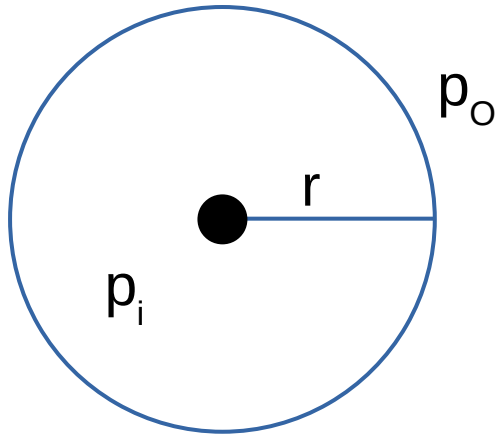
$$X_{A,B} = \frac{n_{A,B}}{\sum n_i}$$

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:

$$dW = \sigma dA$$

σ = surface tension

$A = 4\pi r^2$ = surface area

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

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

$$p_i = p_o + \underbrace{2 \frac{\sigma}{r}}_{\text{Laplace equation (valid for bubble or droplet)}}$$

Important for $r < 50 \mu\text{m}$ (5-10% effect)

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

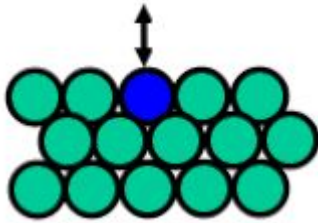
$$\ln \left(\frac{p(r)}{p(\infty)} \right) = \frac{V_{m,liquid} \Delta p}{RT}$$

$p(\infty)$ = vapor pressure over flat surface,
 r = particle radius, R = gas constant,
 $V_{m,liquid}$ = liquid molar volume, e.g. of H₂O,
 σ = surface tension

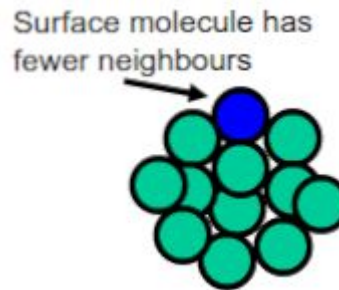
For curved surfaces: $\Delta p = 2\sigma / r$

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

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.:

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

$p_w(\infty)$ = H₂O vapor pressure over liquid water,

σ = surface tension of solution [kg / s²],

r = particle radius [m],

$V_{m,liquid}$ = liquid molar volume of H₂O [m³ / mol],

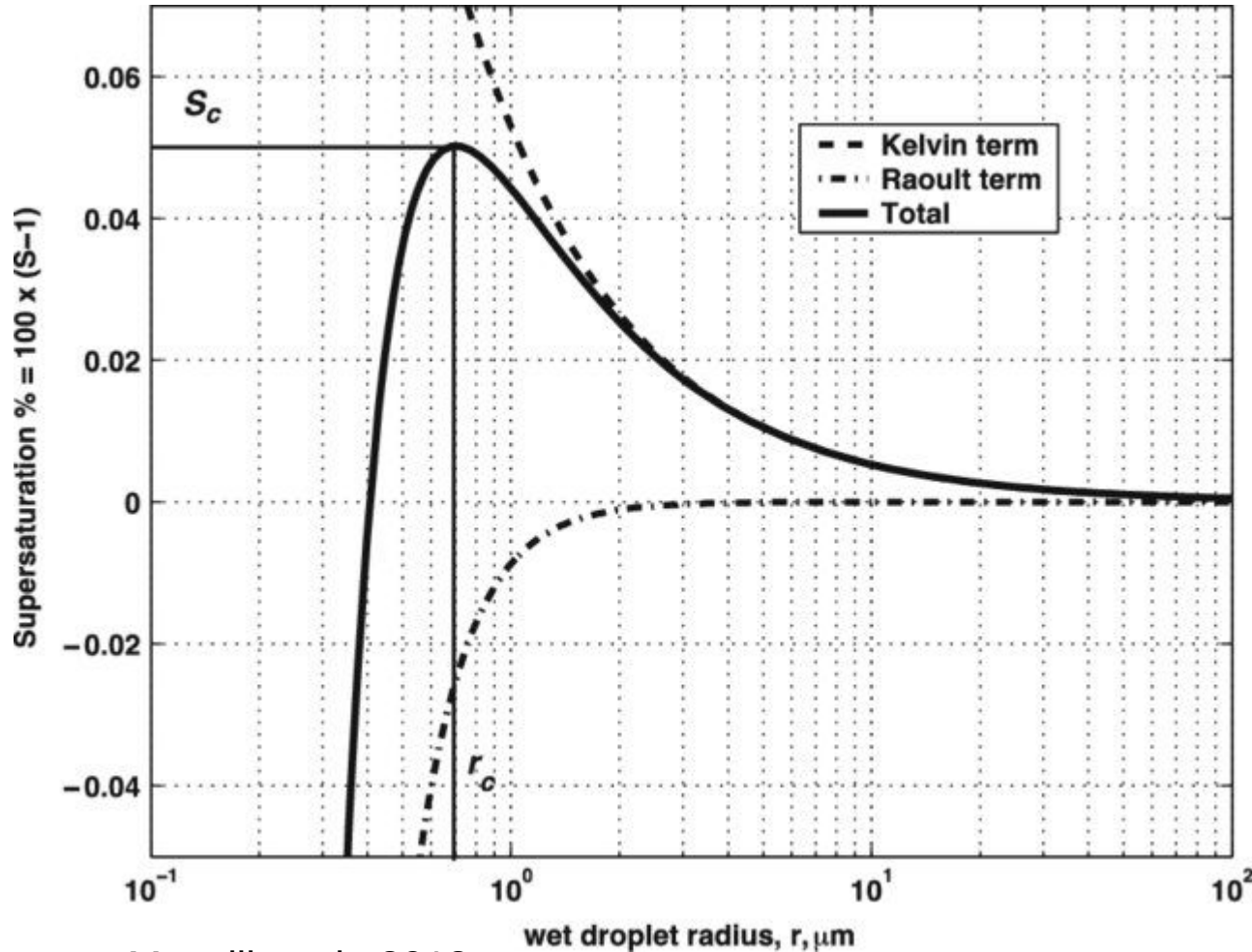
n_s = molar density of solute [1/mol]

R = Gas constant [kg m² / s² / K / mol]

COMBINATION OF RAOULT AND KELVIN (2)

KÖHLER THEORY

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



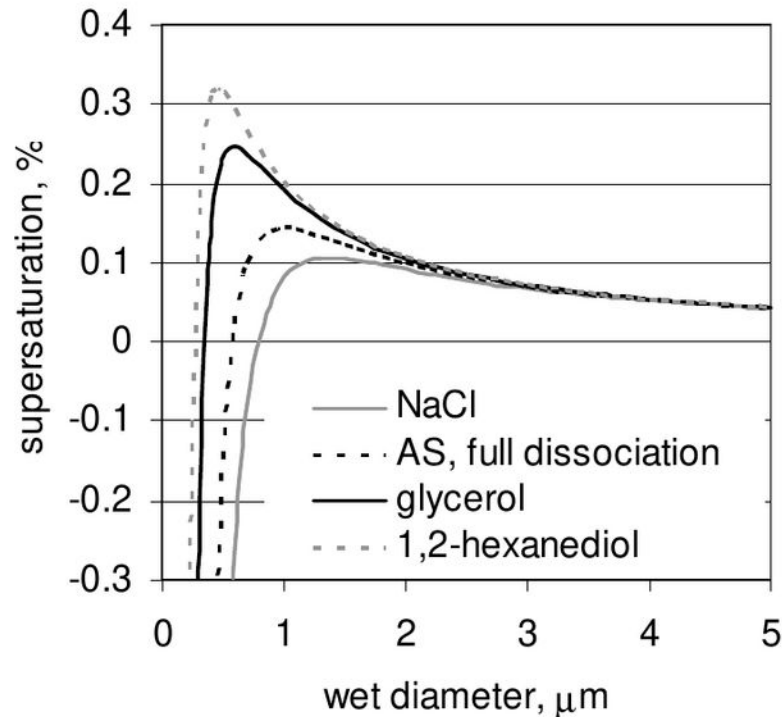
Köhler curve for a NaCl particle with a dry diameter of 50 nm.

r_c : critical radius for further growth

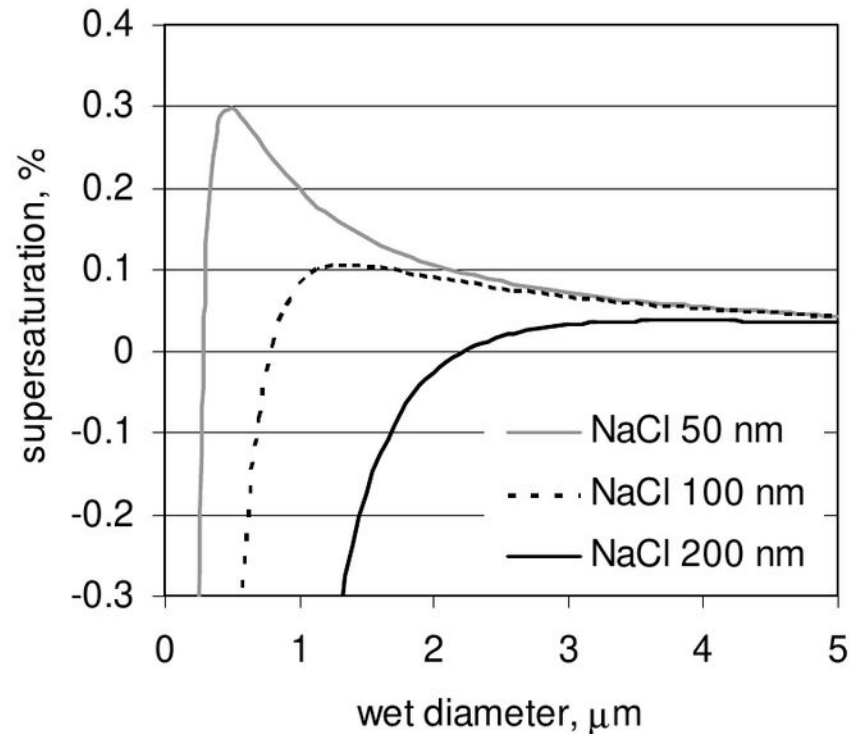
Mcneill et al., 2013

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

SUBTOPICS

3. Cloud formation (water and ice clouds)

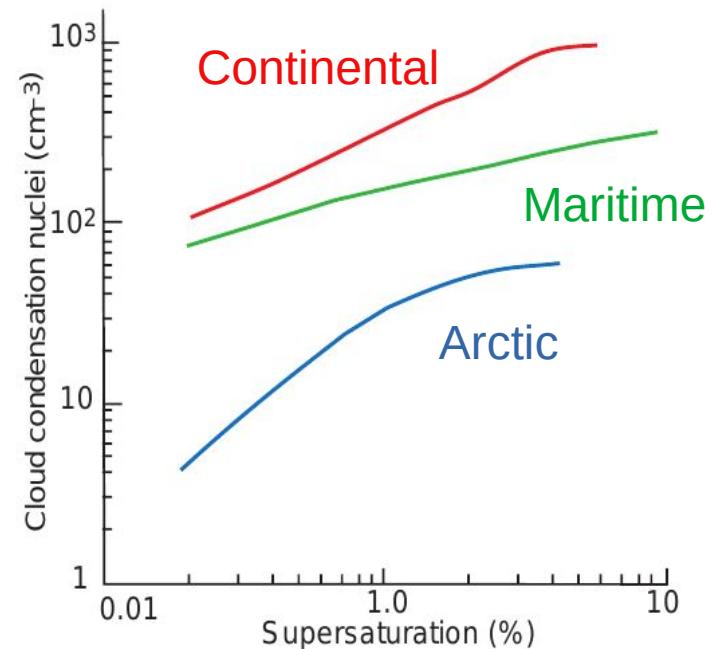
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CLOUD CONDENSATION NUCLEI (CCN)

Random condensation to achieve critical radius is unlikely. The presence of condensation nuclei (CCN) is necessary for condensation process. This lowers the surface tension ($\sigma_{\text{H}_2\text{O}} = 8 \cdot 10^{-2} \text{ N/m}$) and therefore the required saturation level.

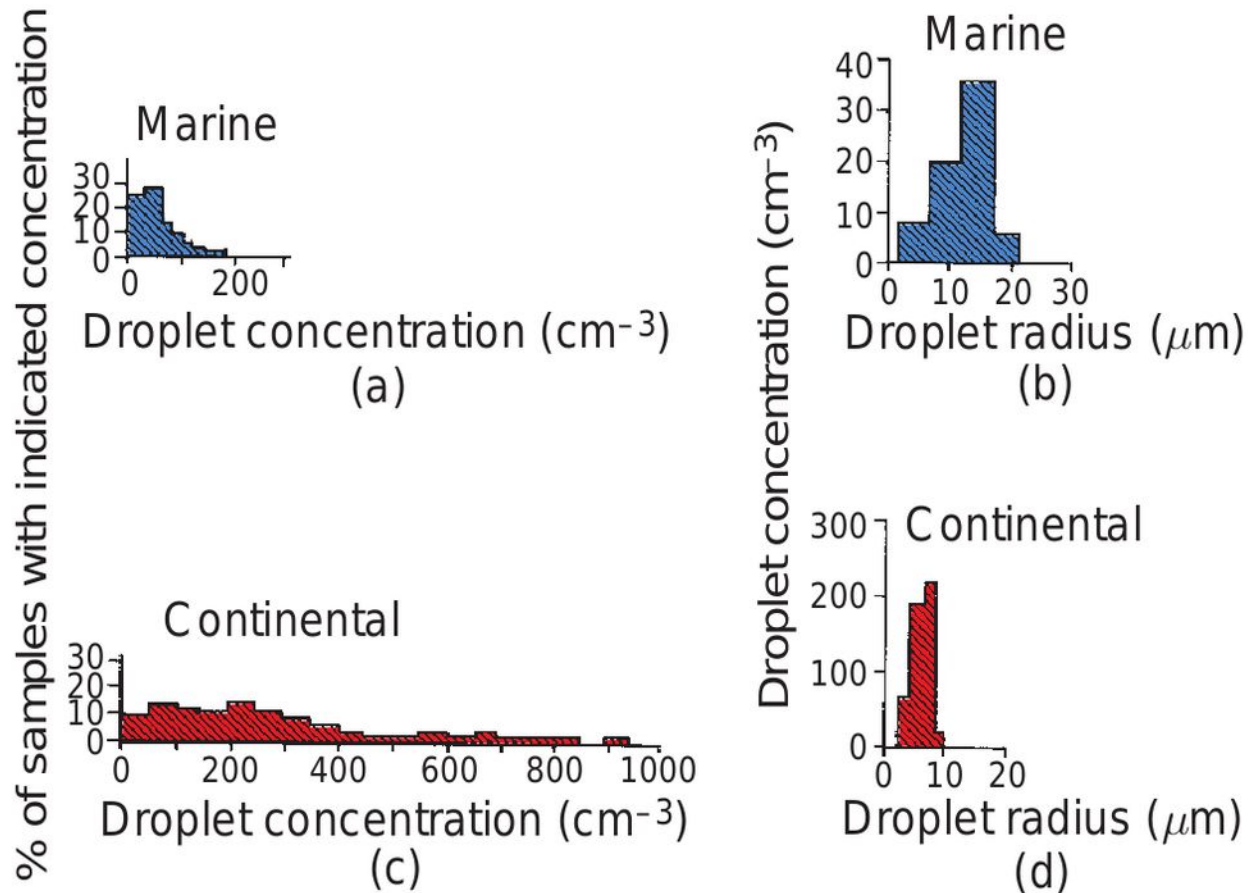
Super-saturation level in natural clouds $S \approx 0.1\%$!
That is mostly insufficient for cloud condensation!

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

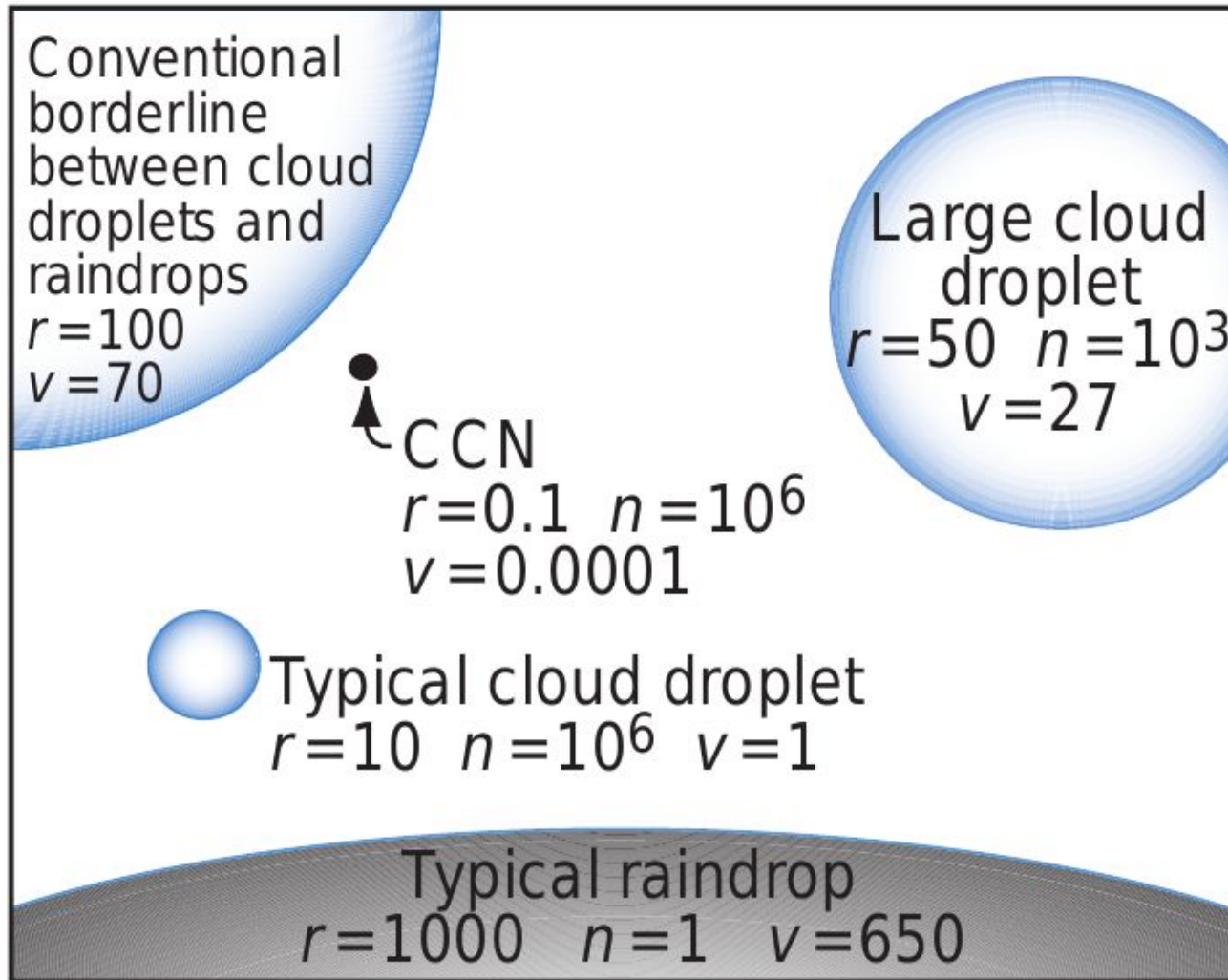
Ship tracks (white lines) in marine stratus clouds over the Atlantic Ocean as viewed from the NASA Aqua satellite on January 27, 2003.

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REL. SIZES OF CLOUD DROPLETS AND RAINDROPS



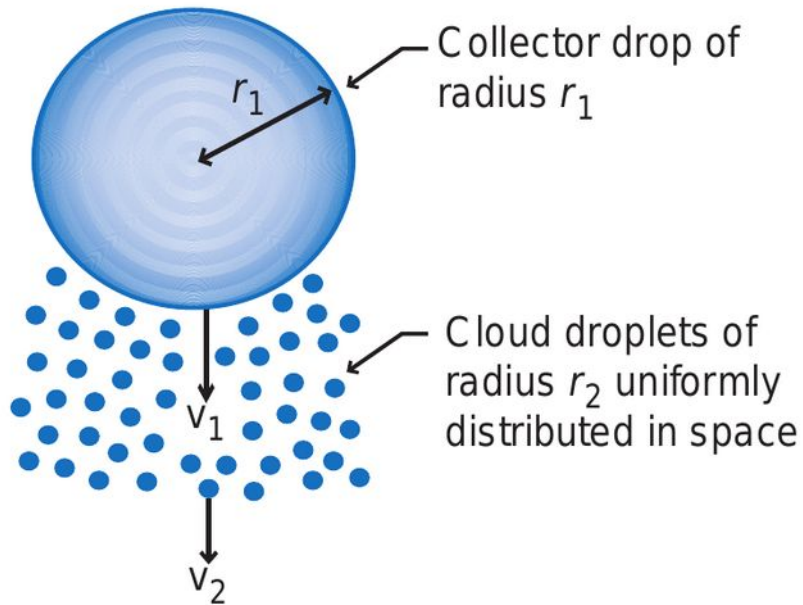
- r is the radius in micrometers
- n is the number per liter of air
- v is the terminal fall speed in cm/s

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

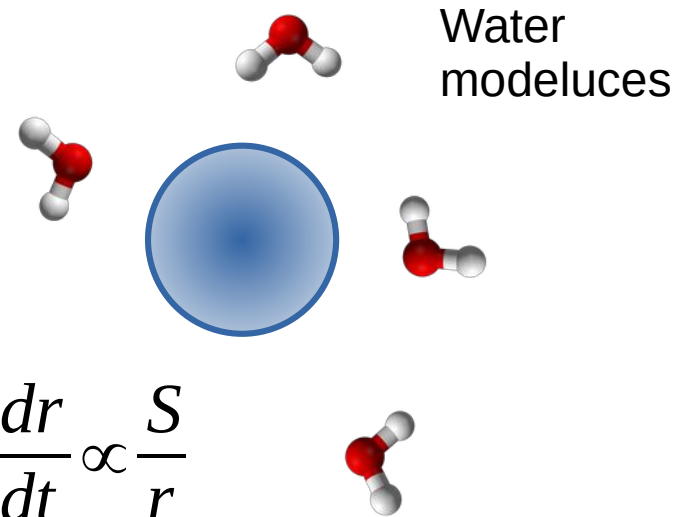
Collision of droplets



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

E: Collection efficiency $\propto r$

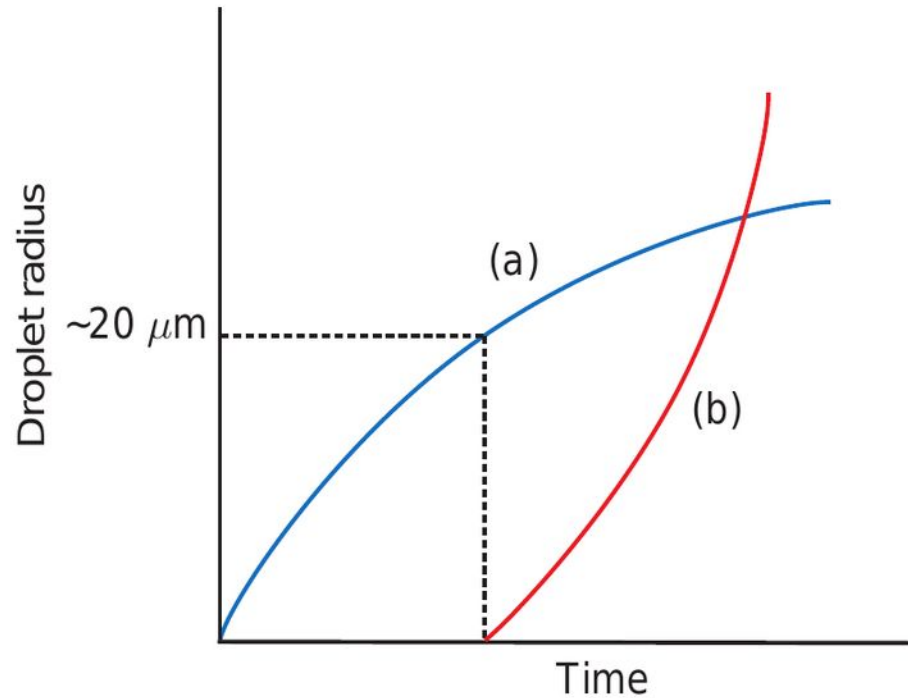
Diffusion of water vapor



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

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

GROWTH OF DROPLETS (COLLISION VS. DIFFUSION)

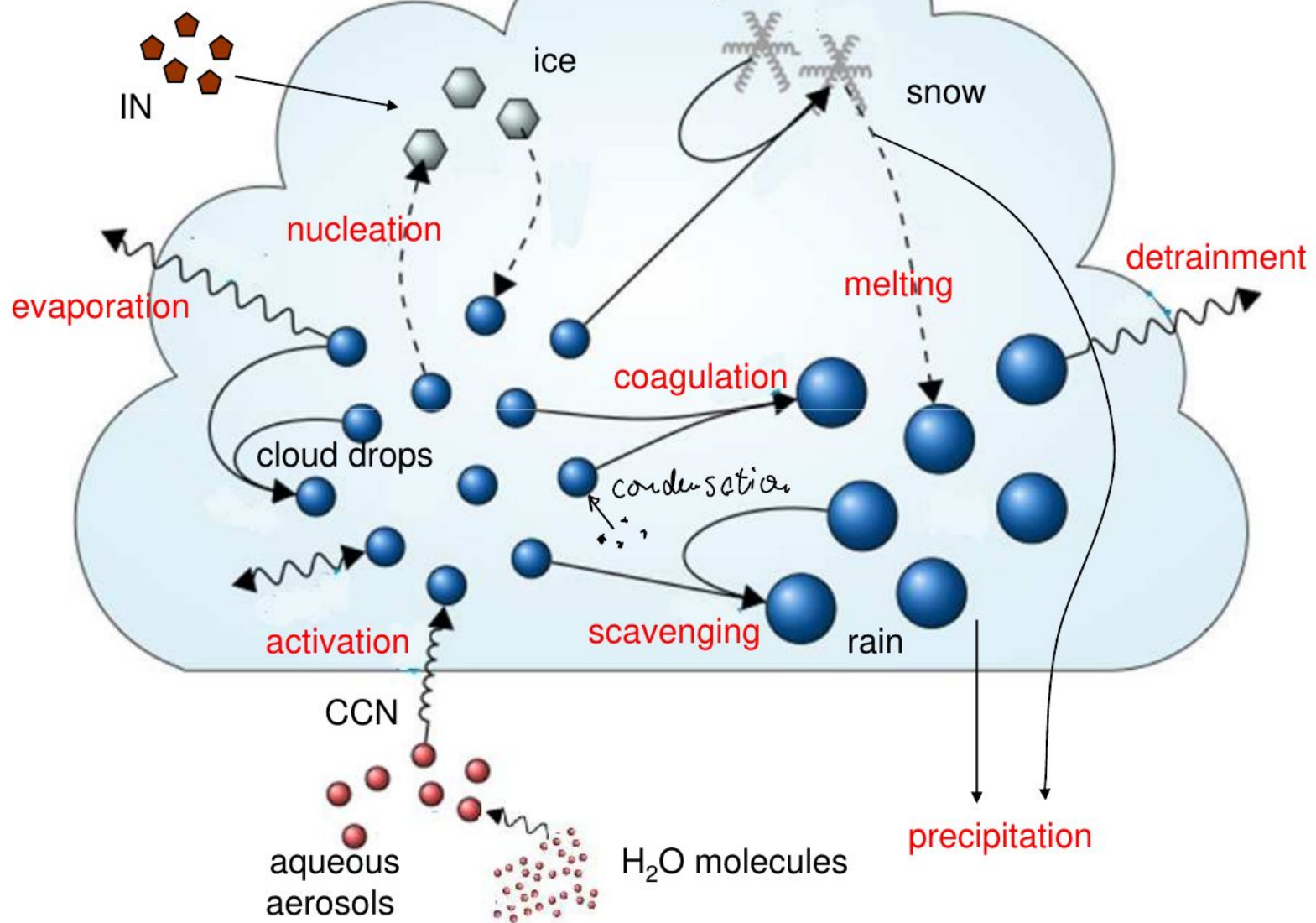


- Collision depends on weight & fall speed
- Diffusion depends on supersaturation surface
- Dommination process depends on the environmental conditions but also on the droplet size

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

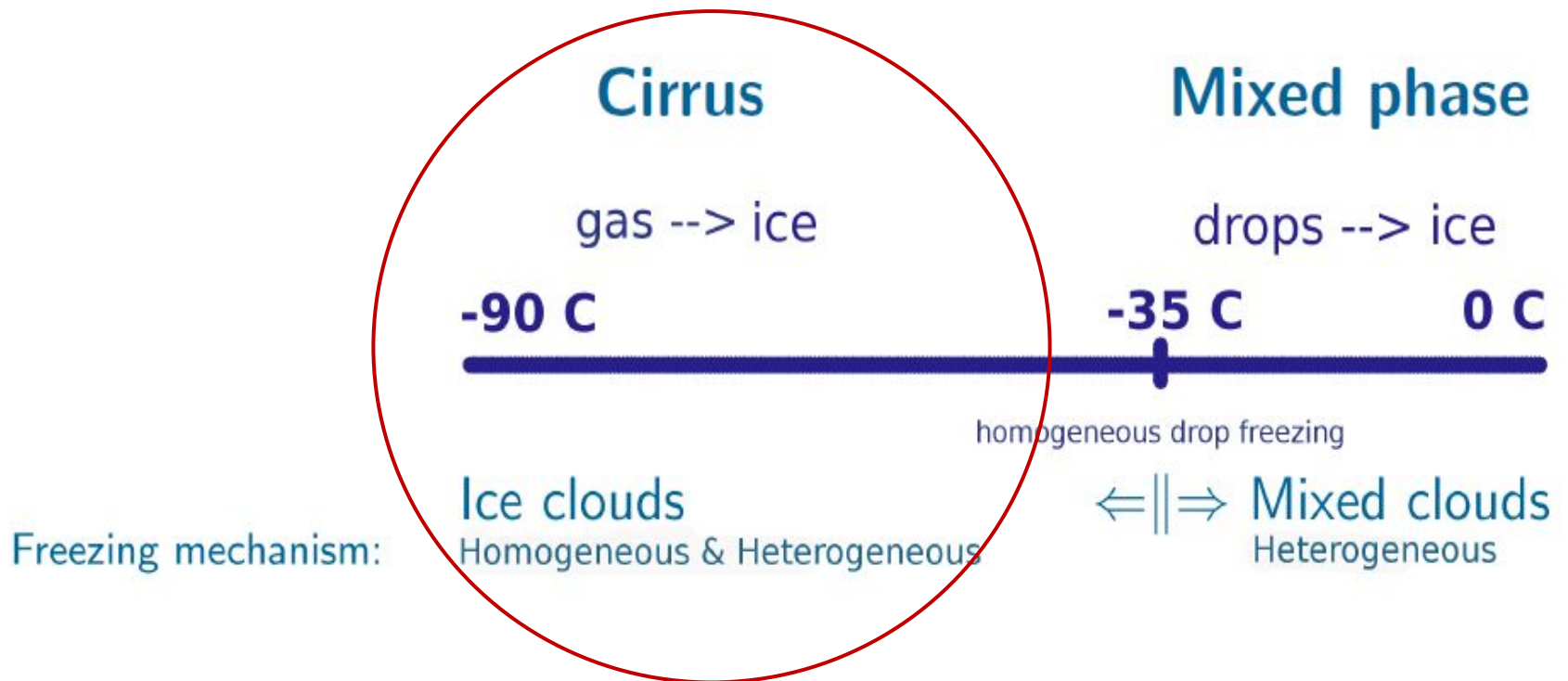


SUBTOPICS

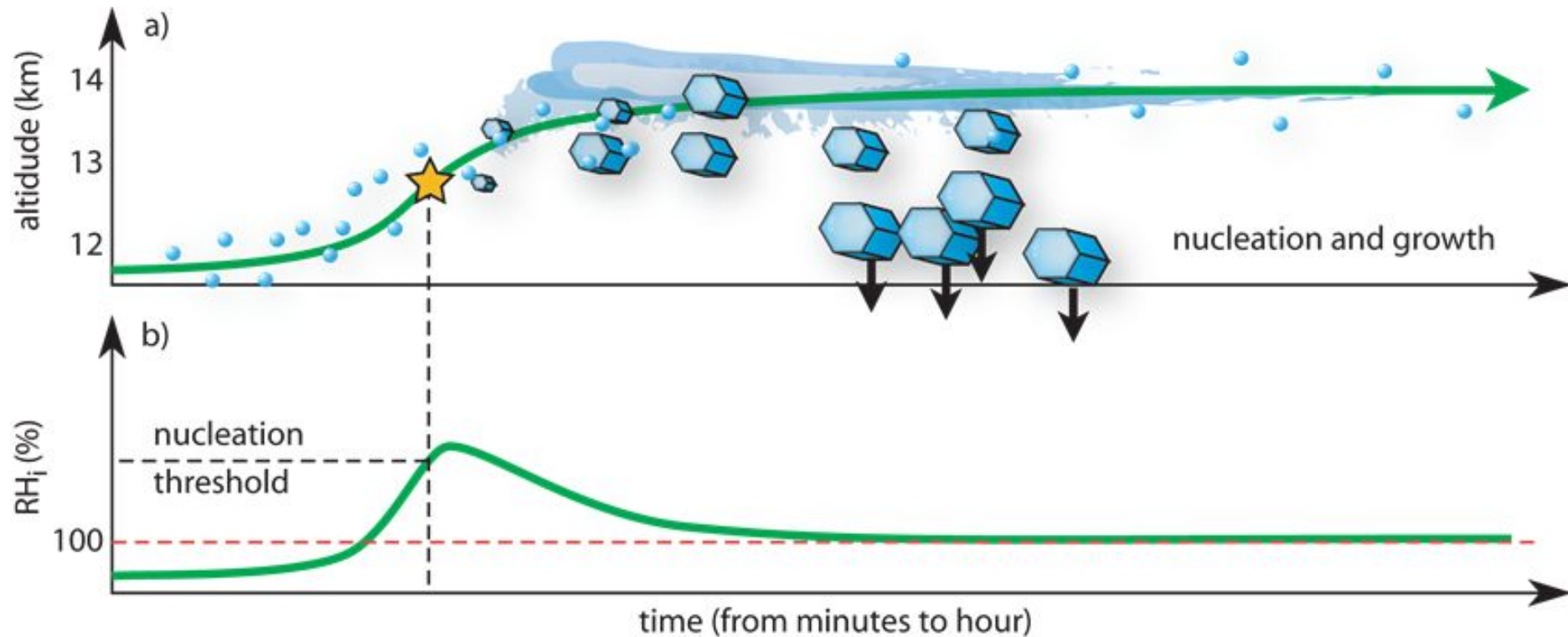
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COLD CLOUDS IN THE ATMOSPHERE



Ice cloud life cycle



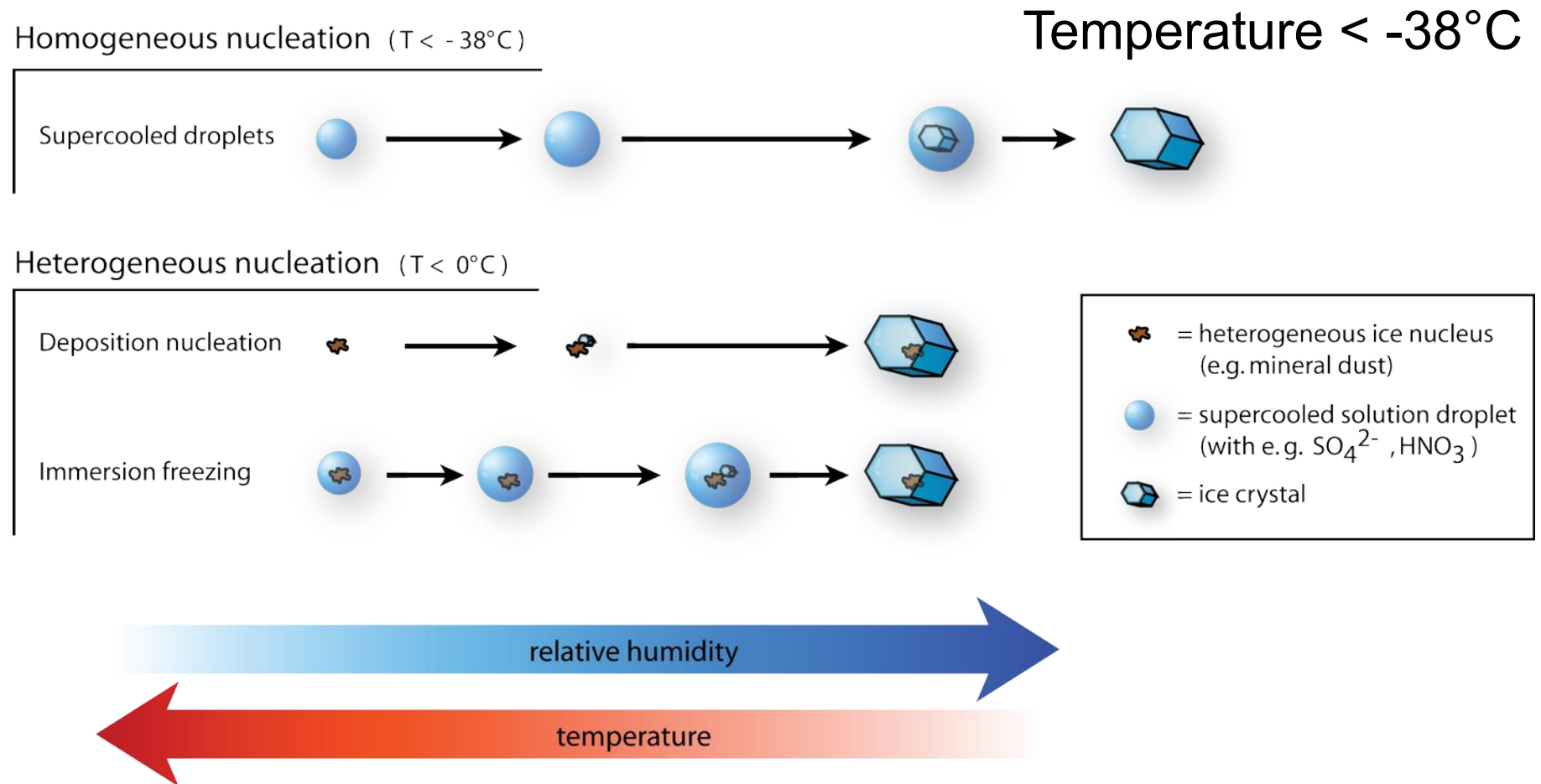
Adapted from Peter et al., 2005

SUBTOPICS

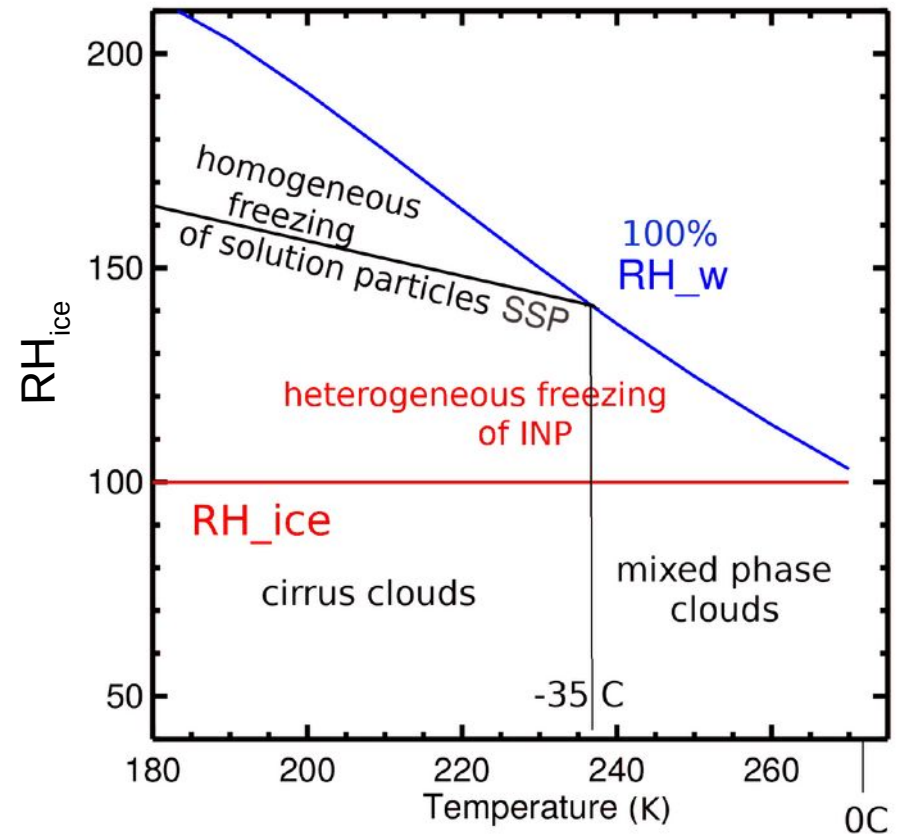
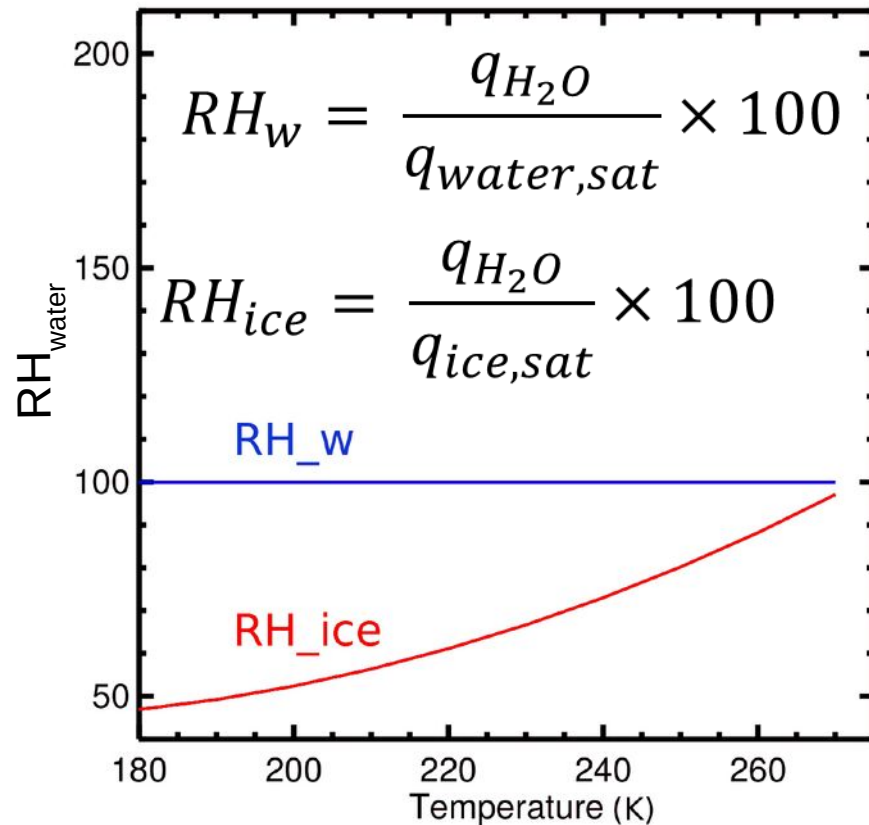
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FREEZING MECHANISM OF CIRRUS CLOUDS



Relative humidity at cold conditions



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

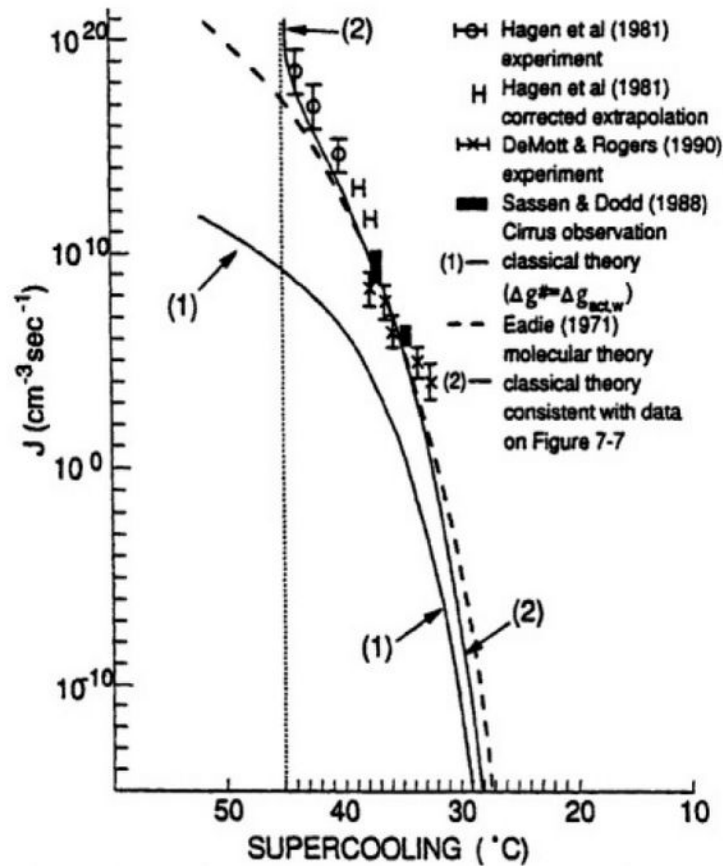
N_c : Number of water molecules

ρ : density of ice (i), liquid (l)

k : Boltzmann constant

T : Temperature

σ : Surface tension



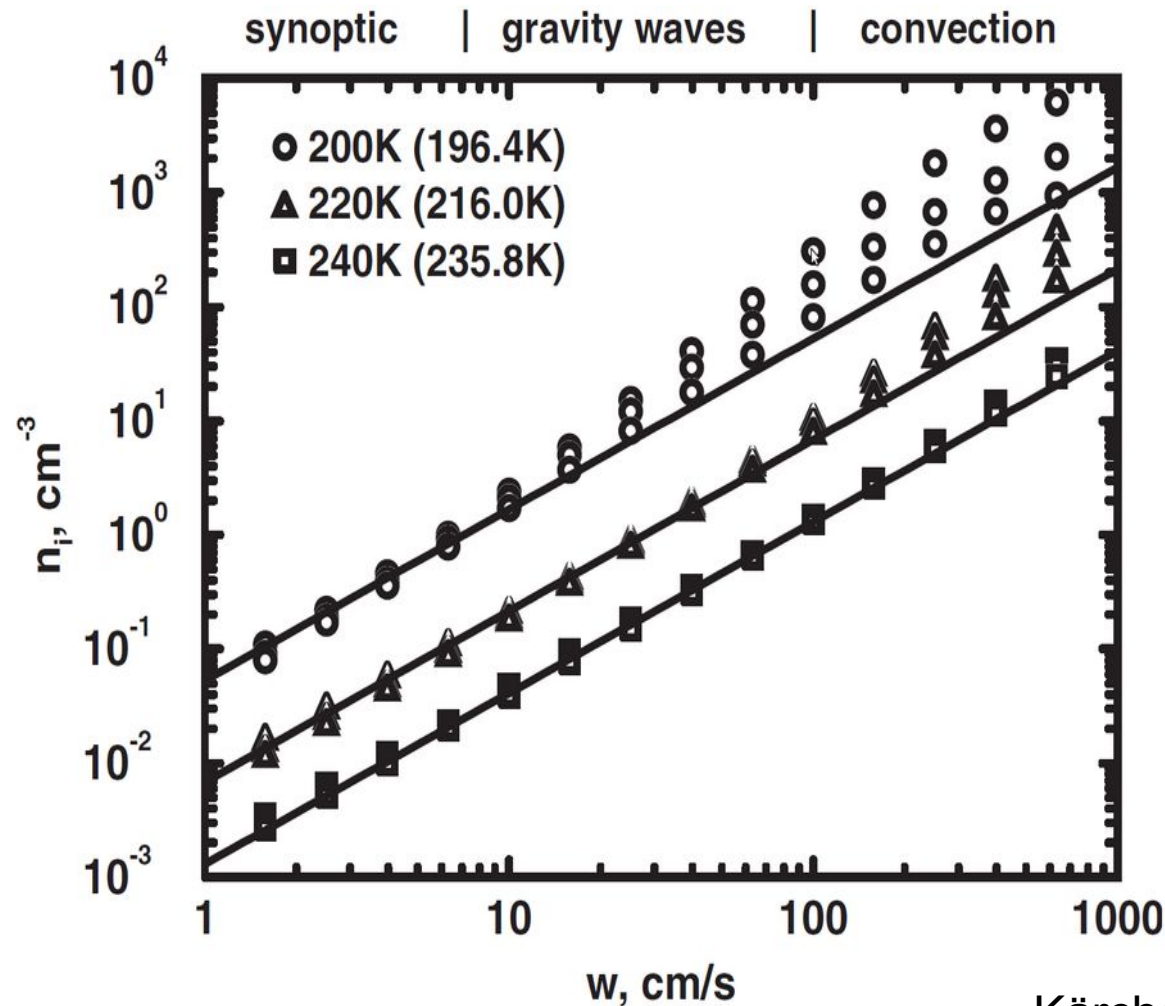
$$J = 2N_c \left(\frac{\rho_l kT}{\rho_i h} \right) \left(\frac{\sigma_{i/l}}{kT} \right)^{1/2} \exp \left(- \frac{\Delta F_{act} + \Delta F_g}{kT} \right)$$

Activation
energy

Germ formation energy
(work against surface
tension)

Pruppacher & Klett 1997

HOMOGENEOUS ICE NUCLEATION RATE



Kärcher and Lohmann (2002)

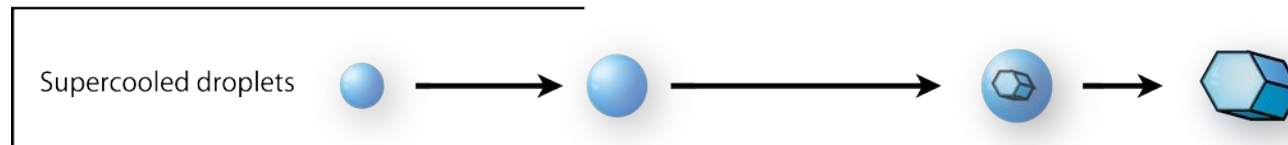
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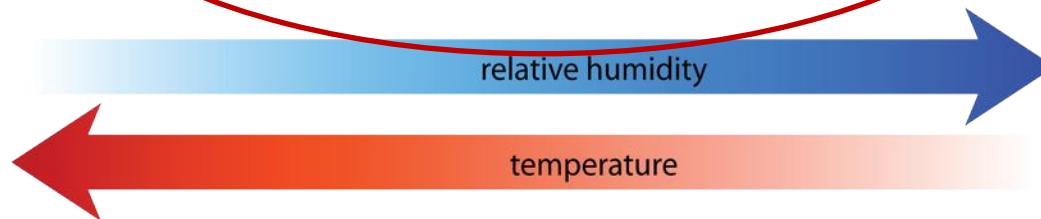
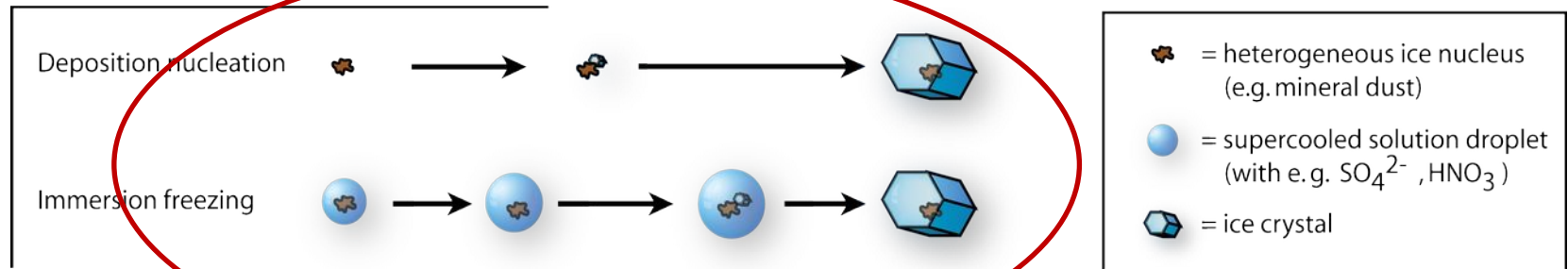
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HETEROGENEOUS FREEZING

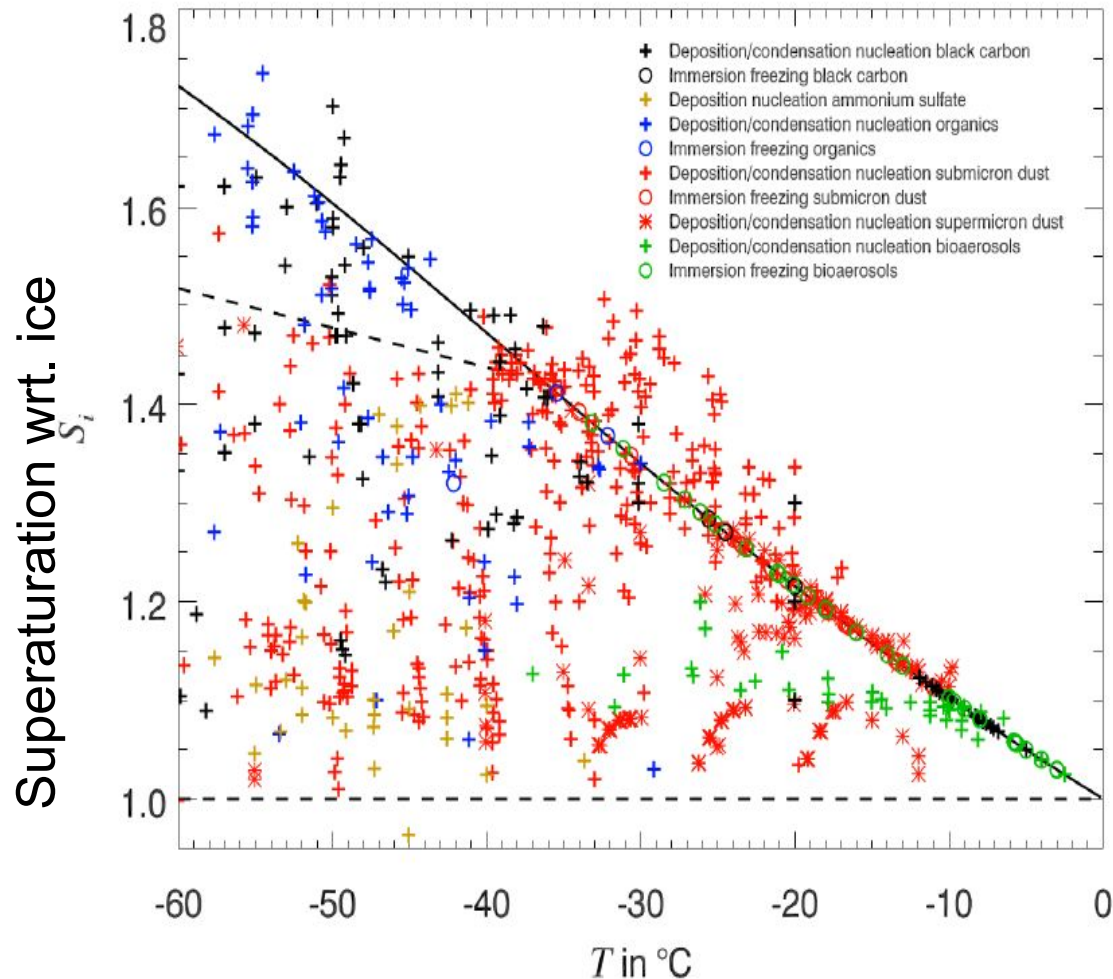
Homogeneous nucleation ($T < -38^{\circ}\text{C}$)



Heterogeneous nucleation ($T < 0^{\circ}\text{C}$)



HETEROGENEOUS FREEZING



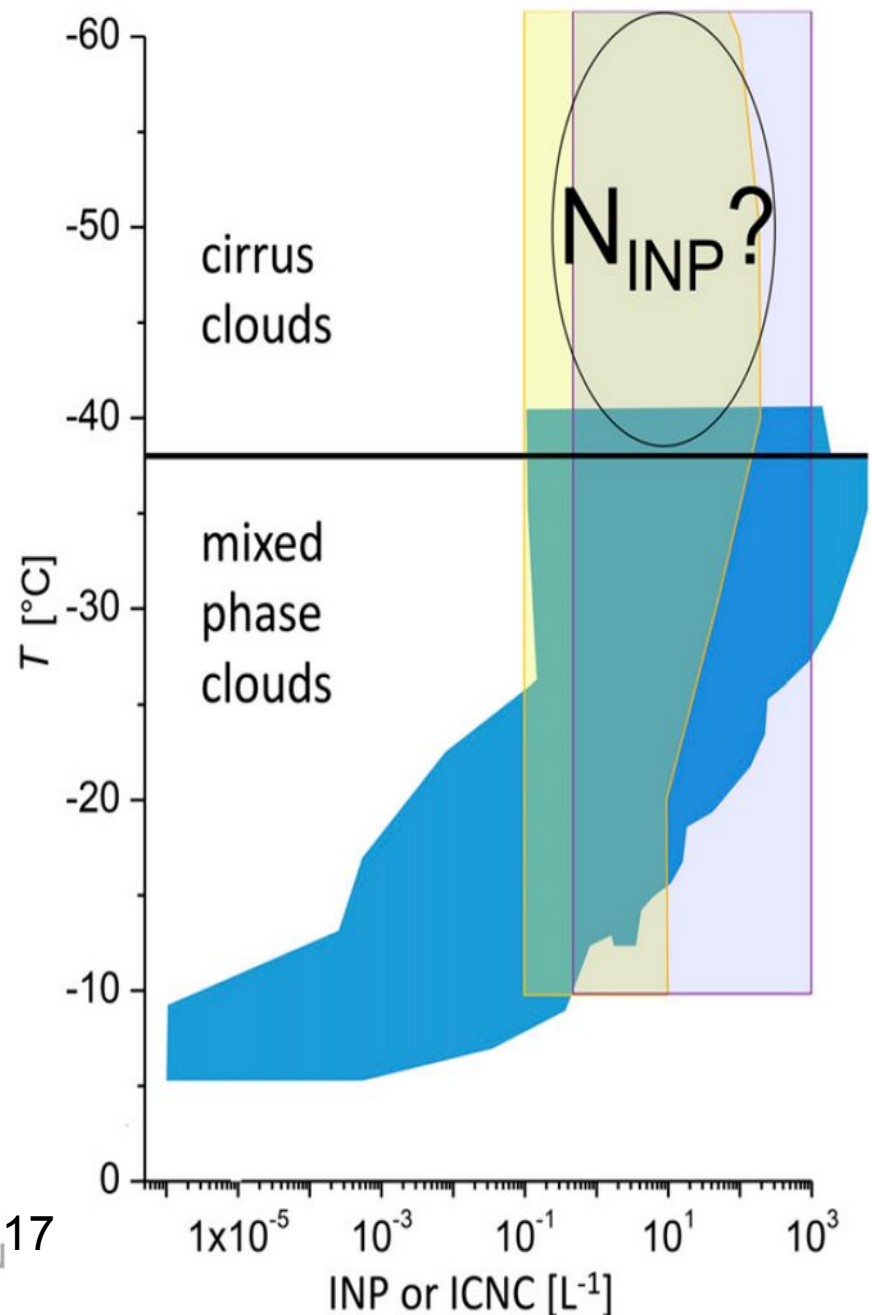
Heterogeneous freezing:

- 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)

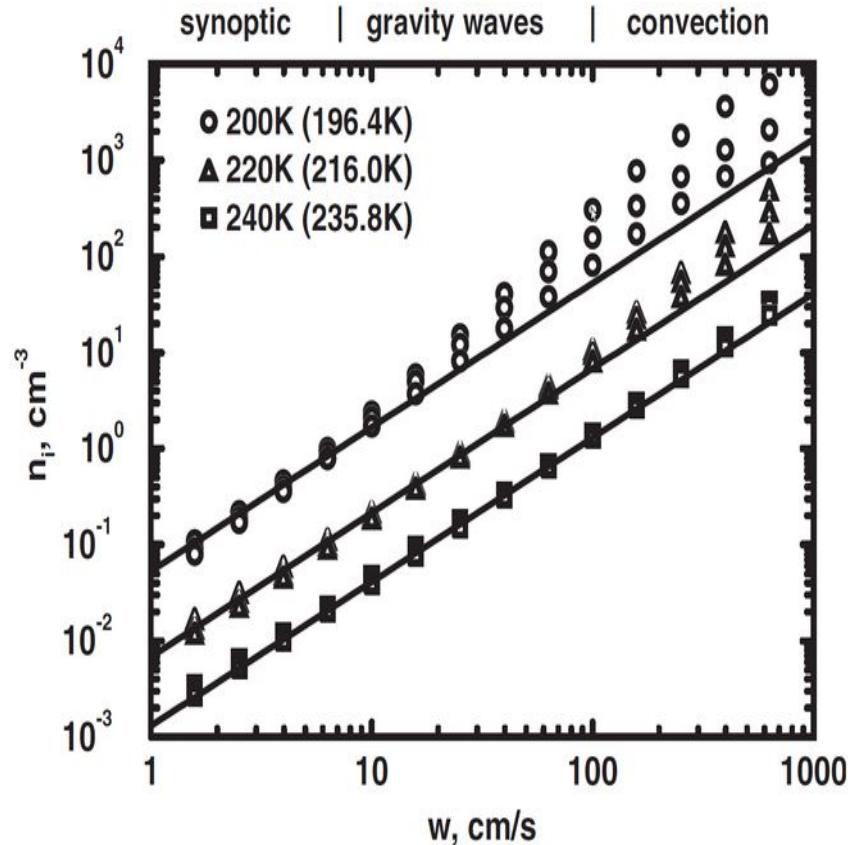
OBSERVATION OF INP

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



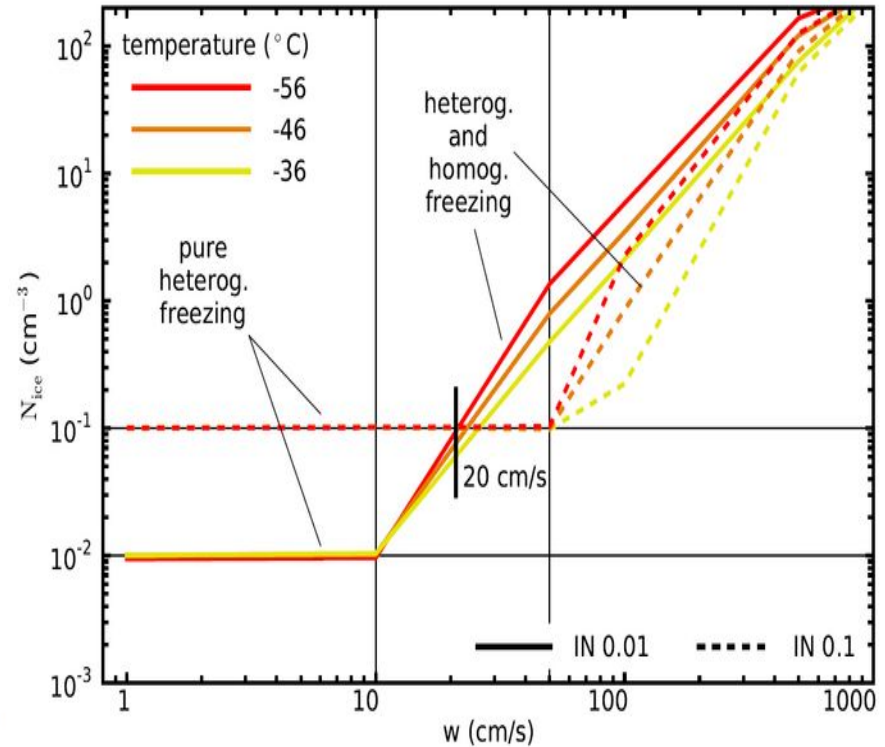
Kanji et al., AMS, 2017

IMPACT ON ICE CRYSTAL NUMBER



Homogeneous
freezing:

nearly no impact



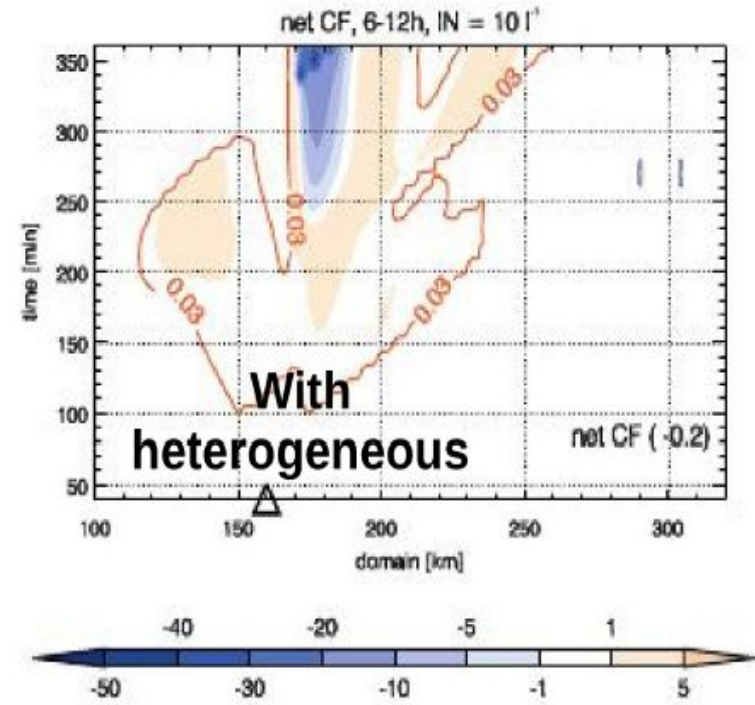
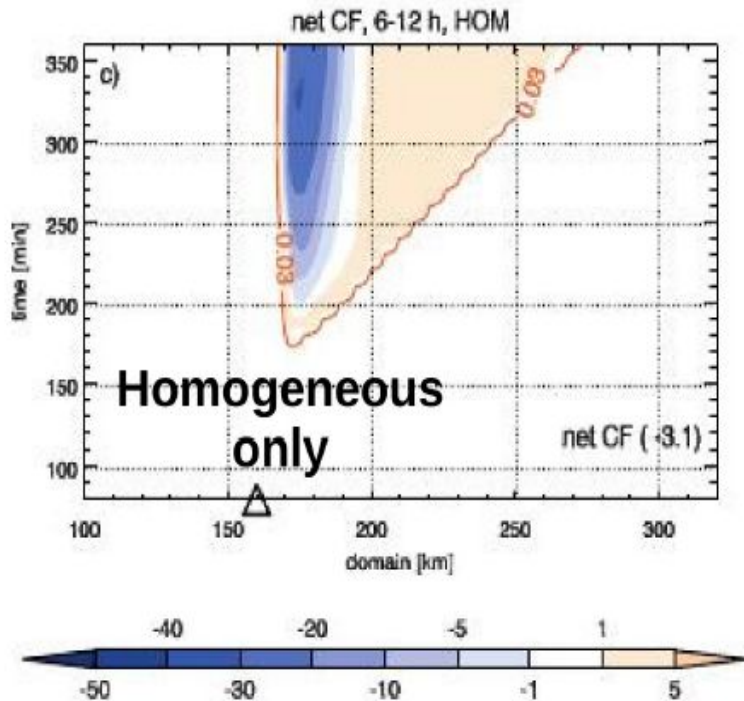
Heterogeneous
freezing:

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

of aerosol number and composition on ice crystal number

HOMOGENEOUS VS. HETEROGENEOUS

Effect of the freezing mechanism on net cirrus forcing



Joos et al. (2014)

- Heterogeneous nucleation change cloud coverage and microphysical properties
- Influence on radiative forcing and thus climate

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ICE CRYSTAL GROWTH

Classical diffusion of water molecules

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

R^* : Universal gas constant

T : temperature

D : diffusion coeff.

β : Mass deposition coeff.

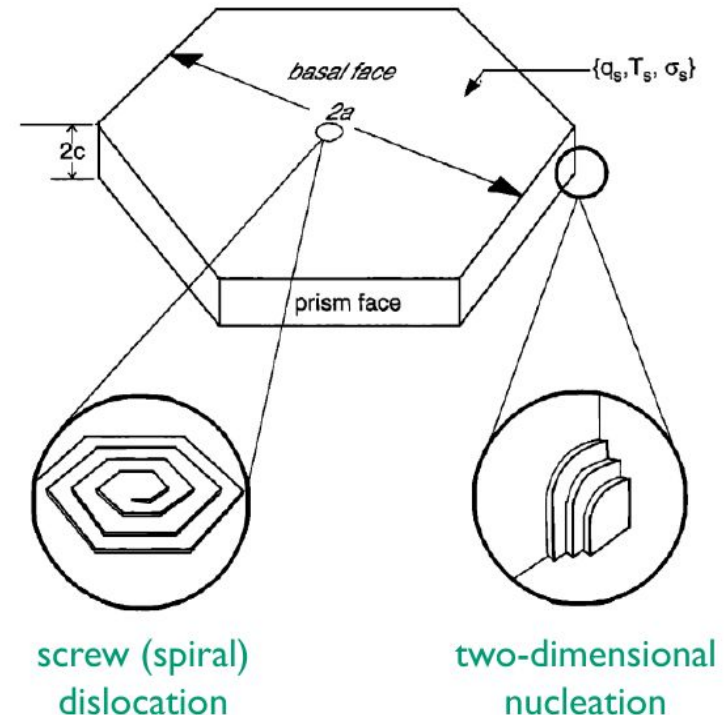
κ : capacity factor (shape dependent)

Φ : ventilation factor

M_w : Molar weight of water

p_w : partial pressure of water vapor

p_i : saturation partial pressure wrt. Ice

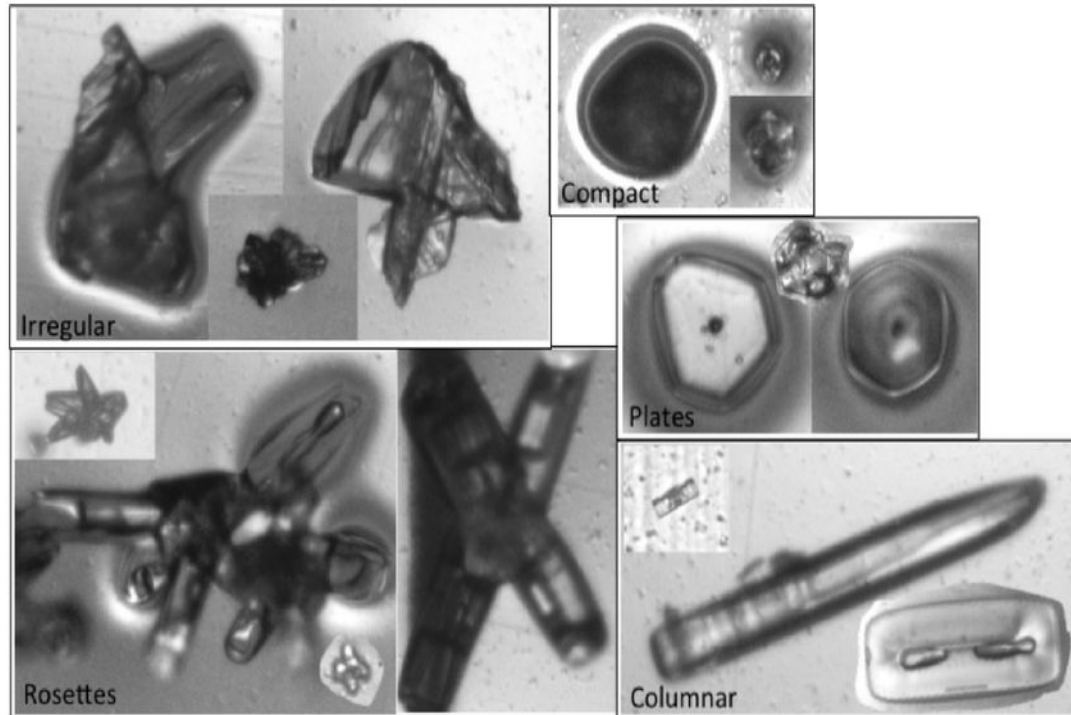


Manly depend on partial pressure difference

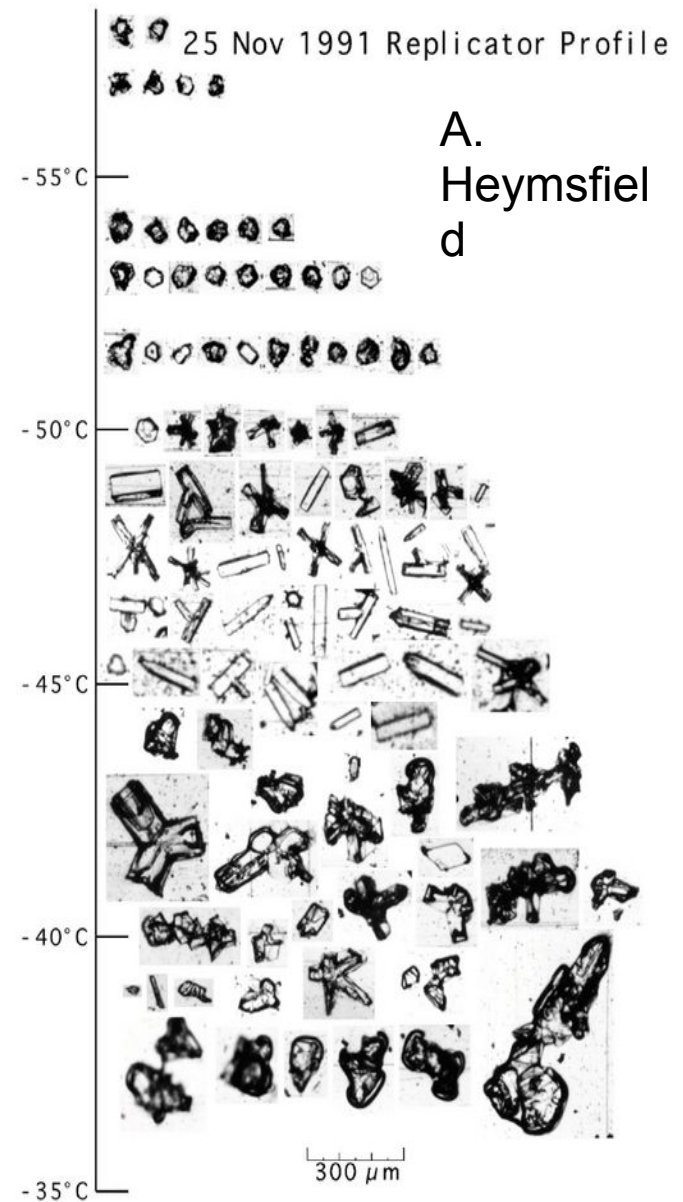
➔ Sub-saturated ➔ negative ➔ decreasing mass

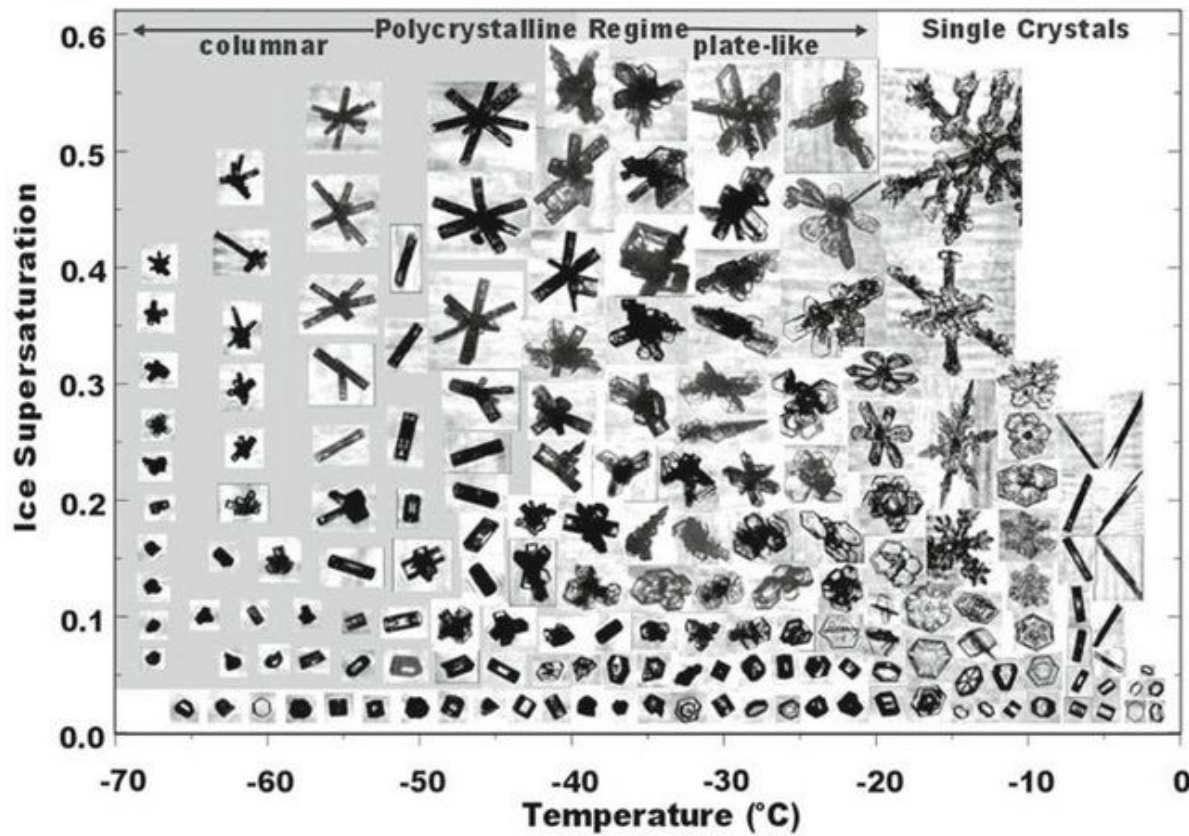
➔ Supersaturated ➔ positive ➔ increasing mass

ICE PARTICLE SHAPES



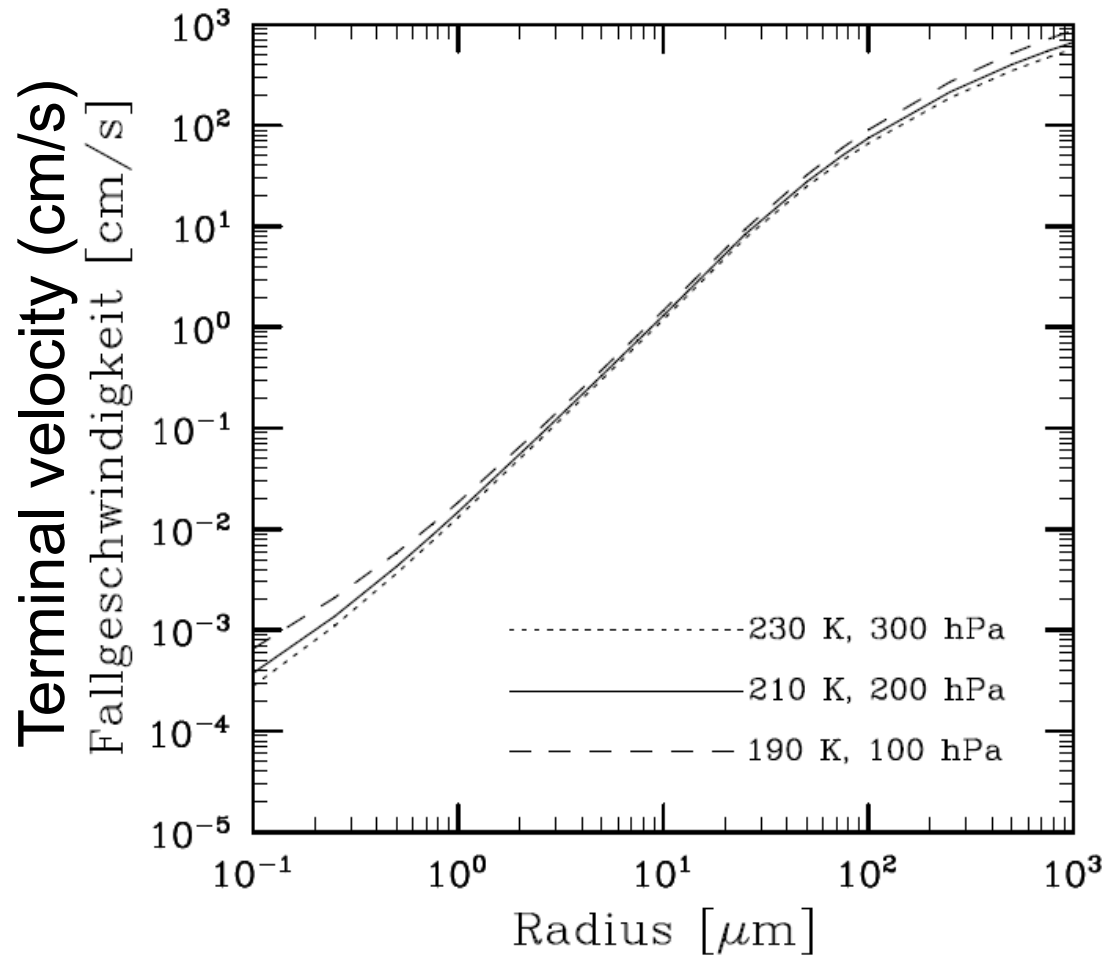
Wolf et al., ACPD
2018





Diedenhoven 2018
Remote Sensing of
Crystal Shapes in Ice
Clouds

SEDIMENTATION OF ICE CRYSTALS



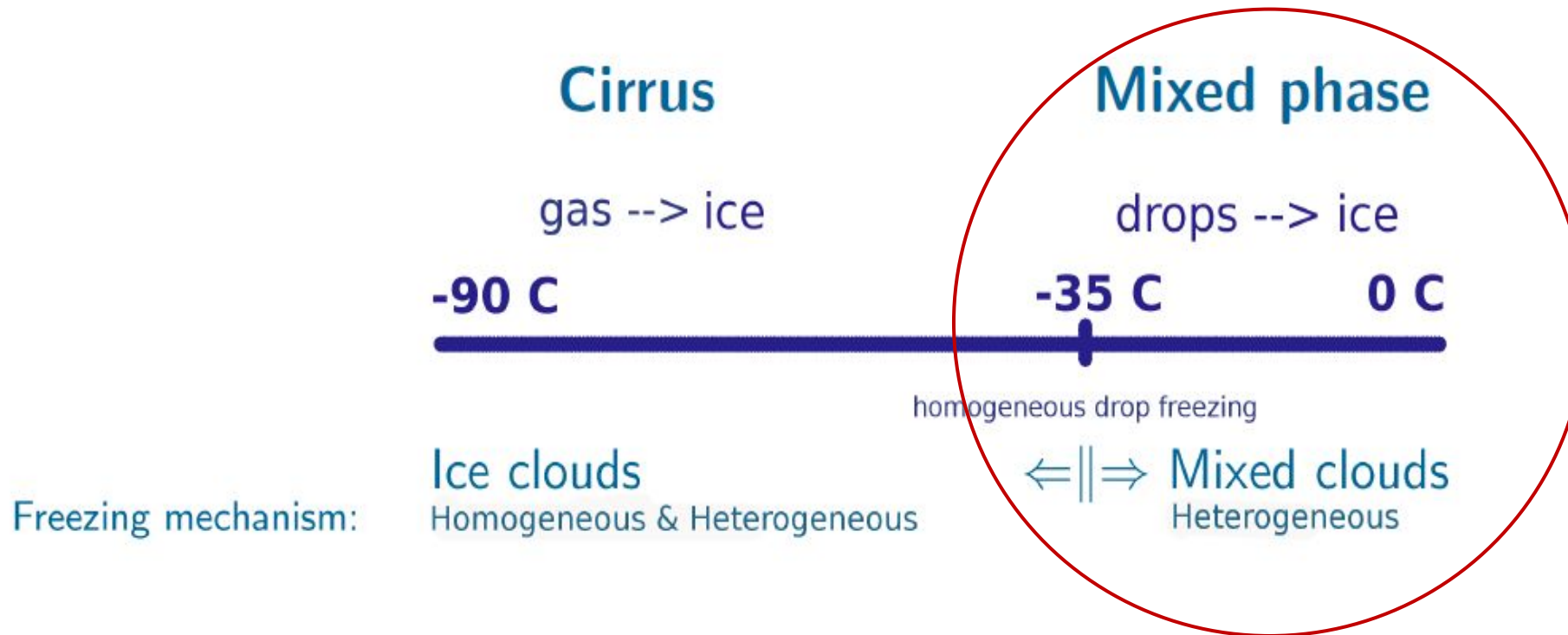
Haag, 2003

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

MIXED PHASE CLOUDS

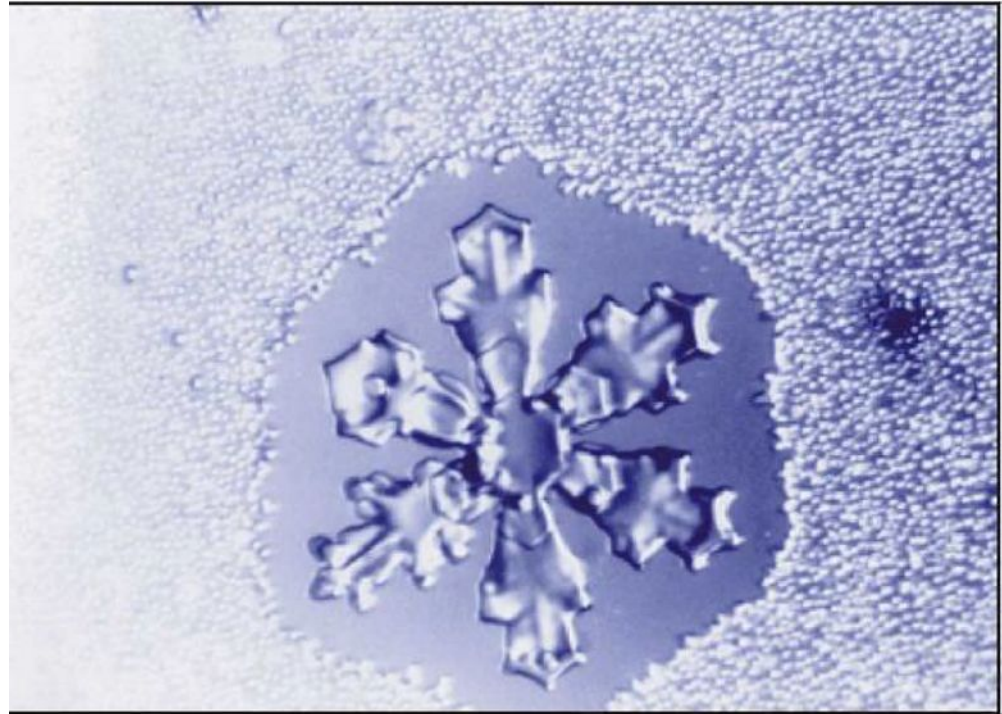


MIXED PHASE CLOUDS

Thunderstrom



EFFECT OF PHASE DIFFERENCE



R.L. Pitter

Coexistence of drops and ice crystals

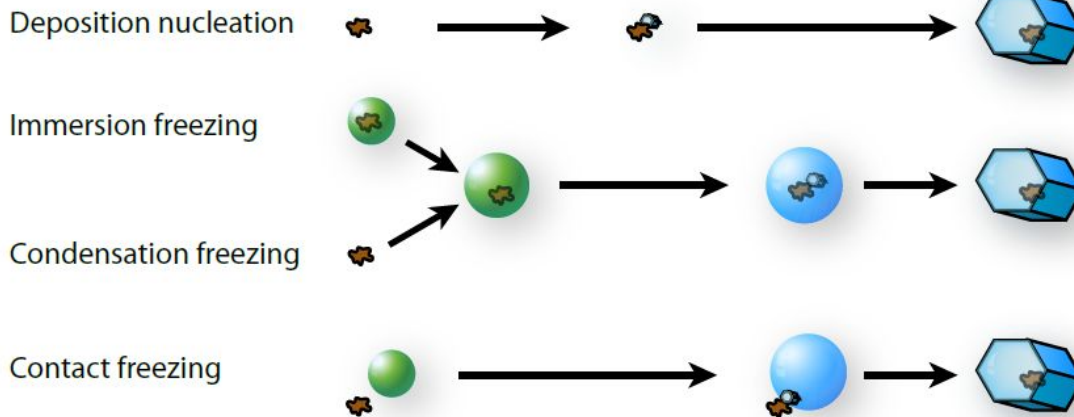
Pictures are taken from
Karlsruher Wolkenatlas
<http://www.wolkenatlas.de>

MIXED PHASE CLOUD FORMATION

Homogeneous nucleation ($T < -38^{\circ}\text{C}$)



Heterogeneous nucleation ($T < 0^{\circ}\text{C}$)



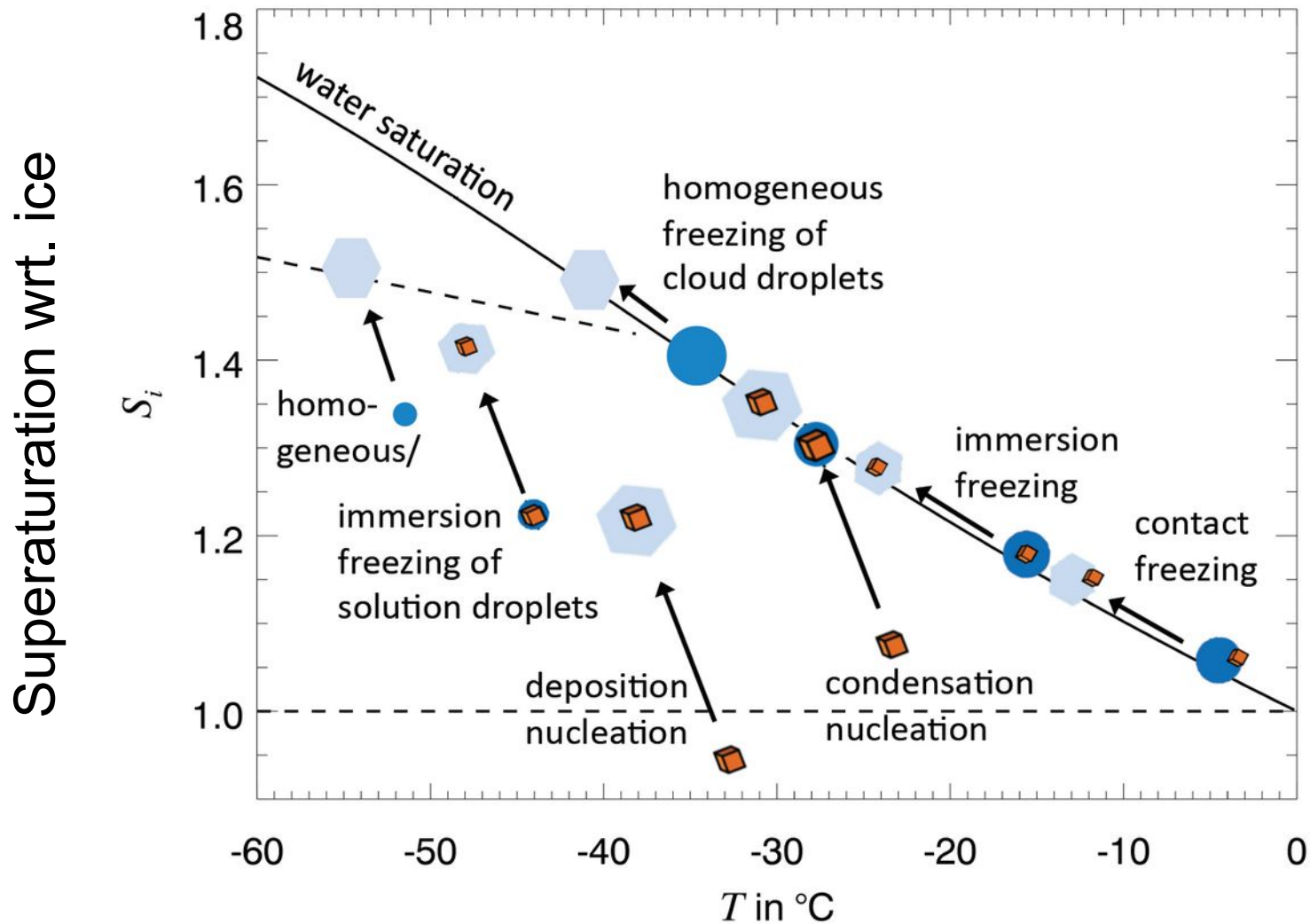
Temperature $< 0^{\circ}\text{C}$ and $> -38^{\circ}\text{C}$

-  = heterogeneous ice nucleus (e.g. mineral dust)
-  = condensation nucleus
-  = supercooled solution droplet (with e.g. SO_4^{2-} , HNO_3)
-  = ice crystal

relative humidity

temperature

MIXED PHASE CLOUDS



Hoose & Möhler, ACP; 2012

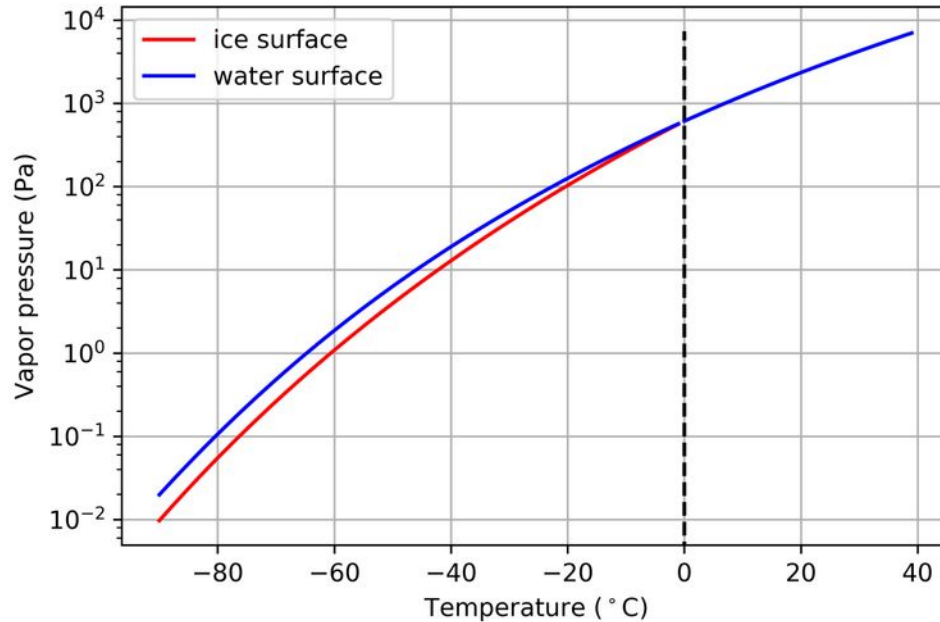
SUBTOPICS

3. Cloud formation (water and ice clouds)

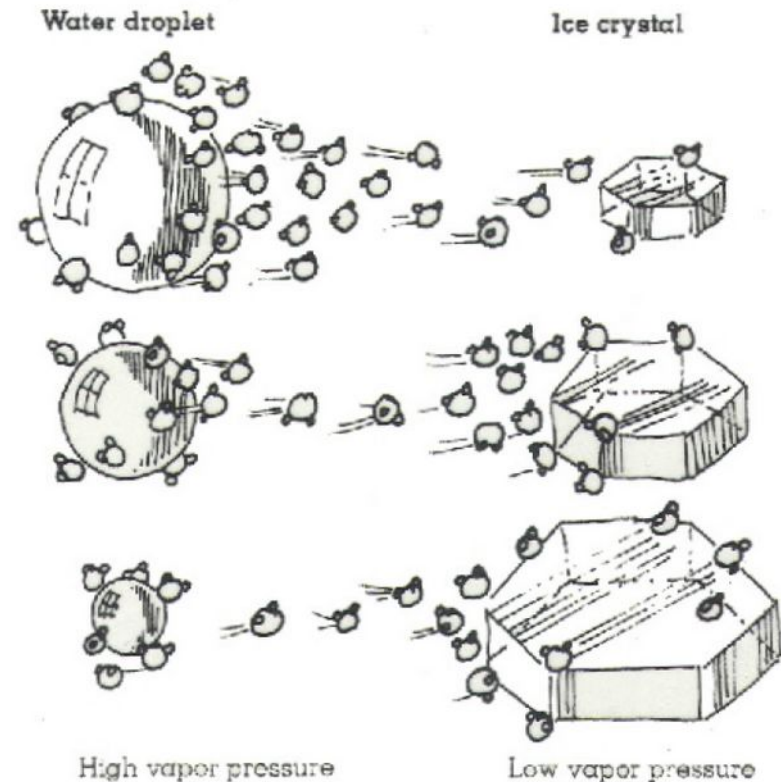
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GLACIATION OF MIXED PHASE CLOUDS

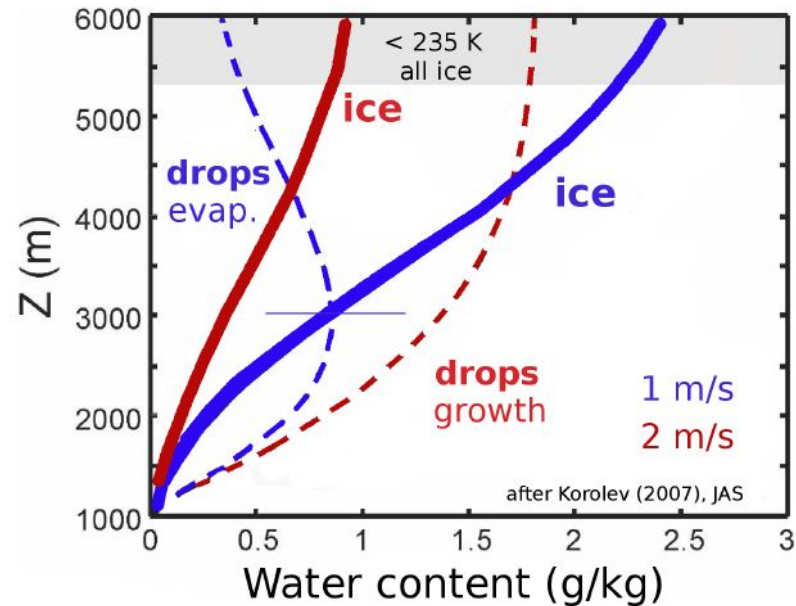
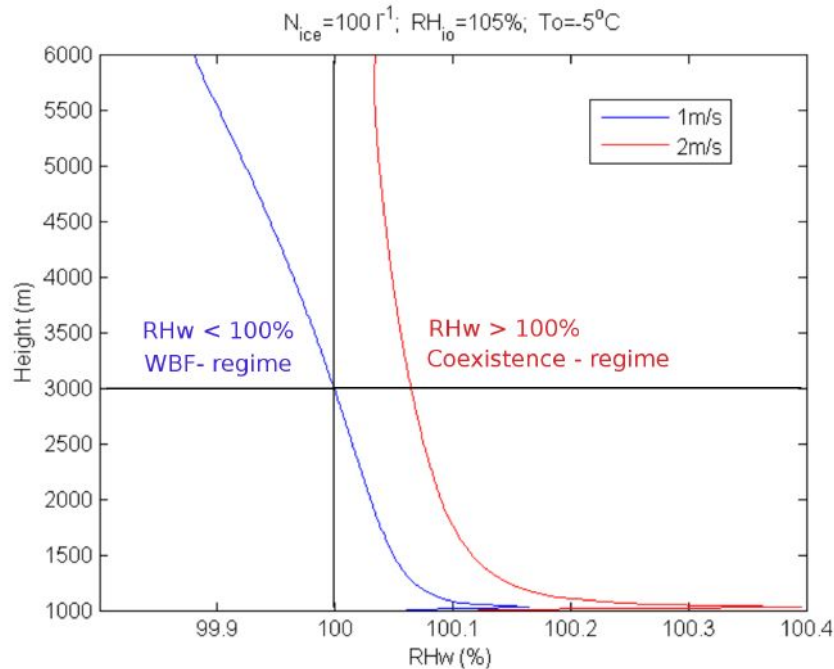
Bergeron - Findeisen Process



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



Bergeron - Findeisen Process



updraft 1 m/s: $RH_w < 100\%$

updraft 2 m/s: $RH_w > 100\%$

→ complete glaciation (Berg-Find)

→ drop - ice coexistence