

Physics and chemistry of the atmosphere

[Download the coursebook \(PDF\)](#)

ENV-320 / 5 credits

Teacher(s): [Huwald Wolf Hendrik](#), [Lehning Michael](#), [Schmale Julia](#), [Nenes Athanasios](#)

Language: English

Summary

The course provides an introduction to the physical and chemical processes that govern the atmospheric dynamics at small and large scales. The basis is laid for an in depth understanding of our atmospheric environment and the climate system.

In the programs

Environmental Sciences and Engineering

Bachelor semester 6

- **Semester:** Spring
- **Exam form:** Written (summer session)
- **Subject examined:** Physics and chemistry of the atmosphere
- **Lecture:** 3 Hour(s) per week x 14 weeks
- **Exercises:** 2 Hour(s) per week x 14 weeks
- **Project:** 1 Hour(s) per week x 14 weeks



Athanasios Nenes

Director of Laboratory of atmospheric processes and their impacts (LAPI)

<http://lapi.epfl.ch>

Topics covered in class: Aerosol-Cloud interactions and Cloud Processes

LAPI – Athanasios (Thanos) Nenes

Laboratory of atmospheric processes and their impacts

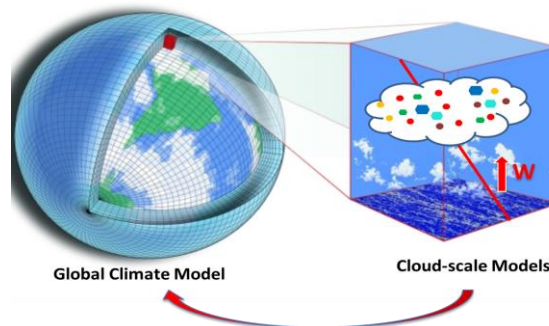


<http://lapi.epfl.ch>

Field and Laboratory Observations



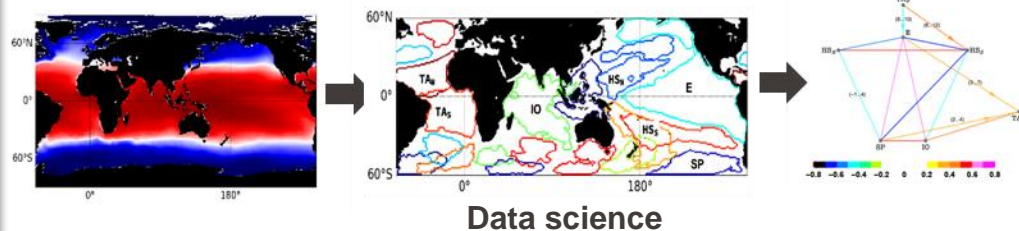
Modeling



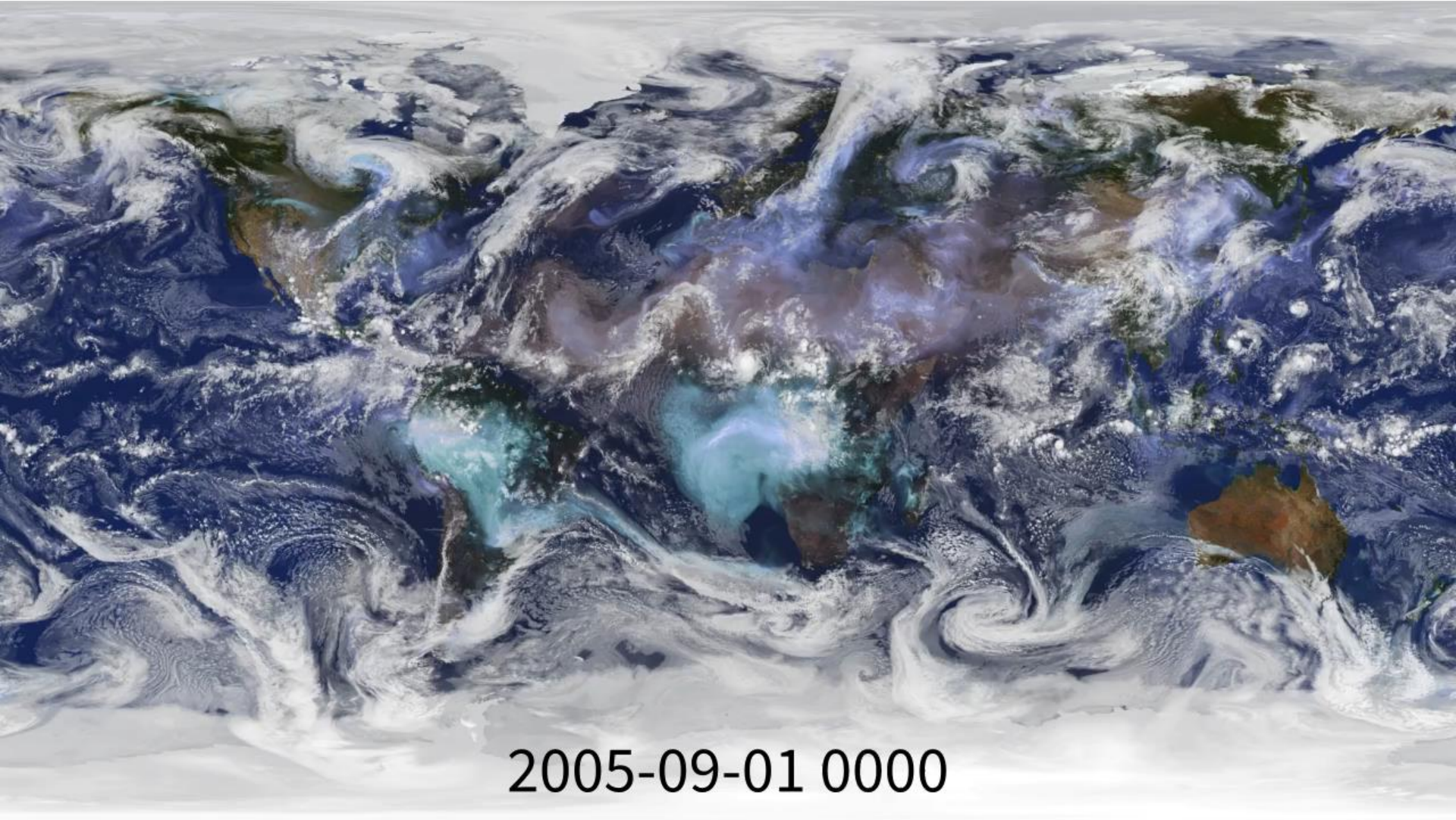
Instrumentation



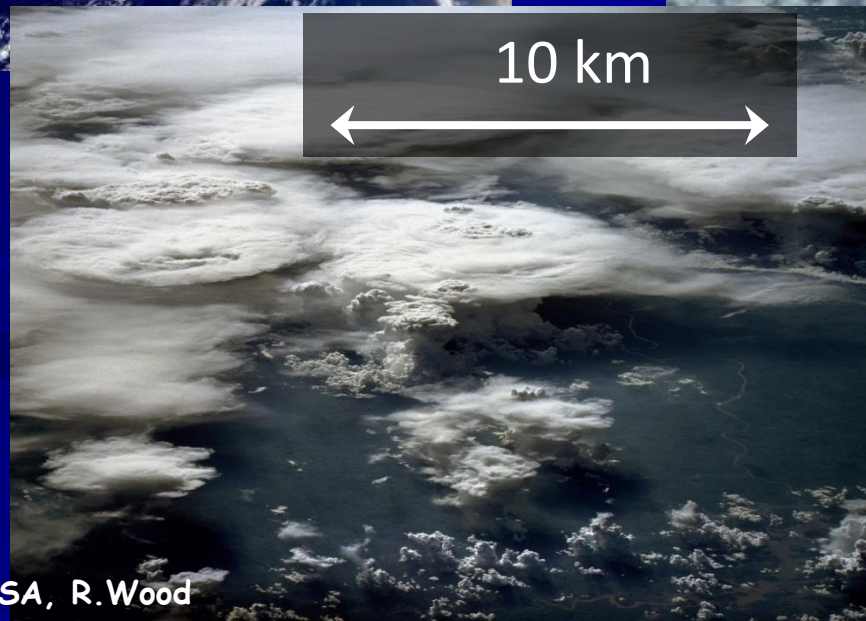
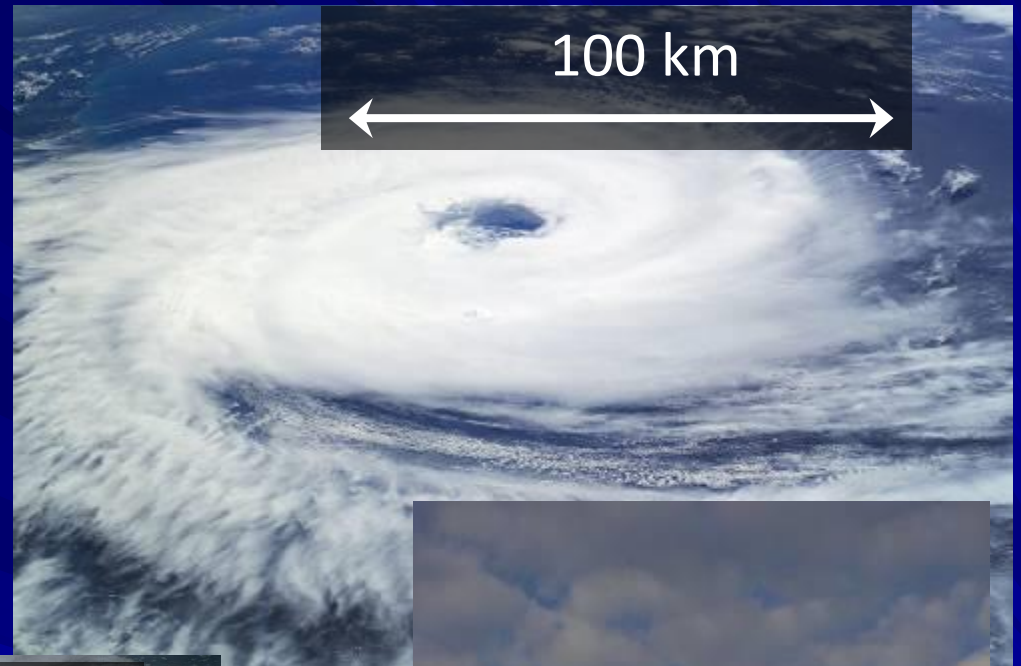
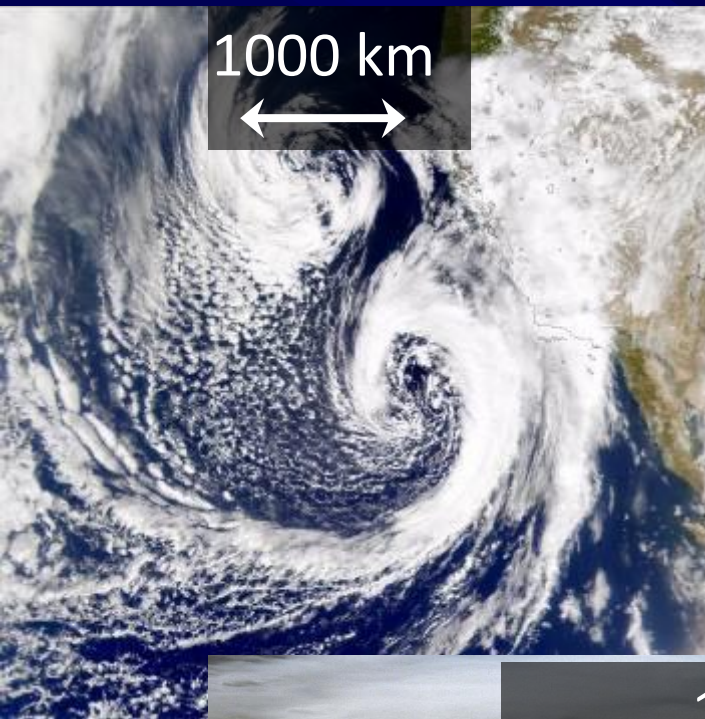
Cloud Condensation Nuclei Counter, US Patent 7,656,510



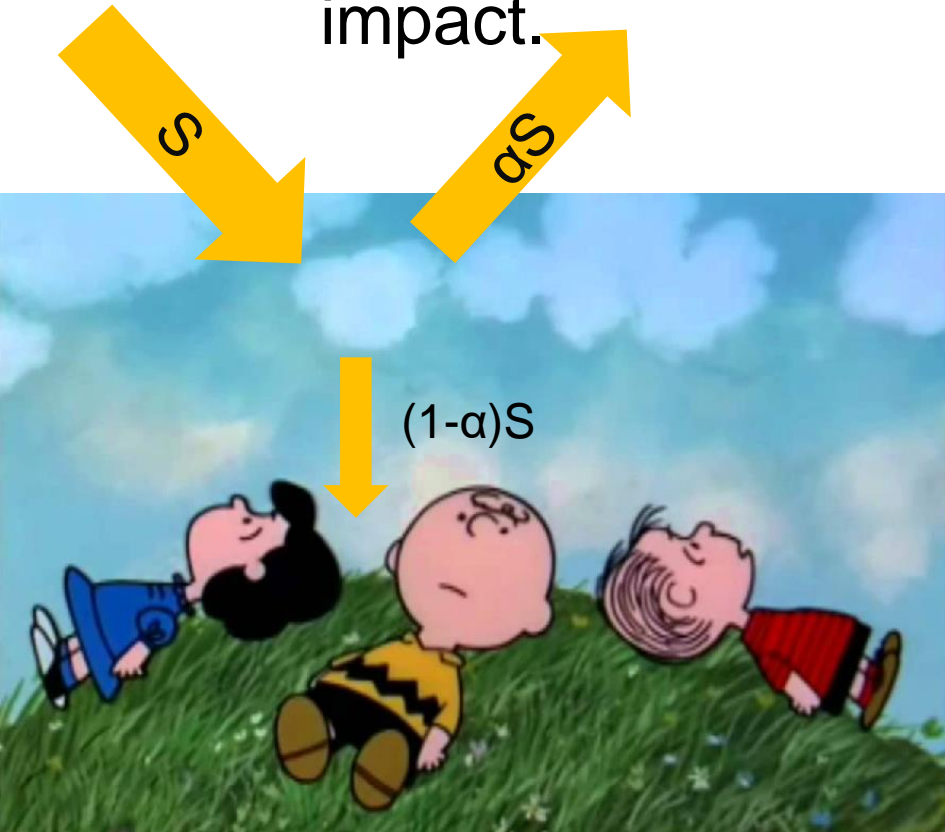
Clouds are everywhere and at all scales...



Clouds are everywhere and at all scales...



Clouds have an important **radiative** impact.



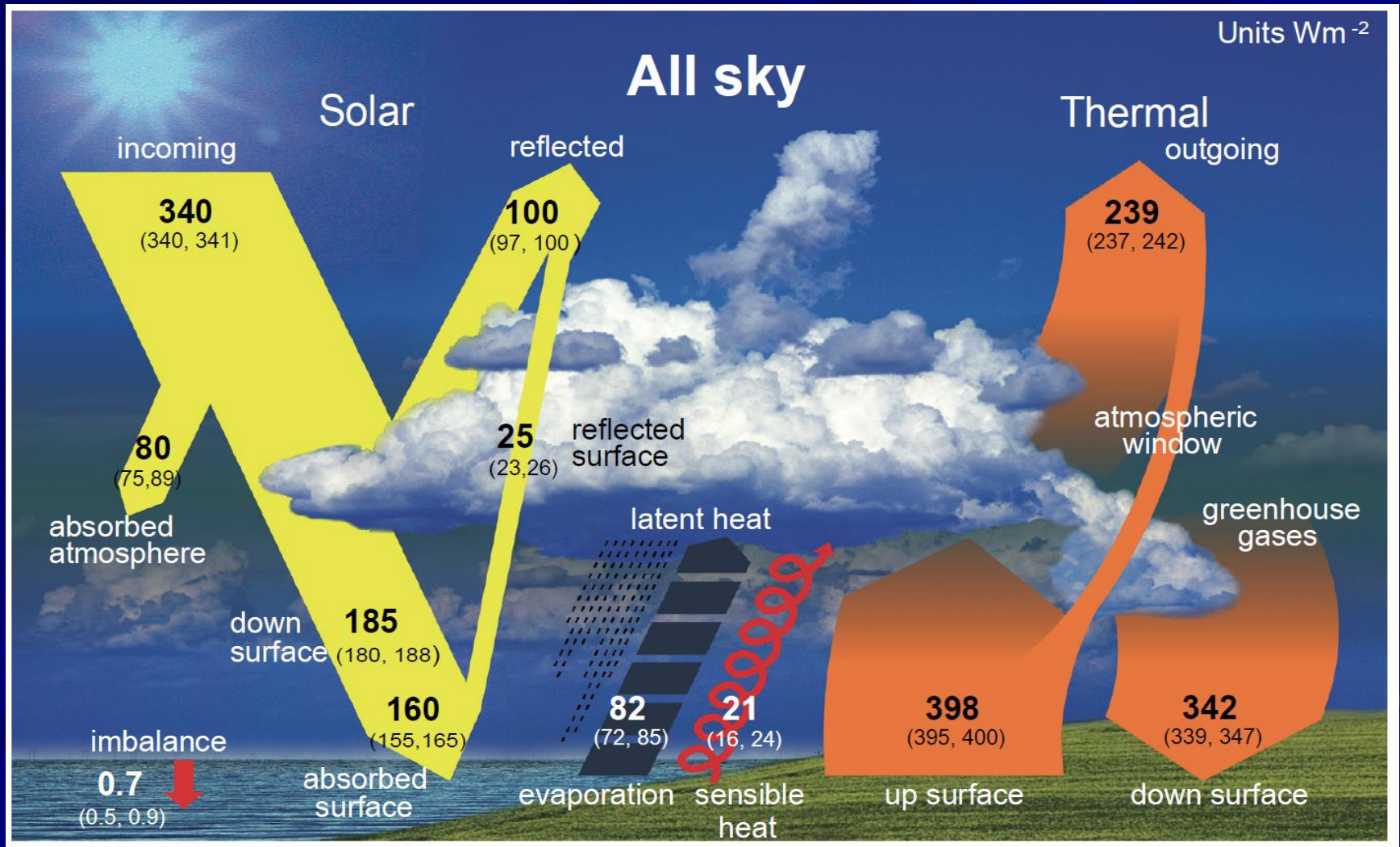
$P(t)$

Photo from Wynn Bullock

Clouds also have an important **hydrological** impact.

Both critically important for regional and global climate

Clouds play a central role in the climate system



Based on J.T. Houghton: "The science of climate change"

Cold cloud processes

- Ice particle formation
- Ice multiplication
- Growth
- Precipitation
- (Evaporation)

Warm cloud processes

- Droplet formation
- Droplet growth
- Precipitation
- (Evaporation)

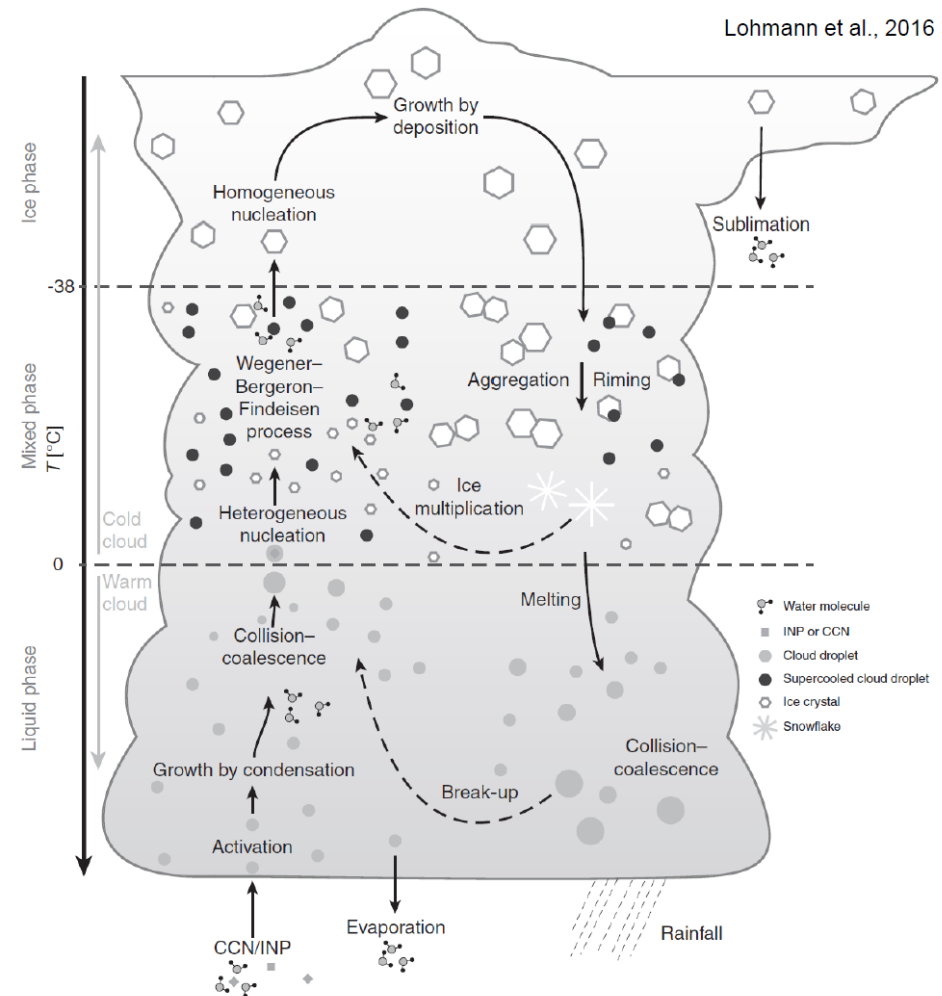


Fig. 8.19

Microphysical processes occurring in a convective cloud with a cloud base temperature higher than 0°C and a cloud top temperature lower than -38°C , so that warm-, mixed- and ice-phase processes take place. The small arrows show typical trajectories of a cloud particle.

Types of clouds

Base height

- decrease in tropopause height with latitude
- decrease in cloud elevation

Structure

- reflects instability and nuclei

Phase

- *warm clouds*: liquid water
- *cold clouds*: ice crystals or mix



Cirro-form
Ice crystals
“Wispy” appearance



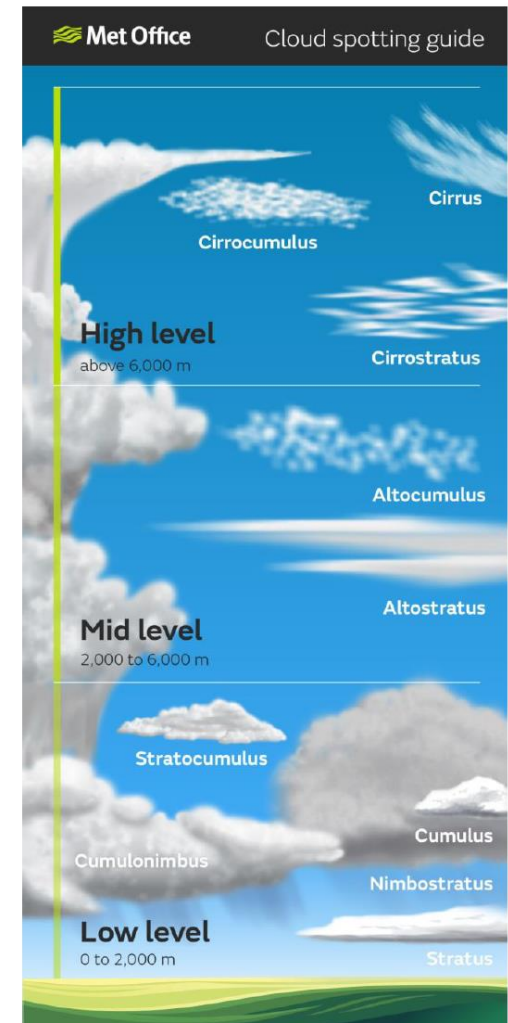
Cumulo-form
Vertical instability
Fluffy appearance



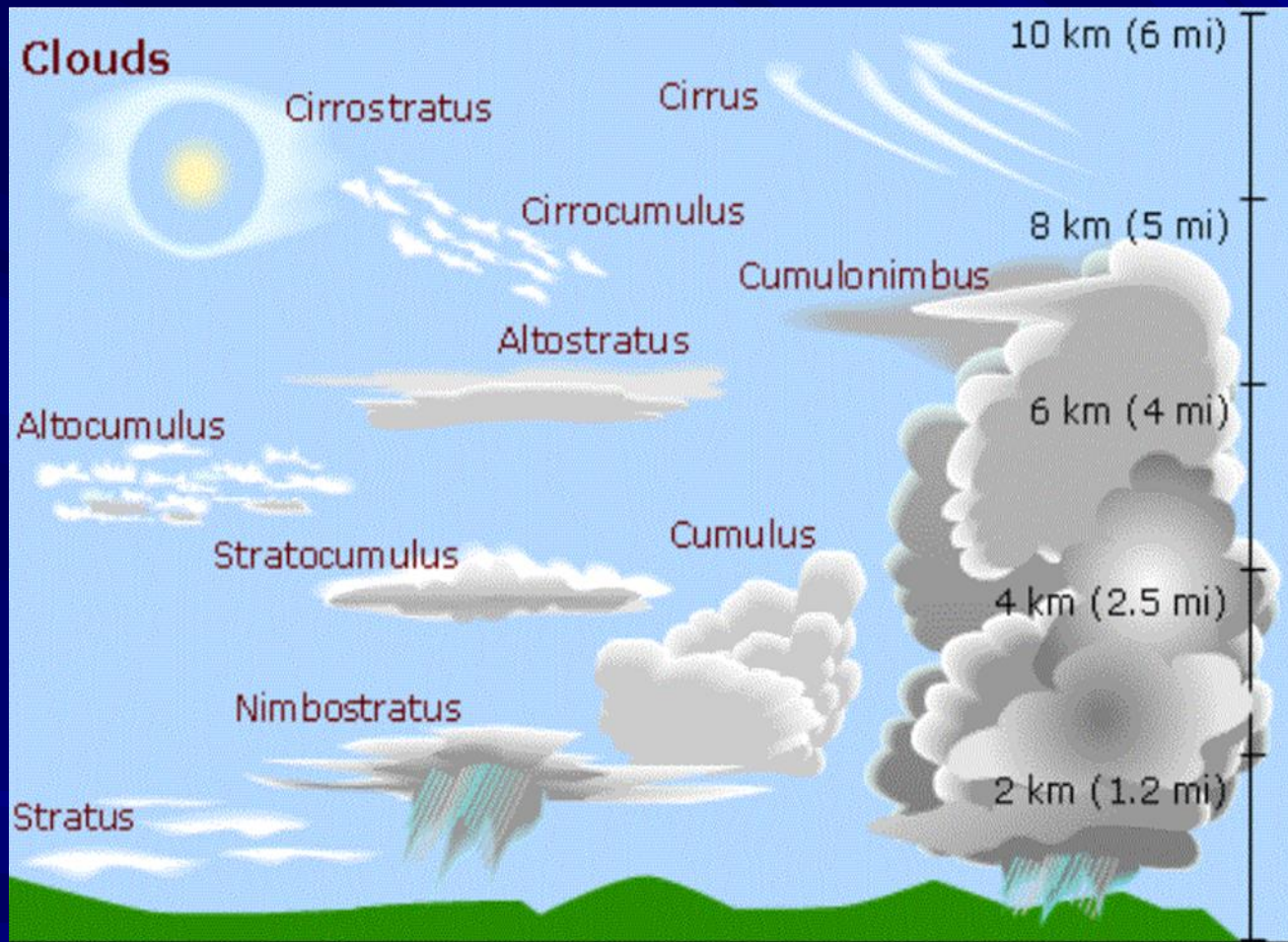
Strato-form
Vertical stability
Broad, layered



Nimbo-form
Great vertical height
Rain clouds



Cloud impacts vary alot

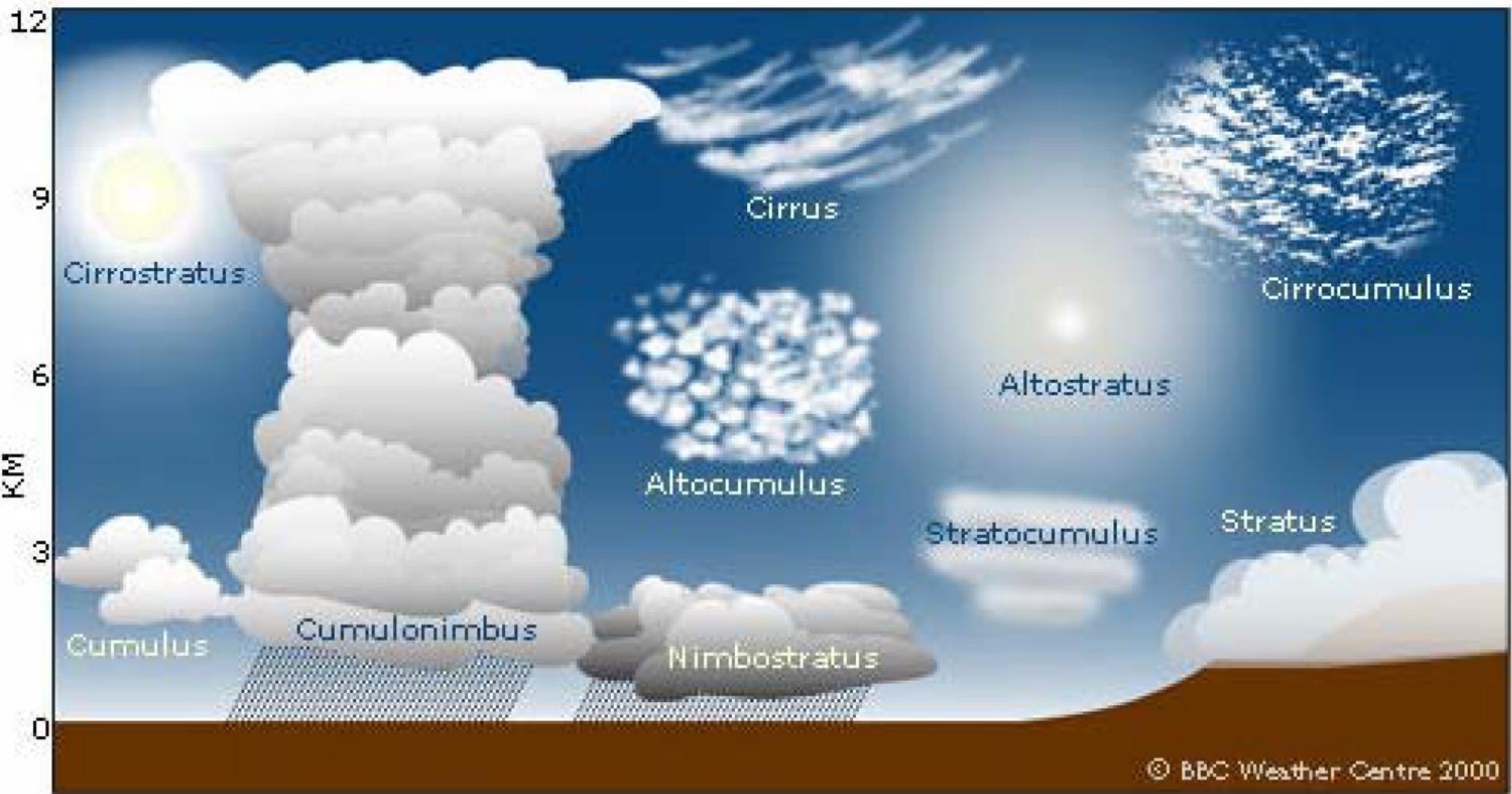


**High clouds
(ice crystals):
warm climate**

Mid-level:
Warm/cool

**Low clouds
(liquid drops):
cool/ climate**

Common Cloud Names, Shapes, and Altitudes:



Low Clouds

Middle Clouds

High Clouds

Genus	Species (can be only one)	Varieties (can be more than one)
Cumulus	humilis mediocris congestus fractus	radiatus
Cumulonimbus (extend through all 3 levels)	calvus capillatus	(none)
Stratus	nebulosus fractus	opacus, translucidus, undulatus
Stratocumulus	stratiformis lenticularis castellanus	translucidus, perlucidus, opacus, duplicatus, undulatus, radiatus, lacunosus
Alto cumulus	stratiformis lenticularis castellanus floccus	translucidus, perlucidus, opacus, duplicatus, undulatus, radiatus, lacunosus
Altostratus	(none)	translucidus, perlucidus, opacus, duplicatus, undulatus, radiatus
Nimbostratus (extend through 1+ levels)	(none)	(none)
Cirrus	fibratus uncinus spissatus castellanus floccus	intortus, radiatus, vertebratus, duplicatus
Cirrocumulus	stratiformis lenticularis castellanus floccus	undulatus, lacunosus
Cirrostratus	fibratus nebulosus	duplicatus, undulatus

Cloud Classification:

Clouds are classified using a Latin “Linnean” system based on genera and species, originally developed by Luke Howard, an amateur meteorologist and Quaker in 1802.

The modern classification scheme is based on Howard’s system and is detailed in *The International Cloud Atlas*, published by the World Meteorological Organization since 1896.

In addition to standardizing the genus-species system, the WMO also classified clouds by altitude and divided the troposphere into 3 levels:

Low-level Clouds: < 6,500 ft.
 Mid-level Clouds: 6,500 to 23,000 ft.
 High-level Clouds: 16,500 to 45,000 ft.

Cumulus Clouds

("The cloud of choice for 6-yr.-olds")



There are three species of cumulus clouds:

- *humilis* are wider than they are tall
- *mediocris* are as wide as they are tall
- *congestus* are taller than they are wide

Often called "fair-weather" clouds, cumulus clouds are common over land on sunny days, when the sun heats the land creating thermal convection currents

Each thermal is distinct, and, consequently, each cumulus cloud is a distinct puff

Fast Facts:

Typical Altitude: 2,000-3,000 ft.

Location: Worldwide (except in Antarctica, where it's too cold)

Precipitation: Generally none, except for brief showers from congestus

Composition: Liquid water

Formation: Thermal convection currents



Cumulonimbus Clouds

("The towering thunderclouds that scare us senseless")

Three critical conditions for cumulonimbus formation:

- Ready supply of warm, moist air, which rises at speeds of up to 25-70 mph
- Tropospheric winds need to increase considerably with height to encourage it to slant forward
- The atmosphere around the cloud needs to be "unstable" – no temp. inversions here

Fast Facts:

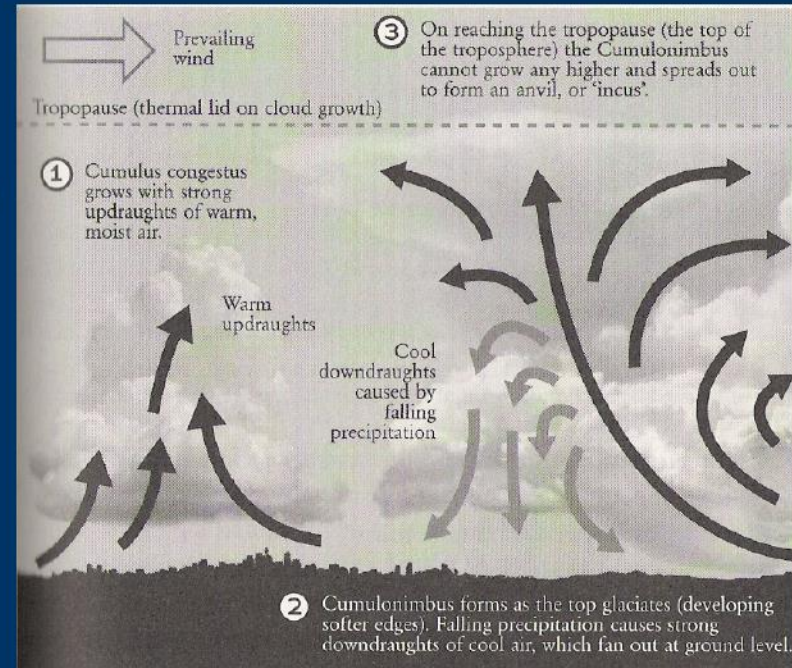
Typical Altitude: 2,000-45,000 ft.

Location: Common in tropics and temperate regions, rare at poles

Precipitation: Heavy downpours, hail

Composition: Liquid water throughout, ice crystals at the top

Formation: Upwardly mobile cumulus congestus clouds (thermals)



Stratus Clouds

("The clouds that weigh heavily on your mood")

Stratus clouds are the lowest forming and are often called fog or mists when they are earth-bound

Stratus clouds are formed when a large air mass cools at the same time (e.g. – a warm air parcel drifts into or above a cooler region)



Fast Facts:

Typical Altitude: 0-6,500 ft.

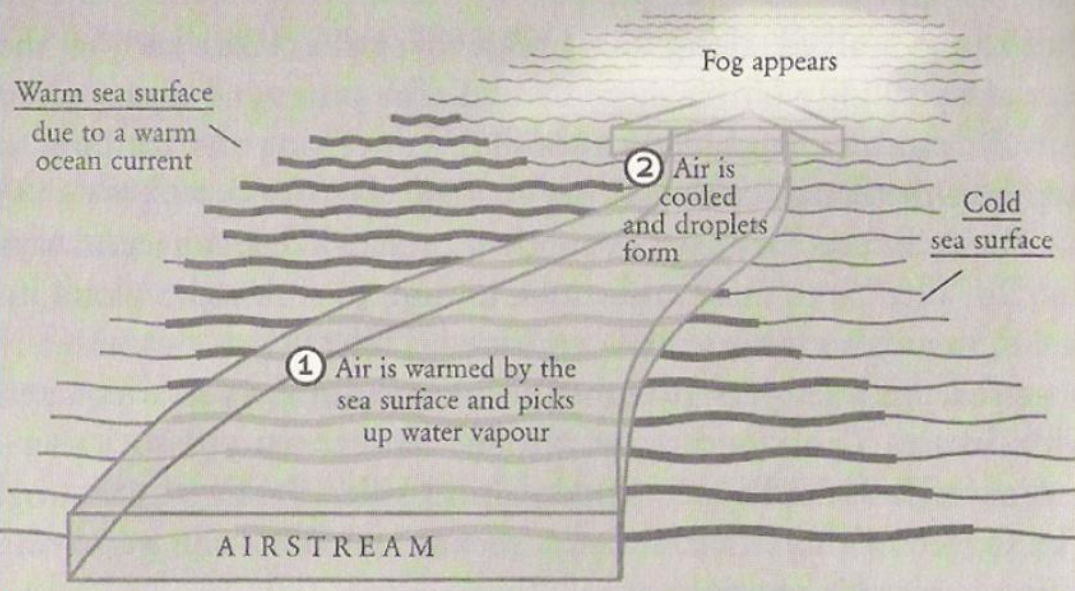
Location: Worldwide, but especially common around coasts and mountains

Precipitation: No more than light drizzle

Composition: Liquid water

Formation: Advective or radiative cooling

ADVECTION FOG



Stratocumulus Clouds

("The low, puffy layers")

Similar to cumulus clouds in form and composition, stratocumulus clouds are textured and puffy, but also joined into a semi-continuous layer

Stratocumulus clouds usually form from cumulus or stratus clouds

Fast Facts:

Typical Altitude: 2,000-6,500 ft.

Location: Worldwide – very common

Precipitation: Occasional light rain, snow

Composition: Liquid water

Formation: Spreading and joining of cumulus clouds below a temperature inversion, wind turbulence in a stratus layer



Altostratus Clouds

("The boring clouds")

Below 6,500 ft. it's stratus

Between 6,500 and 23,000 ft. it's altostratus

Boring! – but being so high up, they do make for nice sunsets.



Fast Facts:

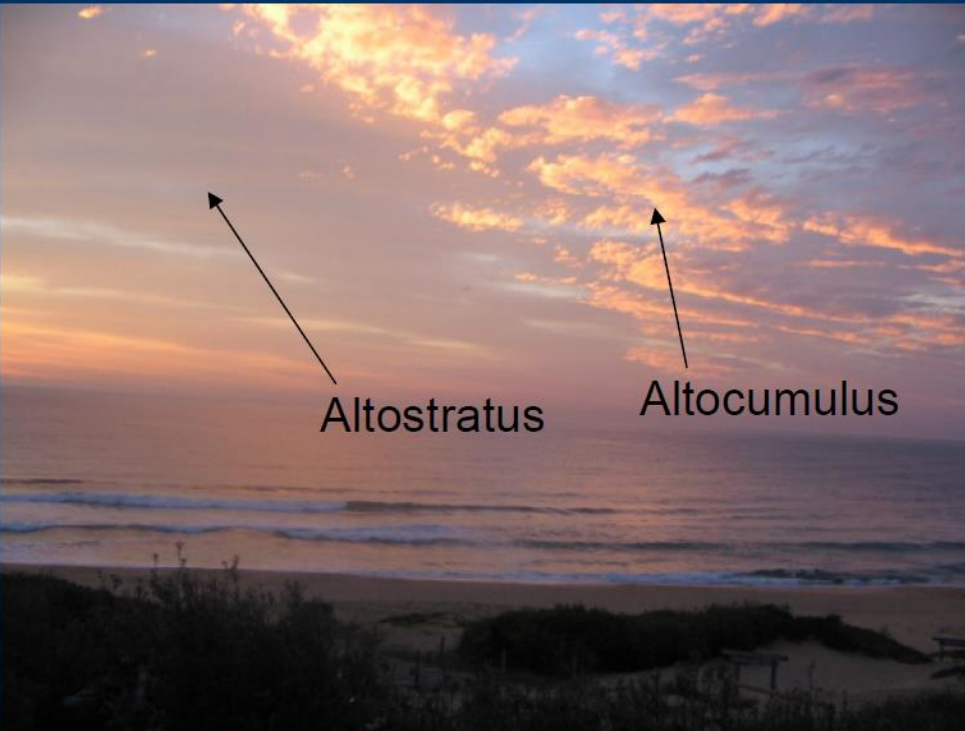
Typical Altitude: 6,500-16,500 ft.

Location: Worldwide, common in middle latitudes

Precipitation: Occasional light rain, snow

Composition: Both liquid water, and ice crystals

Formation: Usually formed from the thickening and lowering of a cirrostratus cloud on its way to becoming a nimbostratus cloud



Altostratus are potentially dangerous to aircraft because they can cause ice accumulation on the wings.

Nimbostratus Clouds

("Rainy day clouds")

The nimbostratus cloud has no species or varieties. It is a thick, wet blanket with a ragged base caused by the continual precipitation



Fast Facts:

Typical Altitude: 2,000-18,000 ft.

Location: Worldwide, common in middle latitudes

Precipitation: Moderate to heavy rain or snow, which is generally steady and prolonged

Composition: Liquid water, raindrops, snowflakes and ice crystals

Formation: Usually formed from the thickening and lowering of an altostratus cloud



Cirrus Clouds

("Delicate cloud streaks")

Cirrus clouds are the highest of all clouds and are composed entirely of ice crystals.

Cirrus clouds are precipitating clouds, although the ice crystals evaporate high above the earth's surface.

The crystals, caught in 100-150 mph winds create wisps of cloud.

Fast Facts:

Typical Altitude: 16,500-45,000 ft.

Location: Worldwide

Precipitation: None that reaches ground

Composition: Ice crystals

Formation: Fall streaks of ice crystals in upper troposphere winds



Cirrocumulus Clouds

("Regularly spaced cloudlets, often rippled")

Cirrocumulus clouds are usually a transitional phase between cirrus and cirrostratus clouds.

Large numbers of cirrocumulus clouds may indicate poor weather is approaching.

Fast Facts:

Typical Altitude: 16,500-45,000 ft.

Location: Worldwide

Precipitation: None that reaches ground

Composition: Ice crystals

Formation: Cloudlets formed by choppy winds and high moisture levels in upper troposphere



Cirrostratus Clouds

("Delicate cloud streaks")

Cirrostratus clouds are difficult to spot and appear as a pale, milky lightening of the sky.

Cirrostratus clouds never block out the sun completely, but rather produce a variety of optical effects.

Fast Facts:

Typical Altitude: 20,000-42,000 ft.

Location: Worldwide

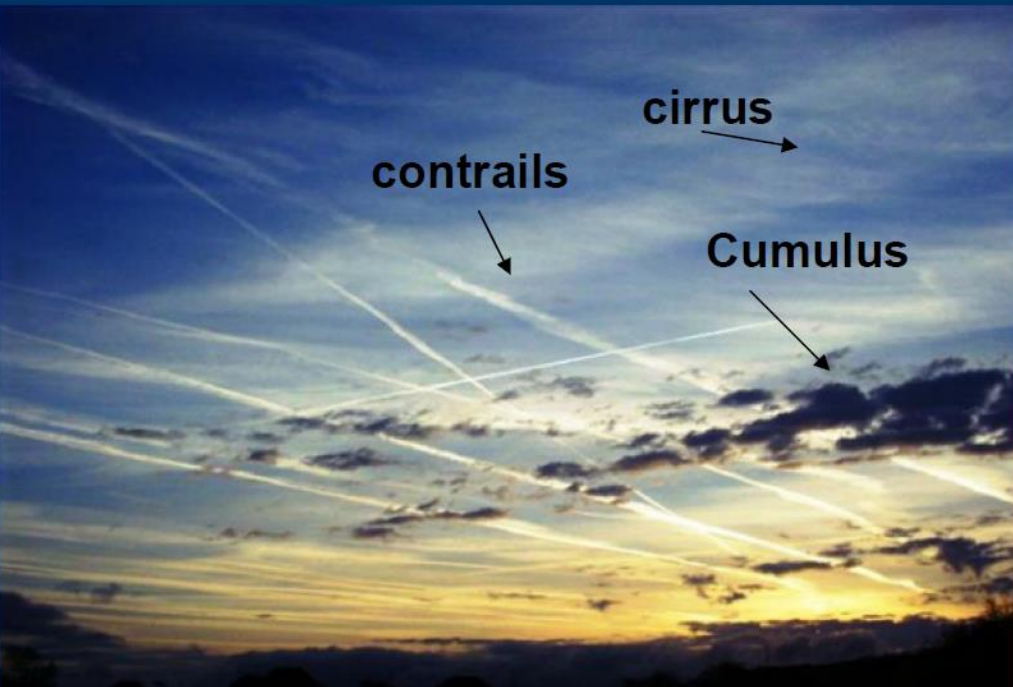
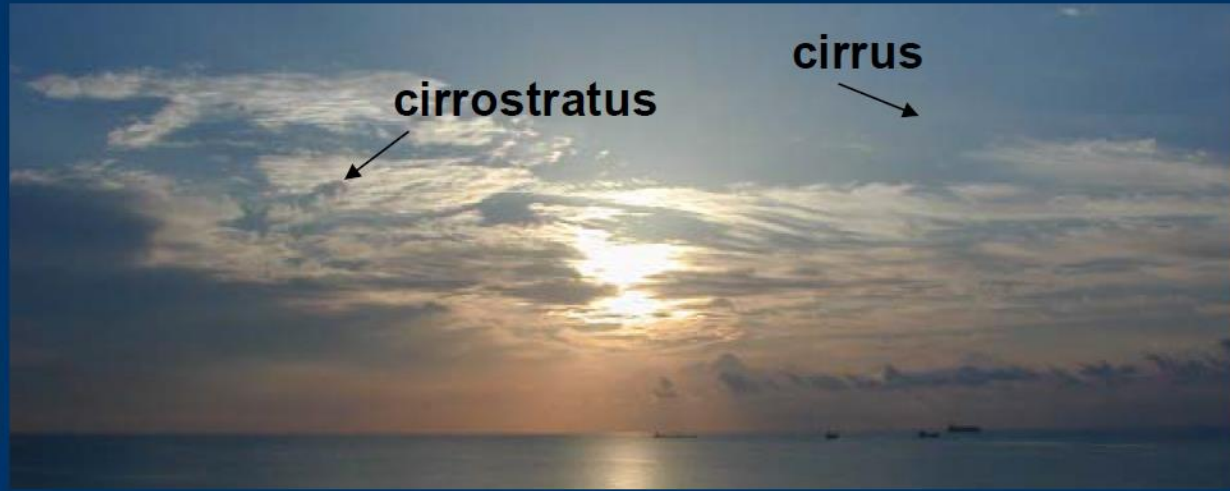
Precipitation: None

Composition: Ice crystals

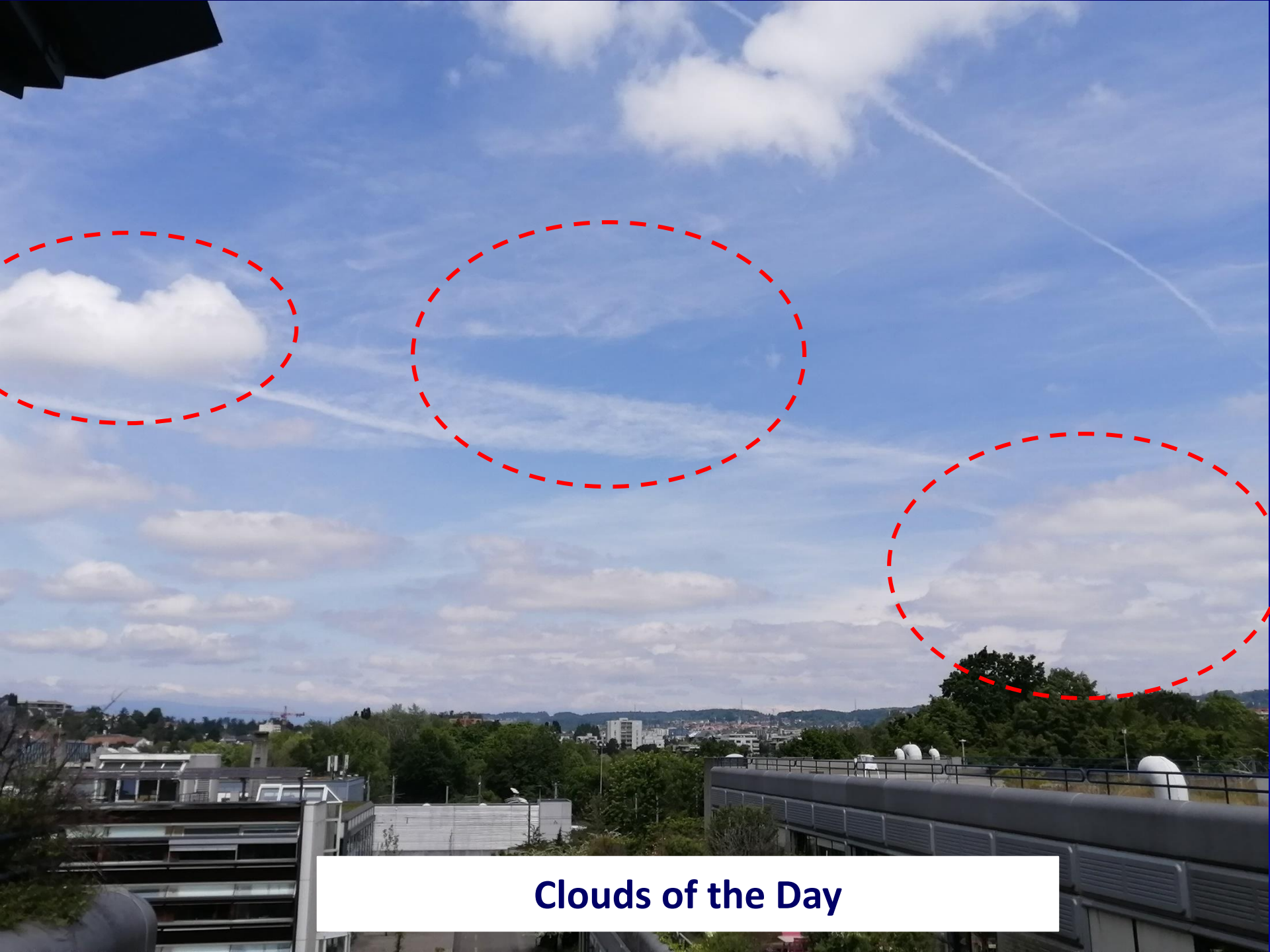
Formation: Spreading and joining of cirrus clouds



Contrails and Others



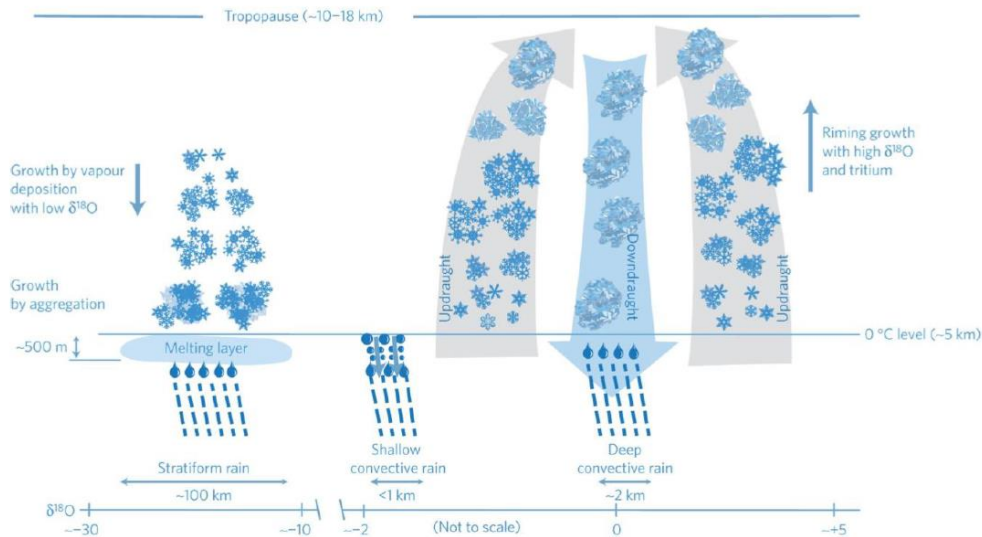
Clouds and precipitation



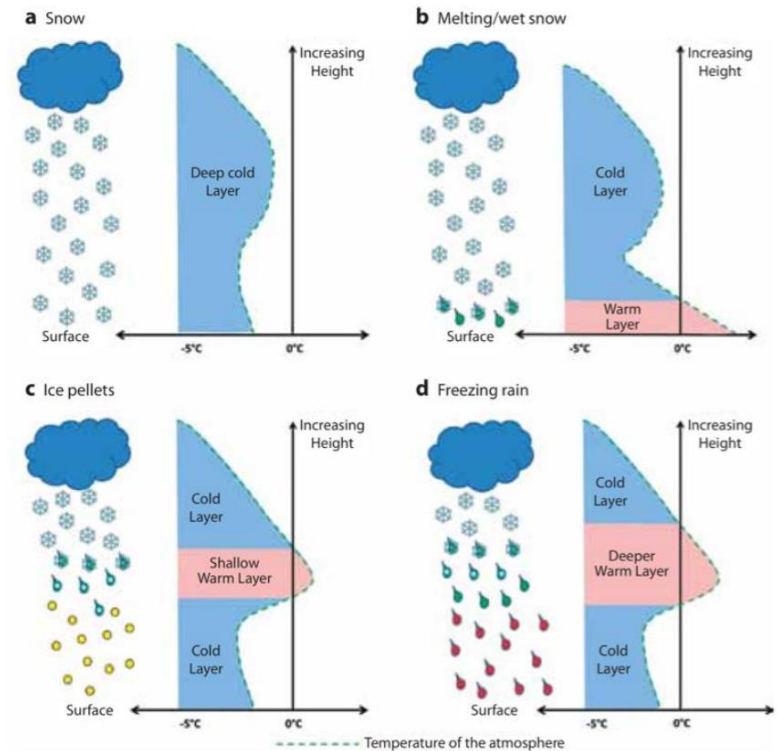
Clouds of the Day

Types of precipitation

Spatial extent and intensity



Phase (winter precipitation)



- Lifetime of water in troposphere:
9-11 days
- 1/3 of precipitation over land originates
from water evaporated over oceans
- Excess precipitation over land feeds
ocean as runoff

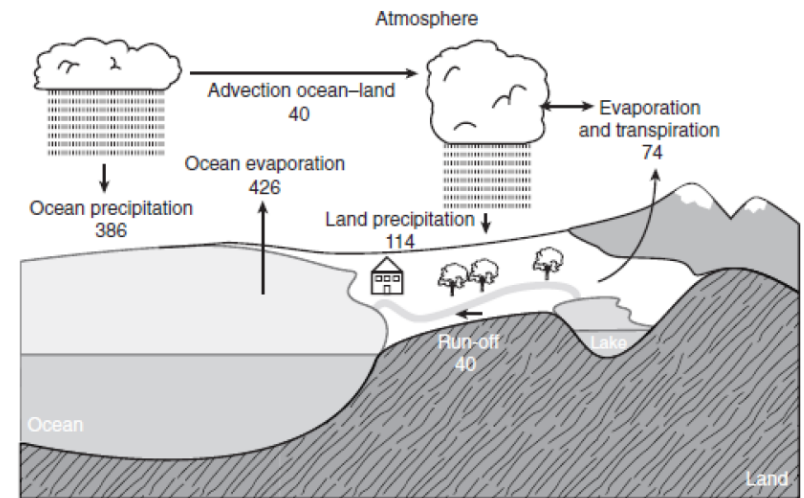


Fig. 9.18

Schematic surface view, with a vertical cross section, of various fluxes and reservoirs within the atmospheric branch of the hydrological cycle, with their annual average magnitudes based on data of Trenberth *et al.* (2011) and references therein. The water vapor reservoirs are given in 10^{15} kg and the fluxes in $10^{15} \text{ kg yr}^{-1}$.

Frequency of occurrence of rain from liquid-, mixed-, and ice-phase clouds derived from A-Train satellite retrievals

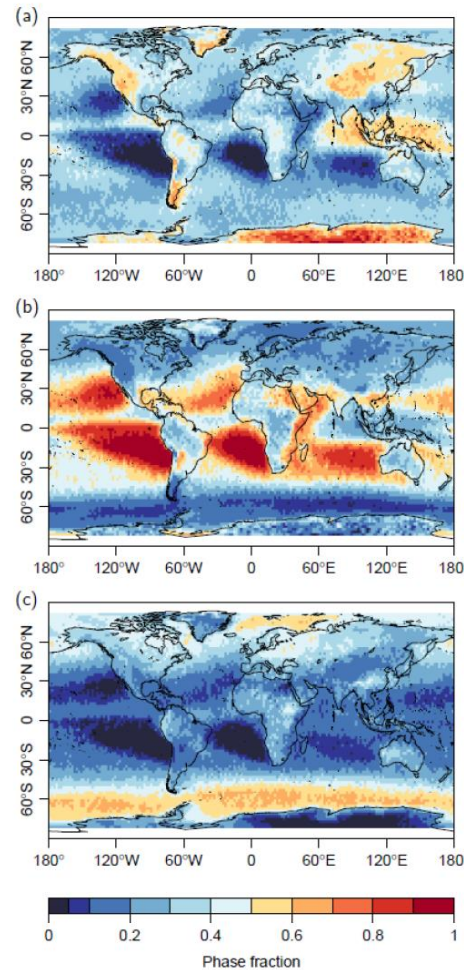
Johannes Mülmenstädt¹, O. Sourdeval¹, J. Delanoë², and J. Quaas¹

¹Institute of Meteorology, Universität Leipzig, Leipzig, Germany, ²Laboratoire Atmosphères, Milieux, Observations Spatiales (LATMOS UVSQ/CNRS/UPMC/IPSL), Guyancourt, France

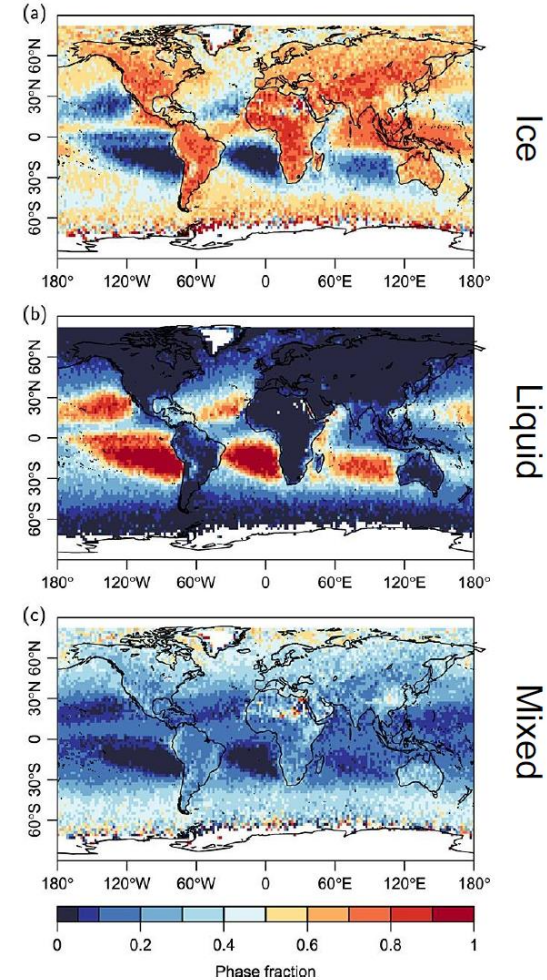
Land-sea contrast in *cloud droplet radii*:

- moisture
- aerosol concentrations
- updrafts (higher supersaturations)

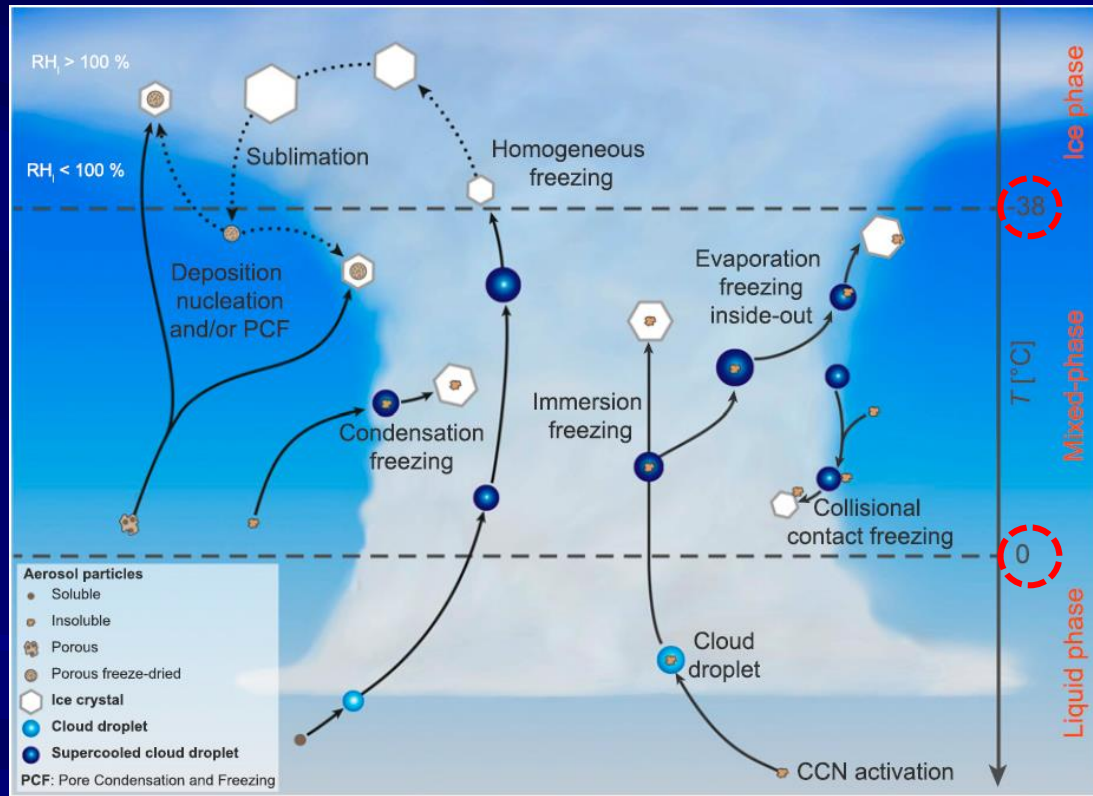
Fraction of cloud types



Fraction of rain



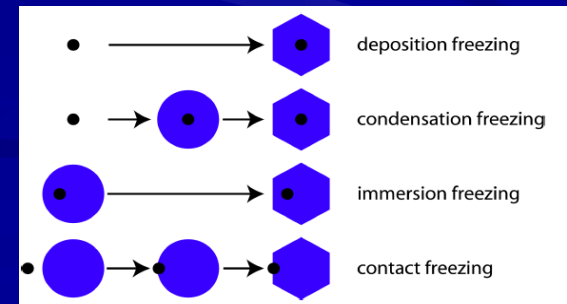
Liquid+ice ("mixed-phase") clouds occur very frequently



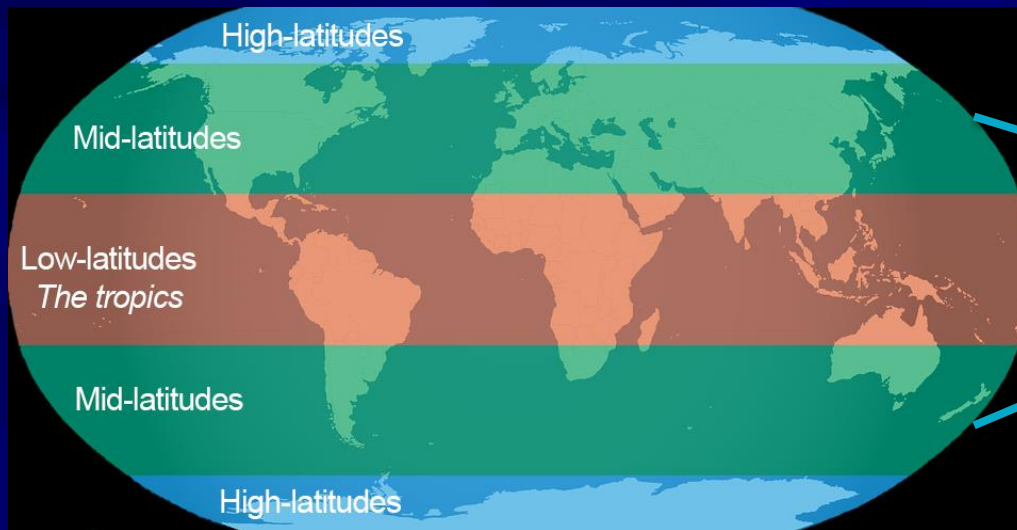
Kanji et al., 2017

Pure ice cloud (glaciated)
Consisting only of ice particles
(formed through homogeneous
freezing of cloud droplets)

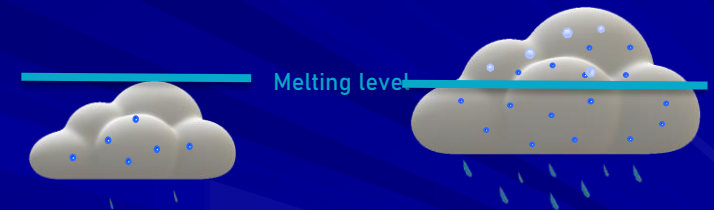
Mixed-phase cloud
Consisting of both supercooled
liquid droplets (in a
metastable phase) and ice
(formed mainly through
heterogeneous ice nucleation)



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



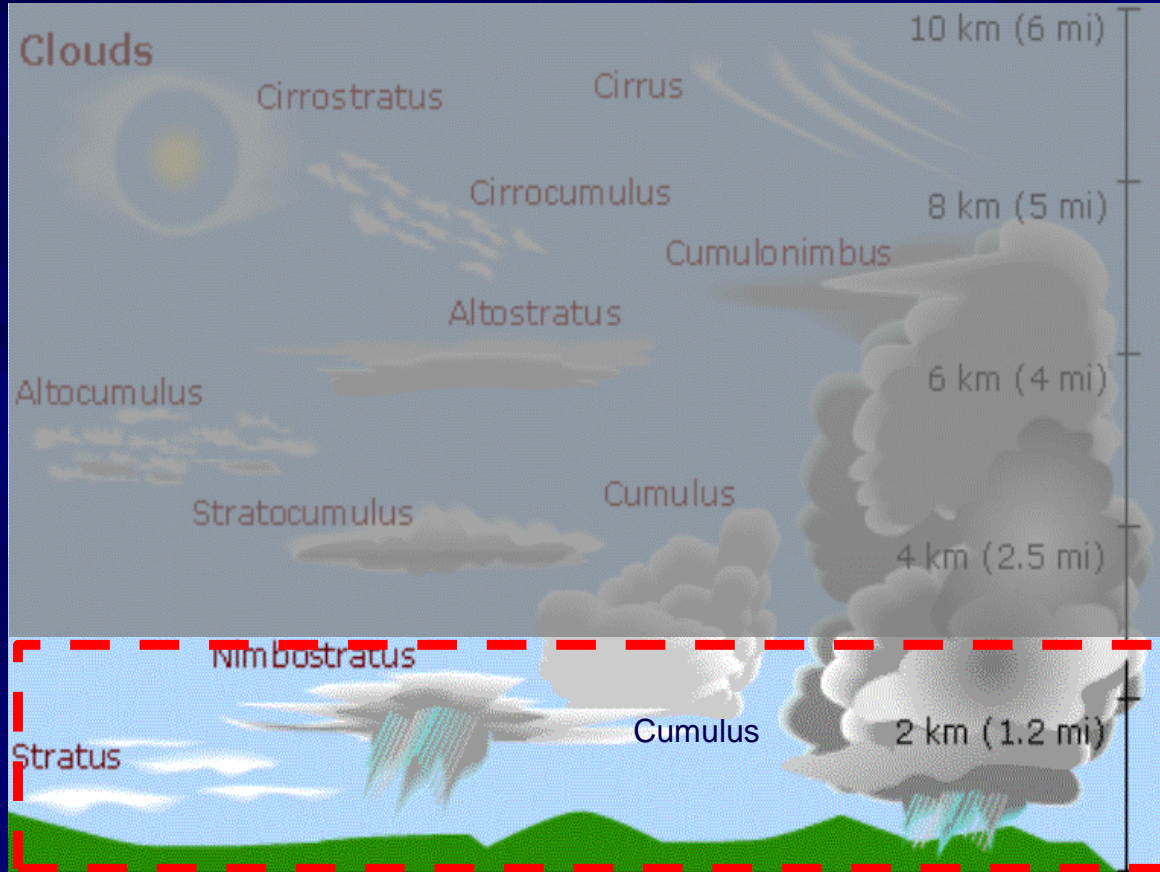
30-50% of precipitation occurs from the ice phase



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 . . .”

Let's focus on LIQUID clouds first



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

Cloud particles are formed on **aerosol particles**.
These aerosol-cloud interactions affect climate

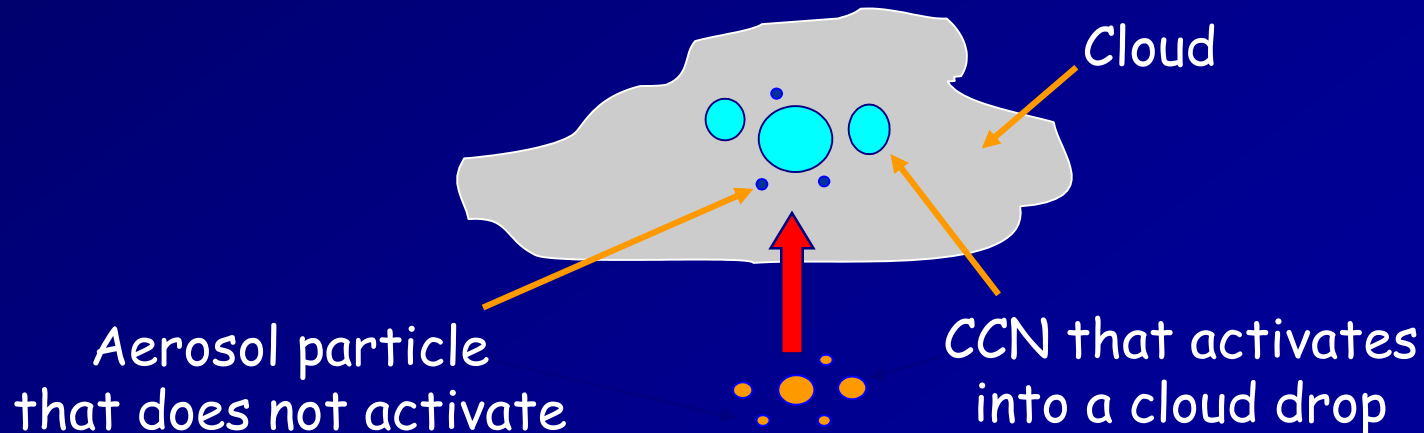
How do (liquid water) clouds form?

Clouds form in regions of the atmosphere where there is too much water vapor (it is "supersaturated").

This happens when air is cooled (primarily through expansion in updraft regions and radiative cooling).

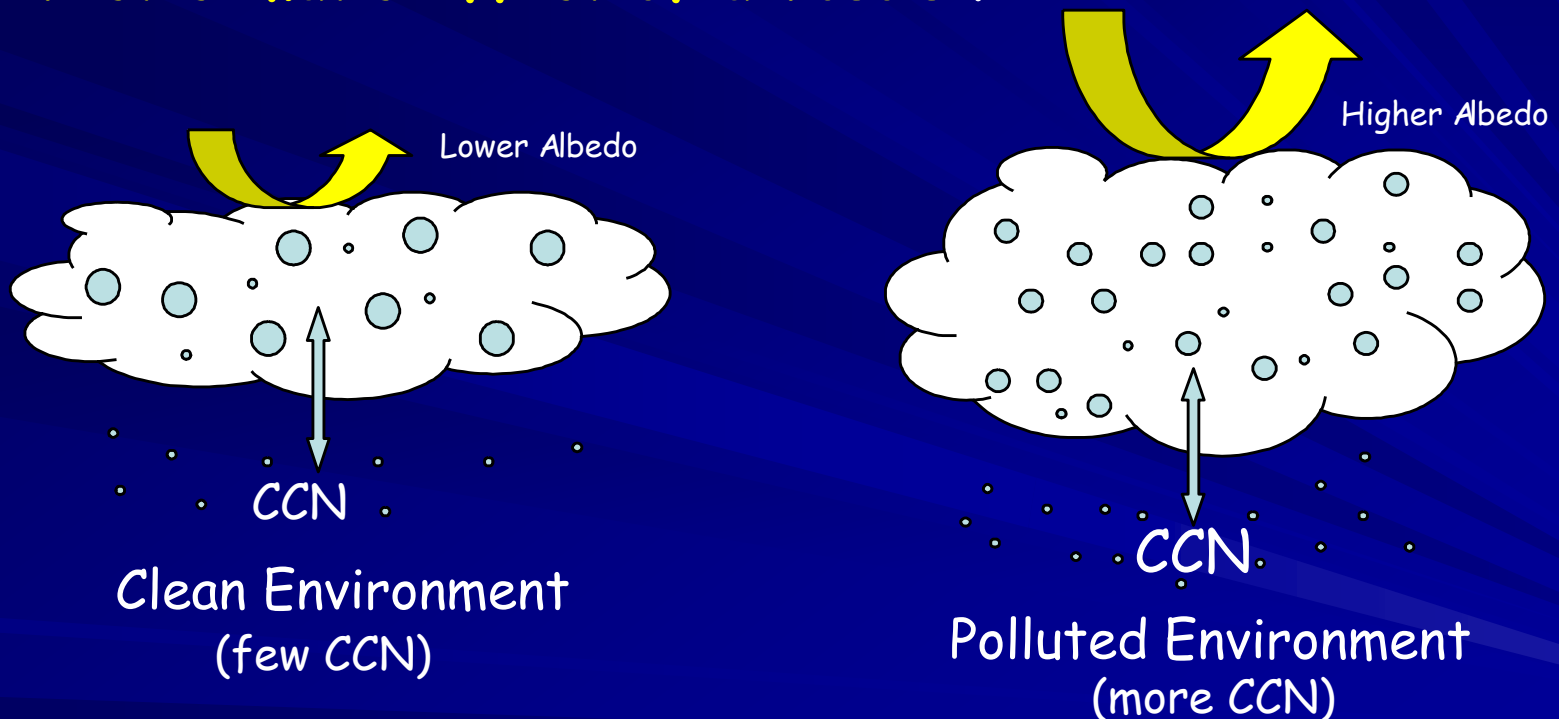
Cloud droplets nucleate on pre-existing particles found in the atmosphere (aerosols) with $\sim 0.1\mu\text{m}$ diameter.

Aerosols that can become droplets are called cloud condensation nuclei (CCN).



Increases in aerosol affects warm clouds

You make clouds that are "whiter", precipitate less (persist longer) and potentially cover larger areas of the globe. This is thought to yield a net cooling on climate and is termed as the **"indirect climatic effect of aerosols"**.



Increasing particles tends to cool climate (potentially alot).
Quantitative assessments done with climate models.

Cloud condensation nuclei (CCN)

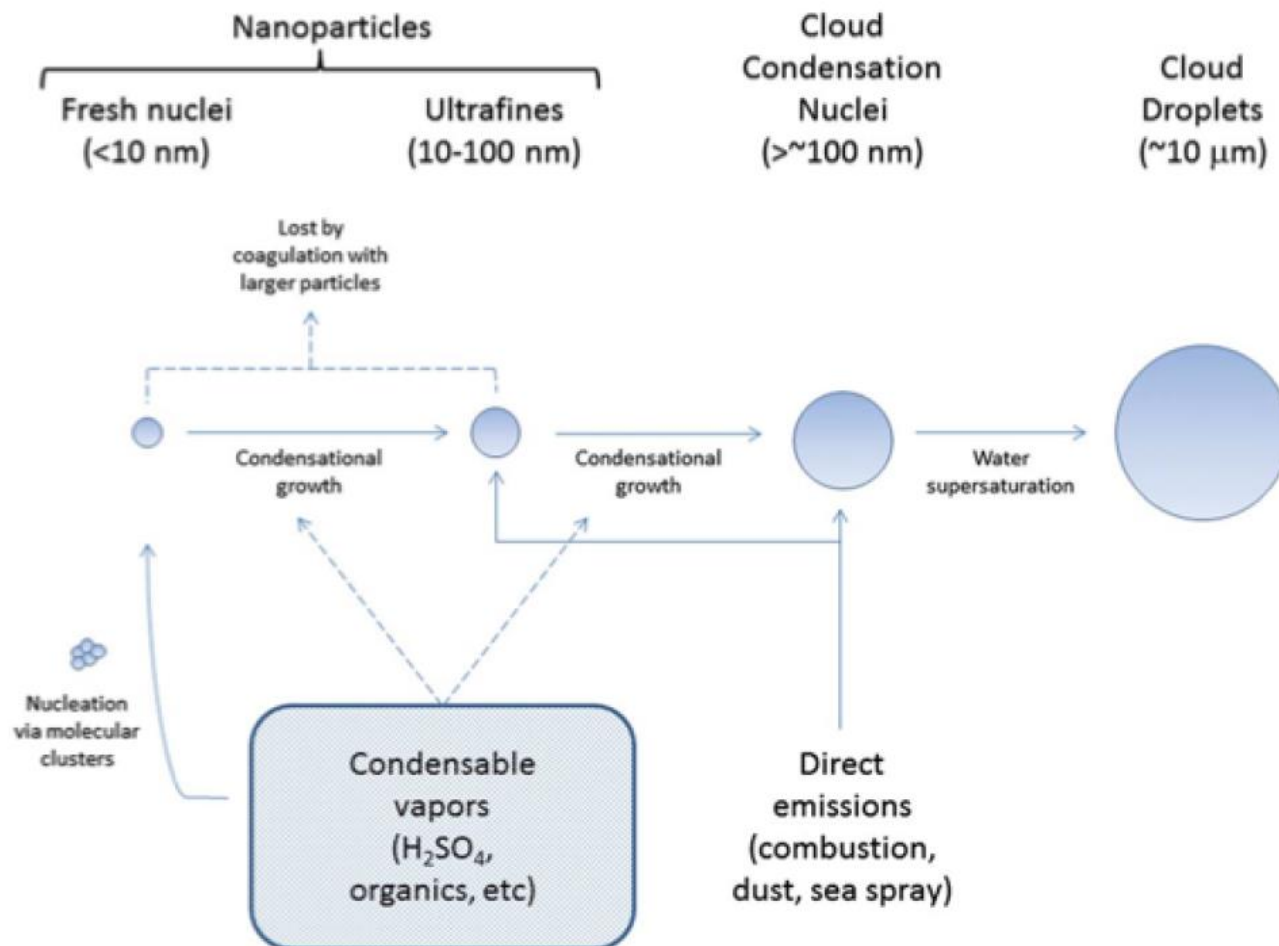


Figure 2. A schematic of processes that form cloud condensation nuclei (CCN).

Particles may either be directly emitted (primary) or nucleated in the atmosphere itself (secondary). Nucleated particles (1–2 nm dia.) must grow substantially via condensation of sulfuric acid and organics to approximately ~100 nm dia. before they can nucleate cloud droplets. Along the way, many of the nucleated particles are lost by coagulating with pre-existing, larger particles.

Adams et al., 2013

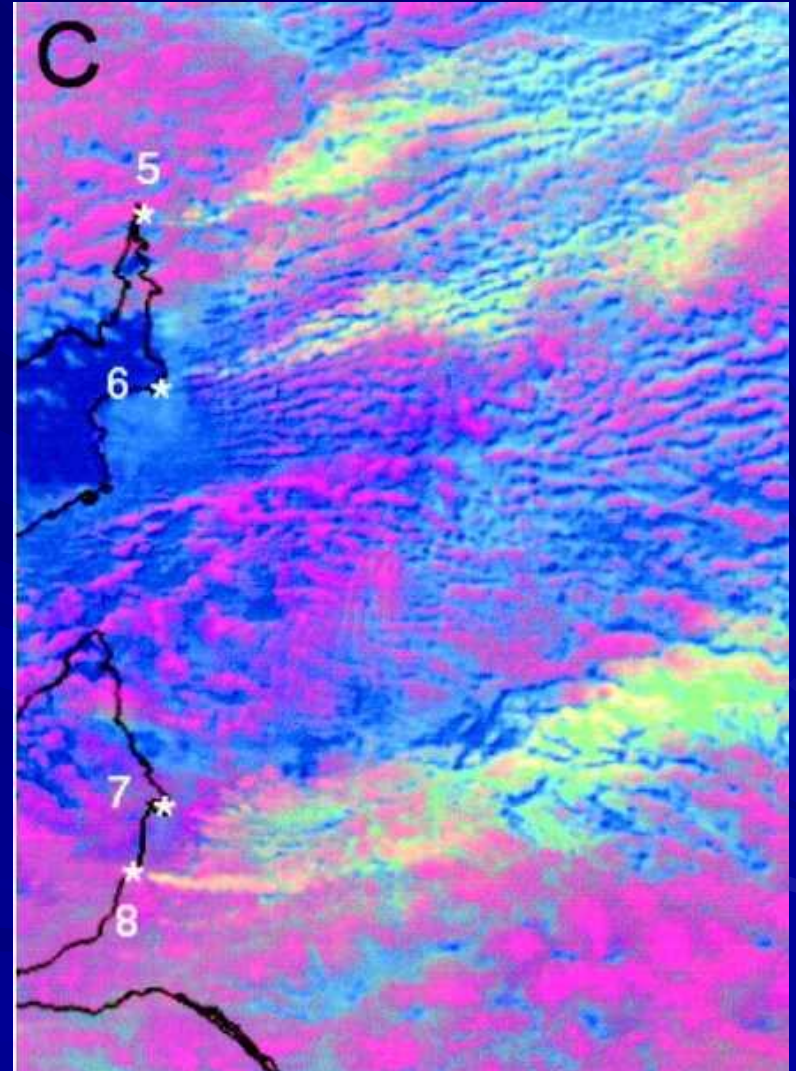
Observational evidence of indirect effect

Satellite observations of clouds off W. Australia.

Red: Clouds with low reflectivity.

White: Clouds that reflect a lot.

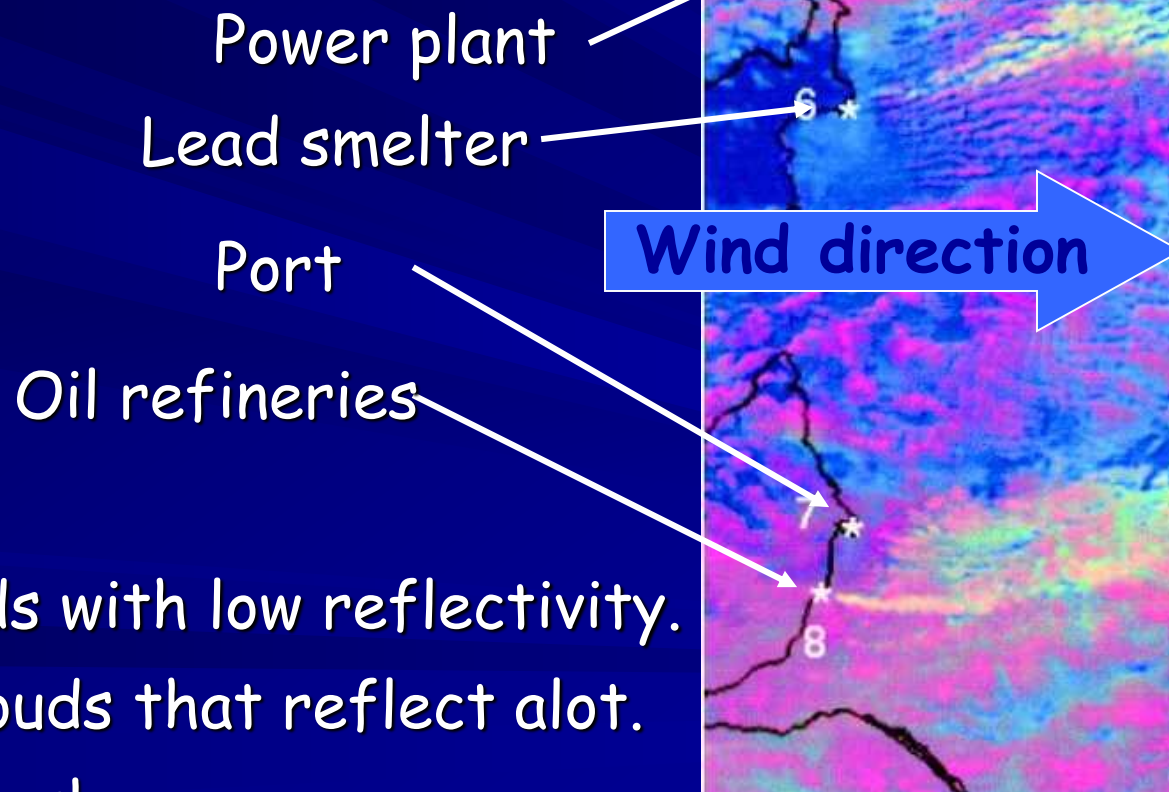
Blue: Clear sky.



Observational evidence of indirect effect

Air pollution can affect cloud properties

Satellite observations of clouds off W. Australia.



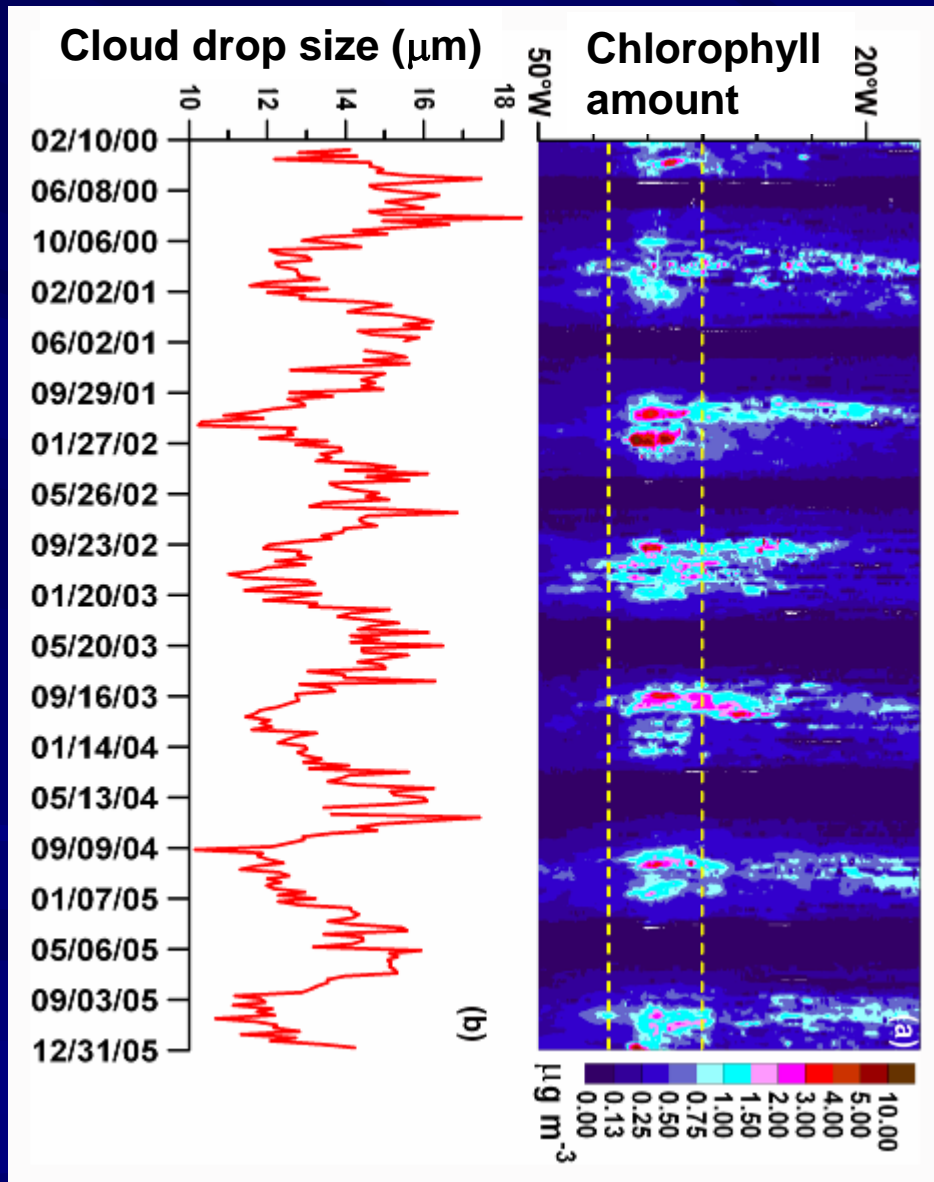
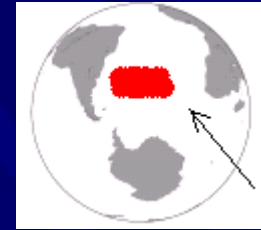
Red: Clouds with low reflectivity.

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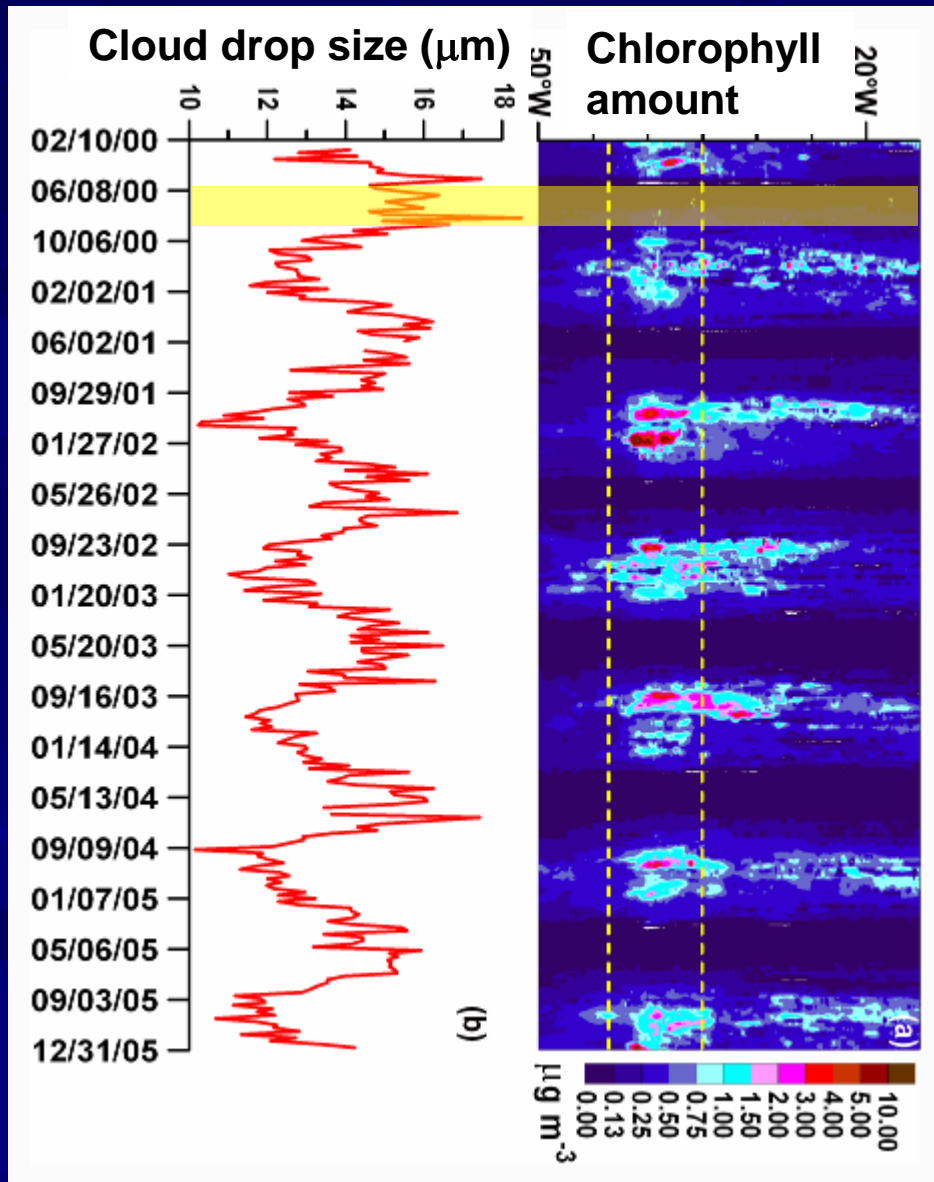
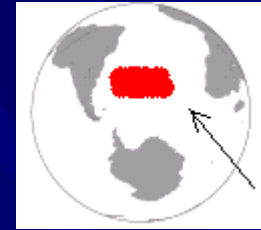
Phytoplankton affect clouds too...

Location: East of Patagonia (South America)



Phytoplankton affect clouds too...

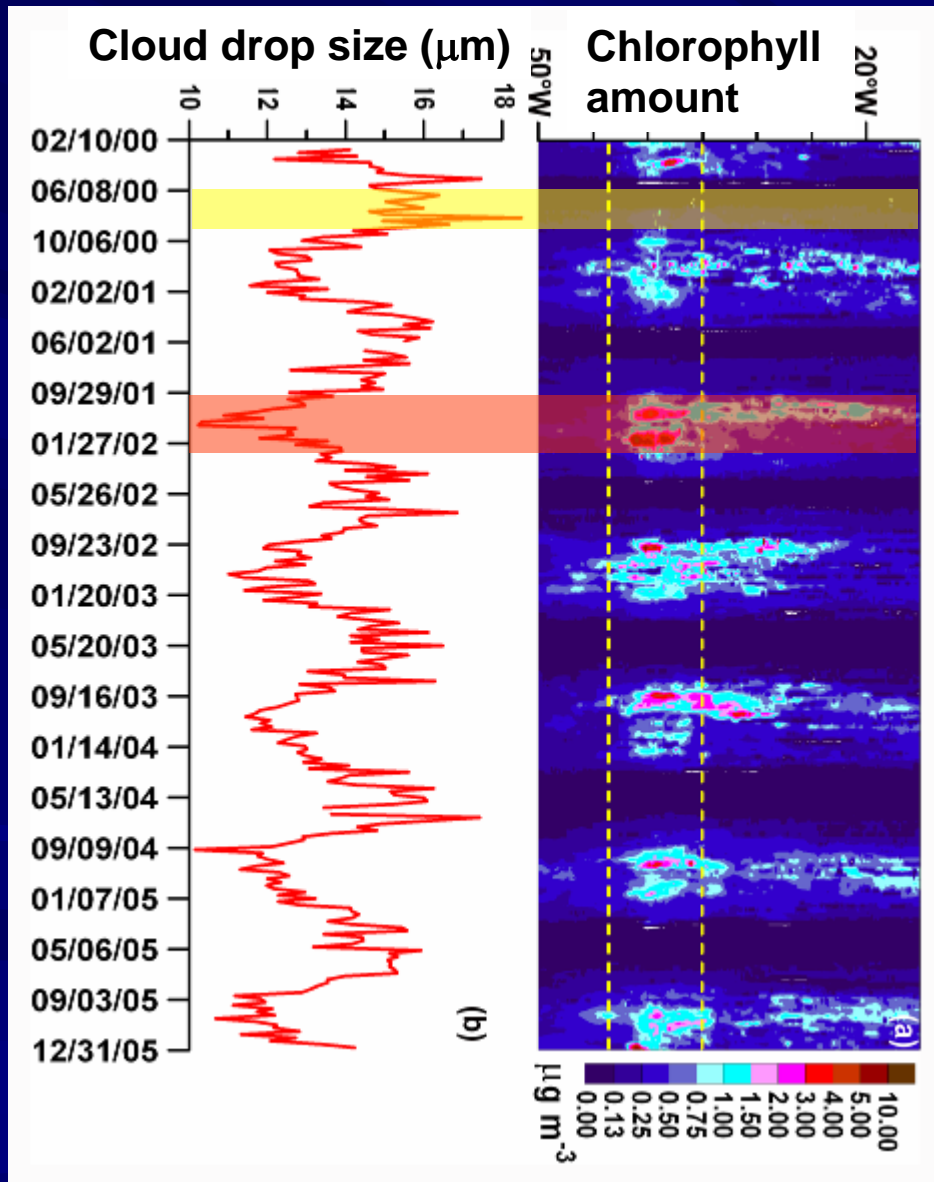
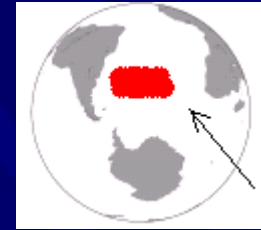
Location: East of Patagonia (South America)



Low chlorophyll period,
clouds have large drops
(not very reflective)

Phytoplankton affect clouds too...

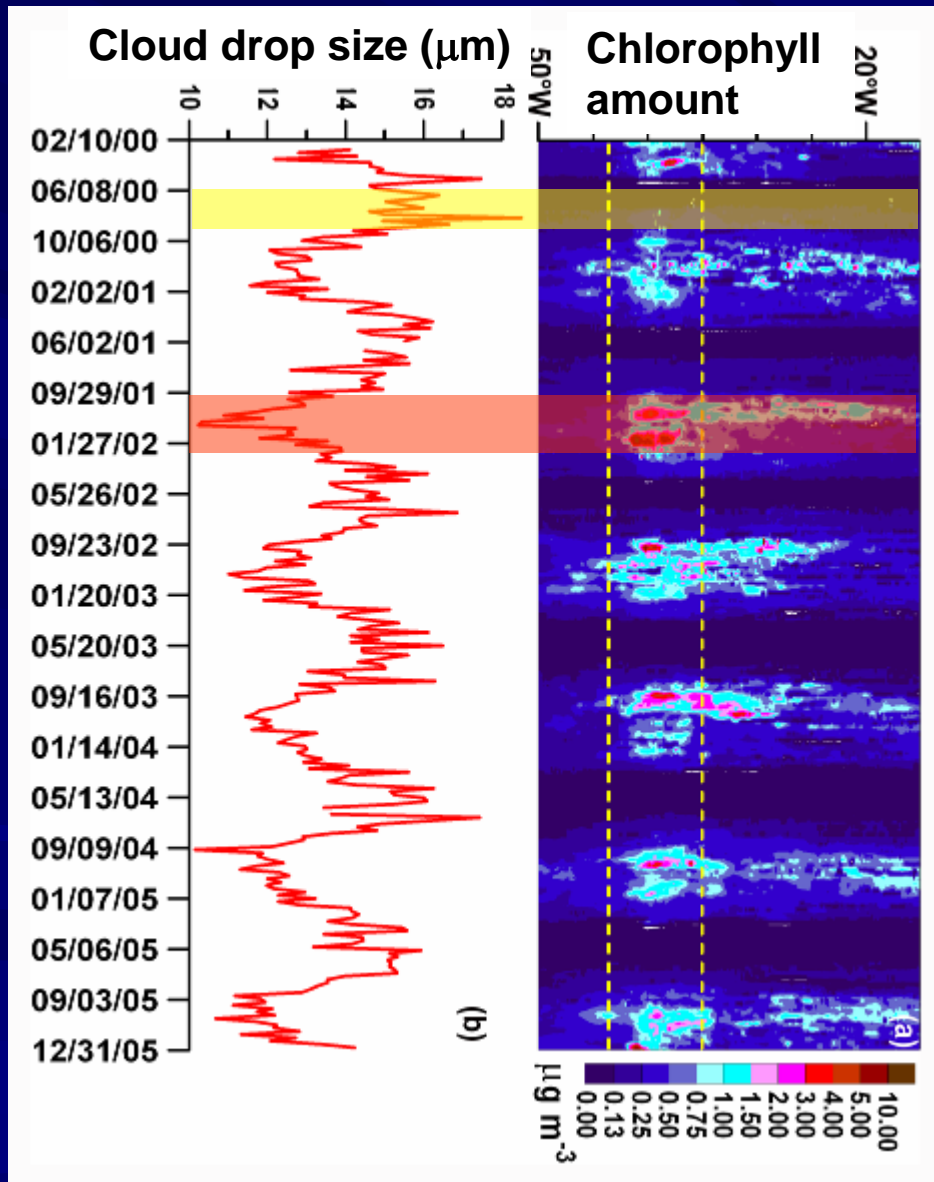
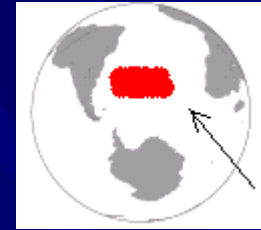
Location: East of Patagonia (South America)



- Low chlorophyll period, clouds have large drops (not very reflective)
- High Chlorophyll period, Clouds have small drops (very reflective)

Phytoplankton affect clouds too...

Location: East of Patagonia (South America)



← Low chlorophyll period,
clouds have large drops
(not very reflective)

← High Chlorophyll period,
Clouds have small drops
(very reflective)

Phytoplankton emissions
increase particle loads, and
strongly impact clouds.

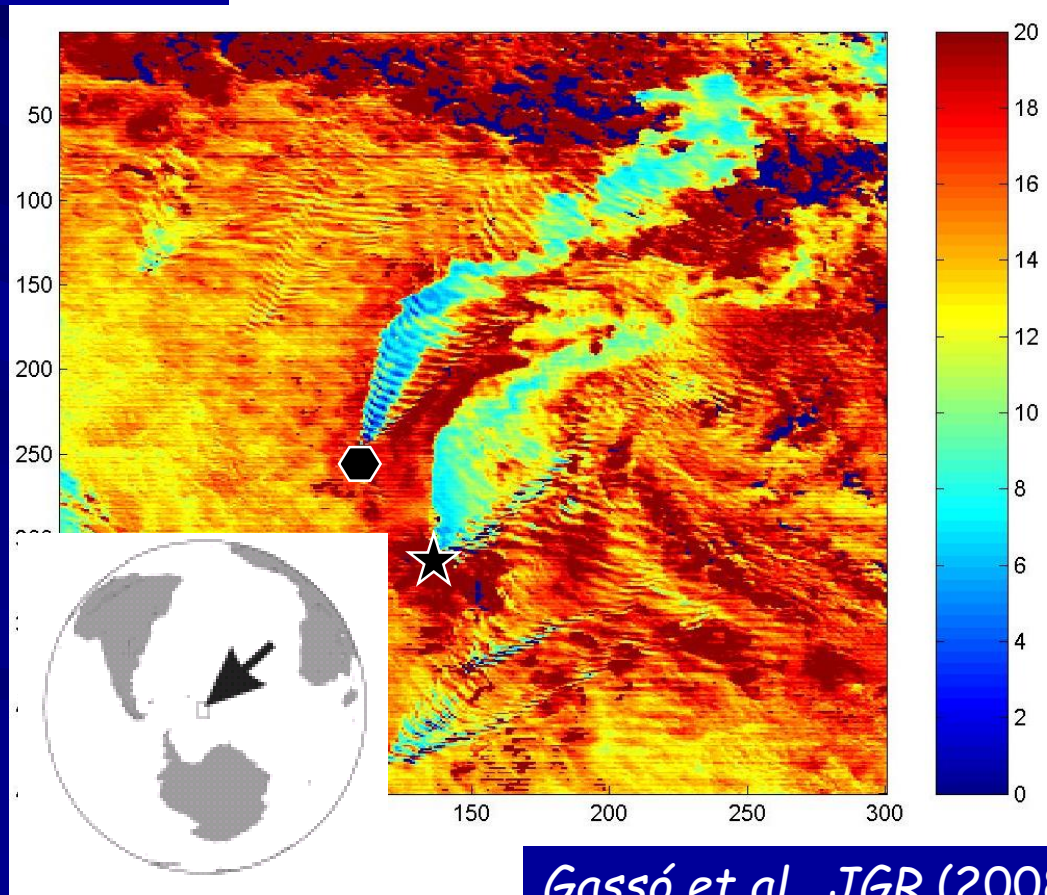
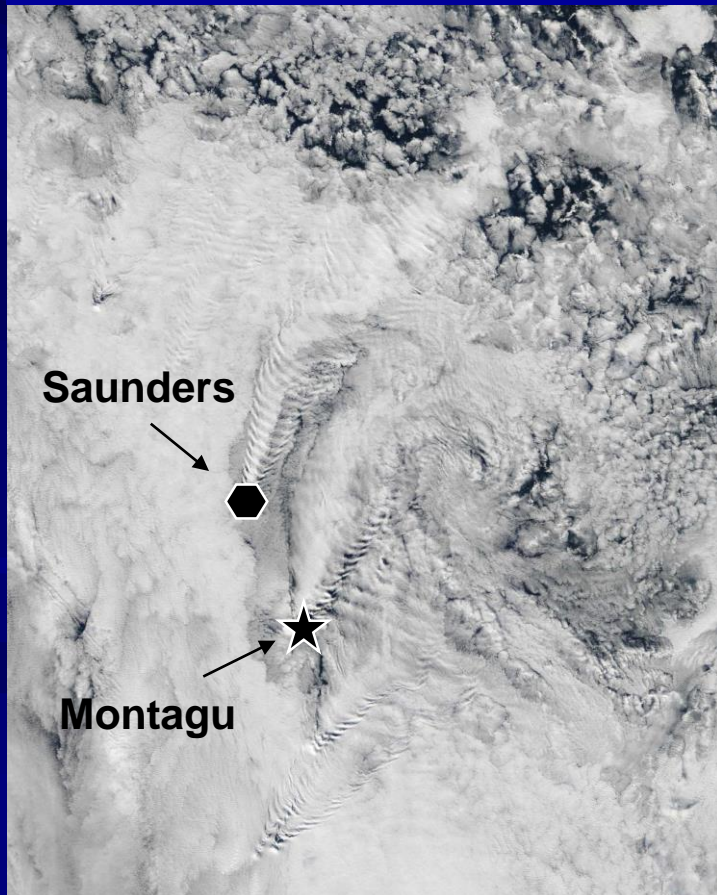
Biology-cloud interactions
affect radiation in the region.

Meskhidze and Nenes, Science, 2006

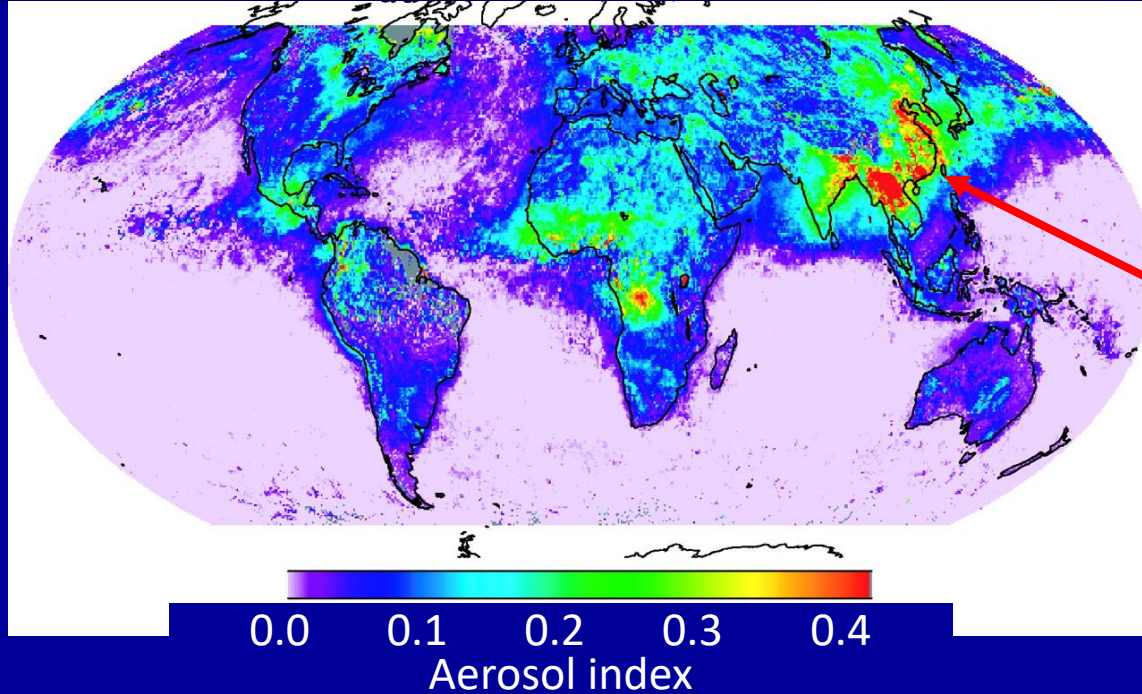
So do volcanoes (even when “sleeping”) ...

Volcanoes continuously emit SO_2 which becomes sulfate aerosol. The aerosol can substantially increase CCN in volcanic plumes. Clouds in the plume are much more reflective than outside.

Location: Sandwich Islands, $\sim 55^\circ\text{S}$, $\sim 30^\circ\text{W}$



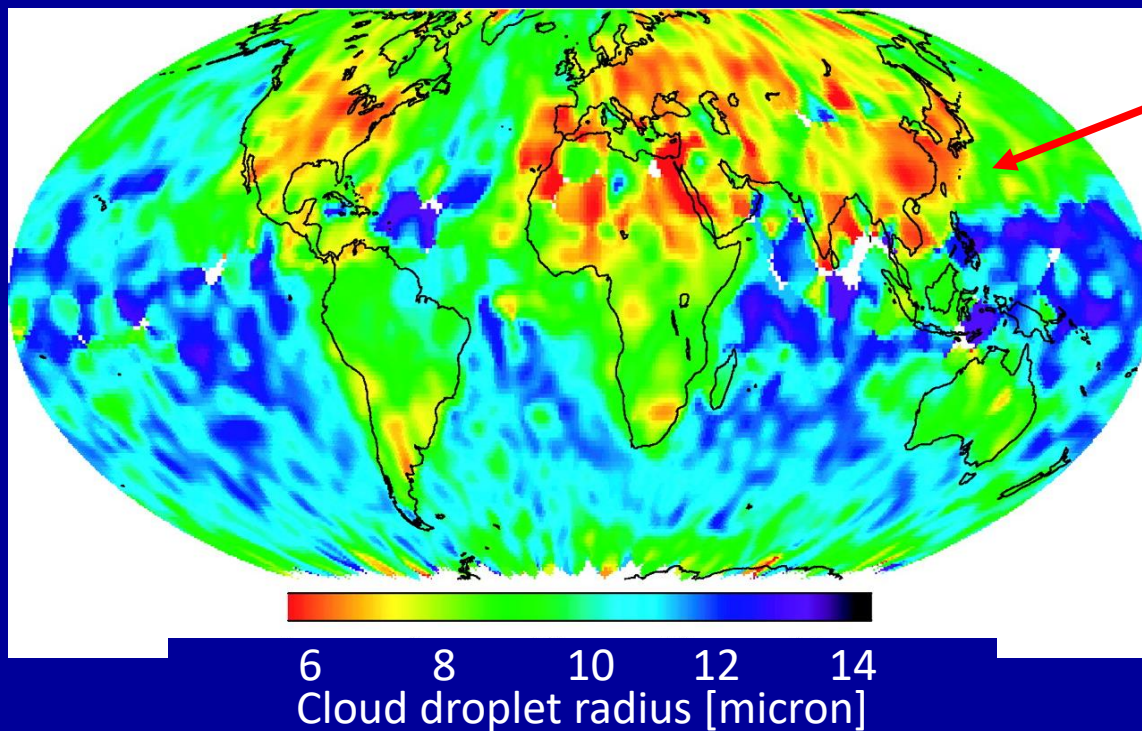
Gassó et al., JGR (2008)



A remote sensing
global picture...

A lot of aerosol...

...gives smallest
cloud droplets

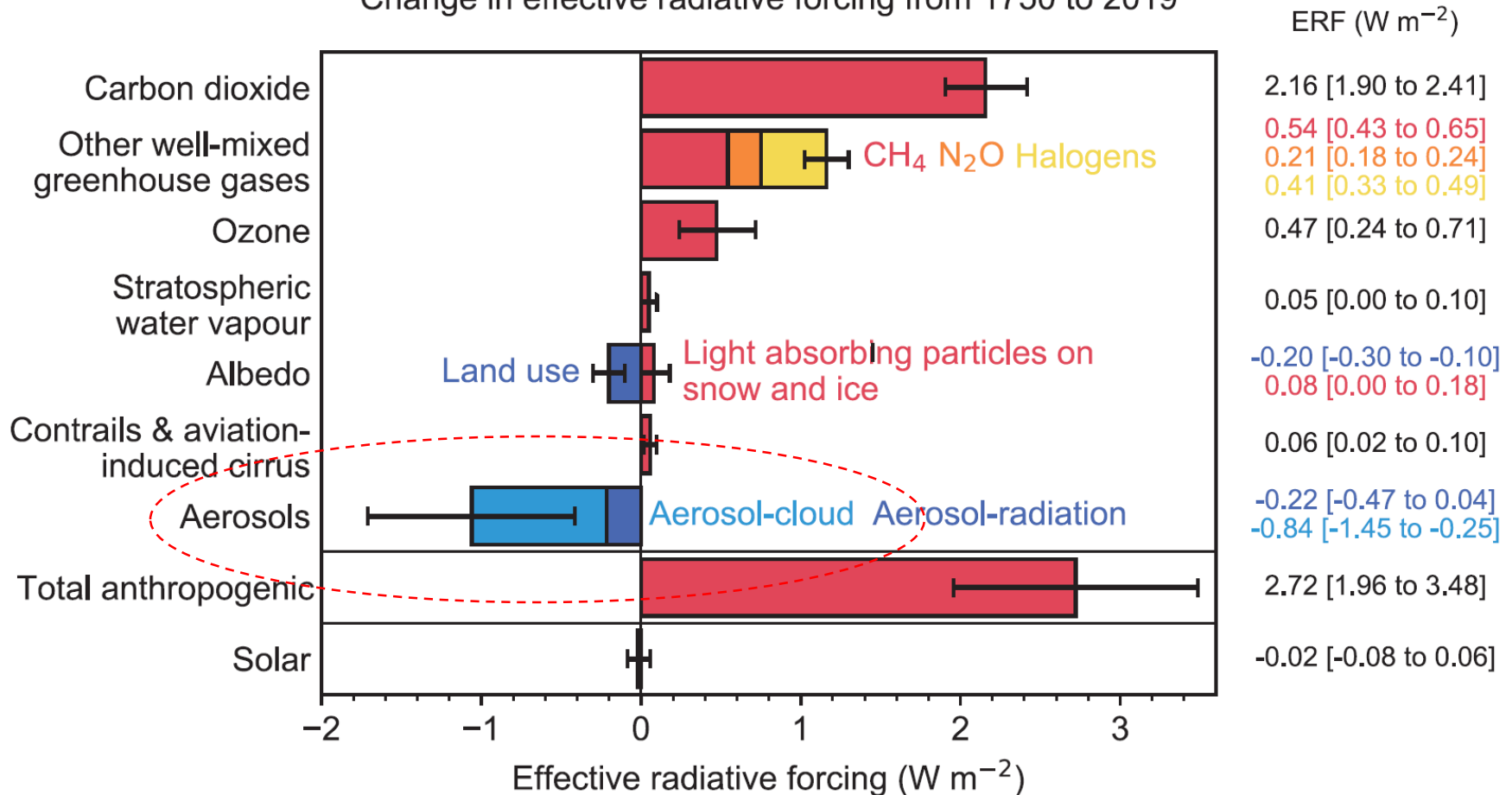


We see the same
on all satellite
platforms...

Breón et al. (2002)

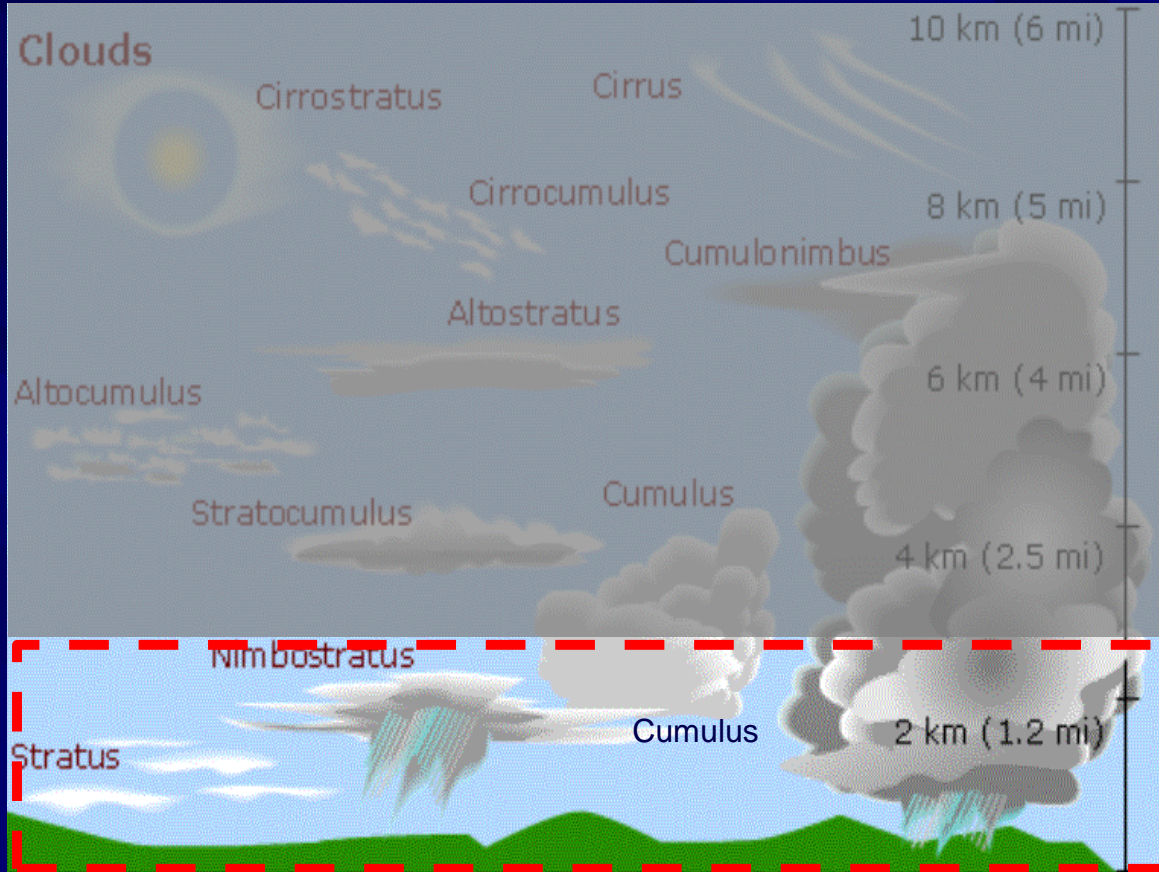
Aerosol-cloud interactions are important for climate - but highly uncertain

Change in effective radiative forcing from 1750 to 2019



Forster et al. (2021), IPCC AR6

LIQUID cloud microphysics



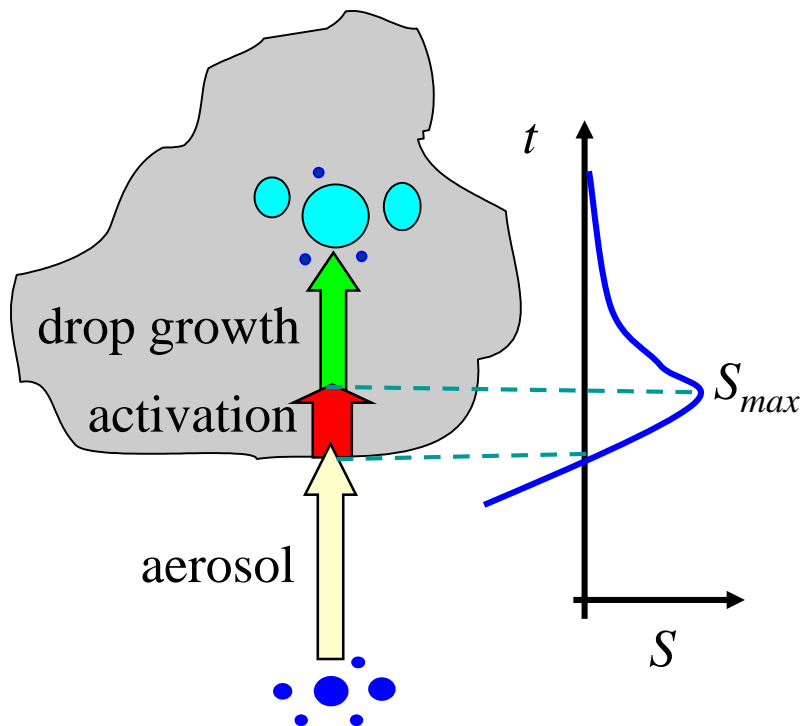
- **Ice (cold) clouds:**
Made of ice crystals at $T < 235$ K.
- **Mixed Phase clouds:**
Mixture of liquid droplets and ice for T between 235 and 273K
- **Liquid (warm) clouds:**
Made of liquid droplets at $T > 273$ K

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

Droplet formation: The essence

Goal: Link cloud droplet concentration with precursor aerosol

Approach: Use the “simple story of cloud formation”.

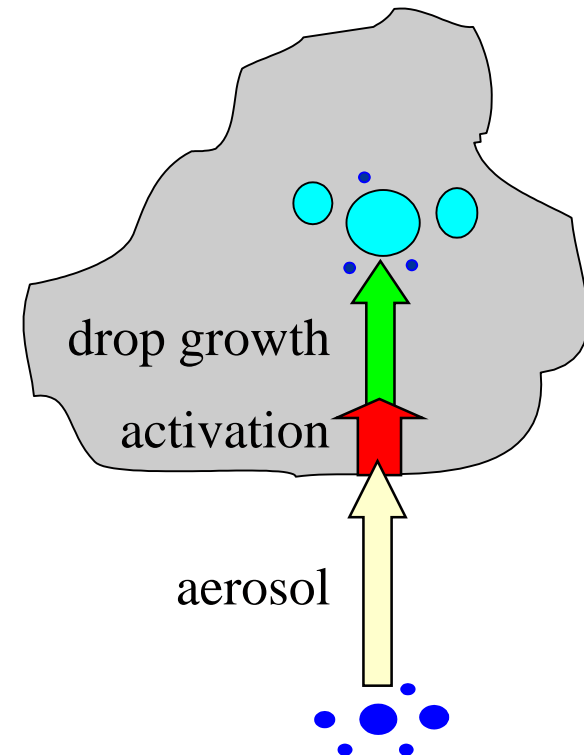
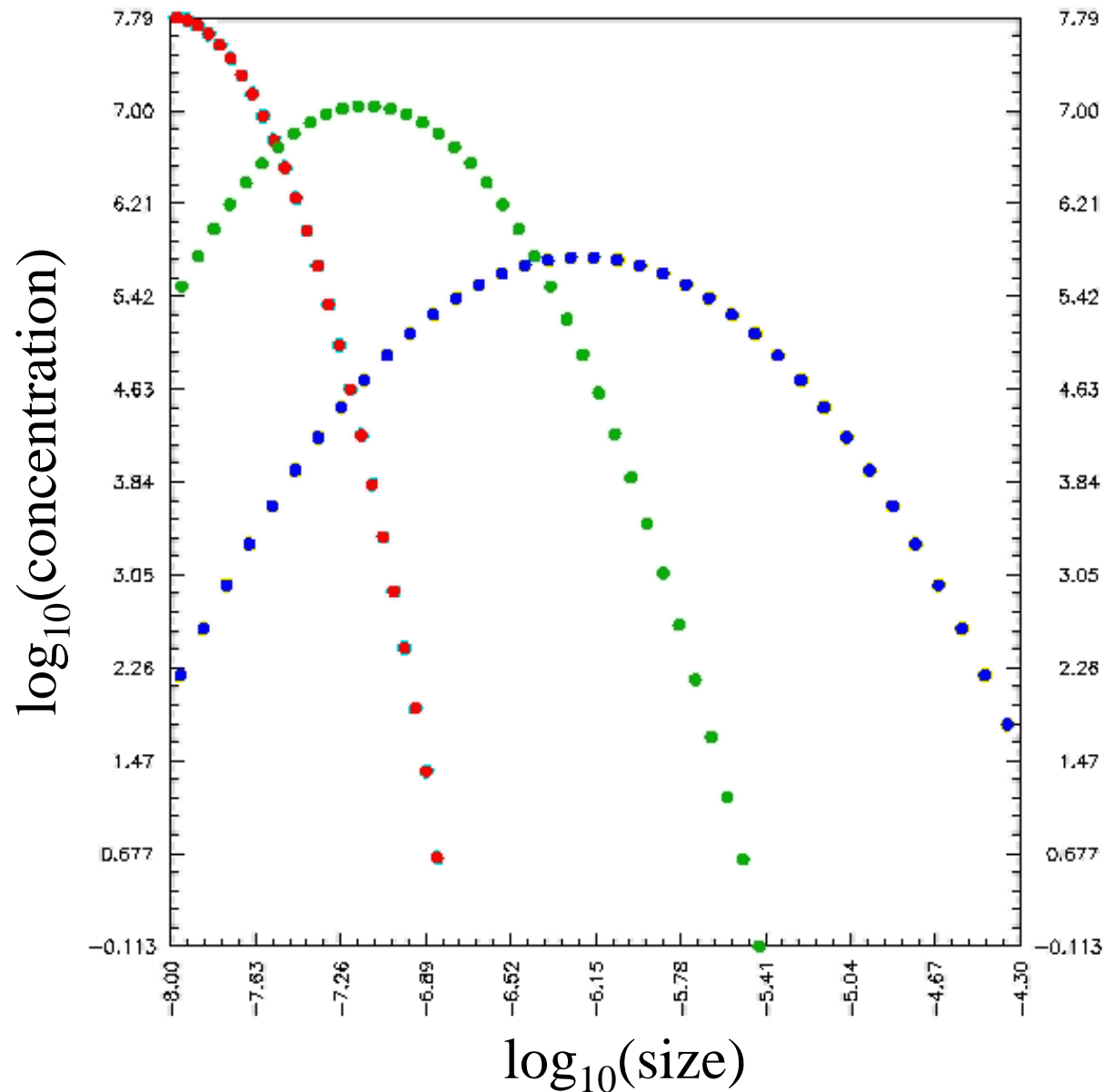


Conceptual steps are:

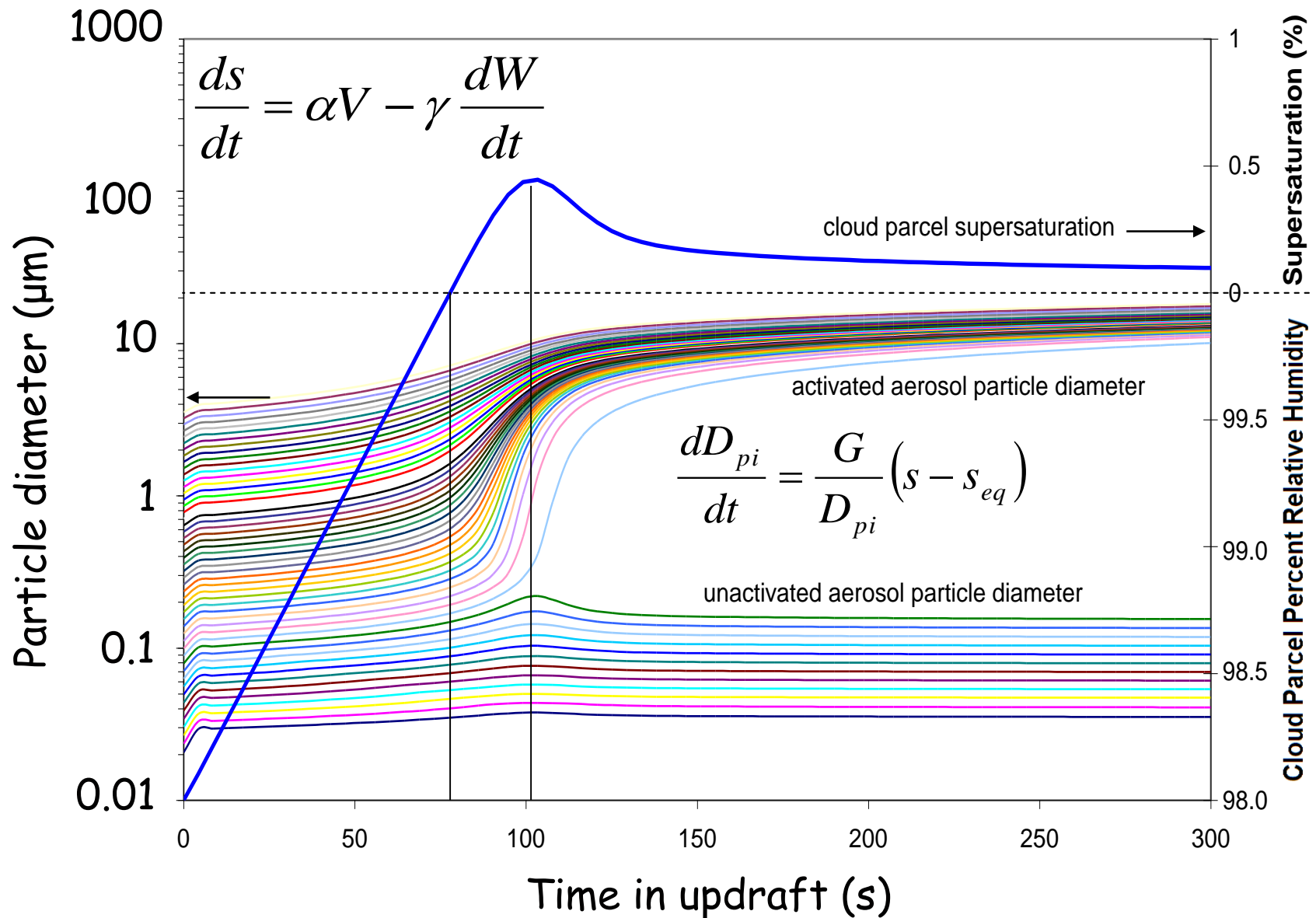
- Air parcel cools, reaches 100% humidity.
- Water vapor is **supersaturated**
- Droplets start forming on existing CCN.
- Condensation of water on droplets becomes intense.
- Humidity reaches a **maximum**
- No more additional drops form

A “classical” nucleation/growth problem

Simulation of cloud droplet formation



Simulation of cloud droplet formation



Drop formation: coupled nonlinear system

Supersaturation (and s_{max}) depends strongly on the expansion rate (updraft velocity V) and the condensation rate of water:

$$\frac{ds}{dt} = \alpha V - \gamma \left(\frac{dW}{dt} \right) \quad \leftarrow \text{Water condensation rate on droplets}$$

Water condensation rate on droplets determined from the contribution of each aerosol "size class" i

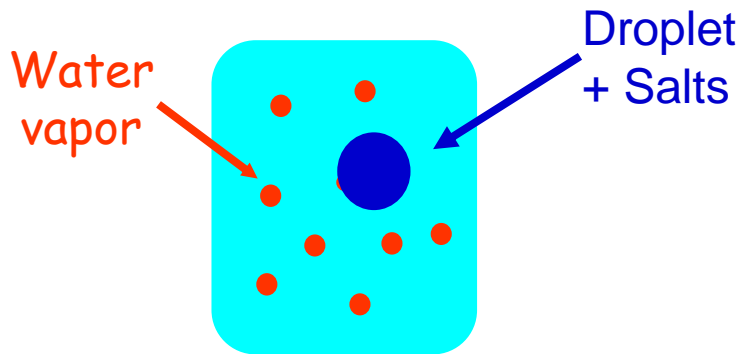
$$\frac{dD_{p,i}}{dt} = \frac{G_i}{D_{p,i}} \left(s - s_{eq,i} \right) \quad \begin{array}{l} \leftarrow \text{Mass transfer coefficient of water from the gas phase} \\ \leftarrow \text{Equilibrium saturation (relative humidity) of droplet} \end{array}$$

We need to know:

1. $S_{eq,i}$ for aerosol and droplets (**thermodynamics**)
2. G_i depends on the water vapor diffusivity (P, T) and the water vapor mass transfer coefficient (**kinetics**).

Thermodynamics 101: essentials

- Equilibrium vapor pressure of water over pure water



$$P_{H_2O(g)} = P_{sat}(T)$$

- Effects of dissolved solutes

Dissolved salts decrease the energy of your system (why?).

This reduces the equilibrium $P_{H_2O(g)} < P_{sat}(T)$

Mol fraction of water in solution $\frac{n_w}{n_w + n_{salts}}$

$$P_{H_2O(g)} = P_{sat}(T) x_{H_2O(l)}$$

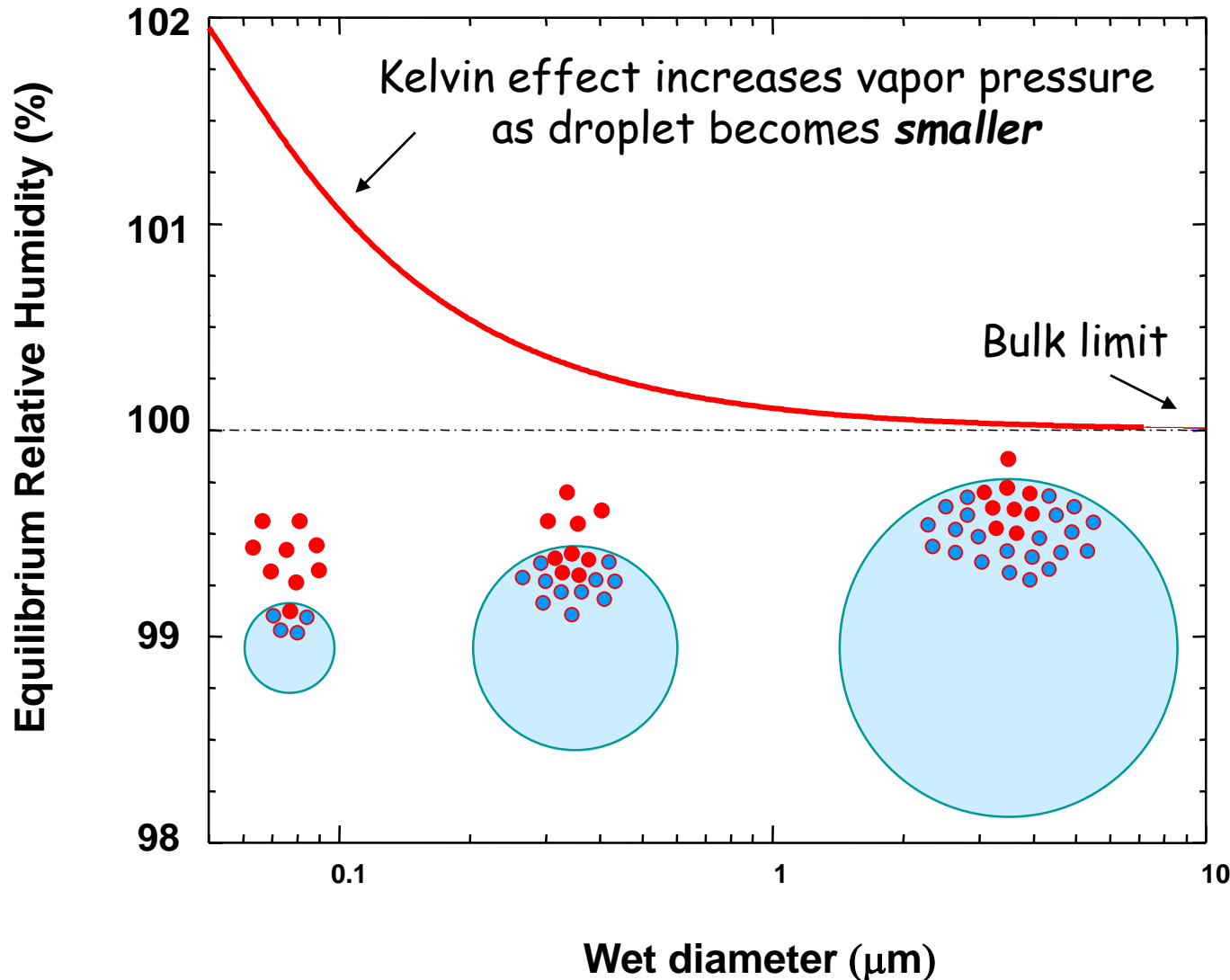
Raoult's Law (for ideal solutions)

Droplet thermo: special considerations

- “Bulk” thermodynamics (like described before) assume that interfaces are “flat”.
- Sometimes this is not a good approximation.
- “Curvature” effects may need to be included in the thermodynamic expressions.
- Main parameters expression curvature effects:
 - Interfacial tension (“surface tension”)
 - Radius of curvature (most often, aerosol/drop radius)

Including curvature: Thermodynamics of droplets

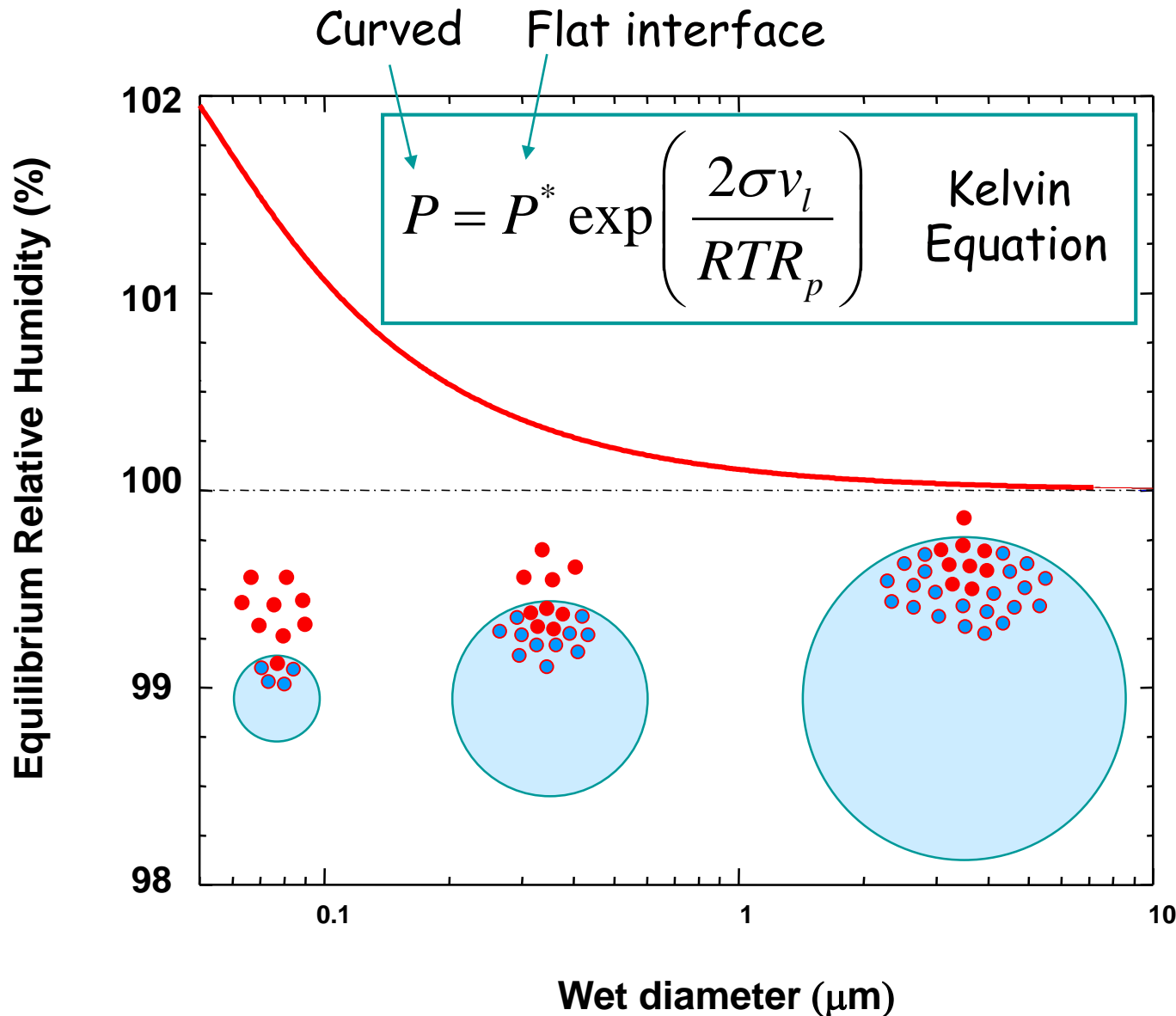
Impact of curvature: "the physical explanation"



As the droplet size decreases, its equilibrium vapor pressure increases ("Kelvin effect")

Less molecules around in small drops to "keep" H_2O in the droplet phase

Including curvature: Thermodynamics of droplets



As the droplet size decreases, its equilibrium vapor pressure increases ("Kelvin effect")

Hugely important equation.

Needed for any nucleation process

Thermodynamics of droplets: Köhler equation

Apply Kelvin equation to a pure water droplet (i.e., σ_w and $v_l = \frac{M_w}{\rho_w}$)

$$P = P^* \exp\left(\frac{4M_w\sigma}{RT\rho_w D_p}\right)$$

Dissolved substances in the drop depress water vapor pressure. If $\sigma_w, v_l \sim \text{const.}$ then only P^* changes (given by Raoult's law: $P^* = P^{sat} \gamma_w x_w$)

$$\frac{P}{P^{sat}} = x_w \gamma_w \exp\left(\frac{4M_w\sigma}{RT\rho_w D_p}\right) \quad \text{Köhler Equation}$$

Equilibrium relative humidity of a particle when it has absorbed water and acquired a wet diameter, D_p

Thermodynamics of droplets: Köhler equation

One can then invoke simplifying assumptions:

$$x_w = \frac{n_w}{n_w + in_s} = 1 - \frac{in_s}{n_w + in_s} \sim 1 - \frac{in_s}{n_w} = 1 - \frac{in_s}{\frac{\pi}{6} D_p^3 \frac{\rho_w}{M_w}} = 1 - \frac{6 M_w in_s}{\pi \rho_w D_p^3}$$

$$= 1 - \frac{B}{D_p^3} \quad \text{where} \quad B = \frac{6 M_w}{\pi \rho_w} in_s$$

Moles of solute in droplet
van't Hoff factor of solute in droplet

$$\gamma_w \sim 1 \quad \text{and} \quad \exp\left(\frac{4M_w\sigma}{RT\rho_w D_p}\right) \sim 1 + \frac{A}{D_p} \quad \text{where} \quad A = \frac{4M_w\sigma}{RT\rho_w}$$

Substitution into full Köhler equation, and considering leading terms:

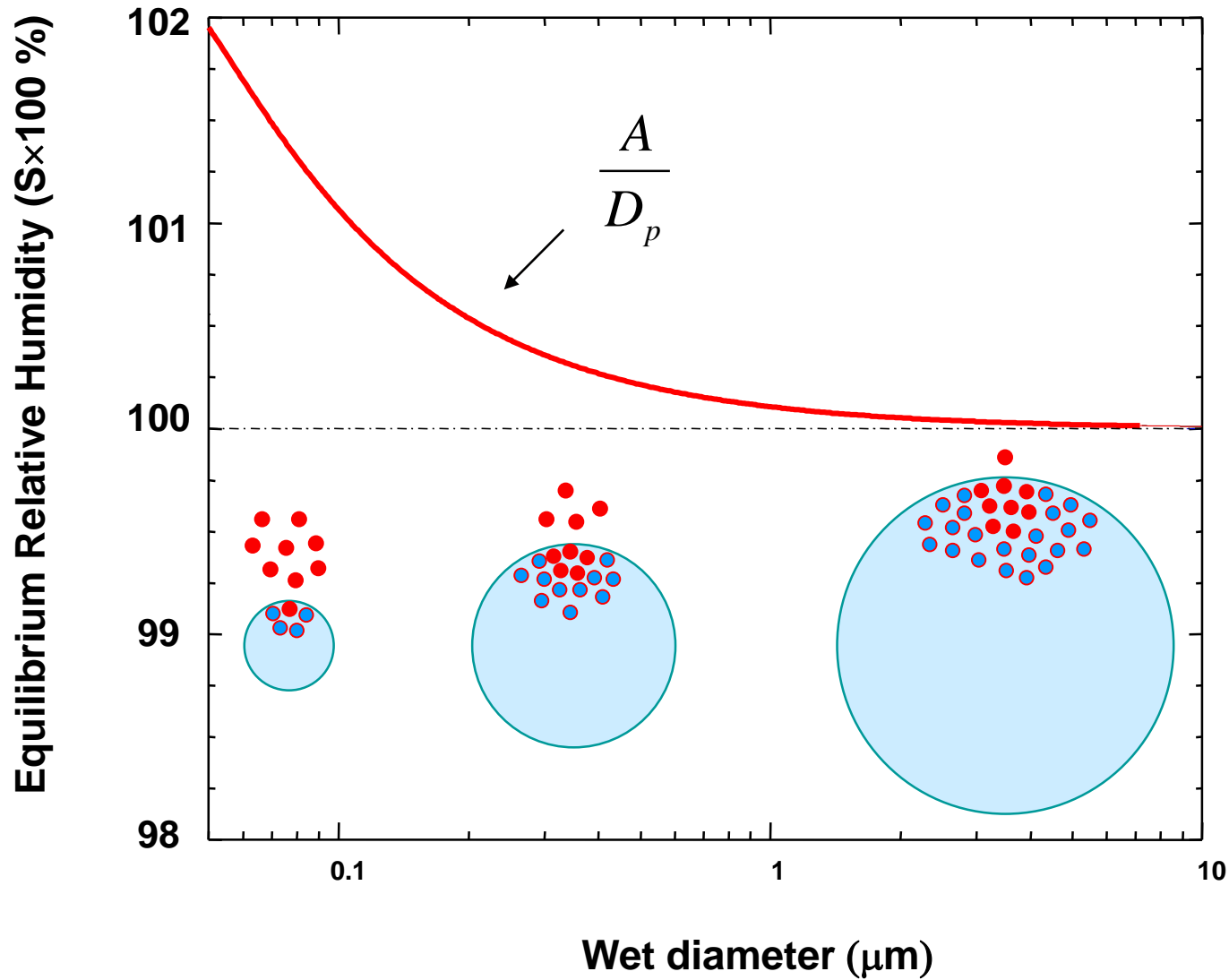
$$S = \frac{P}{P^{sat}} = 1 + \frac{A}{D_p} - \frac{B}{D_p^3}$$

Simplified Köhler equation

↑ Saturation ratio
↑ "Kelvin" term
↑ "Raoult" term

Thermodynamics of droplets: Köhler equation

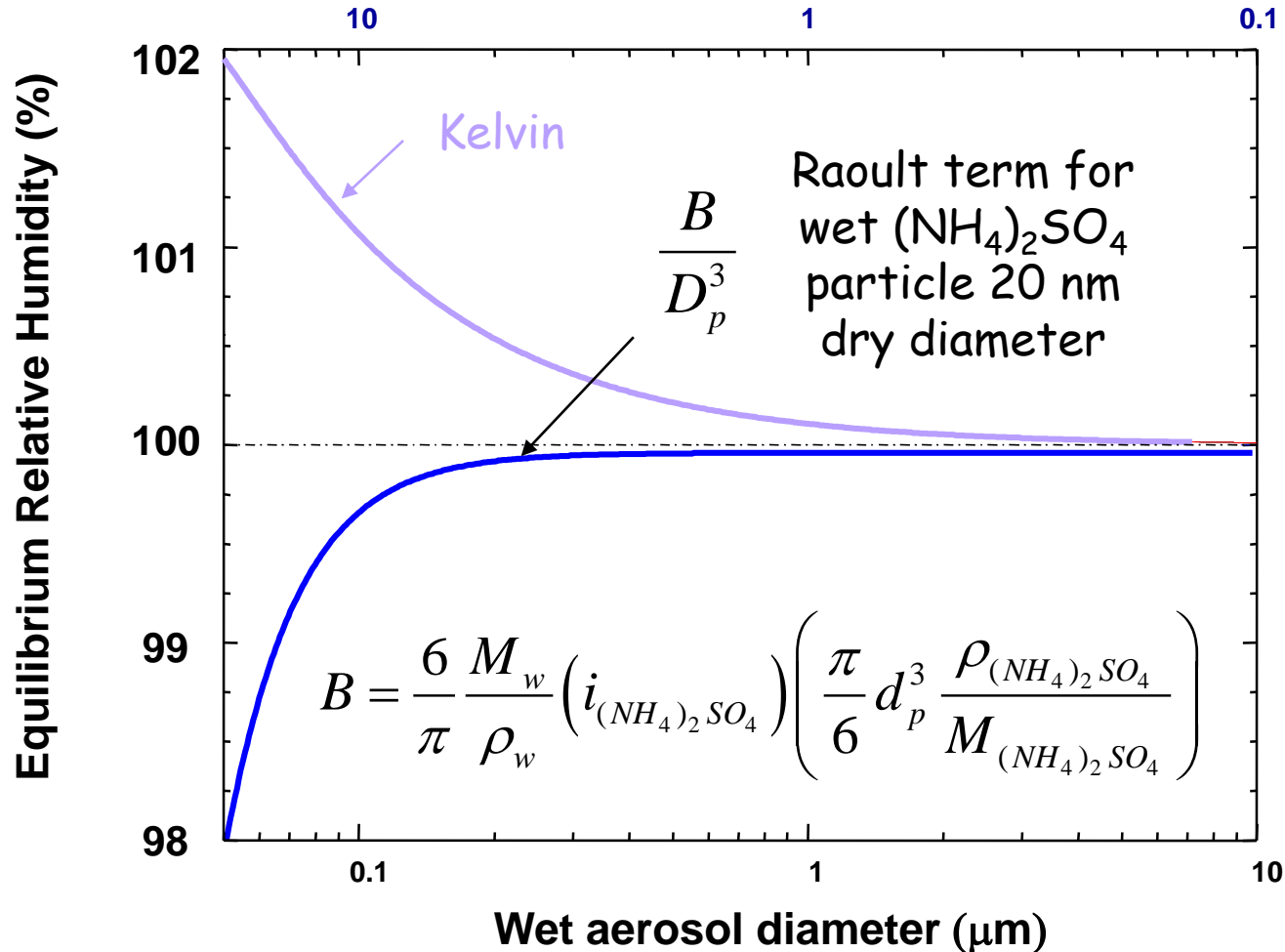
First plot the Kelvin term



Thermodynamics of droplets: Köhler equation

...then plot the Raoult term

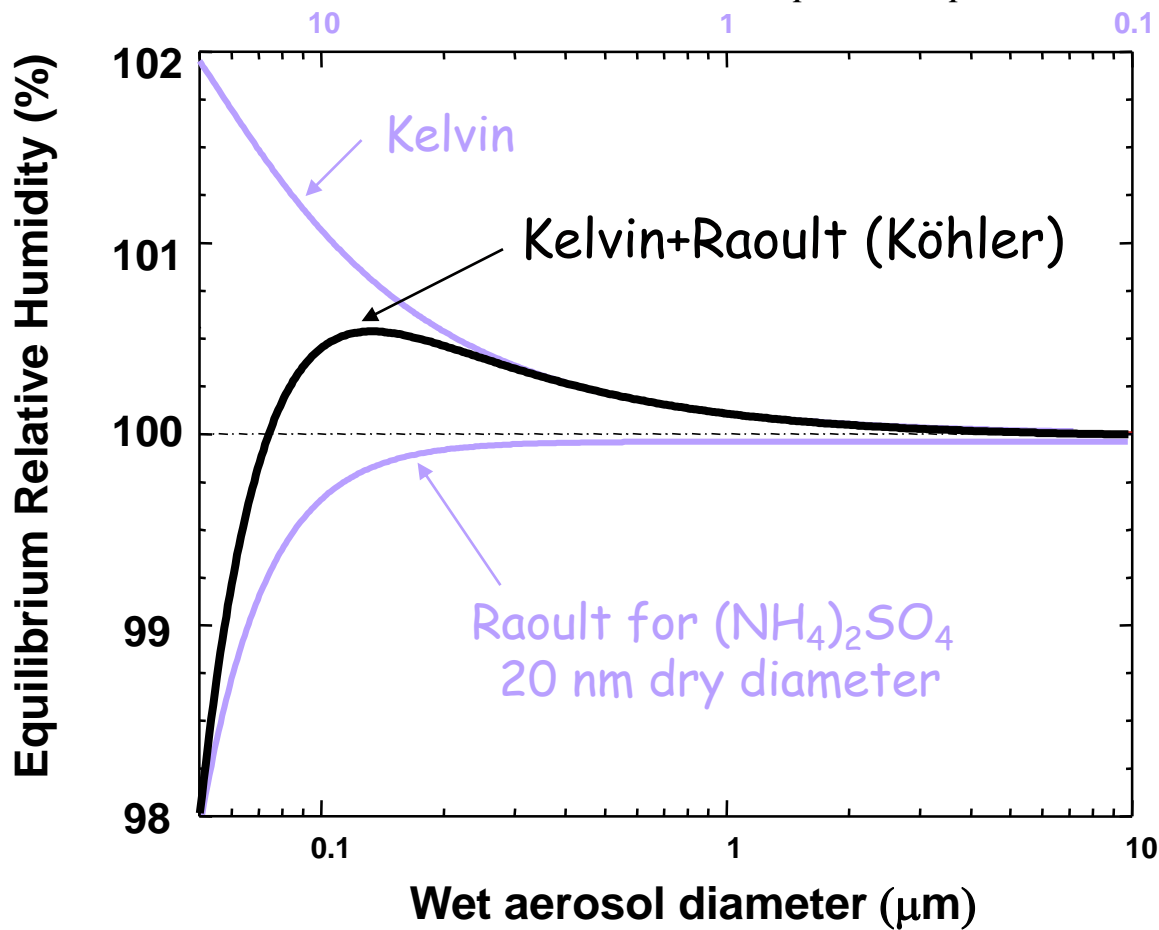
Solute Concentration (M)



Thermodynamics of droplets: Köhler equation

Both effects together: equilibrium vapor pressure of a wet aerosol.

$$S = \frac{P}{P^{sat}} = 1 + \frac{A}{D_p} - \frac{B}{D_p^3}$$



The combined Kelvin and Raoult effects is the simplified **Köhler equation**.

You can be in equilibrium **even if** you are **above saturation**.

Indicative Equilibrium (Kohler) curves for particles

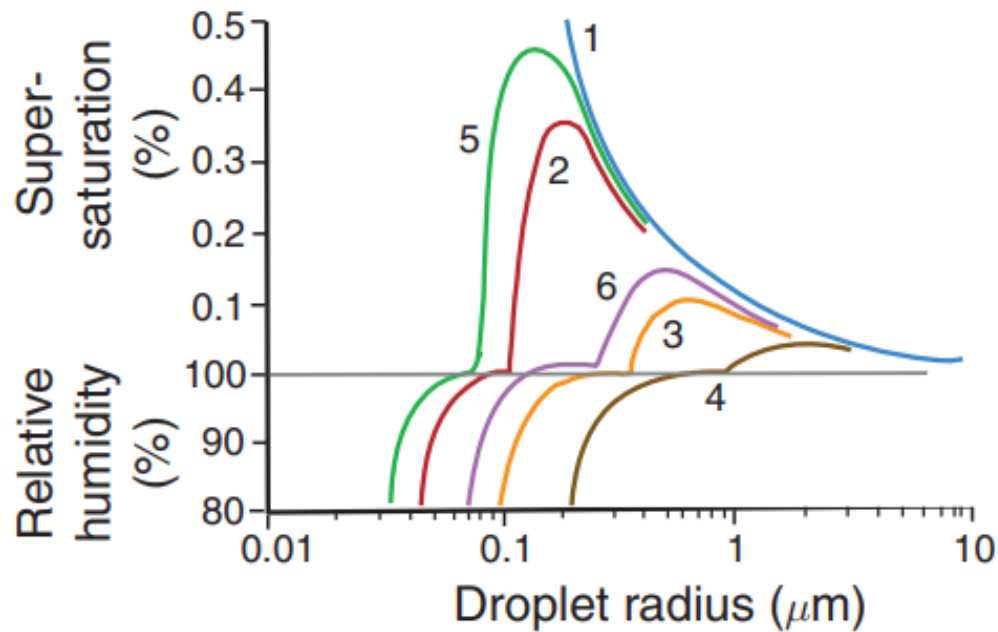


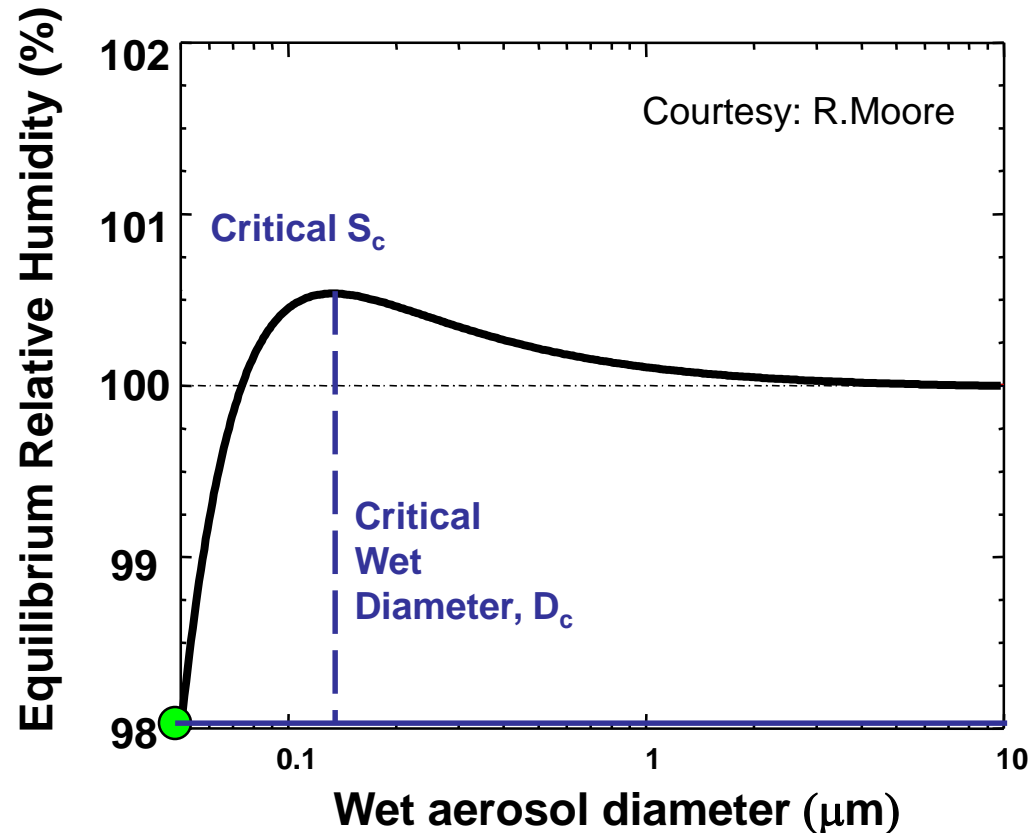
Fig. 6.3 Variations of the relative humidity and supersaturation adjacent to droplets of (1) pure water (blue) and adjacent to solution droplets containing the following fixed masses of salt: (2) 10^{-19} kg of NaCl, (3) 10^{-18} kg of NaCl, (4) 10^{-17} kg of NaCl (5) 10^{-19} kg of $(\text{NH}_4)_2\text{SO}_4$, and (6) 10^{-18} kg of $(\text{NH}_4)_2\text{SO}_4$. Note the discontinuity in the ordinate at 100% relative humidity.

Regions of stability/instability of ambient droplets

Dynamical behavior of an aerosol particle in a variable RH environment.

Wet Aerosol
(Haze)

Cloud Droplet



If ambient S exceeds the maximum, particles grow uncontrollably.
They are said then to act as Cloud Condensation Nuclei (CCN)

Stability analysis of particles in humid environments

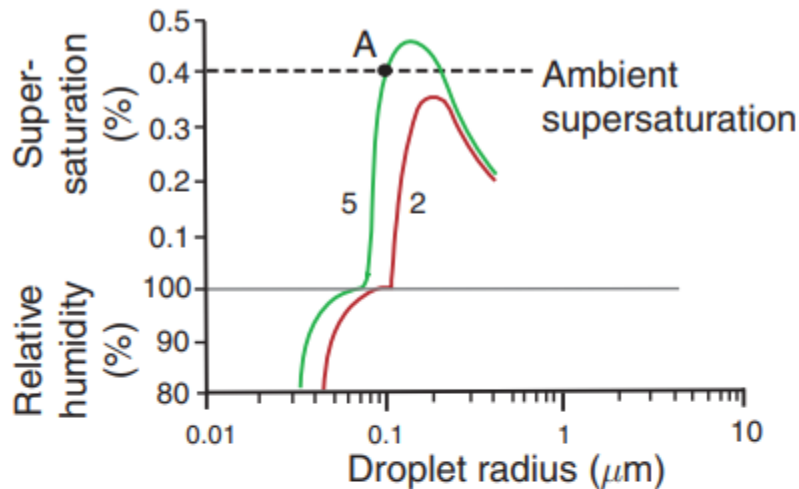


Fig. 6.4 Köhler curves 2 and 5 from Fig. 6.3.

Curve 2 is for a solution droplet containing 10^{-19} kg of NaCl, and curve 5 is for a solution droplet containing 10^{-19} kg of $(\text{NH}_4)_2\text{SO}_4$. The dashed line is an assumed ambient supersaturation discussed in the text.

Haze droplet. A droplet at point A is in equilibrium with its supersaturated vapor phase (with respect to water). Haze droplets in the atmosphere can considerably reduce visibility by scattering light.

Regimes:

- ▶ peak corresponds to $(r_{\text{crit}}, SS_{\text{crit}})$
- ▶ $r < r_{\text{crit}}$: aerosol
- ▶ $r > r_{\text{crit}}$: cloud droplet (*activated droplet*)

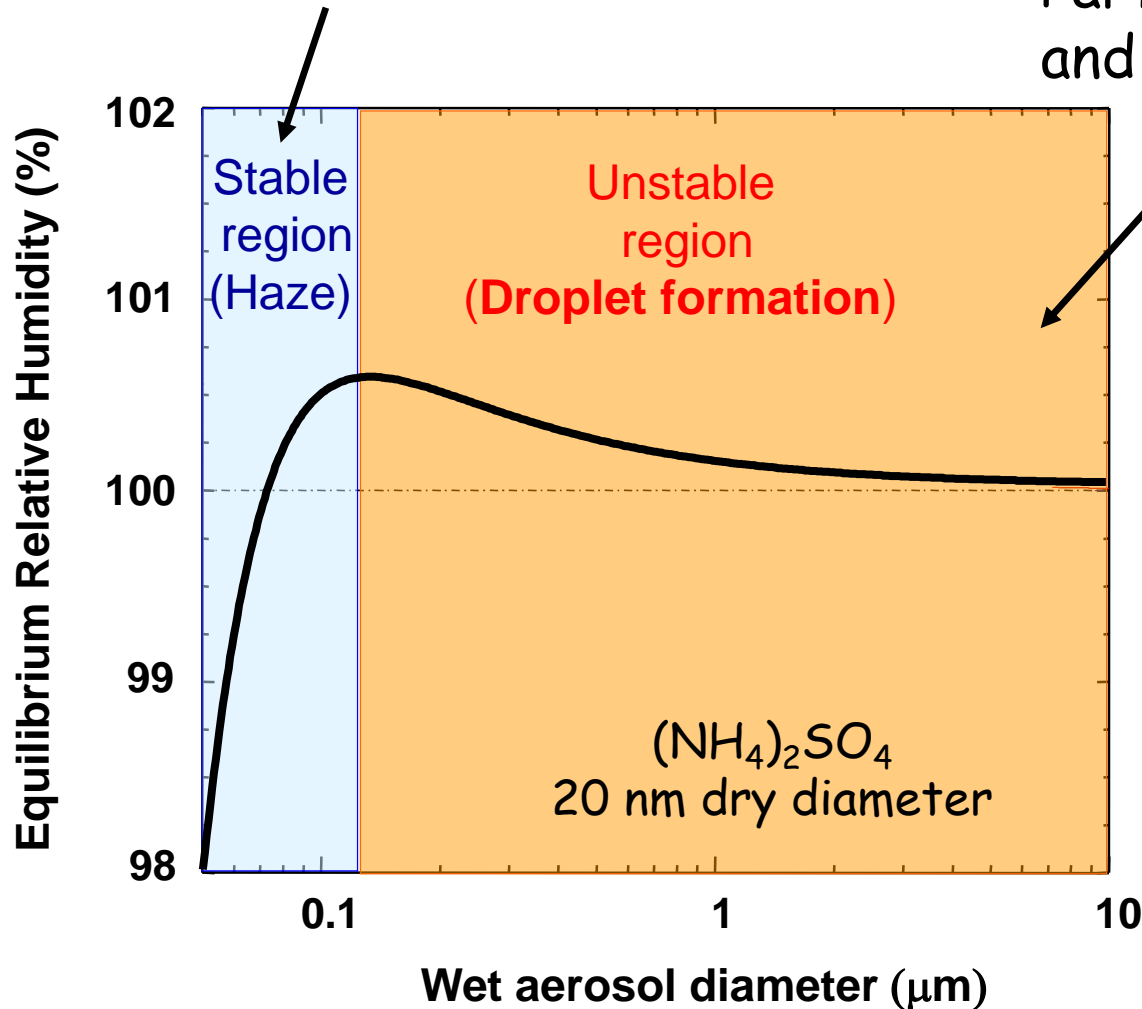
Compare SS_{amb} , SS_{eq} , and SS_{crit} :

- ▶ $r < r_{\text{crit}}$ and $SS_{\text{eq}} < SS_{\text{amb}} < SS_{\text{crit}}$: condensational growth will increase droplet radius and SS_{eq} ; growth is terminated when $SS_{\text{eq}} = SS_{\text{amb}}$
- ▶ $r > r_{\text{crit}}$ and $SS_{\text{crit}} < SS_{\text{amb}}$: aerosol will activate into a cloud droplet
- ▶ $r > r_{\text{crit}}$ and $SS_{\text{eq}} < SS_{\text{amb}}$: growth of droplet will increase SS_{eq} and further increase the driving force for condensational flux $SS_{\text{amb}} - SS_{\text{eq}}$, until water vapor is eventually reduced ($SS_{\text{amb}} \rightarrow SS_{\text{eq}}$)

When does an aerosol particle act as a CCN ?

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

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



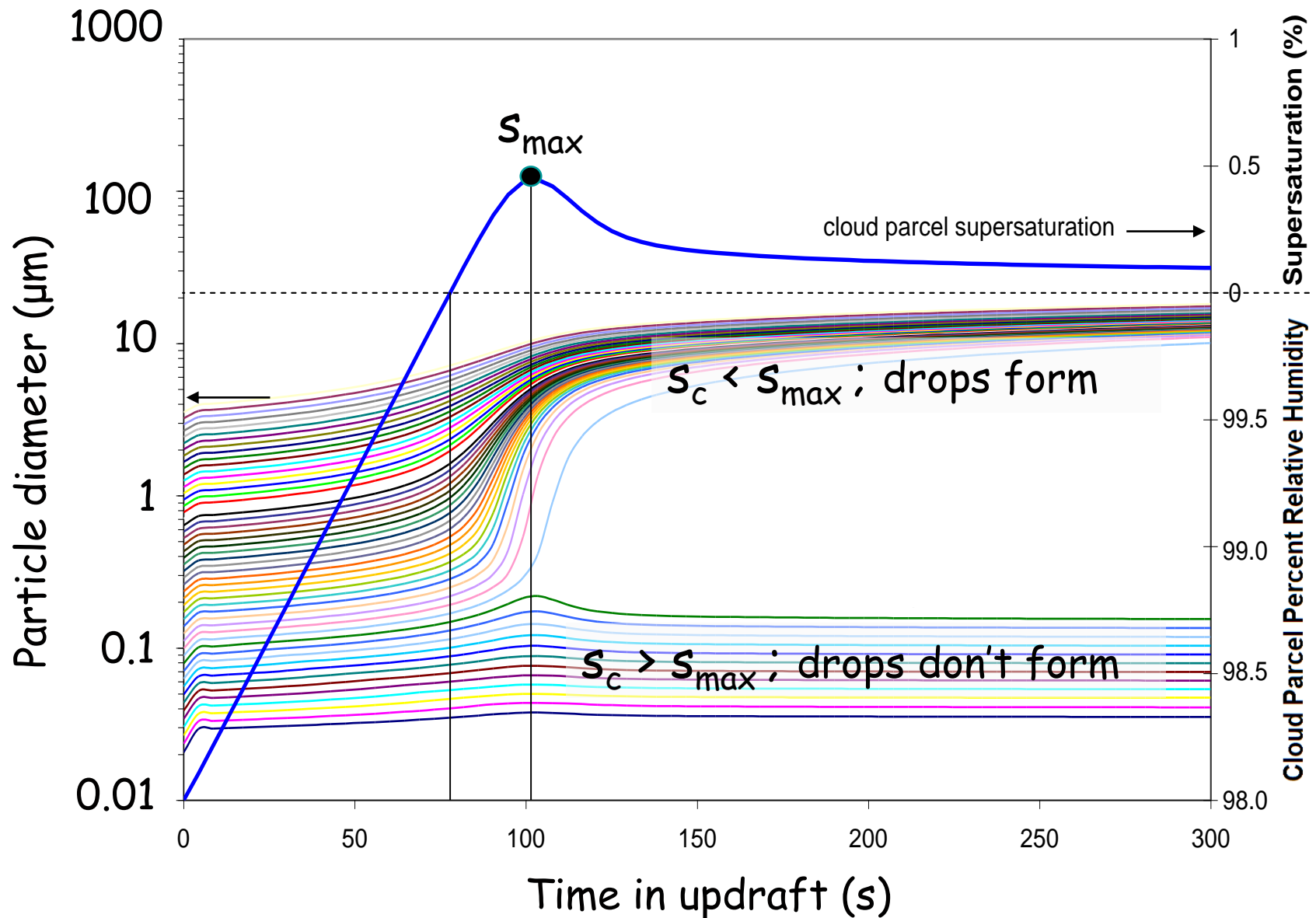
Köhler theory:

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

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

Size is more
important than
composition

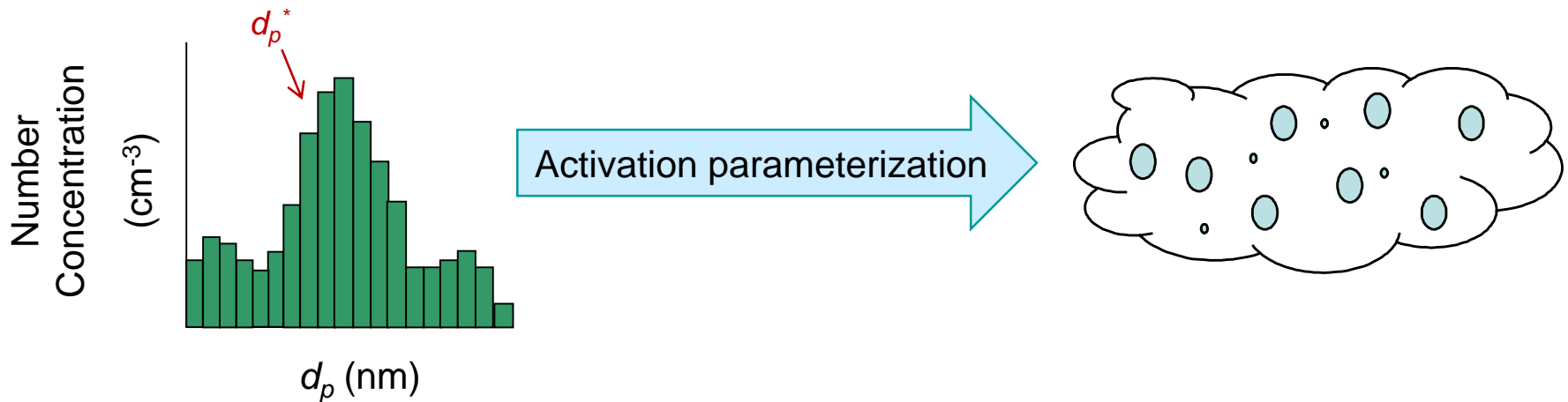
Now we understand droplet formation



Describing droplet formation in models...

Droplet calculation in models then is:

Calculated size distribution + κ + vertical velocity

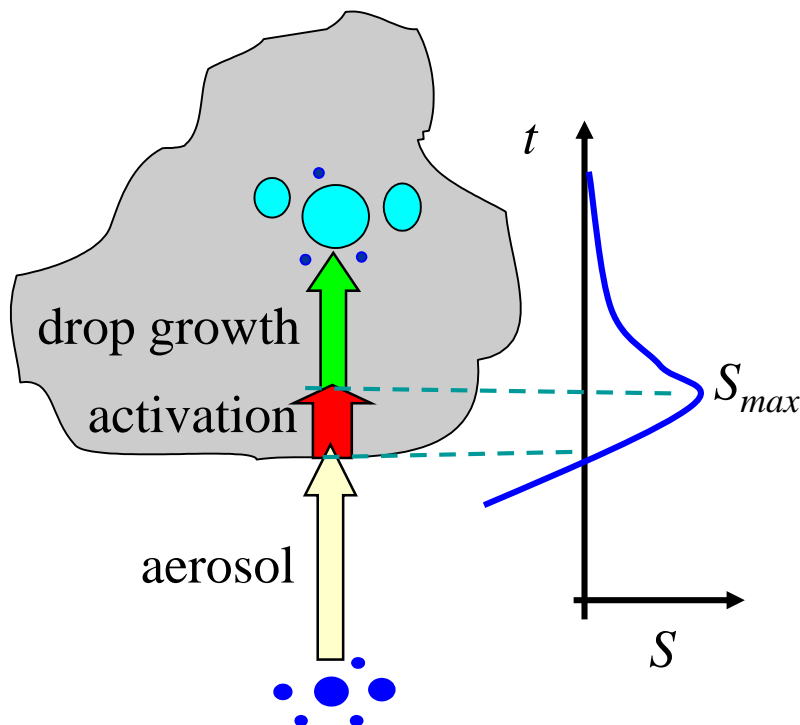


Activation parameterization is either a correlation or a solution to the parcel model equations that describe the activation process in clouds.

Droplet number needs CCN and max.cloud RH...

Algorithm for calculating N_d :
(Mechanistic parameterization)

1. Calculate S_{max} (approach-dependent)
2. N_d is equal to the CCN with $s_c \leq S_{max}$



Mechanistic Parameterizations:

Twomey (1959); Abdul-Razzak et al., (1998); Nenes and Seinfeld, (2003); Fountoukis and Nenes, (2005); Kumar et al. (2009), Morales and Nenes (2014), and others.

Input: P, T, vertical wind, particle size distribution, composition.

Output: Cloud properties (droplet number, size distribution).

Comprehensive review & intercomparison:

Ghan, et al., JAMES (2011); Morales and Nenes (2014)

Is this description of droplet formation real

Evaluate with in-situ data from airborne platforms



Observed Aerosol size
distribution & composition

Observed Cloud updraft
Velocity (PDF)

Predicted Drop Number
(Parameterization)

Compare

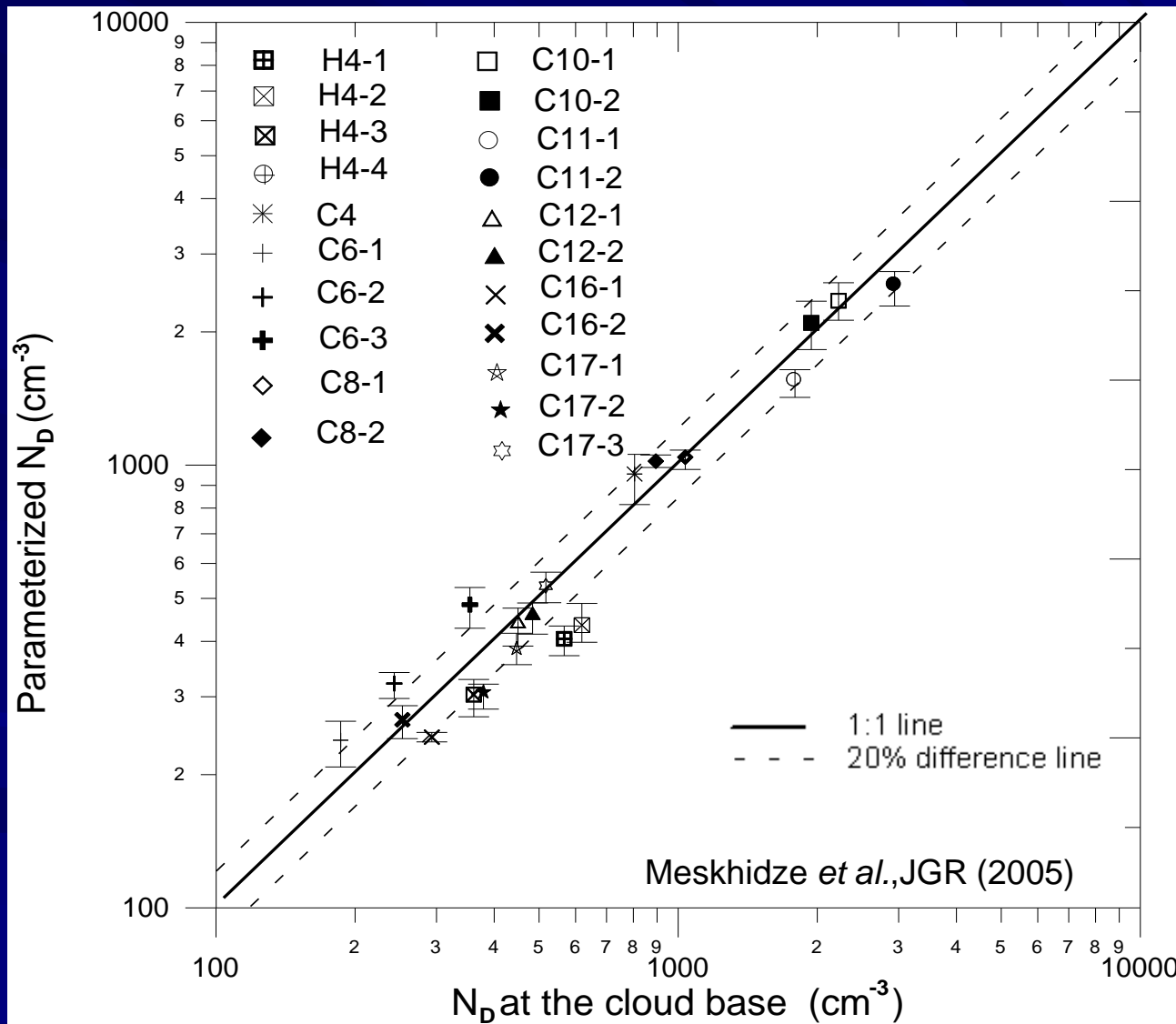
Observed Drop Number
Concentration



CRYSTAL-FACE (2002) Cumulus clouds



CIRPAS Twin Otter



Parameterized
agrees with
observed cloud
droplet number

Agreement to
within a few %
(on average)!

...when you know
everything
about aerosol
composition and
size

Issue: aerosols are complex



Primary emissions

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

Secondary transformations

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

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



Parameterizing “characteristic” CCN activity...

Petters and Kreidenweis (2007) expressed the Kohler theory parameter B in terms of a “hygroscopicity parameter”, κ

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

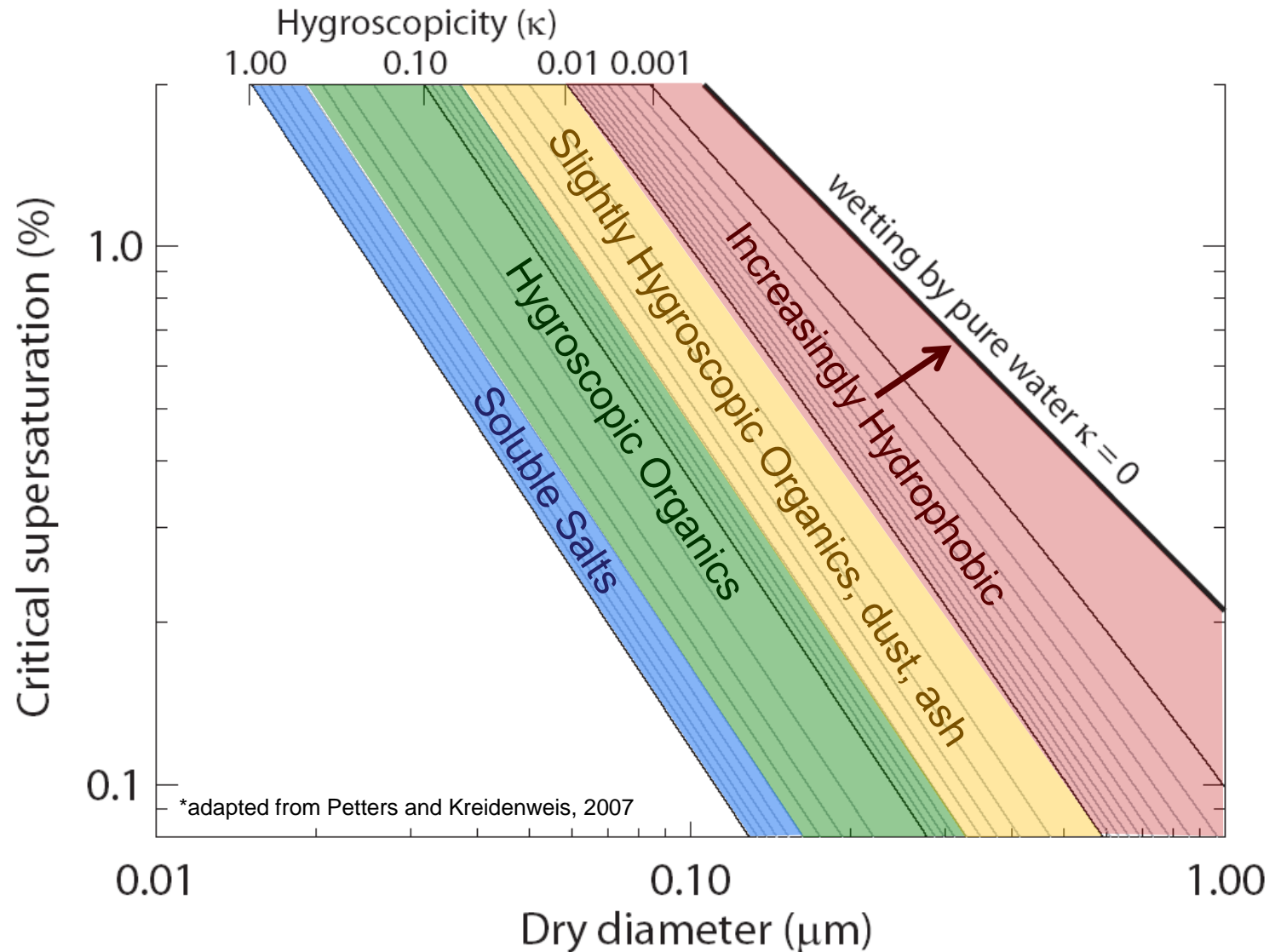
$\kappa \sim 1$ for seasalt, ~ 0.6 for $(\text{NH}_4)_2\text{SO}_4$, $\sim 0\text{-}0.3$ for organics

κ rarely exceeds 1 in atmospheric aerosol

Simple way to think of κ : the “equivalent” volume fraction of seasalt in the aerosol (the rest being insoluble).

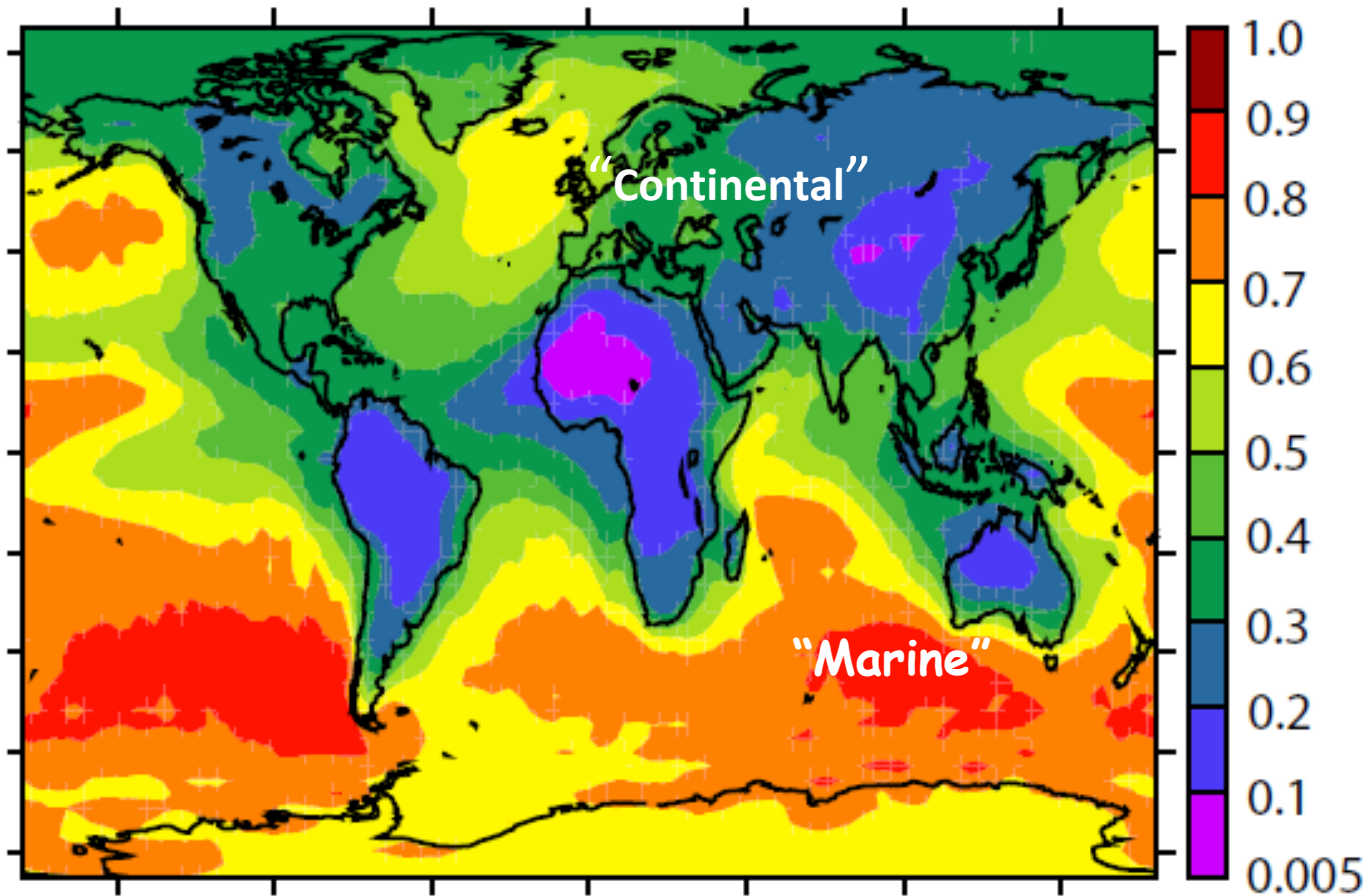
$\kappa \sim 0.6 \Rightarrow$ particle behaves as 60% seasalt, 40% insoluble

Hygroscopicity Space



- κ is used to parameterize the activation of particles in the atmosphere

Global “average” distribution for κ



Pringle et al., ACP, (2010)

Fig. 2. Annual mean distribution of κ at the altitude of the planetary boundary layer.

Some References

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- Petters, M. D., and S. M. Kreidenweis (2007), A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7, 1961–1971.
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- Fountoukis, C. and Nenes, A., ISORROPIA II: A Computationally Efficient Aerosol Thermodynamic Equilibrium Model for K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , Na^+ , SO_4^{2-} , NO_3^- , Cl^- , H_2O Aerosols, *Atmos.Chem.Phys.*, 7, 4639, 2007
- Bougiatioti, A., Fountoukis, C., Kalivitis, N., Pandis, S.N., Nenes, A. and Mihalopoulos, N., Cloud Condensation Nuclei Measurements in the Marine Boundary Layer of the Eastern Mediterranean: CCN closure and droplet growth kinetics, *Atmos.Chem.Phys.*, 9, 7053-7066, 2009
- <http://lapi.epfl.ch> (CCN measurements & techniques)
- <http://lapi.epfl.ch/ISORROPIA> (CCN thermodynamics)