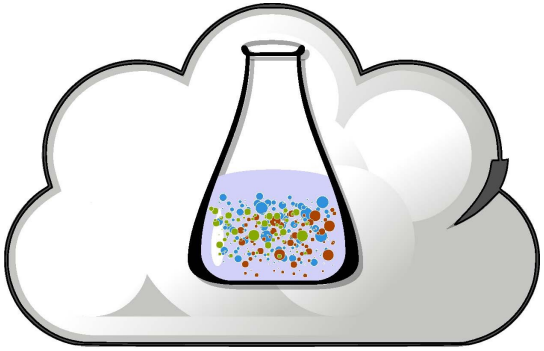
Activation of Cloud Droplets



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Heterogeneous nucleation

Cloud Condensation Nuclei (CCN) activation

Diffusional growth

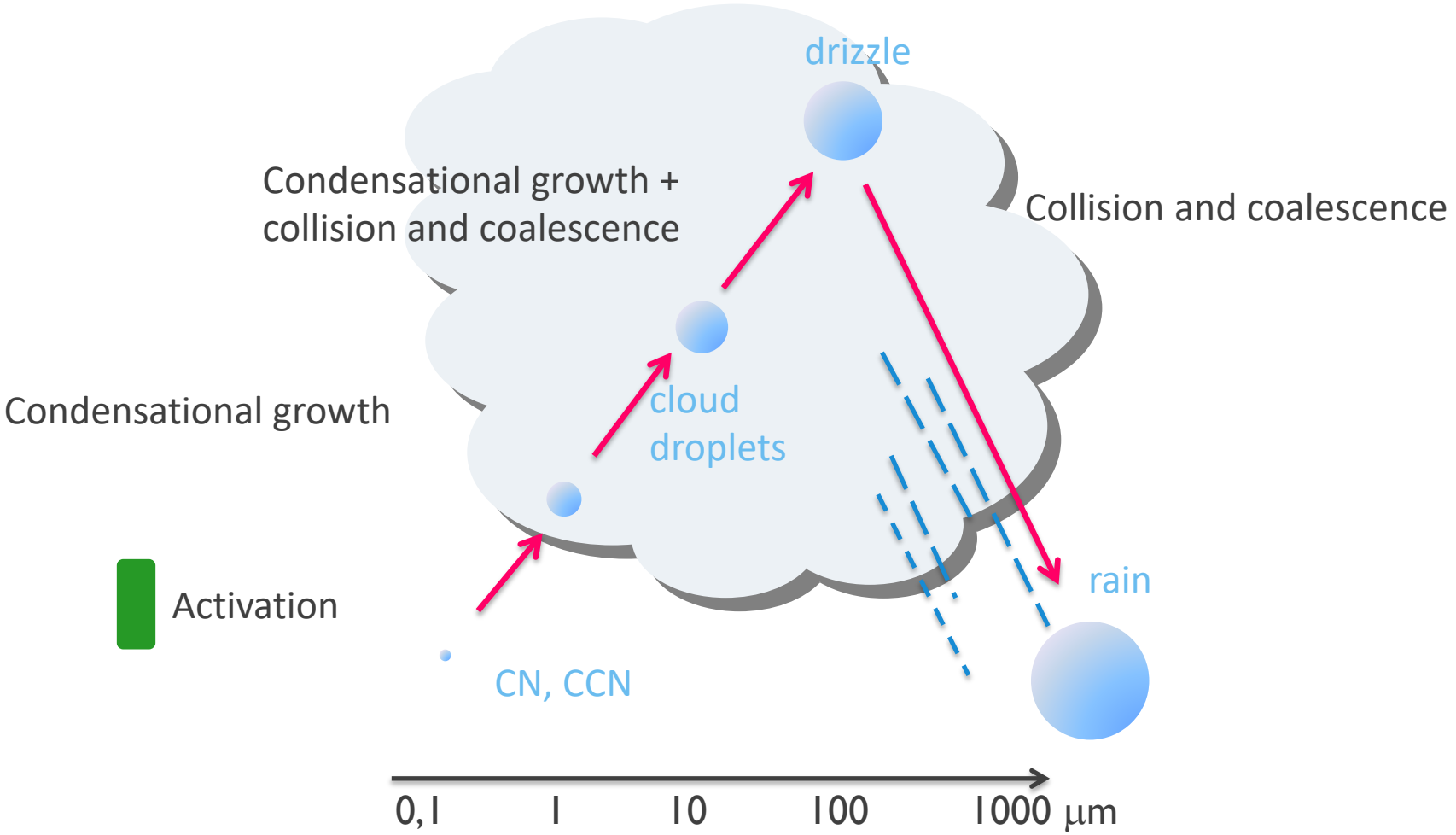
Condensational growth

Collision/coalescence

Drizzle formation

Rain

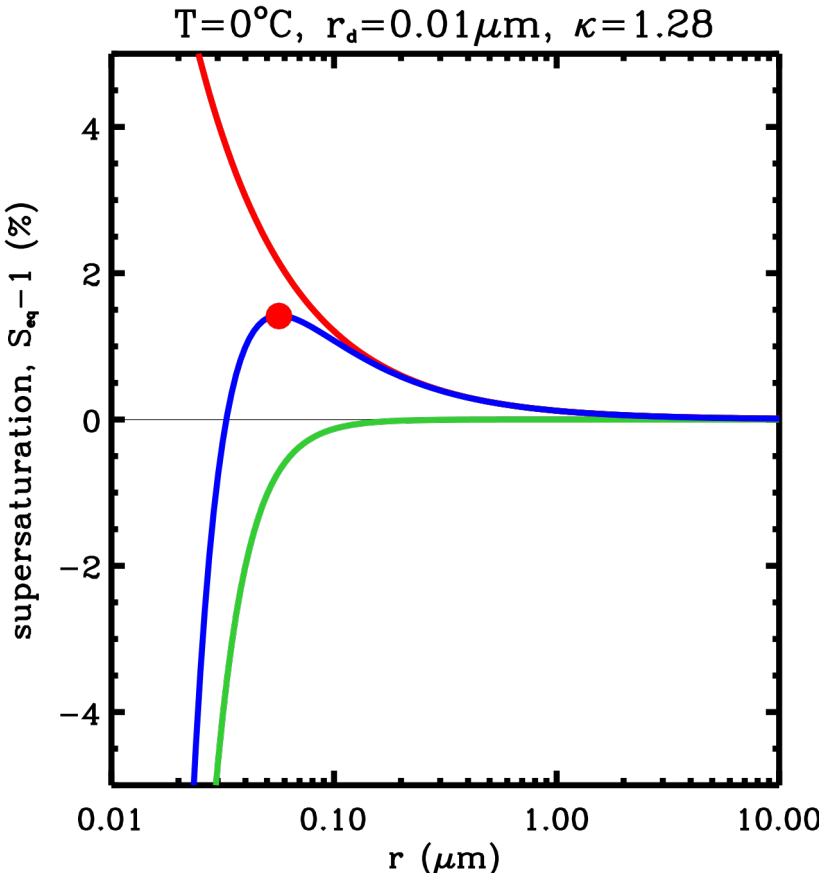
CCN washout


drizzle
Condensational growth +
collision and coalescence
Collision and coalescence
Condensational growth
cloud
droplets
Activation
rain
CN, CCN
0,1
1
10
100
1000 μm

Köhler curve

$$S_{eq}(r, r_d, \kappa, T) = \frac{r^3 - r_d^3}{r^3 - (1-\kappa) r_d^3} \exp\left(\frac{A(T)}{r}\right)$$

$$S_{eq}(r, r_d, \kappa, T) \approx 1 + \frac{A(T)}{r} - \frac{\kappa r_d^3}{r^3}$$



Surface tension of pure water

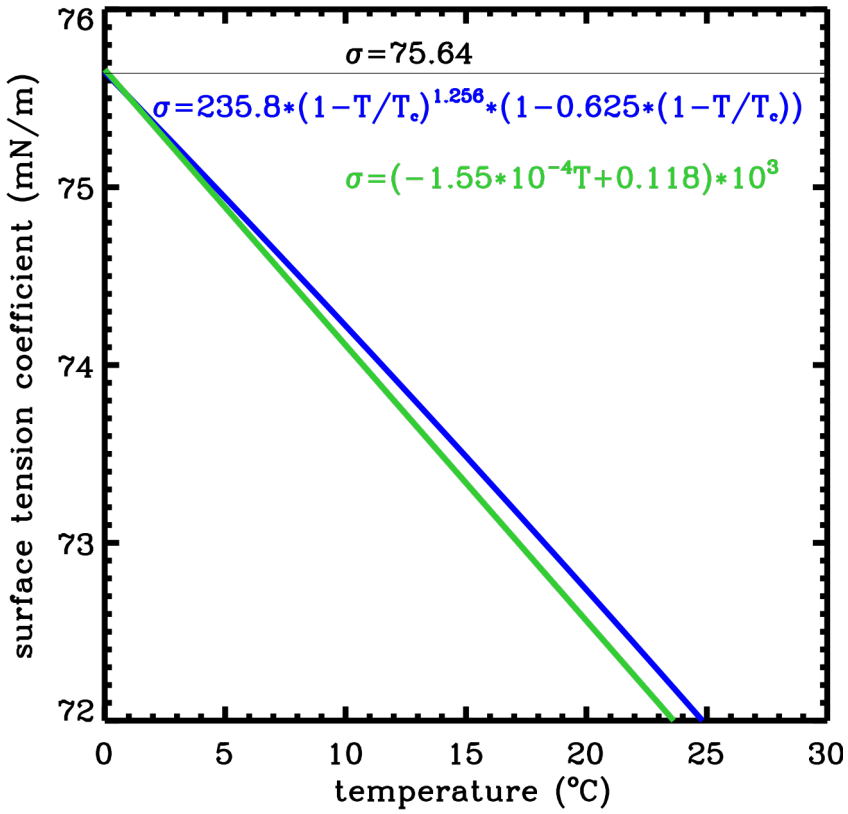
$$S(r,T) = \exp\left(\frac{2 \cdot \sigma(T)}{r \rho_l R_v T}\right)$$

$$\sigma(T) = 235.8\left(1 - \frac{T}{T_c}\right)^{1.256}\left[1 - 0.625\left(1 - \frac{T}{T_c}\right)\right] \cdot 10^{-3}\ [Nm^{-1}];\ T_c = 647.096$$

or

$$\sigma(T) = -1.55 \cdot 10^{-4} T + 0.118\ [Nm^{-1}]$$

$$\sigma(0°\mathrm{C}) = 75.64 \cdot 10^{-3}\ Nm^{-1}$$

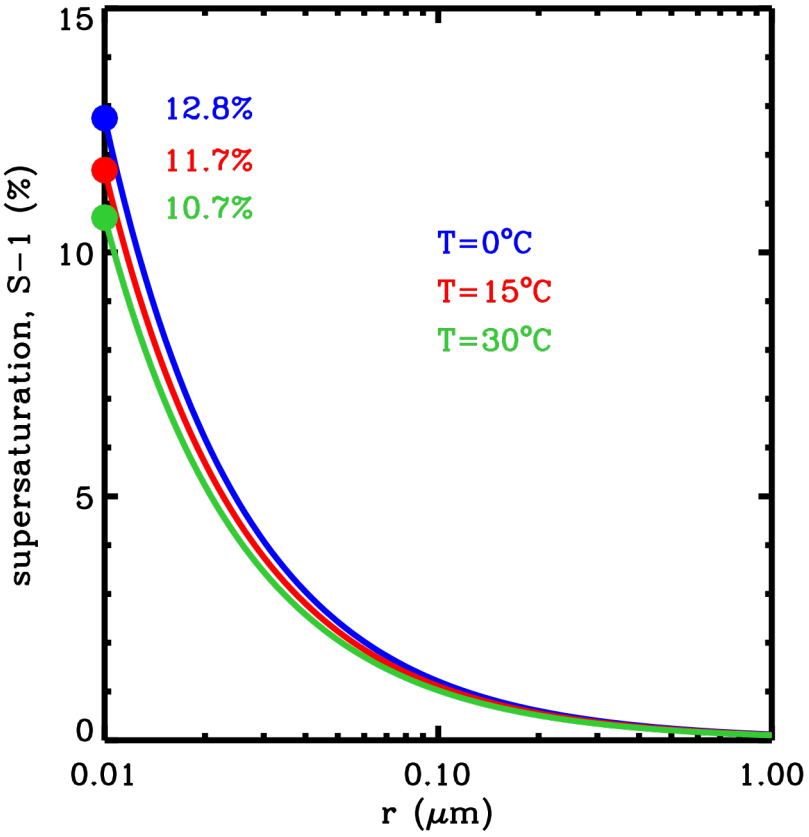


International Association for the Properties of Water and Steam (June 2014). "Revised Release on Surface Tension of Ordinary Water Substance"

Surface tension of pure water

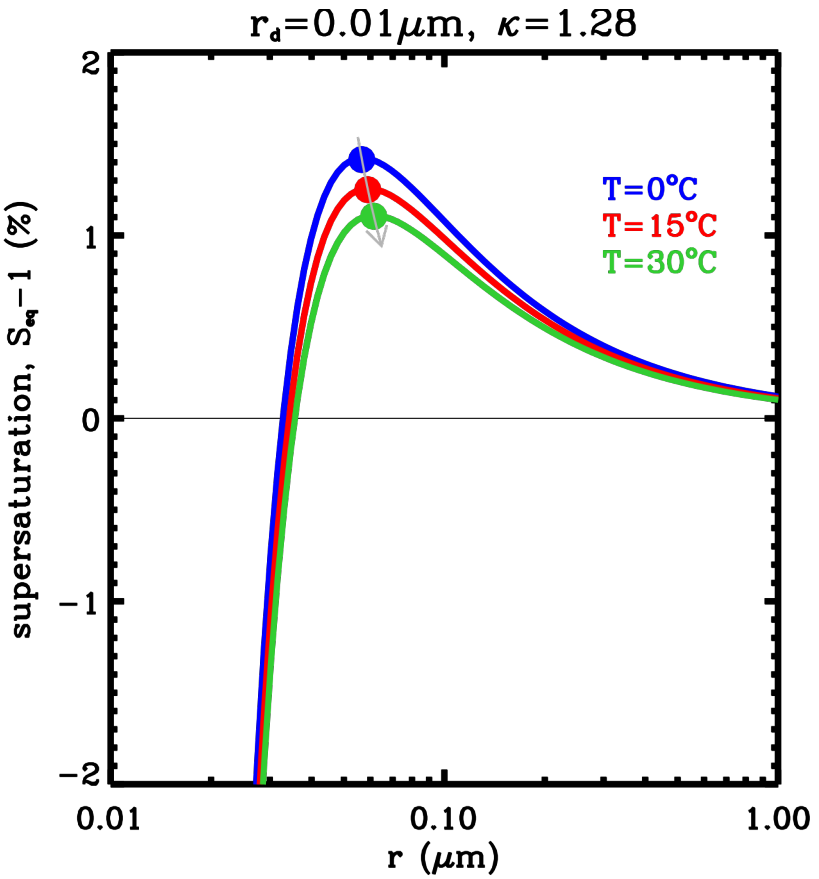
$$S(r,T) = \exp\left(\frac{2 \cdot \sigma(T)}{r \rho_l R_v T}\right) = \exp\left(\frac{A(T)}{r}\right)$$

$$T\nearrow \quad \rightarrow \quad A(T)\searrow$$



Köhler curve temperature dependance

$$S_{eq} - 1 = \frac{A(T)}{r} - \frac{\kappa r_d^3}{r^3}$$



$T \nearrow \quad \rightarrow \quad S^* \searrow \quad r^* \nearrow$

$$r^* = \sqrt{3\,\kappa r_d^3 / A(T)}$$

$$S^* - 1 = \sqrt{4A(T)^3 / 27\kappa r_d^3}$$

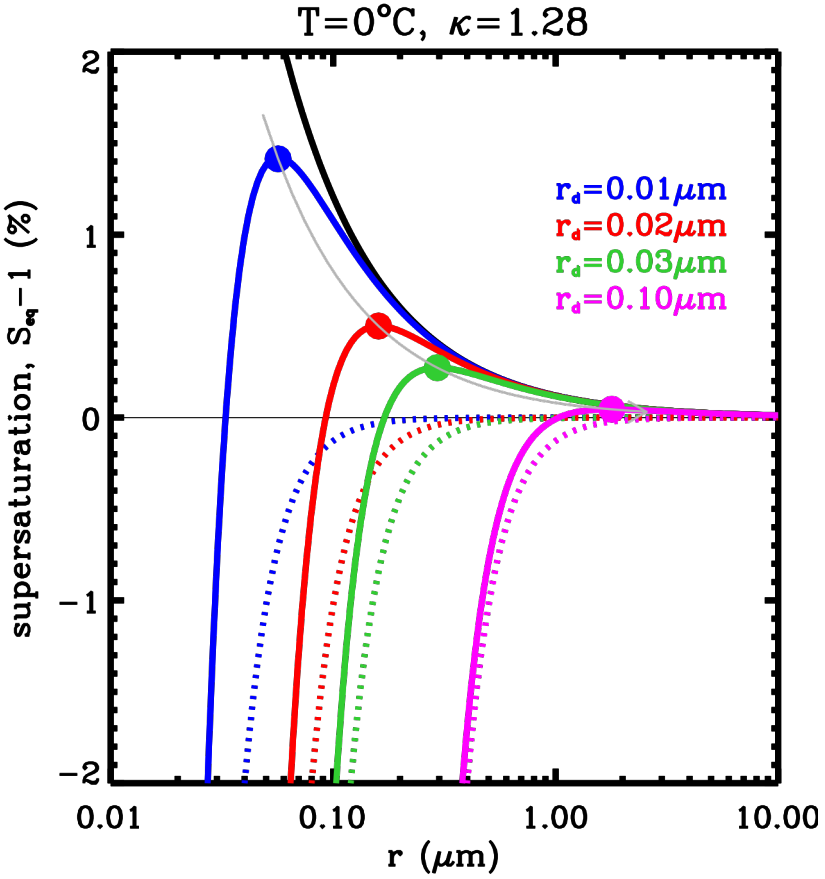
Köhler curve for different solute amount

$$S_{eq} - 1 = \frac{A(T)}{r} - \frac{\kappa r_d^3}{r^3}$$

$r_d \nearrow \rightarrow S^* \searrow \; r^* \nearrow$

$$r^* = \sqrt{3\,\kappa r_d^3 / A(T)}$$

$$S^* - 1 = \sqrt{4A(T)^3 / 27\kappa r_d^3}$$



Köhler curve for different solute

$$S_{eq} - 1 = \frac{A(T)}{r} - \frac{\kappa r_d^3}{r^3}$$

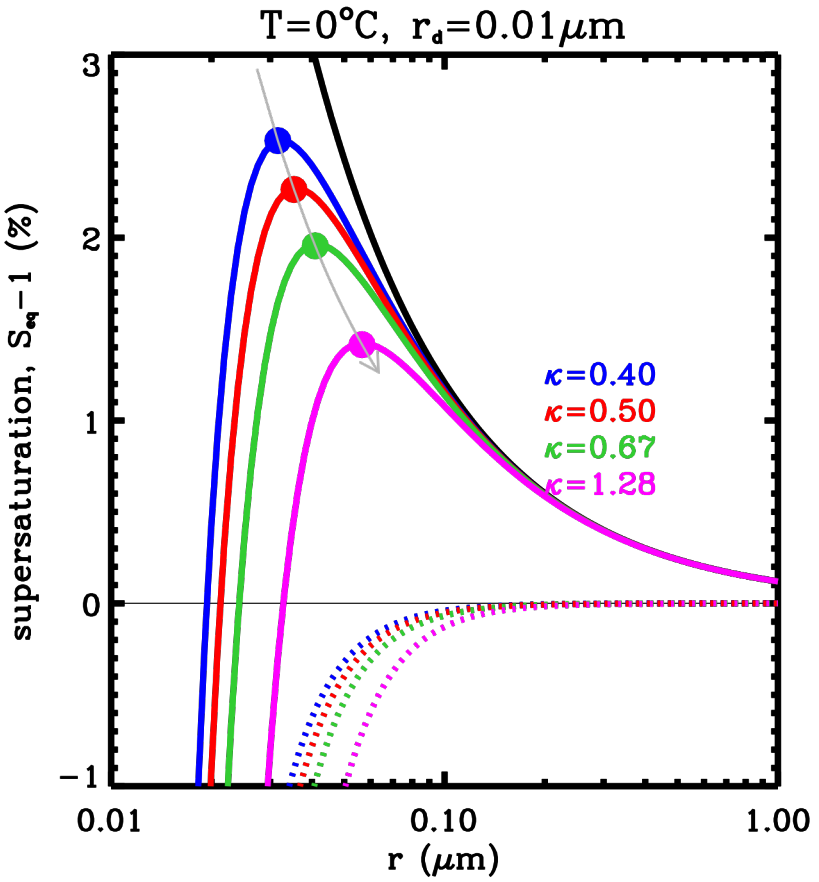
Sodium chloride NaCl
κ=1.28

Ammonium nitrate NH_4NO_3
κ=0.67

$\kappa \nearrow \rightarrow S^* \searrow \; r^* \nearrow$

$$r^* = \sqrt{3\,\kappa r_d^3 / A(T)} \propto \sqrt{\kappa}$$

$$S^* - 1 = \sqrt{4A(T)^3 / 27\kappa r_d^3} \propto {}^{1}/_{\sqrt{\kappa}}$$



$$r^* = \sqrt{3\,\kappa r_d^3 / A(T)}$$

$$S^* - 1 = \sqrt{4A(T)^3 / 27\kappa r_d^3}$$

$$r^*(S^* - 1) = \frac{2}{3}A(T)$$

Dependence of activation/critical parameters on r_d is stronger than on κ:

$$r^* \propto r_d^{3/2} \qquad r^* \propto \sqrt{\kappa}$$

- Hygroscopicity Range ($\kappa \in [0.2, 1.4]$): Variations in chemical composition alter critical parameters (S_c) by a factor of ≤ 2.5.
- Dry Radius Range (r_d spans > 1 order of magnitude): Variations in dry size influence critical parameters by a factor of ≥ 30.

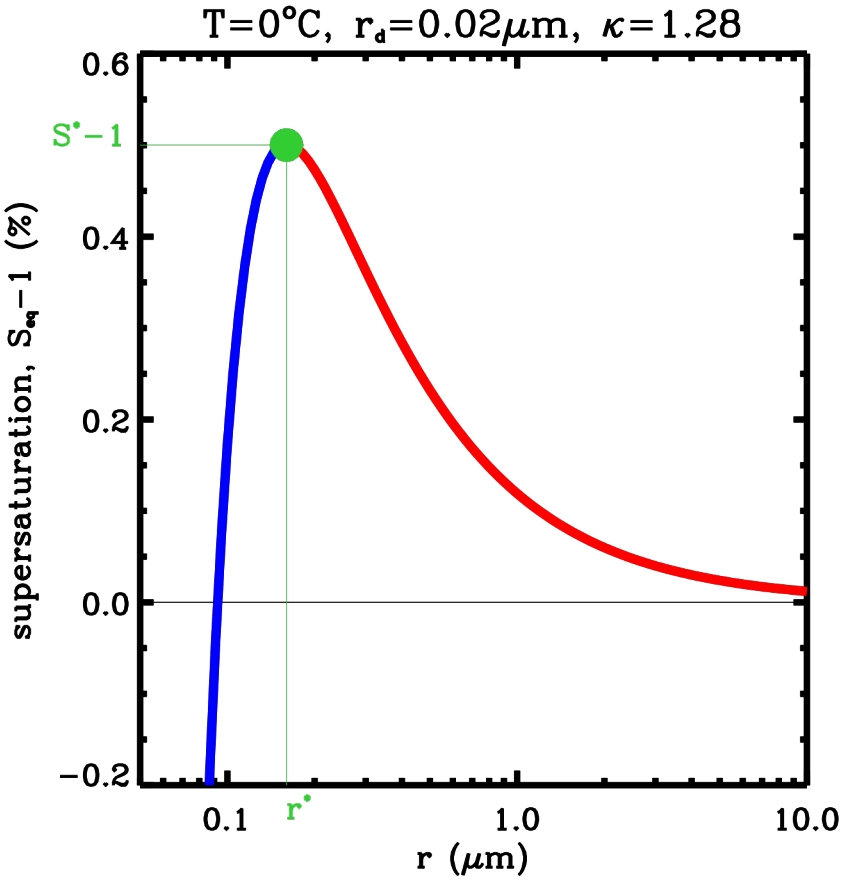
Equilibrium conditions

Stable equilibrium

$$\frac{dS}{dr} > 0$$

$$r < r^*$$

any random fluctuation in droplet size triggers a negative feedback mechanism that restores the droplet to its original equilibrium radius r_{eq}



Unstable equilibrium

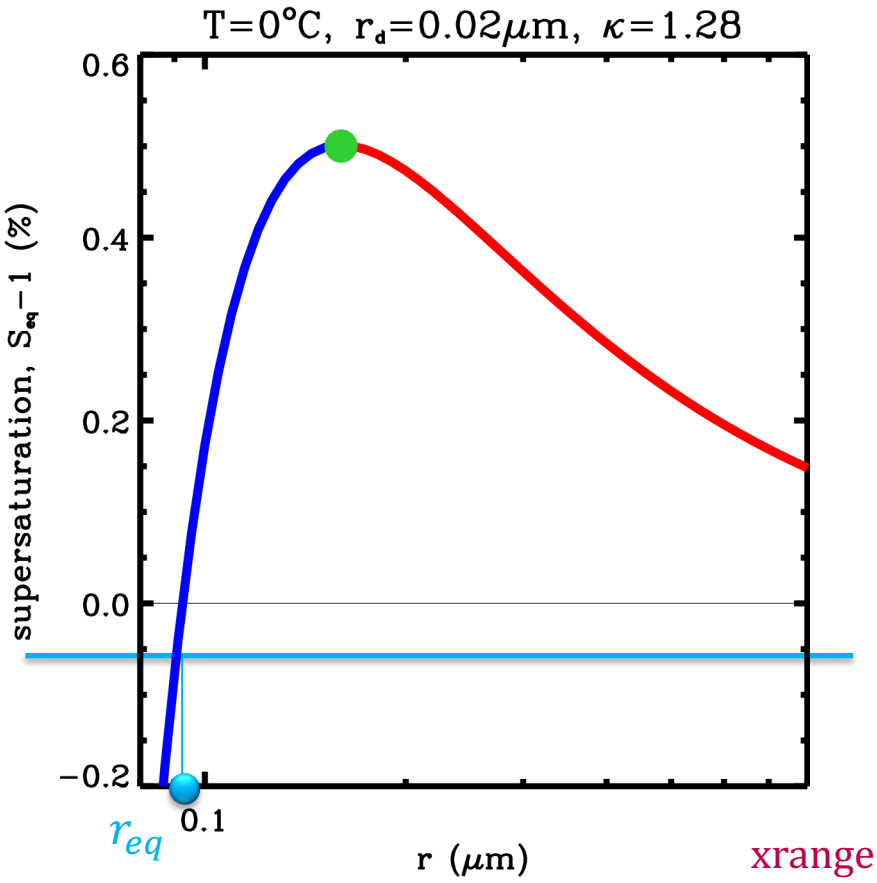
$$\frac{dS}{dr} < 0$$

$$r > r^*$$

any perturbation in droplet size triggers a positive feedback loop, driving the droplet away from its unstable equilibrium state

$$r^* = \sqrt{3\,\kappa r_d^3 / A(T)}$$

$$S^* - 1 = \sqrt{4A(T)^3 / 27\kappa r_d^3}$$

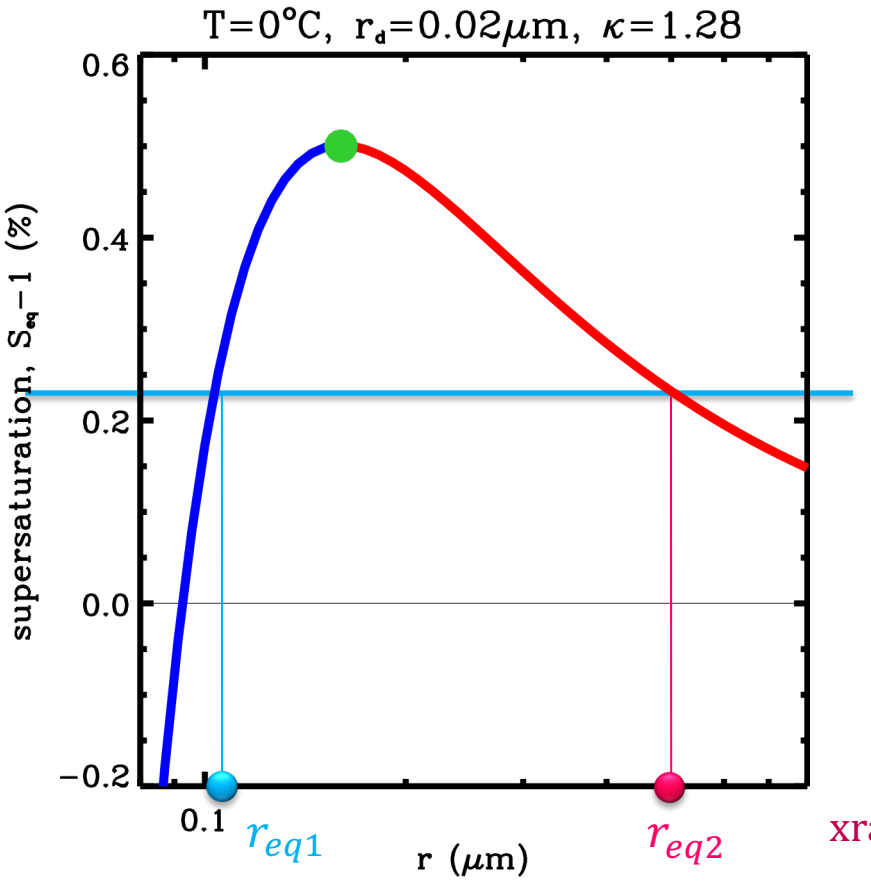


environmental supersaturation

$S - 1 < 0$

xrange $[0.08, 0.8]$ μm

If environmental supersaturation $S - 1 < 0$ (undersaturated!) there is only one equilibrium point.
Droplet with radius r_{eq} is in stable equilibrium with the environment.
Droplets with $r < r_{eq}$ will grow and droplets with $r > r_{eq}$ will evaporate to reach equilibrium at $r = r_{eq}$ where $S = S_{eq}(r_{eq})$.

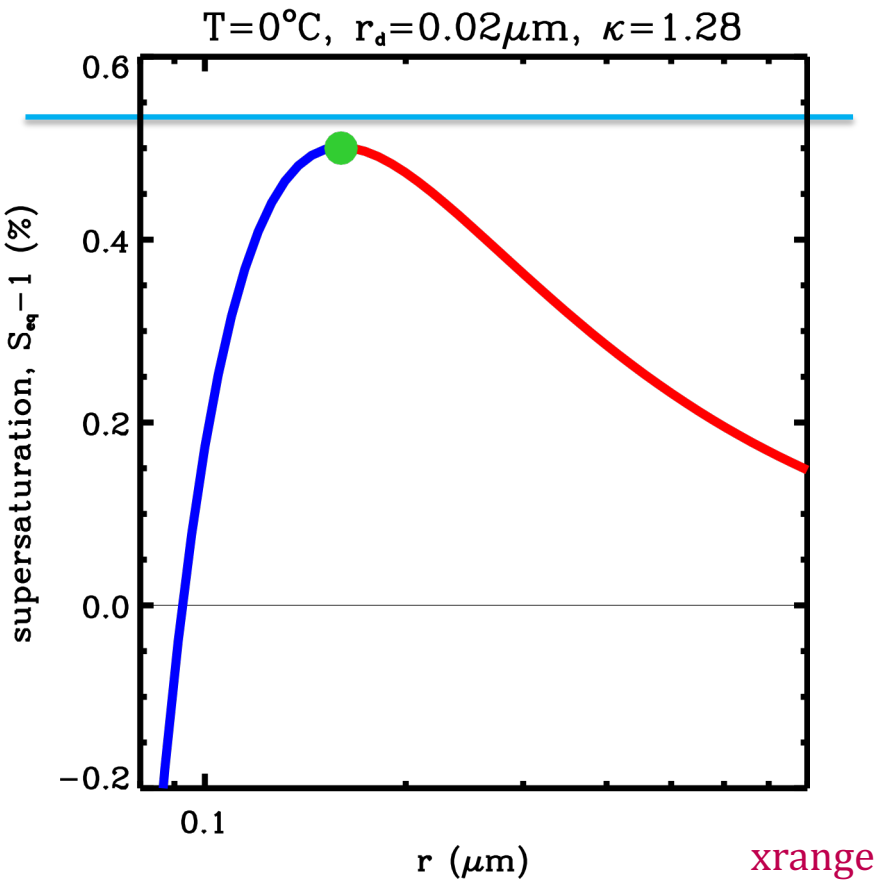


environmental supersaturation

$S - 1 < S^* - 1$

xrange [0.08, 0.8] μm

If $S - 1 > 0$ and $S < S^*$ there are two equilibrium points r_{eq1} and r_{eq2}.
Droplets with $r < r_{eq1}$ will grow and droplets with $r_{eq1} < r < r_{eq2}$ will evaporate to reach equilibrium at $r = r_{eq1}$ where $S = S_{eq}(r_{eq1})$.
Droplets with $r > r_{eq2}$ will spontaneously grow by condensation.



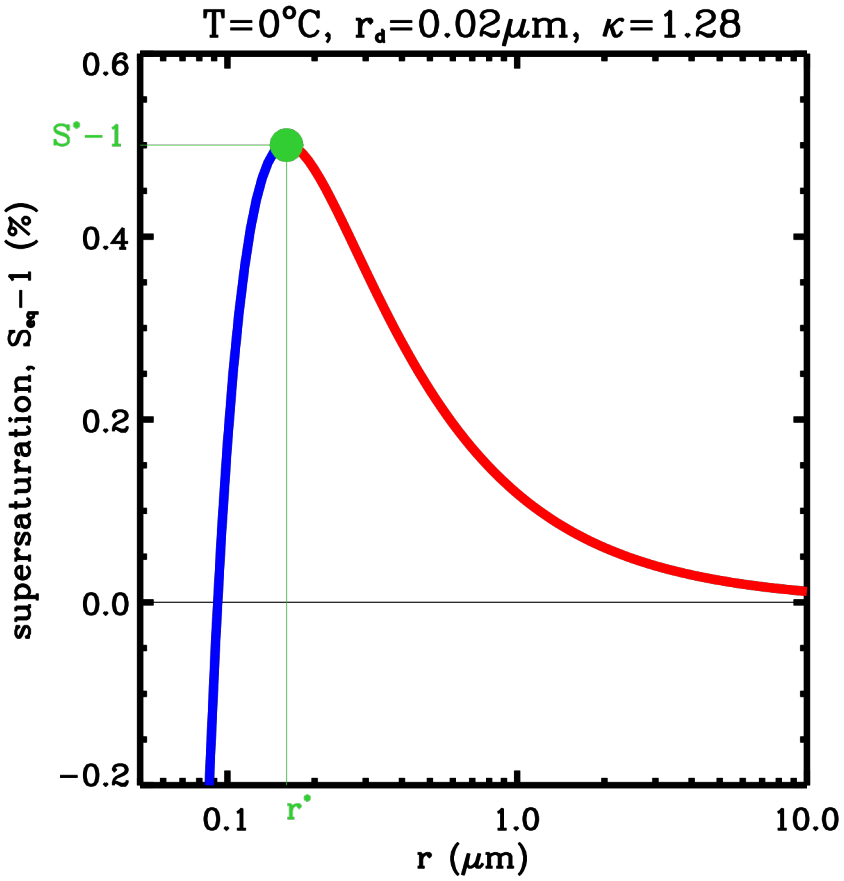
environmental supersaturation

$S - 1 > S^* - 1$

xrange $[0.08, 0.8]$ μm

If $S > S^*$ the environment is supersaturated with respect to all droplets.
All droplets will grow by condensation.

Equilibrium conditions



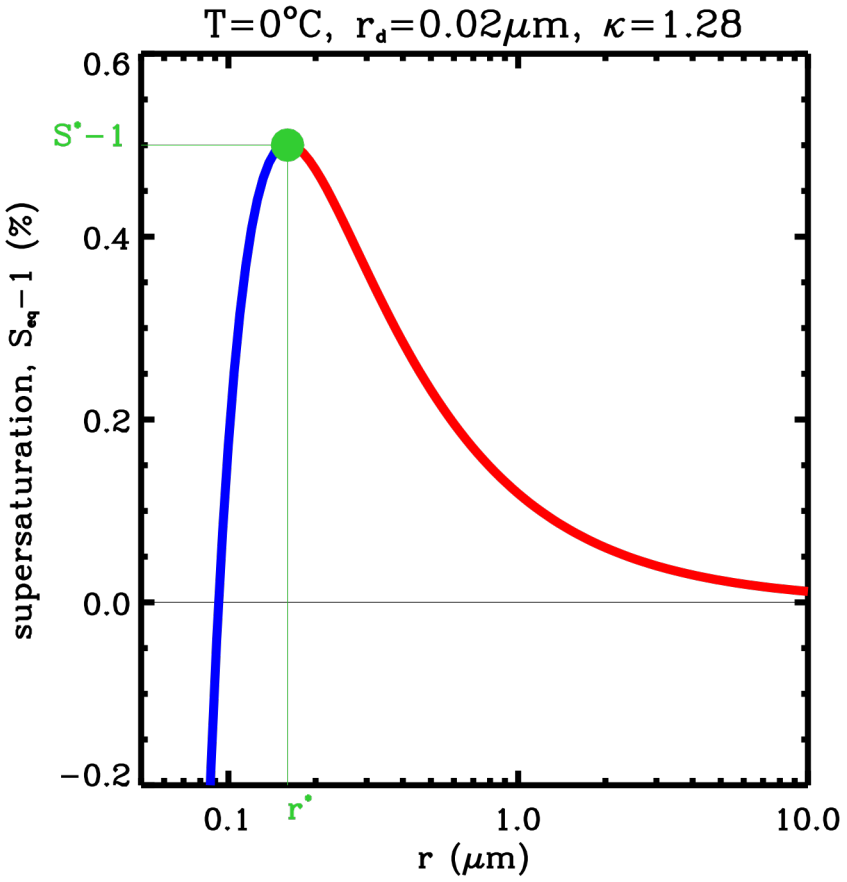
If $S < S^*$:

In the stable regime droplets adopt their size to get into equilibrium with the environment; i.e. $S_{eq}(r_{eq}) = S$.

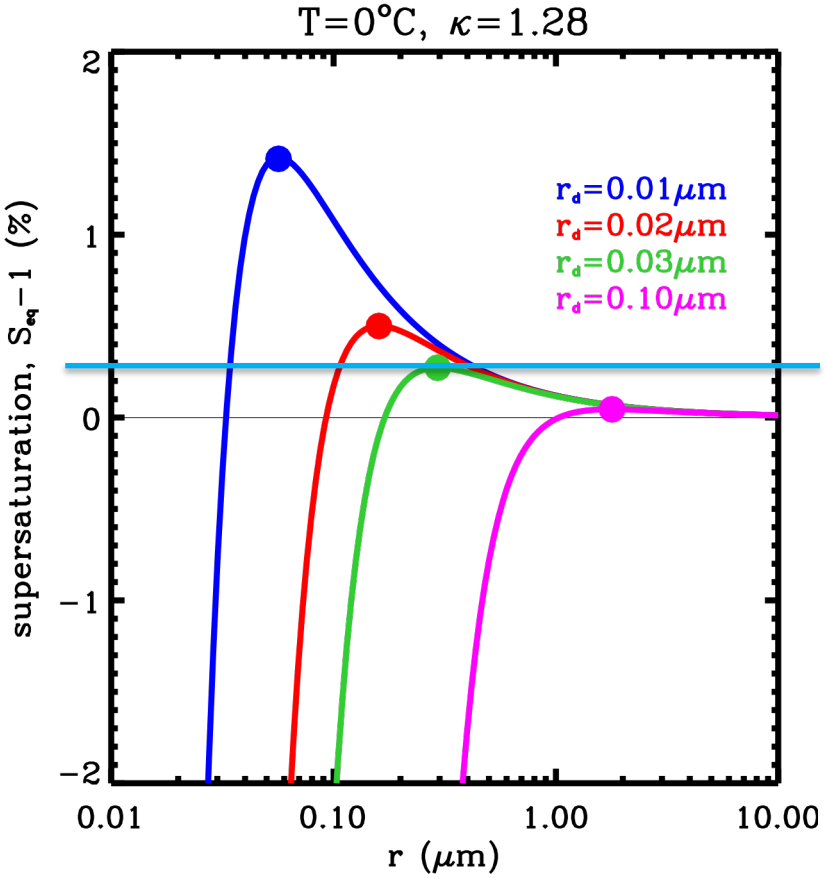
In the unstable regime droplets either evaporate to reach equilibrium $S_{eq}(r_{eq}) = S$ or they grow spontaneously by condensation.

Droplet activation; cloud condensational nuclei

- Activation: The transition from stable equilibrium ($r < r^*$) to an unstable, spontaneous growth regime ($r > r^*$) driven by ambient supersaturation.
- Defines the critical supersaturation (S^*) and critical radius (r^*) required for unconstrained droplet growth.
- Cloud condensation nuclei (CCN) : Aerosol particles with sufficient size and solute content to activate at a prescribed supersaturation.
- The concept of activation is fundamental to determining how aerosol particles function as CCN and establish the initial droplet size distribution and microstructure of clouds.



Activation



environmental supersaturation, $S_{env} - 1$

- **Activation Criterion**: Droplets activate only if their critical supersaturation is lower than the ambient value $S^*(r_d) < S_{env}$.
- **Size Threshold**: In the depicted scenario, only aerosol particles with dry radii $r_d > 0.03\mu m$ activate.
- **Sequential Activation**:
  - Initial/Low S_{env}: Larger CCN activate first (lower S^*).
  - Rising S^*: Progressively smaller CCN cross their activation threshold.

CN condensation nuclei
CCN cloud condensation nuclei

CN (condensation nuclei) aerosol particles that can become water drops for supersaturation <400%

CCN (cloud condensation nuclei) are defined for a given saturation S

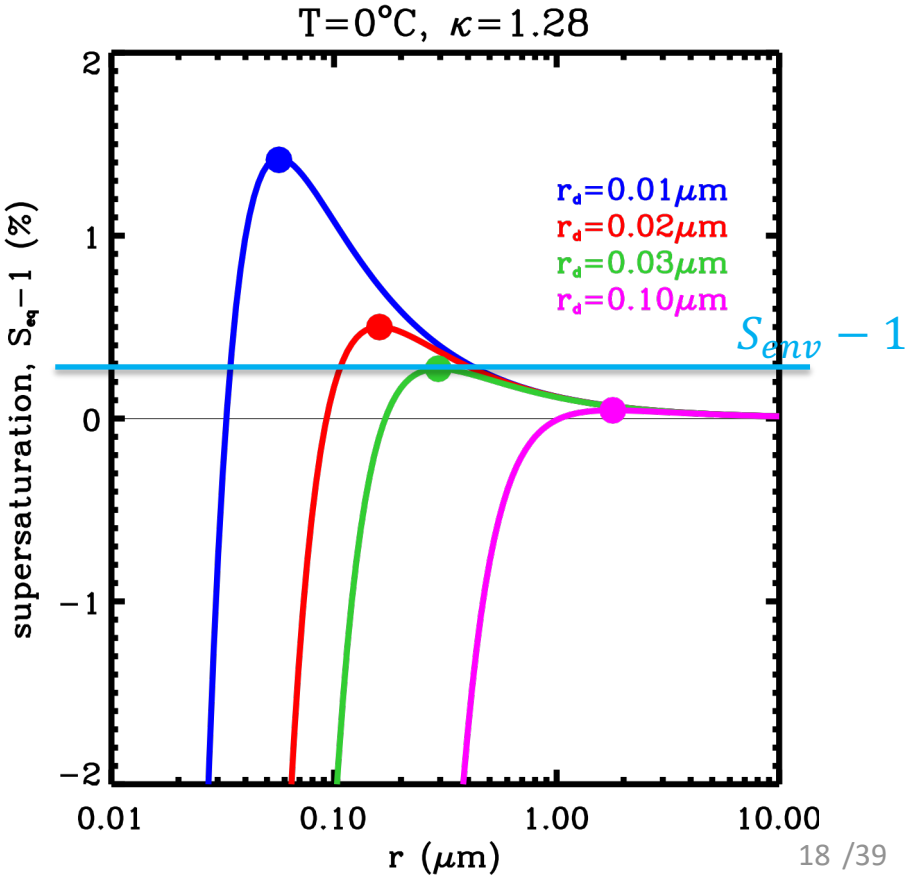
Critical supersaturation

$$S^* - 1 = \sqrt{4A(T)^3/27\kappa r_d^3}$$

For a given environmental saturation S_{env} all particles for which $r_d > r_{d,cr}$ will be activated/

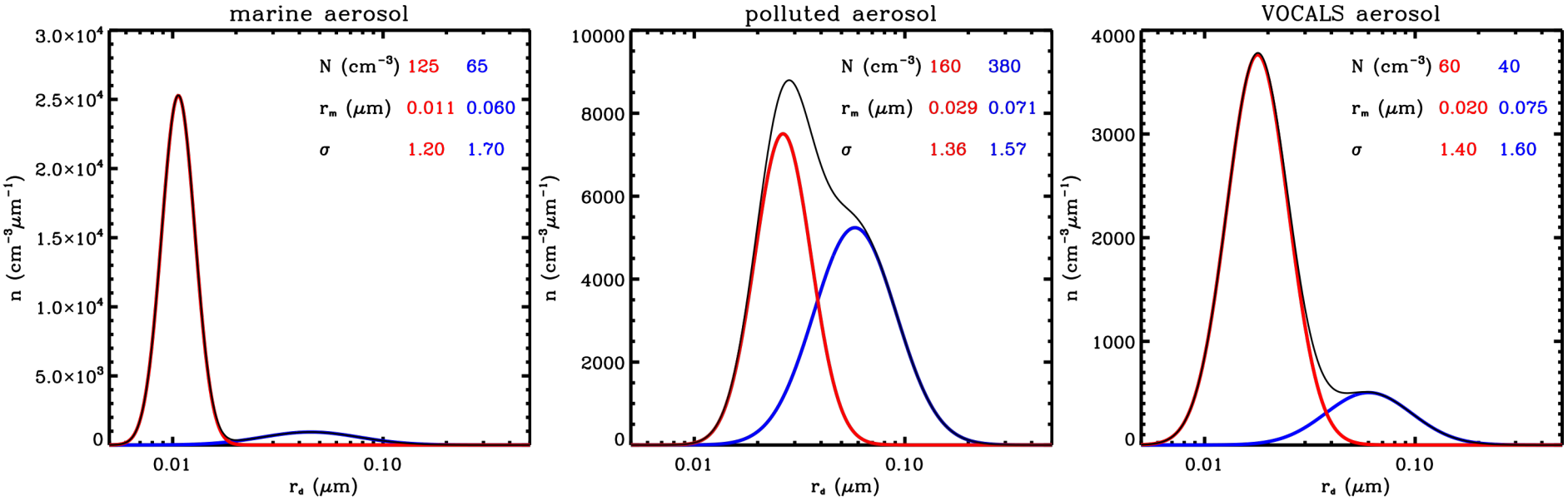
$$r_{d,cr} = \left(\frac{4A^3}{27\kappa(S_{env}-1)^2}\right)^{1/3}$$

In this figure $r_{d,cr} = 0.03\mu m$.



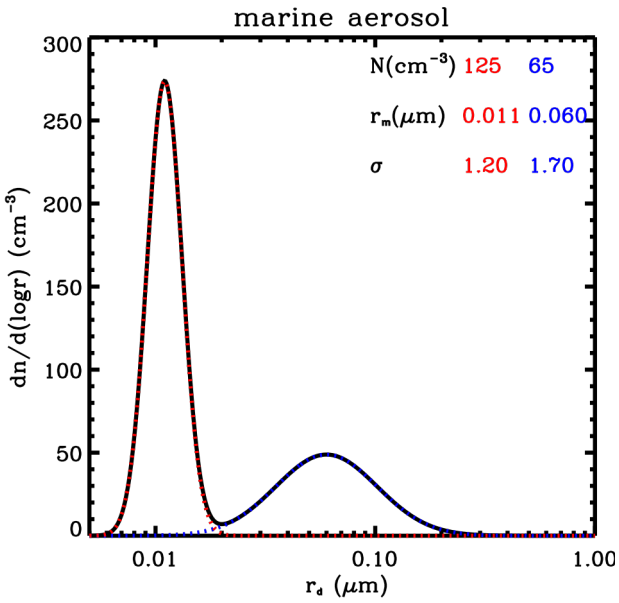
Log-normal distribution

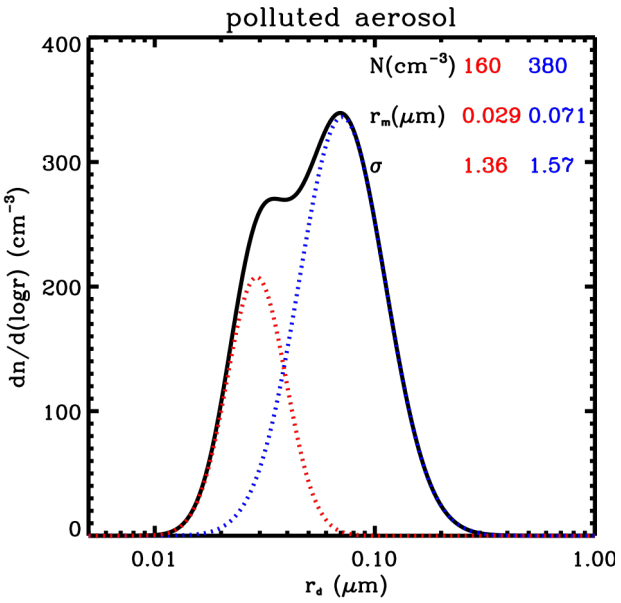
$$n(r_d) = \frac{dN(r_d)}{dr_d} = \frac{N}{\sqrt{2\pi}\, r_d \ln\sigma} \exp\left(-\frac{\left(\ln r_d - \ln r_{d,m}\right)^2}{2\, ln^2\sigma}\right) \quad [cm^{-3}\mu m^{-1}]$$

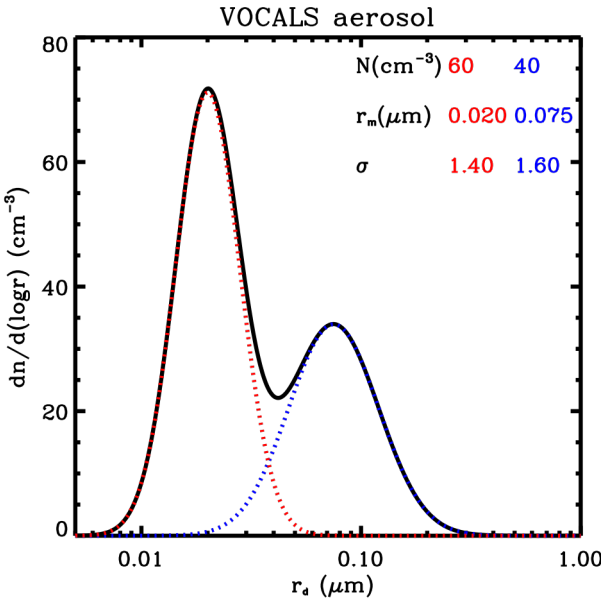


Log-normal distribution

$$n_l(r_d) = \frac{dN(r_d)}{d\log r_d} = r_d\, n(r_d) \ln 10 = \frac{N \ln 10}{\sqrt{2\pi}\, \ln \sigma} \exp\left(-\frac{\left(\ln r_d - \ln r_{d,m}\right)^2}{2\, ln^2 \sigma}\right) \; [cm^{-3}]$$



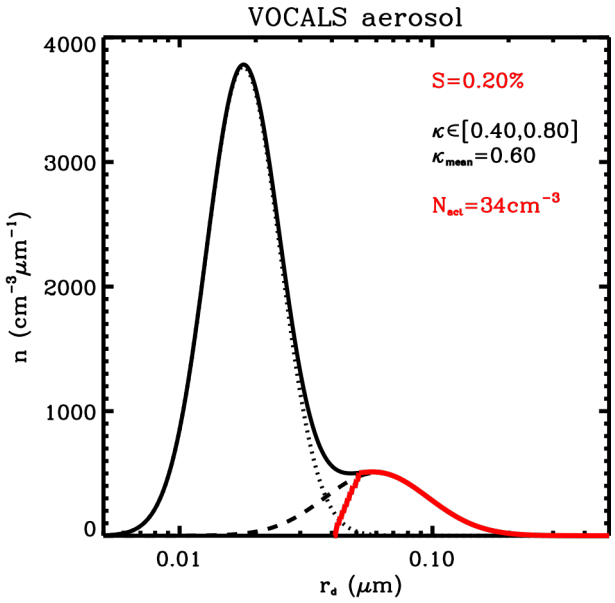


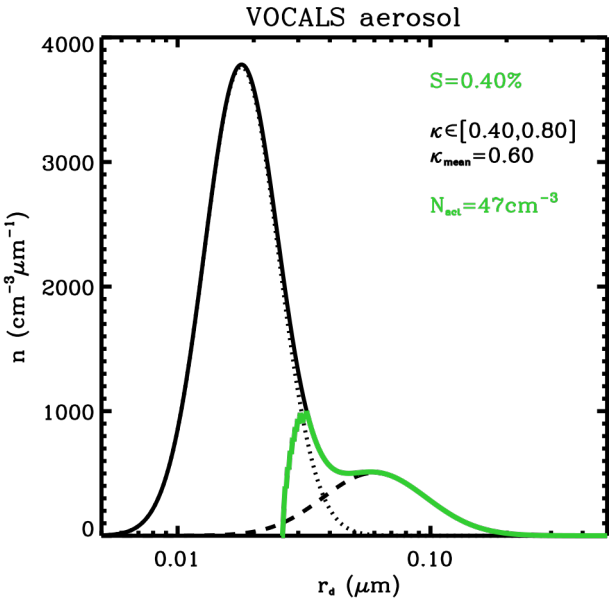


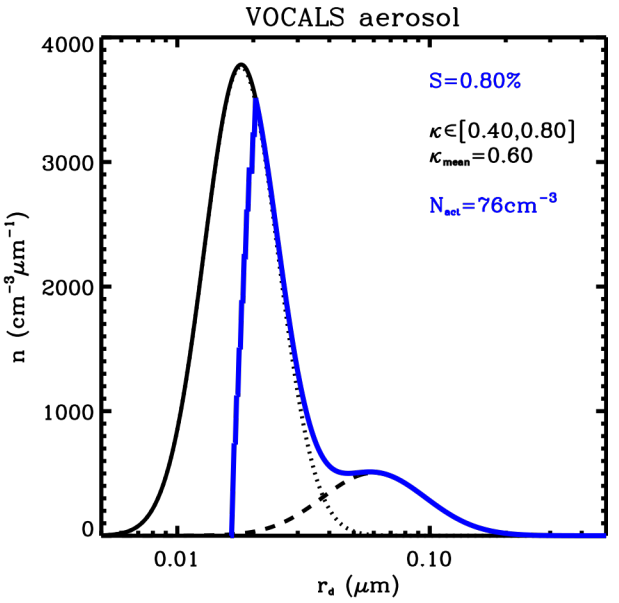
CN condensation nuclei
CCN cloud condensation nuclei

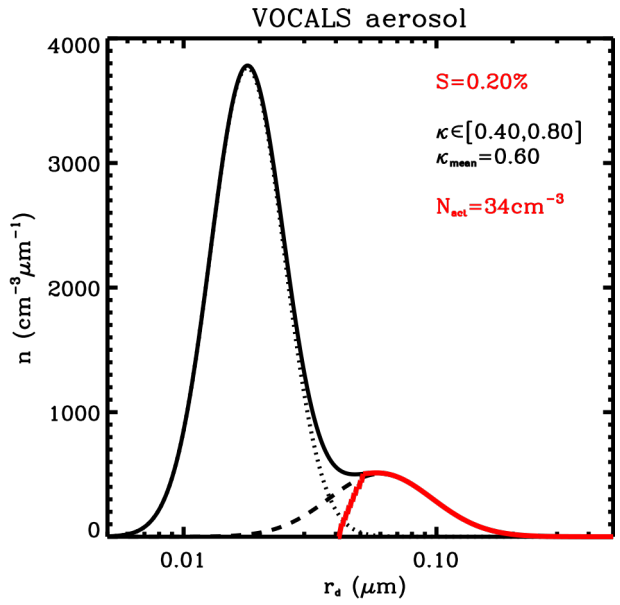
CN (condensation nuclei) aerosol particles that can become water drops for supersaturation <400%

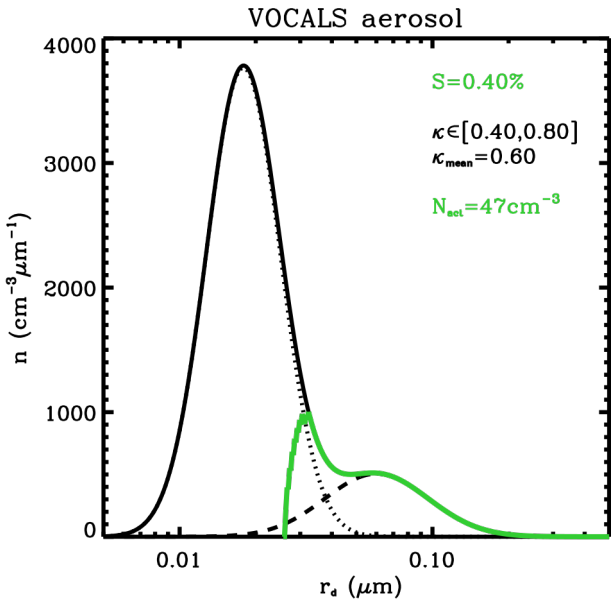
CCN (cloud condensation nuclei) are defined for a given supersaturation S

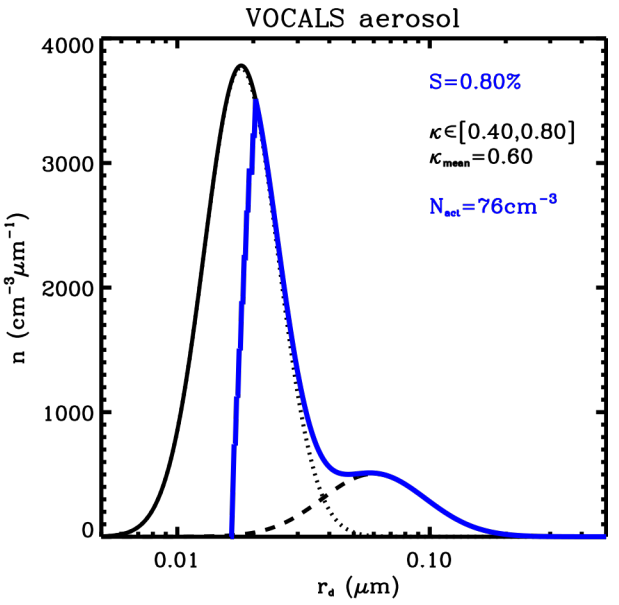


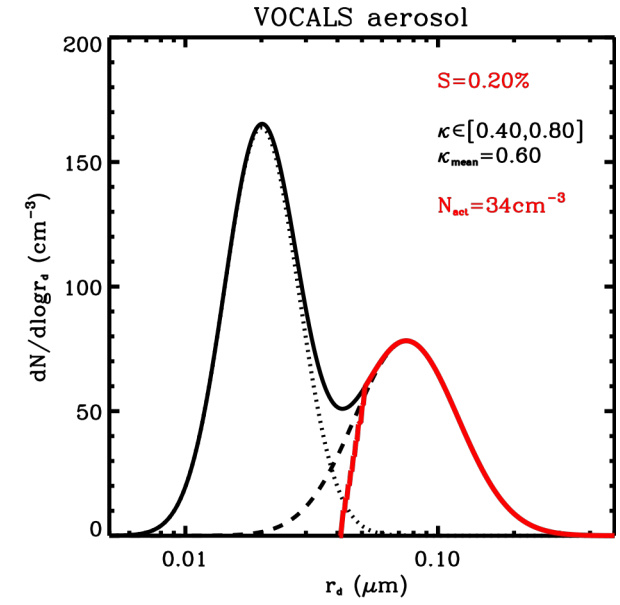


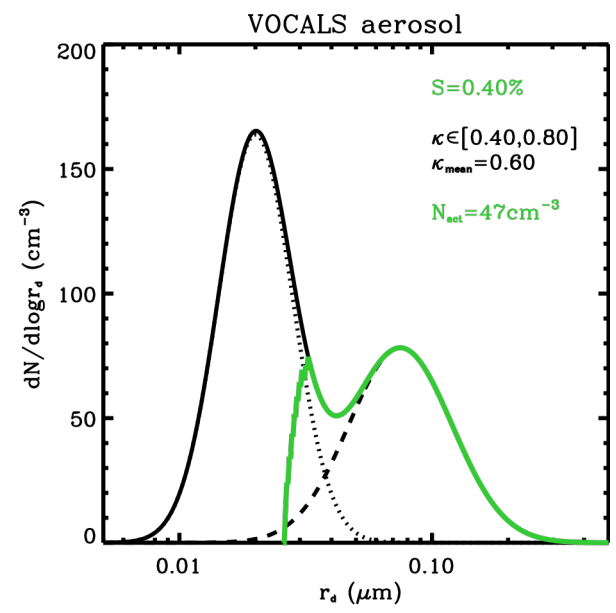


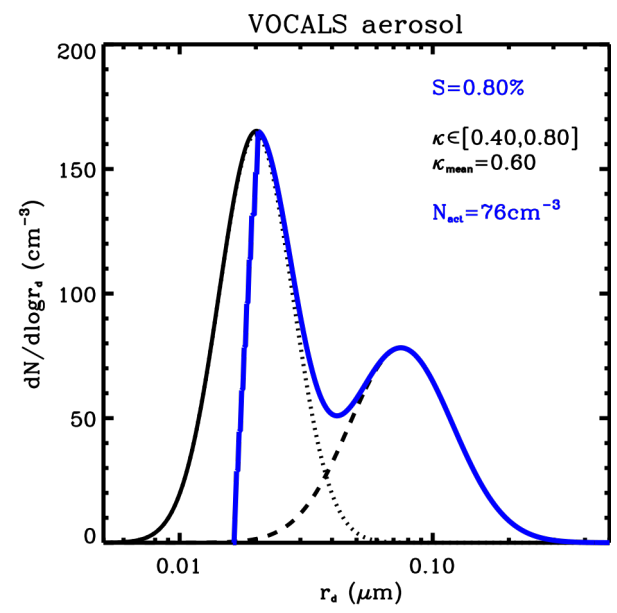

VOCALS aerosol
S=0.20%
κ∈[0.40,0.80]
κmean=0.60
Nact=34cm-3
n (cm-3μm-1)
rd (μm)


VOCALS aerosol
S=0.40%
κ∈[0.40,0.80]
κmean=0.60
Nact=47cm-3
n (cm-3μm-1)
rd (μm)


VOCALS aerosol
S=0.80%
κ∈[0.40,0.80]
κmean=0.60
Nact=76cm-3
n (cm-3μm-1)
rd (μm)


VOCALS aerosol
S=0.20%
κ∈[0.40,0.80]
κmean=0.60
Nact=34cm-3
dN/dlogrd (cm-3)
rd (μm)


VOCALS aerosol
S=0.40%
κ∈[0.40,0.80]
κmean=0.60
Nact=47cm-3
dN/dlogrd (cm-3)
rd (μm)


VOCALS aerosol
S=0.80%
κ∈[0.40,0.80]
κmean=0.60
Nact=76cm-3
dN/dlogrd (cm-3)
rd (μm)

ACTIVATION SPECTRUM

Quantifies the cloud-forming propensity of an aerosol population as a function of supersaturation, $N_{CCN}(S)$.

Represents the cumulative number concentration of particles activated into cloud droplets at or below a given supersaturation, S.

These spectra are experimentally determined using cloud condensation nuclei (CCN) chambers, in which low supersaturations (typically $S \leq 1 - 2\%$) can be precisely generated and controlled.

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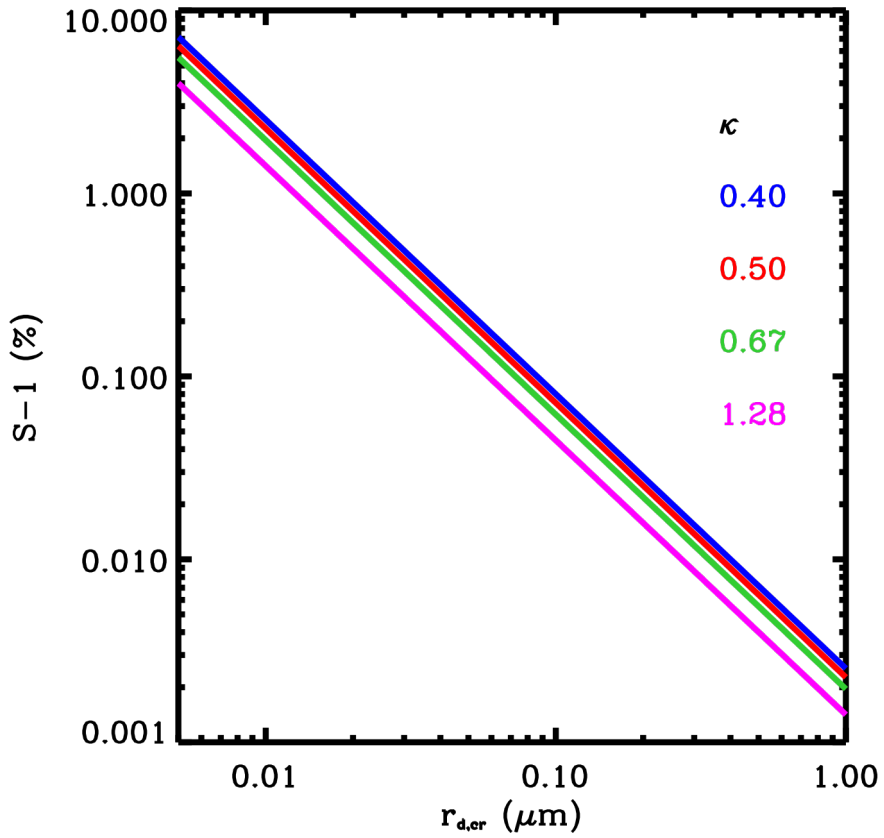
Activation spectrum

$$n(r_d) = \frac{N}{\sqrt{2\pi}\, r_d \ln\sigma} \exp\left(-\frac{\left(\ln r_d - \ln r_{d,m}\right)^2}{2\, ln^2\sigma}\right)$$

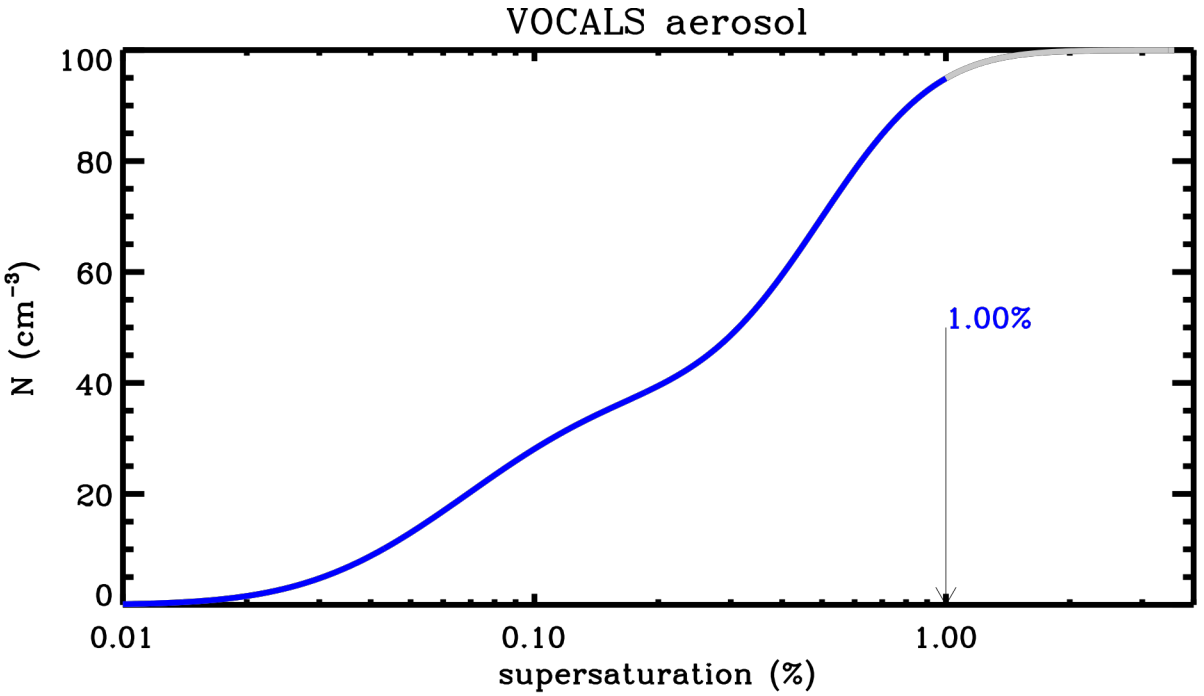
$$N_{CCN} = \int_{r_{d,cr}}^{\infty} n(r_d)dr_d = \frac{N}{2}\left(1 + \mathrm{erf}\frac{\ln {r_{d,m}}/{r_{d,cr}}}{\sqrt{2}\ln\sigma}\right)$$

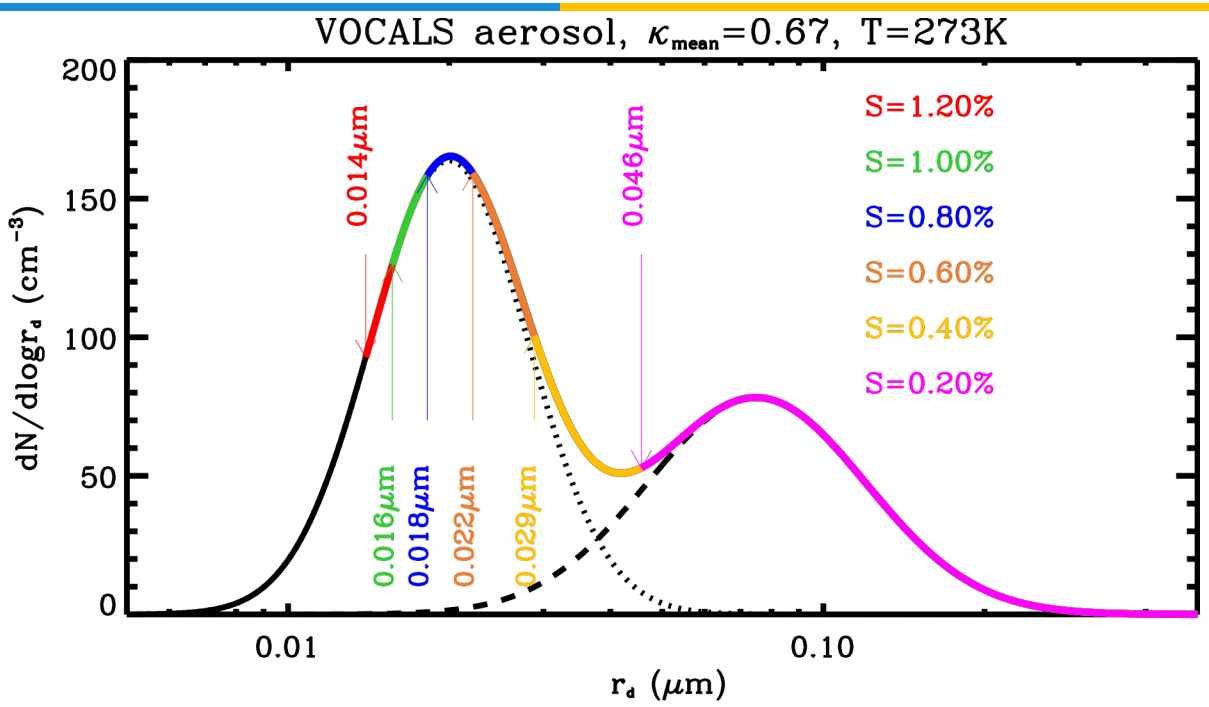
$$r_{d,cr} = \left(\frac{4A^3}{27\kappa(S-1)^2}\right)^{1/3}$$

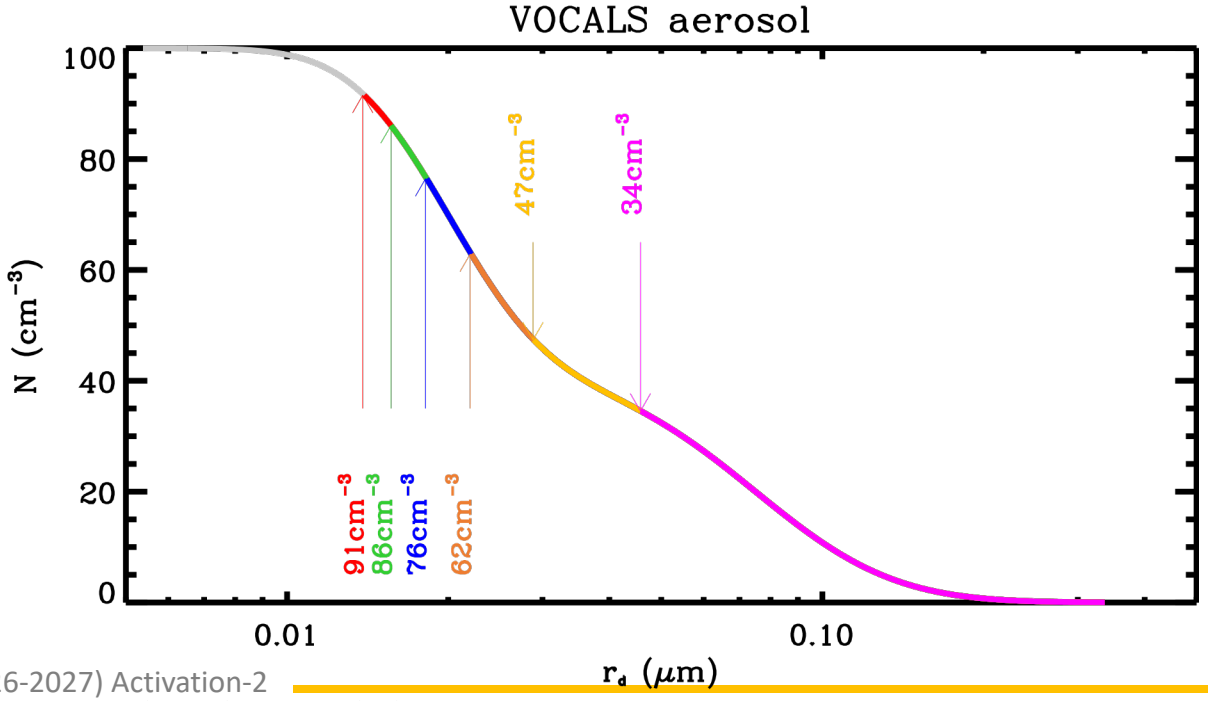
$$r_{d,cr} = \left(\frac{4A^3}{27\kappa(S-1)^2} \right)^{1/3}$$

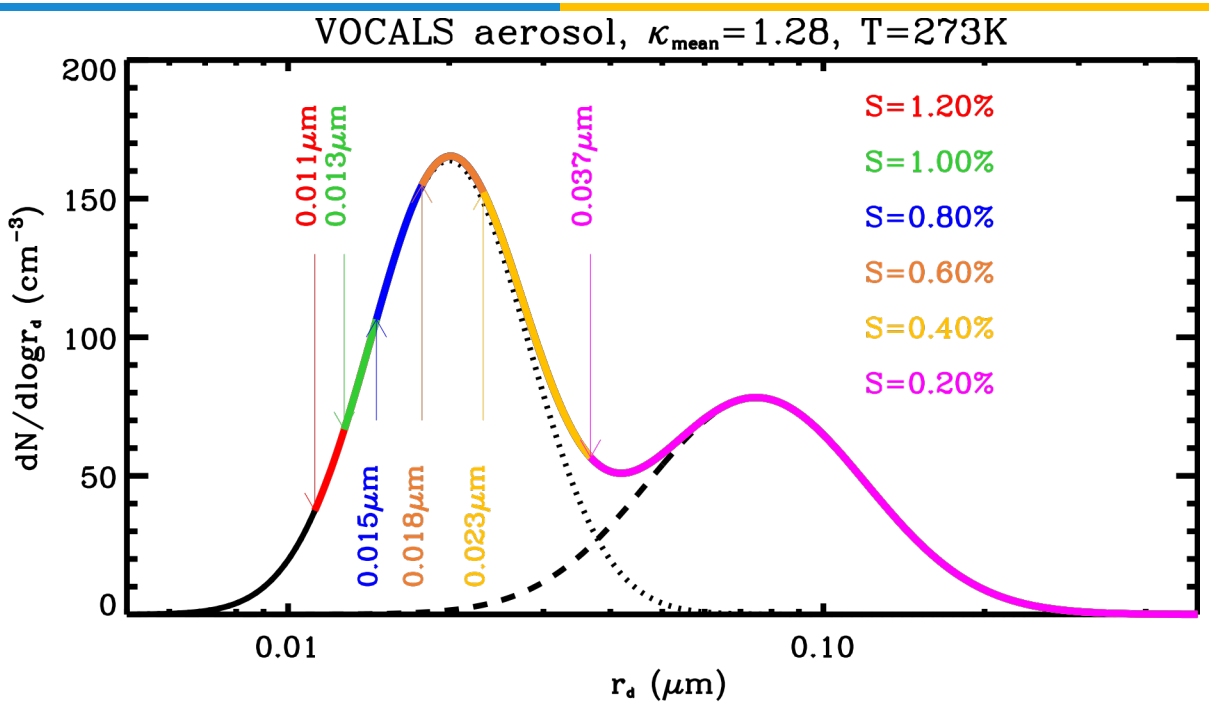


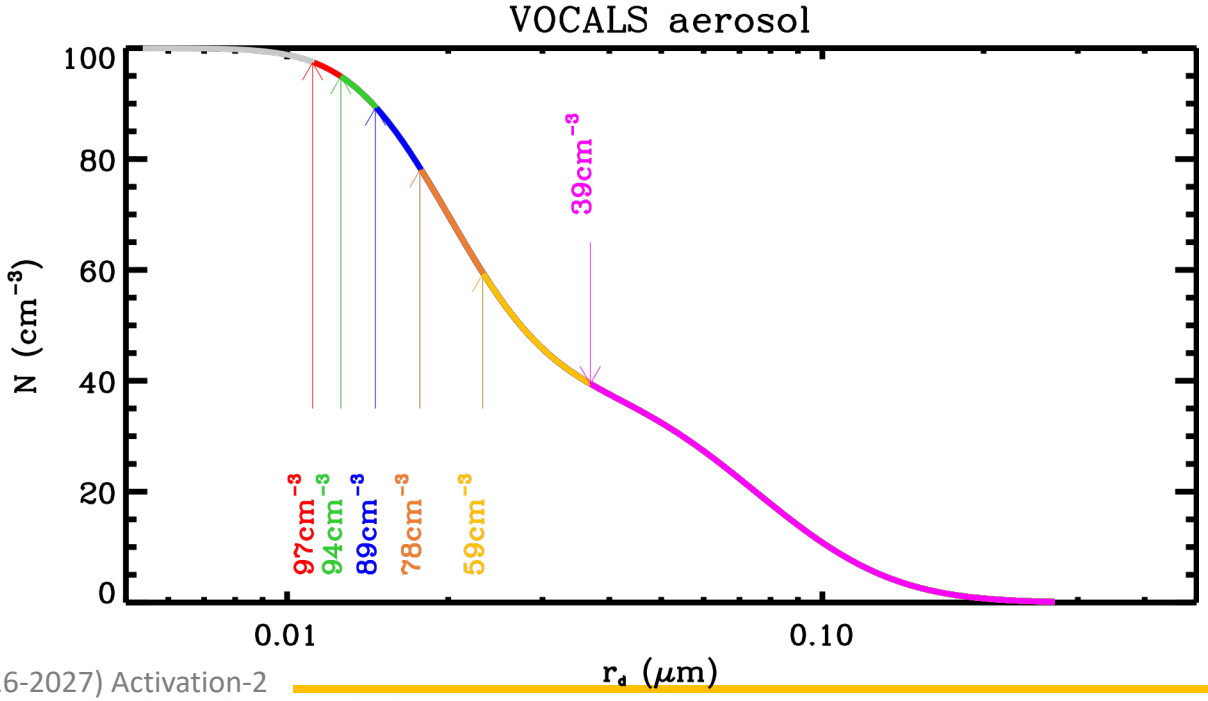
$$\int_{r_{d,cr}}^{\infty} n(r_d)dr_d = \frac{N}{2}\left(1 + \mathrm{erf}\frac{\ln {r_{d,m}}/{r_{d,cr}}}{\sqrt{2}\ln\sigma}\right)$$

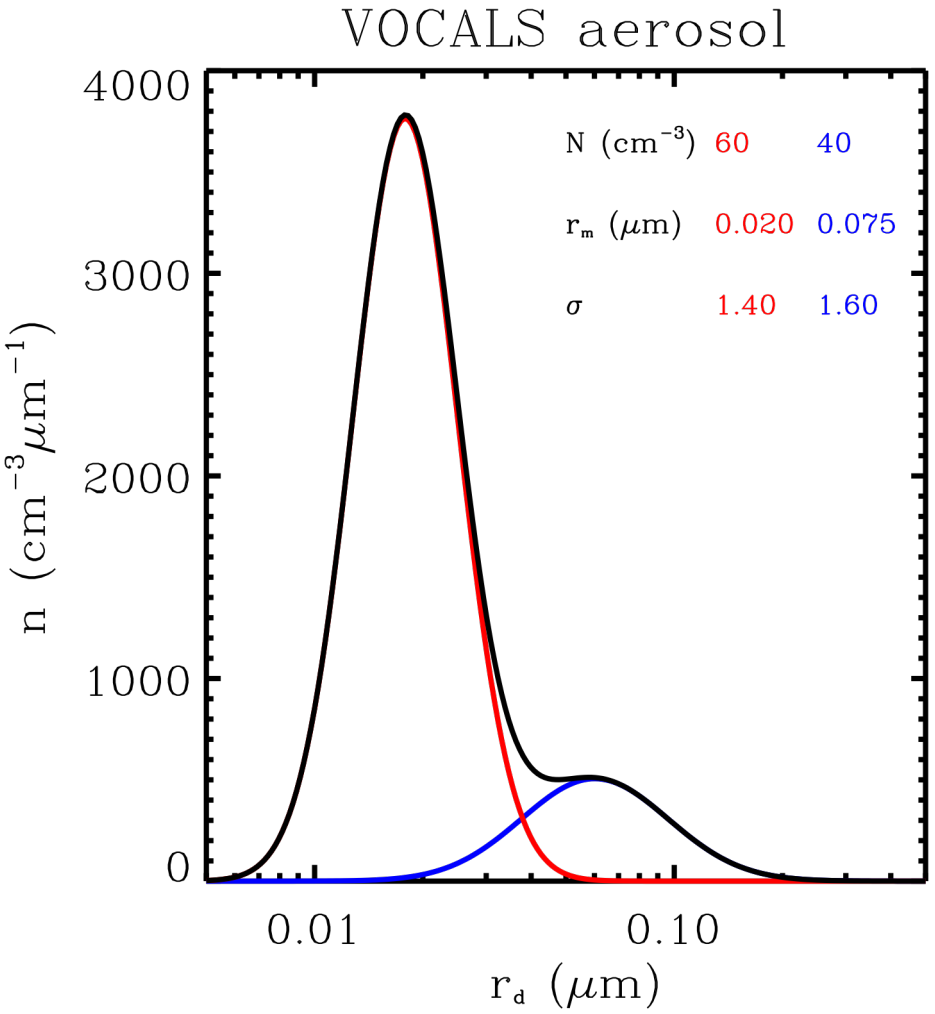


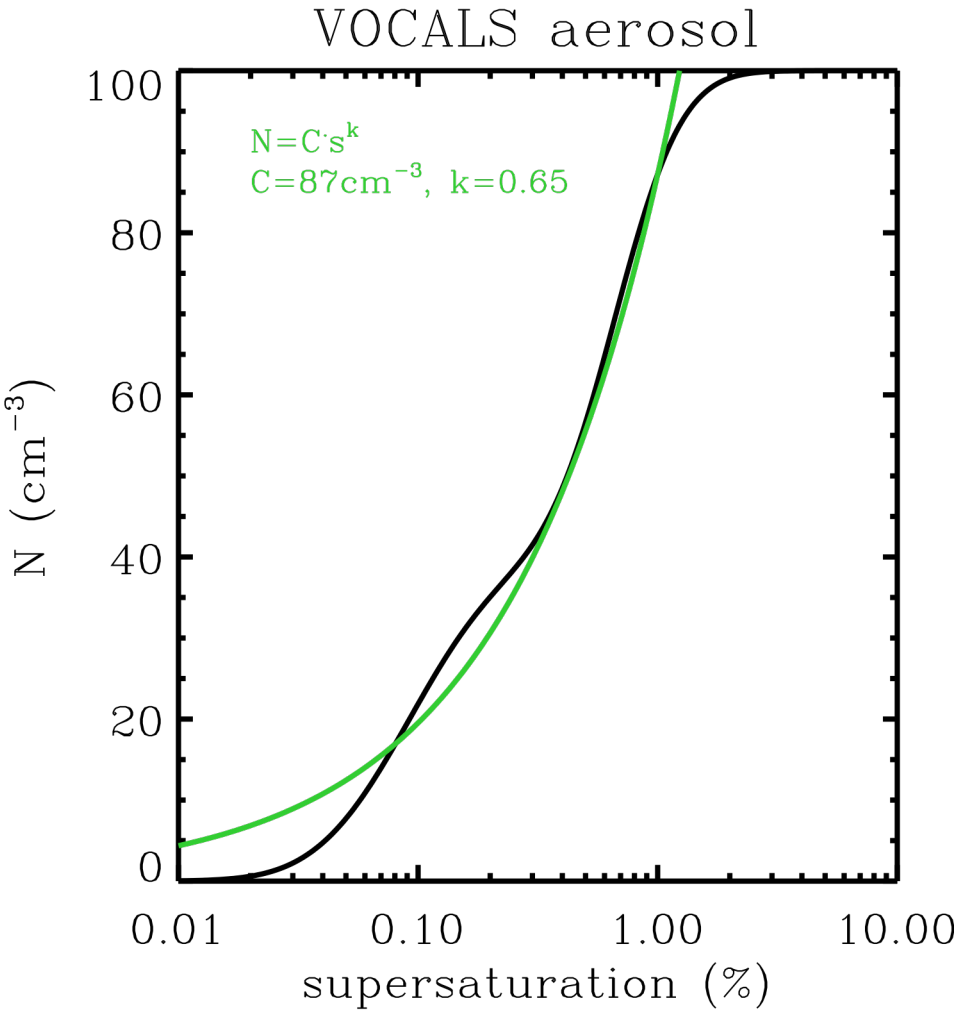

VOCALS aerosol, κ_{mean}=0.67, T=273K
dN/dlogr$_d$ (cm^{-3})
r$_d$ (μm)
0.01
0.10
0
50
100
150
200
0.014μm
0.046μm
0.016μm
0.018μm
0.022μm
0.029μm
S=1.20%
S=1.00%
S=0.80%
S=0.60%
S=0.40%
S=0.20%


VOCALS aerosol
N (cm^{-3})
r$_d$ (μm)
0.01
0.10
0
20
40
60
80
100
47cm^{-3}
34cm^{-3}
91cm^{-3}
86cm^{-3}
76cm^{-3}
62cm^{-3}


VOCALS aerosol, κ_{mean}=1.28, T=273K
dN/dlogr$_d$ (cm^{-3})
r$_d$ (μm)
0.011μm
0.013μm
0.015μm
0.018μm
0.023μm
0.037μm
S=1.20%
S=1.00%
S=0.80%
S=0.60%
S=0.40%
S=0.20%


VOCALS aerosol
N (cm^{-3})
r$_d$ (μm)
97cm^{-3}
94cm^{-3}
89cm^{-3}
78cm^{-3}
59cm^{-3}
39cm^{-3}

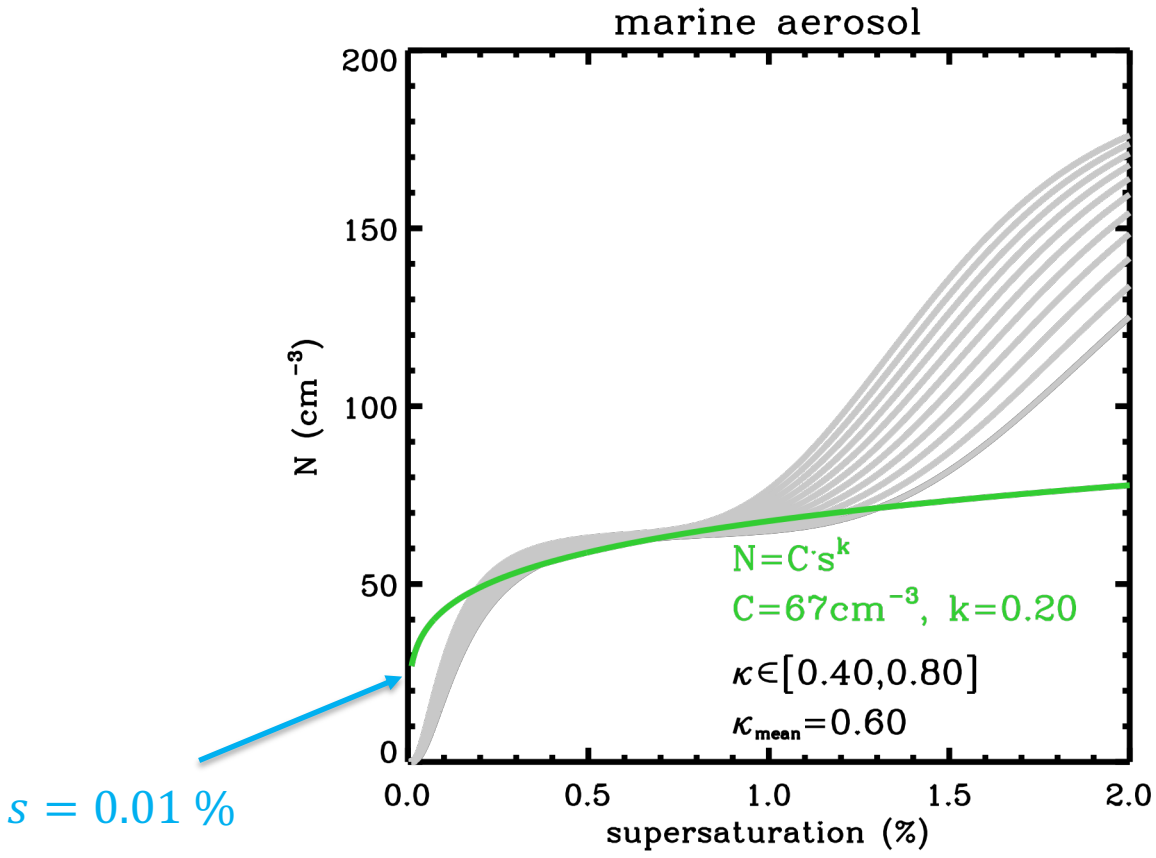




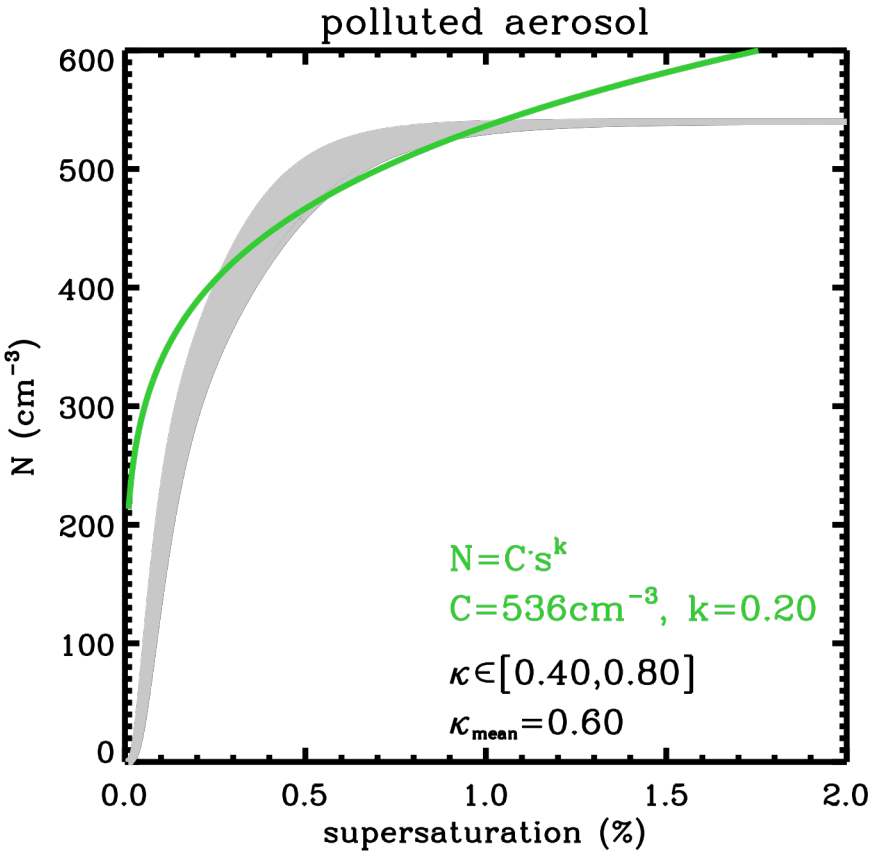
$$N = C \cdot S^k$$

C concentration of particles activated for S=1%

Activation spectrum



Activation spectrum



Activation – where it happens ?

- Activation occurs near cloud base as ascending updrafts drive adiabatic cooling and generate peak supersaturation, S_{max}.
- Under typical atmospheric conditions, CCN activation is confined to the first 30–50 m above cloud base. *However, in vigorous convective updrafts with vertical velocities on the order of* $10\ ms^{-1}$*, supersaturation can exceed 1%, sustaining activation higher into the cloud column.*
- The peak value of the supersaturation determines the fraction of available CCN that are activated. S_{max} dictates the activated fraction (N_{CCN}/N_{CN}), set by the competition between updraft cooling rates and water vapor depletion via diffusional droplet growth.
- Cloud droplet number concentration (N_d) is set by the aerosol activation spectrum $N_{CCN}(S)$ at peak supersaturation achieved during cloud formation.
  - *clouds forming in continental or polluted environments typically exhibit significantly higher droplet number concentrations than those developing in marine or pristine maritime environments, where aerosol background concentrations are substantially lower.*

Aerosol Characterization Experiment ACE2

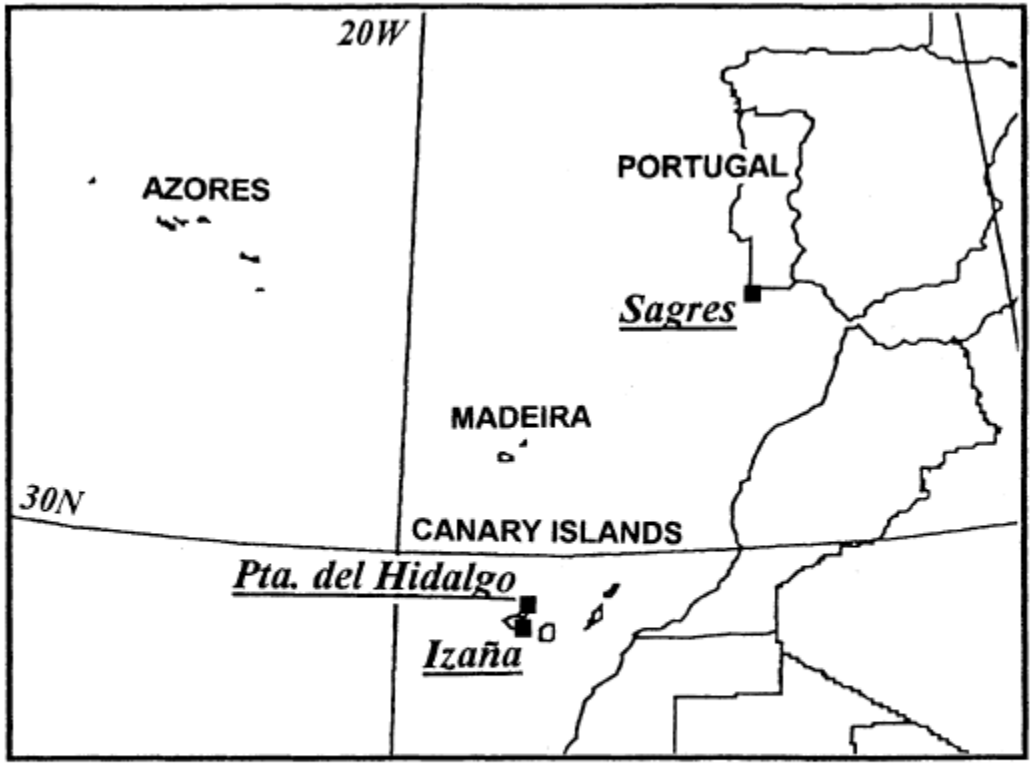
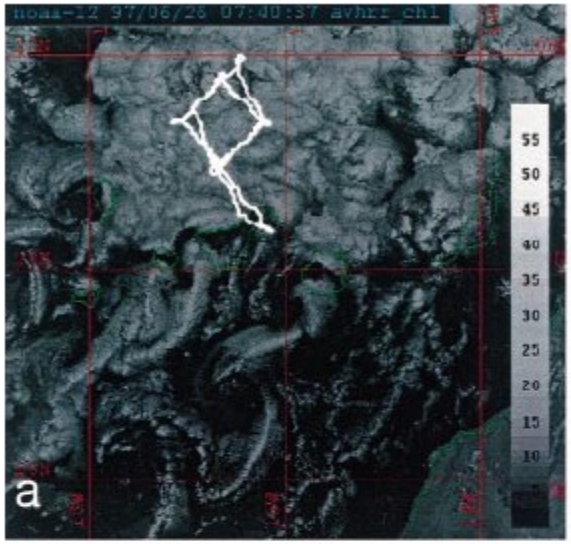
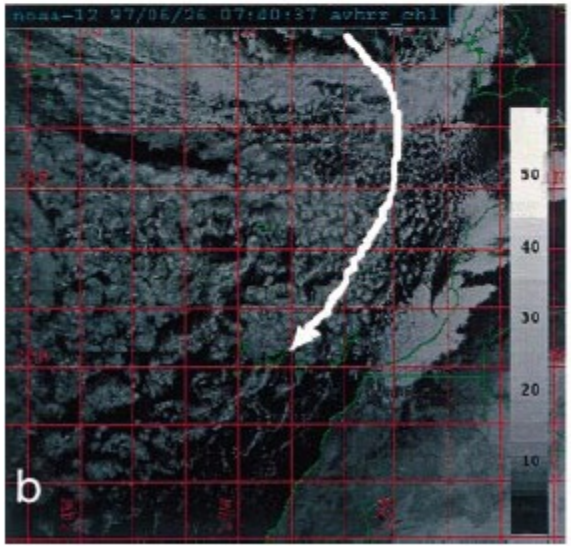


Fig. 1. The ACE-2 intensive area, with indication of the ground based measuring stations at Sagres (40 m asl), Punta del Hidalgo (50 m asl) and Izaña (2360 m asl).

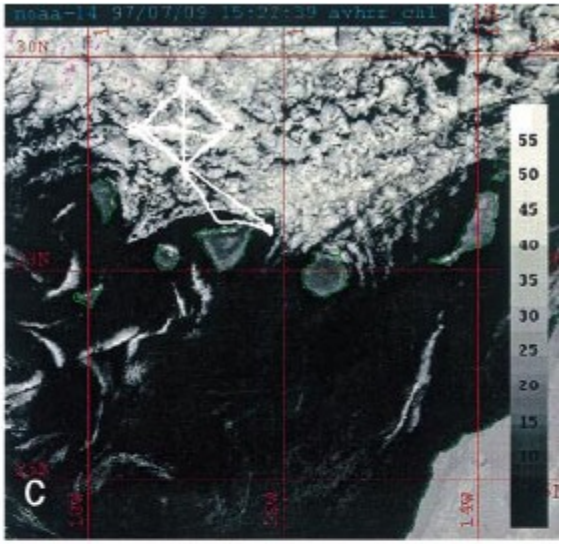
ACE2

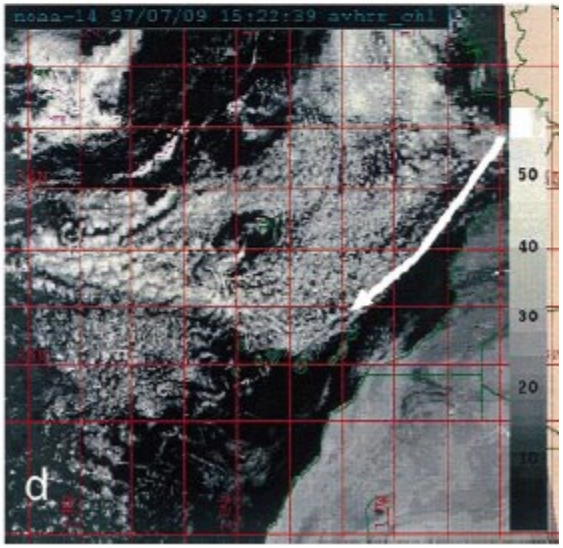
Pristine





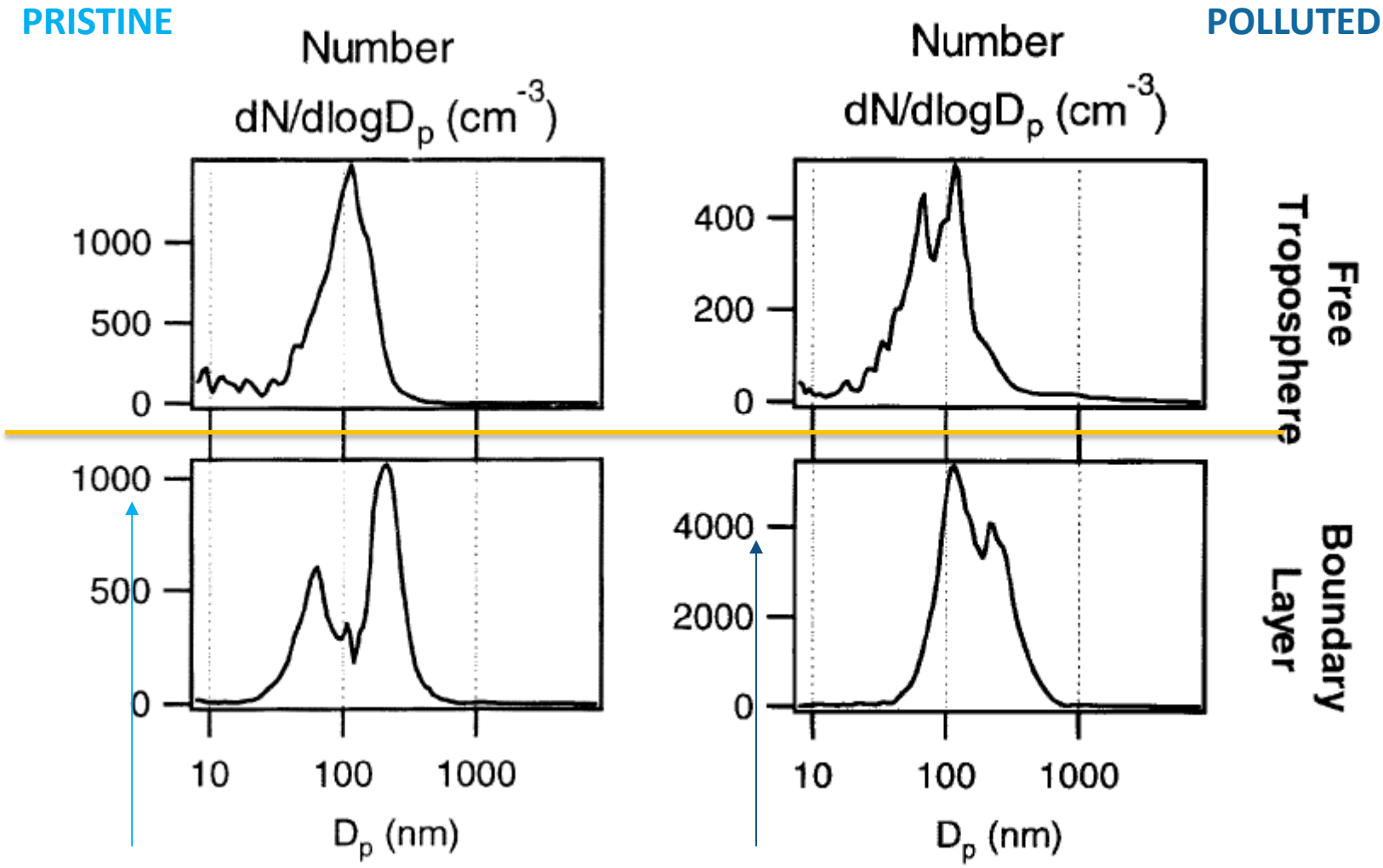
Polluted





Pawlowska, H., and J. L. Brenguier, 2000: Microphysical properties of stratocumulus clouds during ACE2, Tellus, vol. Vol. 52, Issue 2 , pp. 867-886

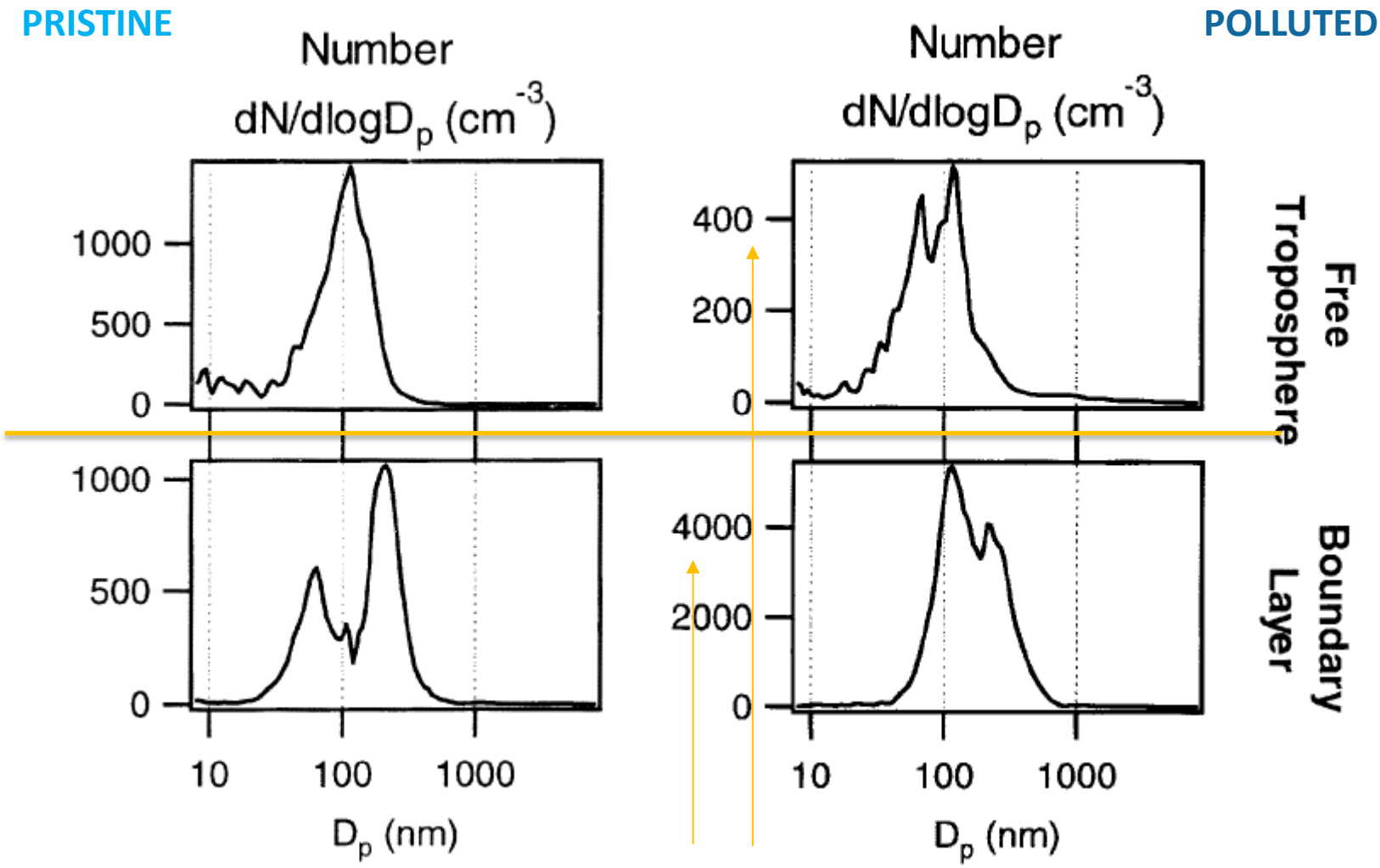
Aerosol



Collins et al., 2000: In-situ aerosol size distribution and clear-column radiative closure during ACE2. Tellus, vol. Vol. 52, Issue 2

More aerosols in the boundary layer in polluted case

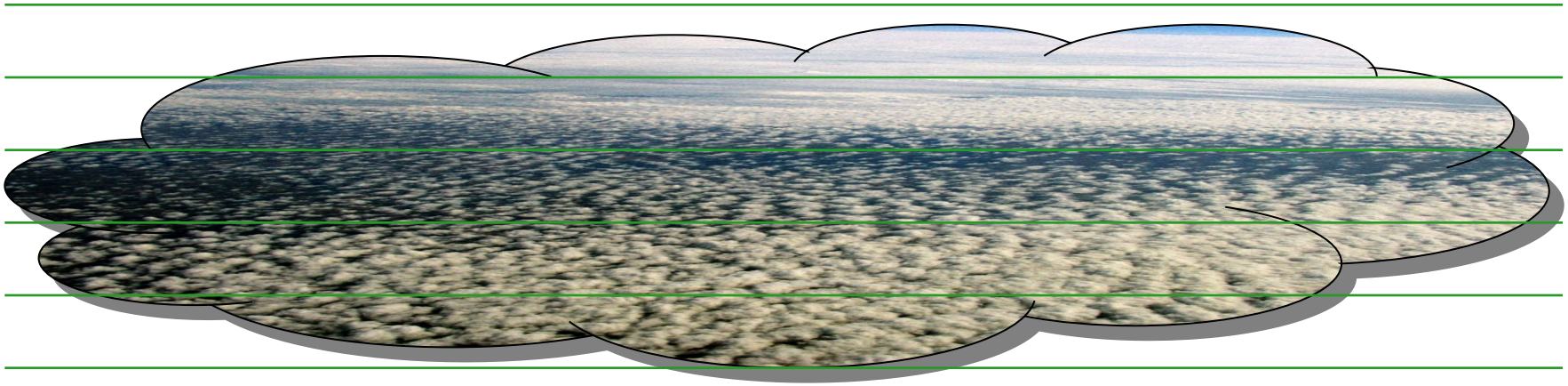
Aerosol



Collins et.al. 2000

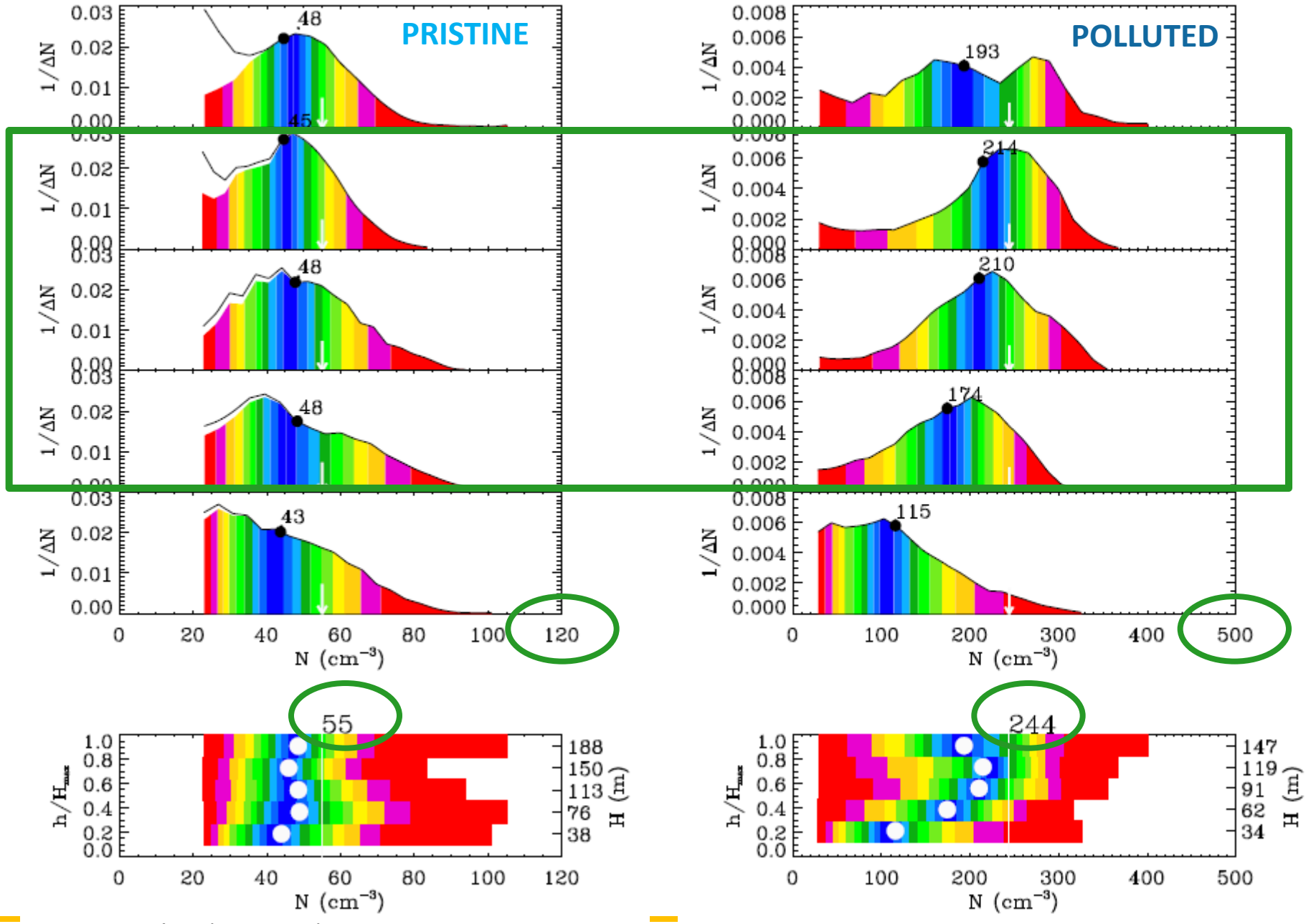
More aerosol in the boundary layer compared to the free troposphere

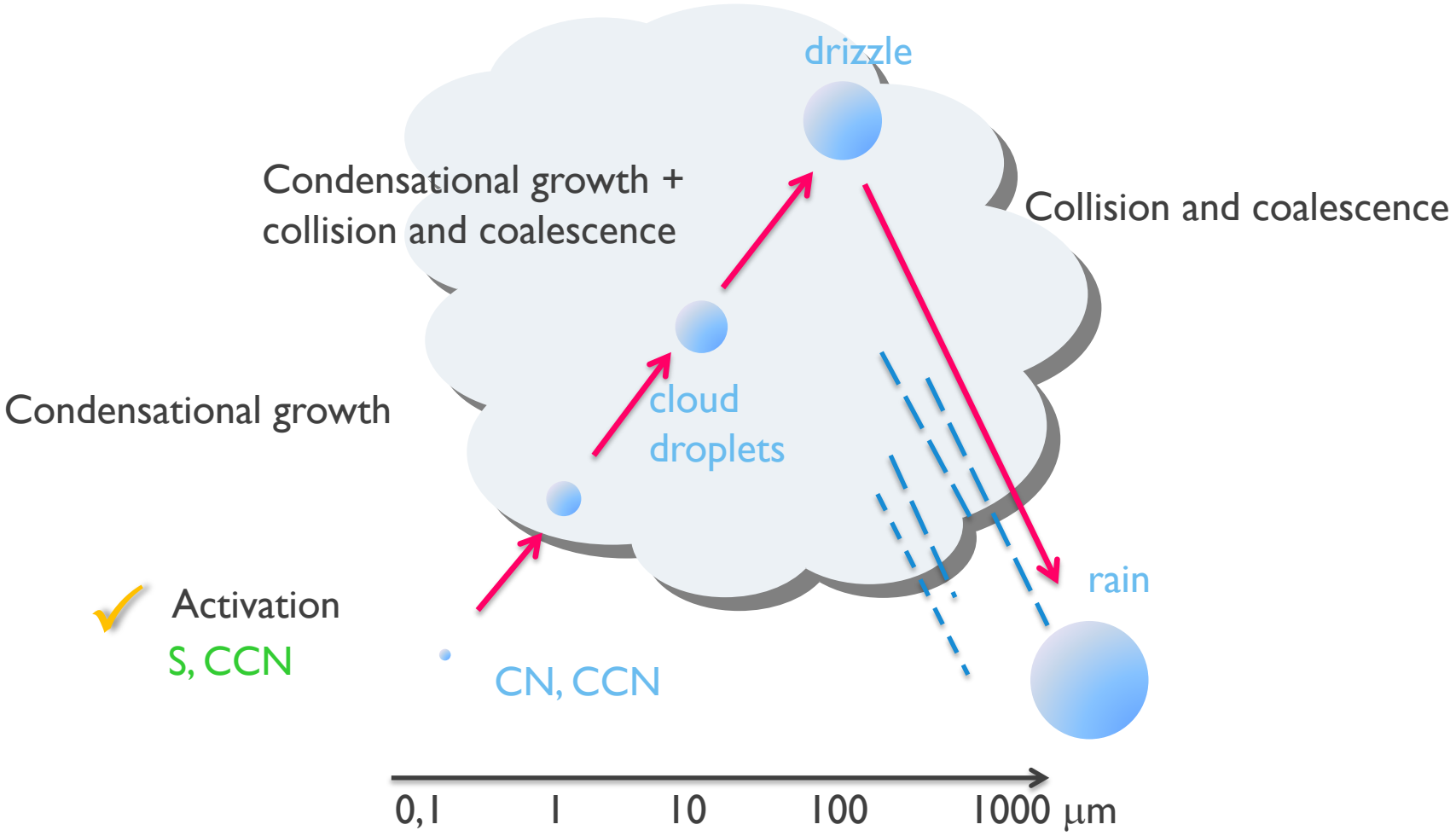
Second Aerosol Characterization Experiment (ACE2) June-July 1997, Stratocumulus clouds over the Atlantic



Cloud divided into 5 layers.
Cloud droplet concentration reflects fairly well the activation process at the cloud base.

Cloud droplets number concentration




drizzle
Condensational growth +
collision and coalescence
Collision and coalescence
Condensational growth
cloud
droplets
rain
Activation
S, CCN
CN, CCN
0,1
1
10
100
1000 μm