

Aerosol life cycles: what can we learn from high altitude sites?

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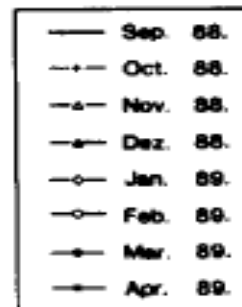
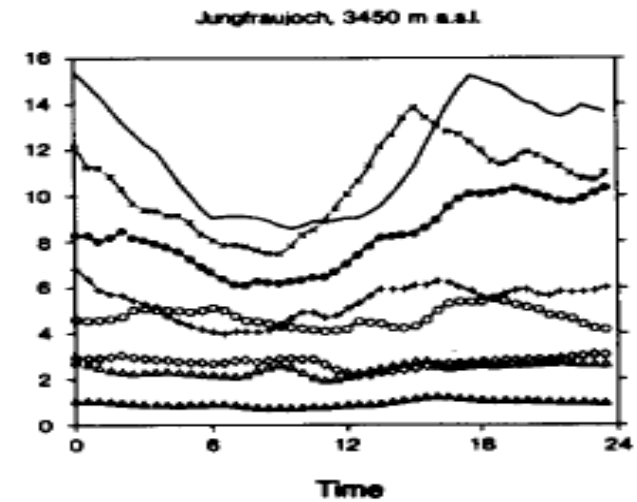
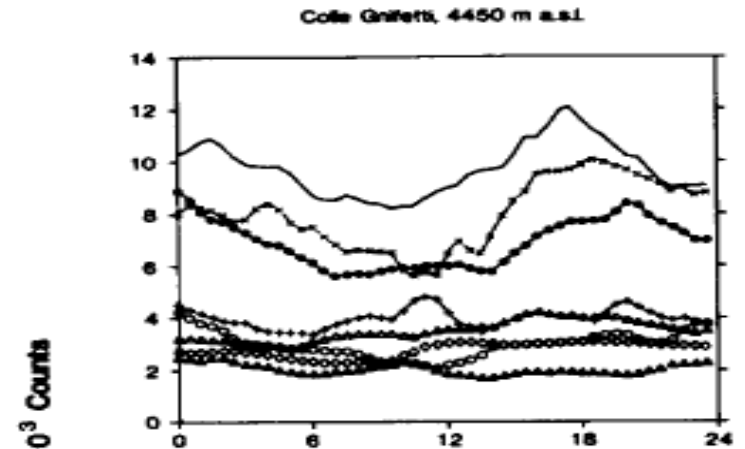
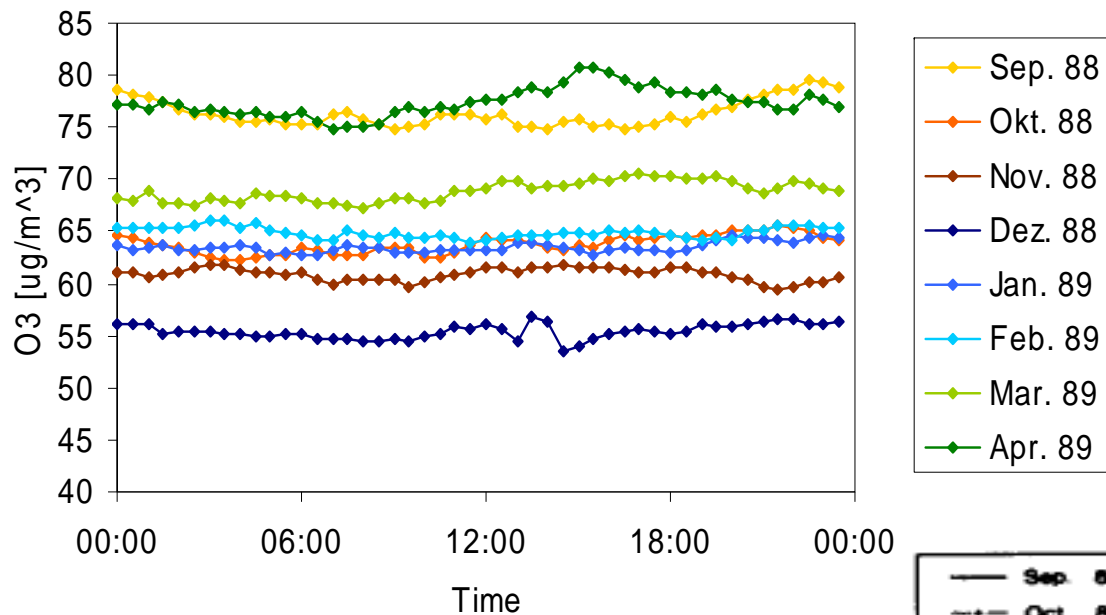
AeroCom Workshop
Hamburg, 23 – 27 September 2013

Research questions

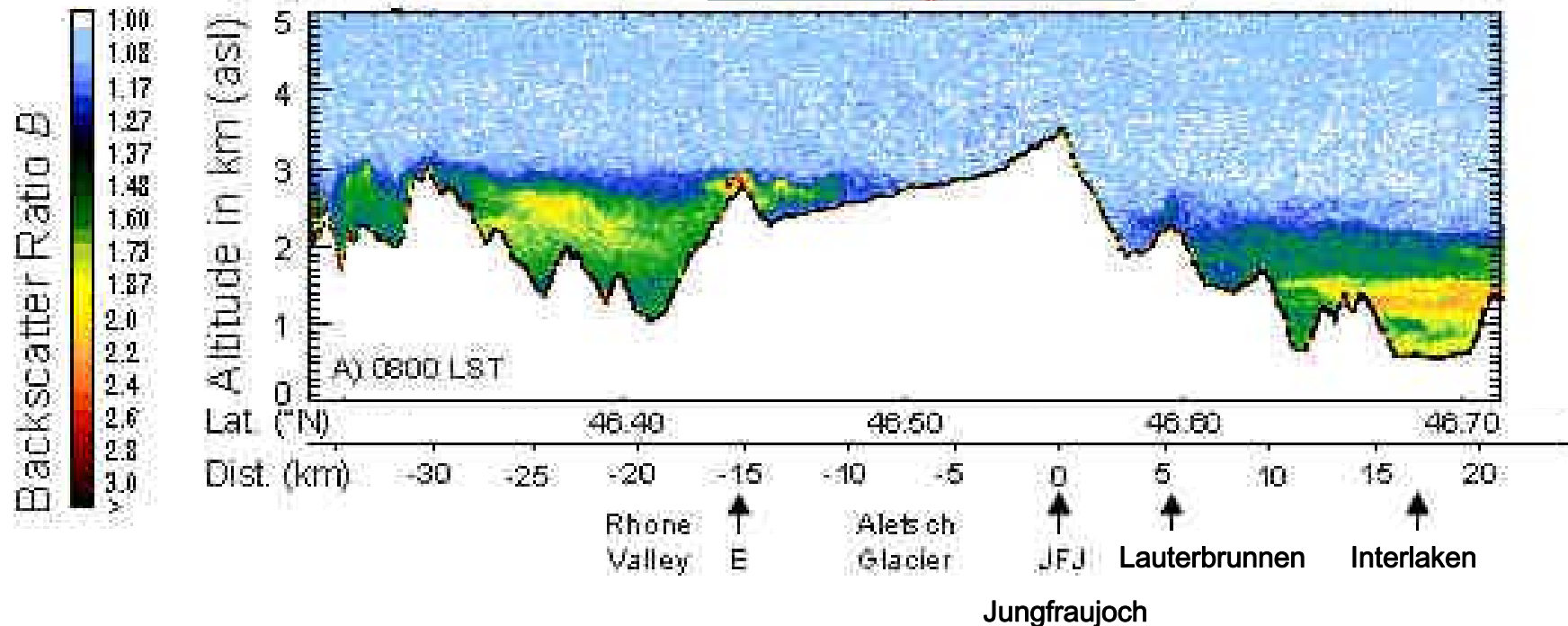
- Topography enhanced vertical transport
- Short lifetime, high spatial variability
- Nucleation
- Away from sources, enabling studies of aged aerosols (physical, optical and chemical properties, source apportionment..)
- If station in cloud: aerosol-cloud interaction, liquid and mixed-phase clouds

Vertical transport does reach the highest mountain tops in Europe

Before, it was believed, based on ozone data, that such sites are always in the free troposphere



Confirmation by airborne lidar



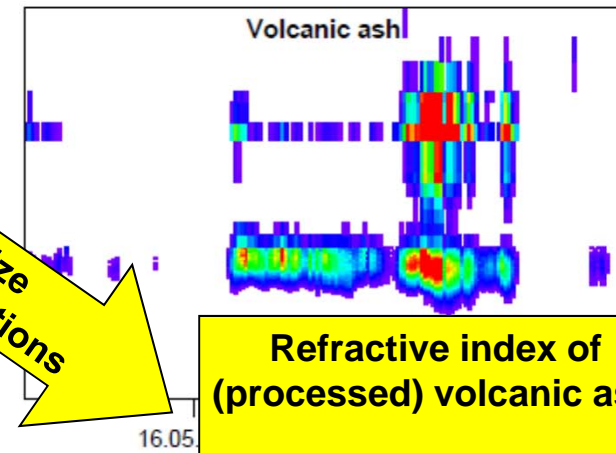
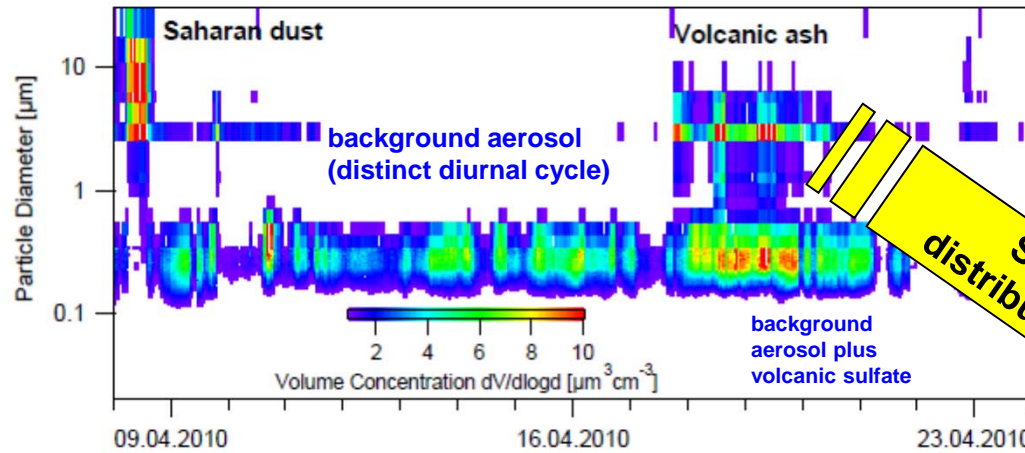
A wide variety of aerosol variables are needed for a full description of the atmospheric aerosol (Recommendations of the GAW Aerosol SAG)

- Multiwavelength optical depth
- Mass in two size fractions
- Major chemical components in two size fractions
- Scattering and hemispheric backscattering coefficient at various wavelengths
- Absorption coefficient
- Aerosol number concentration
- Cloud condensation nuclei (at various supersaturations)
- Aerosol size distribution
- Detailed size fractionated chemical composition
- Dependence on relative humidity
- Vertical distribution of aerosol properties (e.g. LIDAR)

High altitude stations typically provide these properties for an aged aerosol above a continental area, without the influence of a local source

Refractive index and mass extinction efficiency of volcanic ash

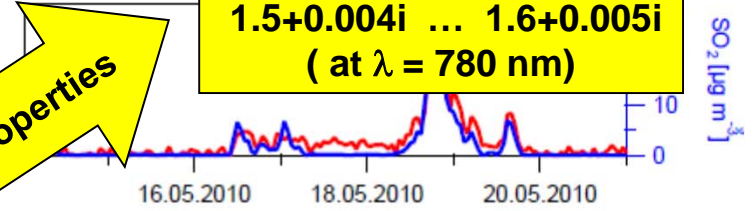
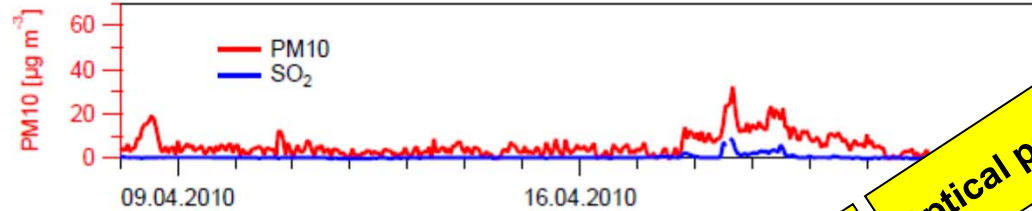
Volume size distribution



Size distributions

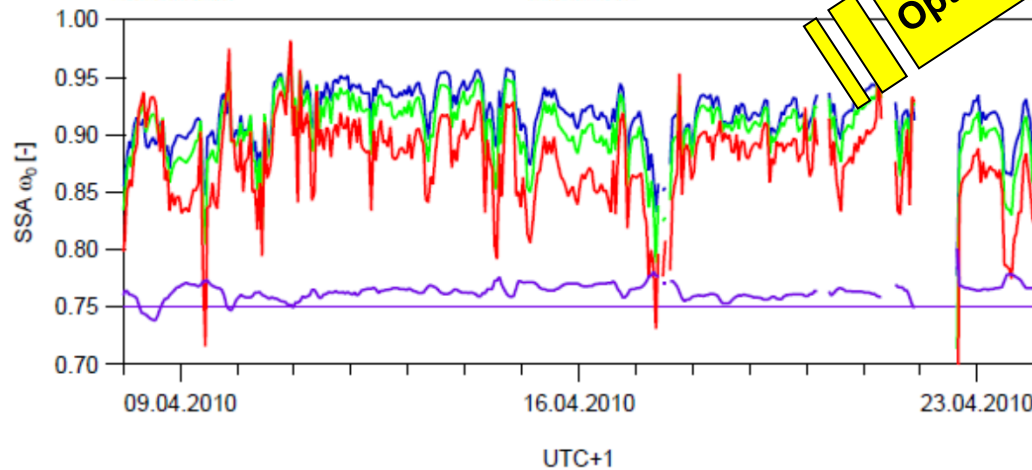
Refractive index of (processed) volcanic ash:
 $1.5+0.004i \dots 1.6+0.005i$
(at $\lambda = 780 \text{ nm}$)

PM₁₀

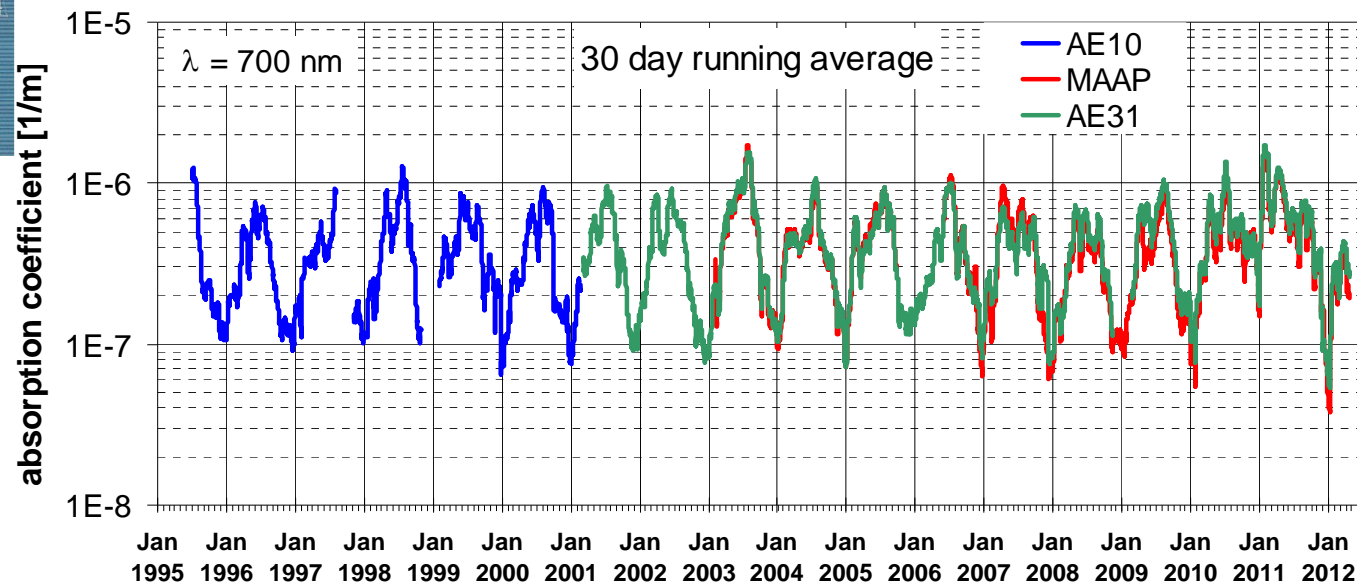
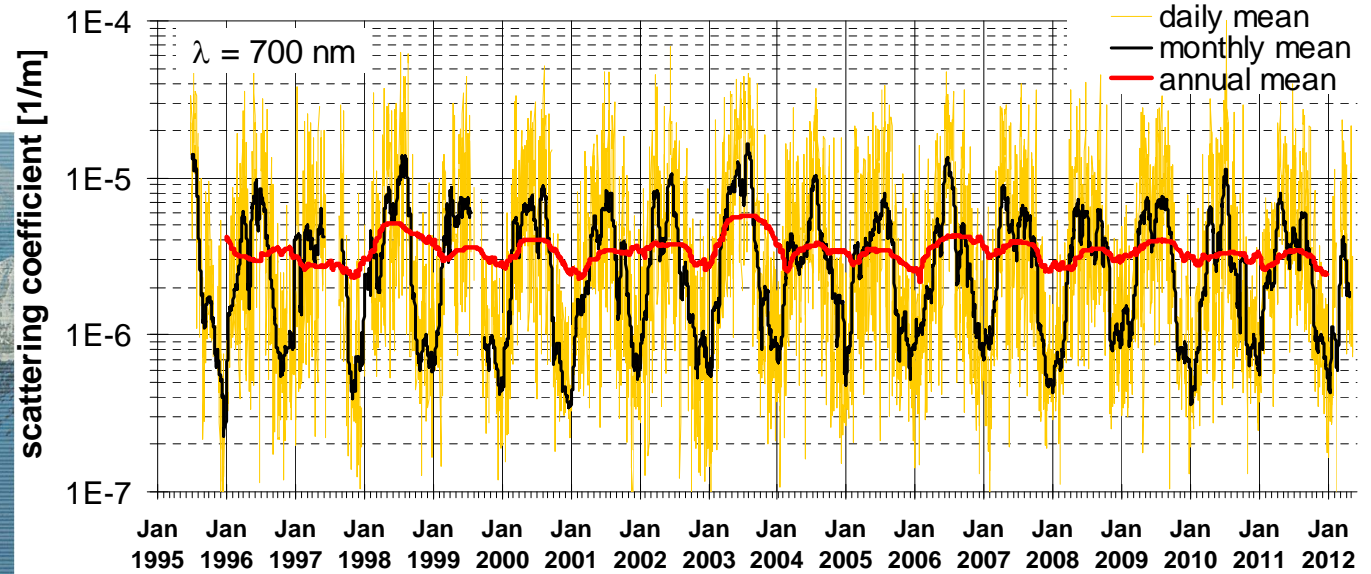
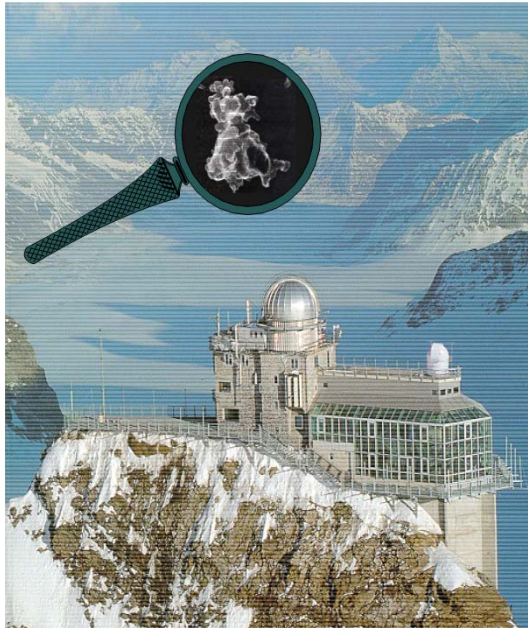


Optical properties

Single Scattering Albedo



18 years of continuous data at the Jungfrauoch (3580 m asl)

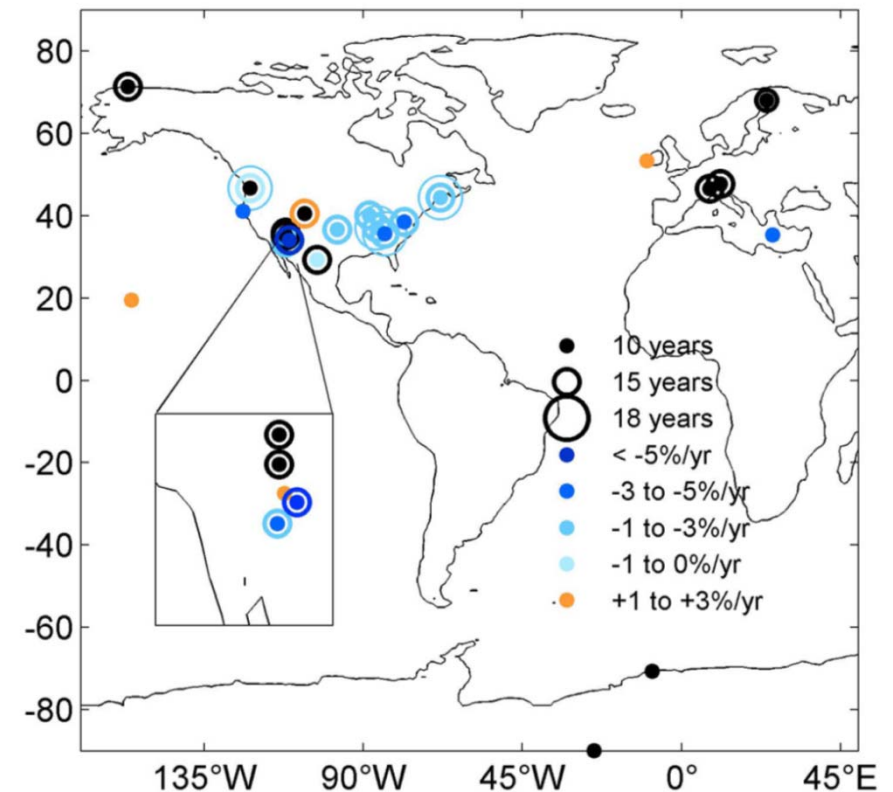


There are only very few stations with long-term measurements of climate relevant properties
 → we need more stations with long-term vision to better assess the impact of aerosols on climate



The ,global' stations of the Global Atmosphere Watch program of the World Meteorological Organization

Trends of the aerosol light scattering coefficient

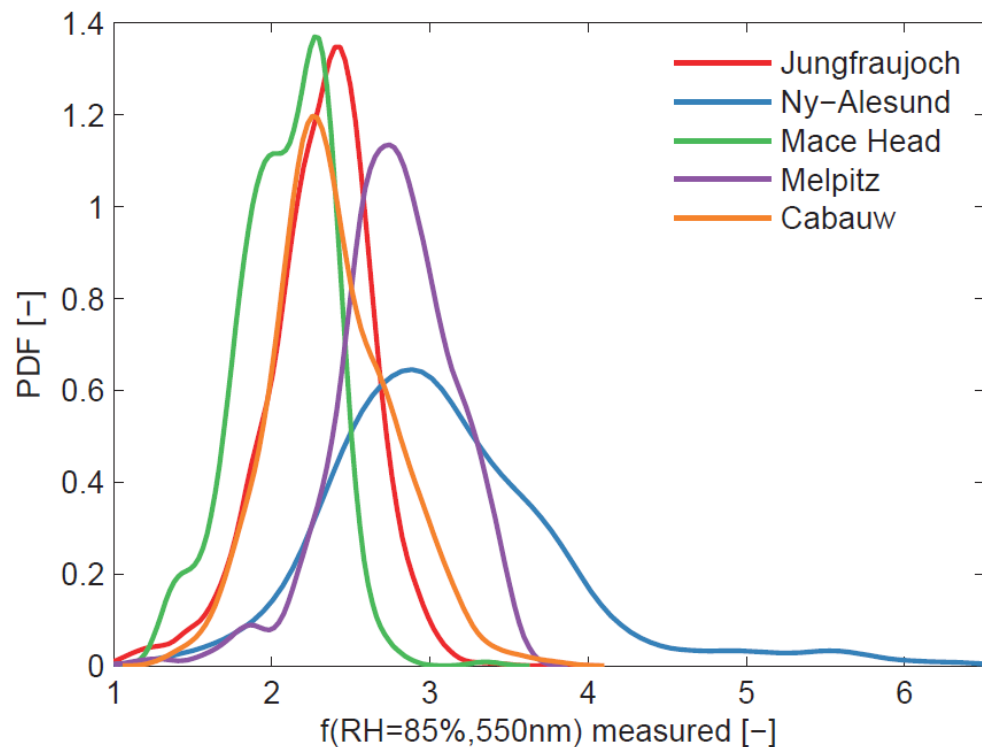


Collaud Coen et al., ACP 2013

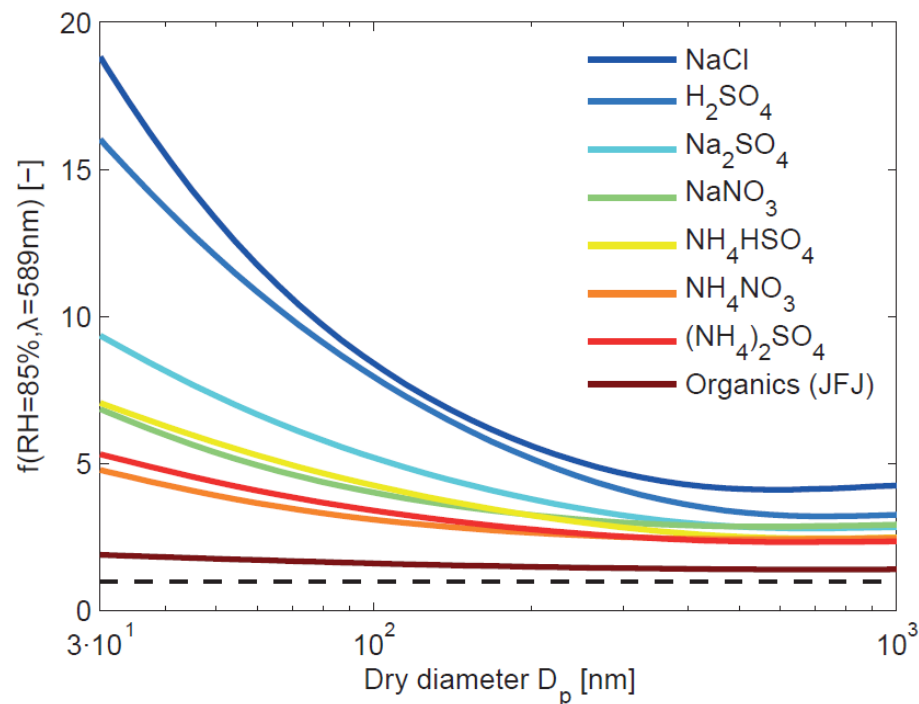
The ambient single scattering albedo requires the ambient scattering coefficient

The scattering enhancement depends on size and chemistry

Scattering enhancement at different sites (at RH = 85%)



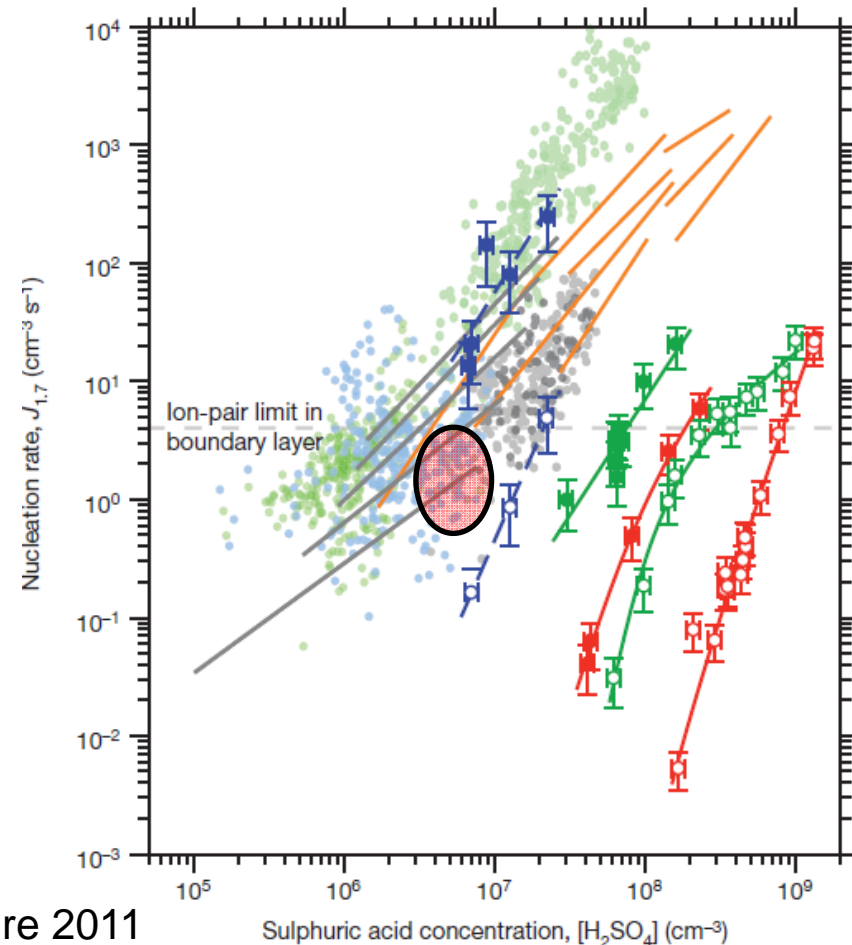
Dependence on size and chemistry



Nucleation occurs frequently in the lower free troposphere, mechanisms still largely unknown

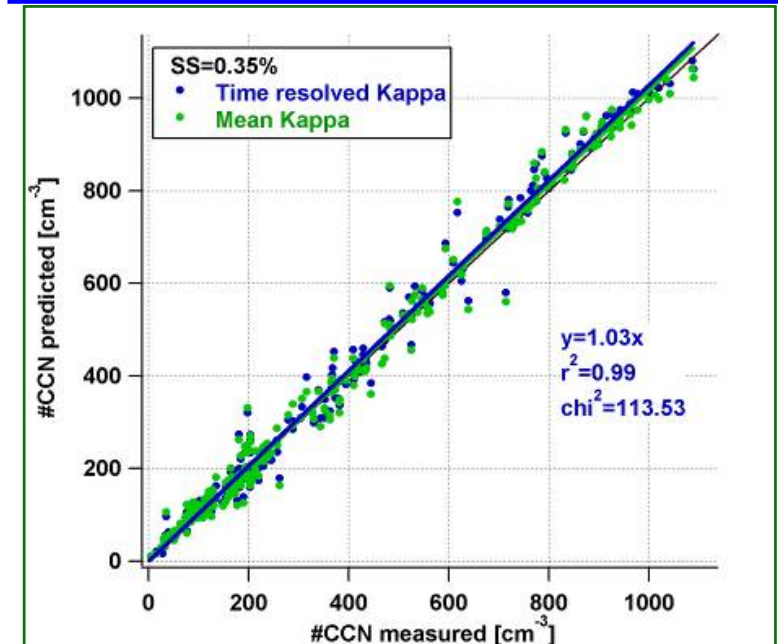
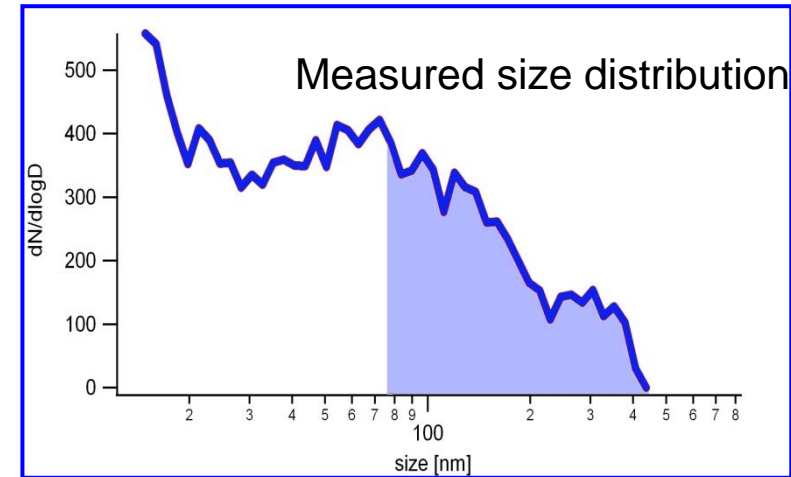
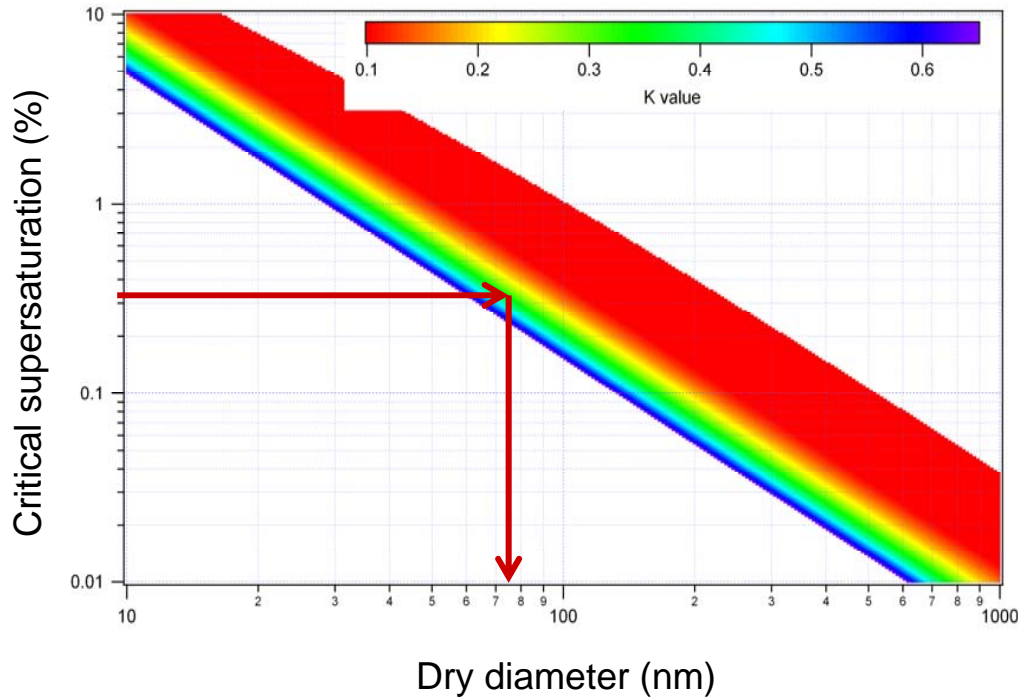
Nucleation rates from CLOUD:
 red: 292 K, green: 278 K, blue: 248 K
 open: pure sulphuric acid; filled: with NH_3

- J_3 nucleation rates around $1 \text{ cm}^{-3} \text{ s}^{-1}$
- Sulfuric acid around $5 \cdot 10^6 - 10^7 \text{ cm}^{-3}$
- Nucleation rates cannot be explained by pure sulfuric acid, even at low T
- Also with NH_3 still difficult at Jungfraujoch temperature



Kirkby et al., Nature 2011

CCN number concentrations by direct measurement and indirectly from size and hygroscopicity measurements

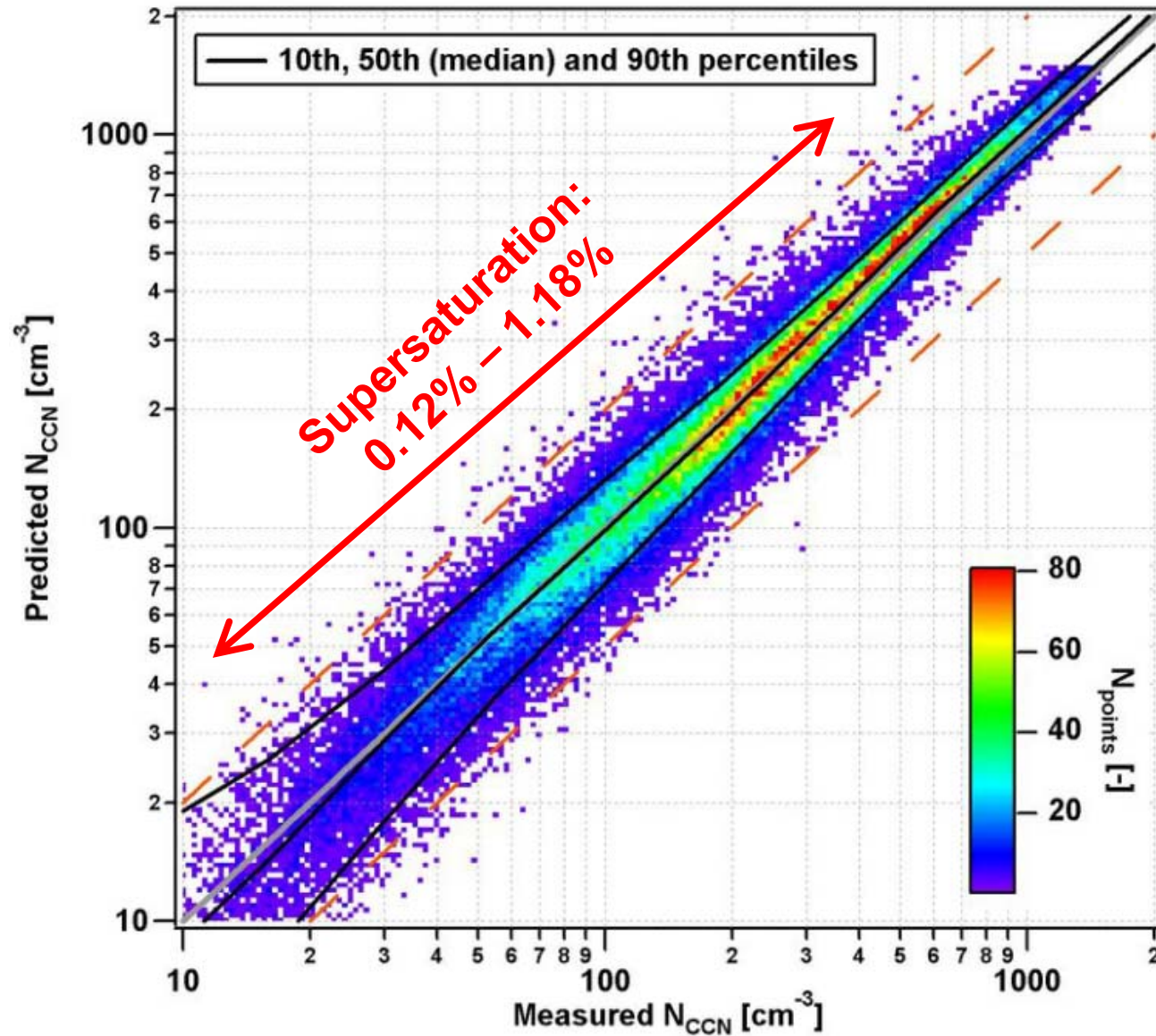


In simple cases, size and chemistry allow to determine CCN number concentration:
Comparison of measured and predicted CCN concentration for the Jungfrauoch



Prediction of the CCN concentration

Prediction only uses $N(D)$ and assumes constant chemical composition in time

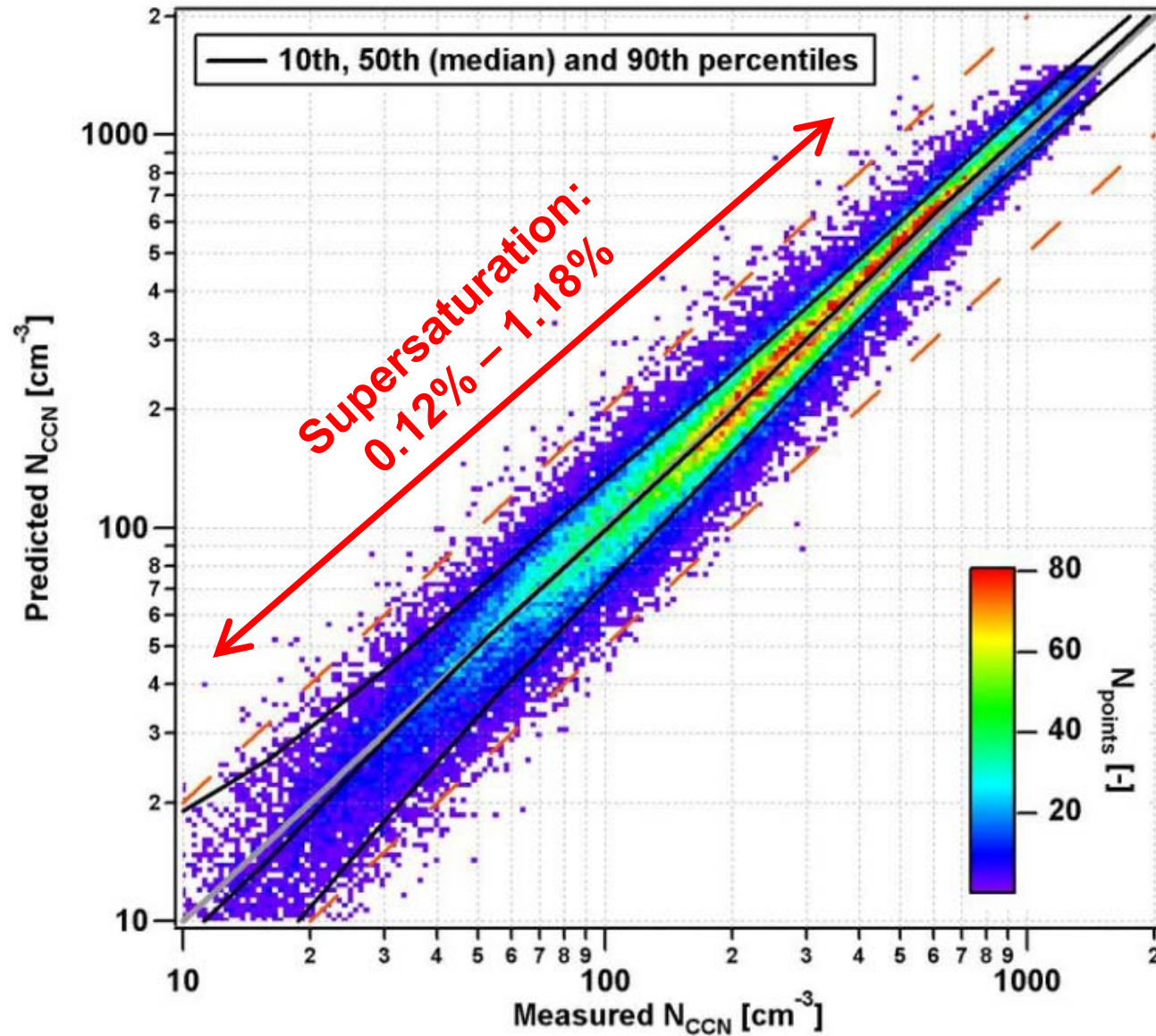


This is great news for the modellers:

With a size distribution and a chemical composition (κ) we can predict the number of CCN for a wide range of supersaturations

Prediction of the CCN concentration

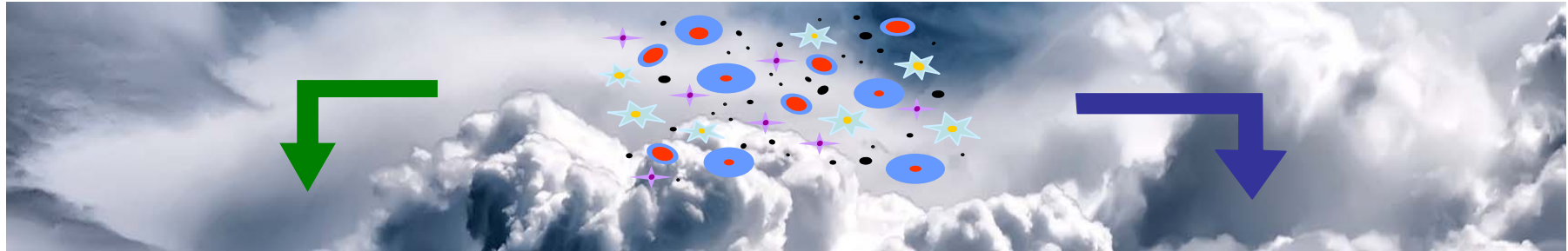
Prediction only uses $N(D)$ and assumes constant chemical composition in time



BUT:

We do not know the supersaturation in the ambient cloud
 → Can we measure it ?

CLACE campaigns



Interstitial inlet:
(no activated particles)
removes :
- droplets
- ice crystals

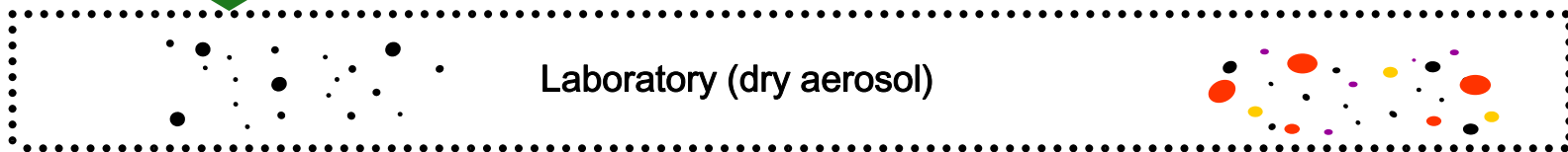
(Size <math>< 2 \mu\text{m}</math>)



- PVM and/or FSSP/FM
- Meteo (T, RH, P, Wind)



**Total inlet :
heated inlet**

• SMPS

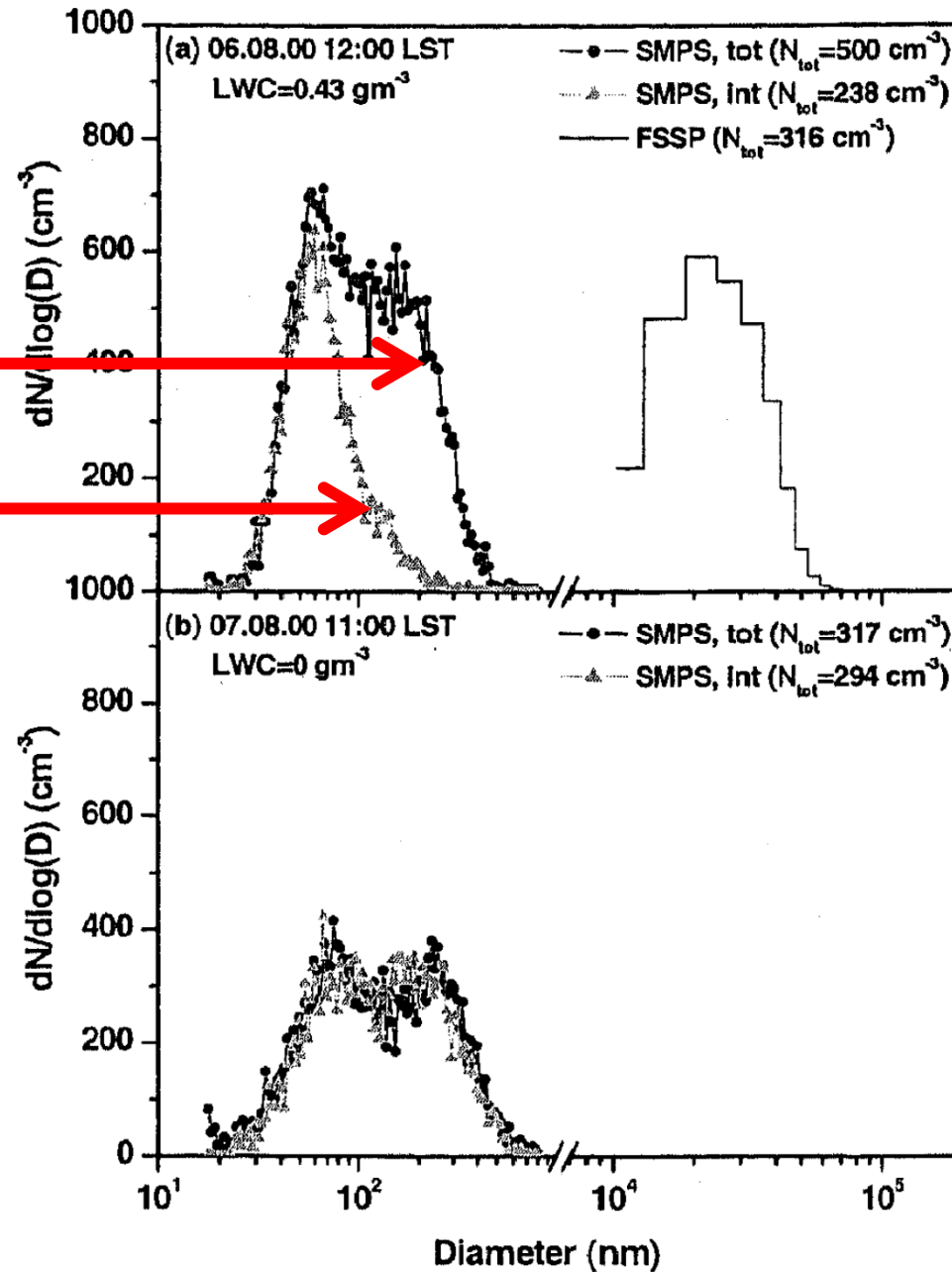
Laboratory (dry aerosol)

• SMPS
• DPM

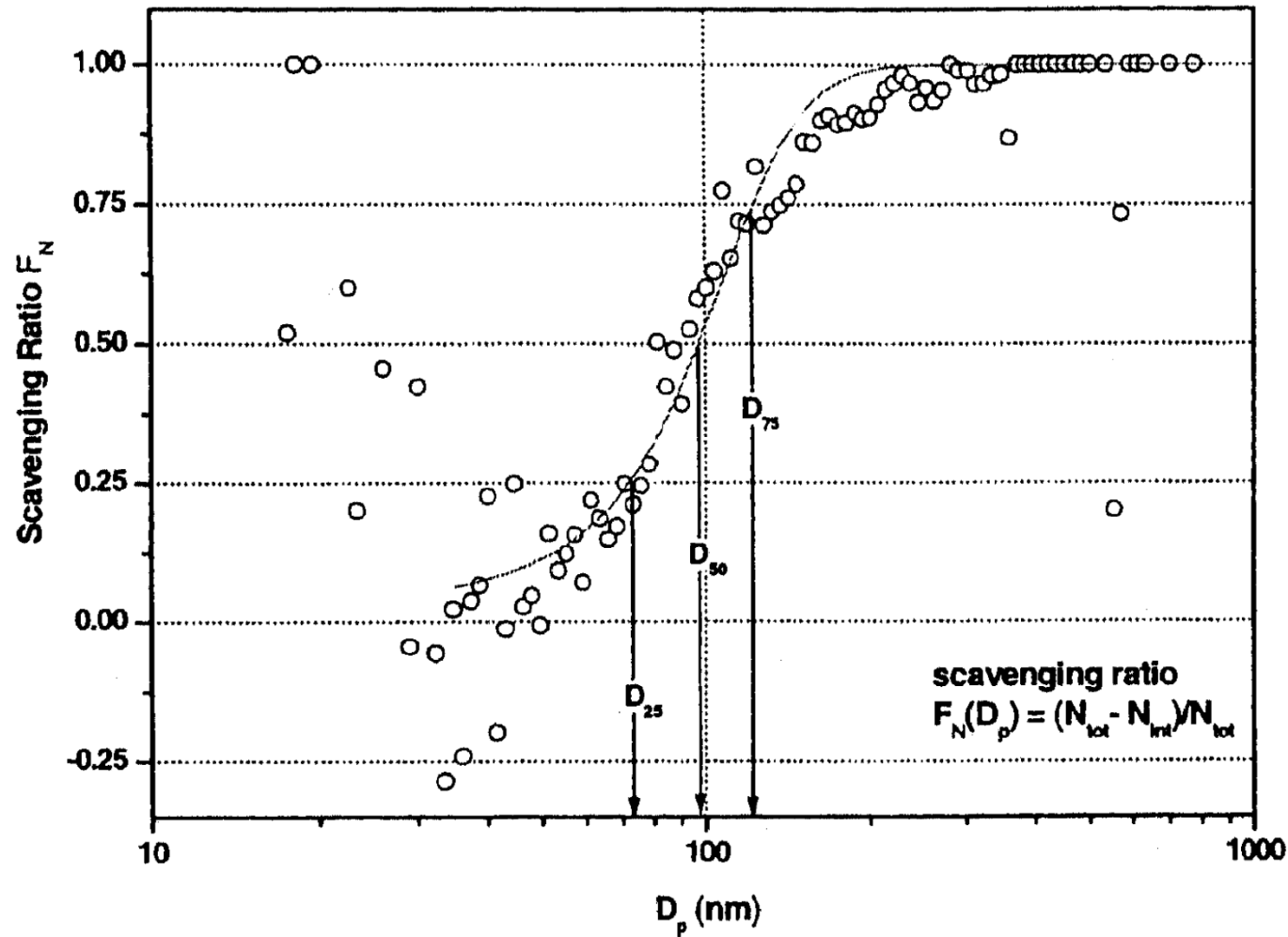
Size distributions in and out of cloud

Total

Interstitial

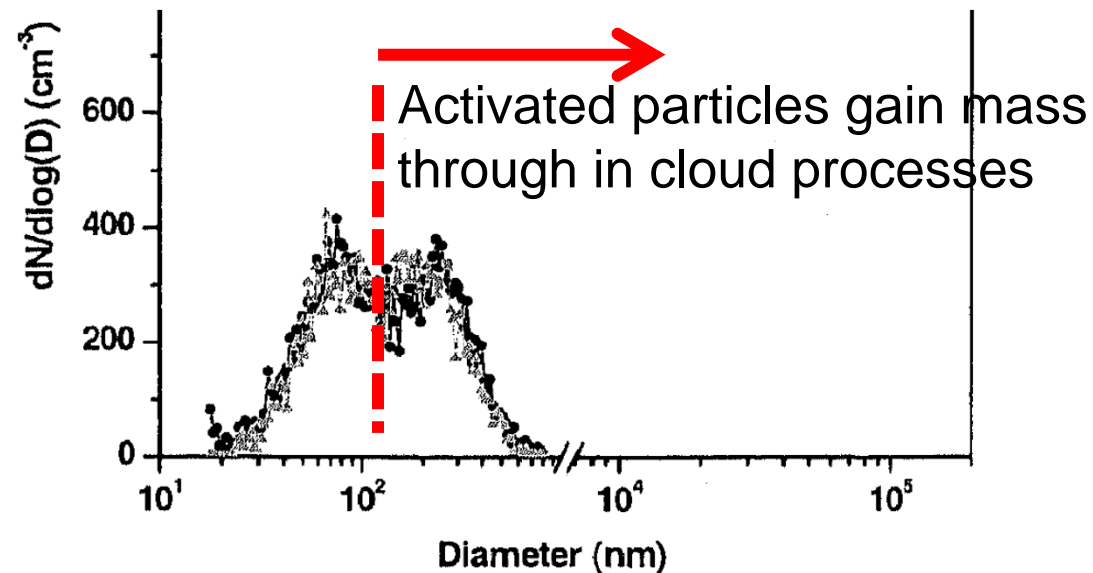


Activated fraction as a function of particle diameter in the real cloud

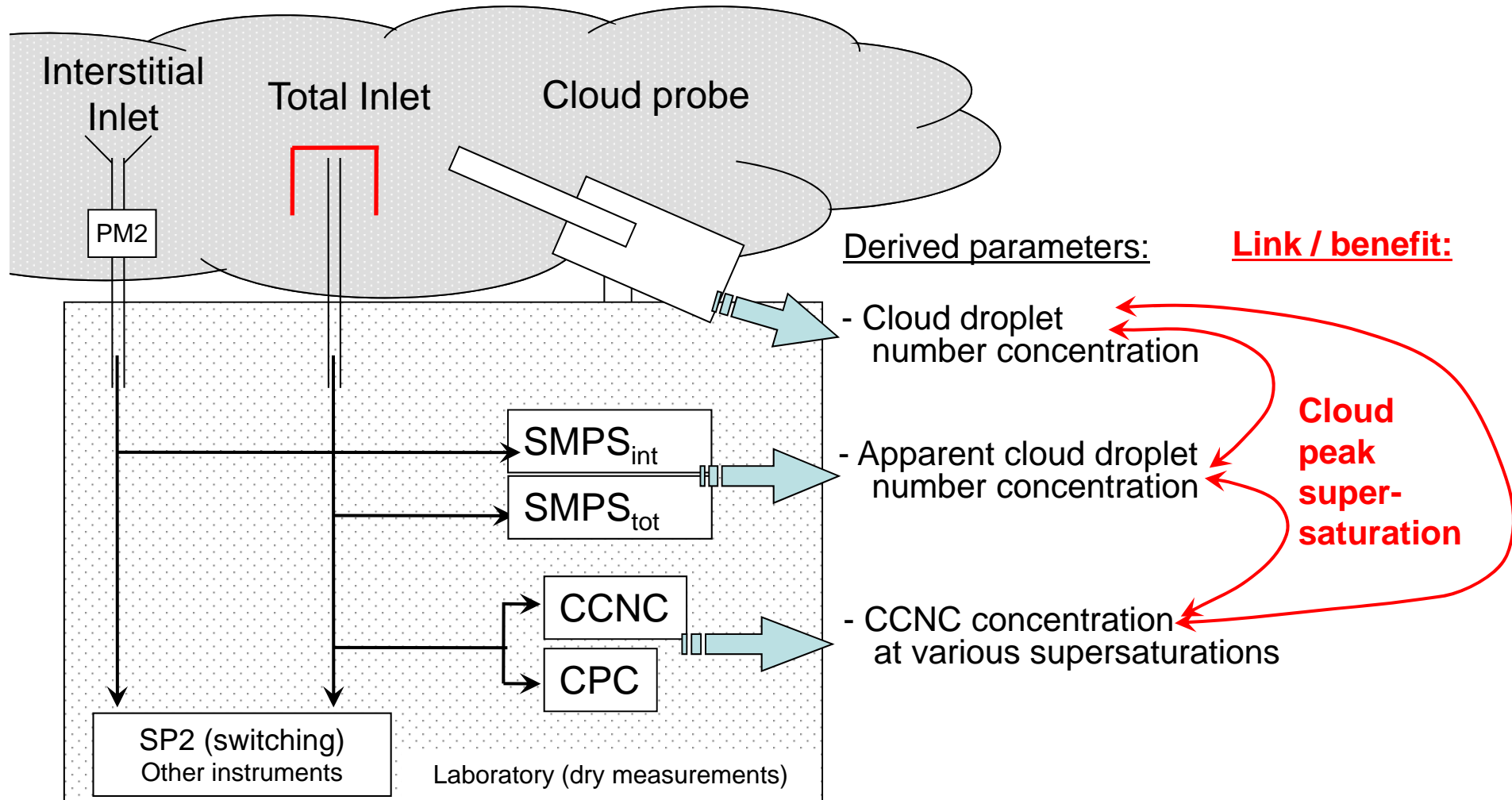


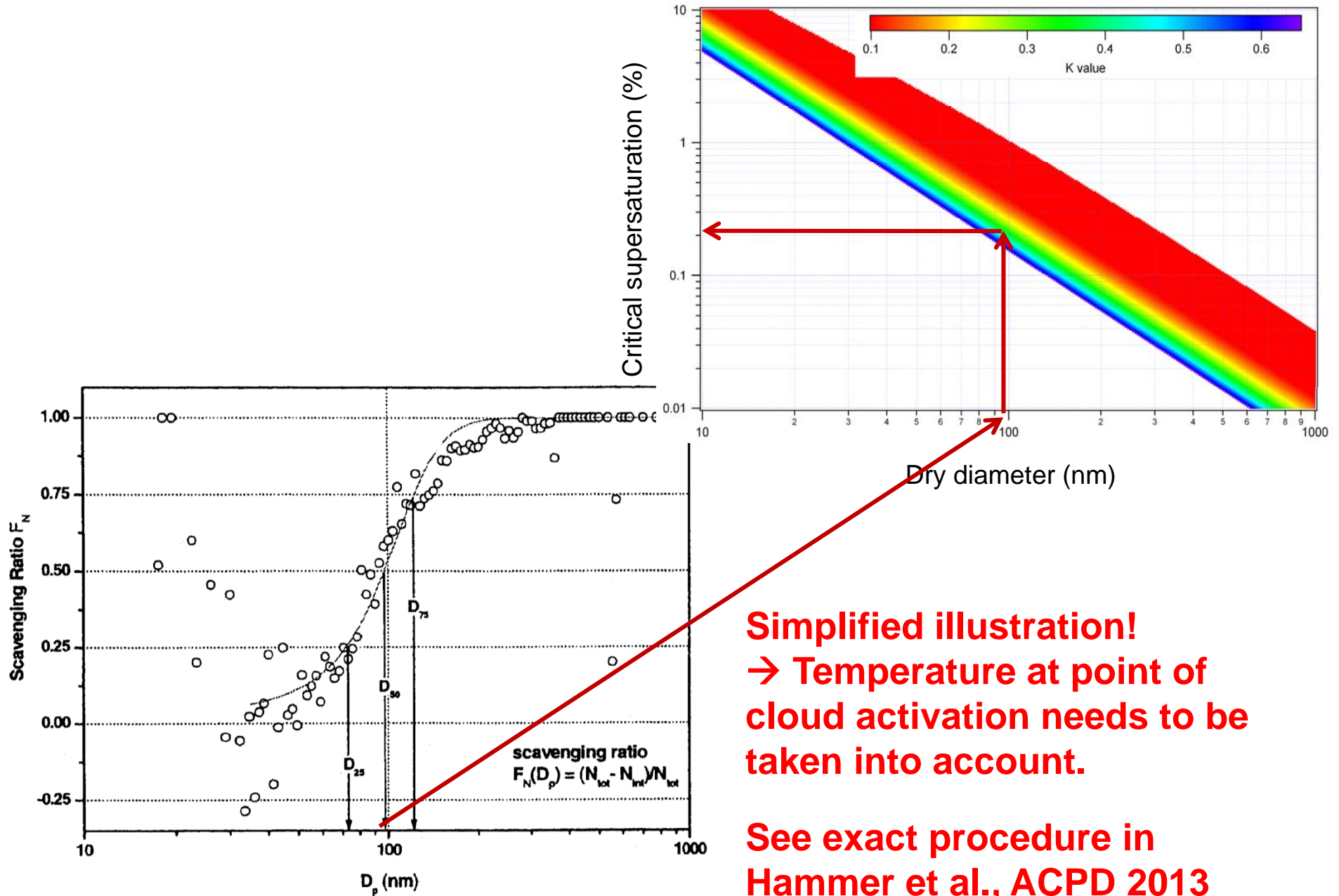
BTW: bimodal size distribution

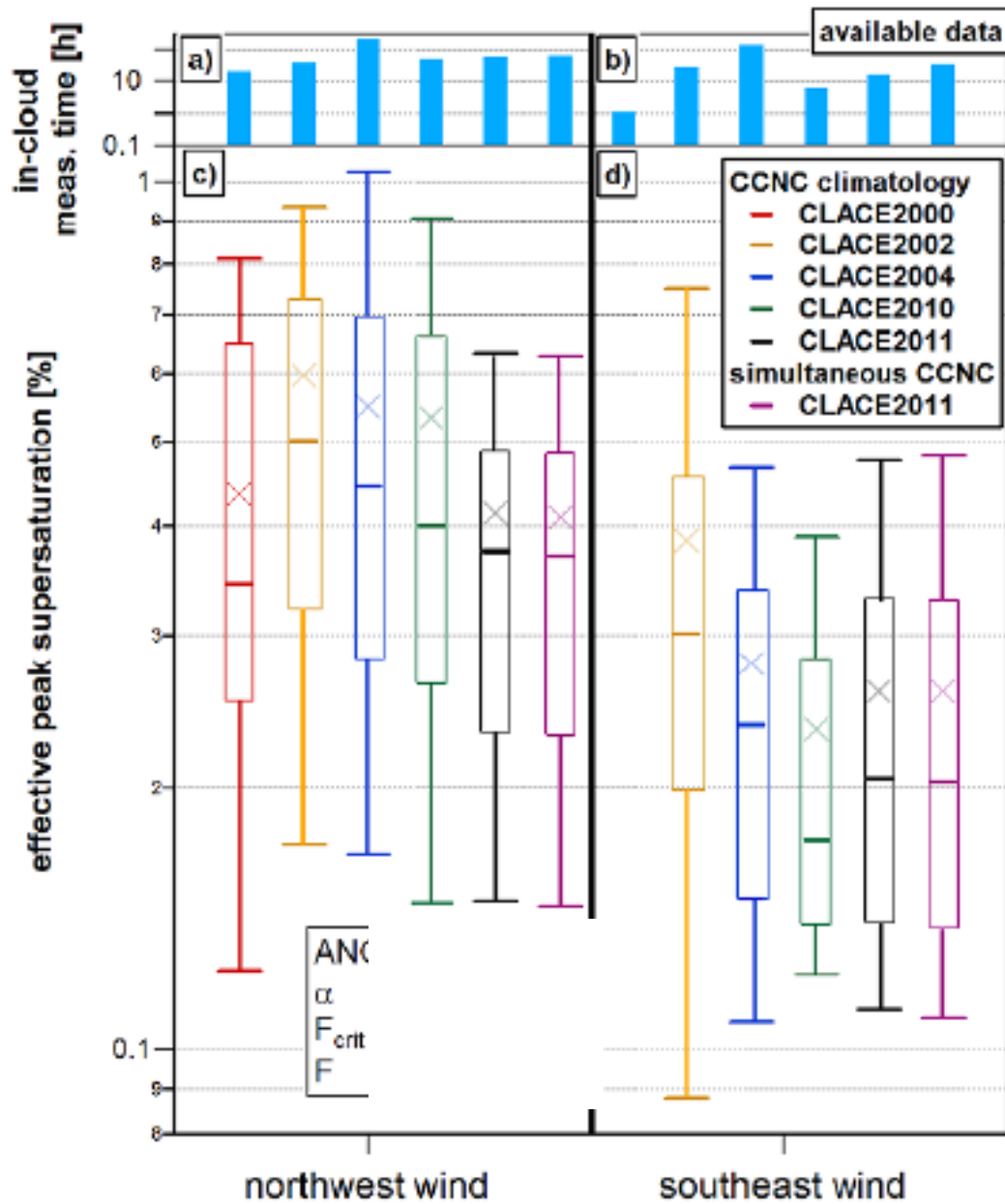
(Hoppel minimum) in the fine mode explained by this preferential activation of larger particles followed by mass gain through in cloud oxidation of water soluble gases (such as SO_2 and oxidized organics)



Determination of the actual supersaturation in clouds



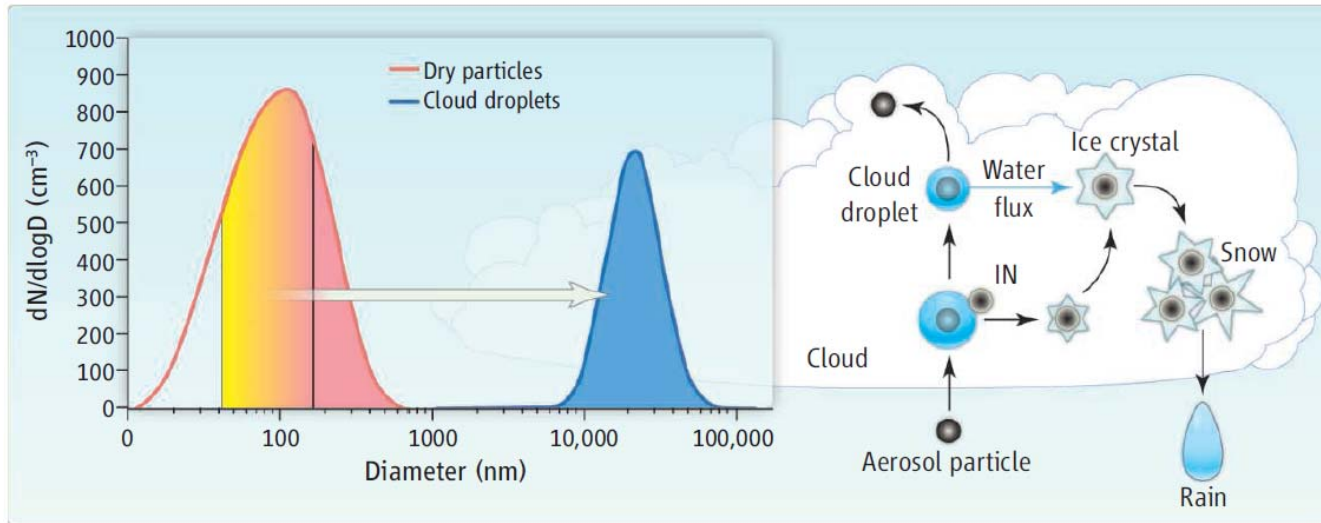




Higher for north wind than for south wind conditions because of steeper North slopes



Which particles act as ice nuclei?



Baltensperger, Science 2010

In an ice cloud, cloud droplets evaporate while few, but large ice crystals grow on ice nuclei, changing cloud radiative properties and enhancing precipitation.

Therefore it is important which particles can act as ice nuclei.



Installation of the new ice selective inlet at the Jungfrauoch

- A large number of aerosol properties needs to be determined for a full characterization
- High-altitude stations can provide highly valuable information on an aged aerosol above a continental area
- Variables need to be measured dry, but for comparison with ambient data their transformation to wet conditions is required
- Nucleation is thought to be important in the free troposphere but very few data exist on the ingredients contributing to nucleation
- High-altitude stations, when in clouds, provide highly valuable information on aerosol-cloud interaction. Even more important: characterization of ice clouds, including the determination of ice nuclei (e.g., are BC particles IN at low temperature or not?)



Thank you for your attention

**And thanks to
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