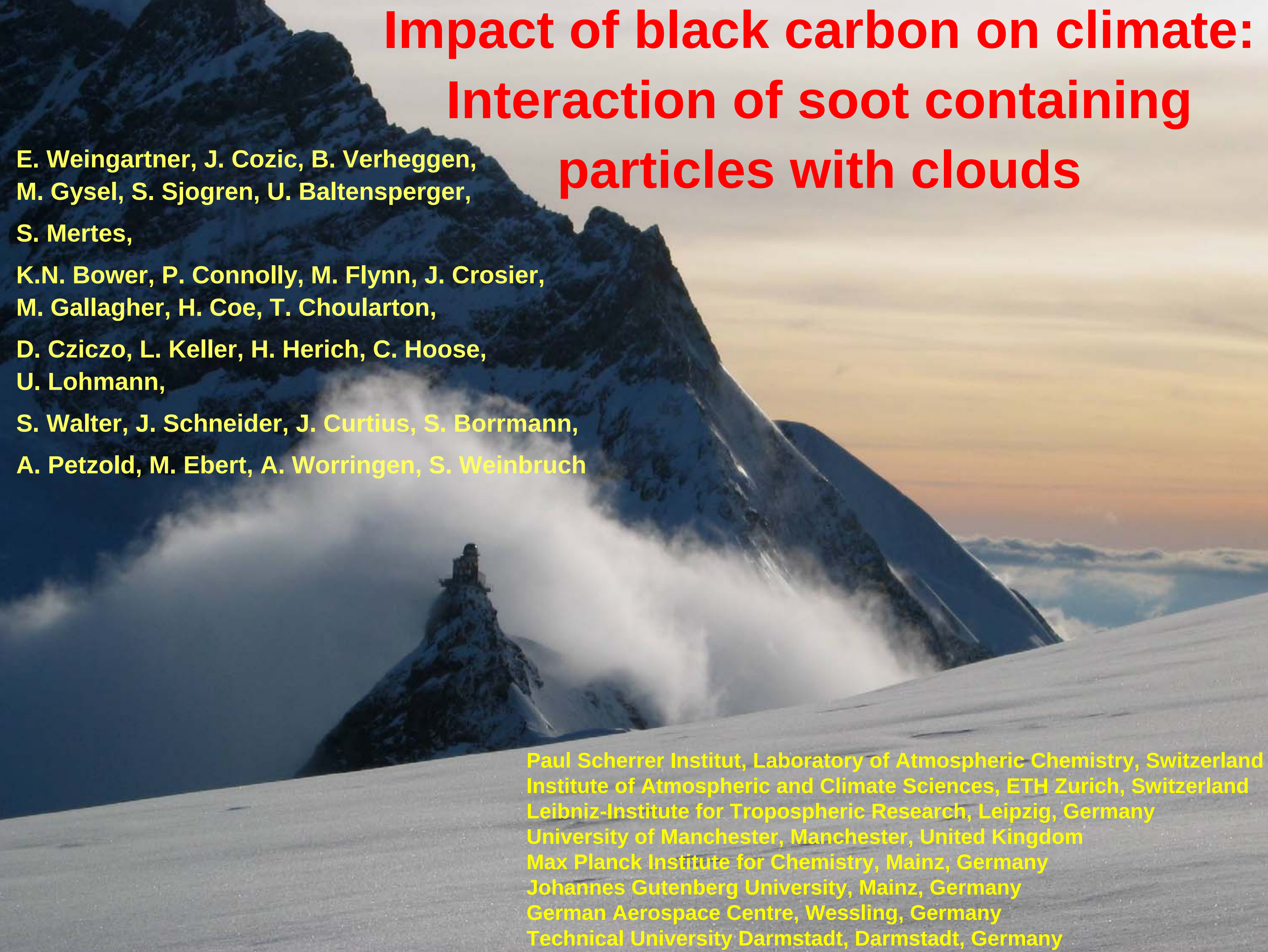


Impact of black carbon on climate: Interaction of soot containing particles with clouds

E. Weingartner, J. Cozic, B. Verheggen,
M. Gysel, S. Sjogren, U. Baltensperger,

S. Mertes,

K.N. Bower, P. Connolly, M. Flynn, J. Crosier,
M. Gallagher, H. Coe, T. Choularton,

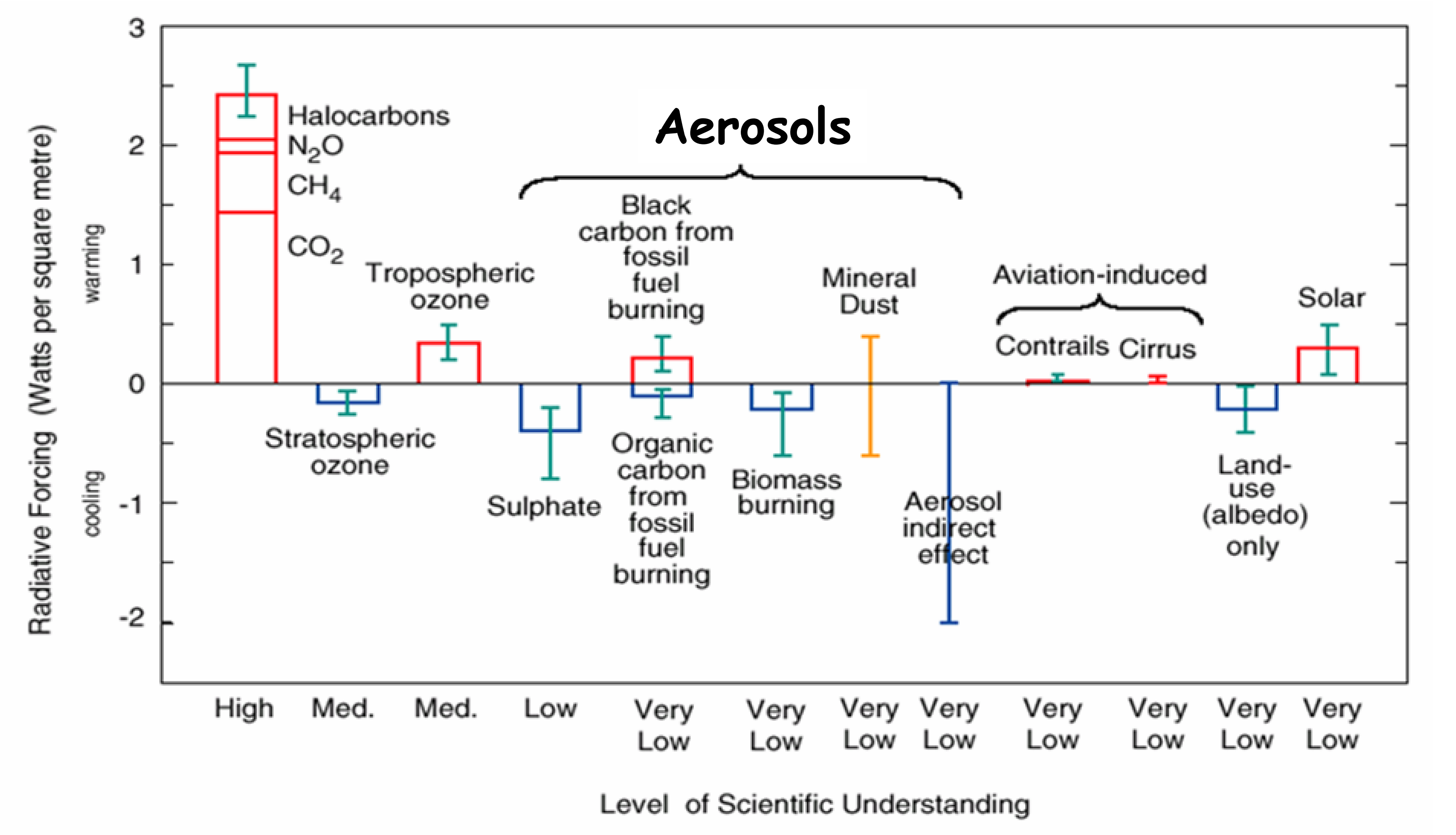
D. Cziczo, L. Keller, H. Herich, C. Hoose,
U. Lohmann,

S. Walter, J. Schneider, J. Curtius, S. Borrmann,

A. Petzold, M. Ebert, A. Worringen, S. Weinbruch

Paul Scherrer Institut, Laboratory of Atmospheric Chemistry, Switzerland
Institute of Atmospheric and Climate Sciences, ETH Zurich, Switzerland
Leibniz-Institute for Tropospheric Research, Leipzig, Germany
University of Manchester, Manchester, United Kingdom
Max Planck Institute for Chemistry, Mainz, Germany
Johannes Gutenberg University, Mainz, Germany
German Aerospace Centre, Wessling, Germany
Technical University Darmstadt, Darmstadt, Germany

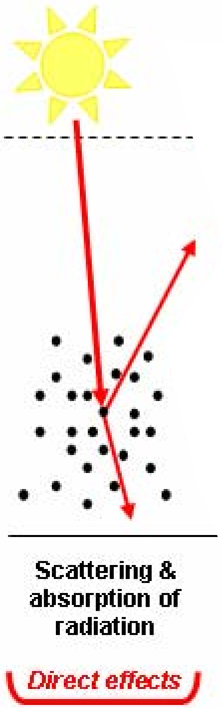
The global mean radiative forcing of the climate system



for the year 2000, relative to 1750

Source: www.ipcc.ch, 3rd assessment repot (2001)

Aerosol direct and indirect effects

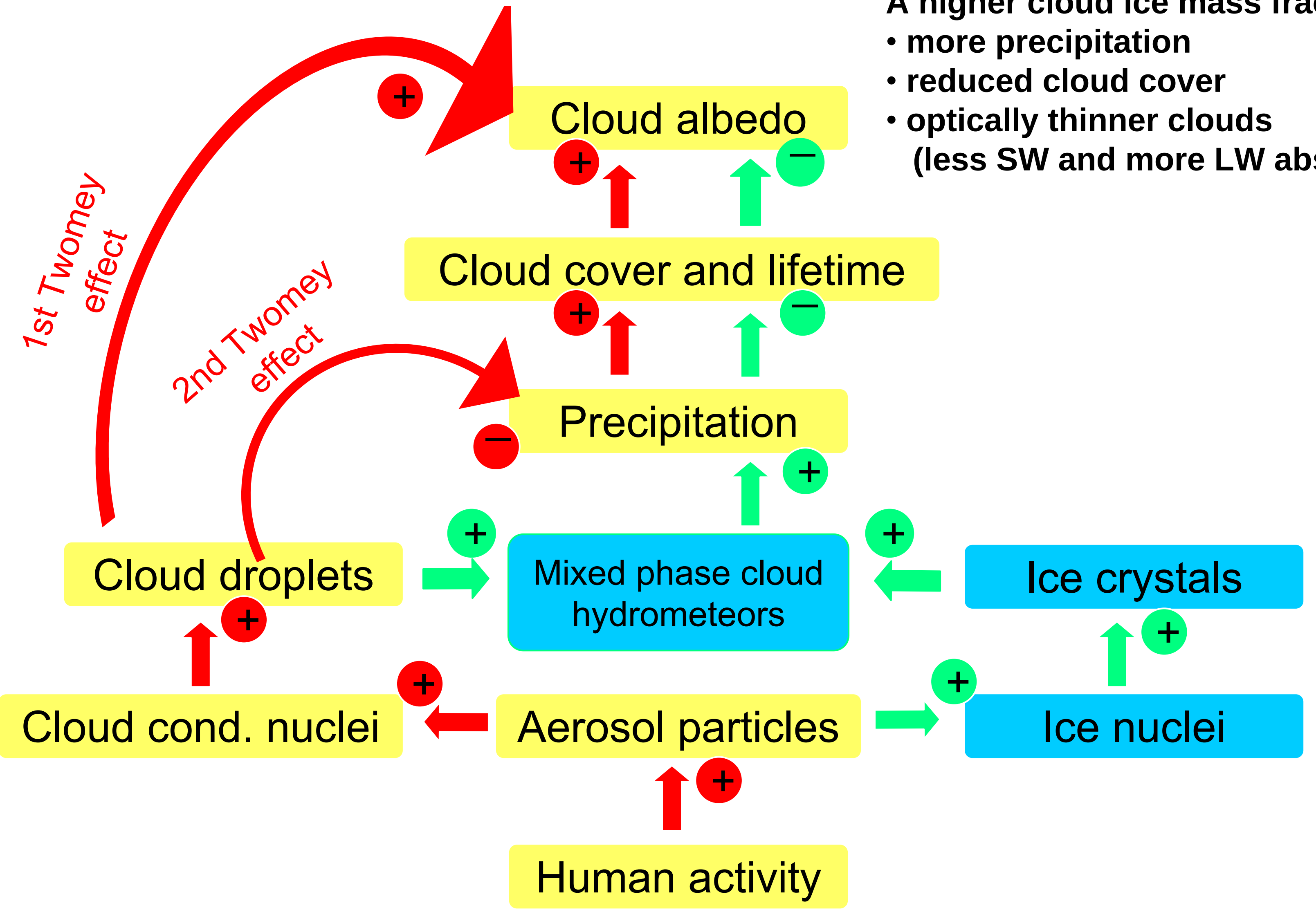


Radiative forcing:

-

adapted from IPCC 2007

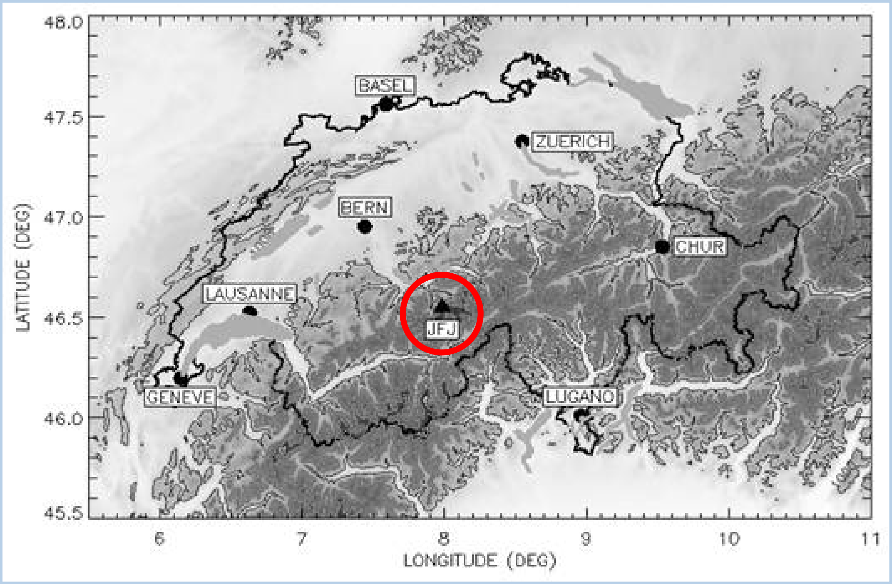
Pathways of the Traditional *Warm Indirect Aerosol Effect*



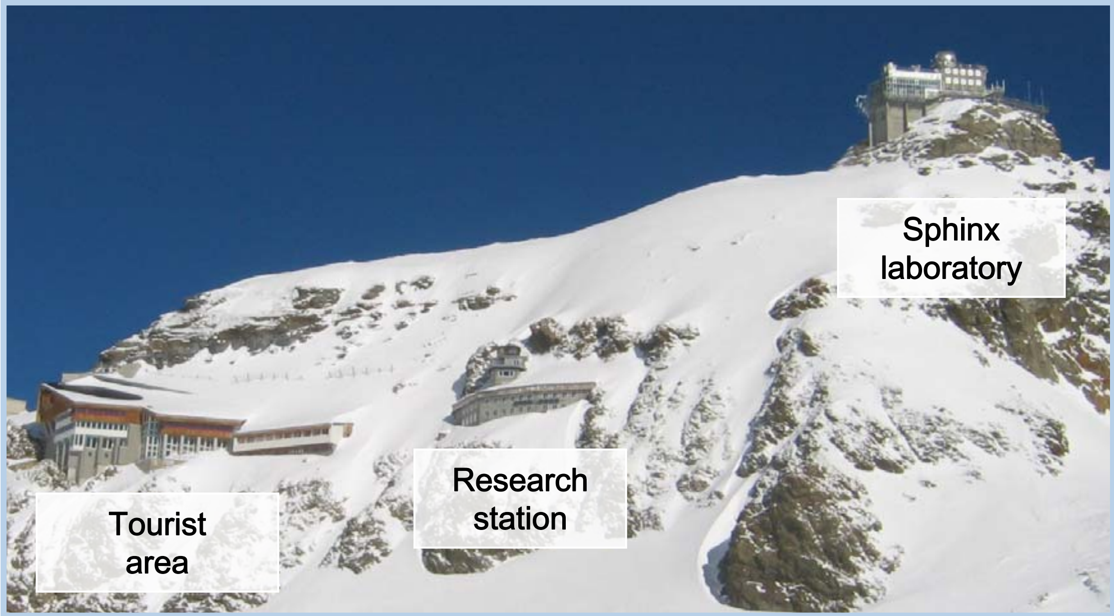
A higher cloud ice mass fraction results in
- more precipitation
- reduced cloud cover
- optically thinner clouds (less SW and more LW absorption)

From U. Lohmann, GRL, 2002

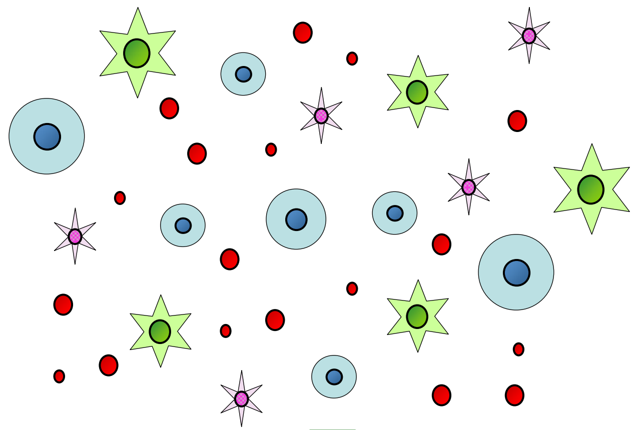
Jungfraujoch 3580 m a.s.l.



- GAW station
- Few local emissions
- Good infrastructure
- Free troposphere
- Aged aerosol
- 40% cloud occurrence



Inlets



Ice CVI inlet:
removes :
- droplets
– int. particles
– large ice crystals

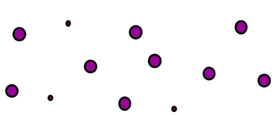
(Size : 5-20 µm)

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

(Size < 2µm)

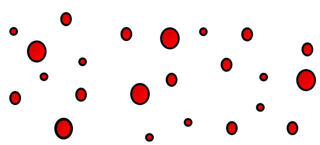
Total inlet :
(all particles, including activated ones)

heated inlet

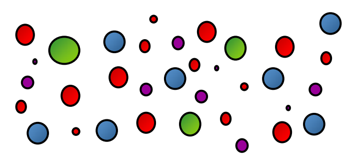


Ice residuals

Laboratory (dry aerosol)

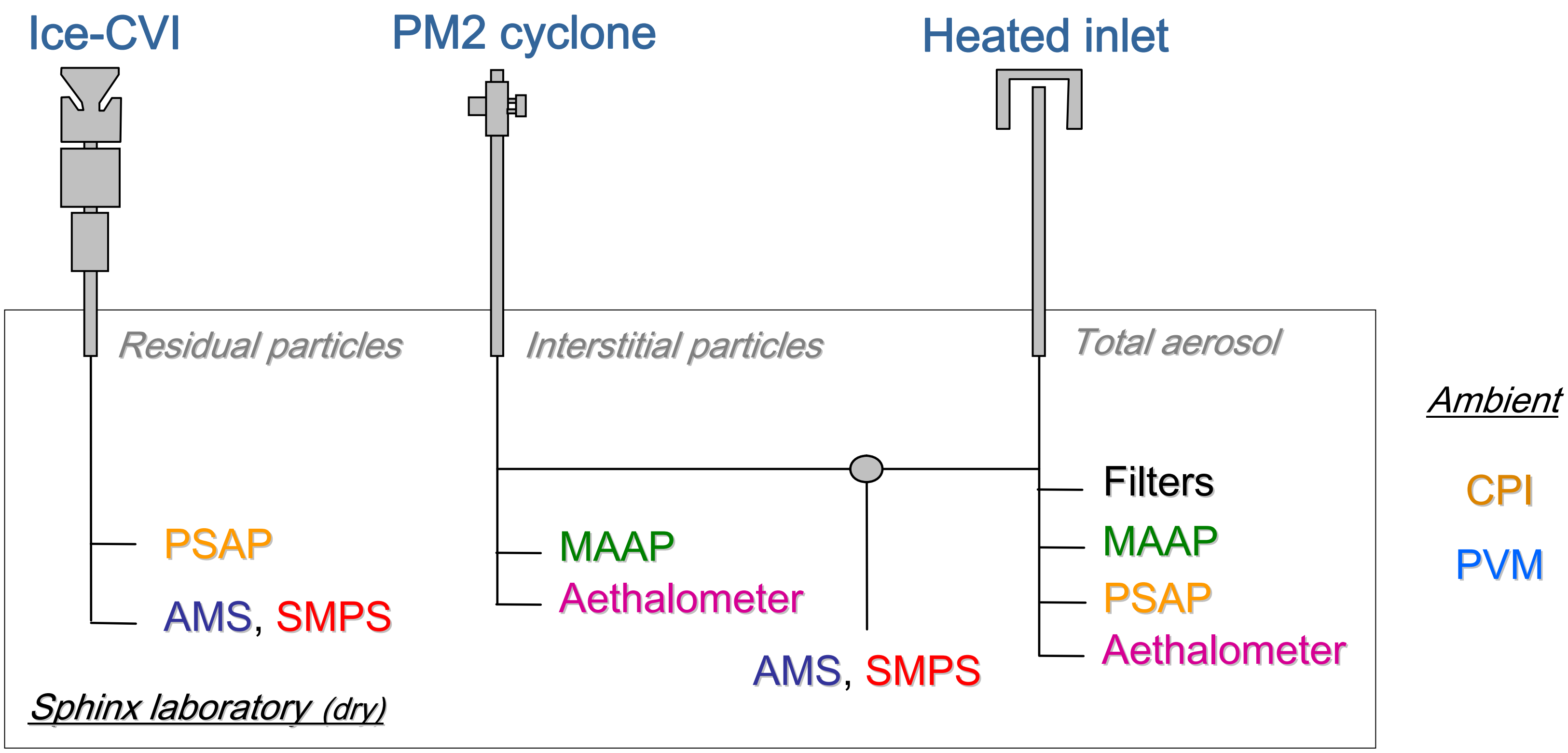


Interstitial particles



All particles

CLACE instrumentation



BC measurements:

- MAAP = Multi Angle Absorption Photometer
- PSAP = Particle Soot Absorption Photometer
- Aethalometer

Chemical composition measurements:

- AMS = Aerosol Mass Spectrometer

Cloud microphysics:

- PVM = Particulate Volume Monitor
- CPI = Cloud Particle Imager

Size distribution:

- SMPS = Scanning Mobility Particle Sizers

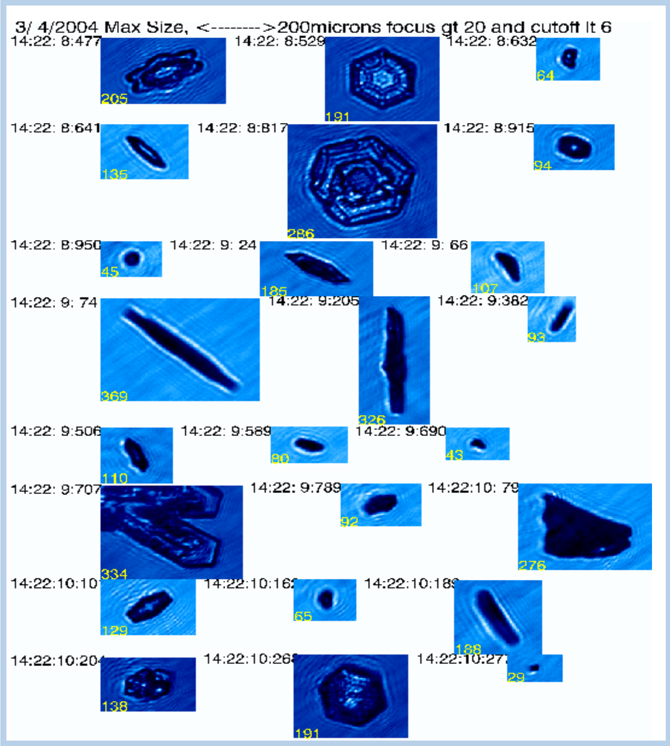
Cloud Microphysics

K. Bower, M. Flynn, Uni Manchester

MANCHESTER 1824

FSSP: size distribution of cloud particles

CPI: images of cloud particles

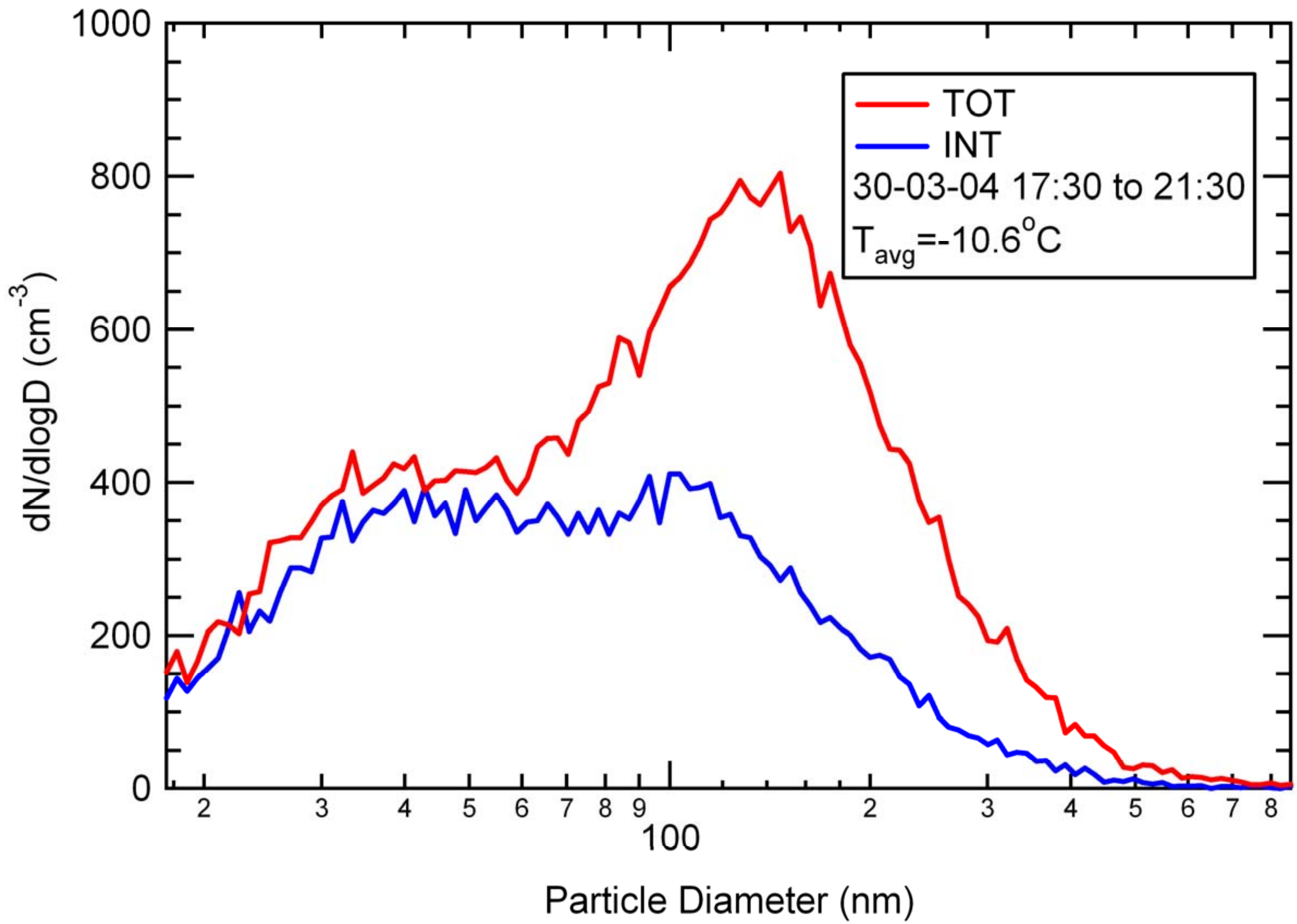


PVM: liquid water content (LWC)

Combining these permit to calculate the **ice mass fraction (IMF)**

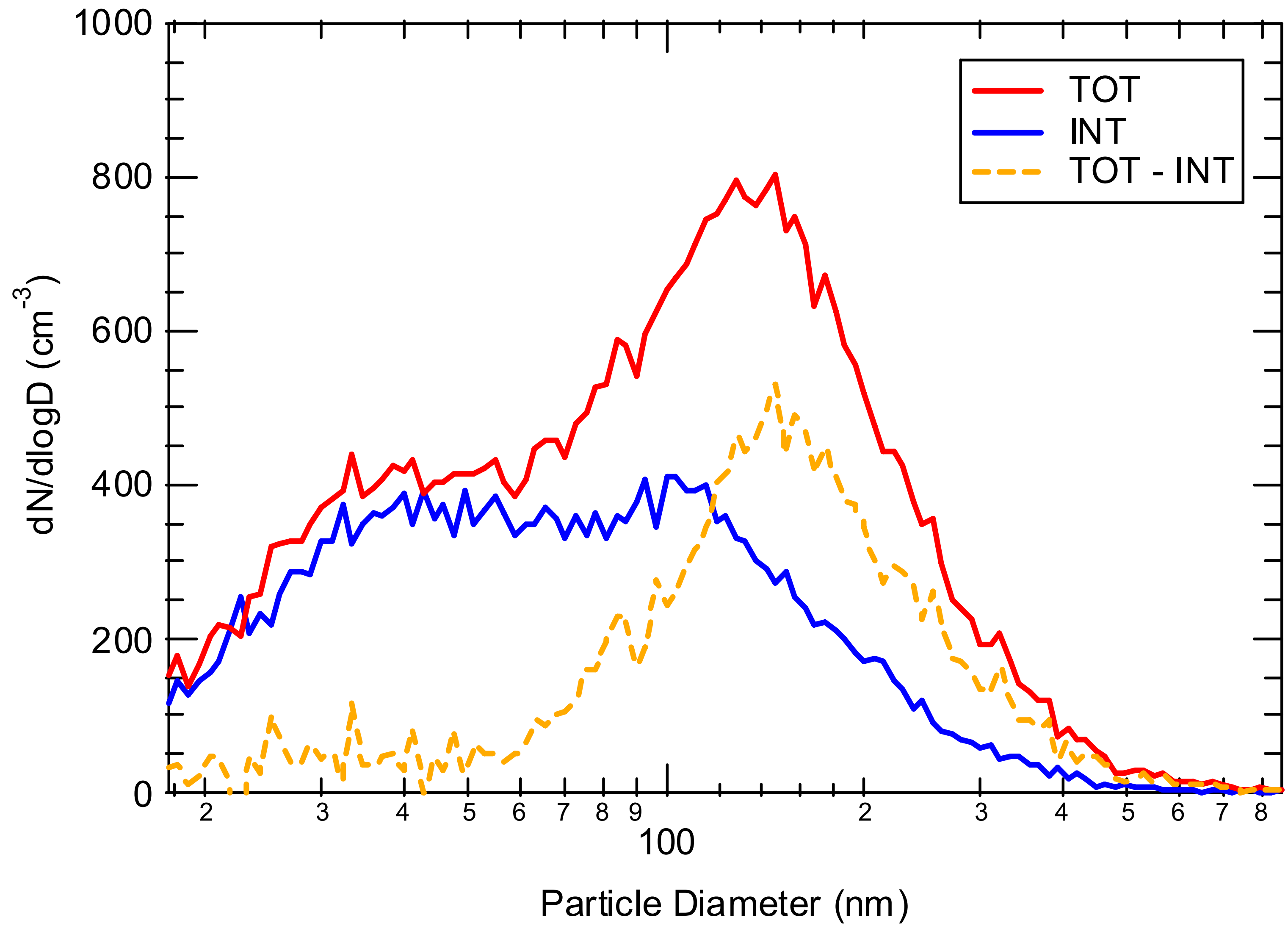
Average size spectra during a liquid cloud

17:30 – 21:30; T_{avg} = -10.6 °C; activated fraction = ~0.6

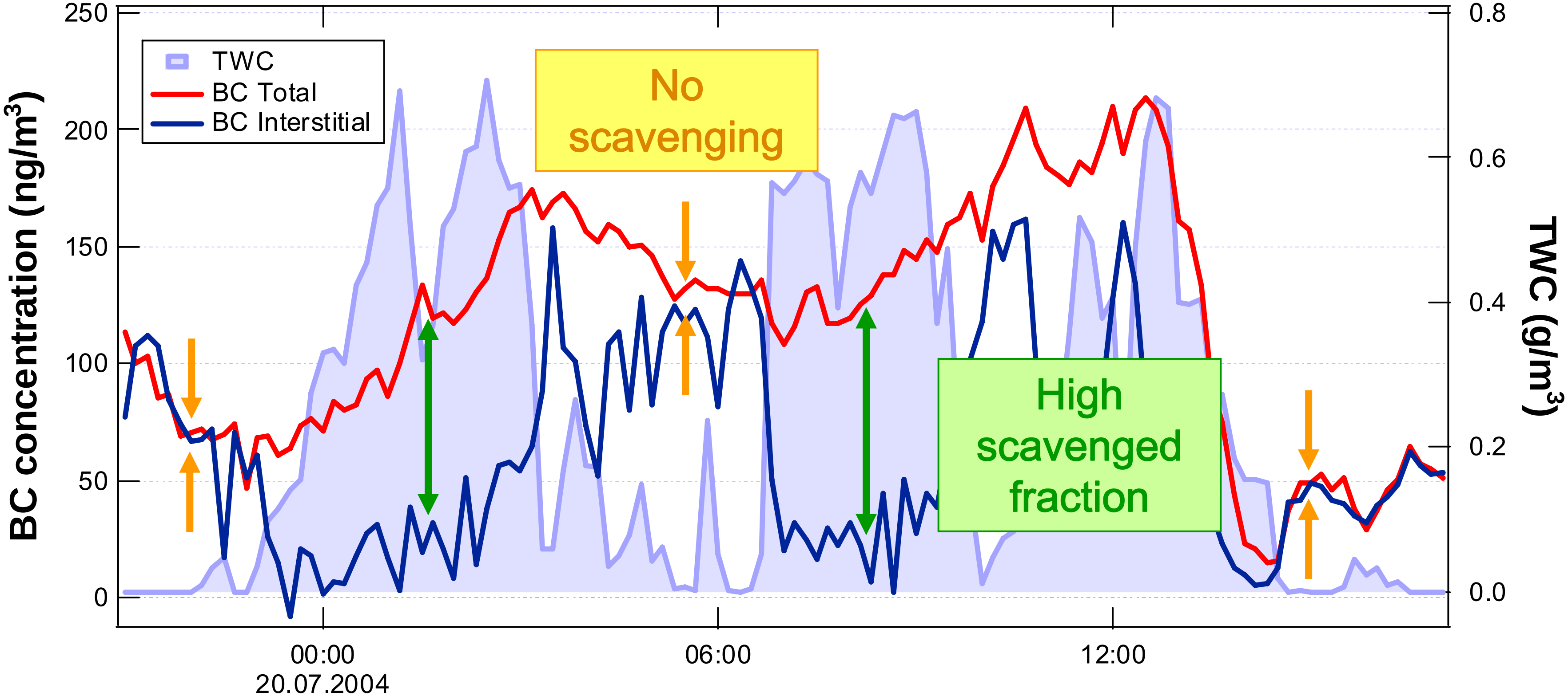


Average size spectra during a liquid cloud

17:30 – 21:30; T_{avg} = -10.6 °C; activated fraction = ~0.6



Scavenging of Black Carbon in a liquid cloud



$$\text{Scavenged fraction} = \frac{C_{cloud}}{C_{total}} = \frac{C_{tot} - C_{int}}{C_{total}}$$

= Fraction of BC mass that is incorporated into a cloud droplet or an ice crystal

Scavenging of Black Carbon

Aging effect

Close to sources

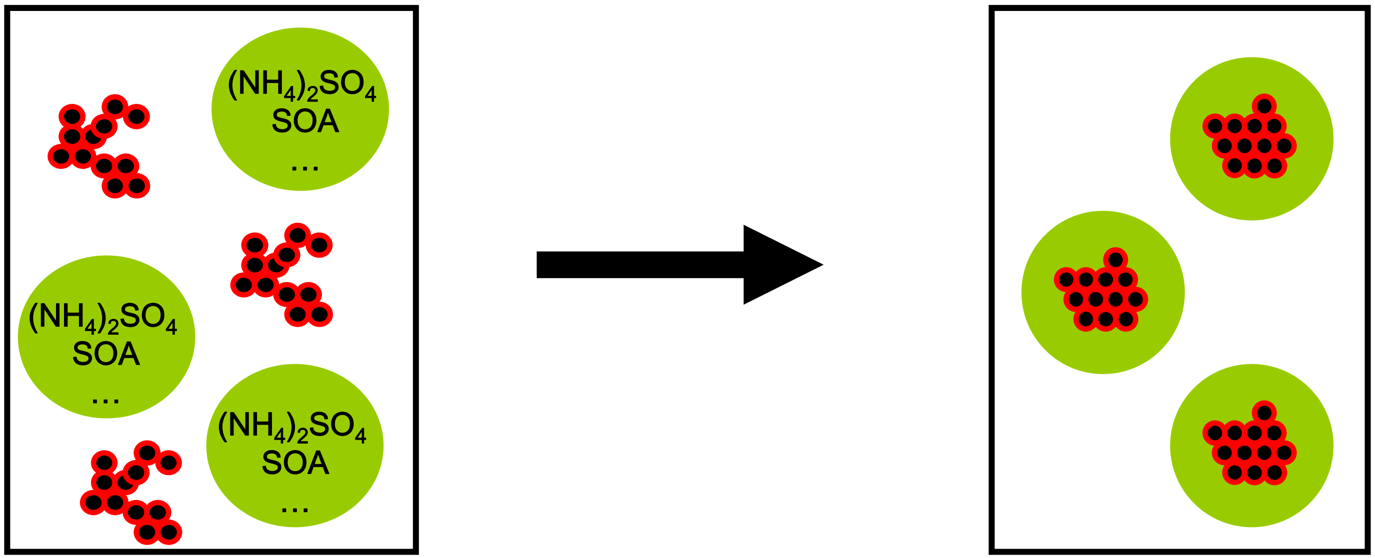
Far from sources

Sampling site	$F_{scav,BC}$	Type of site	Reference
Po Valley (Italy)	0.06	Urban	Hallberg et al. (1992)
Kleiner Feldberg (Germany)	0.15	Rural	Hallberg et al. (1994)
Puy de Dôme (France)	0.33	Mid altitude (1465m)	Sellegri et al. (2003)
Mt Sonnblick (Austria)	0.45	High altitude (3106m)	Kasper-Gielb et al. (2000)
Rax (Austria)	0.54	Mid altitude (1644m)	Hitzenberger et al. (2001)
Great Dun Fell (U.K.)	0.57	Rural - Coastal	Gieray et al. (1997)
Jungfraujoch (Switzerland)	0.61	High altitude (3850m)	Cozic et al. (2007)
Mt Sonnblick (Austria)	0.74	High altitude (3106m)	Hitzenberger et al. (2000)
Spitzbergen (Norway)	0.80	Artic	Heintzenberg and Leck (1994)

Cozic J. et al., ACP, 7, 1797–1807, 2007

Atmospheric aging processes change the mixing state

important for e.g. modeling the radiative forcing of black carbon

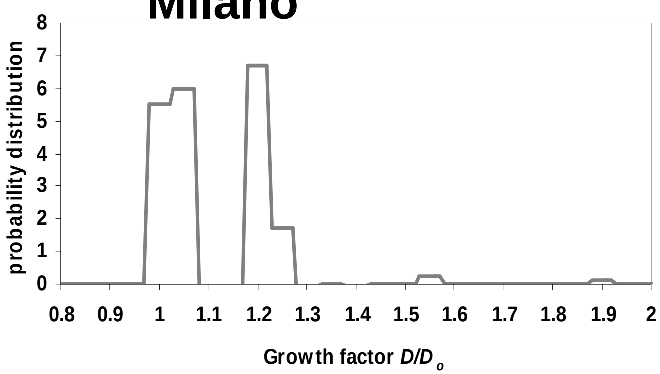


External Mixture
BC particles are separated from scattering particles

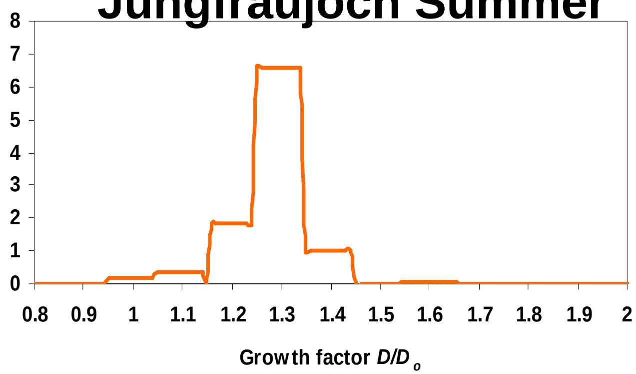
Coated Internal Mixture
BC particles are coated with scattering material

Hygroscopic growth factor distributions (inverted):

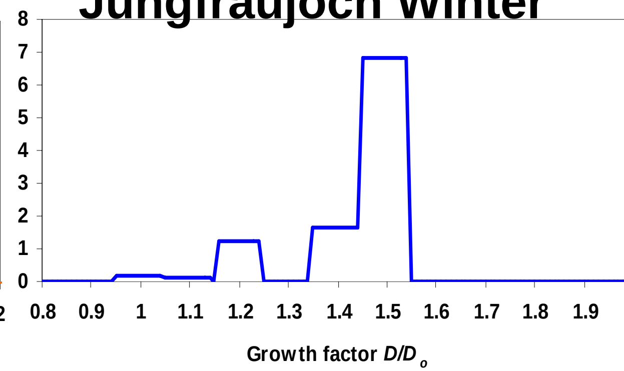
Milano



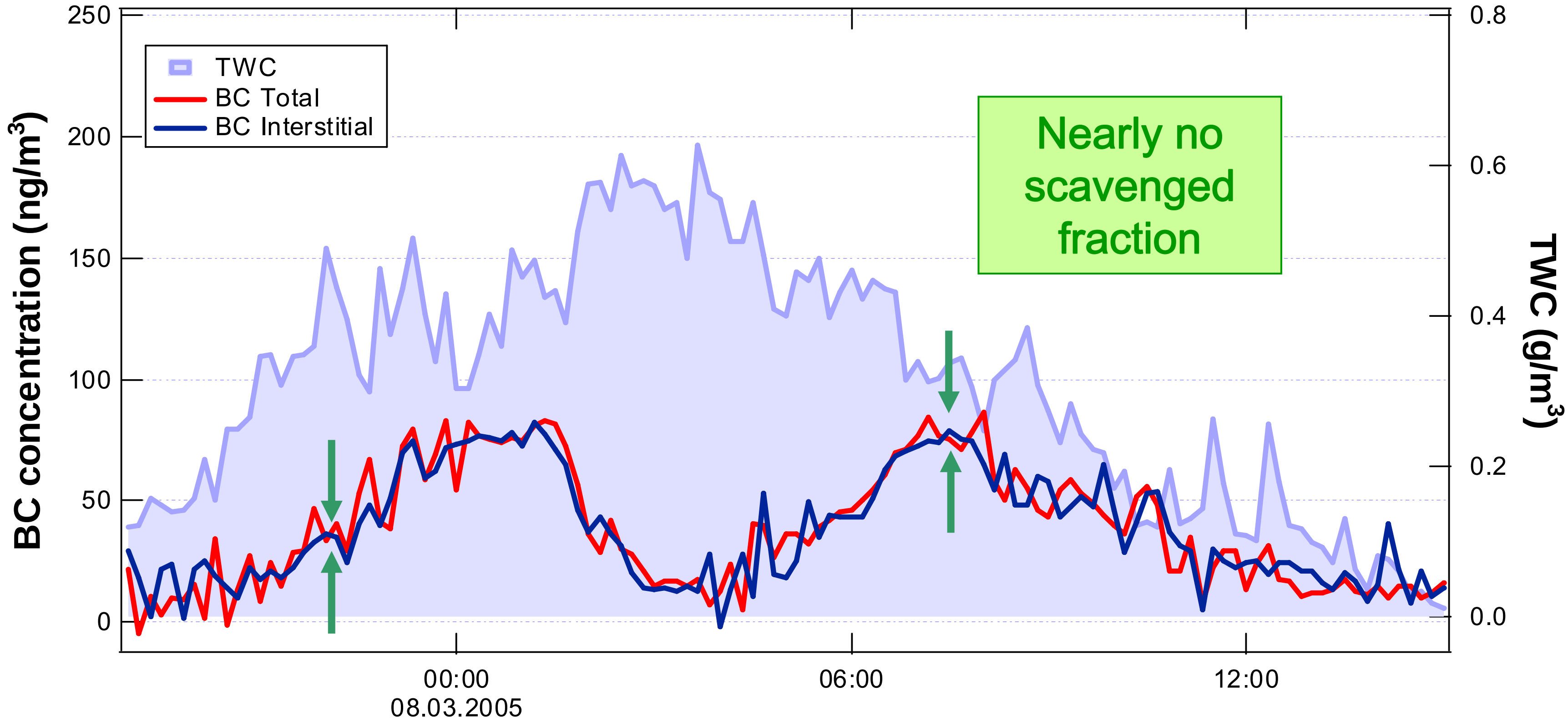
Jungfraujoch Summer



Jungfraujoch Winter



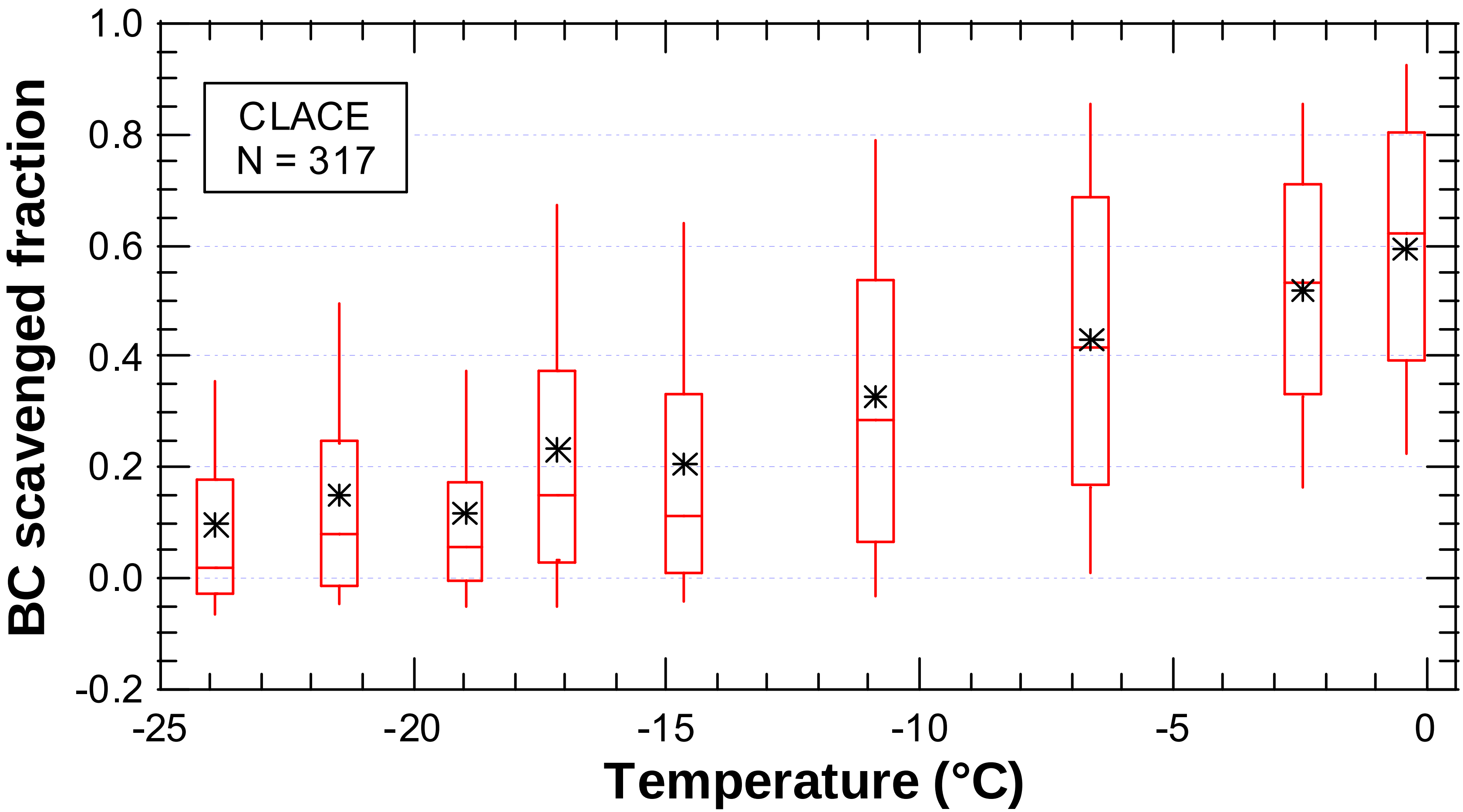
Scavenging of Black Carbon in a mixed phase cloud



Scavenged fraction < 10%

Scavenged BC fraction evolution with temperature

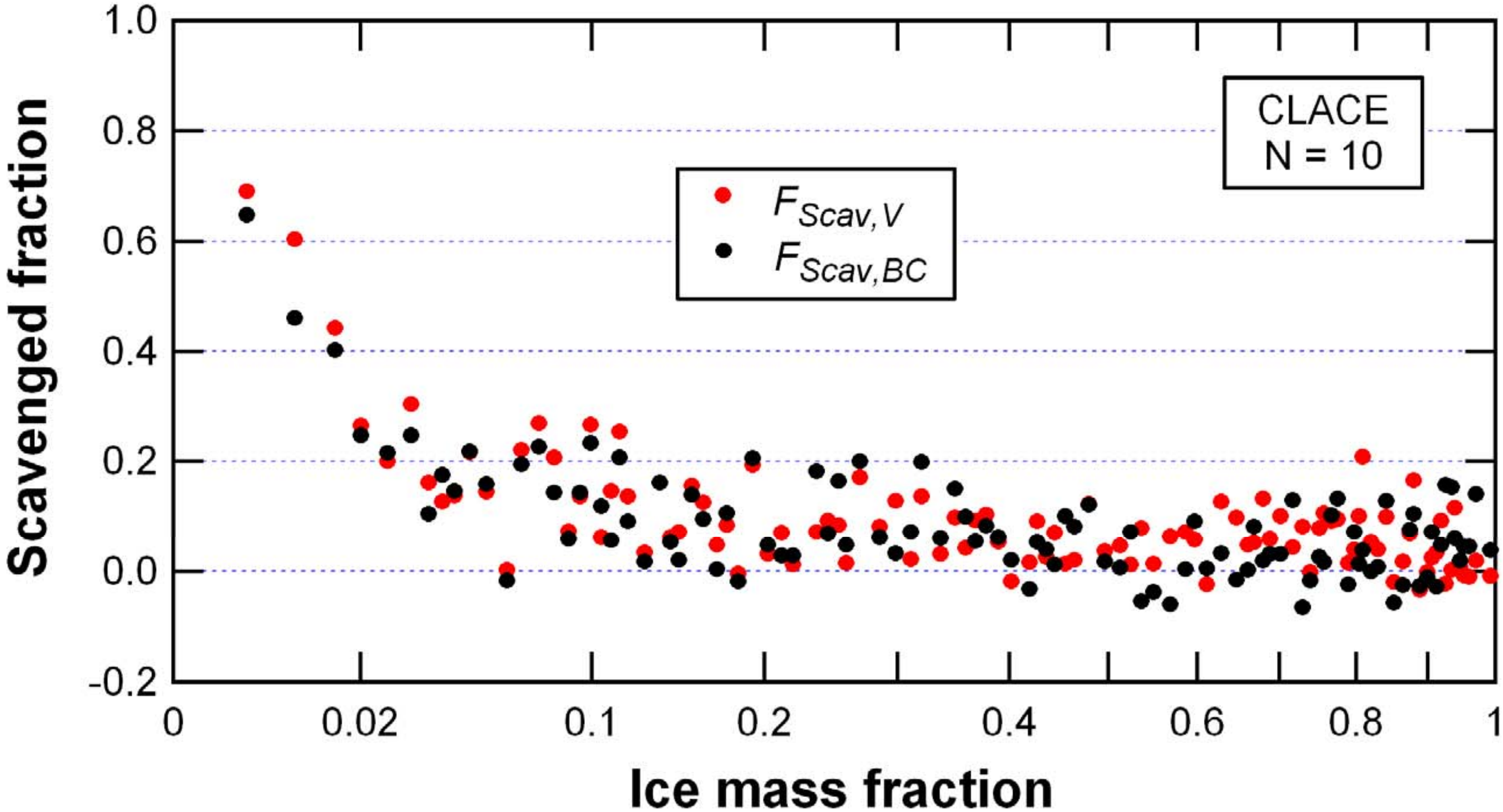
(based on 581 hours of in-cloud measurements)



Cozic J. et al., ACP, 7, 1797–1807, 2007

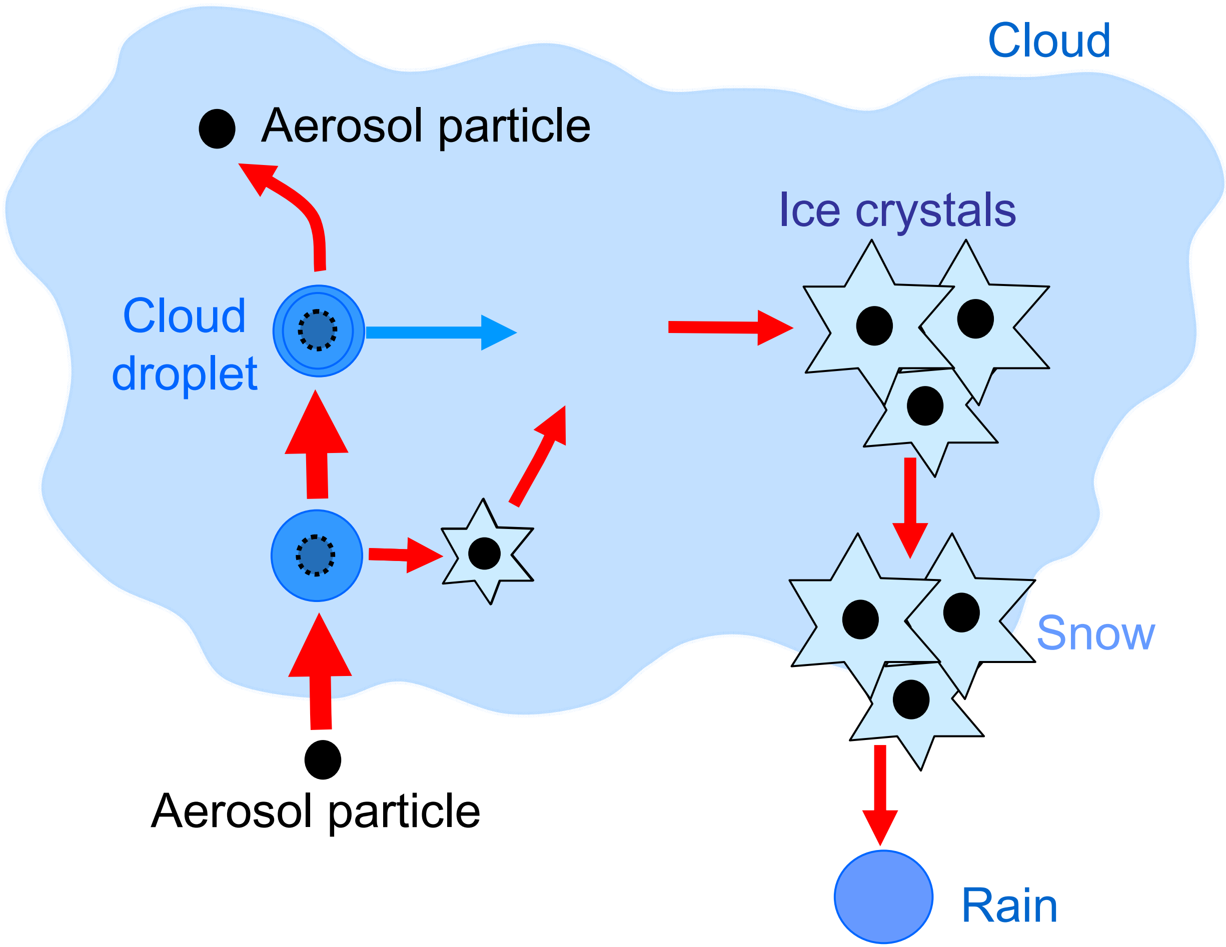
BC is activated as the bulk aerosol

Based on more than 170 hour of in-cloud measurements



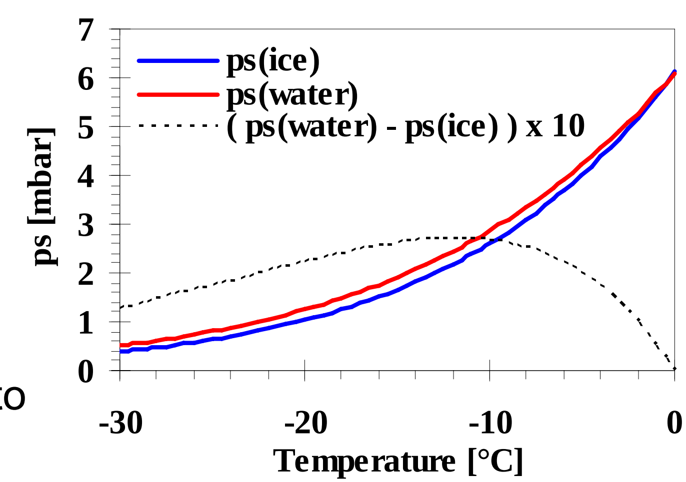
Cozic J. et al., ACP, 7, 1797–1807, 2007

Evolution of particles in cloud: Bergeron-Findeisen process



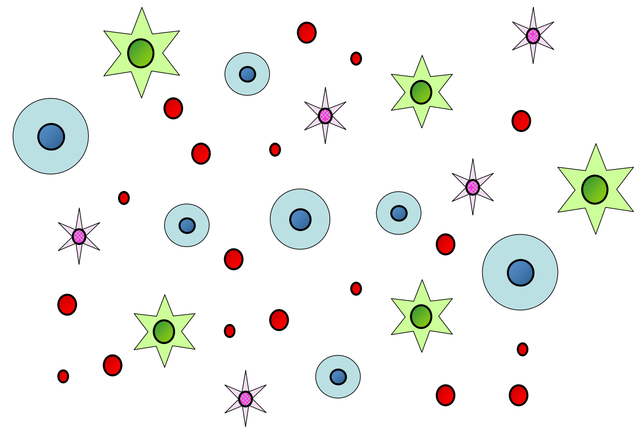
CCN:
10-50% (in terms of number, @ SS= 0.2-0.3%), depending on particle size (D > ~60 nm) and hygroscopic properties

Saturation vapor pressures over water and ice



Flux of water vapor from liquid droplets to ice crystals
The WBF mechanism converts many small supercooled drops to only few, large ice crystals.

Inlets

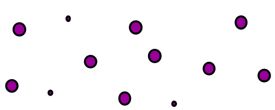


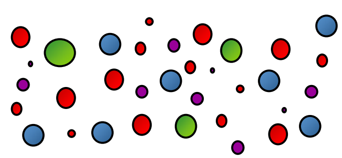
Ice CVI inlet:
removes :
- droplets
- int. particles
- large ice crystals

(Size : 5-20 µm)

Total inlet :
(all particles, including activated ones)

heated inlet

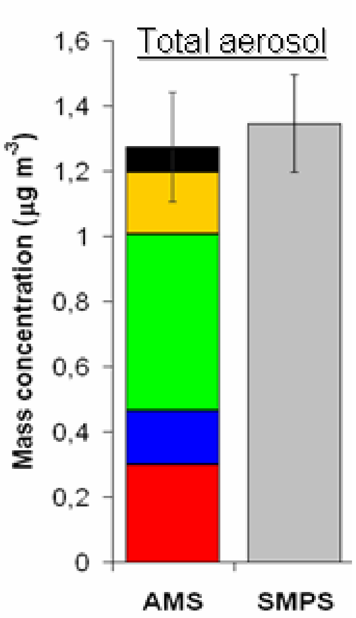


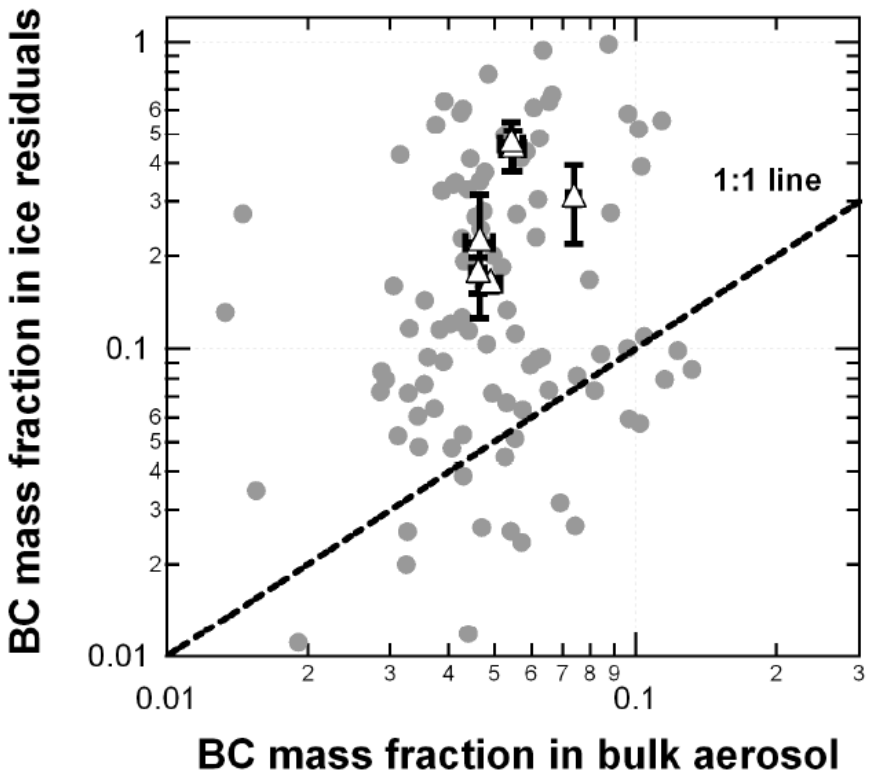


Laboratory (dry aerosol)

Max-Planck-Institut für Chemie (Otto-Hahn-Institut)

Ice residuals chemical composition





BC
Ammonium
Organics
Nitrate
Sulphate

➢ Enrichment of BC in small ice crystals (most points above 1:1 line)

➢ Ice residuals mainly consisted of BC and refractory material (mineral dust,…)

Summary

- The formation of ice inside supercooled clouds is very important and poorly understood.

- The partitioning of aerosol particles between the interstitial and cloud phase strongly depends on ice mass fraction and on temperature

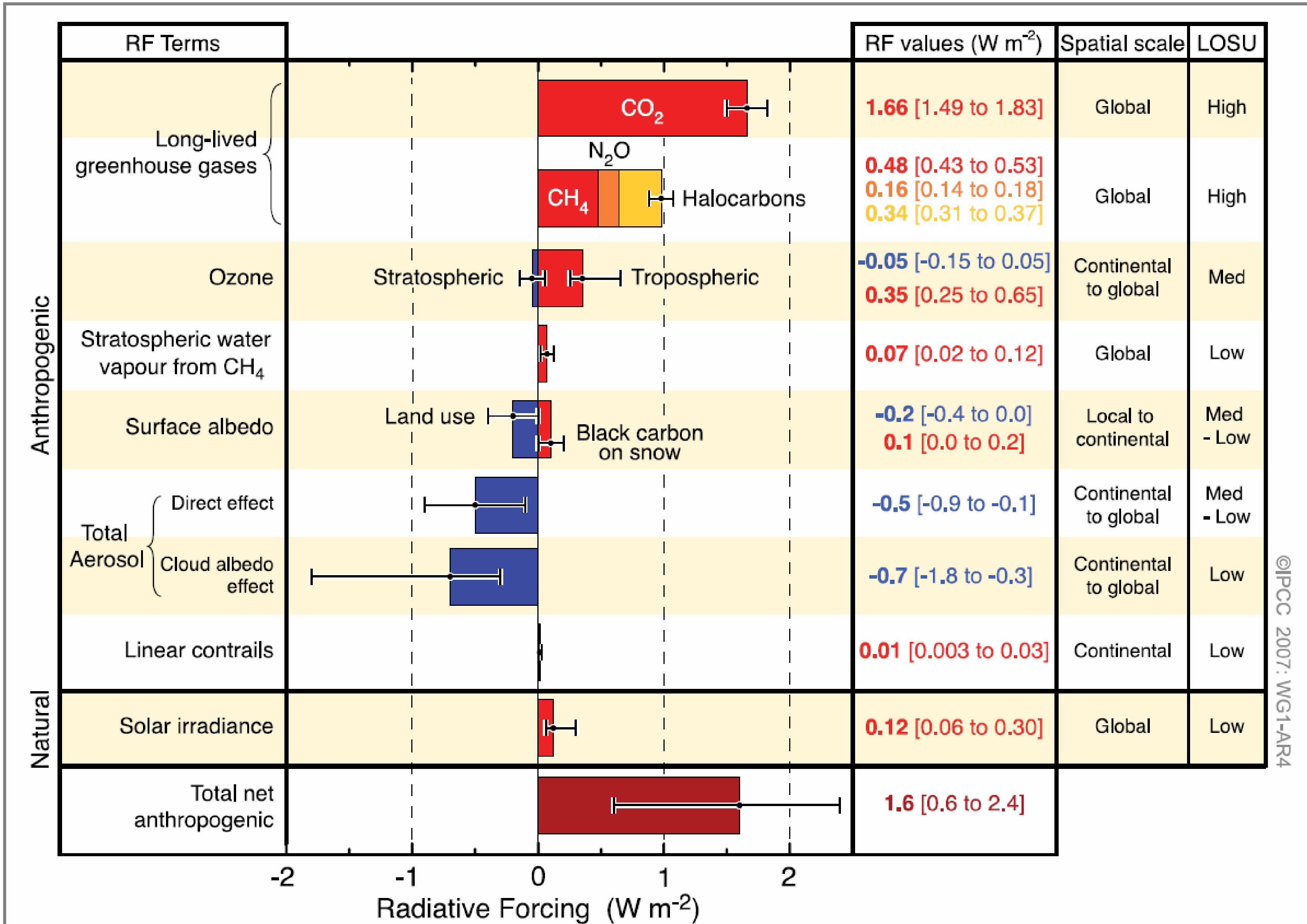
- Only a few particles (< 1% in terms of number) act as ice nuclei in our atmosphere

- The accumulation mode aerosol is internally mixed.
  Therefore, in liquid clouds, BC is scavenged as the bulk aerosol
- But: In mixed-phase cloud, BC is enriched in the ice phase
- Ice residuals mainly consist of BC and refractory material (mineral dust)

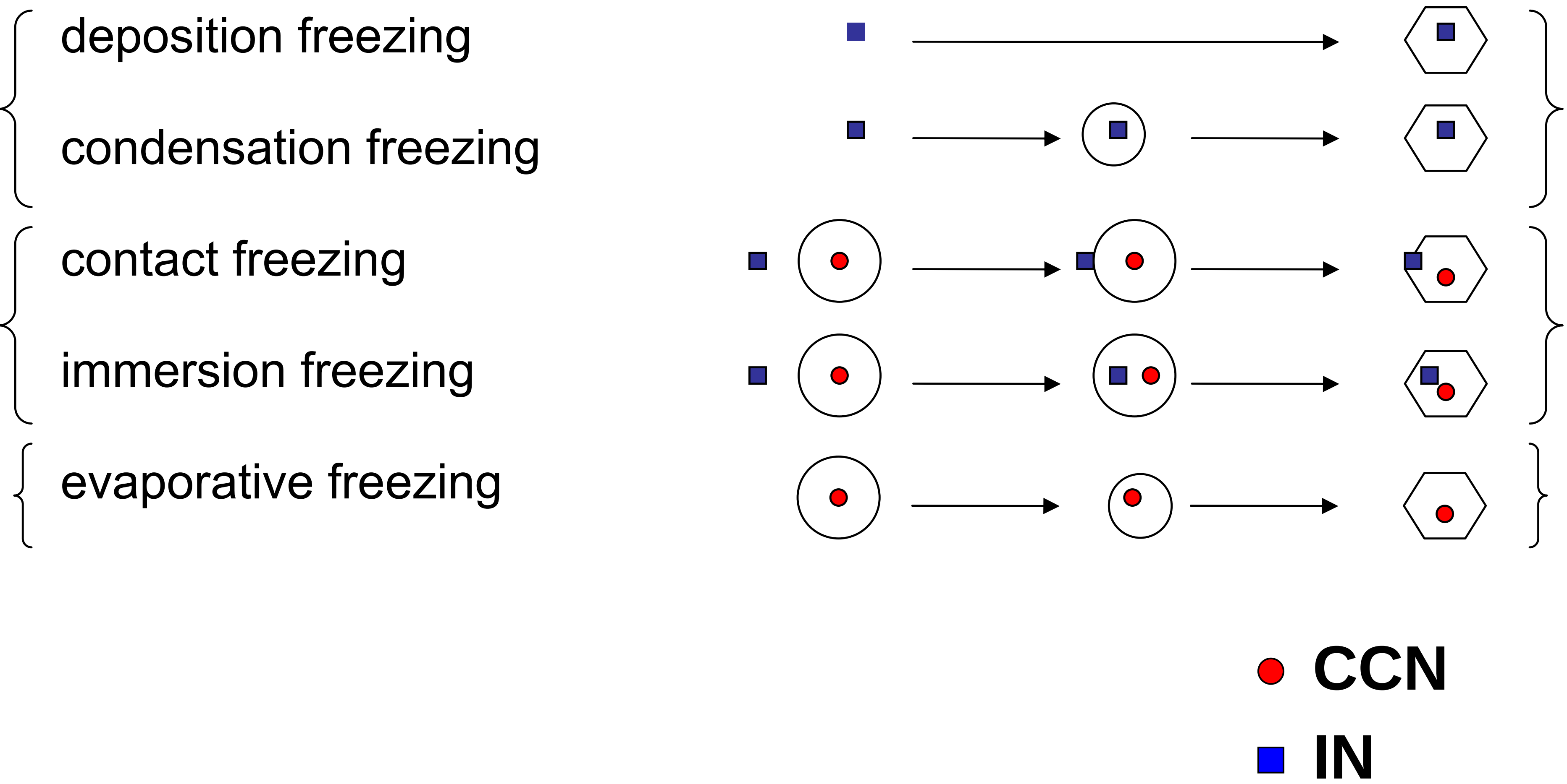
- The interaction of aerosol particles with the ice phase has to be incorporated into global climate models (effect on the radiative balance)


Thank you for your attention

The global mean radiative forcing of the climate system



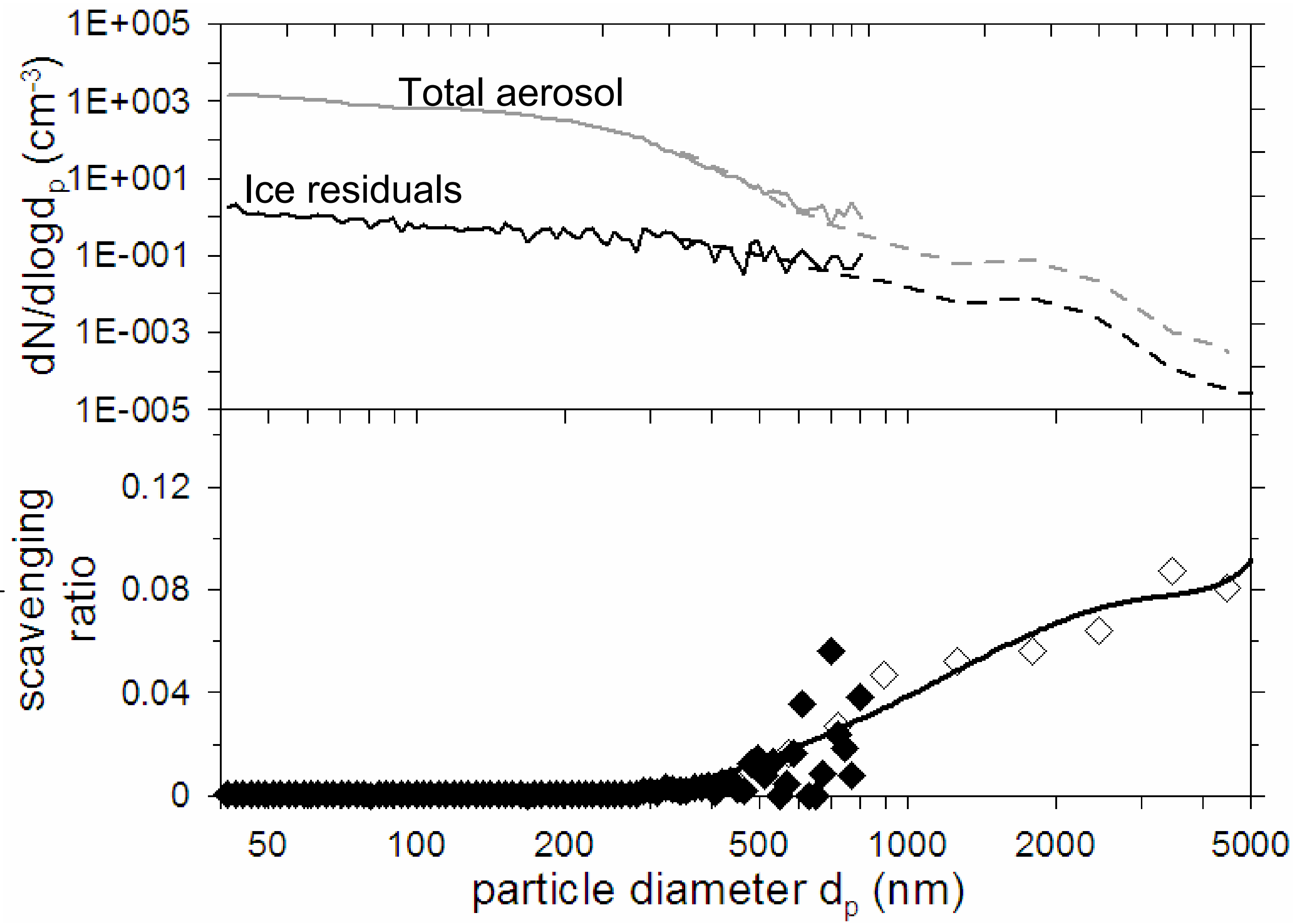
Ice crystal formation



Number size distribution from SMPS and OPC:

Courtesy of
S. Mertes, IFT

$$\frac{N_{Ice\ residuals}}{N_{tot}}$$



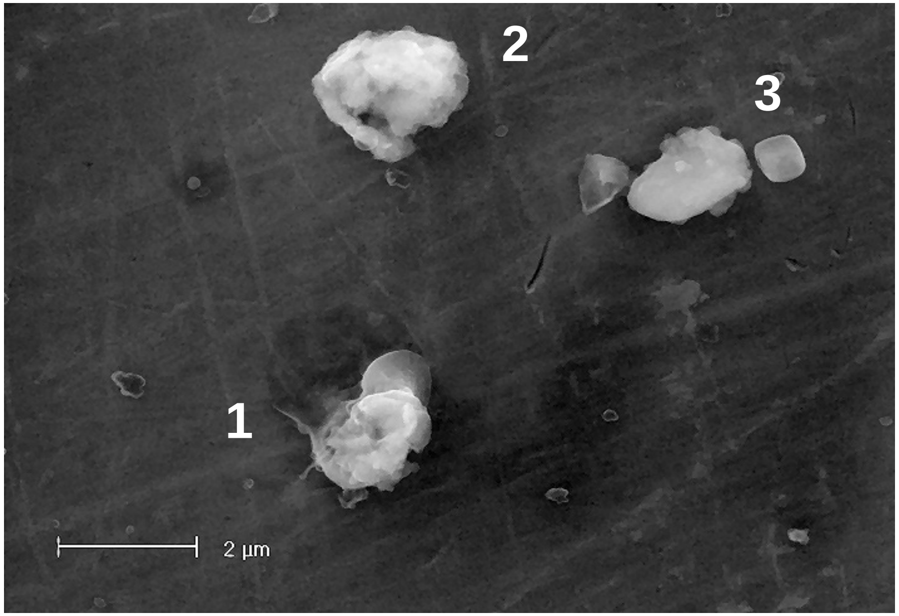
- large particles are preferred to serve as IN:

  ratio IN / total aerosol particles: 1/1000 (submicron)

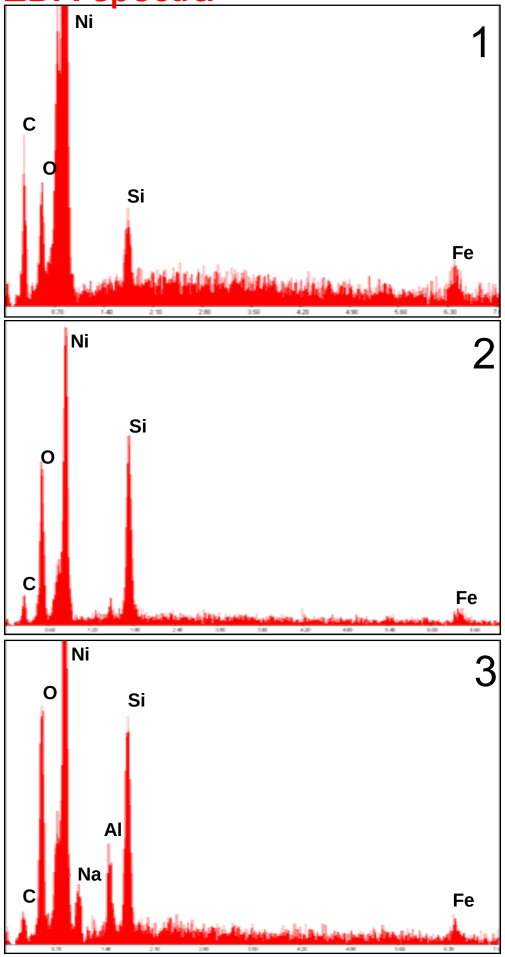
  1/10 (supermicron)

ESEM analysis of single particles sampled downstream of Ice-CVI and interstitial inlet

Courtesy of
M. Ebert, M. Inerle-Hof
TU Darmstadt



EDX-spectra

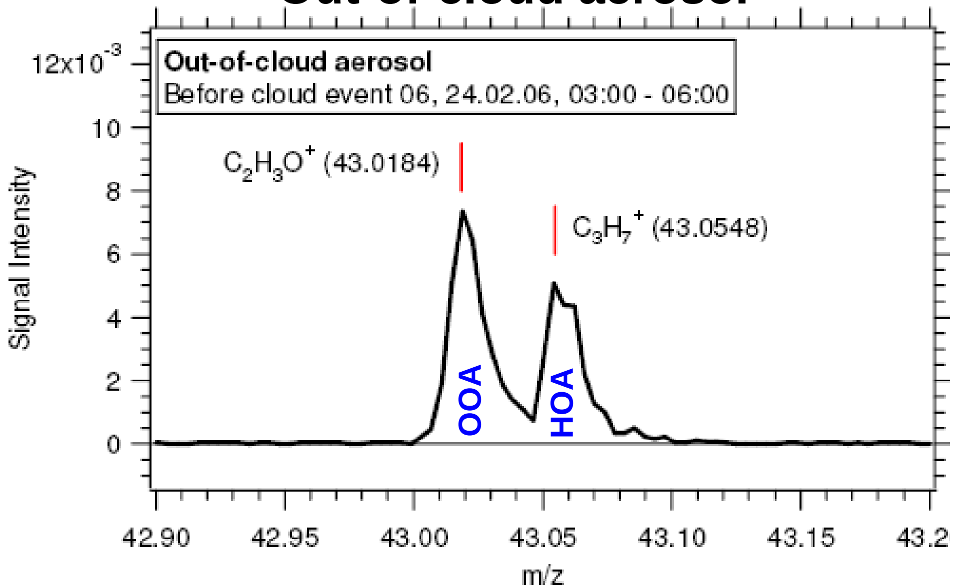


- Carbonaceous material and silicates are enriched in ice residual

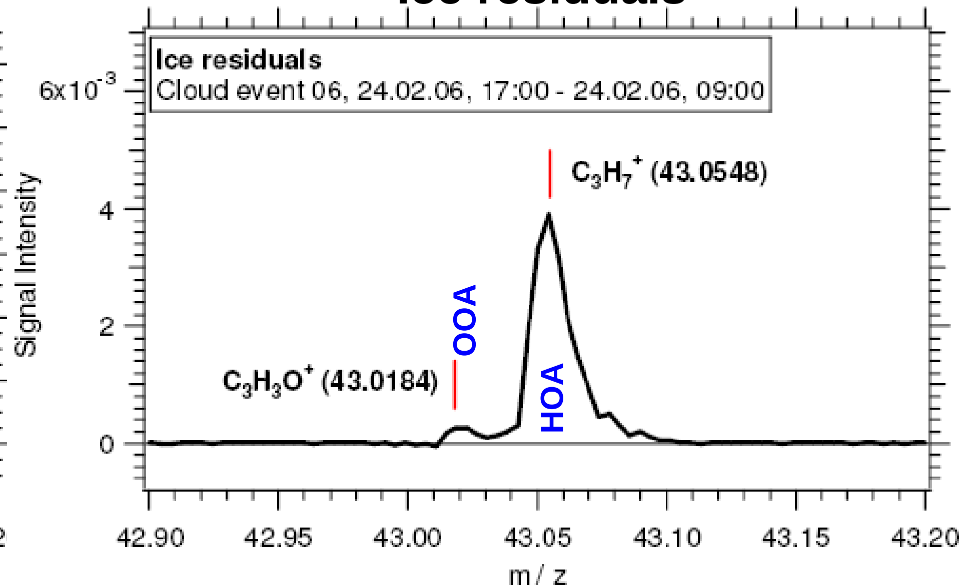
HR-ToF-AMS: Partitioning of Oxygenated Organic Aerosol (OOA) and Hydrocarbon-like Organic Aerosol (HOA):

Courtesy of
J. Schneider MPI Mainz

Out-of-cloud aerosol



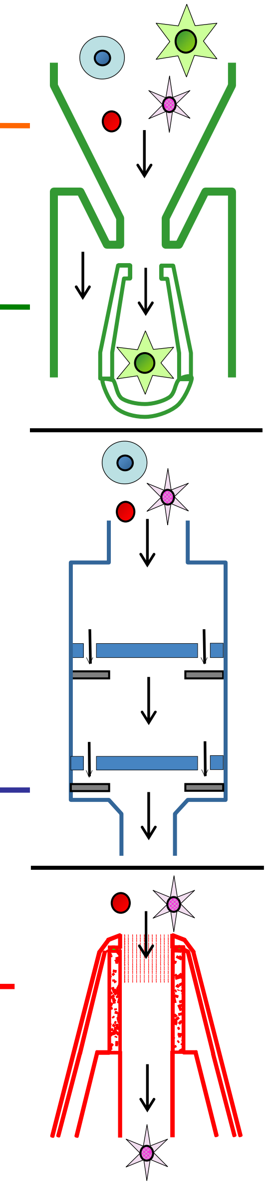
Ice residuals



- Enrichment of hydrocarbon-like organic aerosol (HOA) in ice residuals.

Operating principle of Ice-CVI

S. Mertes, IfT Leipzig



Omni-directional inlet
removes precipitation particles > 50 µm

Virtual Impactor
removes cloud particles > 20 µm

Pre Impactor
removes supercooled drops > 5 µm

Counterflow Virtual Impactor
removes interstitial particles < 5 µm

➢ sampling residual particles (ice nuclei) of small ice crystals (5-20 µm diameter)

Abundances and properties of CCN and IN

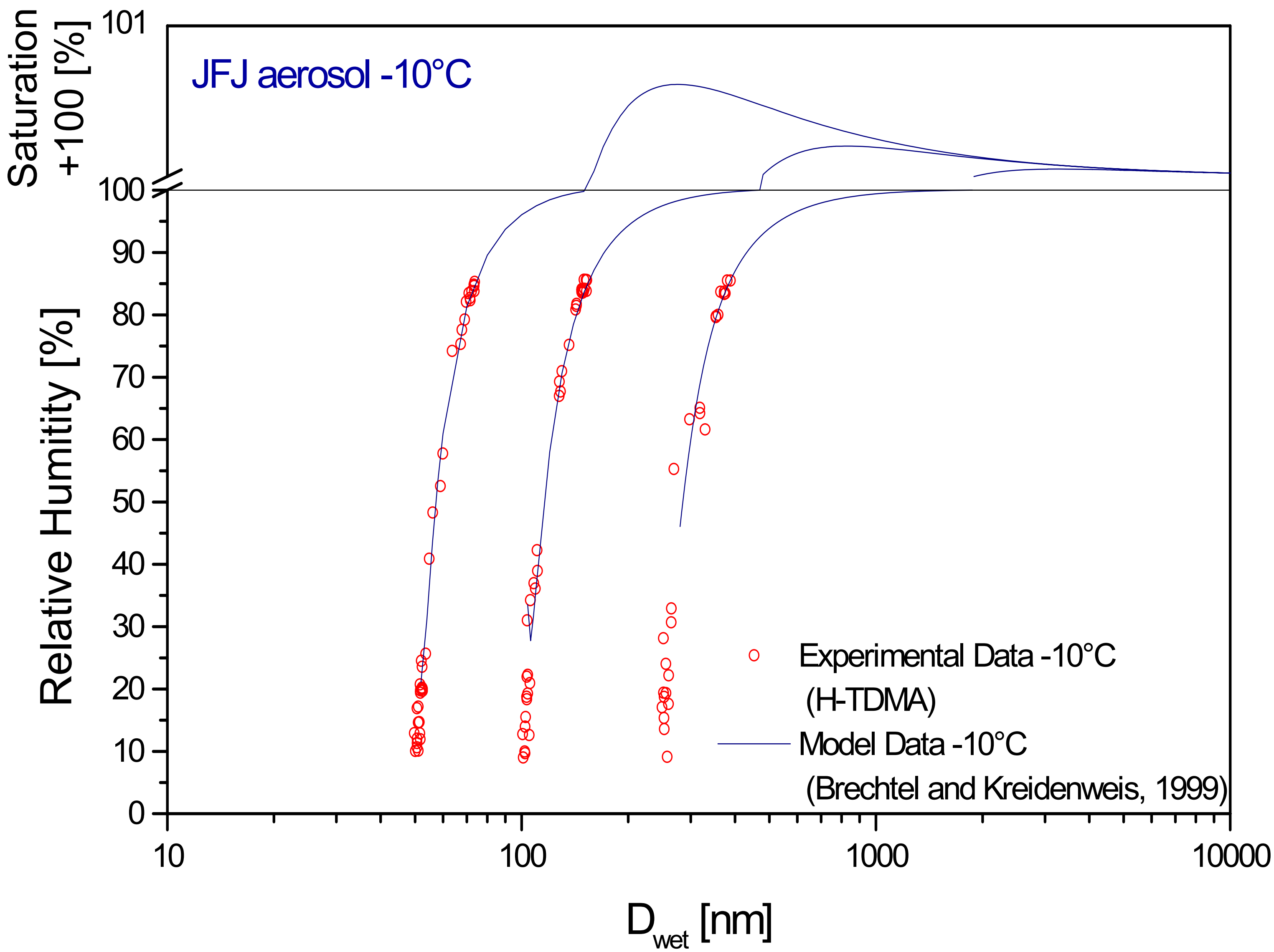
- **CCN:**
  10-50% (@ SS= 0.2-0.3%),
  depending on particle size (D > ~60 nm) and hygroscopic properties

- **IN:**
  < ~1%,
  typically insoluble and large particles
  (bacteria, leaf litter, pollen, mineral dust components, soot)

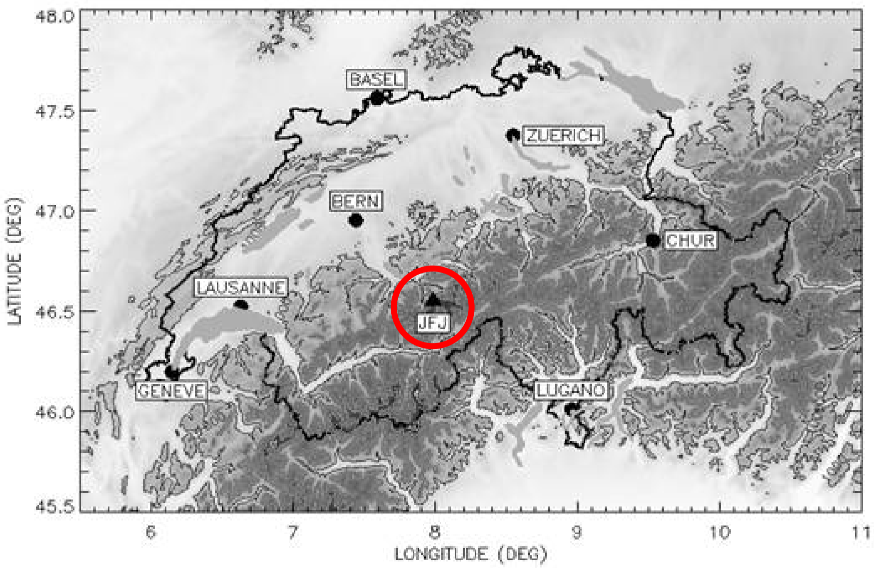
Conclusions

- Aging processes result in coating of BC with soluble components
  - ✓ Internal mixture of JFJ aerosol
  - ✓ Influence on hygroscopic properties of soot particles
- In liquid clouds
  - ✓ BC is incorporated into cloud droplets as bulk aerosol
  - ✓ 60% of BC mass is incorporated into cloud droplets and ice crystals (wet deposition of BC increases)
- In mixed-phase clouds
  - ✓ Incorporation of BC is considerably lower (Bergeron-Findeisen process)
  - ✓ BC is enriched by 20% in the ice phase (influence on cloud optical properties)
  - ✓ Ice nuclei mainly consist of BC and refractory material
- Summary:
  Incorporation of BC into cloud droplets and ice crystals for an aged aerosol
  - ✓ Increases the wet deposition of BC (influence on lifetime of soot particles)
  - ✓ Influence the optical properties of cloud by possibly increasing the number of CCN and by acting as IN

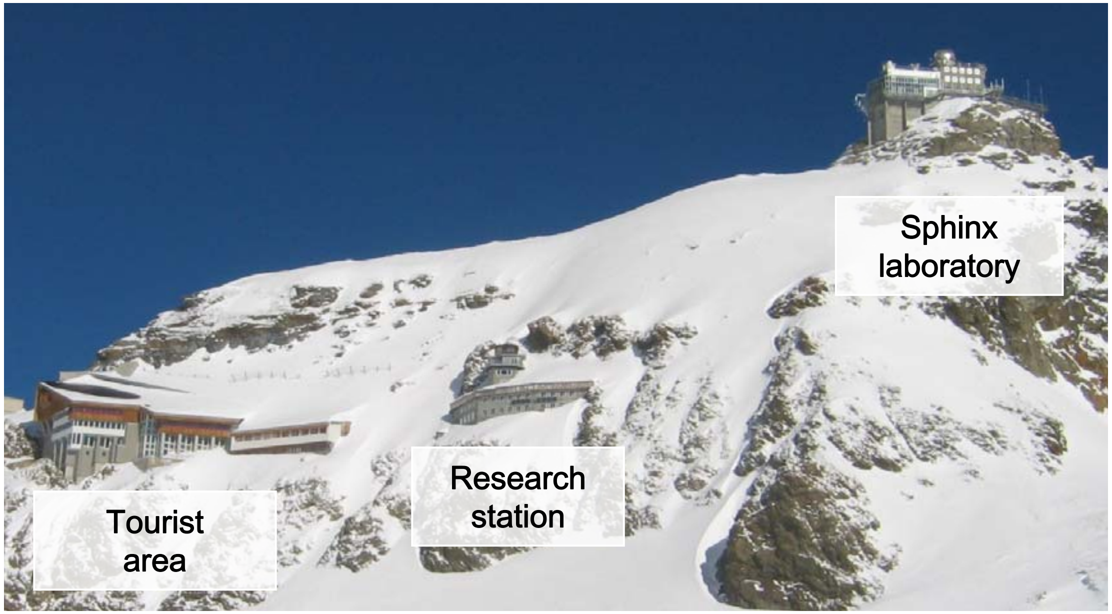
H-TDMA & Modelling data



Jungfraujoch, 3580 m a.s.l.

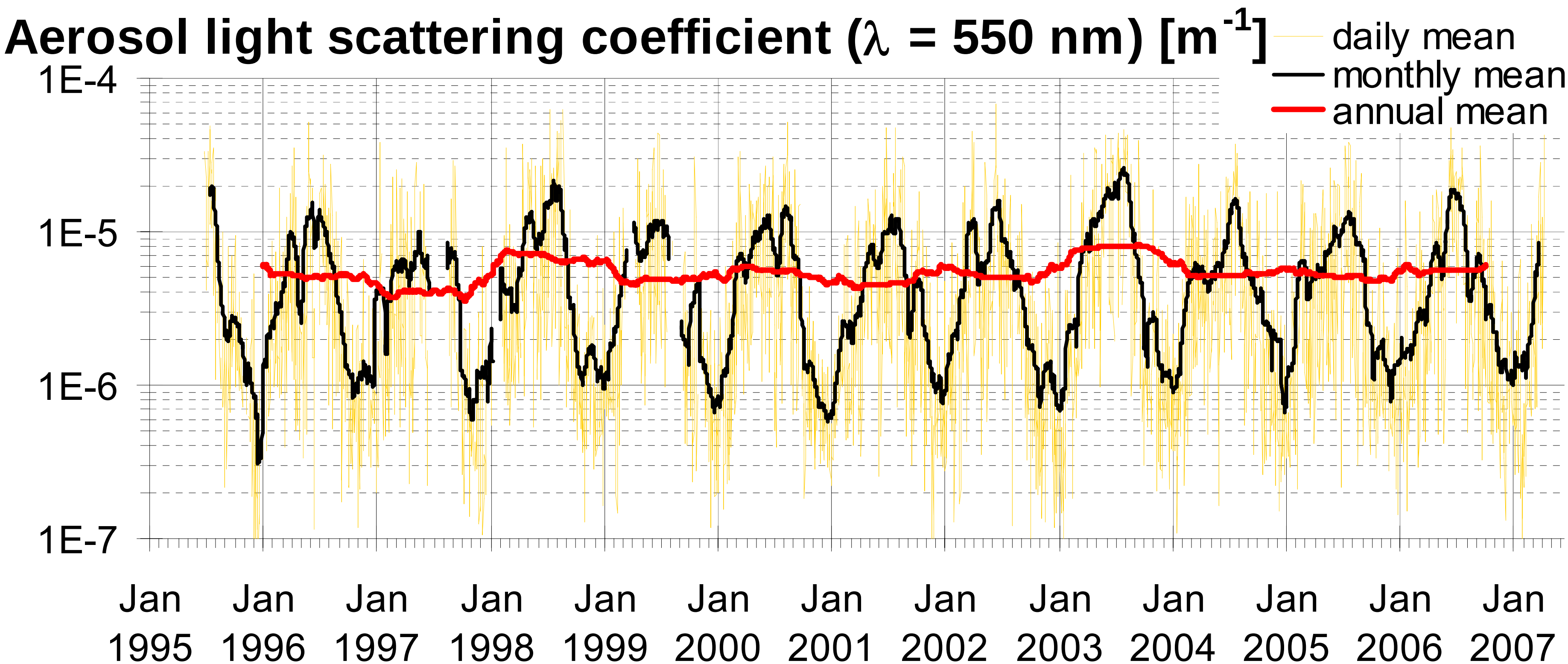


- GAW station
- Few local emissions
- Good infrastructure
- Free troposphere
- Aged aerosol
- 37% cloud occurrence



Current GAW aerosol instrumentation on the Jungfraujoch:

Parameter	Instrument
➢ Particle number concentration	CPC (TSI 3010)
➢ Scattering coefficient at various wavelengths	Nephelometer (TSI 3563)
➢ Absorption coefficient at various wavelengths	Aethalometer (AE-31)
➢ Black carbon concentration	MAAP
➢ Aerosol major ionic composition (PM1 and TSP)	Filter packs
➢ Aerosol mass (PM1 and TSP)	Betameter and HiVol

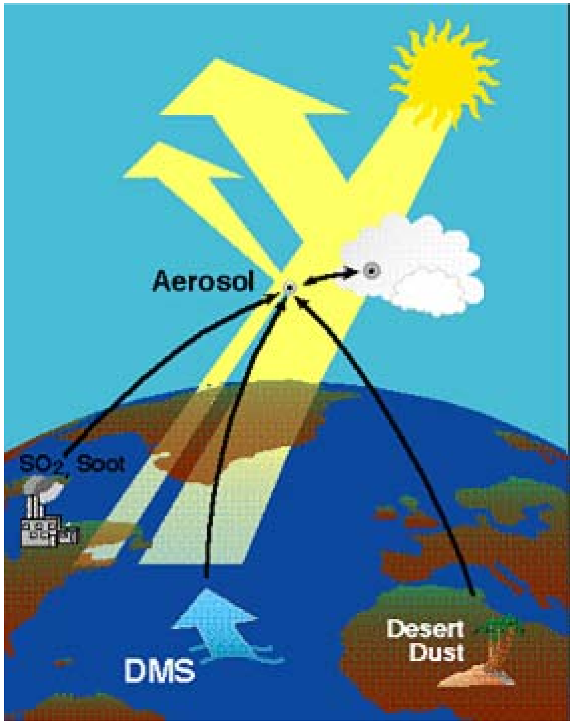


Objectives of CLACE

CLACE = Cloud and Aerosol Characterization Experiments

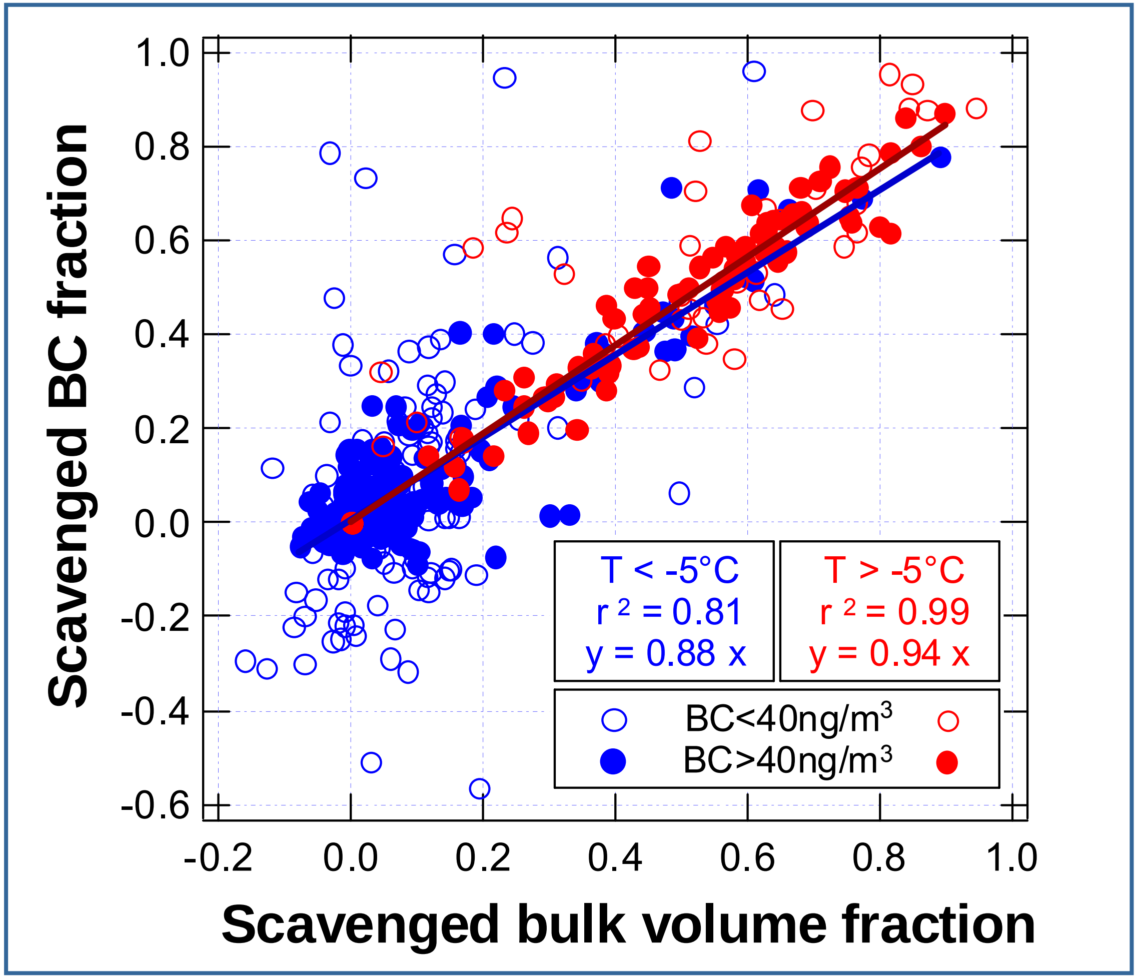
- Physical and chemical characterization of aerosol particles in general and of CCN and IN in particular

- Quantification of aerosol partitioning in mixed-phase clouds



BC scavenging versus bulk volume scavenging

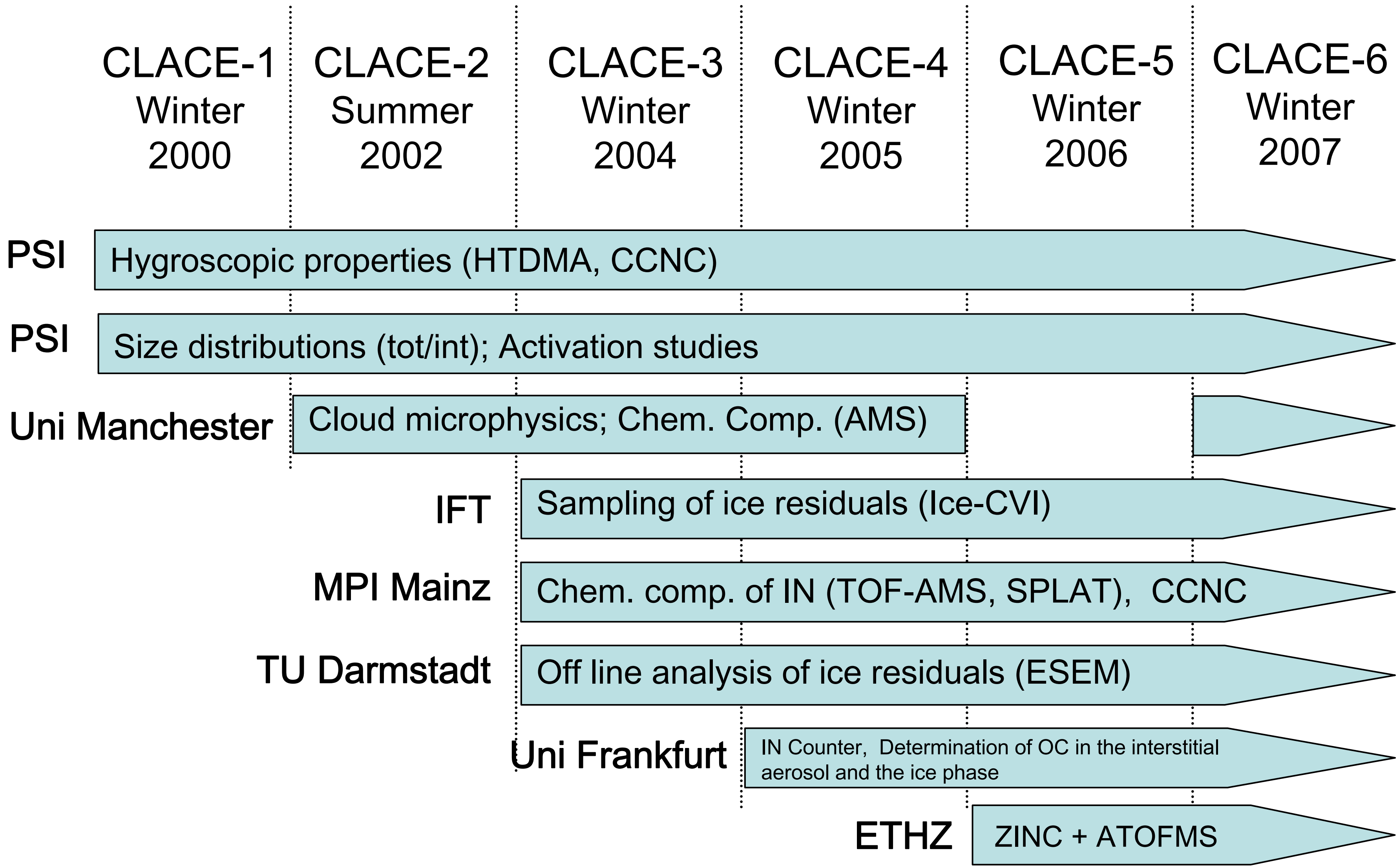
Cozic J. et al., ACP, 7, 1797–1807, 2007



BC mass is scavenged as total volume aerosol

CLACE Experiments

Typically last about 5 weeks, mainly in winter (February/March)



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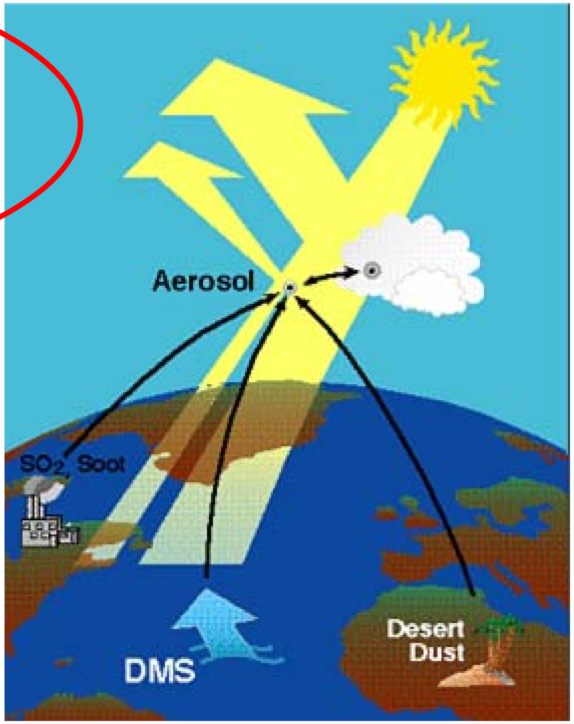
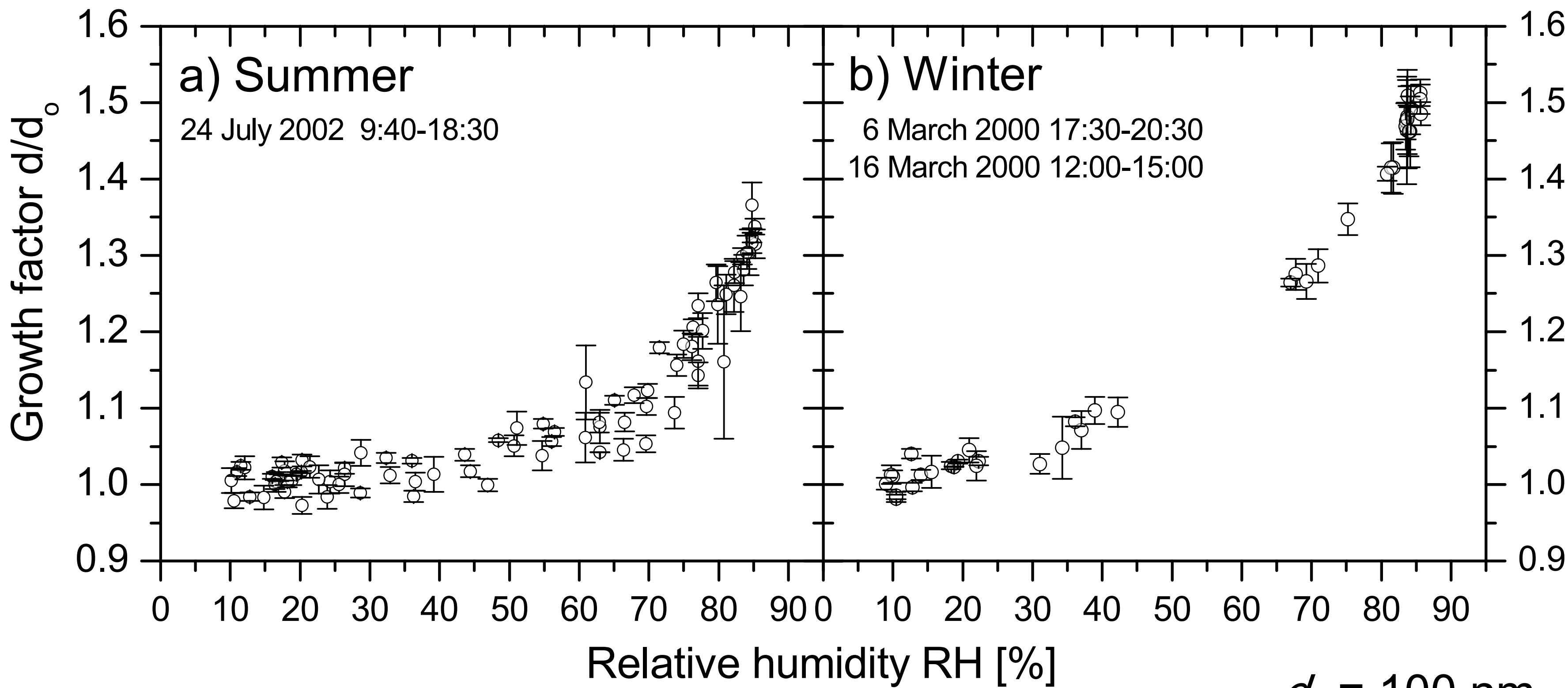


Typical humidograms during Summer (at T = 0°C) and Winter (T = -10°C)

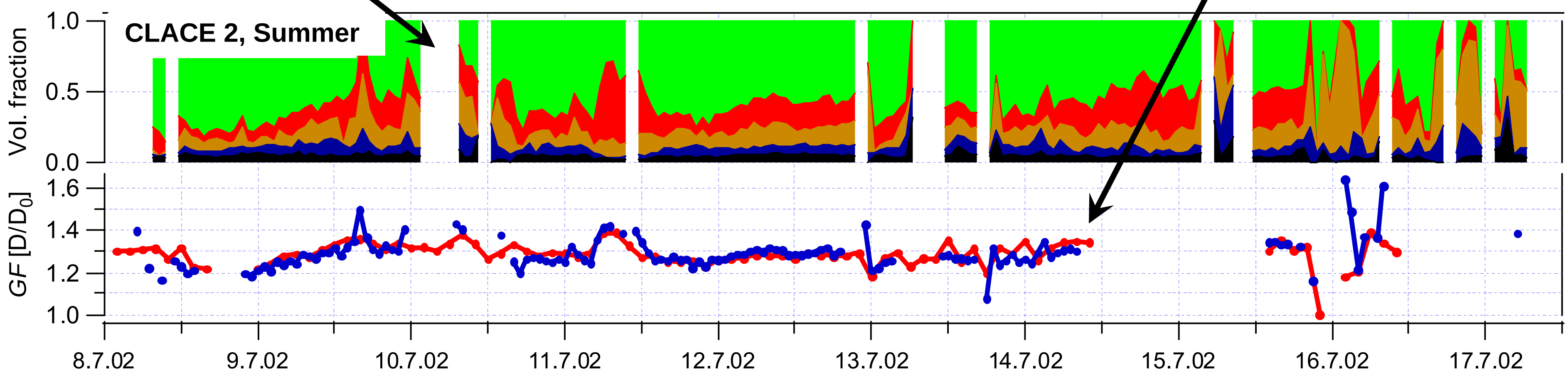


d_o = 100 nm

- Continuous increase of d/d_o as function of RH
- no distinct efflorescence or deliquescence behaviour
- particles are at least partially liquid over a broad RH range

Composition (vol. fractions):
Organics
Sulphate
Ammonium
Nitrate
EC

Measured and modelled hygroscopic growth factors (@85% RH)

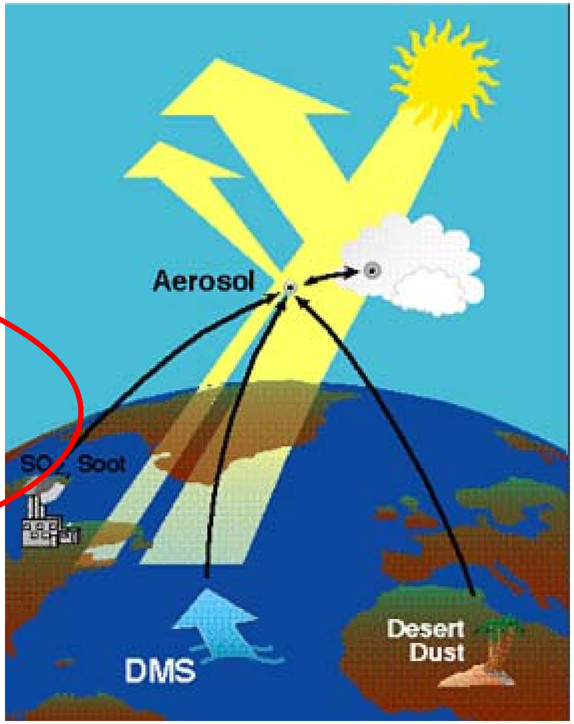


The growth factor measured with the HTDMA agrees with the chemistry measured with the AMS

Objectives of CLACE

CLACE = Cloud and Aerosol Characterization Experiments

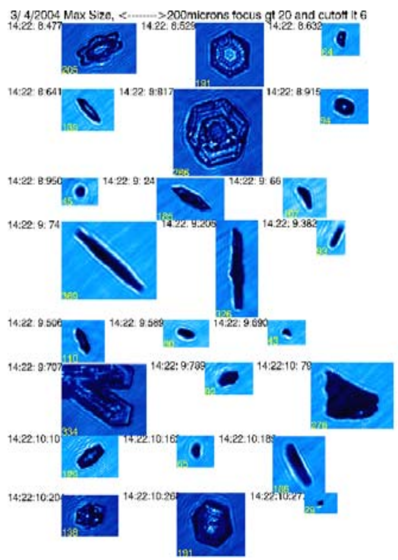
- Physical and chemical characterization of aerosol particles in general and of CCN and IN in particular
- Quantification of aerosol partitioning in mixed-phase clouds



Microphysics, K. Bower, M. Gallagher, M. Flynn, P. Connolly

Size distributions of cloud particles
(FSSP,
phase doppler particle analyzer, TSI PDPA)

Images of cloud particles
-> ice water content
(Cloud particle imager, CPI)



Liquid water content
Particulate volume monitor, PVM)

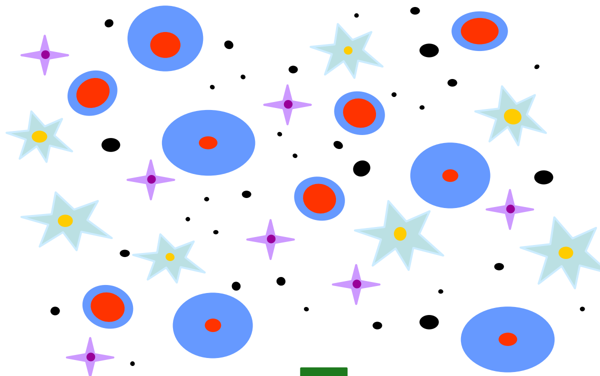
Combining these:

- correcting the PVM for its response to ice
- **ice mass fraction (IMF)**

Inlets during CLACE-3 & 4

1 Total
2 Interstitial
3 Ice-CVI (IFT)
4 Cloud microphysics (UMan)

Inlets during CLACE



Ice CVI inlet:

removes :
- droplets
- int. particles
- large ice crystals

(Size : 5-30 μm)

Interstitial inlet:

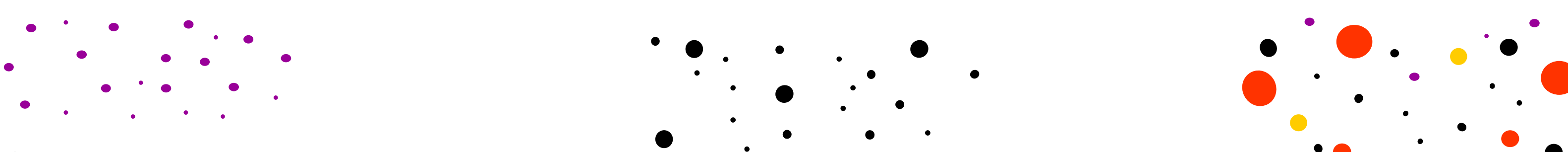
(no activated particles)

removes :
- droplets
- ice crystals

(Size < 2 μm)

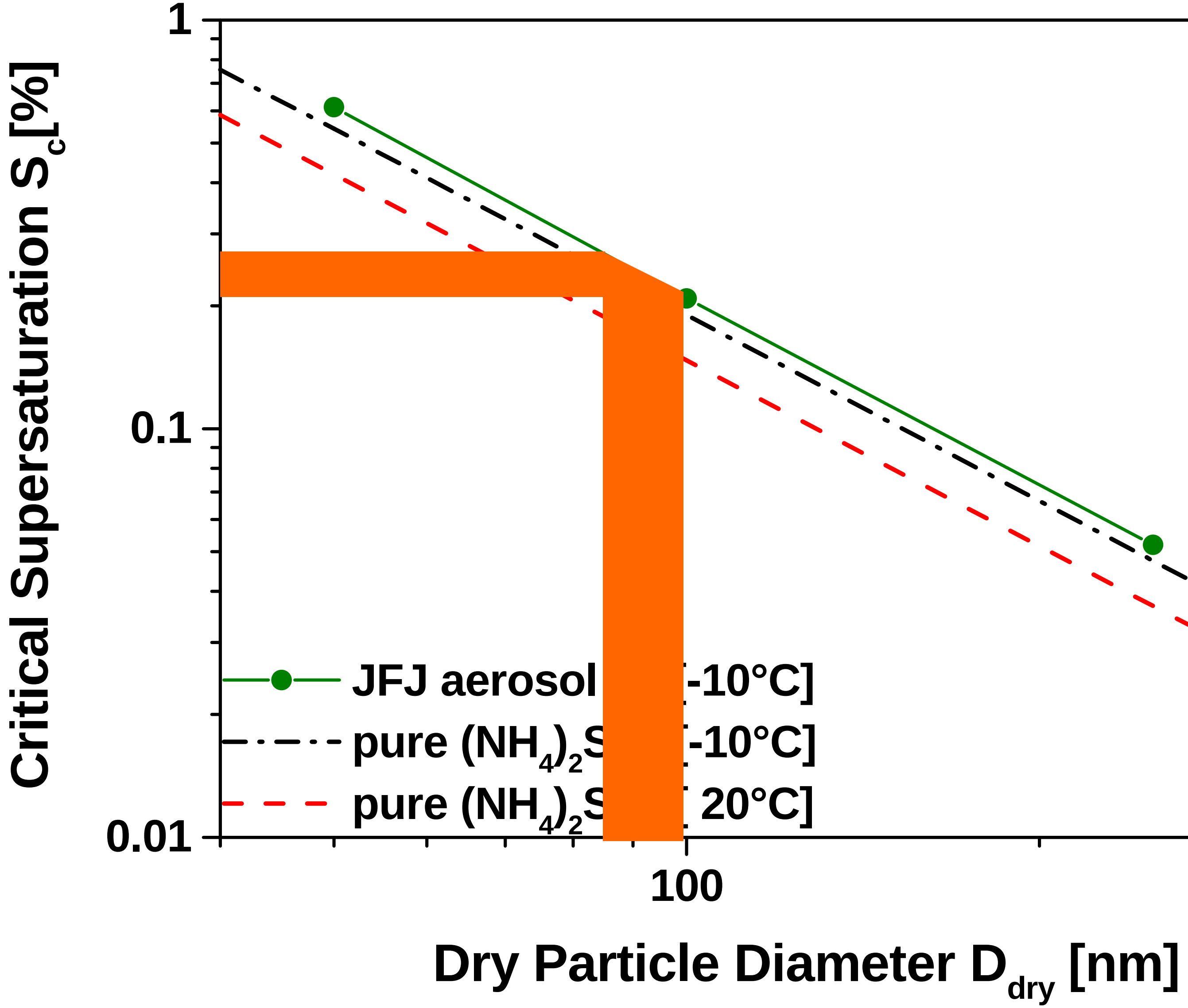
Total inlet :

heated inlet

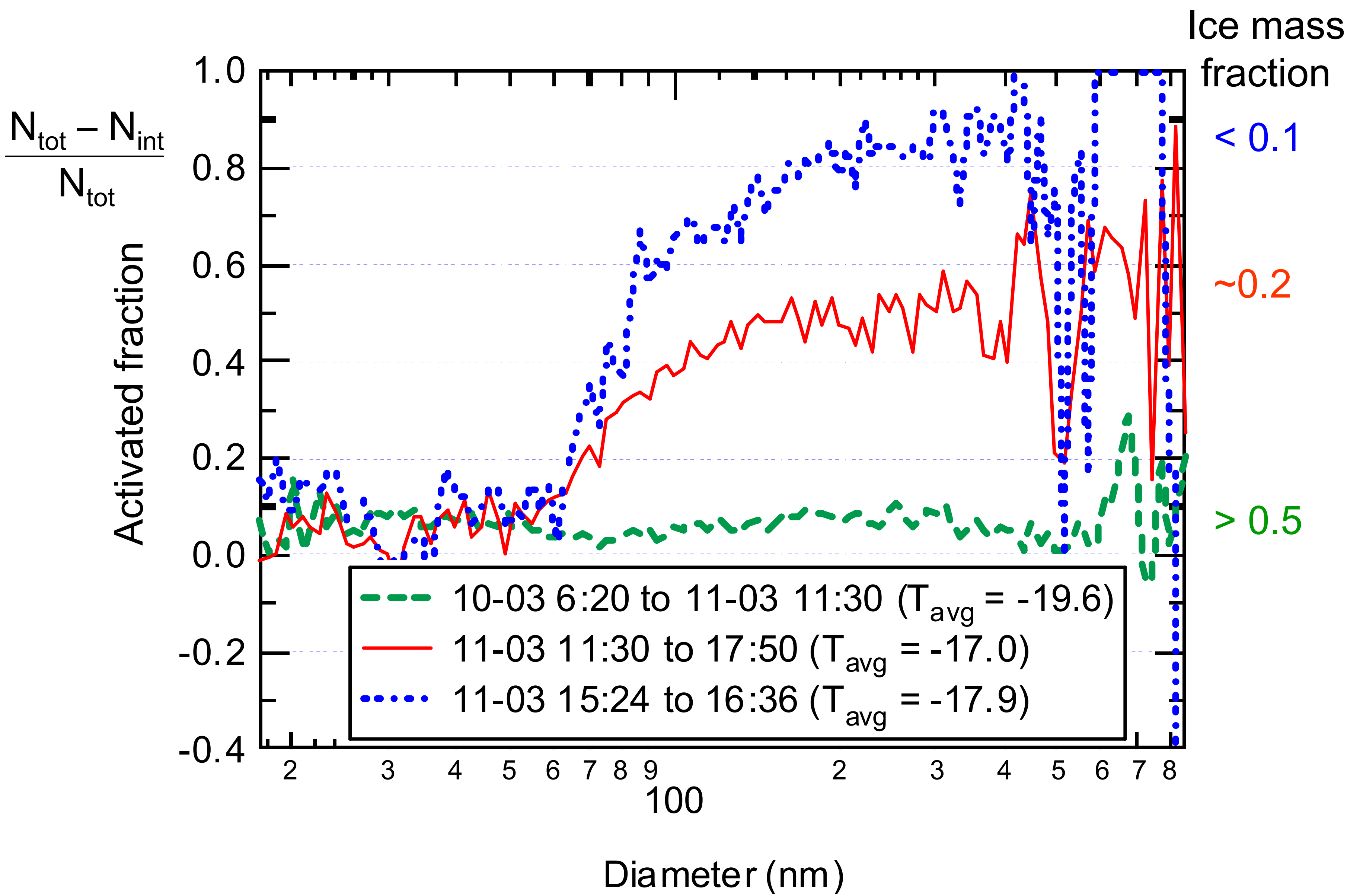


Laboratory (dry aerosol)

Estimated critical supersaturation for the Jungfraujoch aerosol

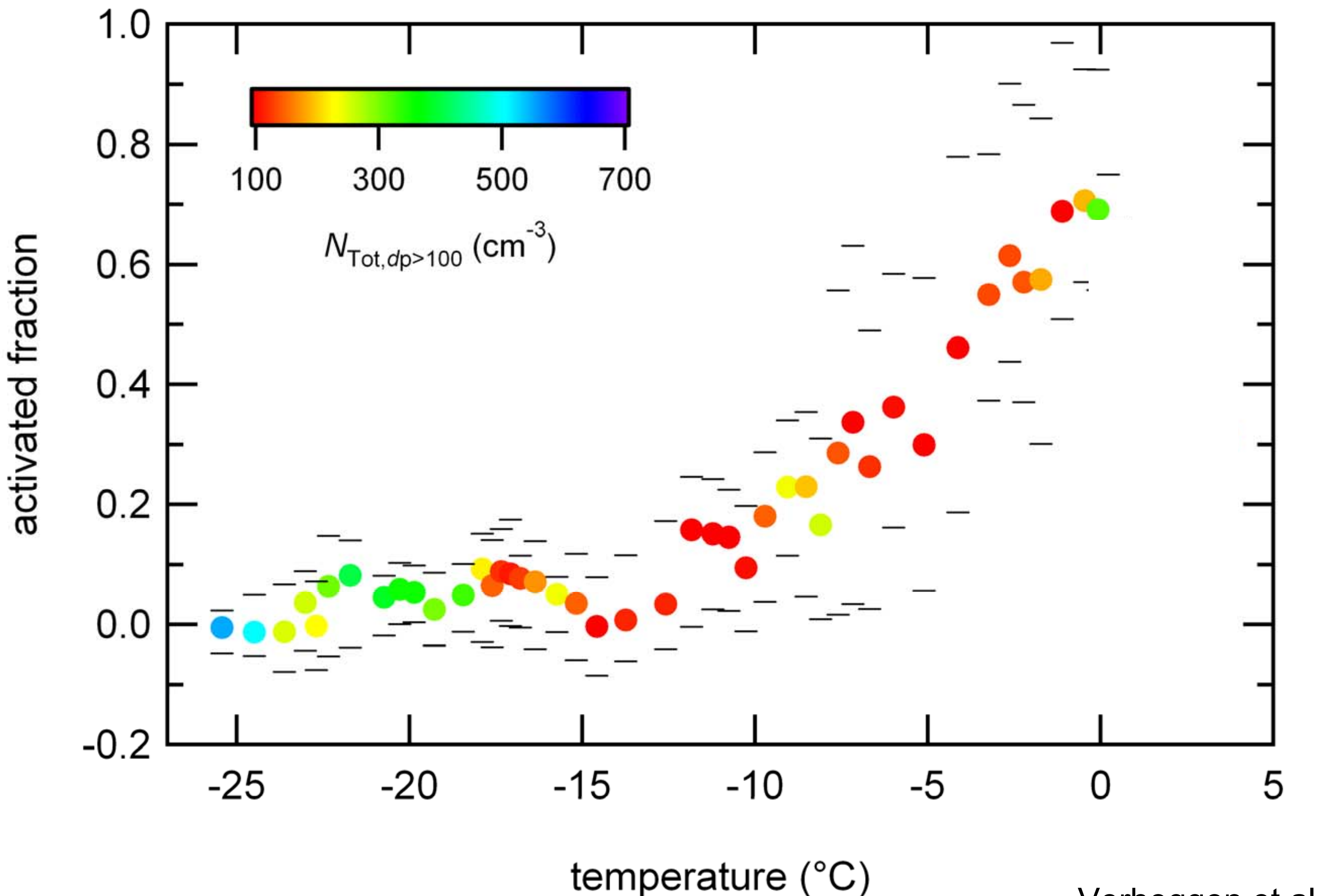


Size resolved activated fractions (for mixed phase clouds)



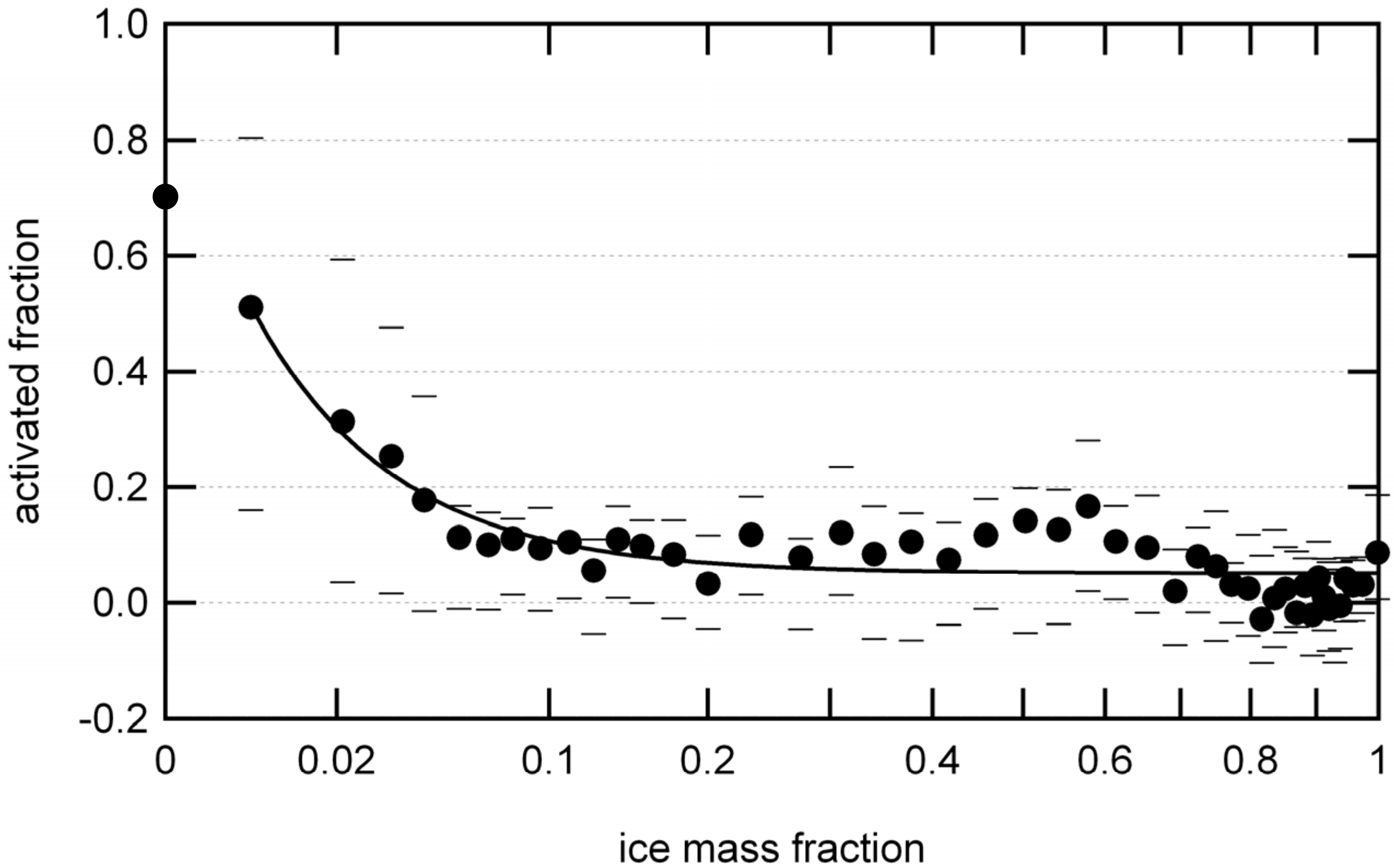
Activated fraction vs. temperature

(over 900 hours of in-cloud measurements)



Verheggen et al. (2007)

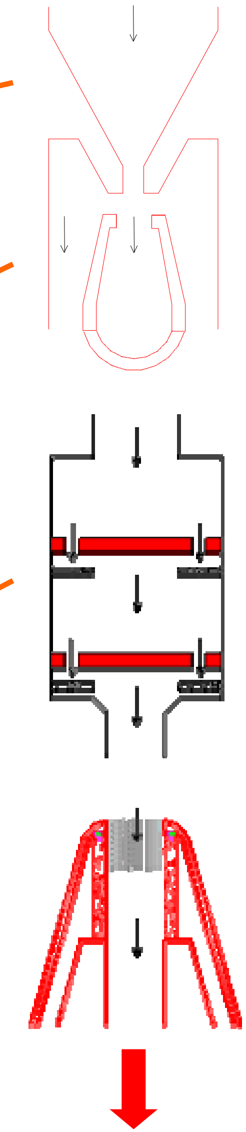
Activated fraction vs. ice mass fraction
(440 hours of in-cloud measurements)



Verheggen et al., submitted to JGR

Operating principle of Ice-CVI

S. Mertes, IfT Leipzig



Virtual Impactor
removes particles
> 30 µm diameter

Pre Impactor
removes
liquid droplets > 5 µm

Counterflow Virtual Impactor
removes particles
< 5 µm diameter

large ice crystals
small ice particles
supercooled droplets
interstitial aerosol

↓

small ice particles
supercooled droplets
interstitial aerosol

↓

small ice particles
interstitial aerosol

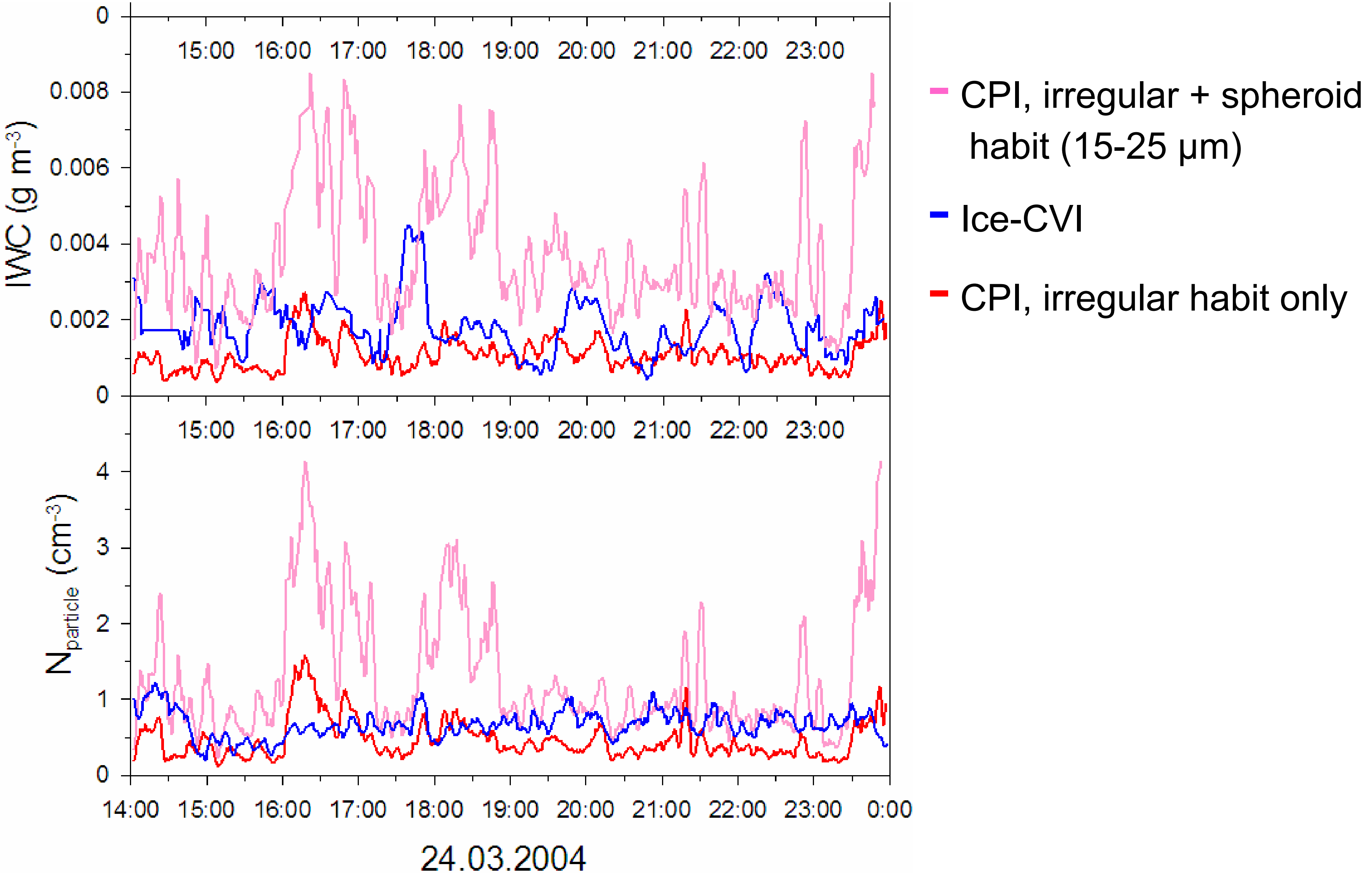
↓

small ice particles

- sampling residual particles (ice nuclei?) of small ice crystals (5-30 µm diameter)

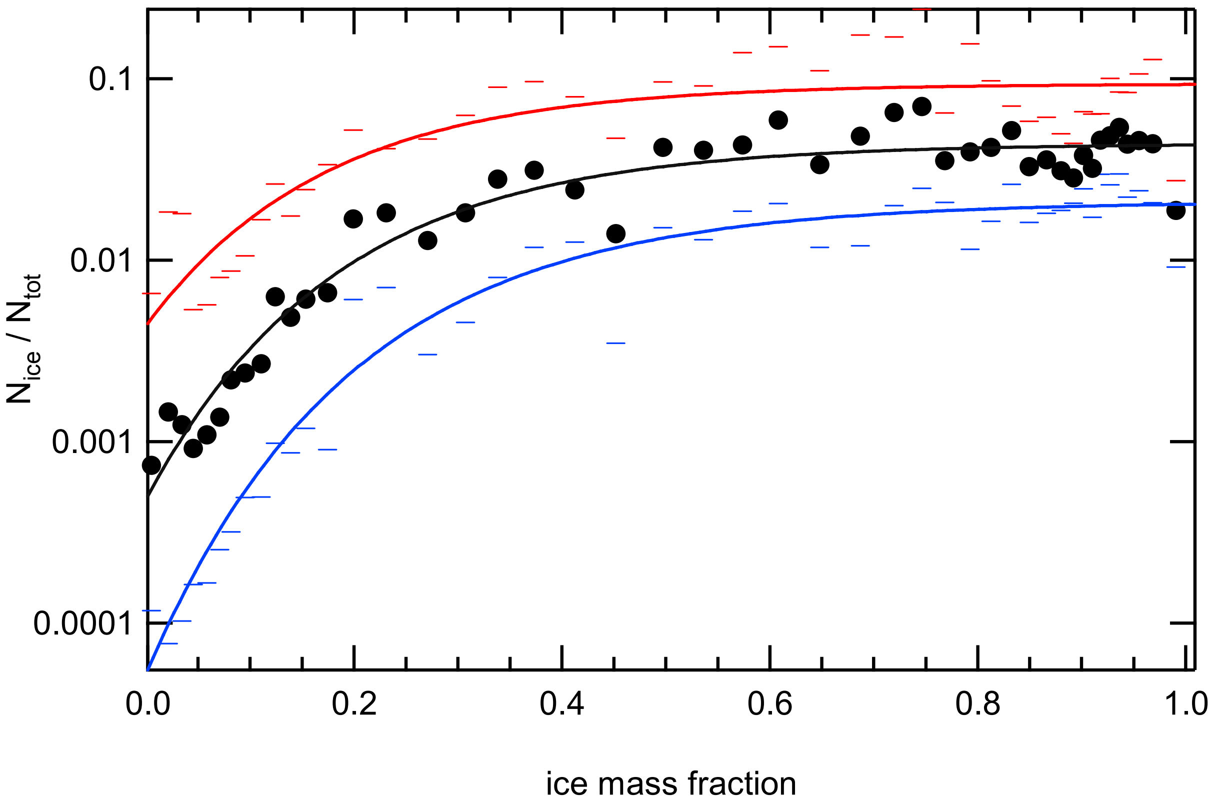
ICE CVI: ice particle sampling efficiency

Courtesy of
S. Mertes, IFT



Ulrike Lohmann's ECHAM4 general circulation model (GCM)

440 hours of non-precipitating, in-cloud observations during winter used **as input for the freezing parameterization** in the ECHAM4 general circulation model. Used as input into ECHAM4 GCM to predict N_{ice} as f(N_{TOT}, IMF)

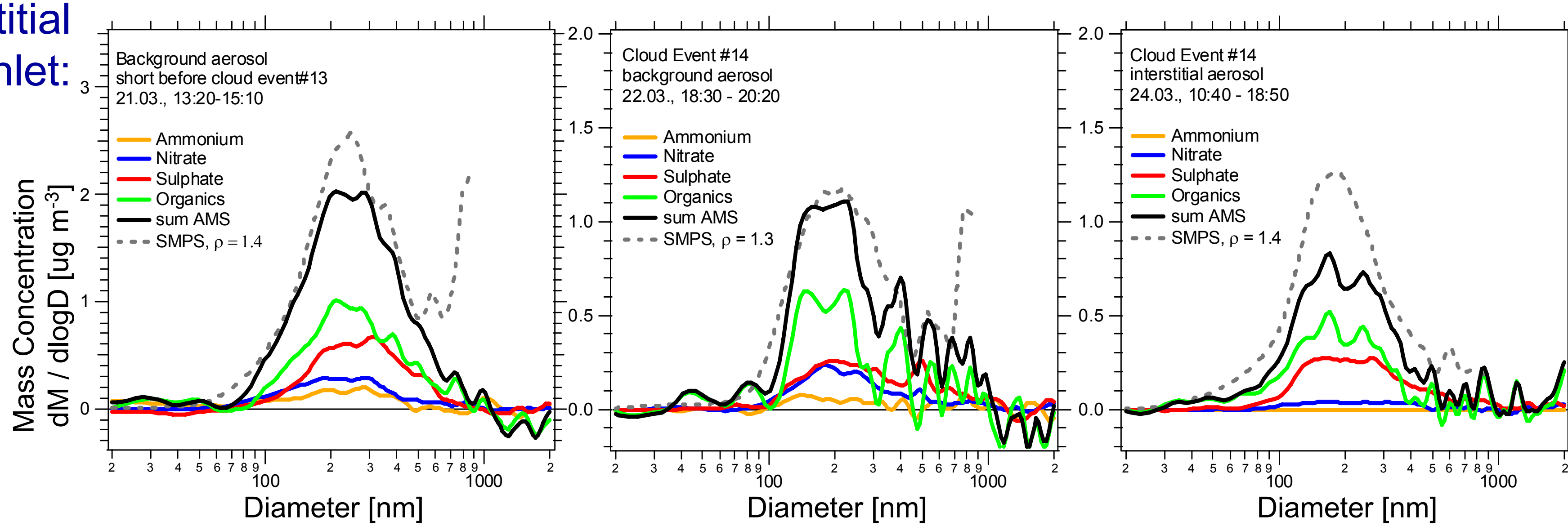


- Model inputs are spatially and chemically resolved aerosol size distributions
- Model treats: transport, chemical transformation of aerosols and precursors, dry, and wet deposition, …
- Includes a fully coupled aerosol-cloud microphysics module (mixed phase clouds)

Comparison AMS-SMPS behind the different inlets…

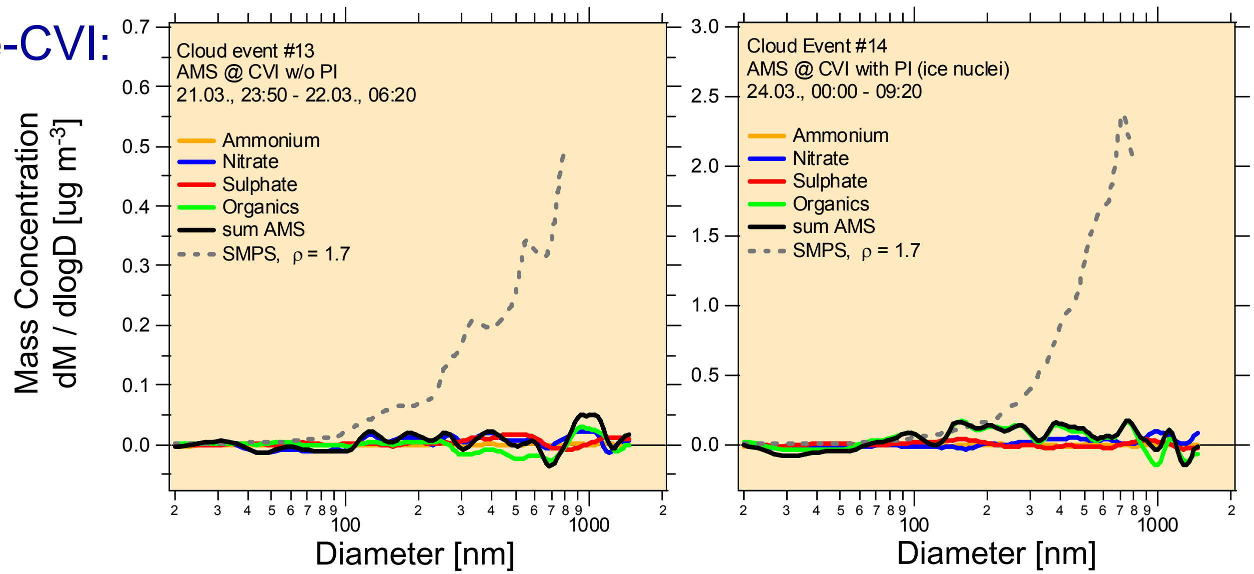
…behind interstitial inlet:

- ➢ Background and interstitial aerosol is composed to about 95% of volatile material



…behind Ice-CVI:

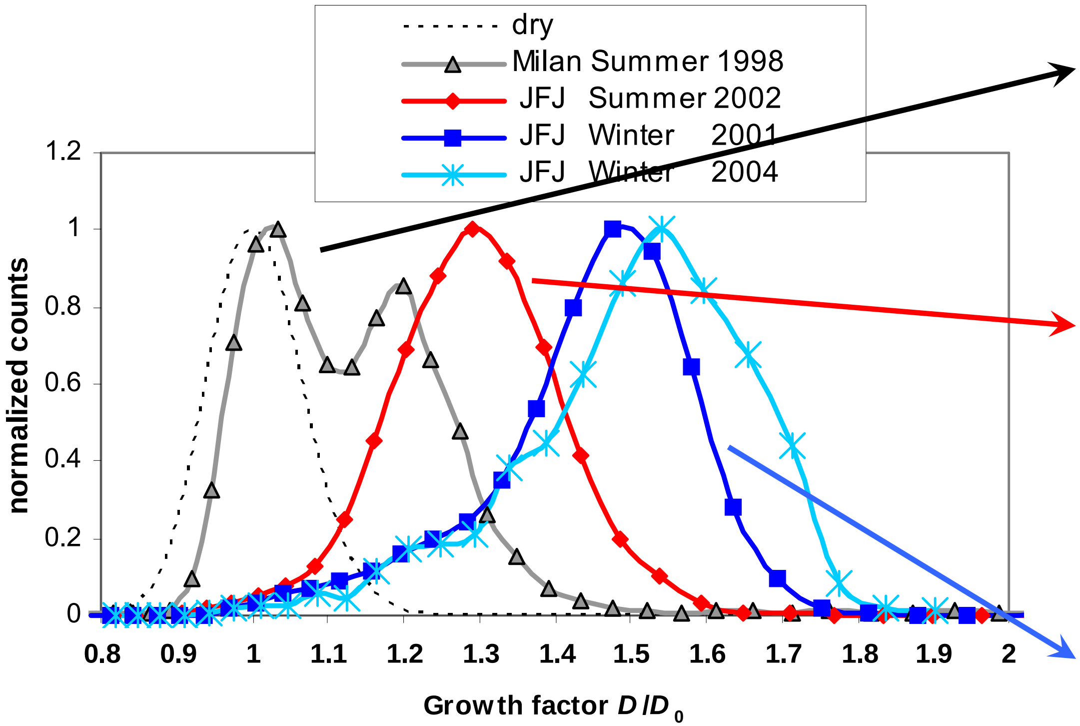
- ➢ Residuals mainly consist of non-volatile material (BC, mineral dust). Larger particles (> 600 nm) dominate the mass distribution of the residuals.



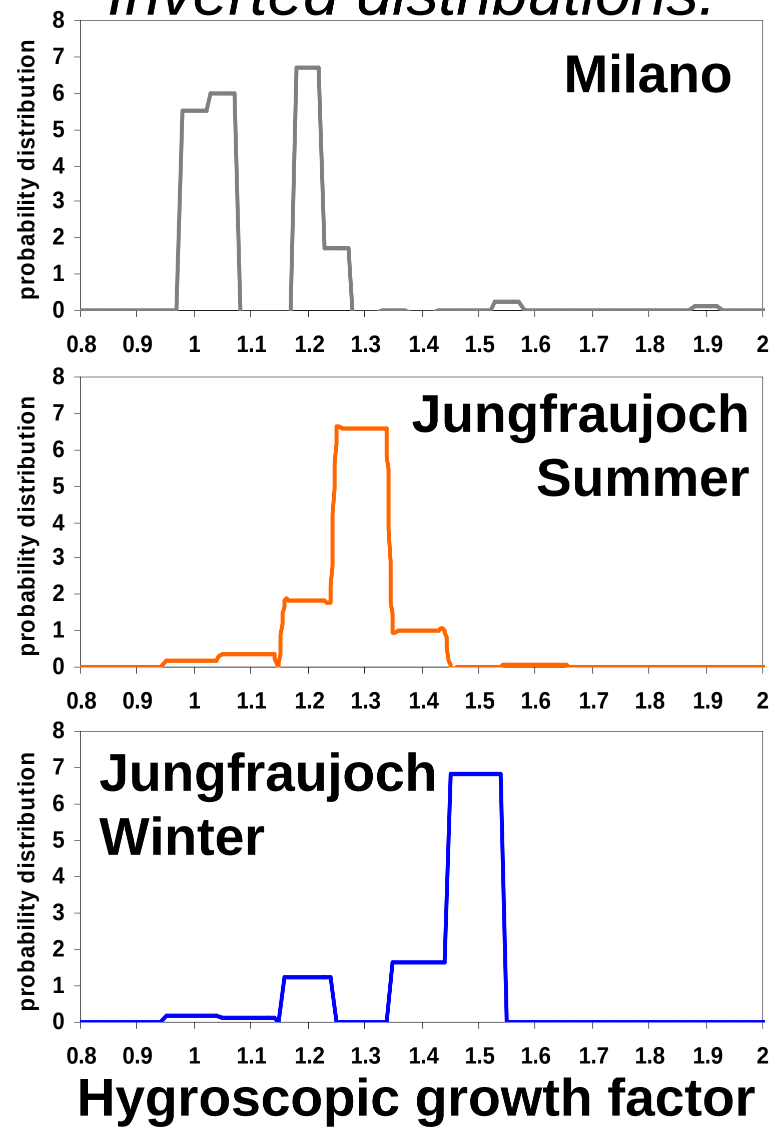
Courtesy of
J. Schneider,
S. Walter; MPI Mainz

H-TDMA growth size distributions at RH = 85% and D_o = 100 nm

Raw distributions:



Inverted distributions:

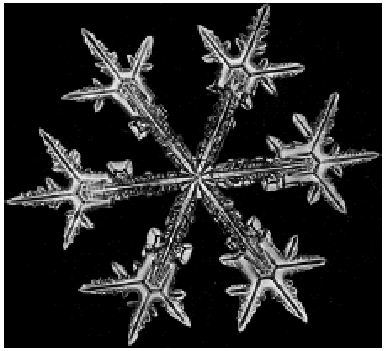


Summer:
~94% of particles with growth factor between 1.2 and 1.5
average GF=1.28
Winter:
“more” and “less” hygroscopic particles
81-85% of particles with GF>1.3
15-19% of particles with GF<1.3

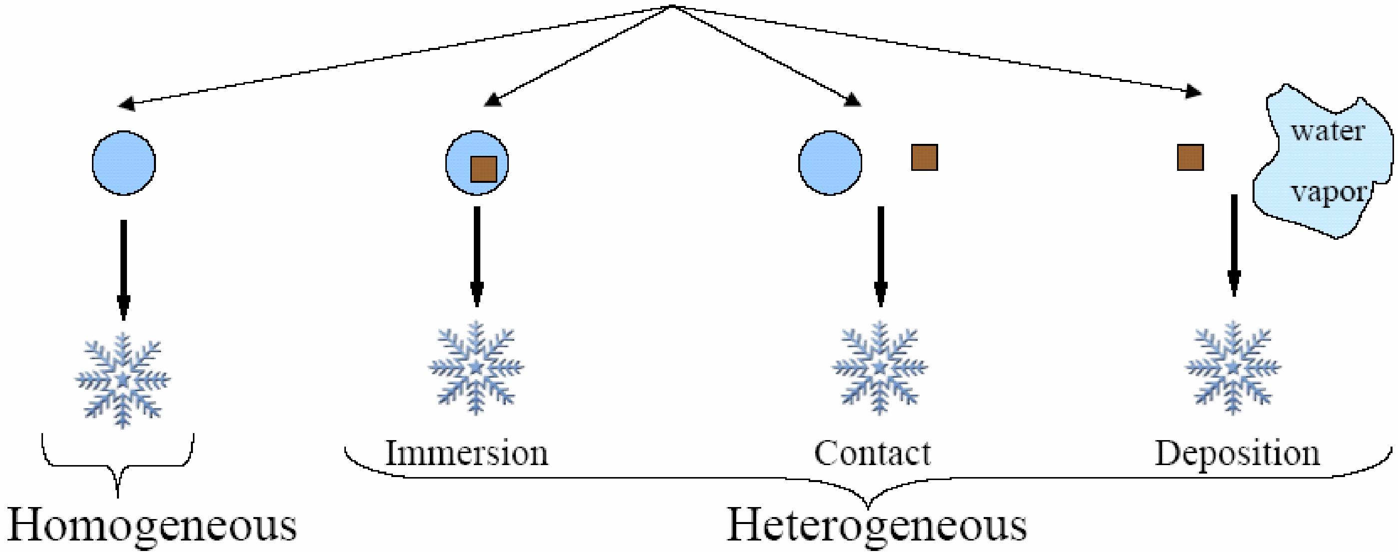
- Aging processes in the atmosphere result in significantly higher growth factors at the Jungfraujoch than in Milano; the less hygroscopic mode nearly disappears.

Ice crystal formation

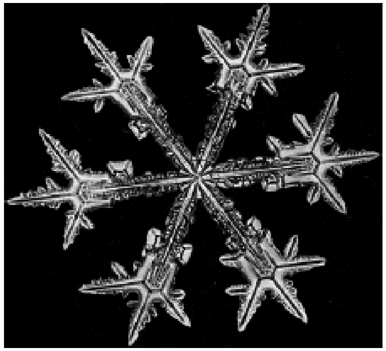
We Know Atmospheric Ice Formation is Important...



But How Does it Occur?



Ice formation is important



- The formation of ice inside supercooled clouds is poorly understood
- Ice formation is important because the majority of precipitation on Earth is initiated via the ice phase
- Ice formation also influences cloud dynamics, latent heat release, chemical processes, particle scavenging, and cloud radiation properties
- Only a few particles (< 1 % in terms of number) act as ice nuclei in our atmosphere

Questions:

- How does ice formation occur?
- Which particles act as ice nuclei in our atmosphere?

ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Incorporating JFJ measurements into GCM

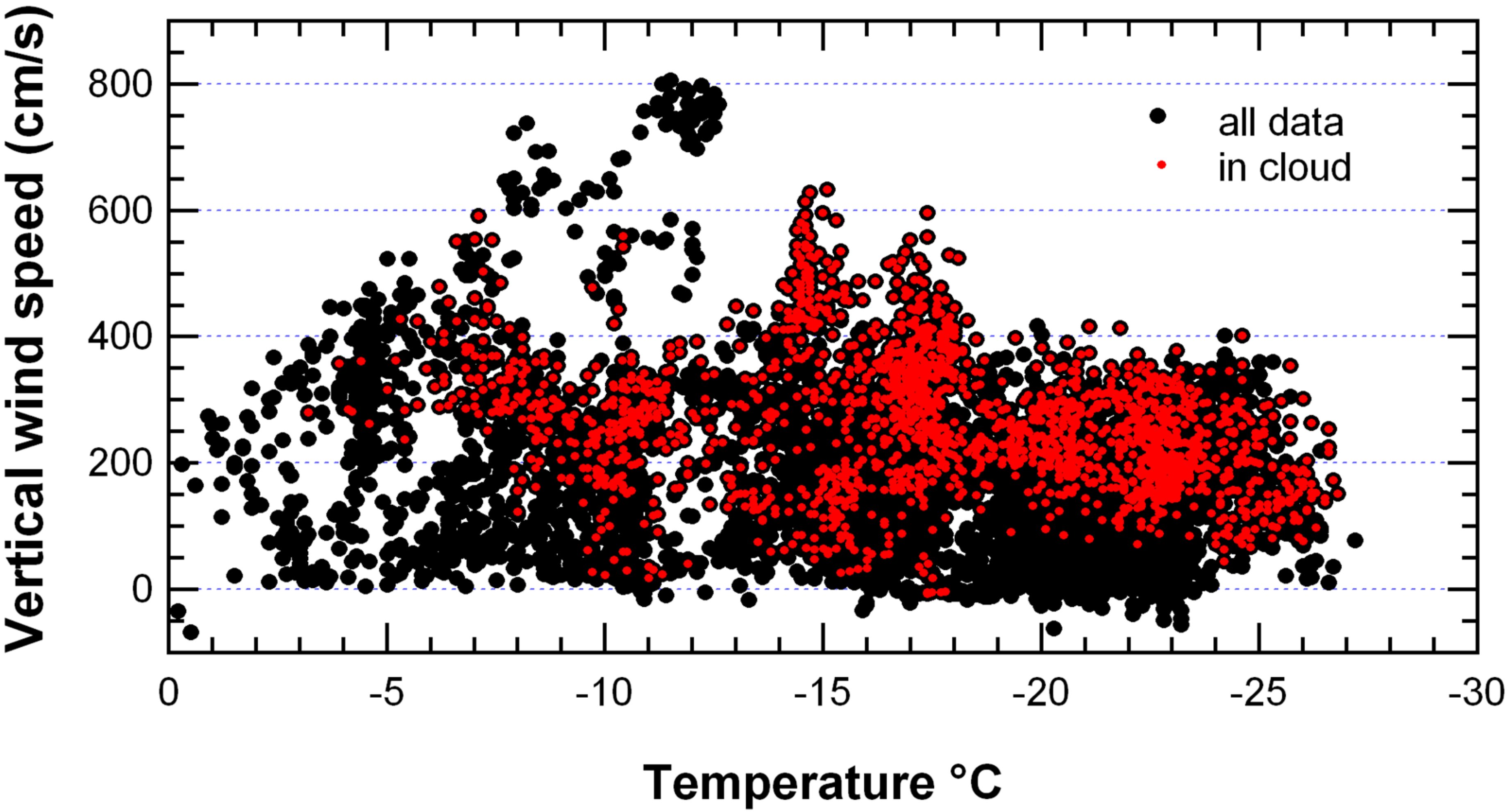
		ECHAM-JFJ	ECHAM-CTL
SW radiation TOA ↑	W/m^2	-0.38	-1.63
LW radiation TOA ↓	W/m^2	-1.14	0.59
net radiation TOA ↓	W/m^2	-1.52	-1.04
IWP ↑	g/m^2	0.4	0.20
LWP ↓	g/m^2	9.52	10.5
Cloud cover ↓	%	-0.77	0.07
Precipitation ↑	mm/d	-0.007	-0.051

CTL:
N_{ice} depends on temperature; *not* on N_{aer}

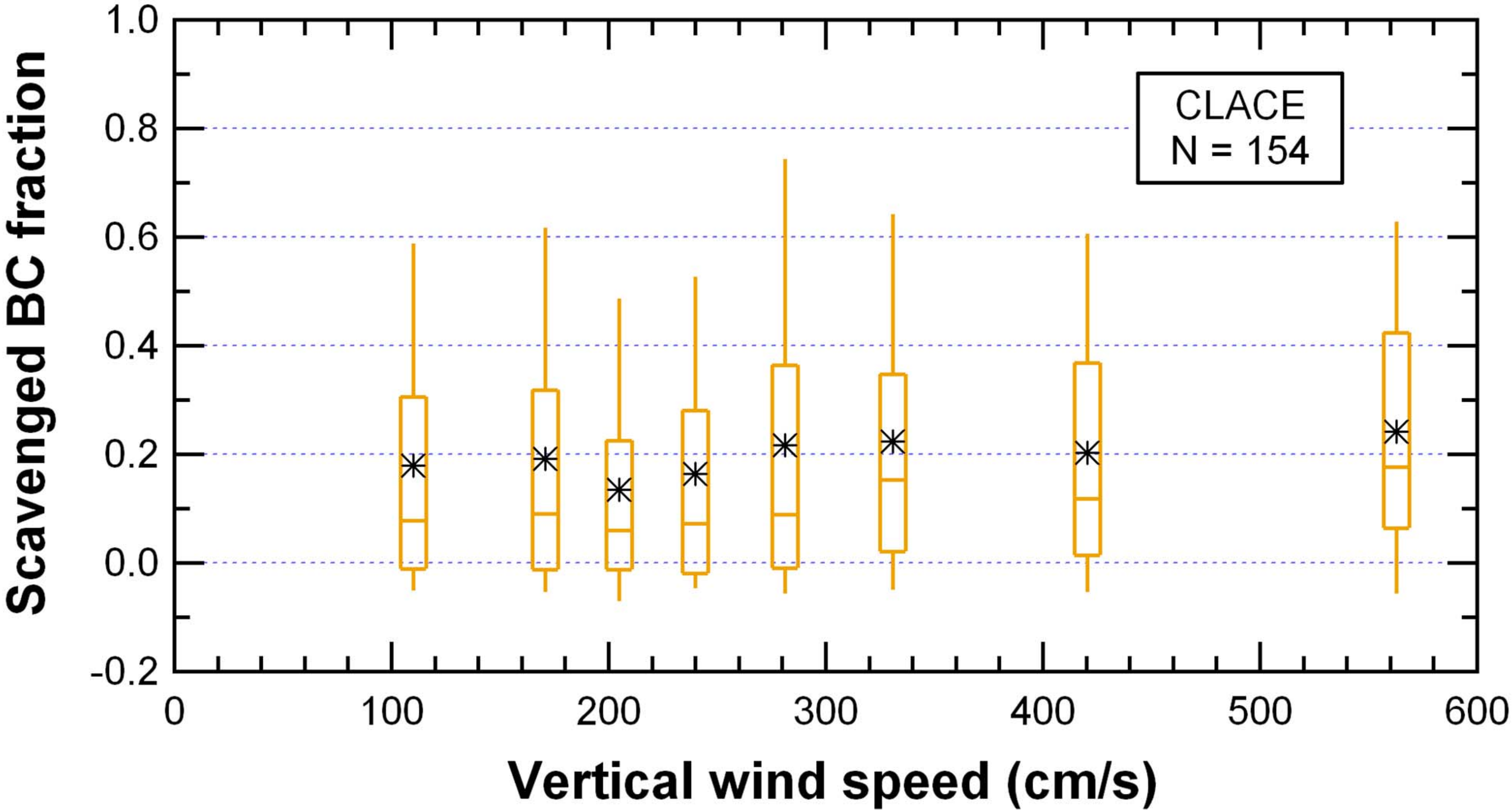
JFJ:
N_{ice} depends on ice mass fraction and on N_{aer}

- JFJ parameterization leads to an enhanced ice phase in the model, which results in a smaller shortwave aerosol indirect effect by increasing precipitation.

Vertical wind speed vs. temperature

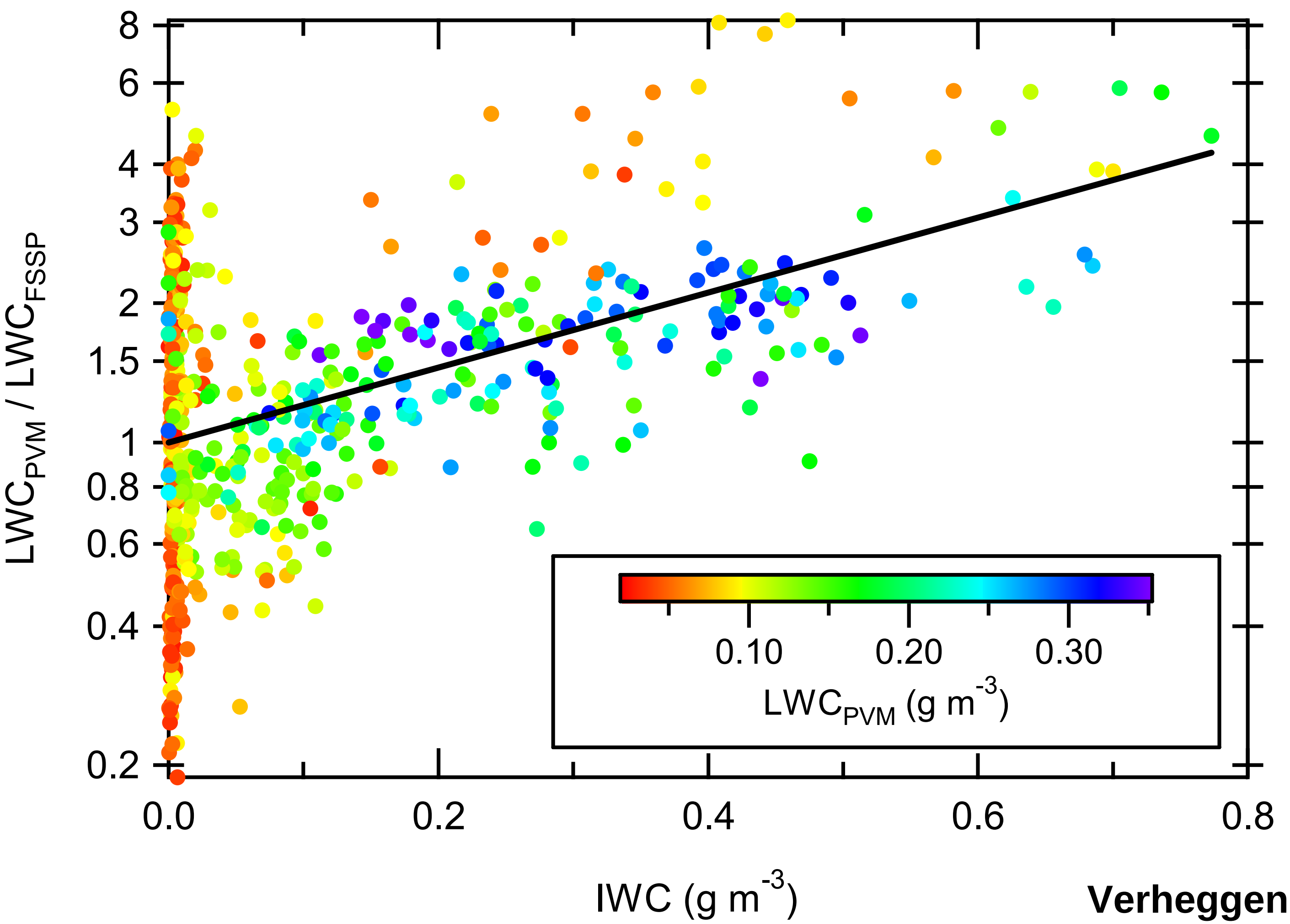


Scavenged BC fraction vs. vertical wind speed



Empirical correction for the PVM response to presence of ice crystals

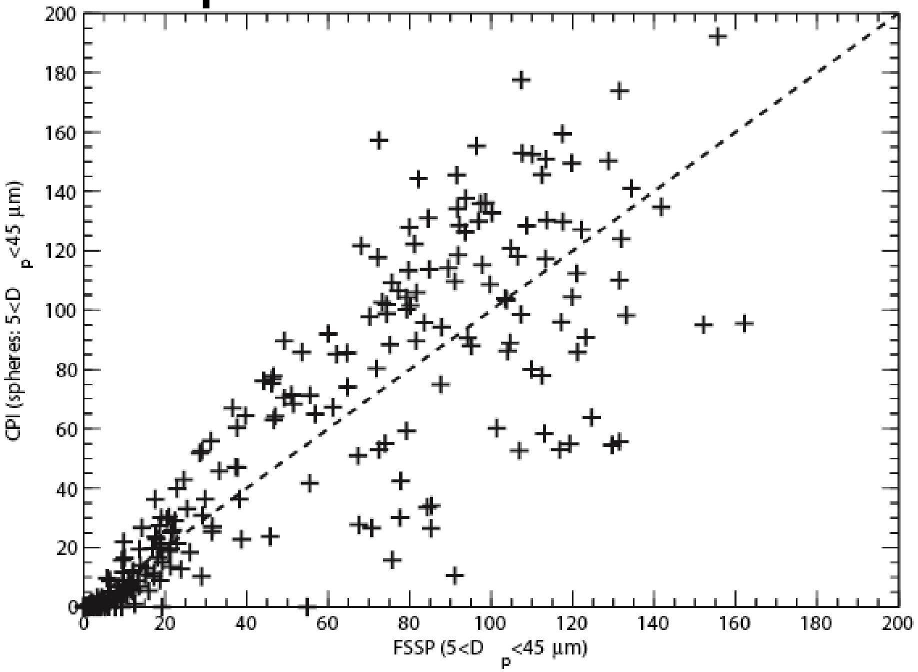
$$LWC_{PVM}(corrected) = \frac{LWC_{PVM}(measured)}{10^{0.81 \times IWC}}$$



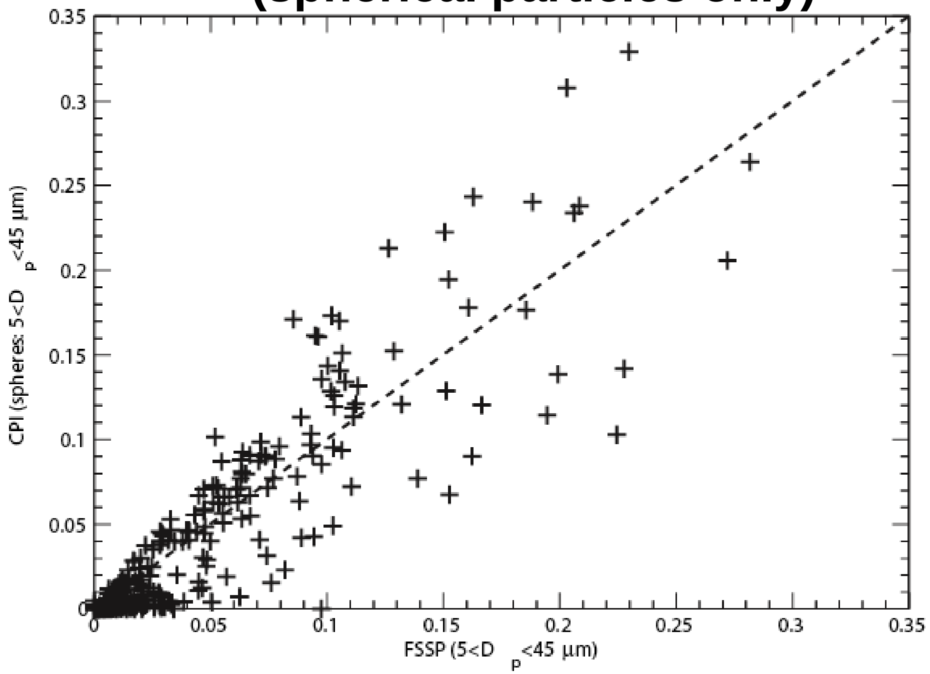
Verheggen et al., submitted to JGR

Intercomparison between FSSP and CPI in the overlapping size range 5 < D < 45 μm in a mixed-phase cloud

droplet number concentration

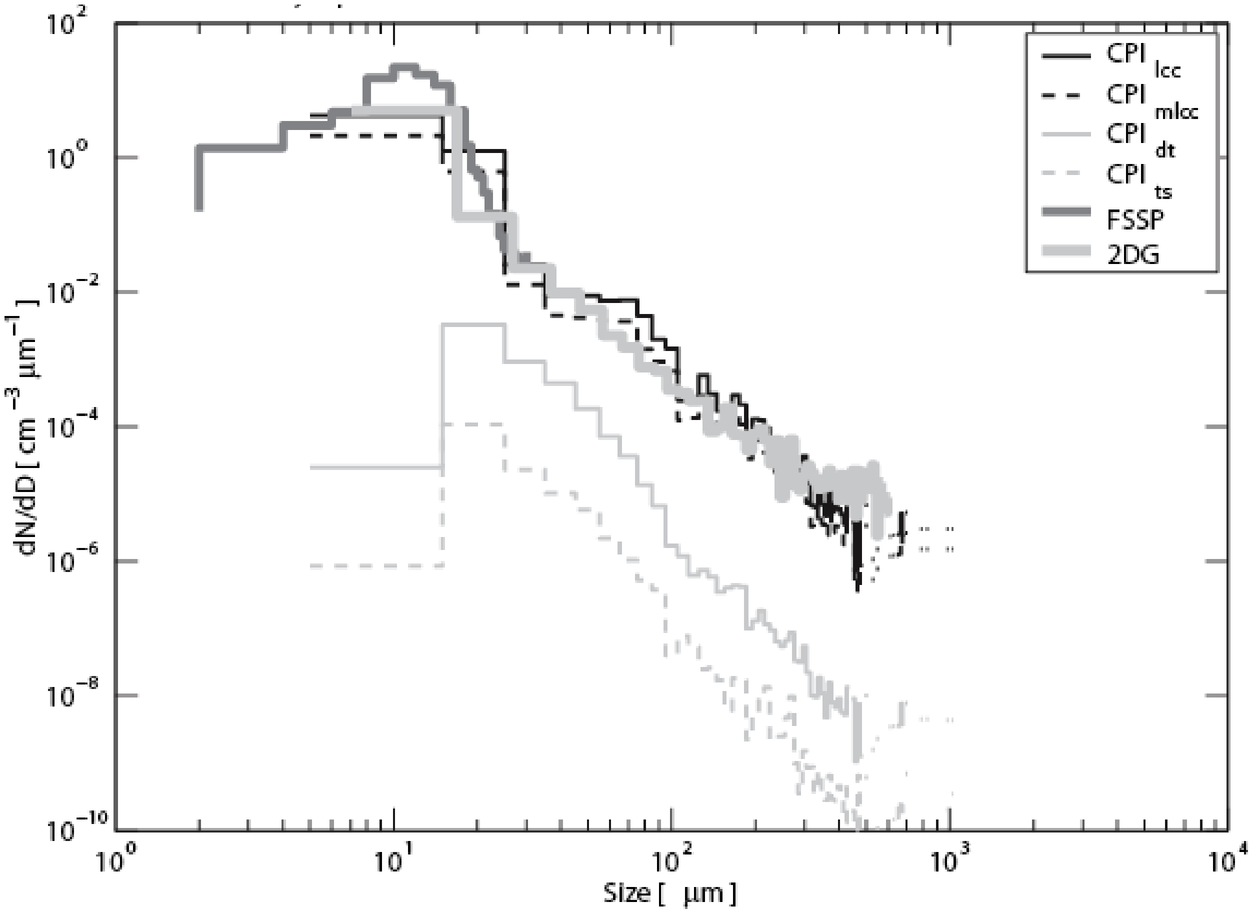


liquid water content (spherical particles only)



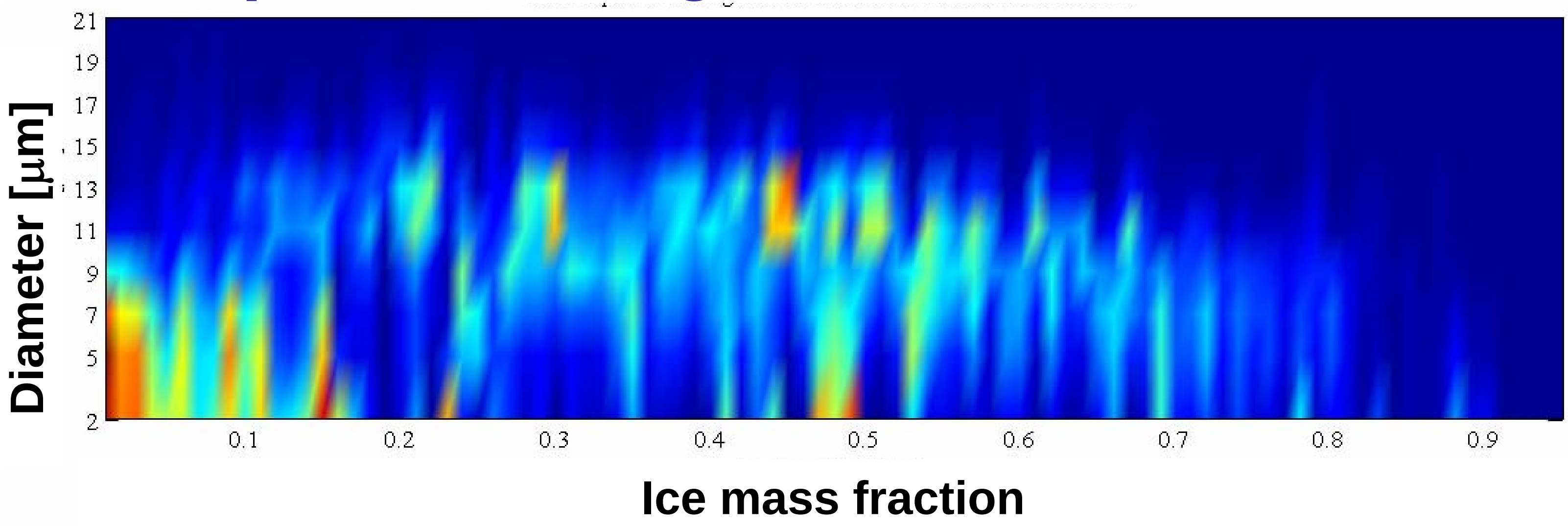
From Connolly *et al*. (2006)

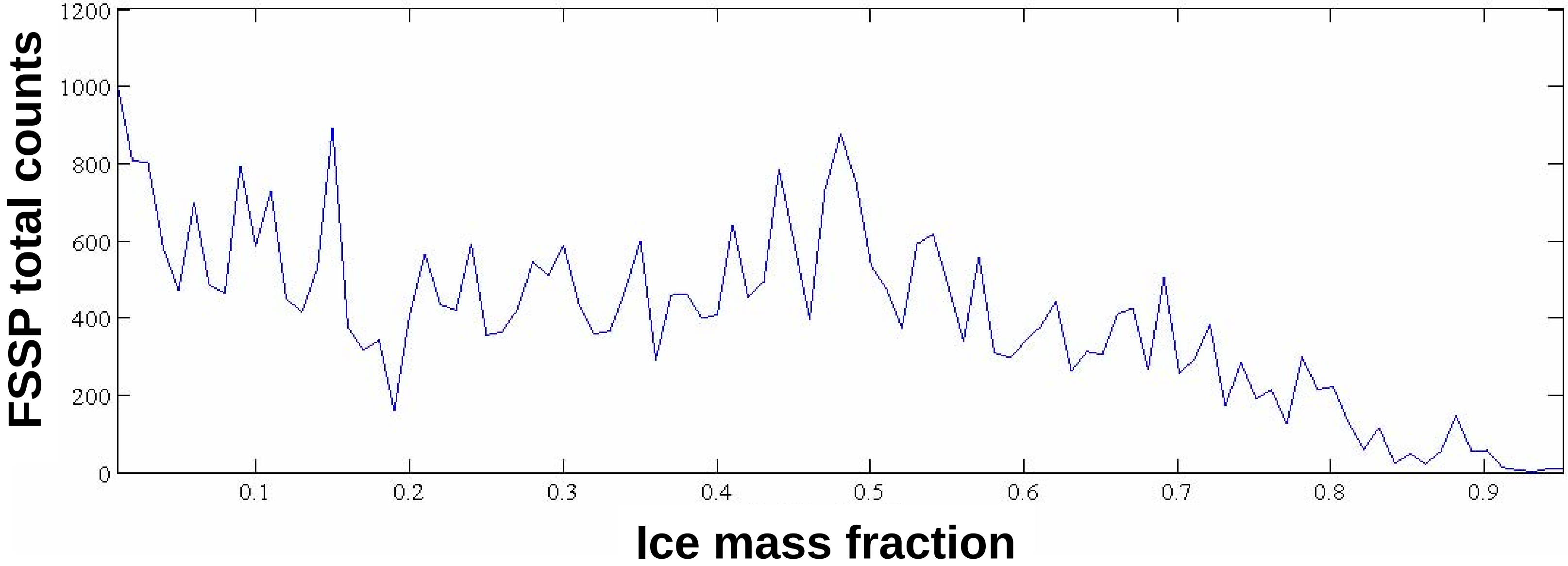
Comparison of hydrometeor size distributions from CPI, FSSP and the 2D probe (2DG) in mixed phase clouds



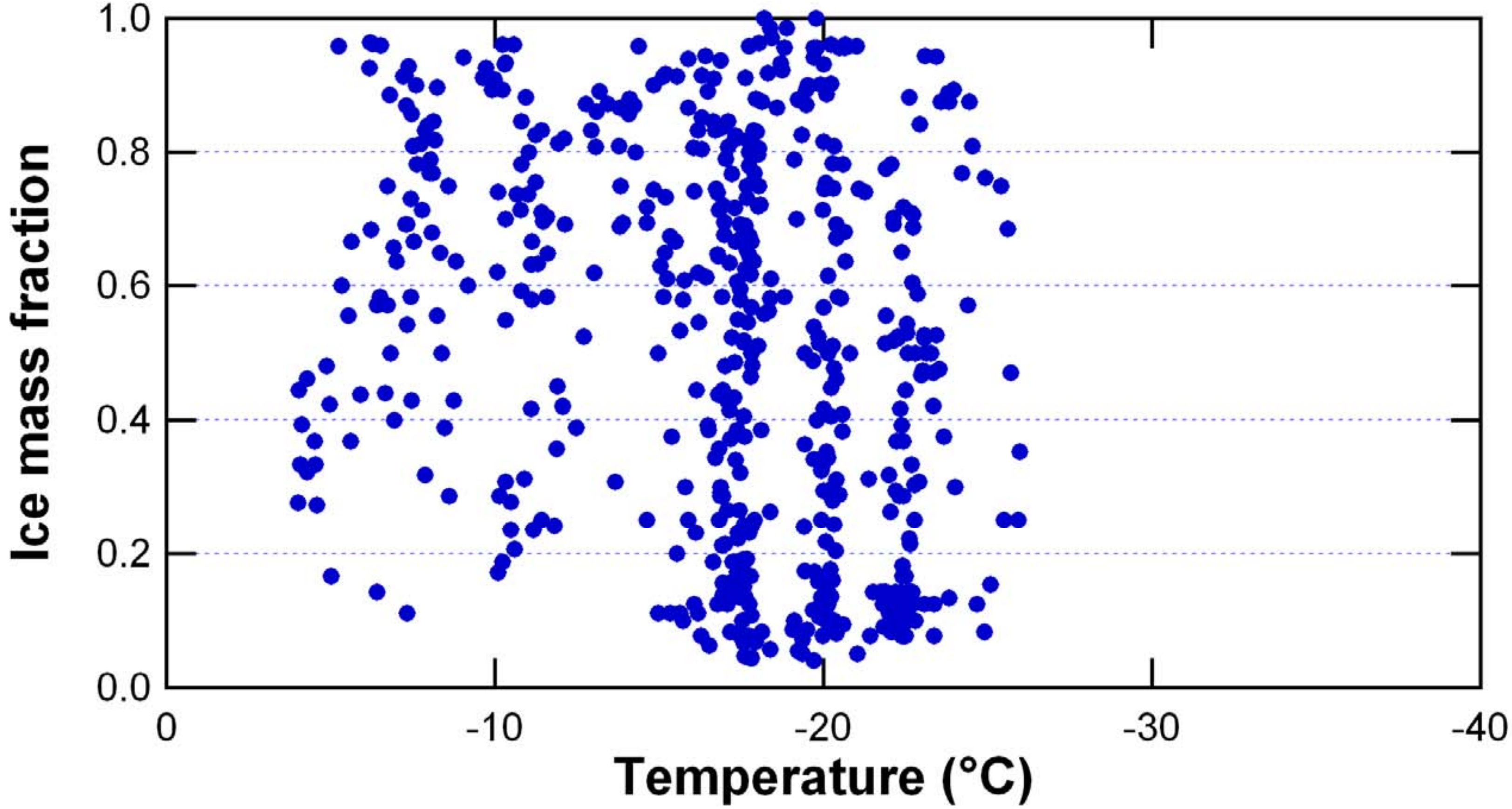
CPIlcc and CPImlcc are the CPI spectra corrected with the procedure described above. In the mlcc calculation one assumes that images are taken at the maximum possible frame rate. The other (lcc) uses the actual frame rate, which is the correct thing to do. **The lower two curves (CPIdt and CPIts) were obtained using the manufacturers basic software which do not account for sample volume and depth of field corrections.** dt means the concentration scaled by deadtime (no other corrections have been applied). ts means the size distribution that is measured by the CPI is scaled so the total concentration is equal to the total strobes.

FSSP Spectra averaged over ice mass fraction



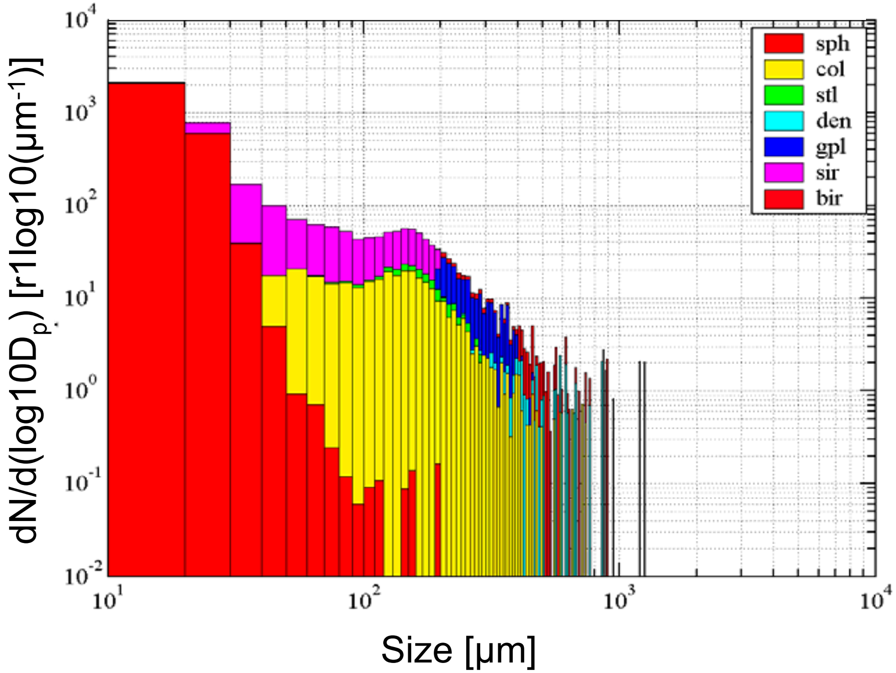


Ice mass fraction vs. temperature

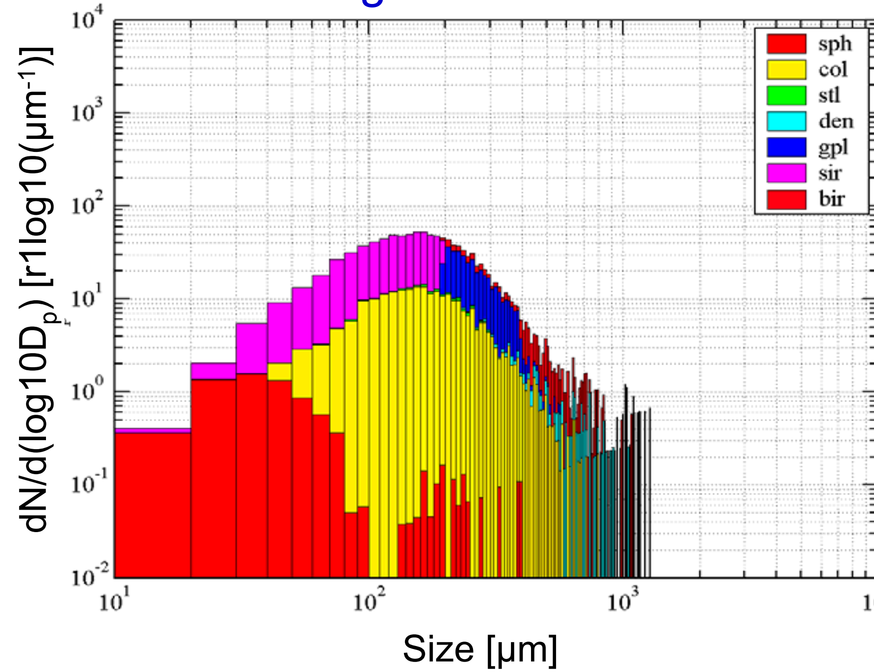


Cloud Particle Imager (CPI) Data

March11, 14:00 – 16:00,
Low ice fraction



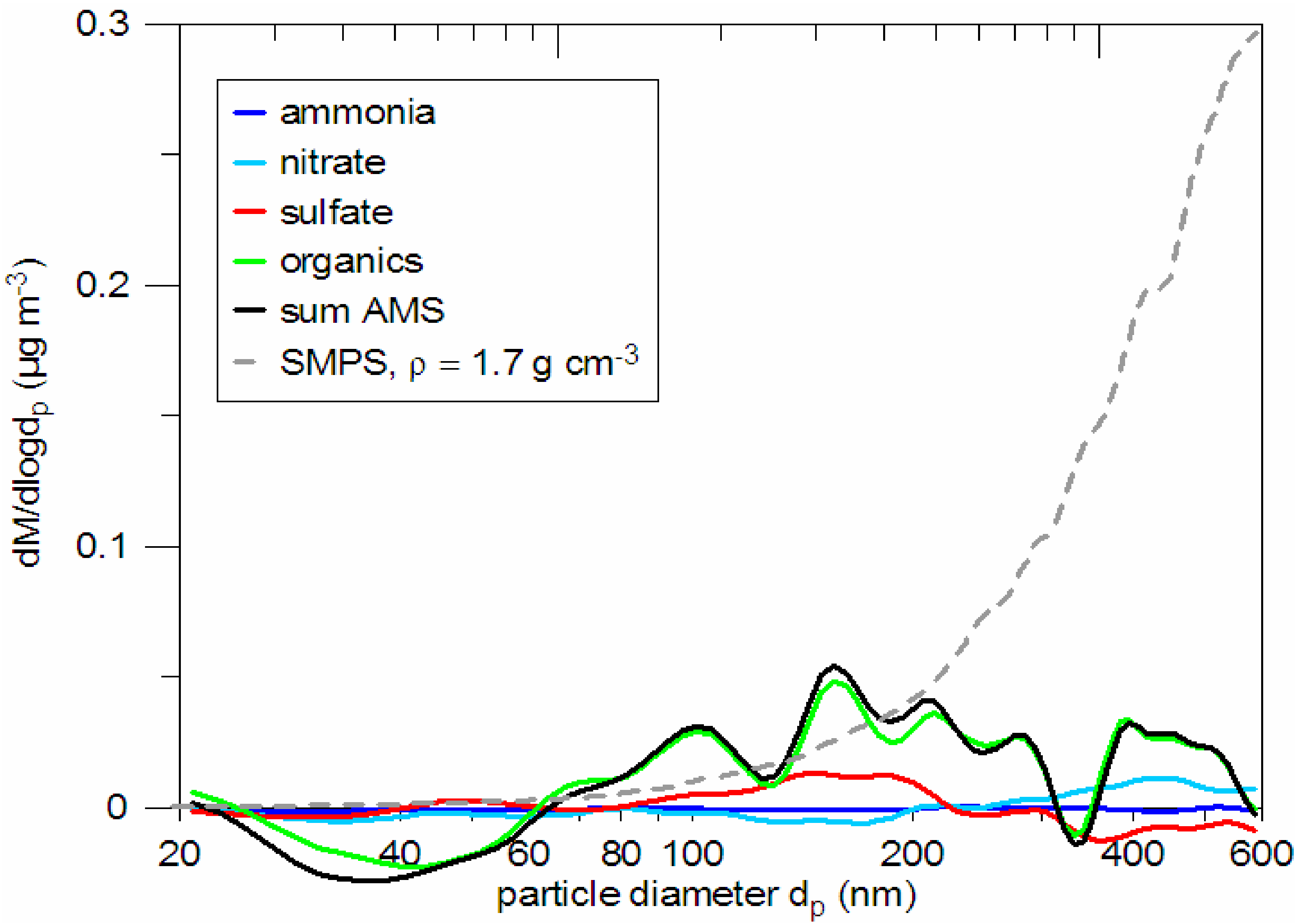
March11, 8:00 – 10:00
High ice fraction



The hydrometeors are classified into the following habits:

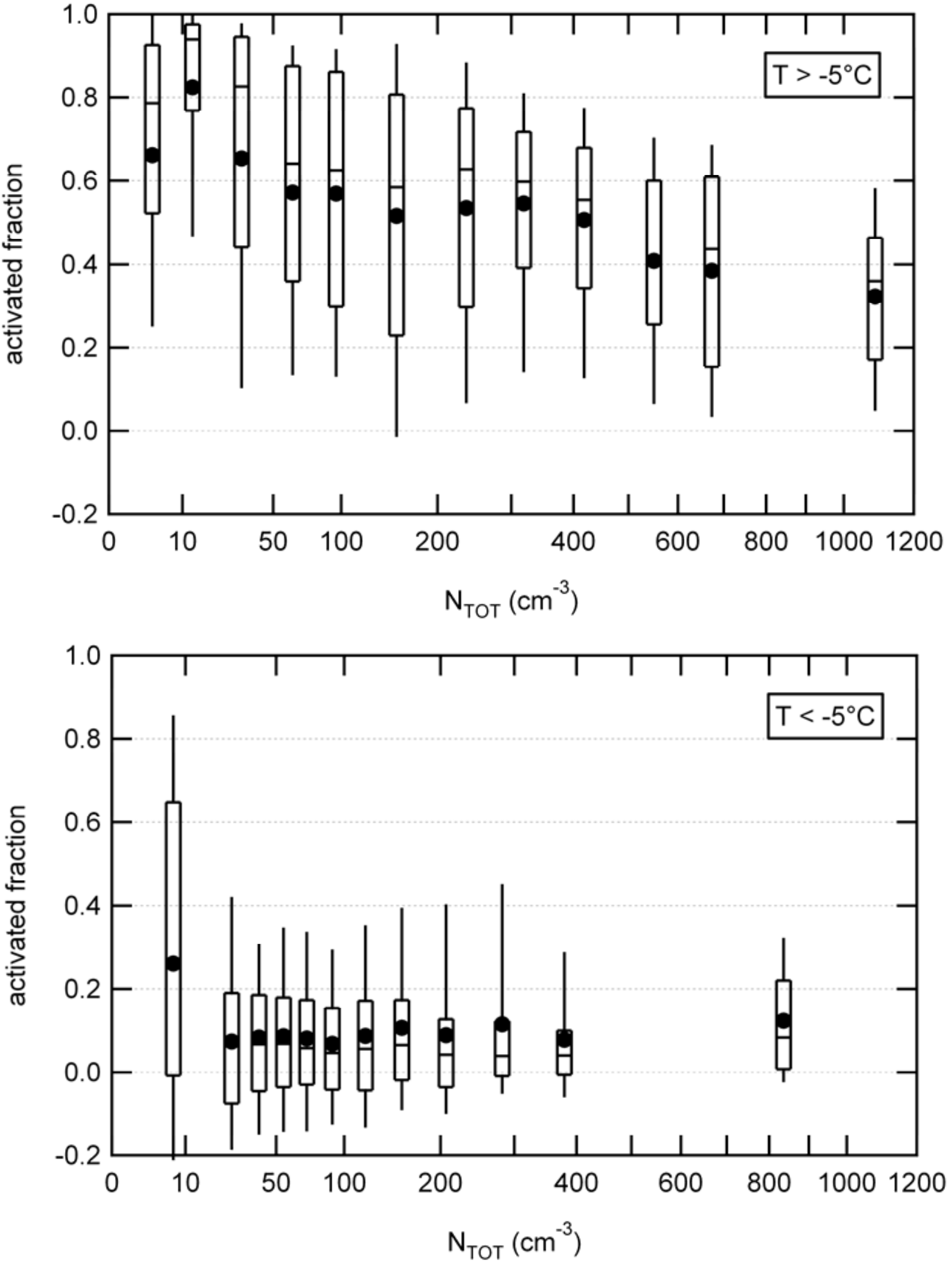
Droplets	Ice crystals	
Spheroid (sph)	**Columns (col)** **Big Irregular (bir)** **Stellar (stl)**	**Dendrite (den)** **Graupel (gpl)** **Small Irregular (sir)**

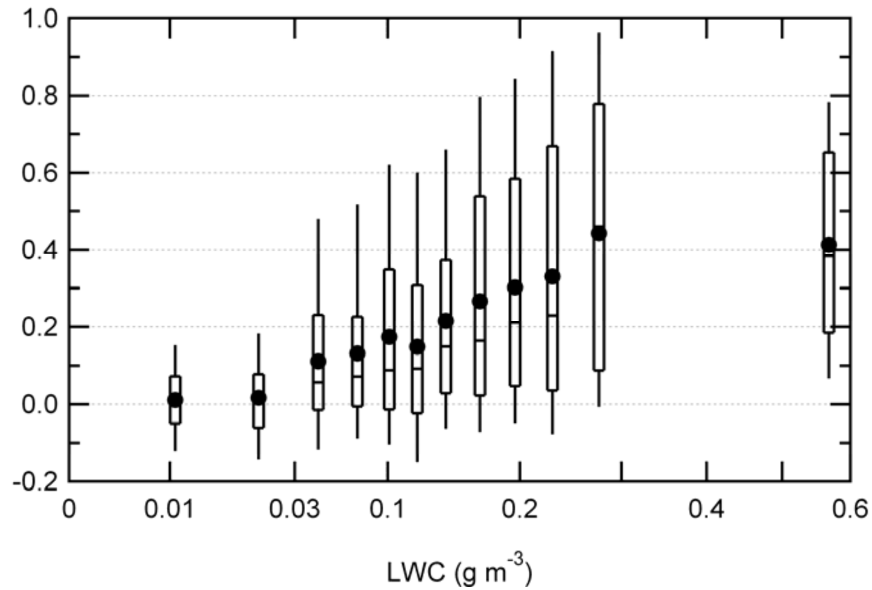
Comparison mass size distribution SMPS and AMS:



- IN consist mainly of refractory particles

Activated fraction vs. temperature and LWC

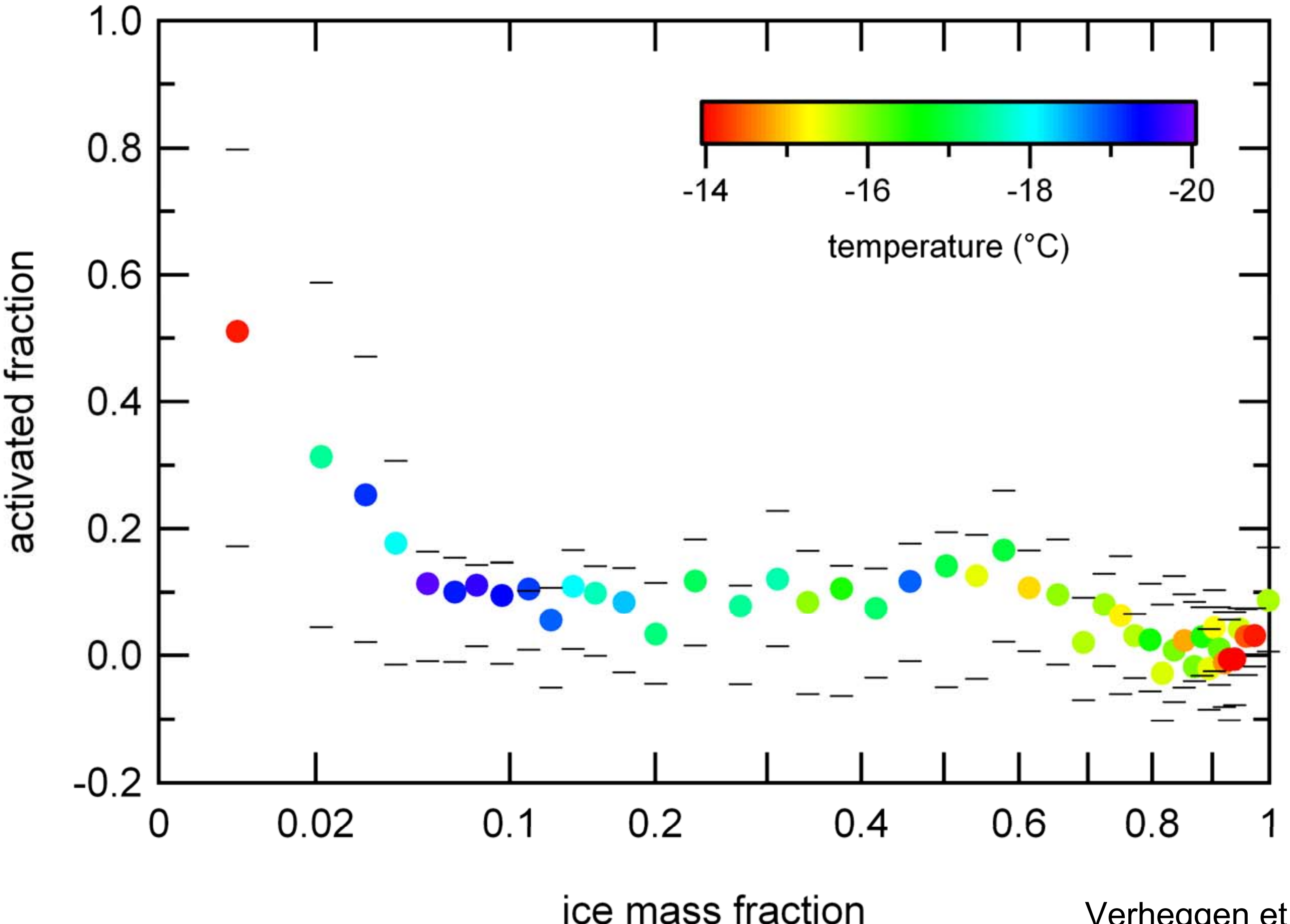




Verheggen et al., submitted to JGR

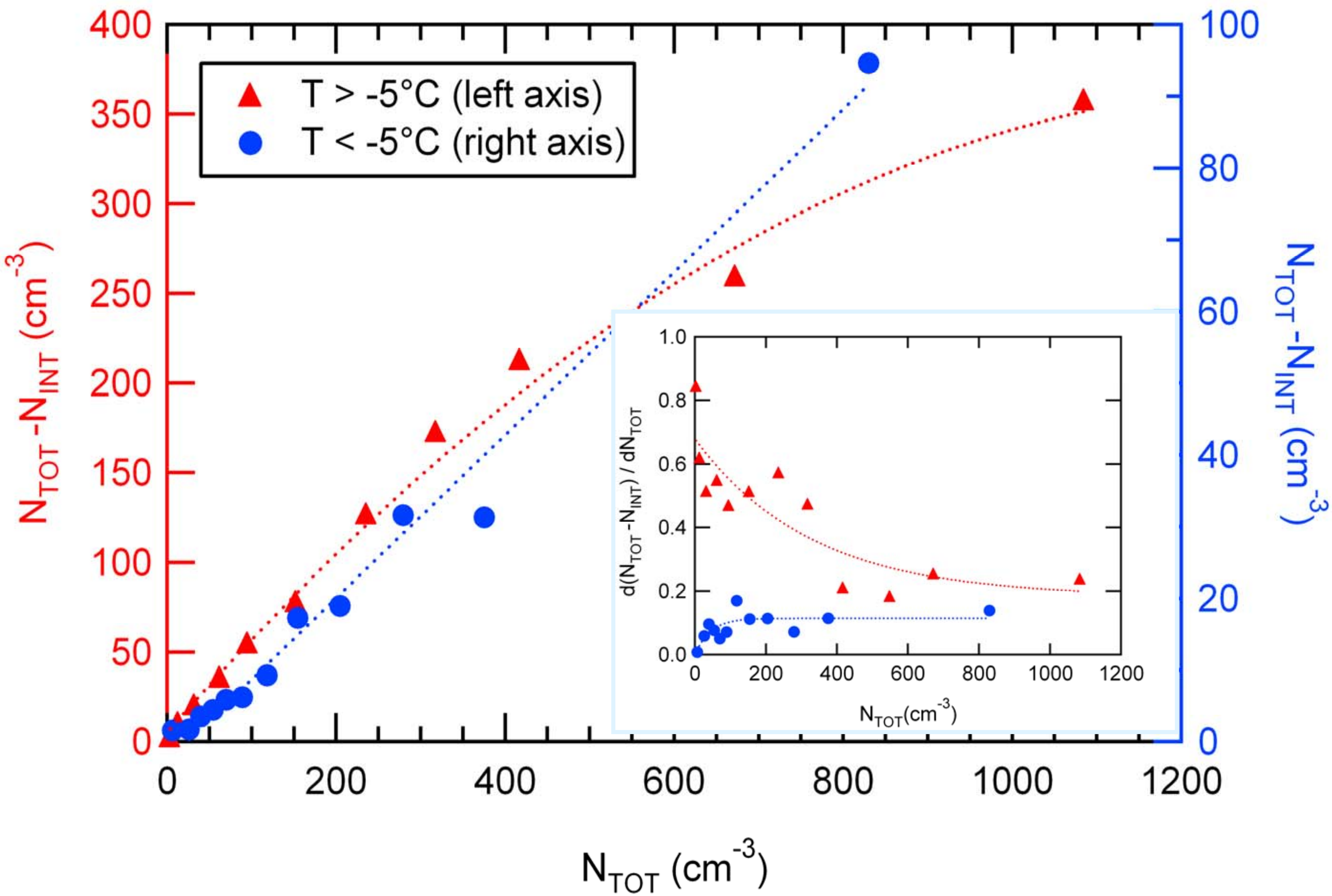
Activated fraction vs. ice mass fraction

(440 hours of in-cloud measurements)

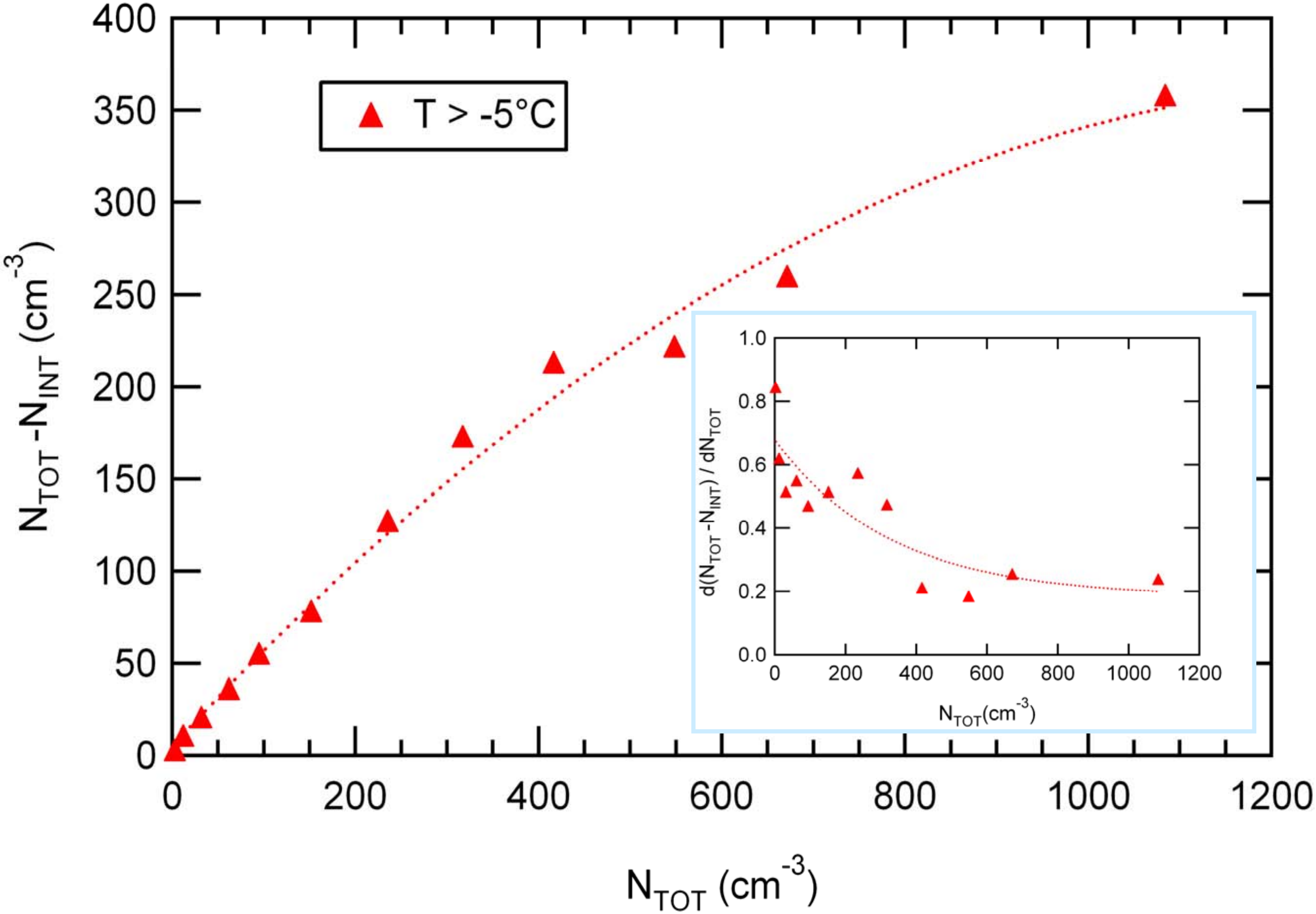


Verheggen et al. (2007)

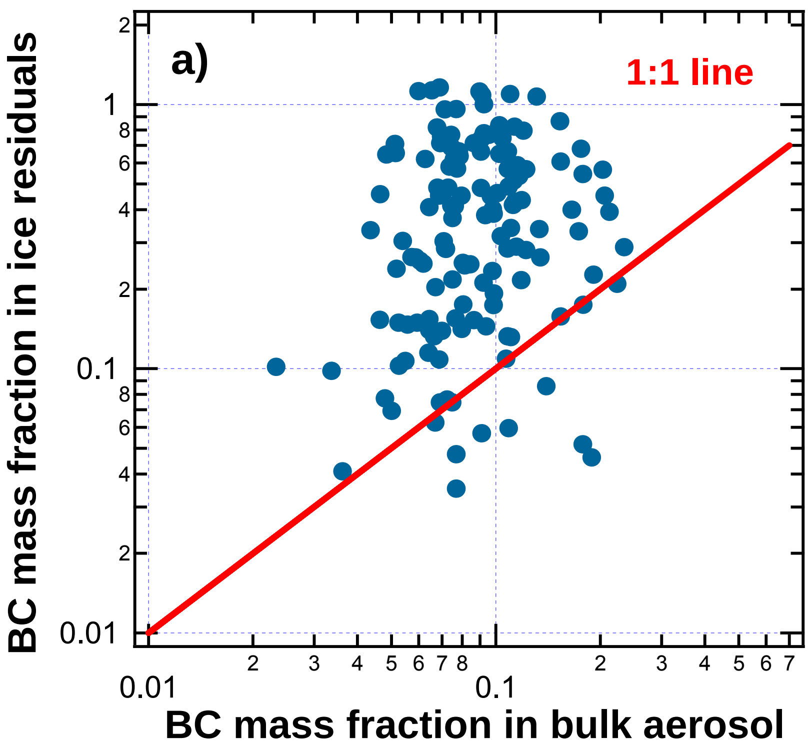
Activated vs. total particle number (d_p>100 nm)

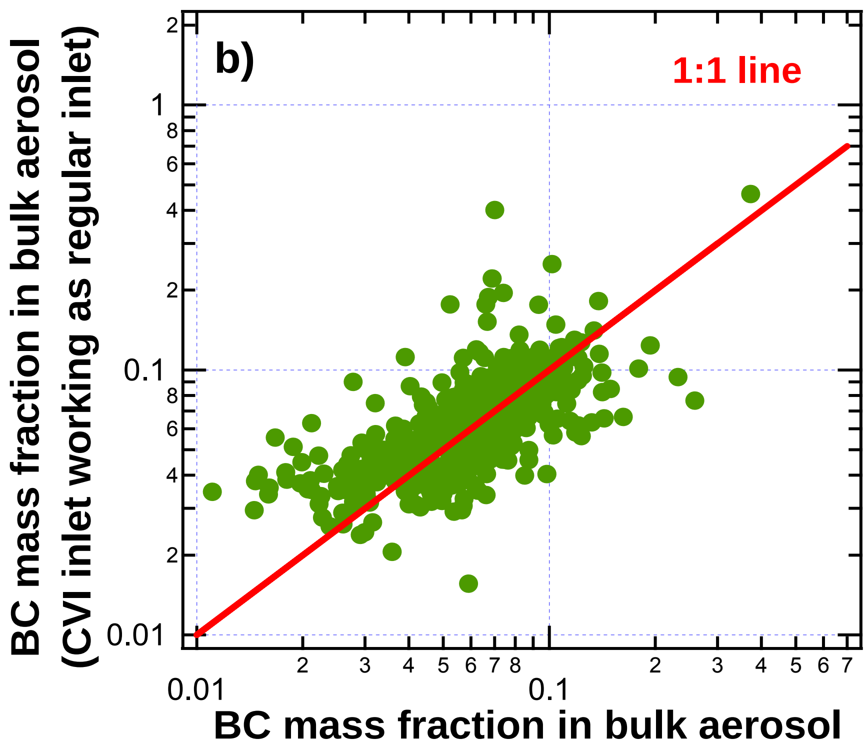


Activated vs. total particle number (d_p>100 nm)



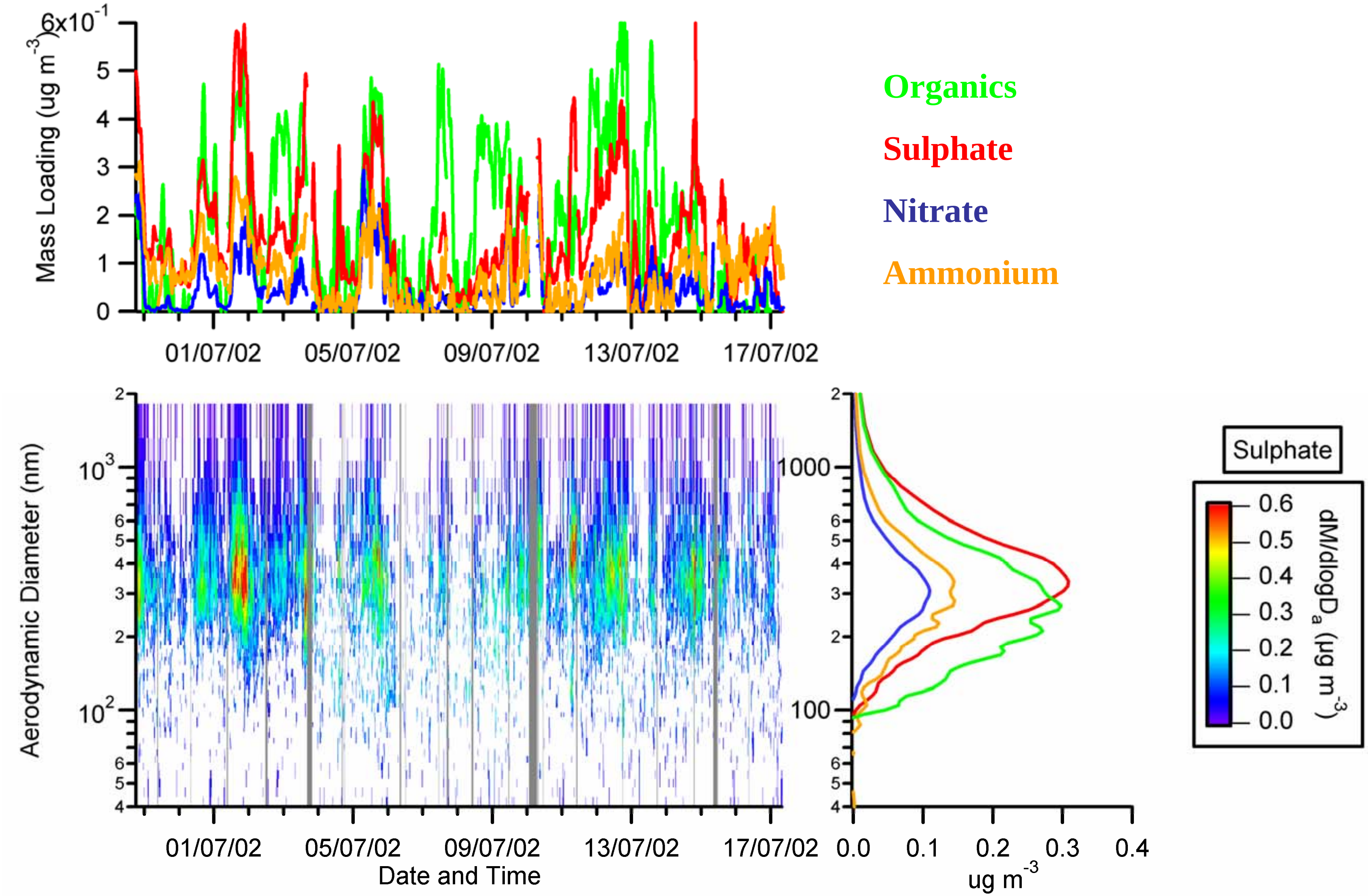
Comparison of the BC mass fraction in the ice residual phase with the mass fraction in the bulk aerosol phase in cloud (23 hours) (a), out of cloud (CVI operating as a regular inlet without counterflow) (62 hours) (b)



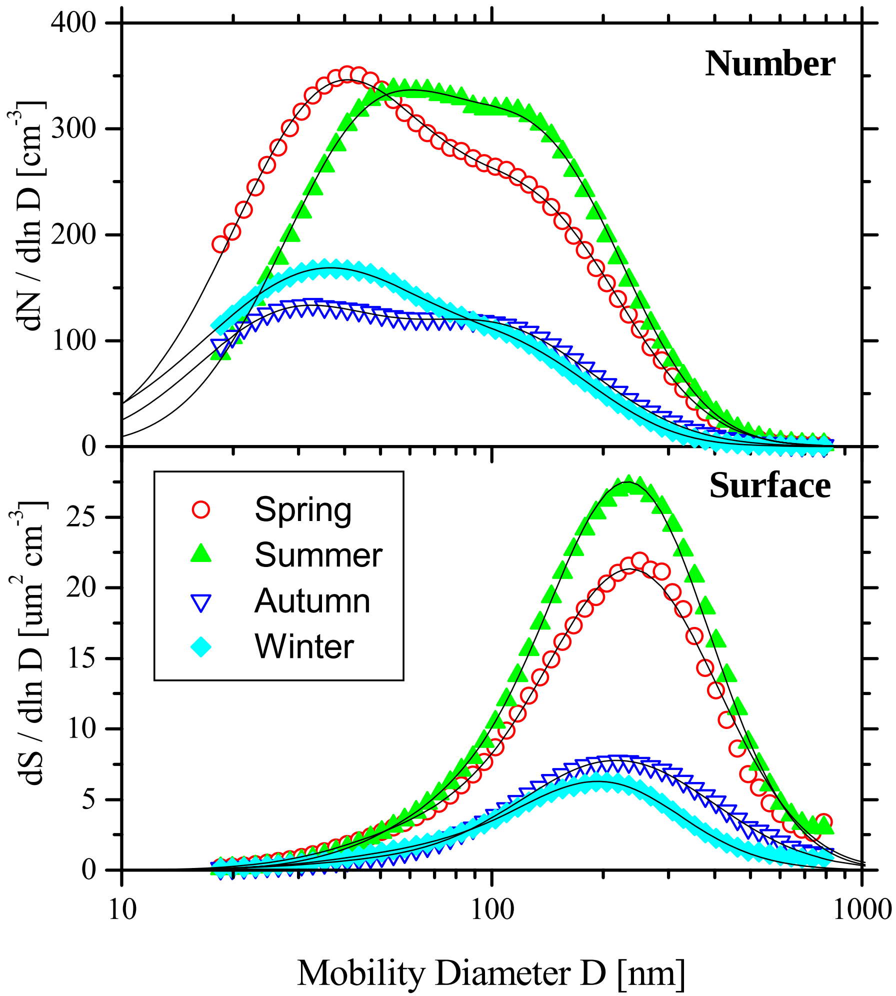


On-line chemistry data with an Aerodyne Aerosol Mass Spectrometer

Hugh Coe, UMIST

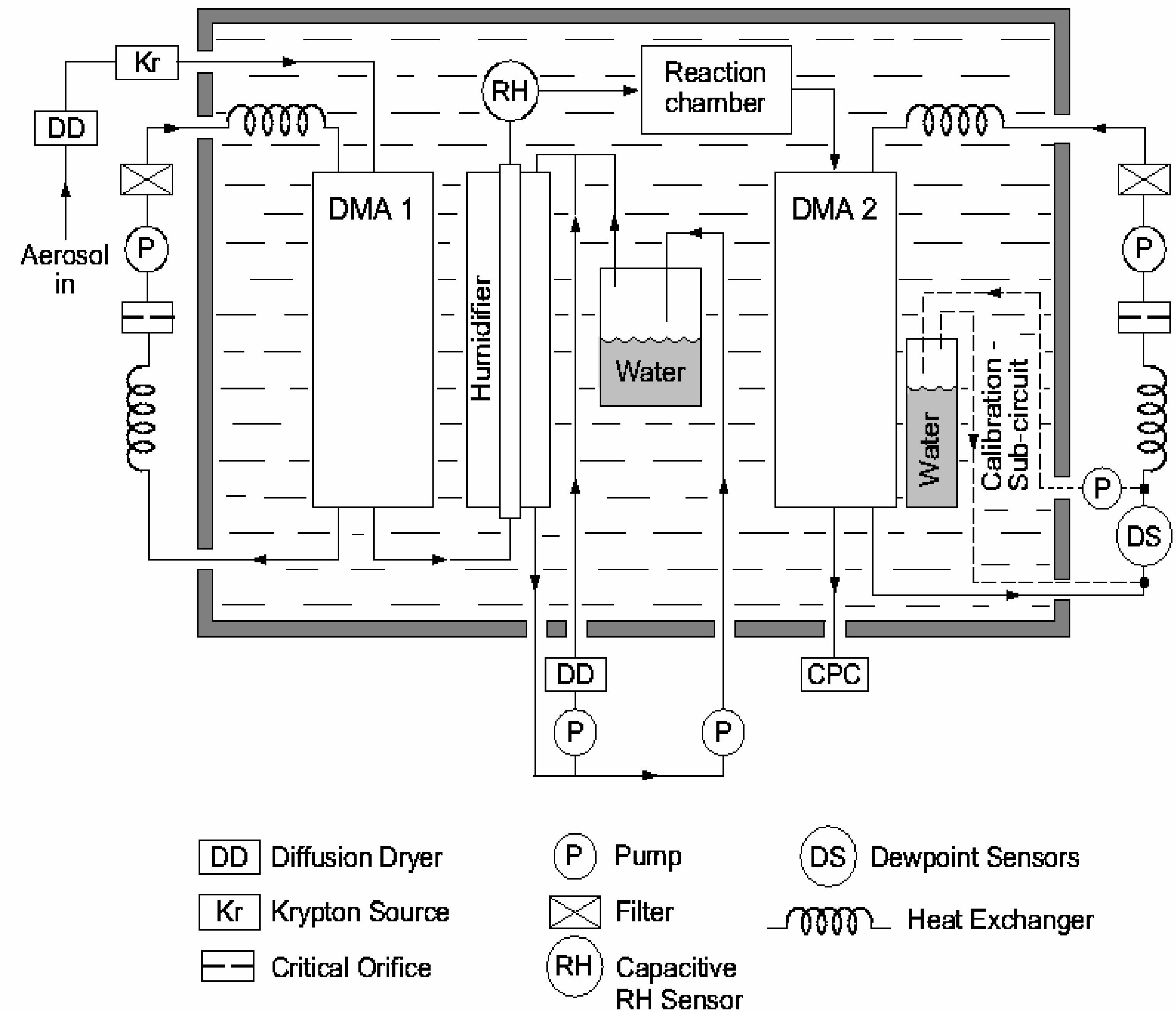


Number and surface area size distributions of the Jungfraujoch aerosol; seasonal averages

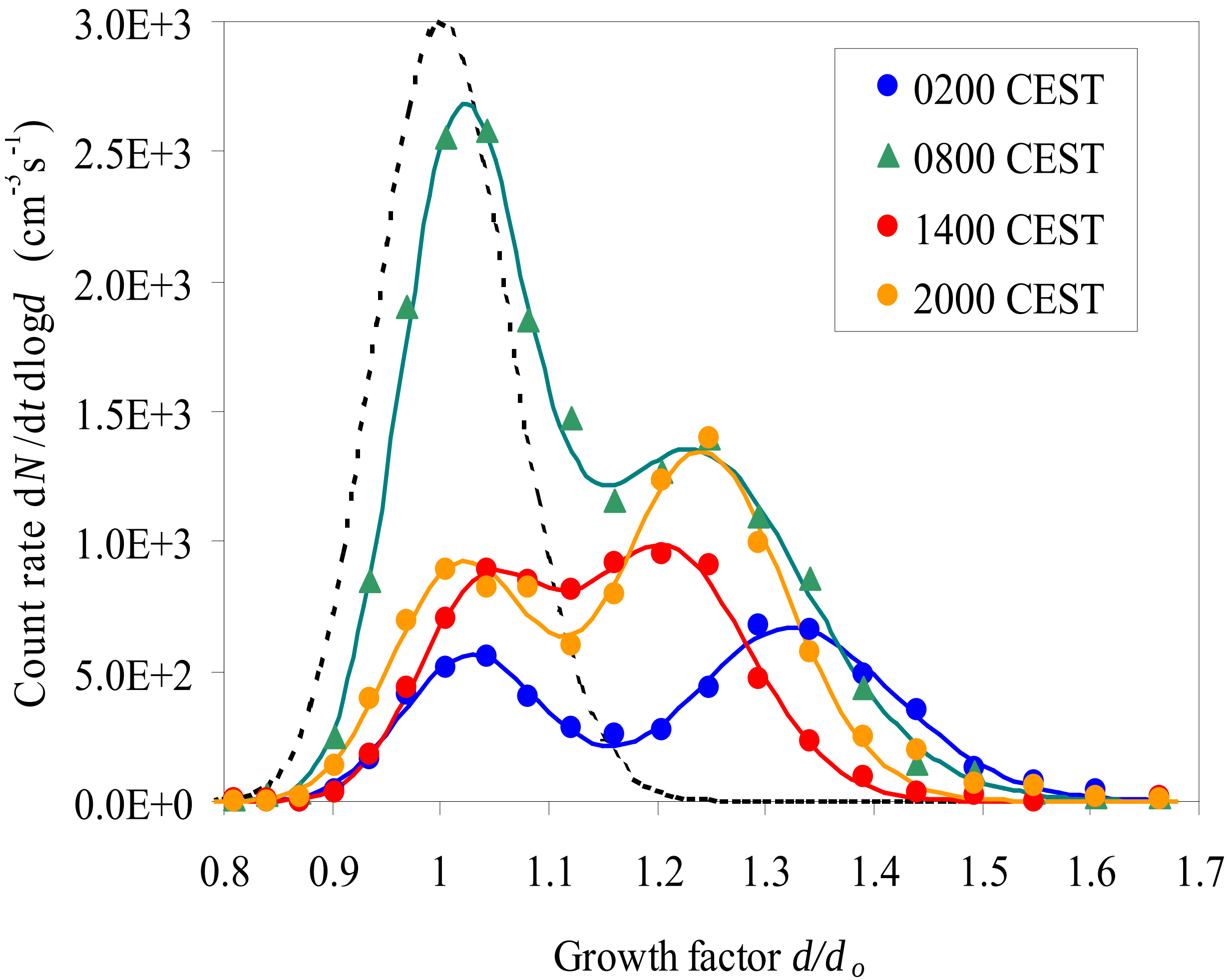


Weingartner et al., JGR (1999)

Experimental Setup of the HTDMA

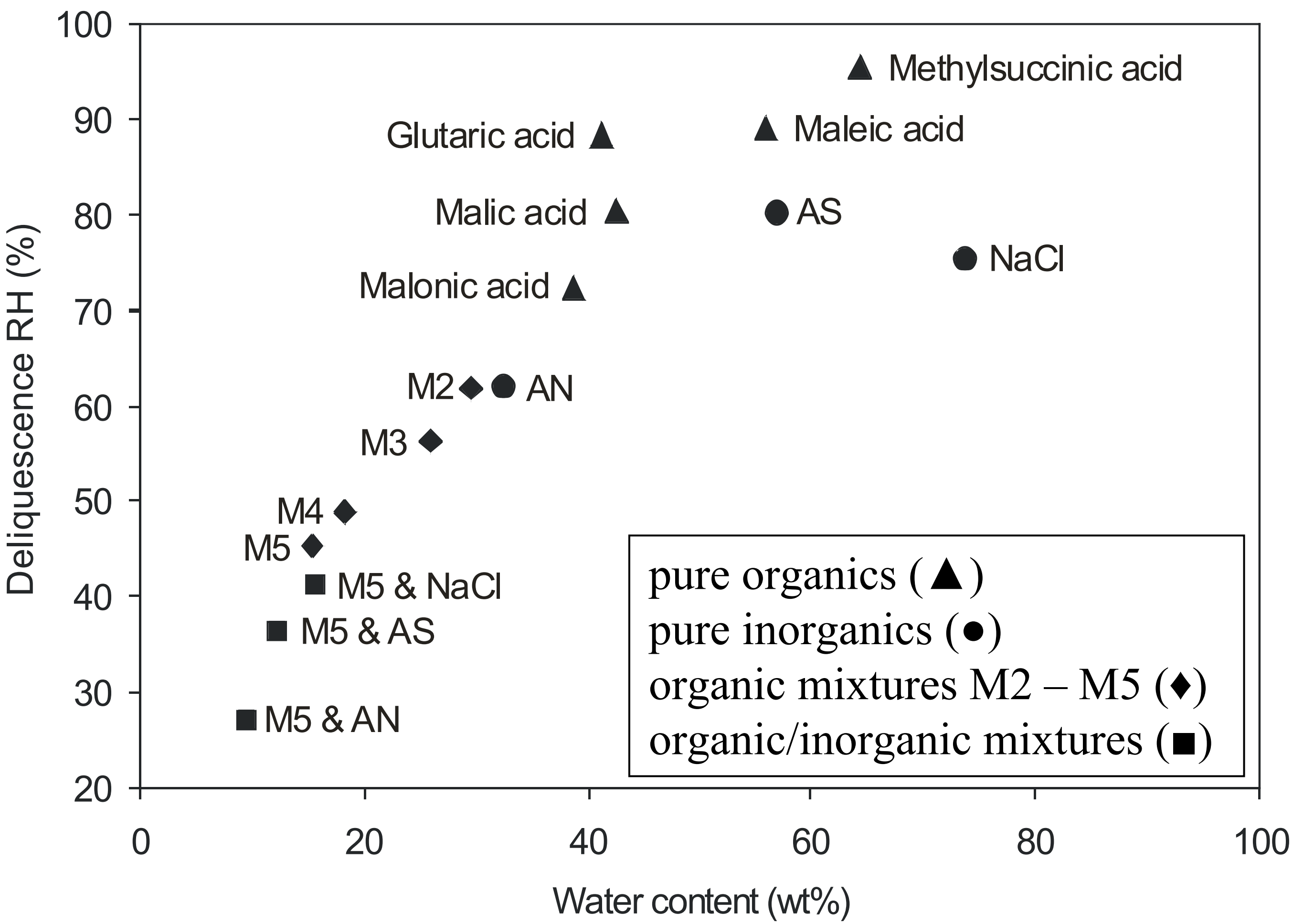


Growth factor (d/d_o) distributions at an urban site (Milano).



Dry monodisperse particles (d_o = 100 nm, dashed line) were exposed to RH = 90% (T=20°C).

Deliquescence relative humidity (DRH) and water content at DRH for various substances



Marcolli et al. (2004), J. Phys. Chem

Zdanovskii-Stokes-Robinson relation (ZSR)

Chen et al., 1973

Growth factors @ RH=85%: 1.52 ($(NH_4)_2SO_4$), 1.1, 1.0

$$\left(\frac{d}{d_o}\right)_{calculated} = \left(\varepsilon_{Inorg}\left(\frac{d}{d_o}\right)^3_{Inorg} + \varepsilon_{organics}\left(\frac{d}{d_o}\right)^3_{organics} + \varepsilon_{EC}\left(\frac{d}{d_o}\right)^3_{EC}\right)^{1/3}$$

volume fraction ε

density ρ: $1.7\ g/cm^{-3}$, $1.5\ g/cm^{-3}$, $2\ g/cm^{-3}$

inorganics mass fraction

Organic mass fraction

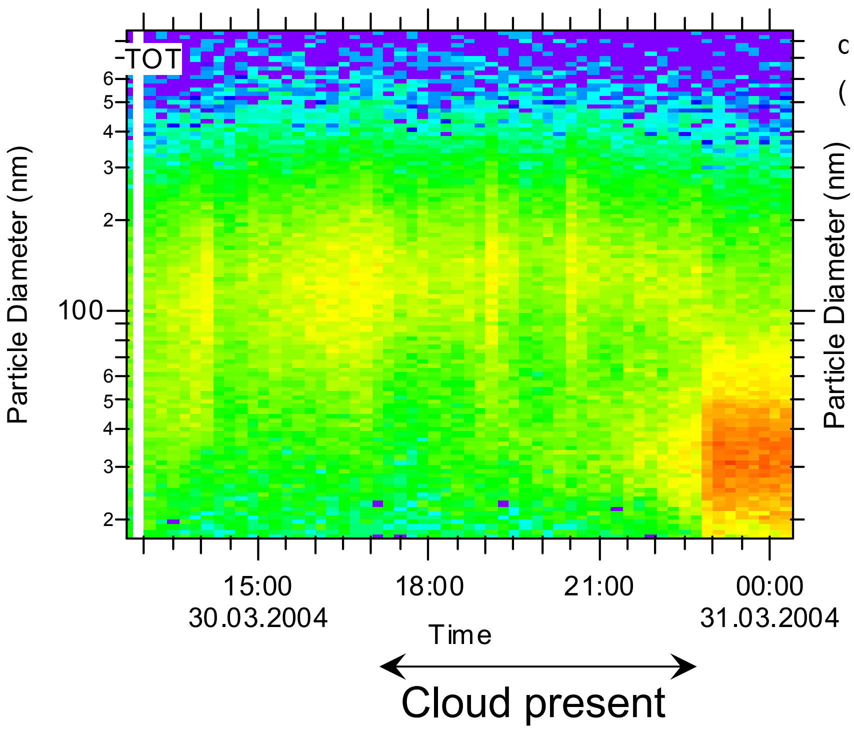
EC mass fraction (9.3 m²/g)

AMS + Aethalometer

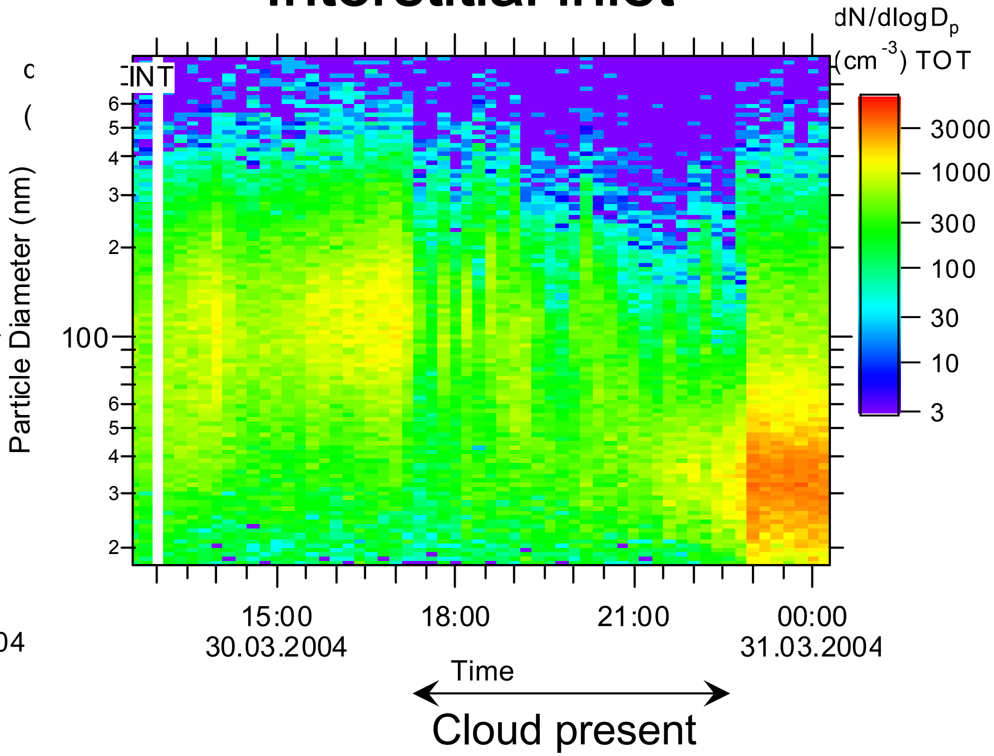
Case Study I: Liquid cloud event

March 30, 2004

Total inlet



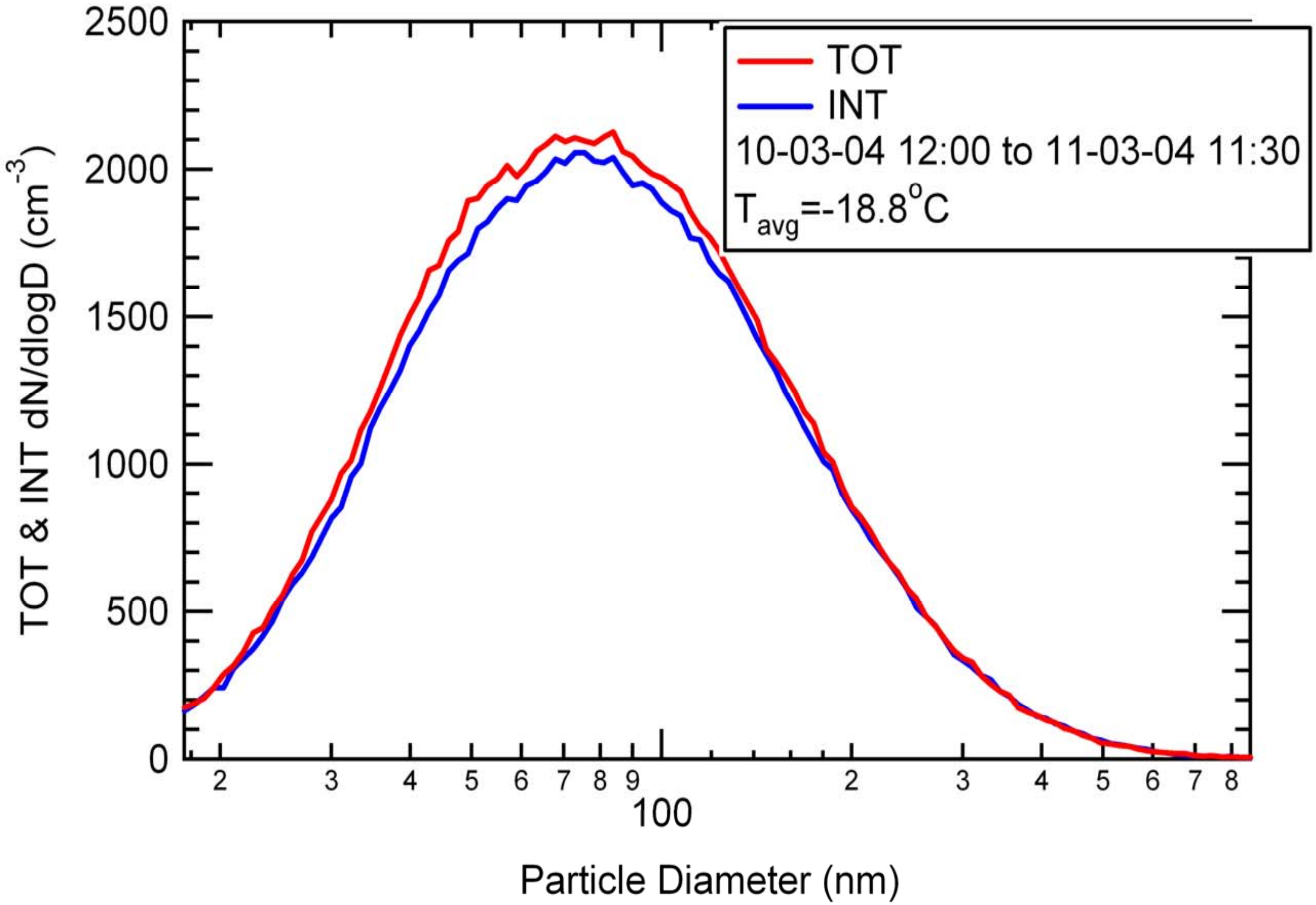
Interstitial inlet



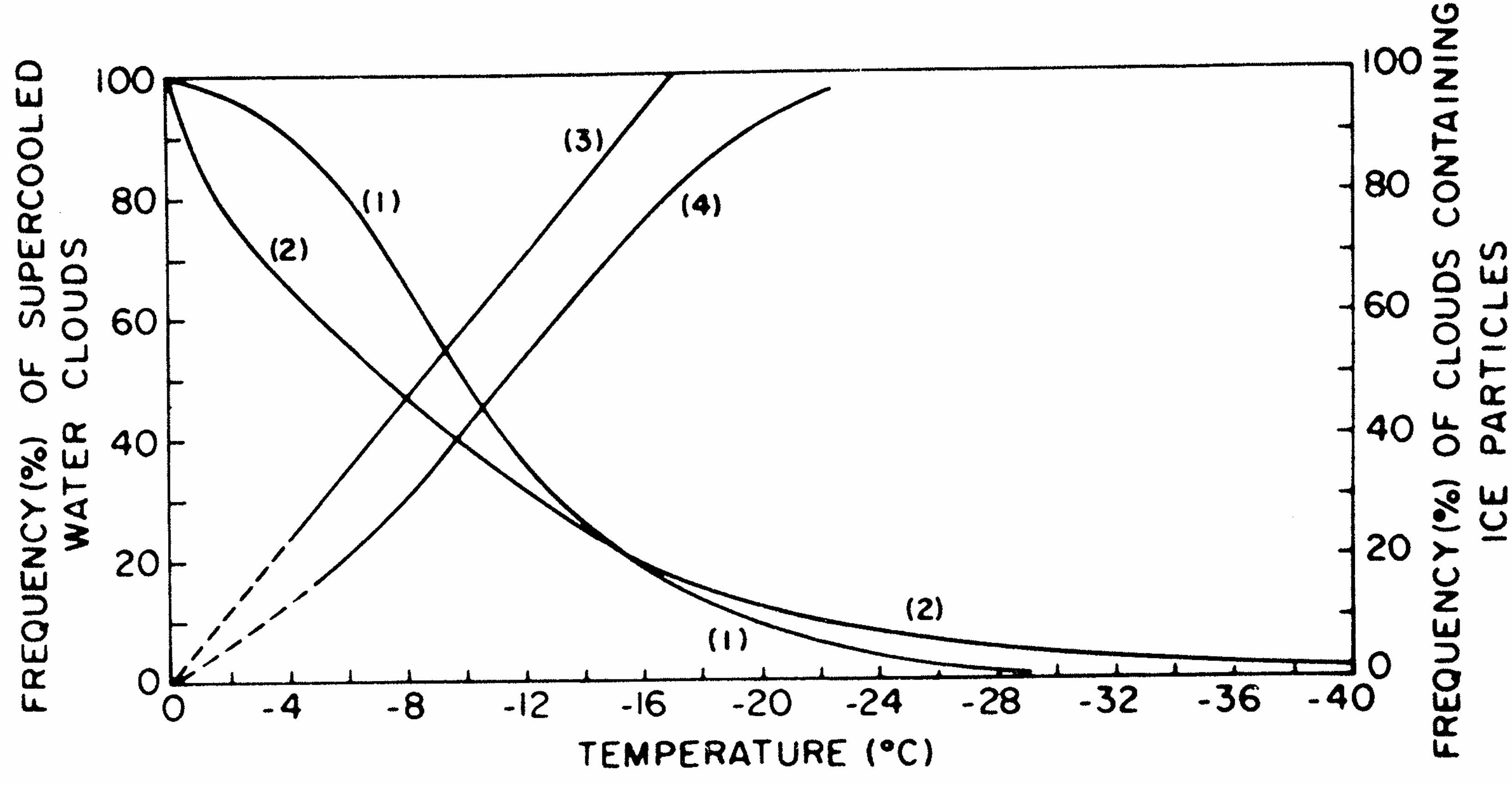
T_{avg} = -10.6 °C

Size spectra are different in a mainly glaciated cloud

Ice mass fraction > 0.5; activated fraction <0.1

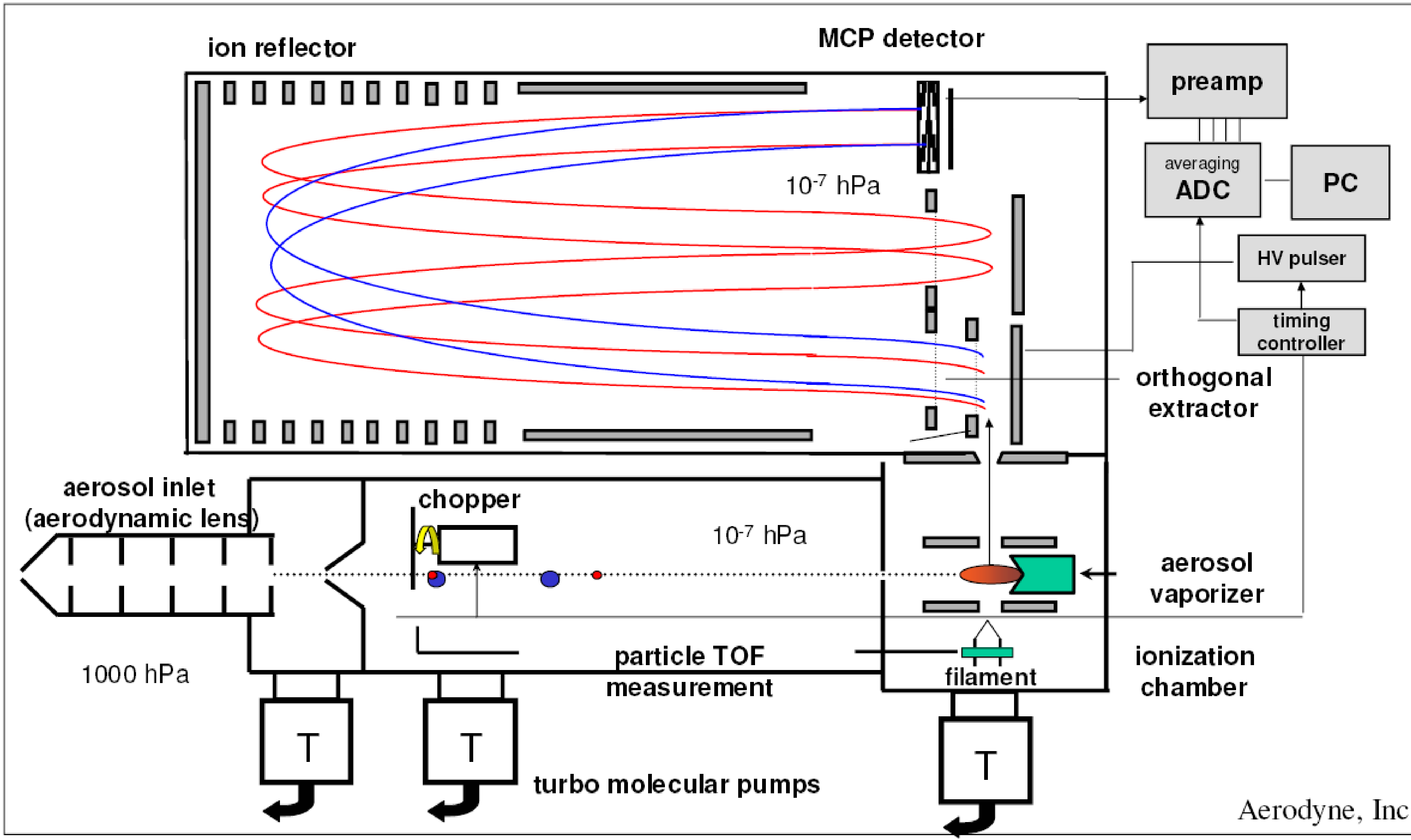


Between -5 and -20°C a significant amount of clouds contain supercooled droplets and ice crystals

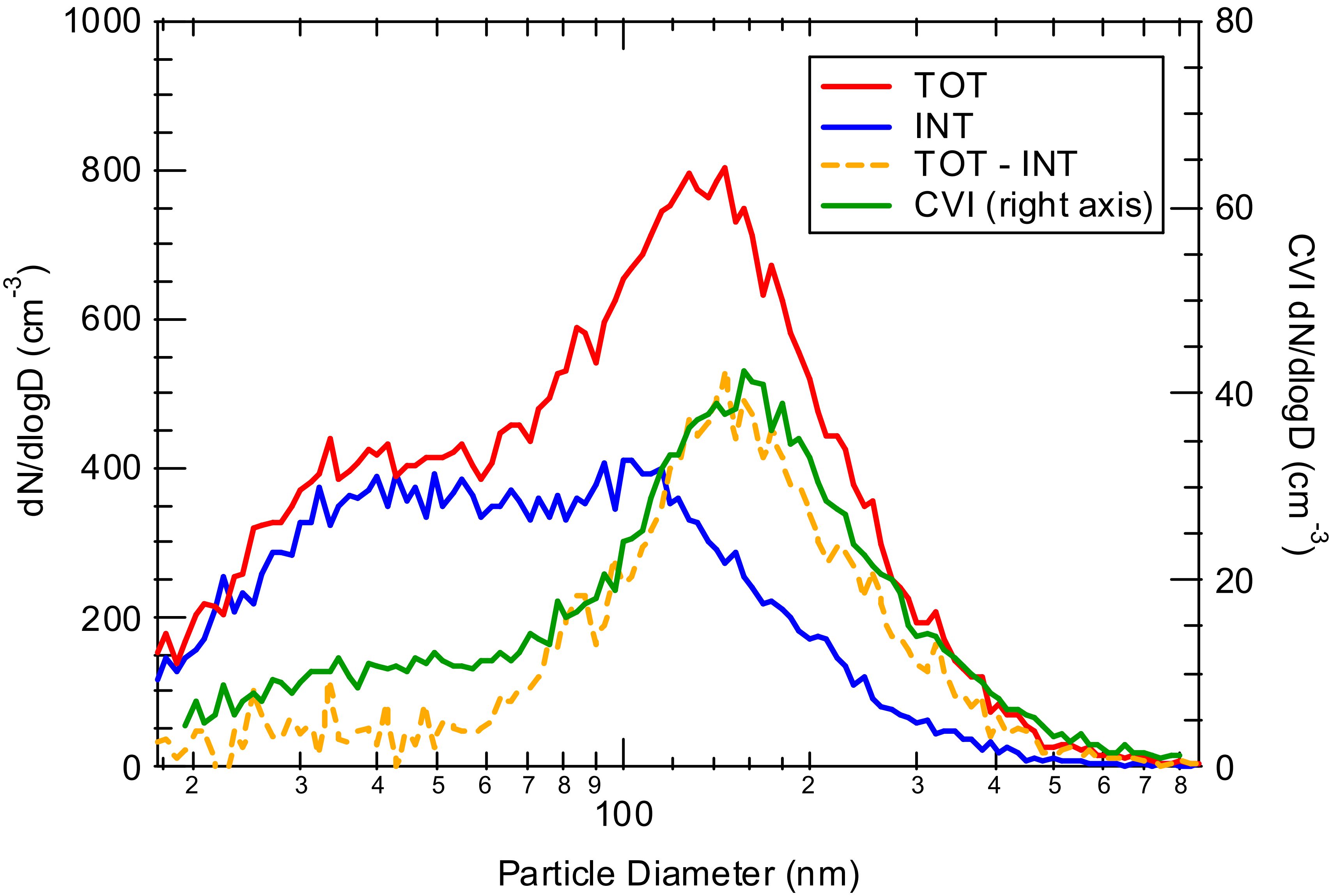


Pruppacher and Klett (1997)

High-Resolution Time-of-Flight AMS(1)

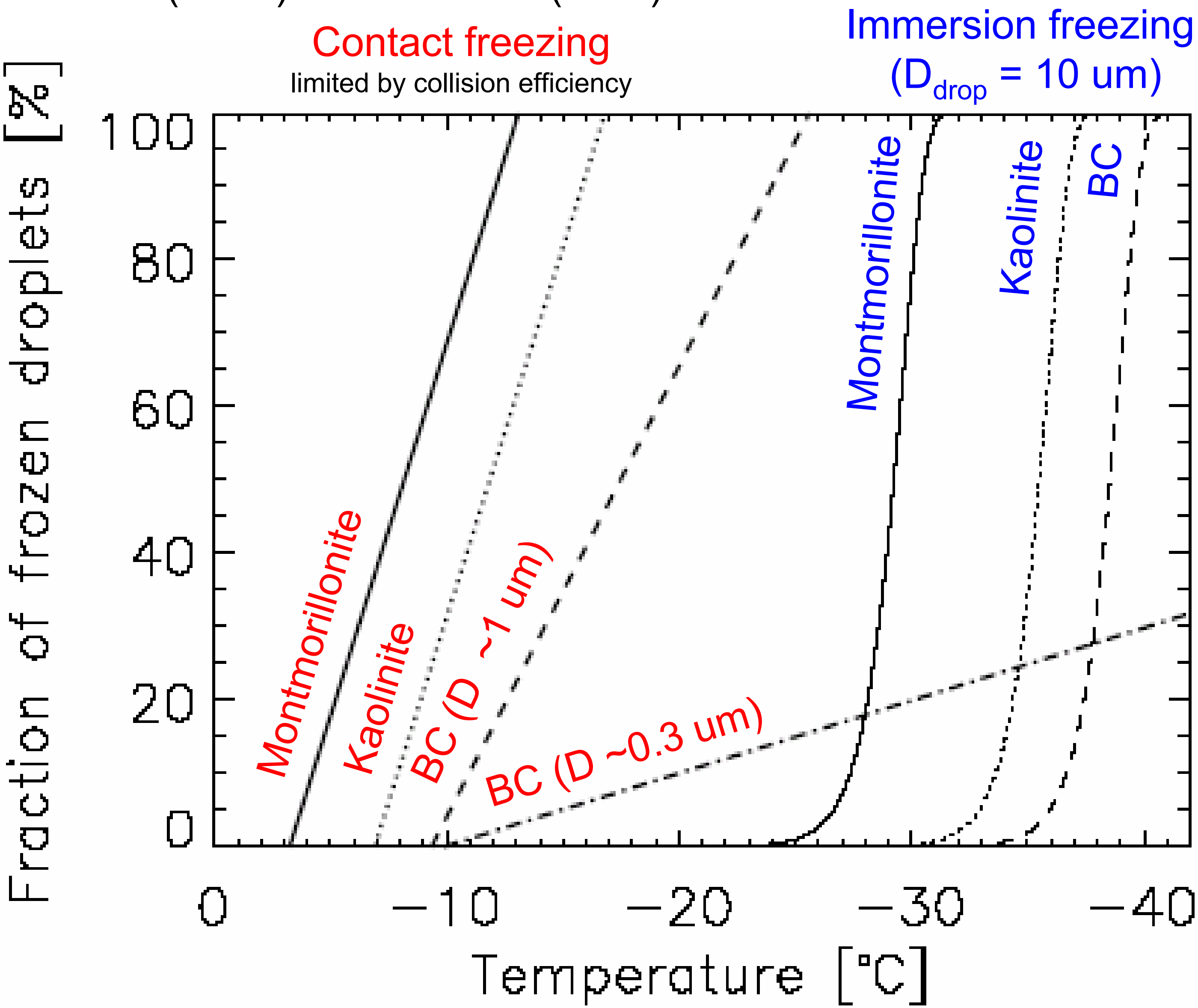


Average size distributions (4 hours in-cloud)



Model input from a compilation of lab data

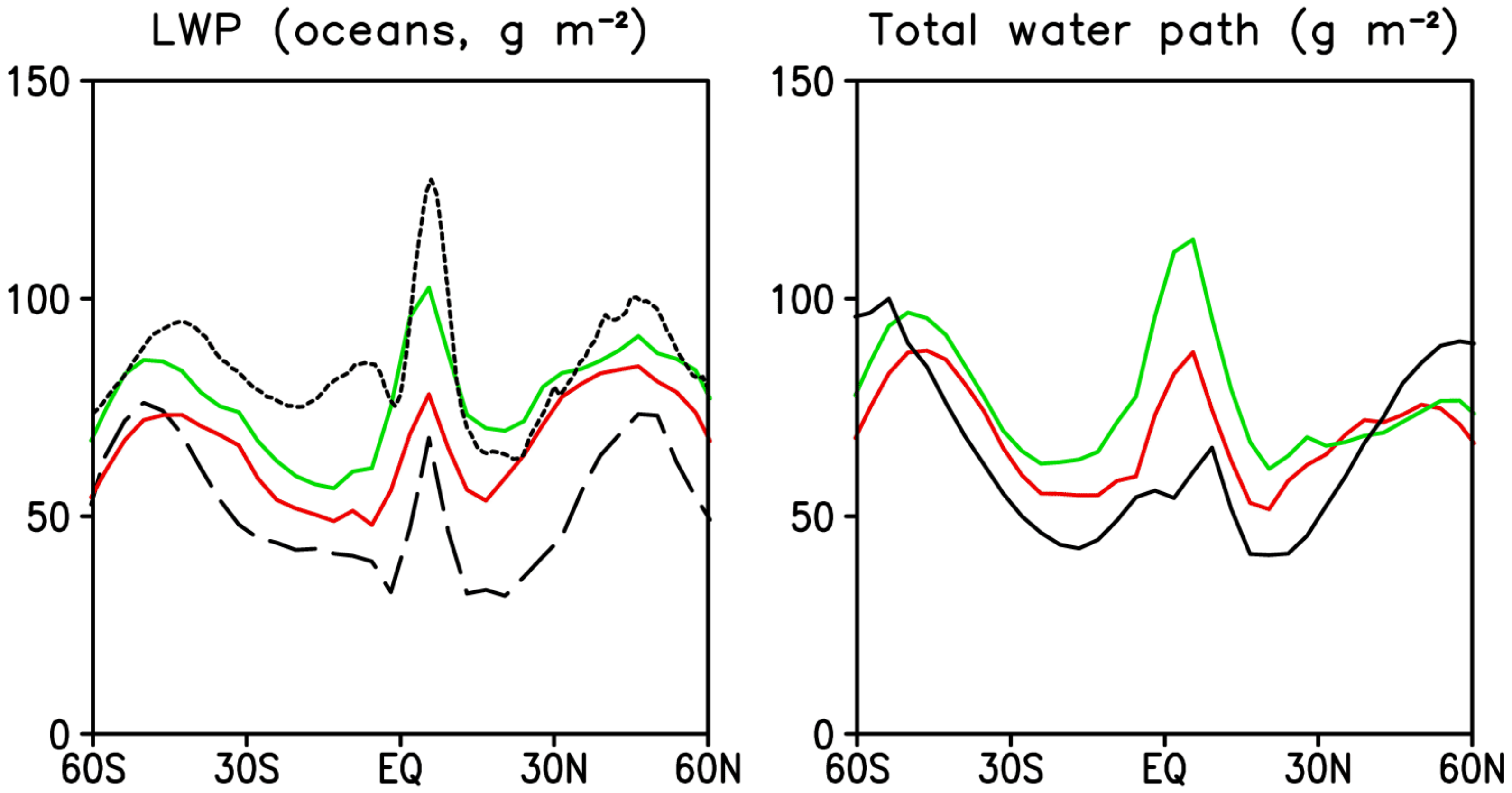
Montmorillonite (MON) and Kaolinite (KAO) as model substances for mineral dust



Adapted from Lohmann und Diehl (2006), JAS

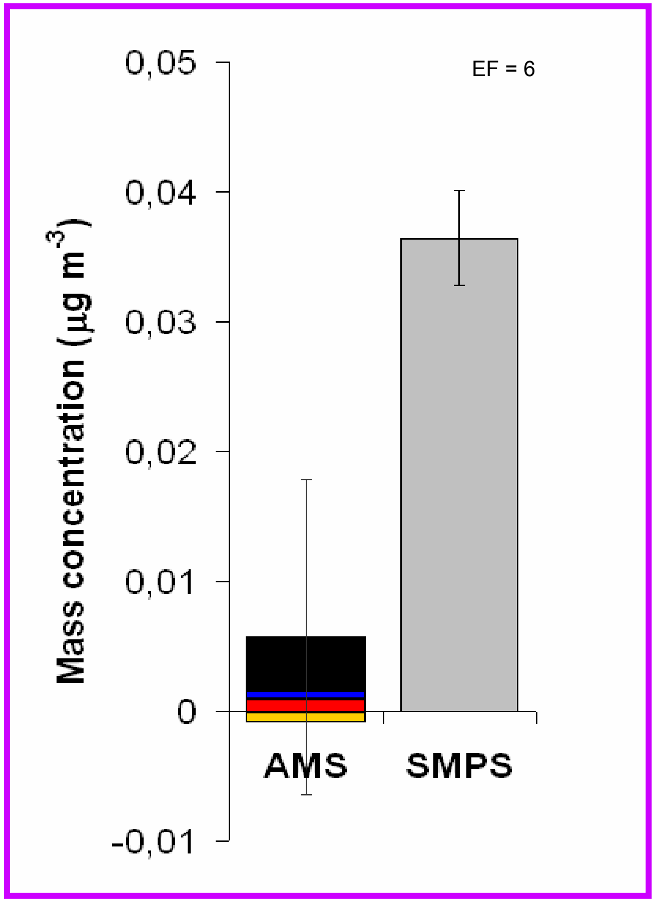
Model results: zonal annual means

Green: ECHAM4 Simulation with JFJ data
Red: Standard simulation that considered freezing of dust and black carbon
Black: “Validation” with observations deduced from satellite measurements



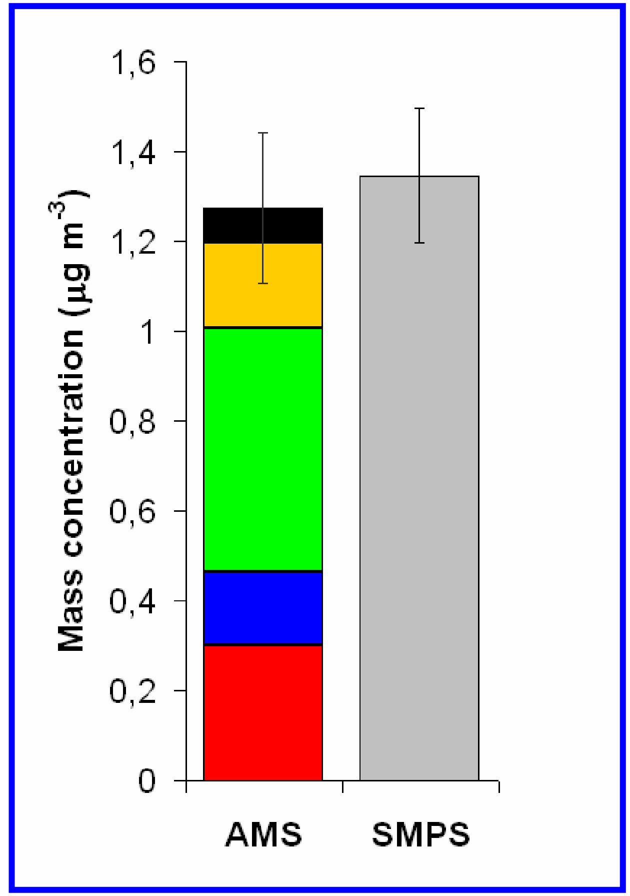
Ice nuclei chemical composition

Ice residual particles



Ammonium
Organics
Nitrate
Sulphate
BC

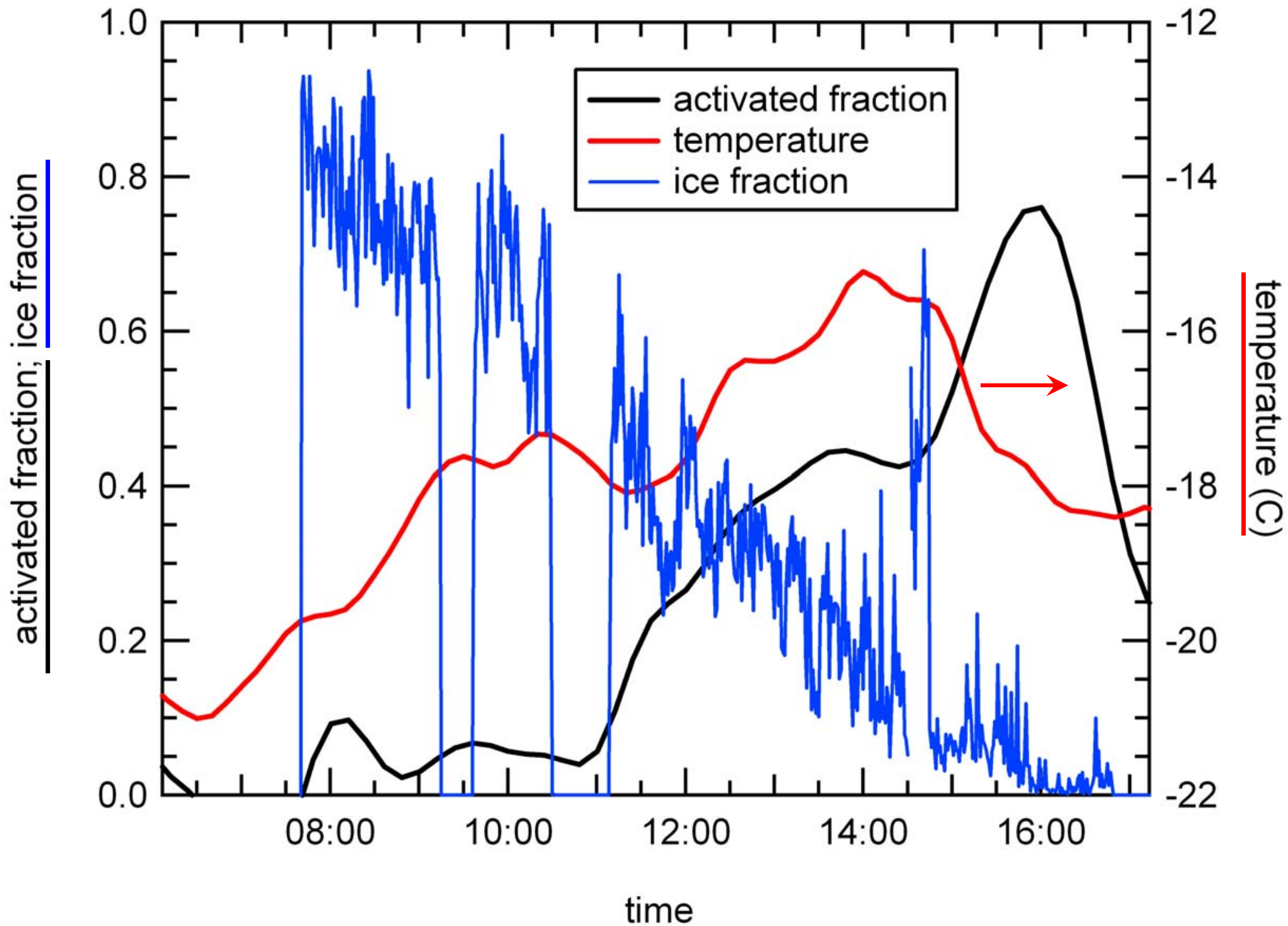
Total aerosol



- Ice residuals mainly consisted of BC and refractory material (mineral dust,…)

Case study II: Mixed phase cloud

March 11, 2004



Analysis of ice particle residuals

ICE-CVI

Lyman-α hygrometer
Condensed water content
Inst. f. Tropos. Res., Leipzig, D

Aerosol Mass Spectrometer (AMS)
SO_4^{2-}, NO_3^-, NH_4^+, OC concentration
Max-Planck-Institute Mainz, D

Condensation Particle Counter (CPC)
Particle number concentration
Inst. f. Tropos. Res., Leipzig, D

Scanning Mobility Particle Sizer (SMPS)
Particle number size distribution
Paul-Scherrer-Institute, Villigen, CH

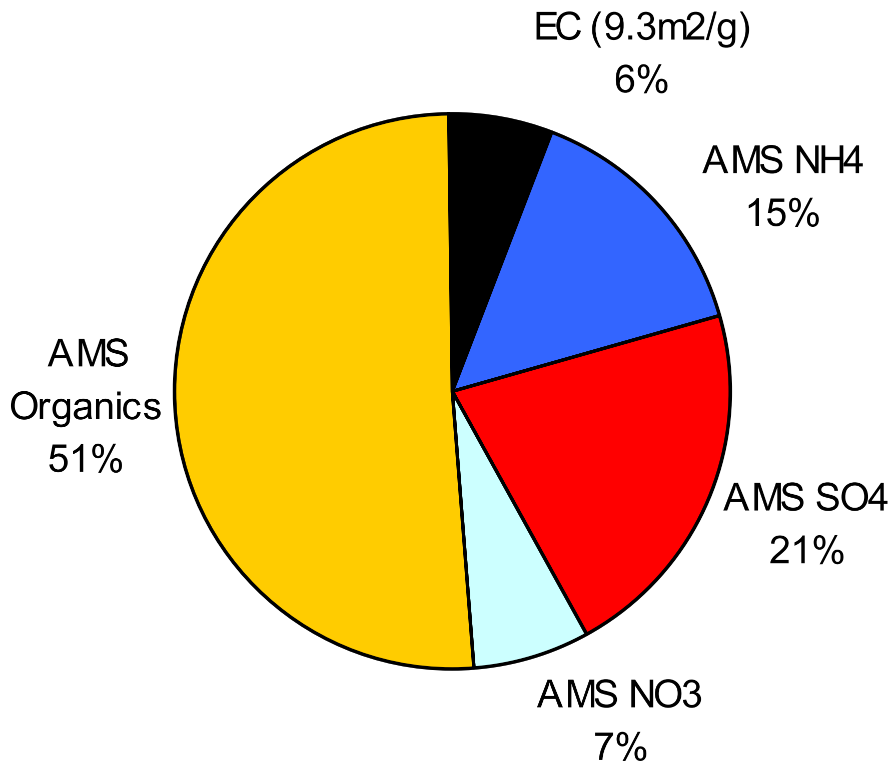
Particle Soot Absorption Photometer (PSAP)
Graphitic carbon concentration
Inst. f. Tropos. Res., Leipzig, D

Environmental Scanning Electron Microscope (ESEM)
Elemental composition, morphology
Technical University Darmstadt, D

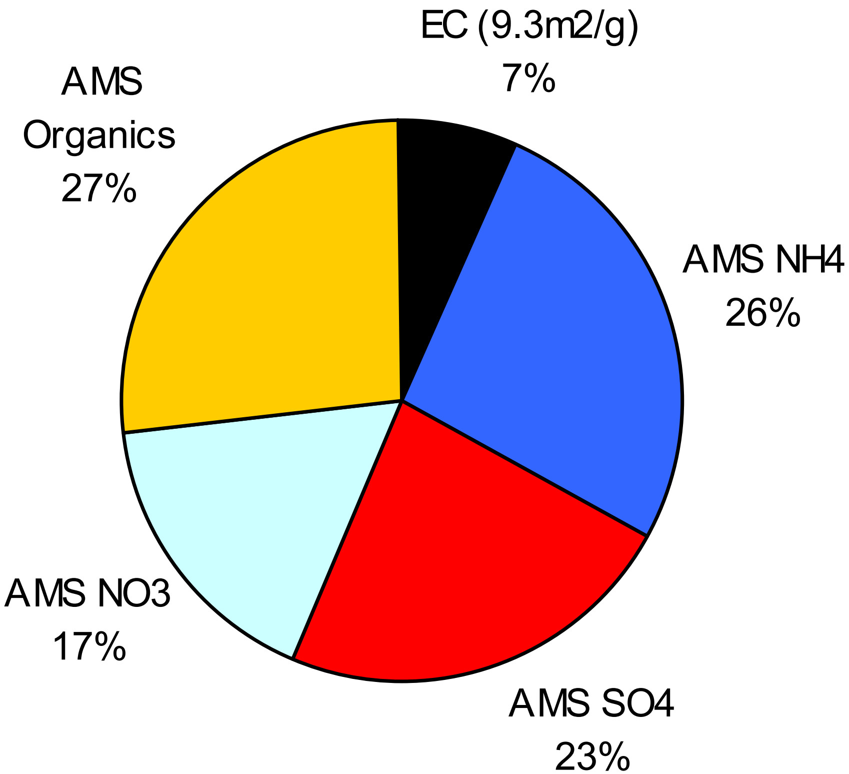
Instrumentation downstream the ICE-CVI inlet during CLACE-3

Chemical Composition (PM1) at the JFJ

Grand average, from AMS and Aethalometer



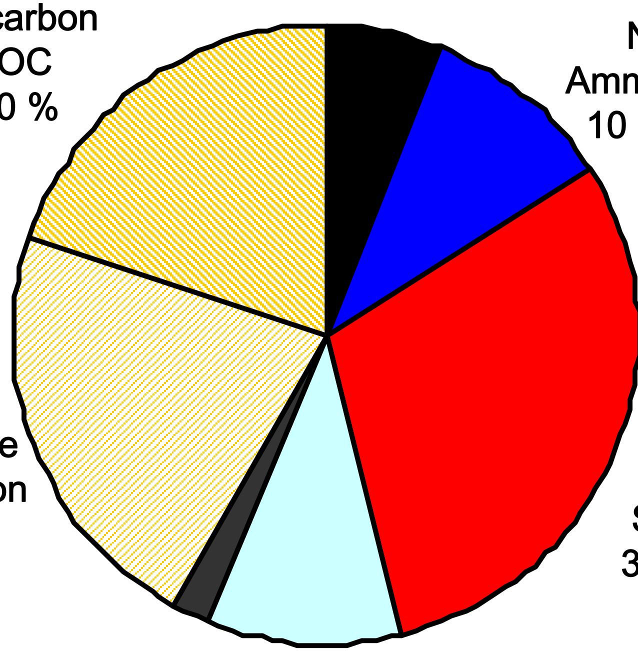
Summer 2002 (CLACE-2)



Winter 2004 (CLACE-3)

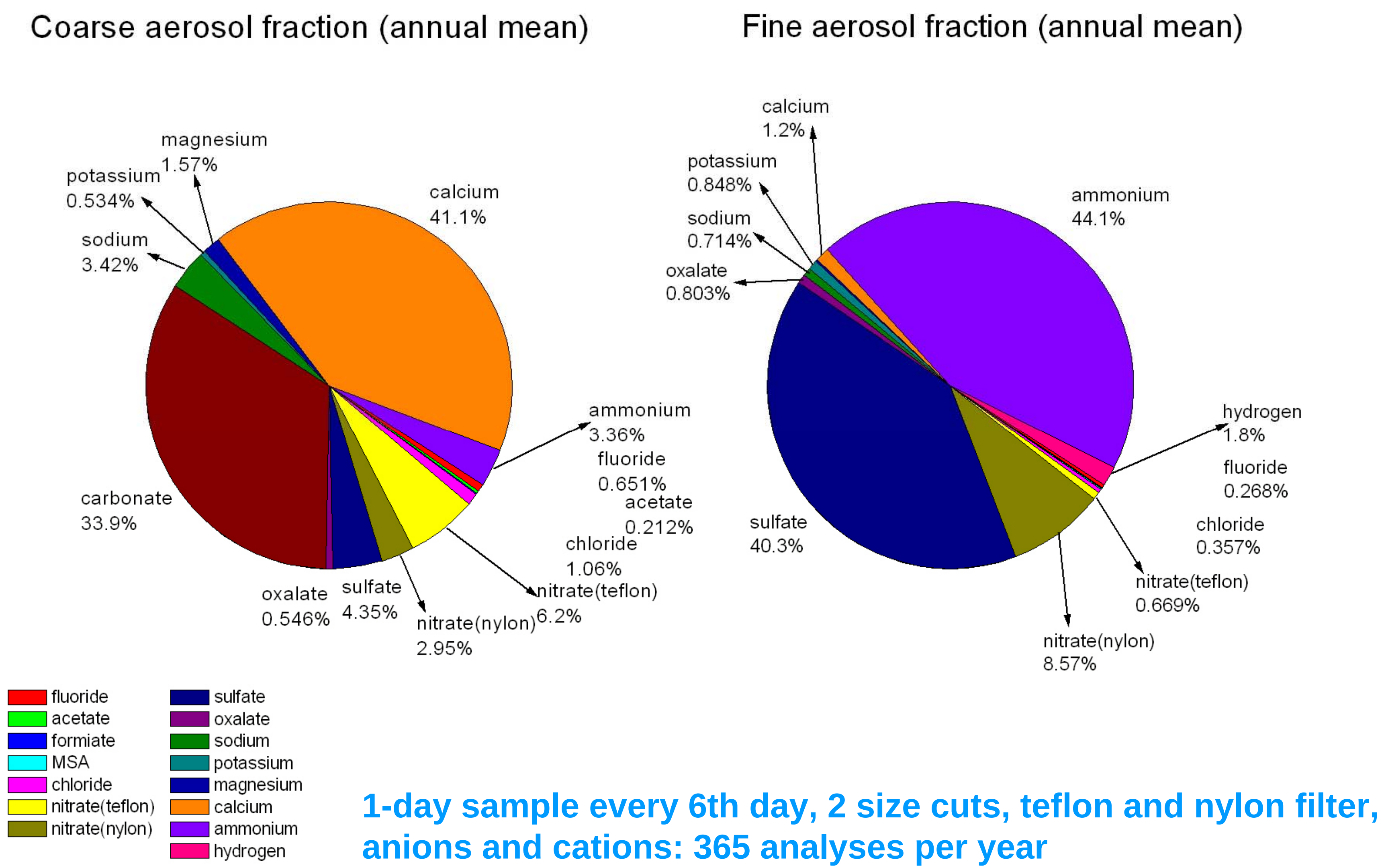
Chemical composition of the Jungfraujoch aerosol during summer

Analyzed offline with IC, solid phase extraction combined with TOC Analyzer
Summer 1998, PM2.5, 8 High Vol Samples



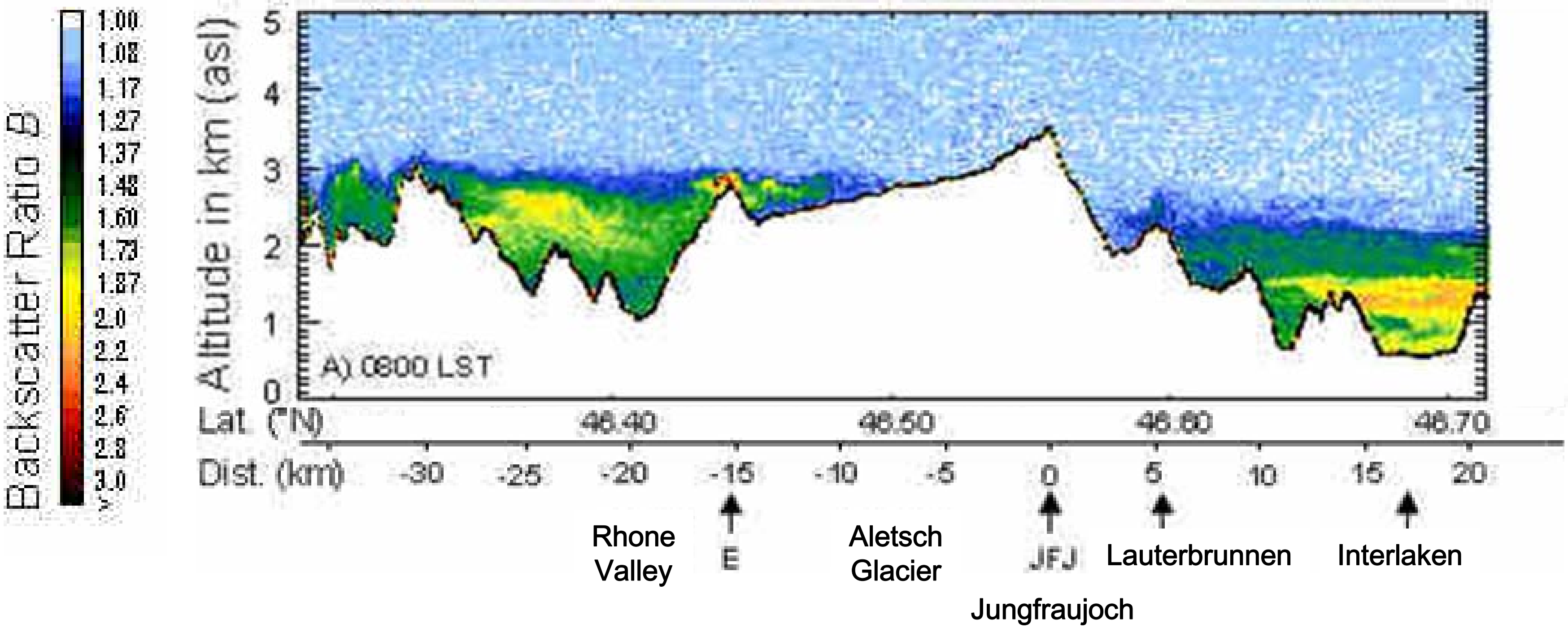
Krivácsy et al. (2001)
J. Atmos. Chem, 39

Water soluble components of the Jungfraujoch aerosol



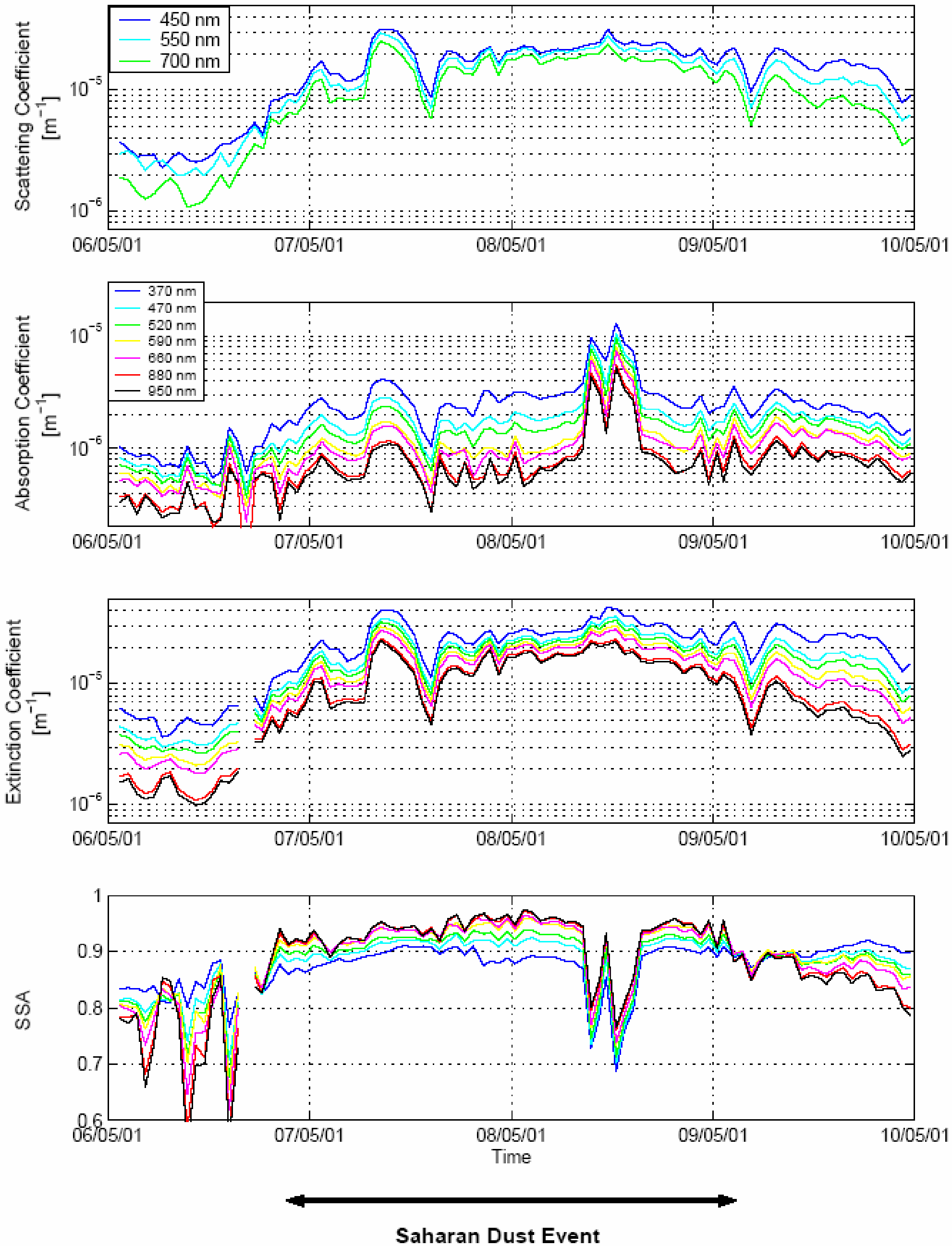
1-day sample every 6th day, 2 size cuts, teflon and nylon filter, anions and cations: 365 analyses per year
Henning et al., J. Geophys. Res. (2003)

Evolution of the planetary boundary layer during a summer day in July 1997 over the Jungfraujoch massif

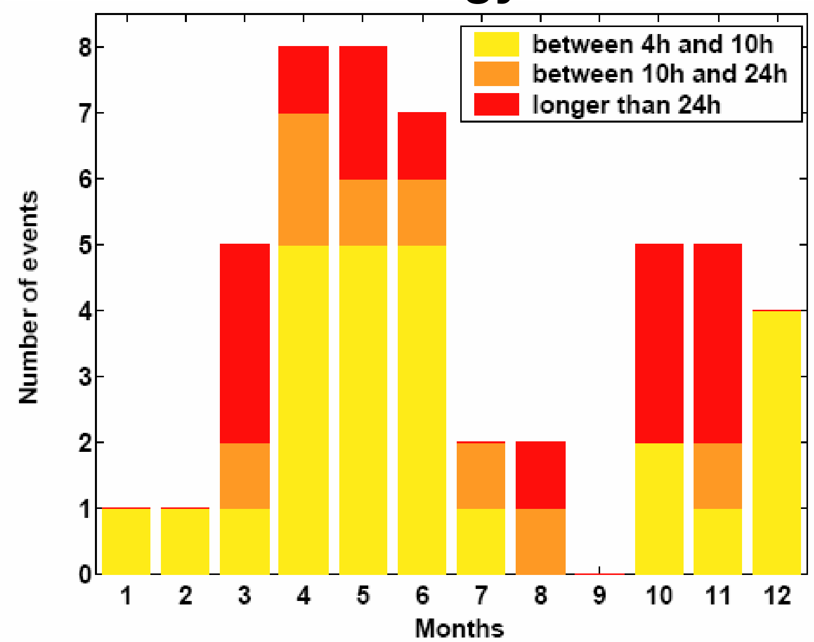


Nyeki *et al., Geophys. Res. Lett., 27*, 689-692, 2000

The single scattering albedo can be used to detect Saharan dust events



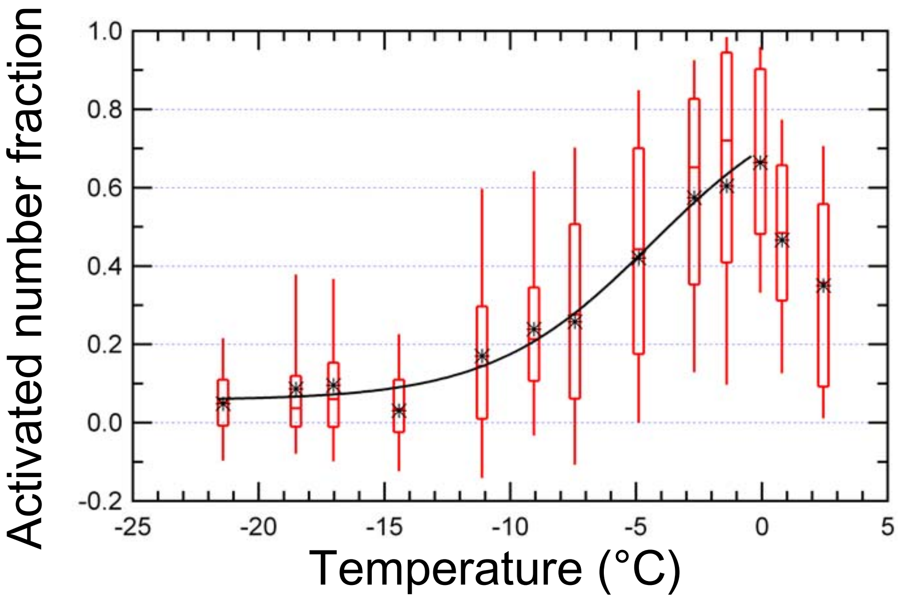
Climatology of SDE

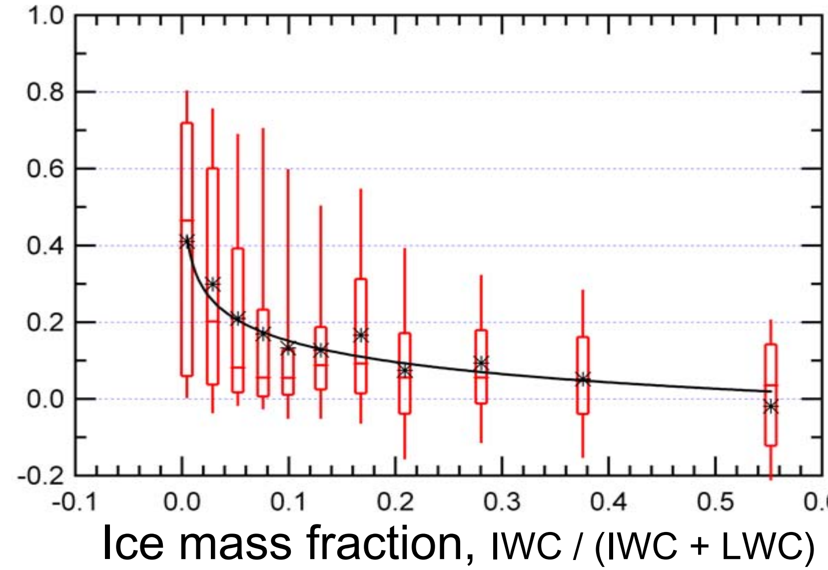


Collaud Coen et al., ACP 2004

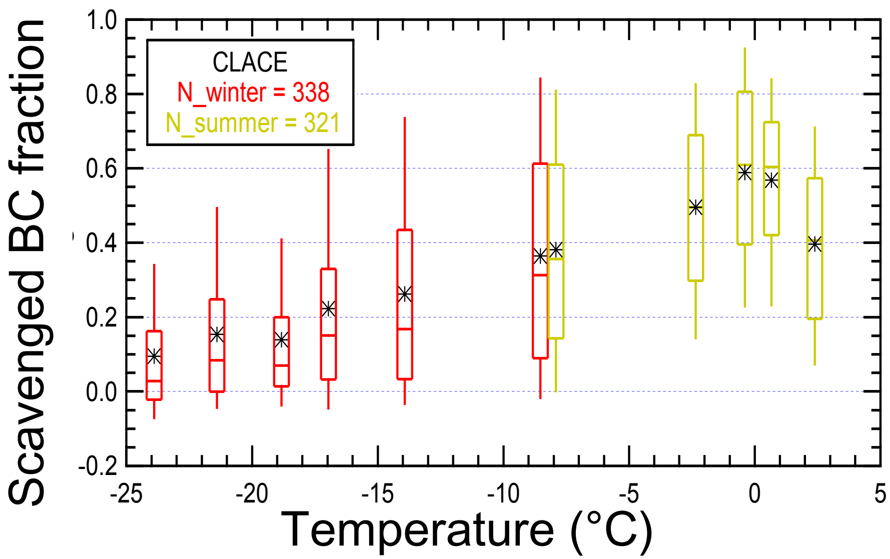
Statistical analysis of more than 600 hour of in-cloud observations…

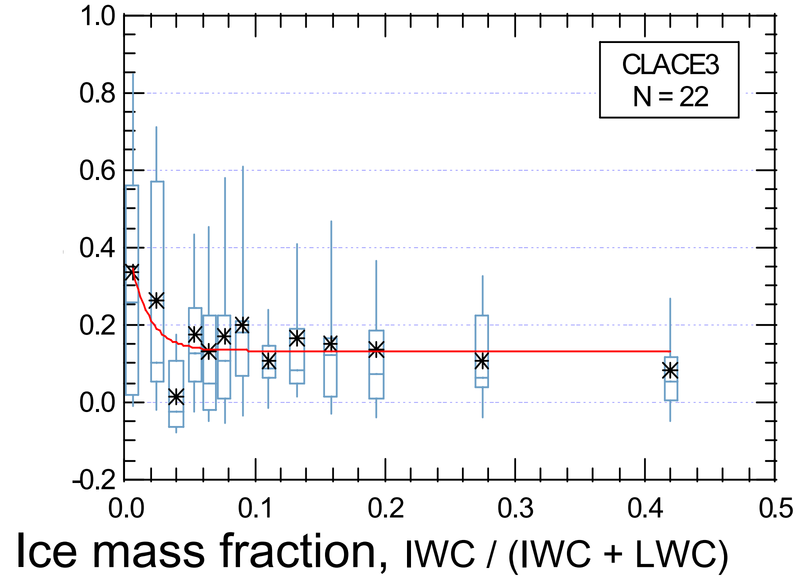
… of activated number:





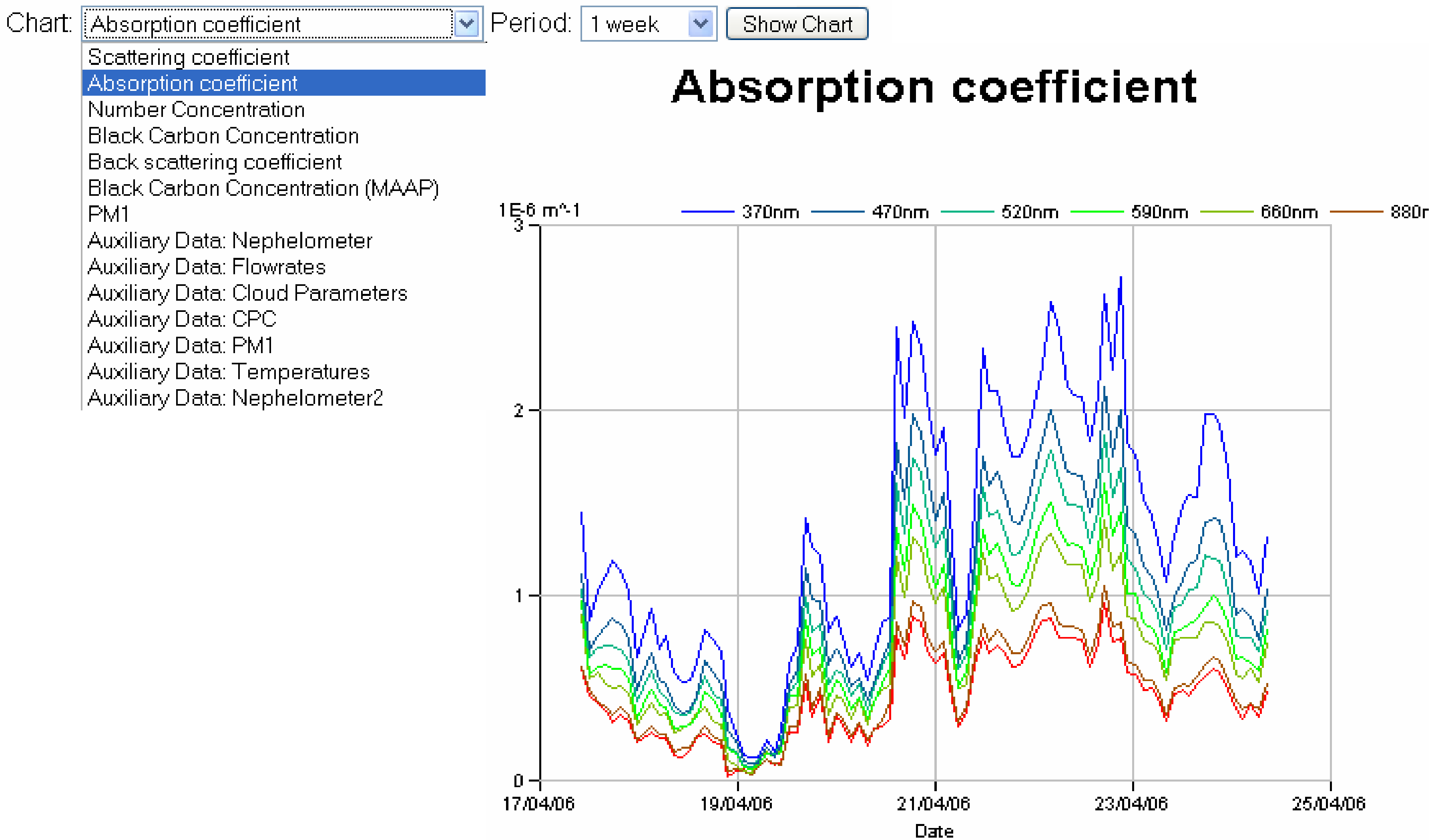
… of scavenged black carbon (BC):



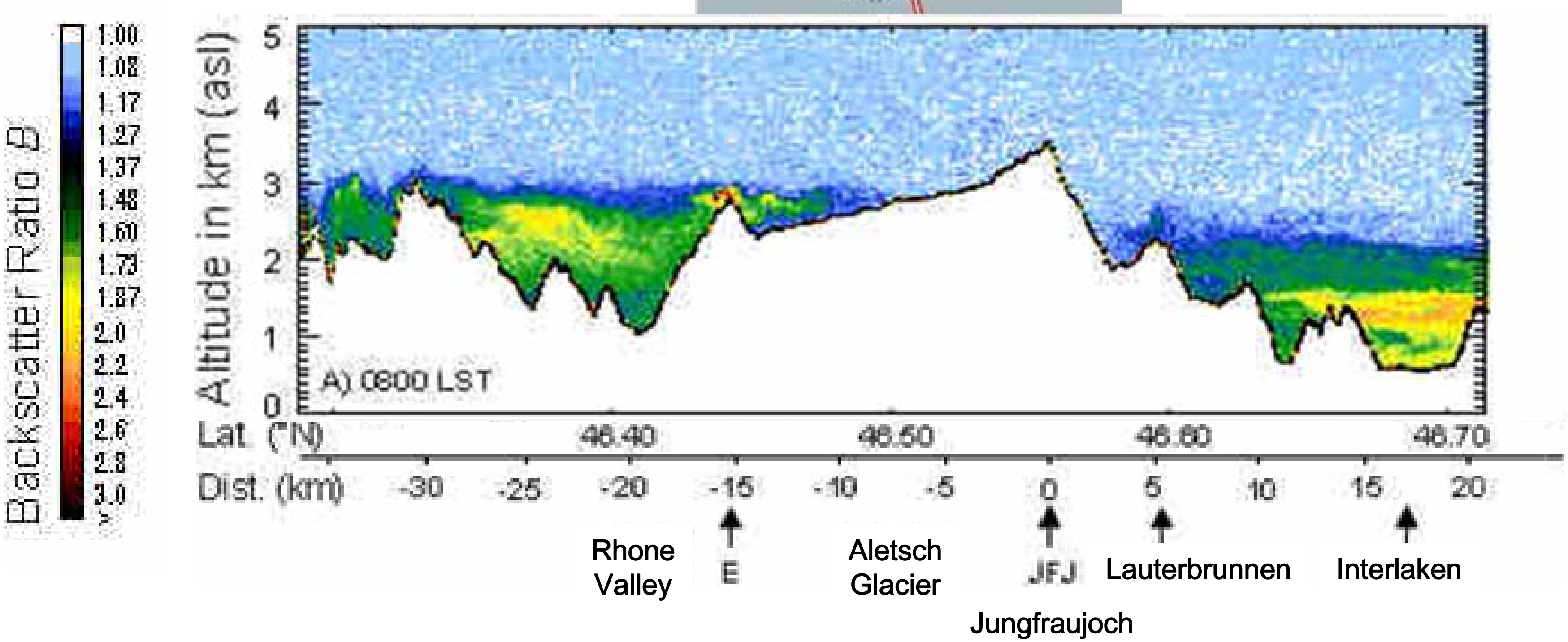


On-line data from the JFJ

http://aerosolforschung.web.psi.ch/



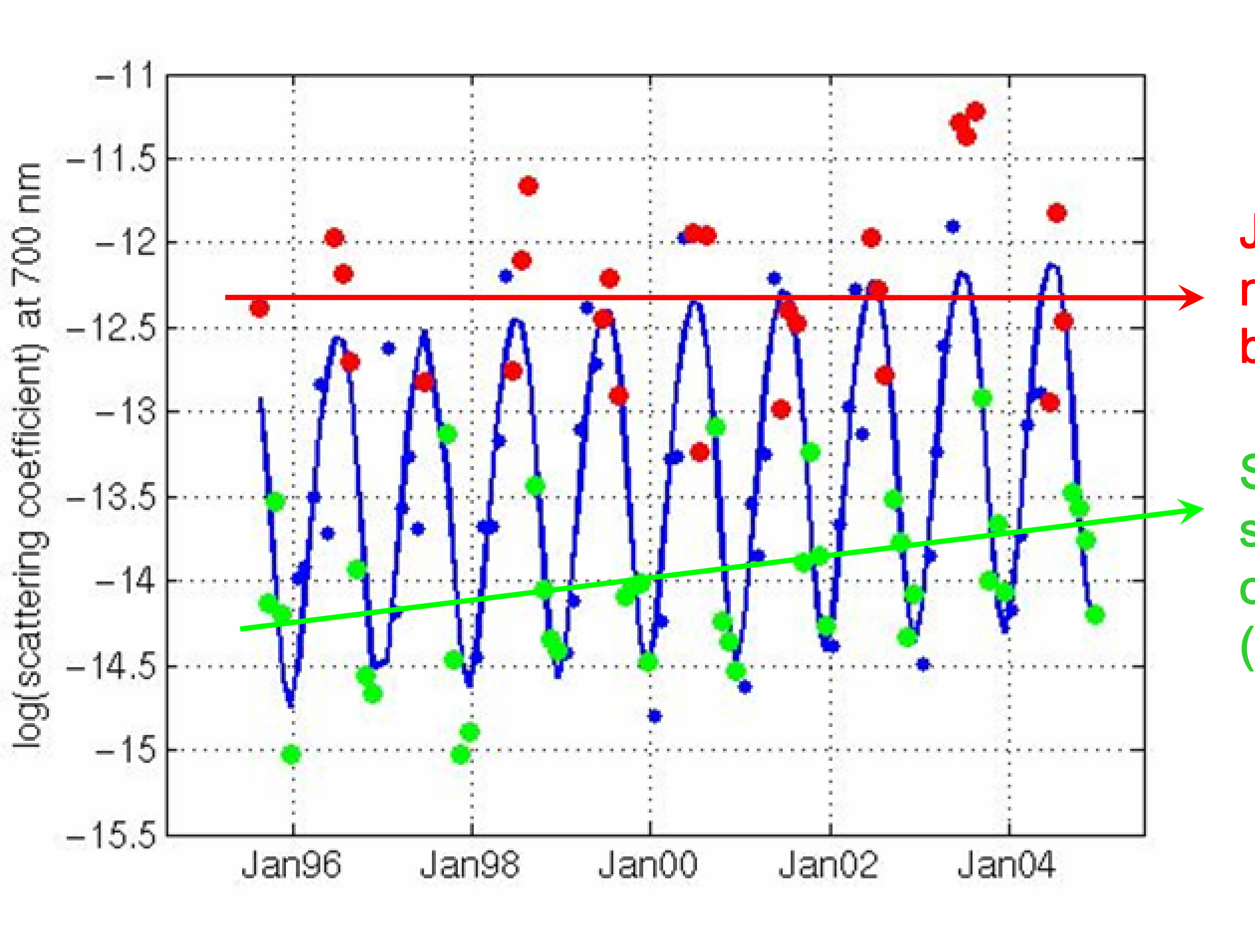
Evolution of the planetary boundary layer during a summer day in July 1997 over the Jungfraujoch massif



Nyeki *et al., Geophys. Res. Lett., 27*, 689-692, 2000

Long-term trend calculations by Martine Collaud Coen (MeteoSwiss)

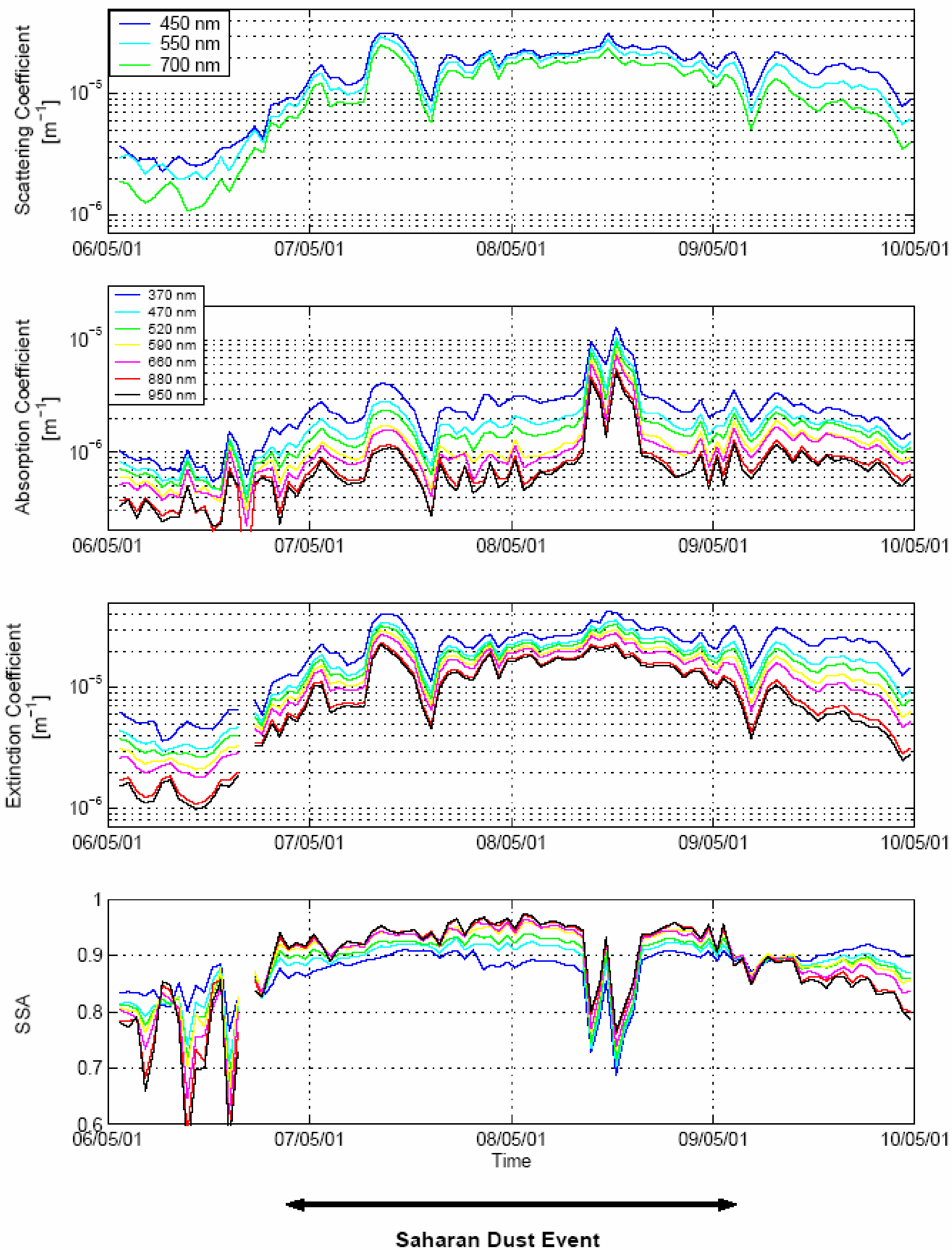
10 years of data are necessary



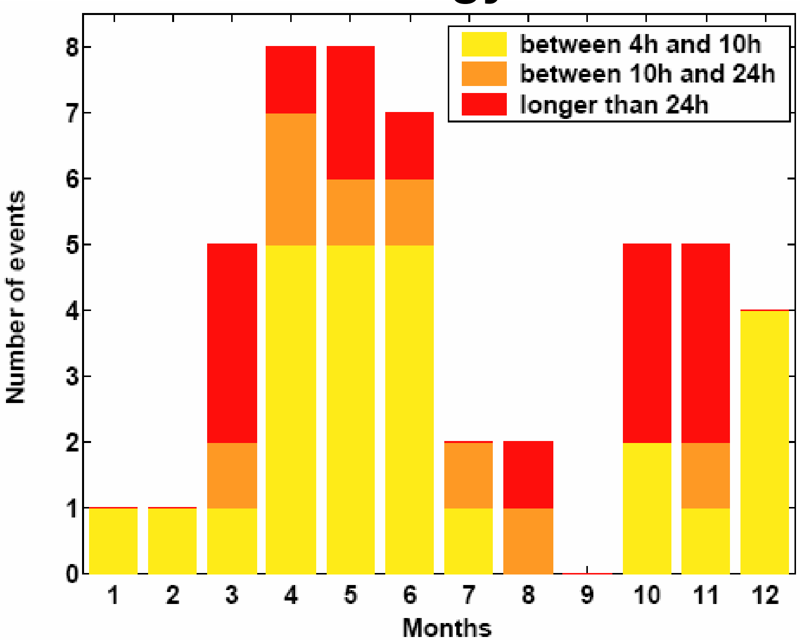
June - August: no significant trend of b_s, b_{abs}, and CN

September - December: significant positive trend of 2 - 4% per year (for b_s, b_{abs}, and CN)

The single scattering albedo can be used to detect Saharan dust events



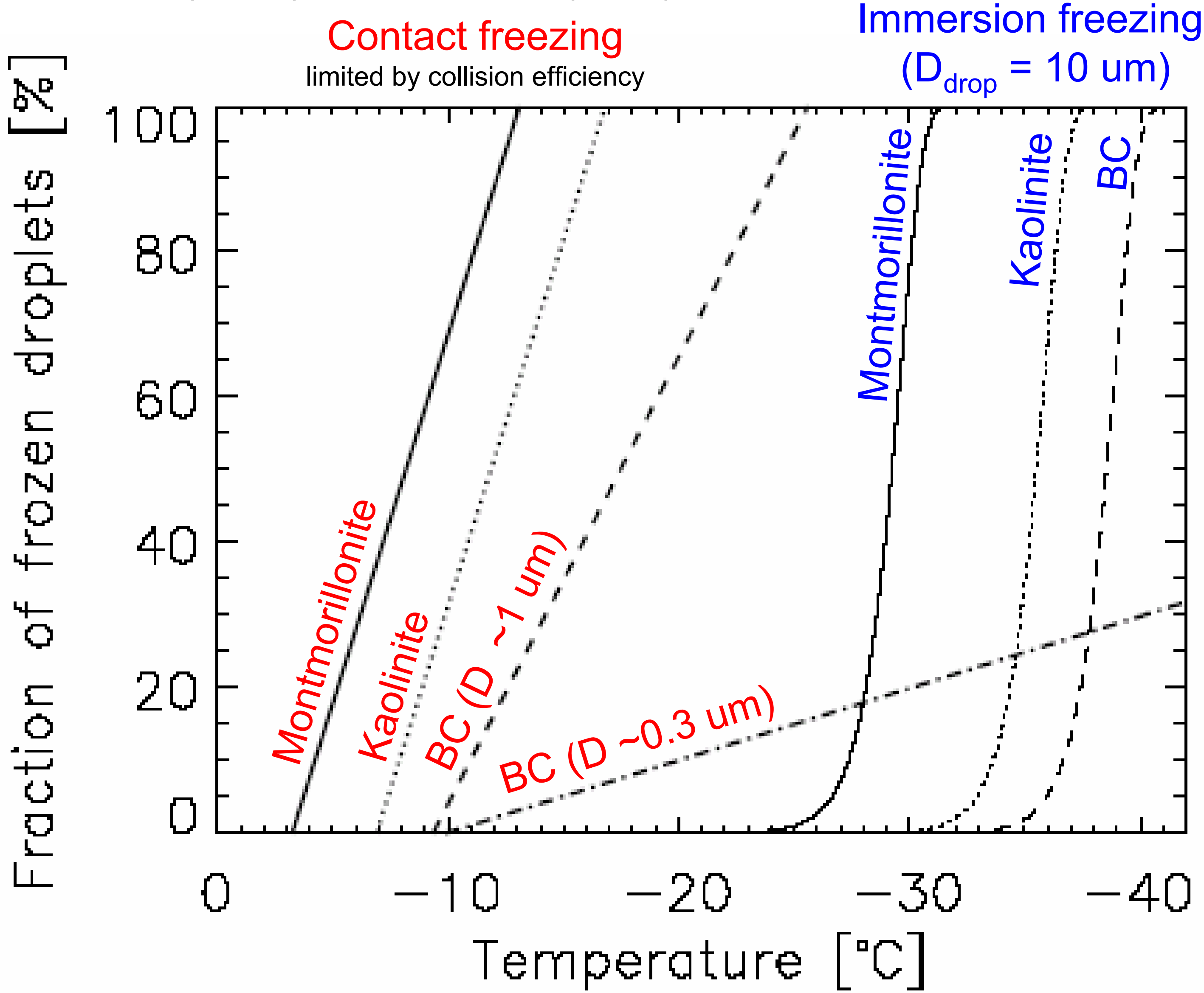
Climatology of SDE



Collaud Coen et al., ACP 2004

Model input from a compilation of lab data

Montmorillonite (MON) and Kaolinite (KAO) as model substances for mineral dust



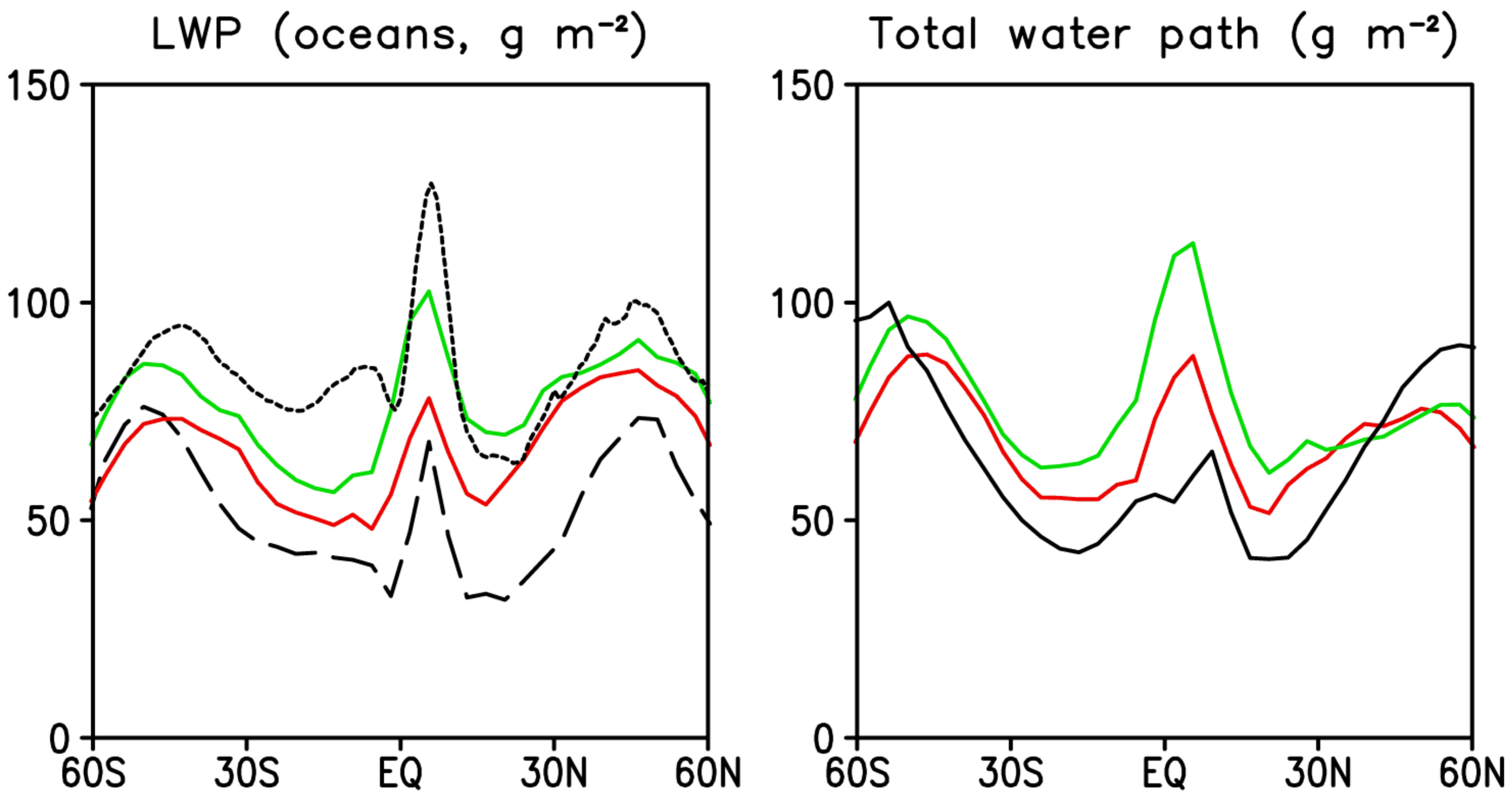
Adapted from Lohmann und Diehl (2006), JAS

Model results: zonal annual means

Green: ECHAM4 Simulation with JFJ data

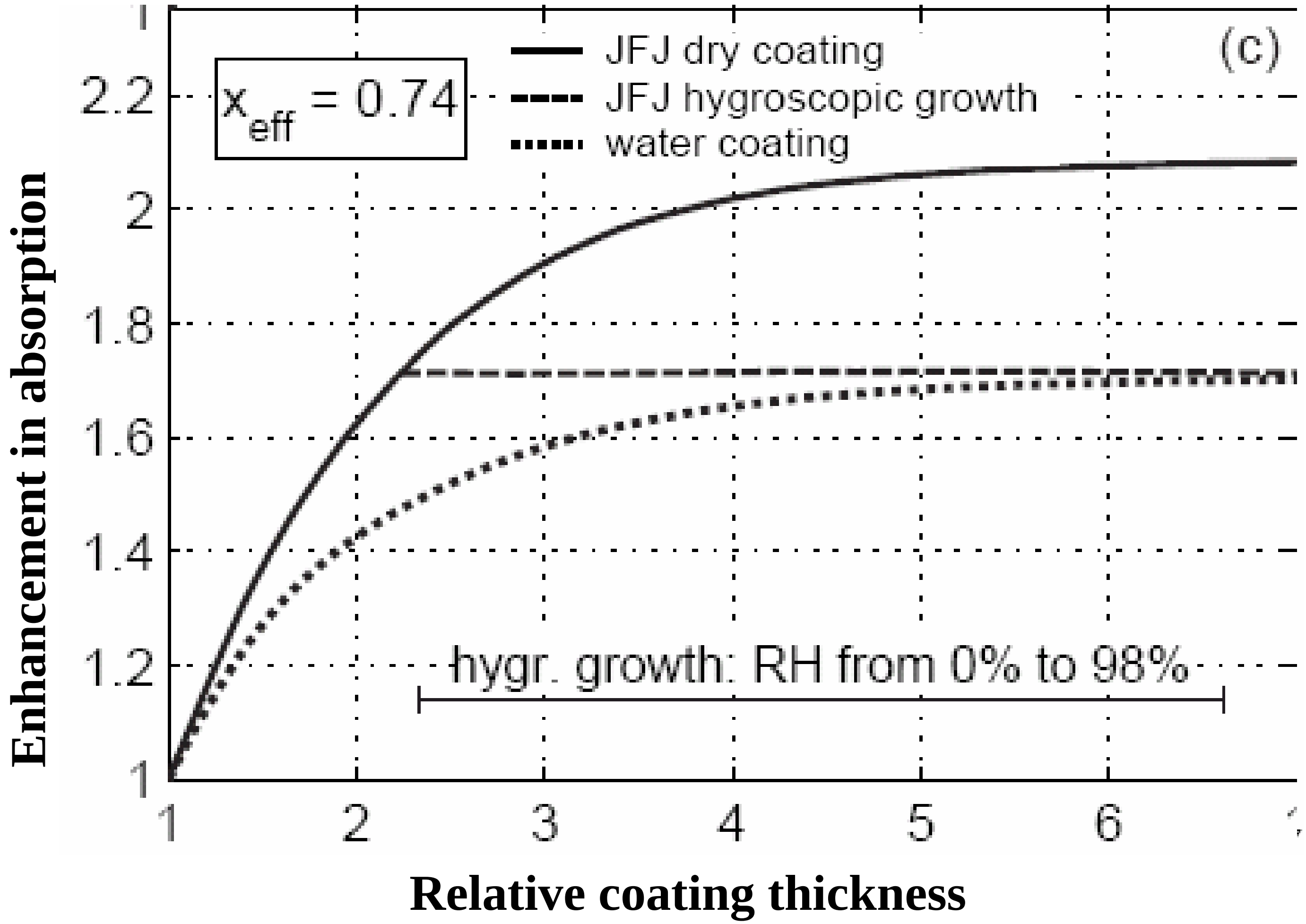
Red: Standard simulation that considered freezing of dust and black carbon

Black: "Validation" with observations deduced from satellite measurements



Model calculation for the conditions at the Jungfraujoch

(polydisperse size distribution and coating including the hygroscopic properties)

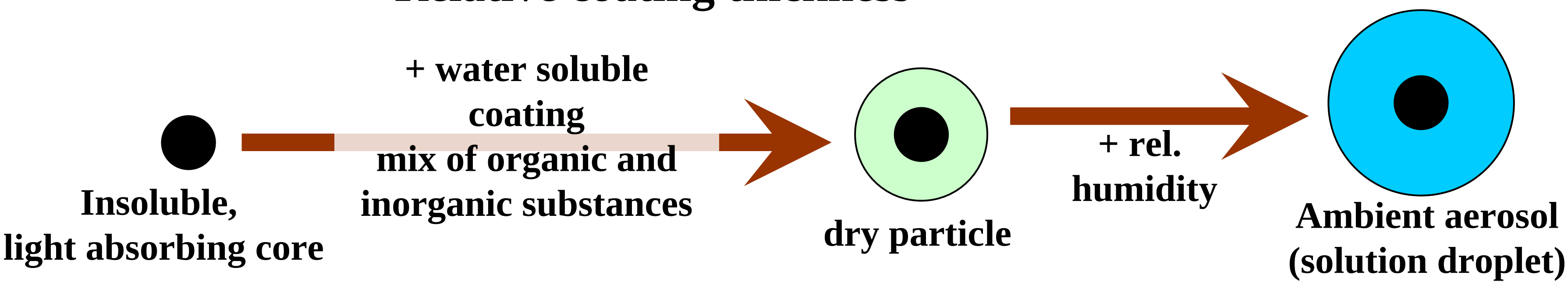


Effective size parameter

$$x_{\text{eff}} = \frac{\pi\, D_{\text{eff}}}{\lambda}$$

$$D_{\text{eff}} = \frac{\int_0^\infty n(D)\, D^3\, \mathrm{d}D}{\int_0^\infty n(D)\, D^2\, \mathrm{d}D}$$

R. Nessler et al., Submitted to JAS 2004



[IN] << [CCN]

- Ice nuclei (IN) are far less abundant in the atmosphere than cloud condensation nuclei (CCN). (Typical CCN and IN concentrations: 100 cm-3 and 0.01 cm-3)
- Hence, in an ice cloud, cloud water is typically distributed on fewer cloud particles than in a liquid cloud.
- Consequently, the ice crystals are larger than the cloud droplets and therefore more likely to fall out as precipitation

Main requirements for IN (Pruppacher & Klett)

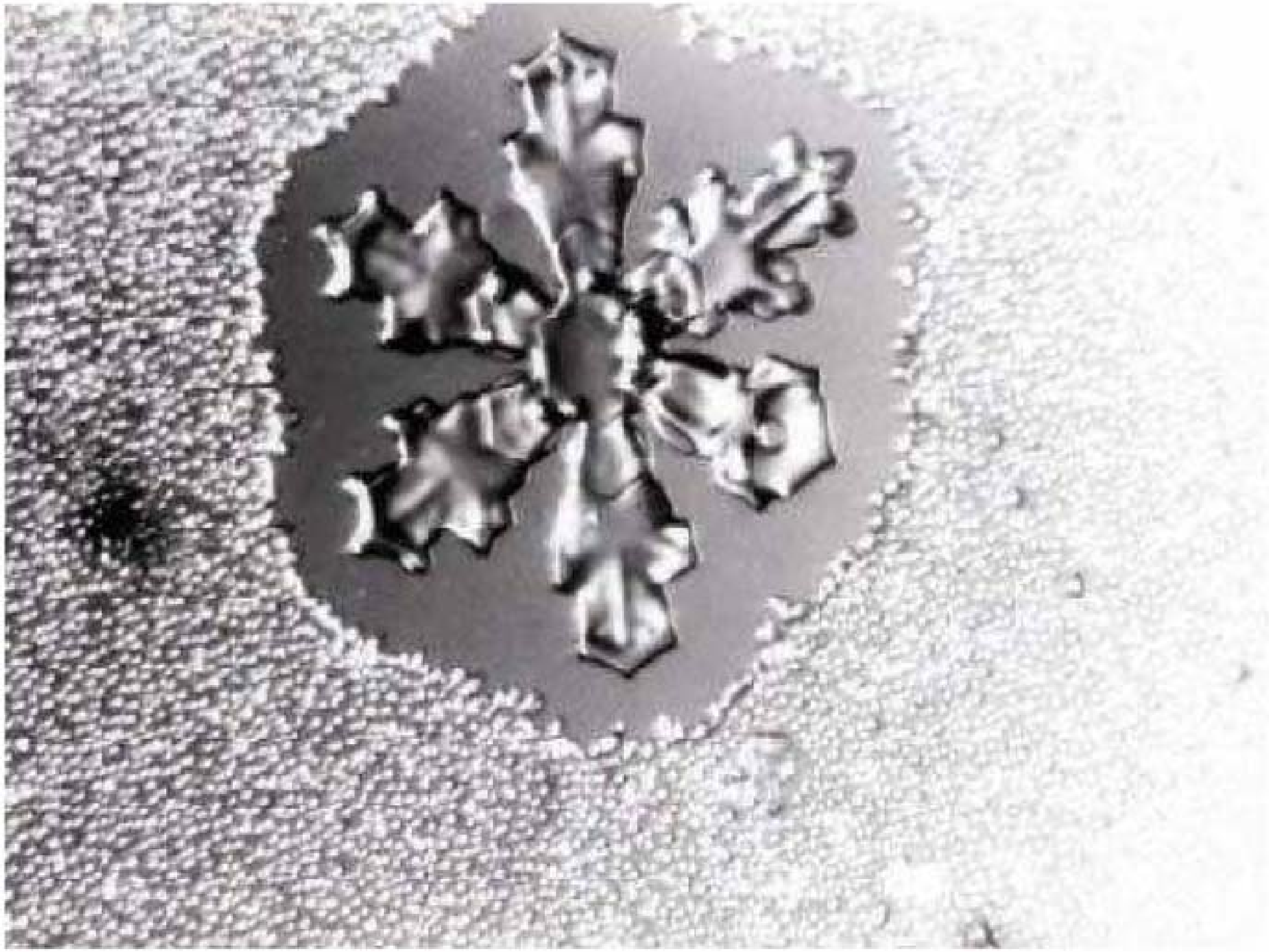
- Insolubility requirement: A rigid substrate is needed for the ice ”germ” formation.
- Size requirement: The Aitken range is less efficient than the accumulation mode range.
- Chemical bond requirement: A similar bond as the ice crystal lattice is beneficial.
- Crystallographic requirements. The geometrical arrangement of the aerosol is important.

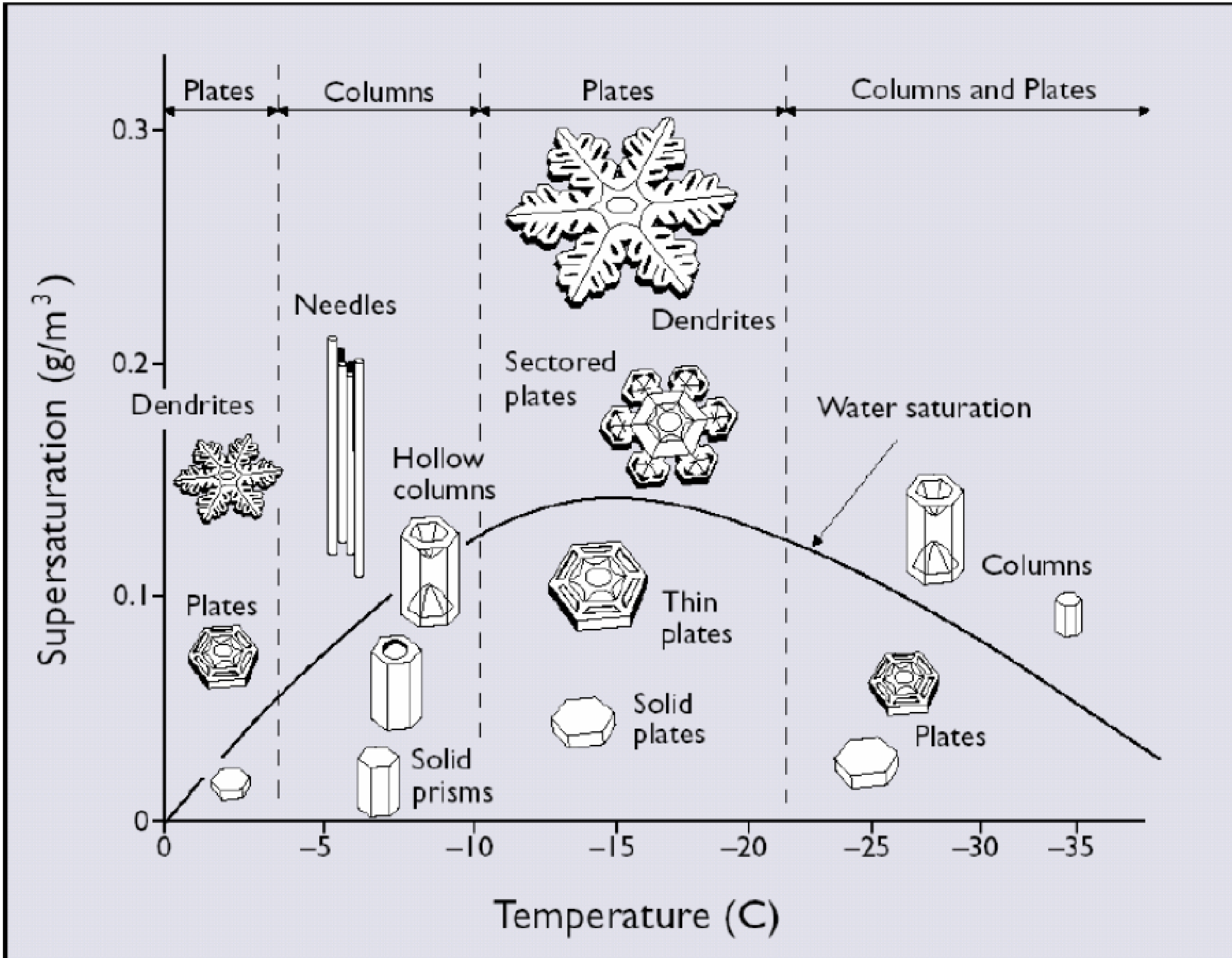
In what ways are cold clouds different?

- Ice crystals form by various heterogeneous freezing processes between 0°C and approximately -35°C. This refers to ice crystals forming with the aid of ice nuclei (IN).
- IN are typically insoluble aerosols.
- Homogeneous freezing becomes effective below approximately -35°C. This is the process by which cloud droplets freeze spontaneously without any further aid from aerosols.
- Ice clouds are often optically thin, such that changes in cloud optical depth become important also in the LW

Homogeneous vs. Heterogeneous Freezing

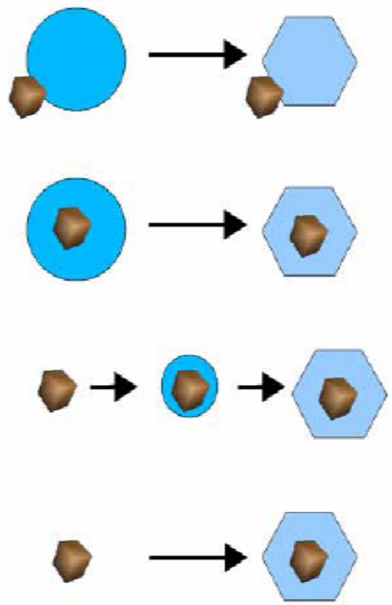
- At temperatures T < -35°C ice crystals can form through homogeneous freezing of aqueous solution droplets, only if ice supersaturations exceed ~40% *(Koop et al., Nature 2000)*.
- Such supersaturations have been observed in the upper troposphere (*Jensen et al., JGR 2001*).
- However, if there are sufficient IN present, heterogeneous freezing processes are believed to dominate, as they require lower supersaturations.




Plates
Columns
Plates
Columns and Plates
0.3
0.2
0.1
0
Supersaturation (g/m^3)
Needles
Dendrites
Dendrites
Sectored plates
Water saturation
Hollow columns
Columns
Plates
Thin plates
Solid plates
Plates
Solid prisms
0
−5
−10
−15
−20
−25
−30
−35
Temperature (C)

Heterogeneous Freezing

- 4 nucleation modes



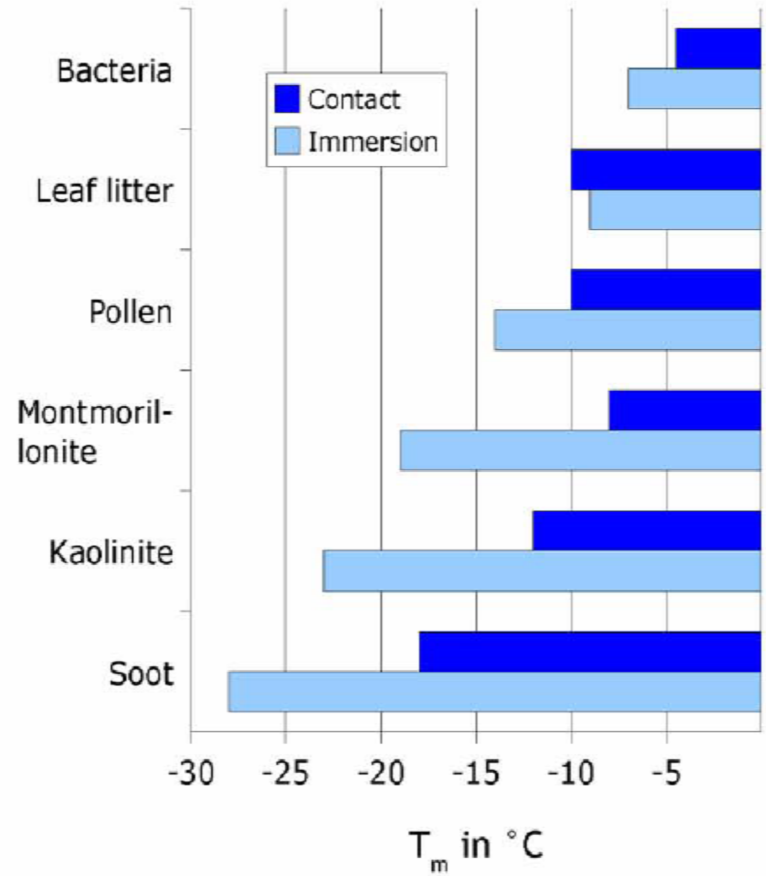
contact freezing

immersion freezing

condensation freezing

deposition

- IN efficiencies depend on material and drop volume
- Efficient IN: bioaerosols, dust, soot



Median freezing temperatures for different IN from lab experiments. Drop radii 250-350 µm. Adapted from *Diehl et al. (subm.)*.

Conclusions

Atmospheric aging processes lead to an **increase in the hygroscopic growth** of aerosol particles, this changes the particle's **optical properties**, their **lifetime** and is of importance for their **radiative climate forcing**.

At the Jungfraujoch, in the lower free troposphere:

The aerosol is predominately **internally mixed**, no less hygroscopic mode is visible

No deliquescence behavior was observed (**liquid state**)

The particles hygroscopicity at the Jungfraujoch is higher in winter than in summer. This is mainly a result of varying fractions of organic/inorganic mass.

In mixed phase clouds the **Bergeron-Findeisen process** determines the aerosol partitioning

- The activation in mixed phase clouds strongly depends on temperature.
- This is valid for particle number, volume and BC content.

Promising results from the **novel ICE-CVI**, sampling ice crystal residuals:

- Comparison of SMPS and AMS data indicates that preferably non volatile particles act as ice nuclei.
- Large residuals (D>500 nm) are dominated by mineral dust particles.

Currently, the observed partitioning in the mixed phase clouds is included into a **general circulation model**.

Conclusions

- The particles hygroscopicity at the Jungfraujoch is higher in winter than in summer. This is mainly a result of varying fractions of organic/inorganic mass.

- Observational evidence for *Bergeron-Findeisen* process provided:
  - The activation in mixed phase clouds strongly depends on temperature.
  - This is valid for particle number, volume and BC content.

- Promising results from the novel ICE-CVI, sampling ice crystal residuals:
  - Comparison of SMPS and AMS data indicates that preferably non volatile particles act as ice nuclei.
  - Large residuals (D>500 nm) are dominated by mineral dust particles.

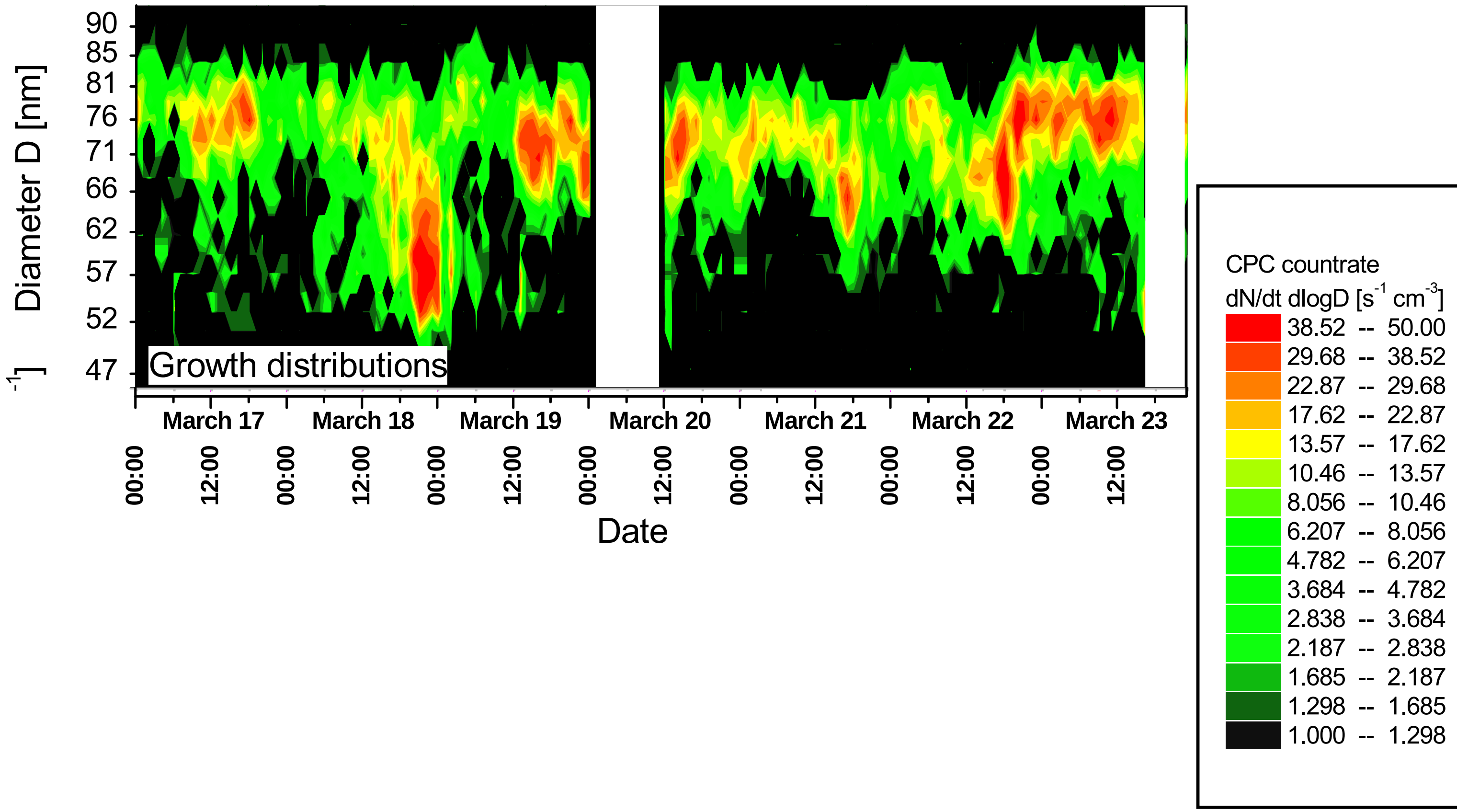
- Currently, the observed partitioning in the mixed phase clouds is included into a general circulation model.

- Future collaborations with the collaborative research centre TROPEIS investigating the tropospheric ice phase funded by the German Science Foundation DFG.

CLACE 1
Winter 2000

Hygroscopic growth of d_o = 50 nm particles

at T = –10°C
RH = 85%



CLACE 1
Winter 2000

Hygroscopic growth of d_o = 50 nm particles

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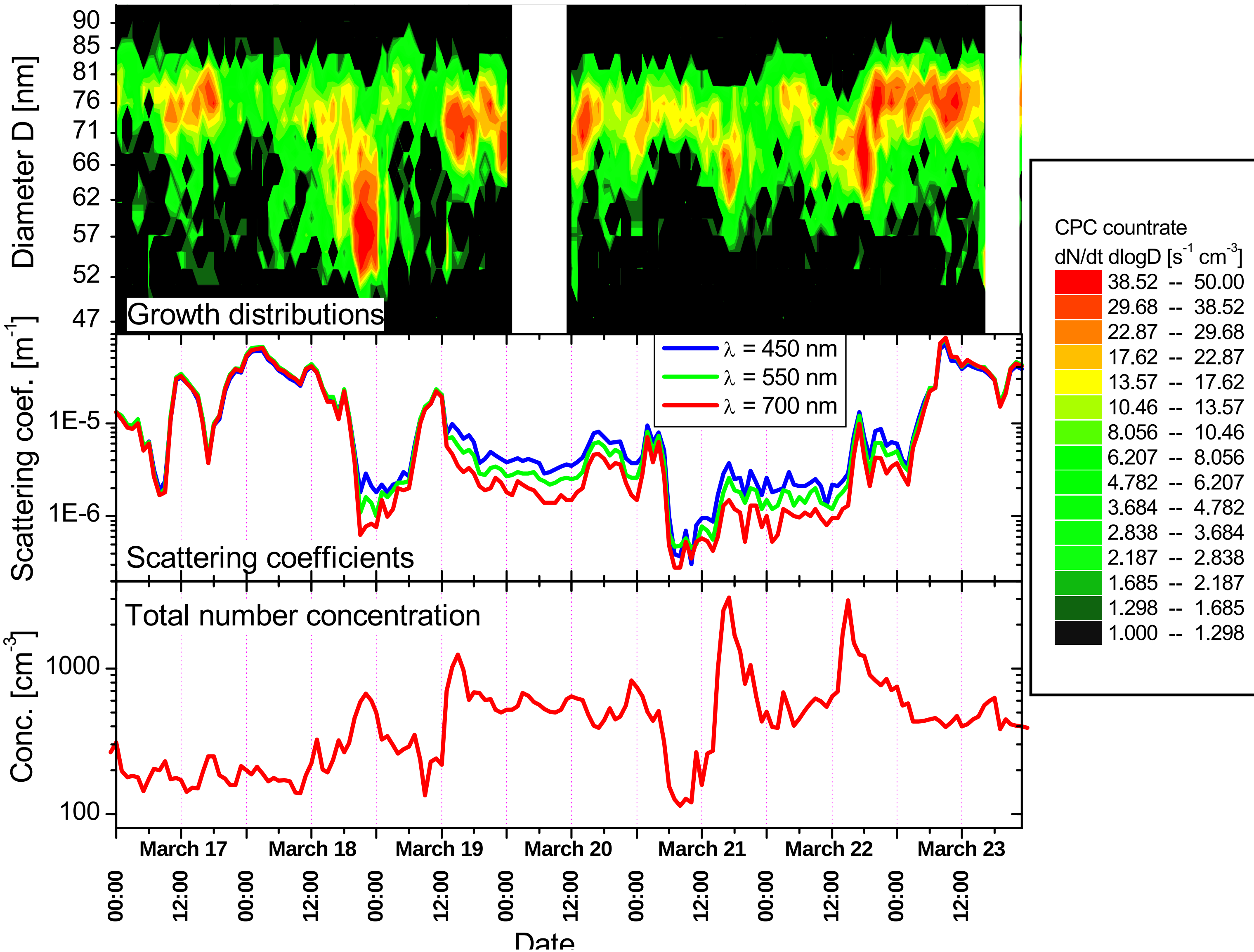
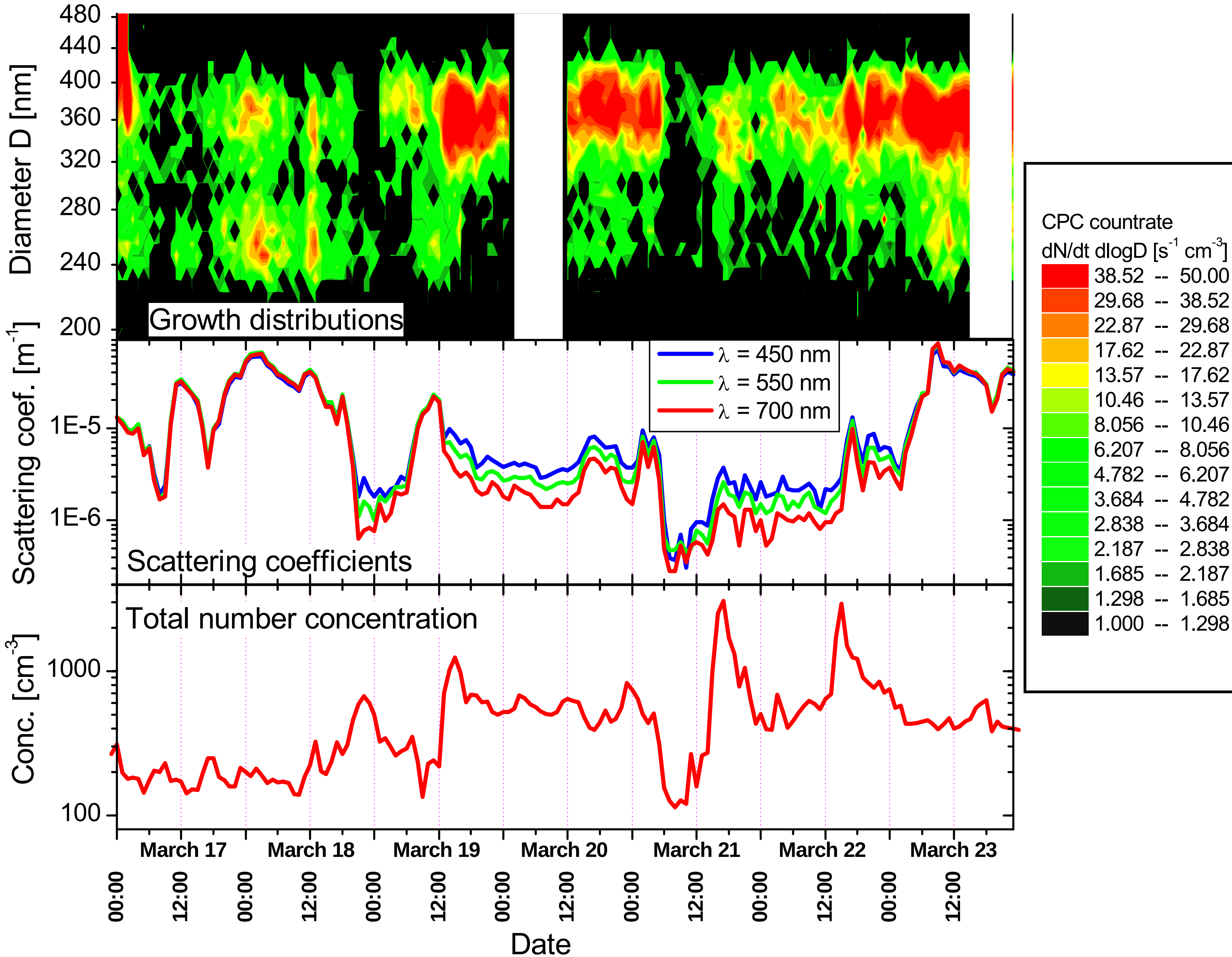
RH = 85%



CLACE 1
Winter 2000

Hygroscopic growth of d_o = 250 nm particles

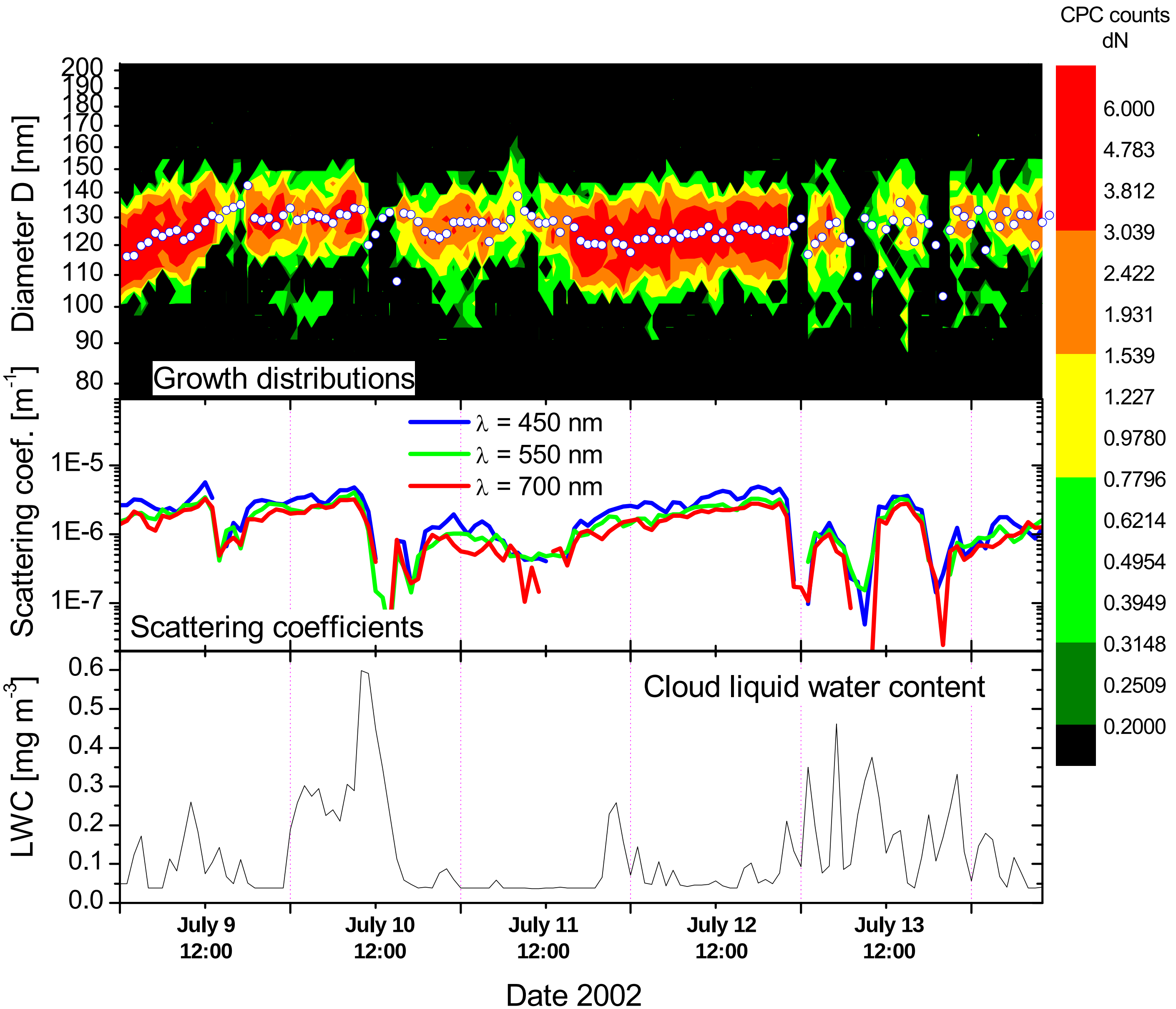
at T = –10°C
RH = 85%



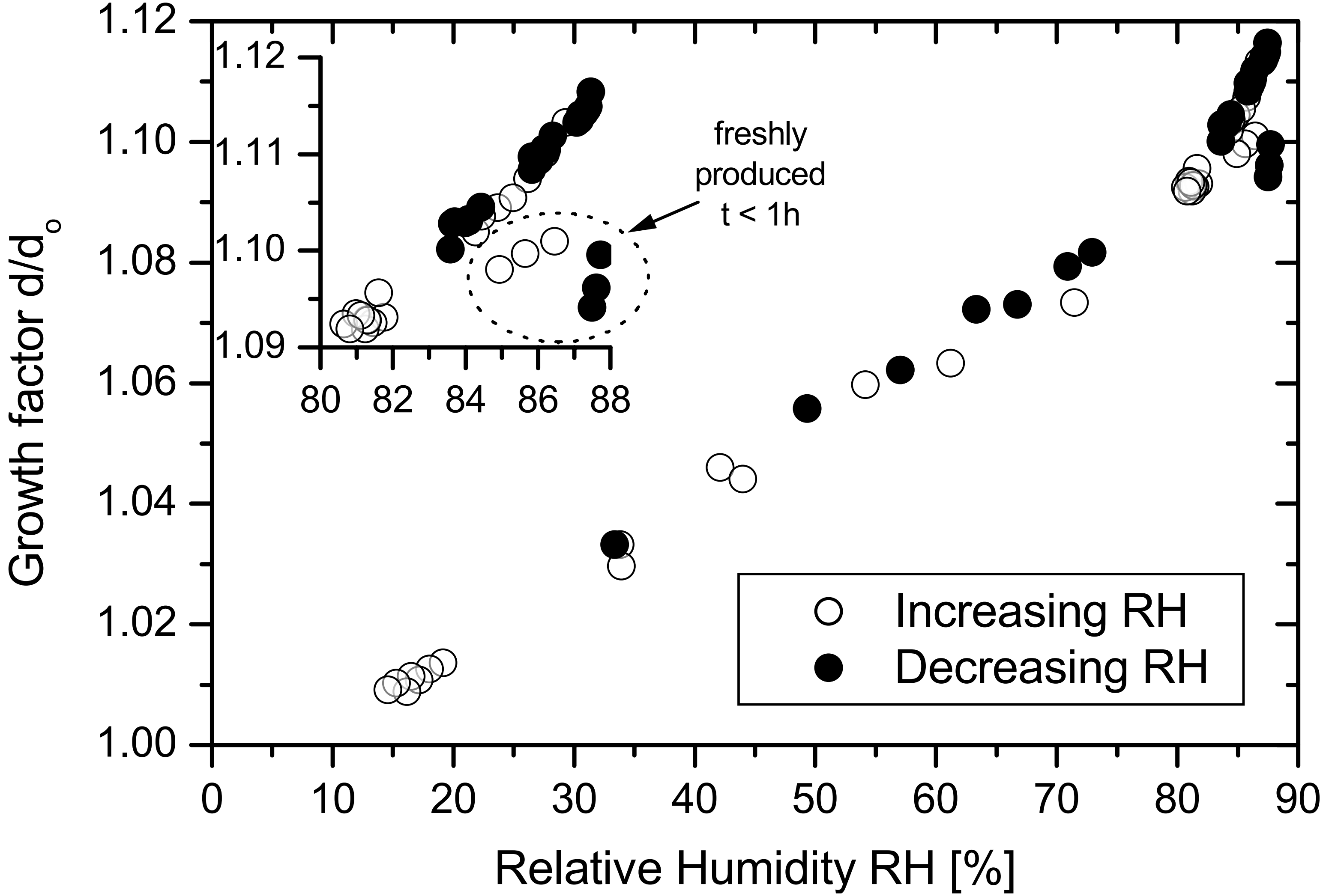
CLACE 2
Summer 2002

Hygroscopic growth of d_o = 100 nm particles

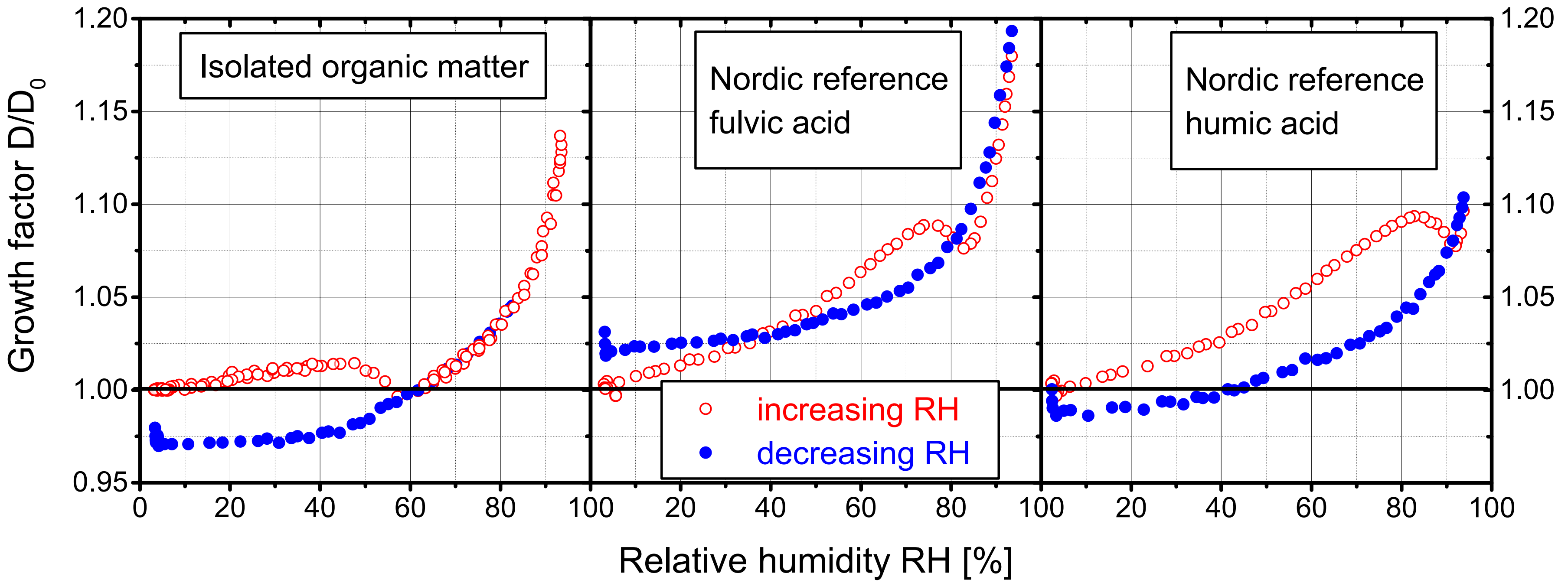
at T = 0°C
RH = 85%



Hygroscopic growth of organic particles (produced by the ozonolysis of α-pinene)



Hygroscopic growth factors of isolated organic matter (ISOM) from atmospheric aerosols and selected reference substances



Aerosol sample from K'puzta

Gysel et al., submitted to ACPD