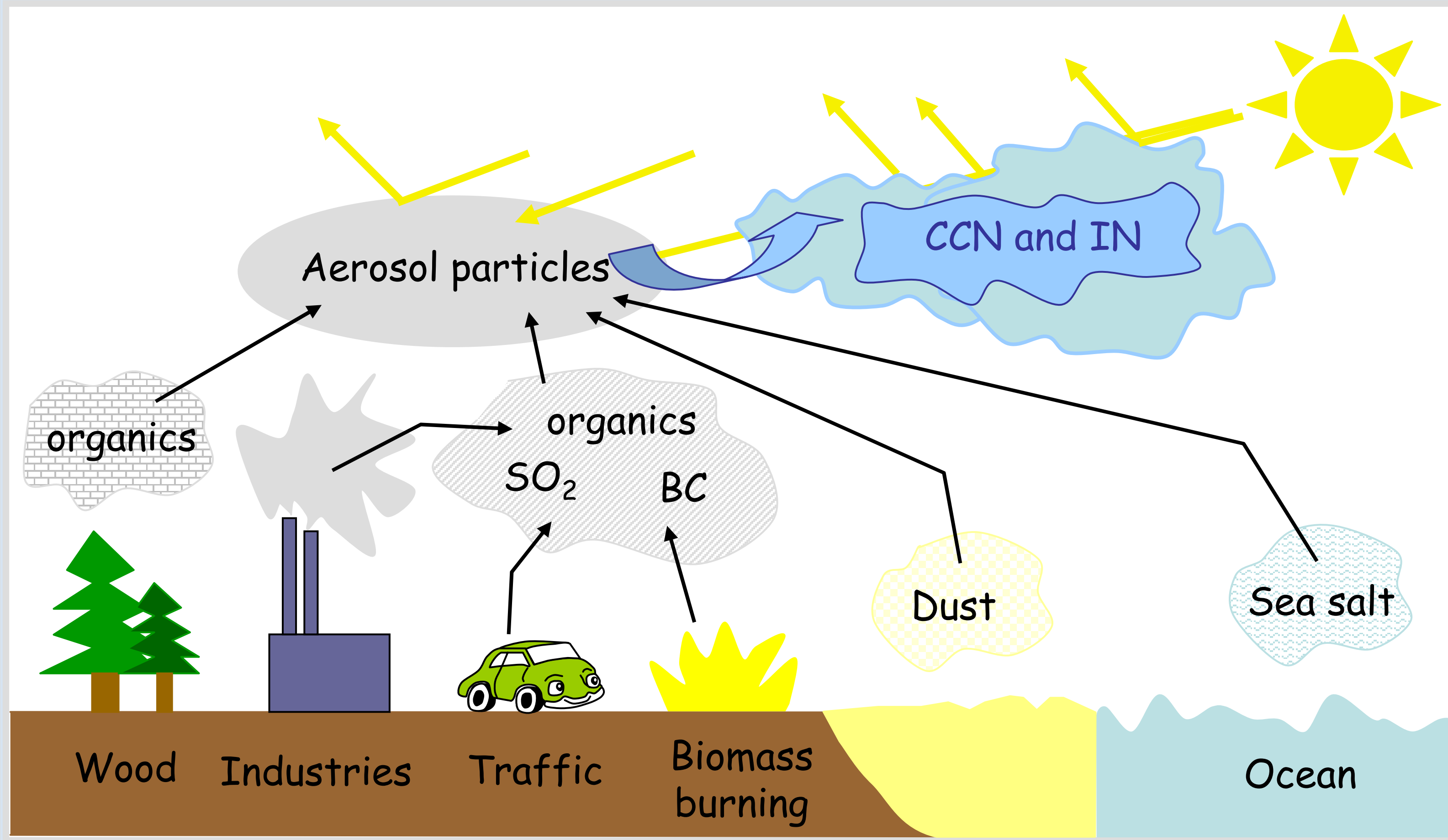
The Fate of Black Carbon in the Atmosphere: Rapid Removal by Wet Deposition after Aging

J. Cozic
Laboratory of Atmospheric Chemistry
(Paul Scherrer Institute)

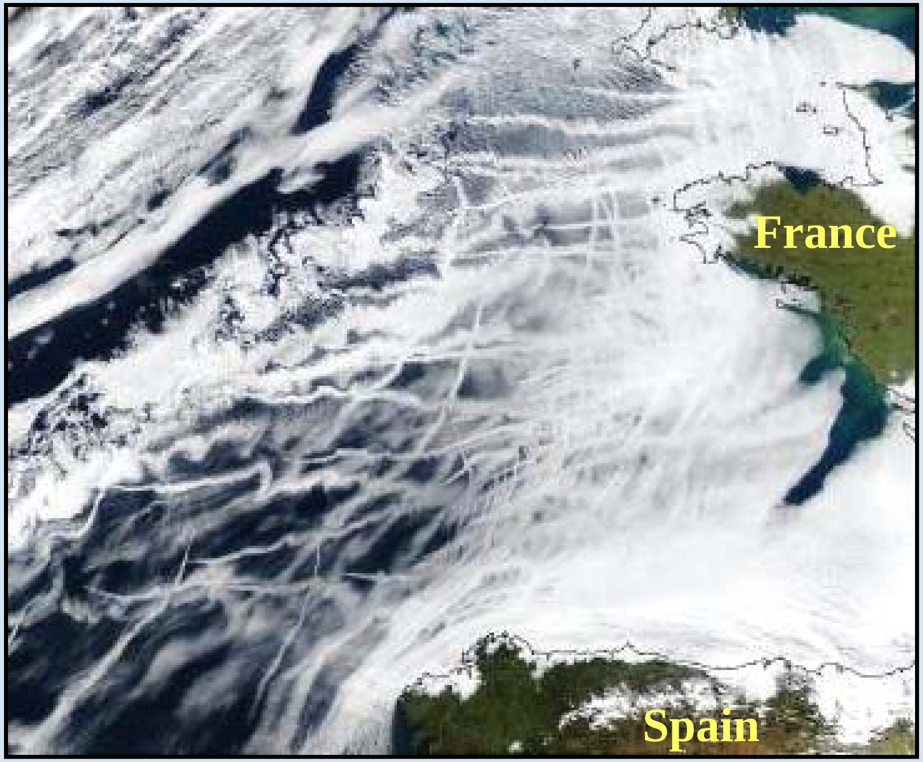
Radiative forcing by Tropospheric aerosol



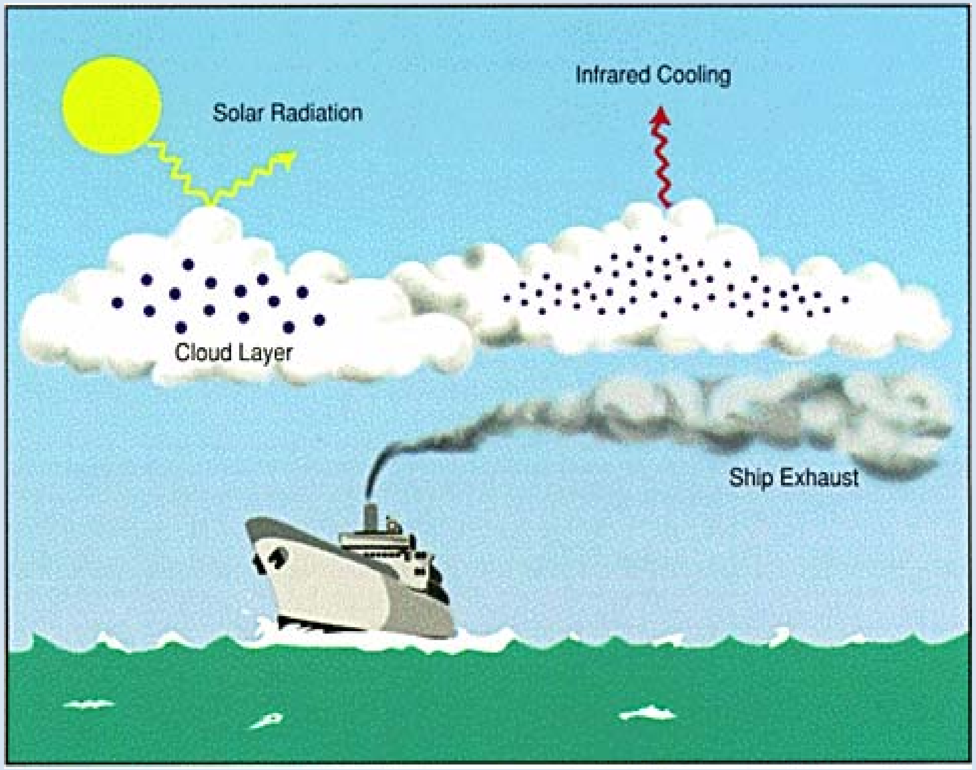
Direct Effect: Scattering and Absorption of incoming sunlight by aerosol particles

Indirect Effect: The number of CCN influences the cloud droplet size and thereby changes the cloud albedo and lifetime

Indirect effect of carbonaceous particles: Ship tracks

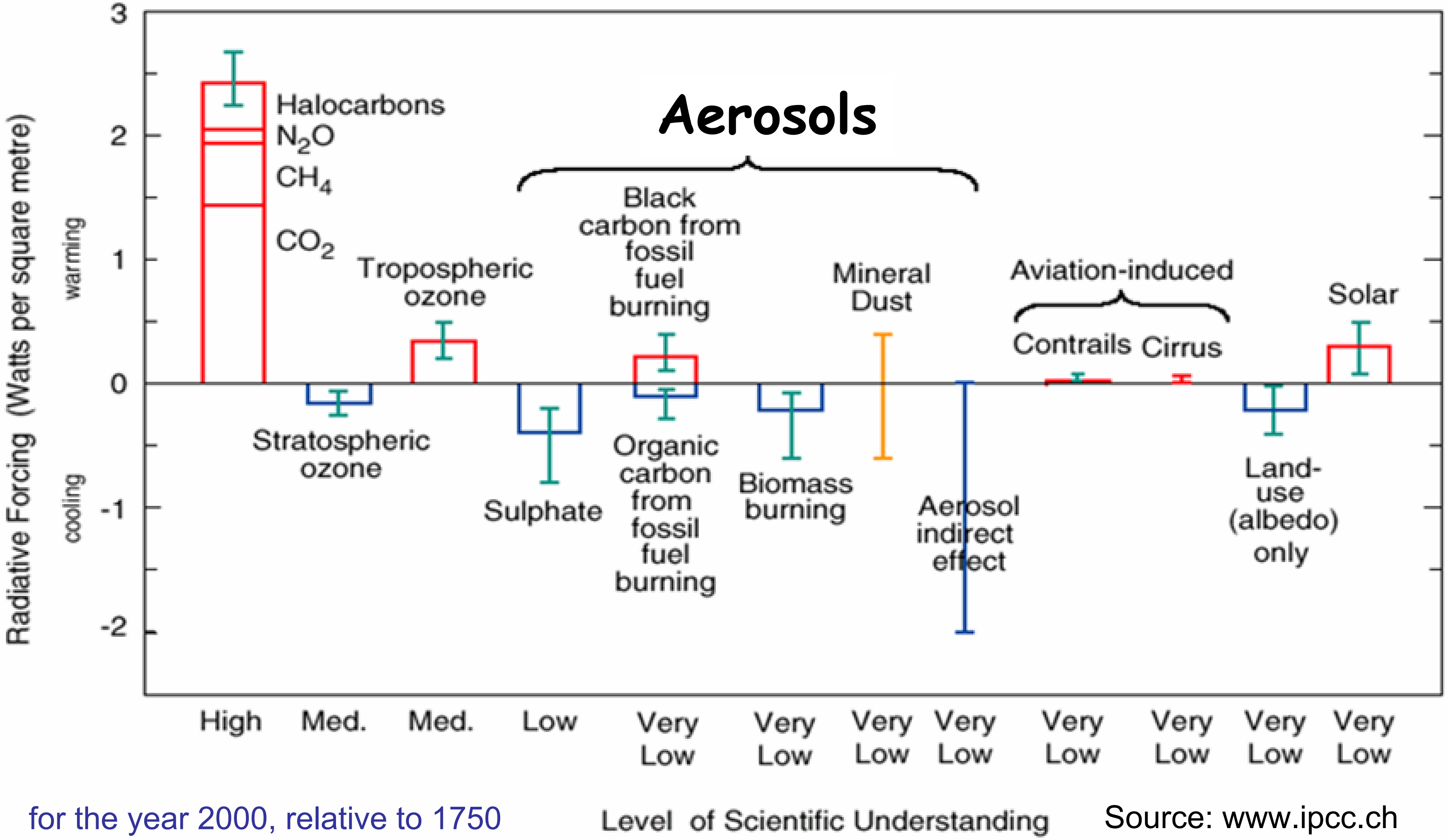


Ship tracks on the East Atlantic



Aerosol particles emitted by ships (soot particles with a high sulfur content) act as CCN and form clouds and enhance cloud reflectivity

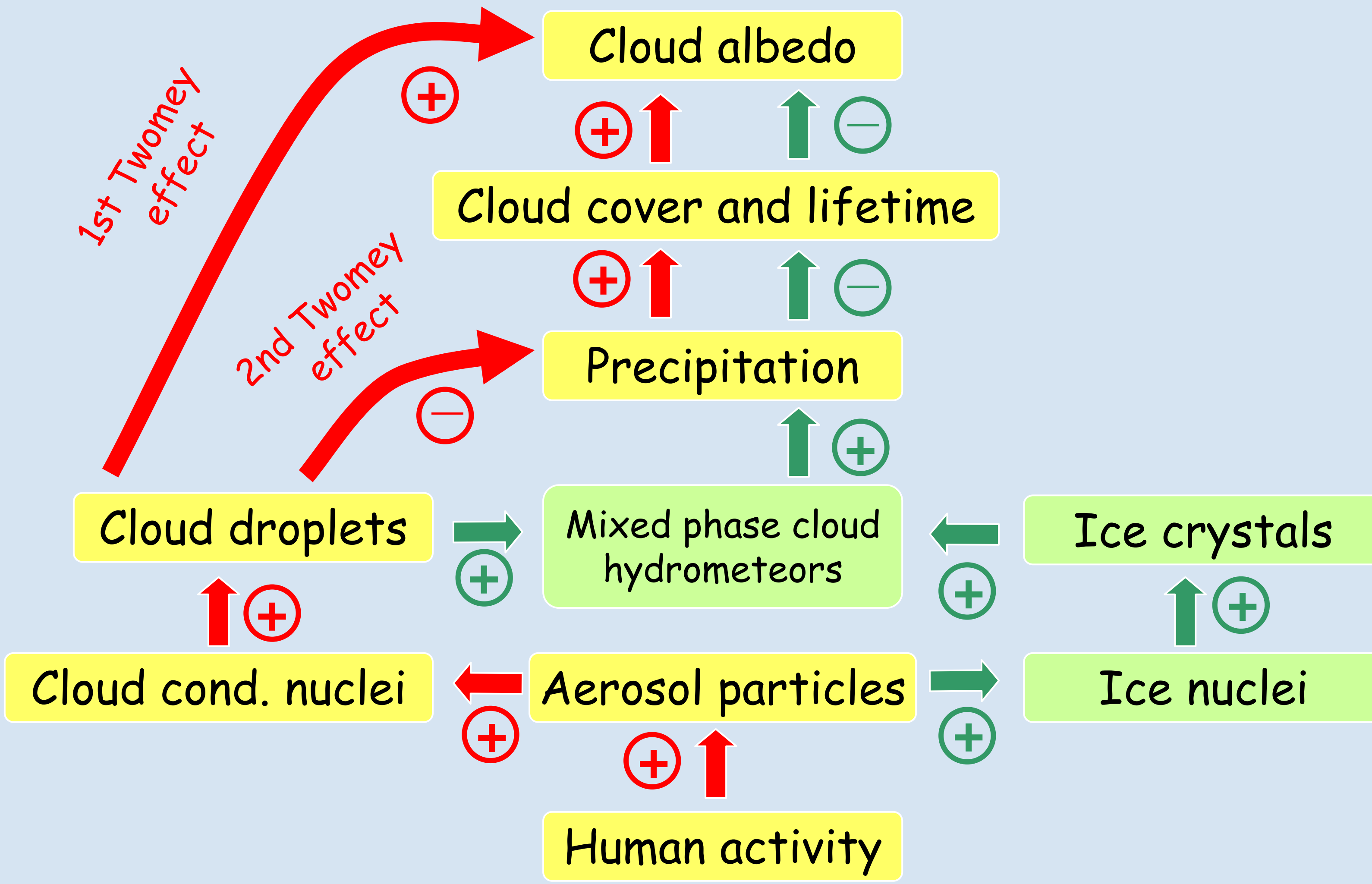
The global mean radiative forcing of the climate system



for the year 2000, relative to 1750

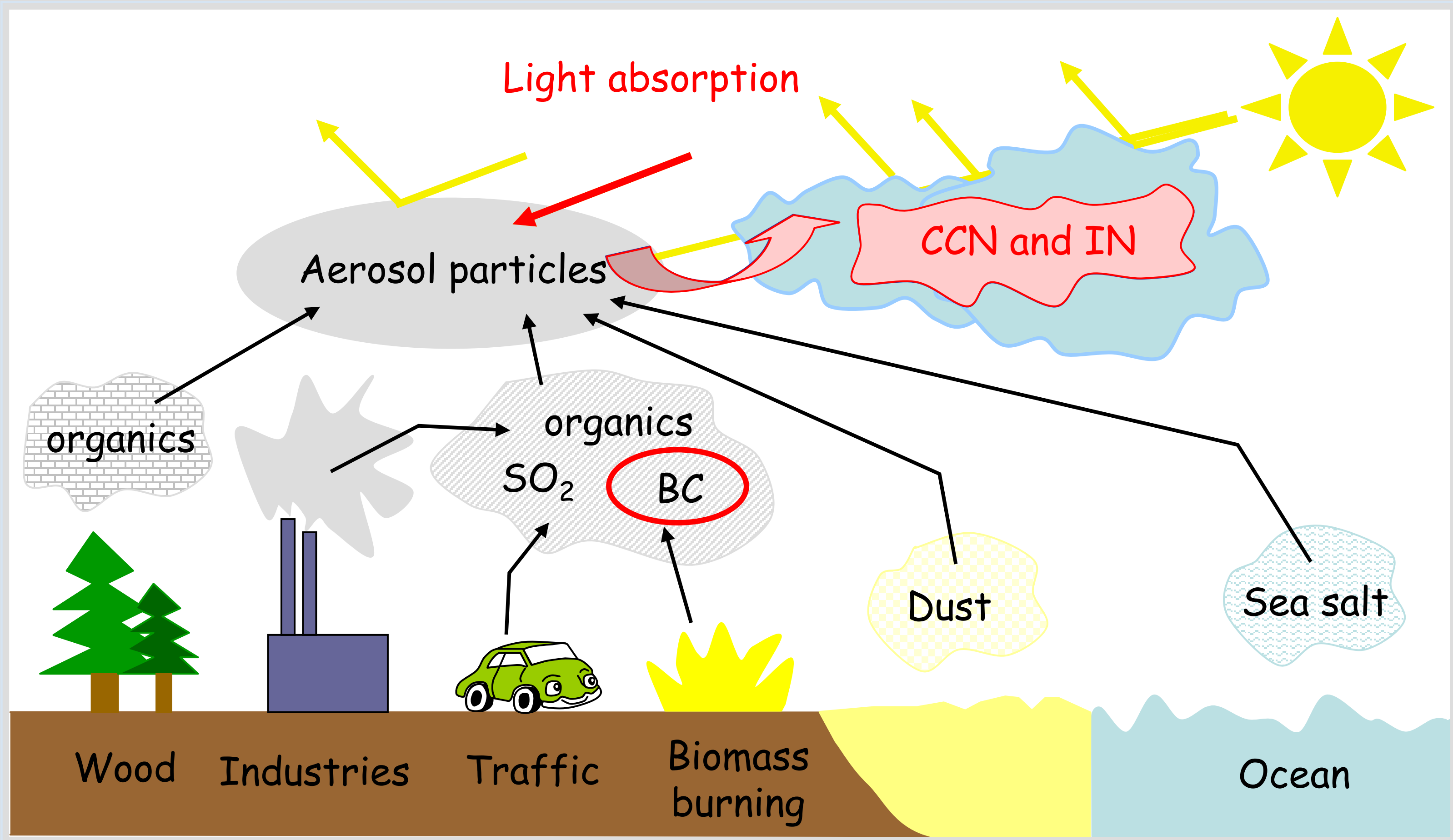
Source: www.ipcc.ch

Pathways of the Traditional *Warm Indirect Aerosol Effect* and the *Glaciation Indirect Aerosol Effect*



Lohmann, GRL, 2002

Radiative forcing by BC

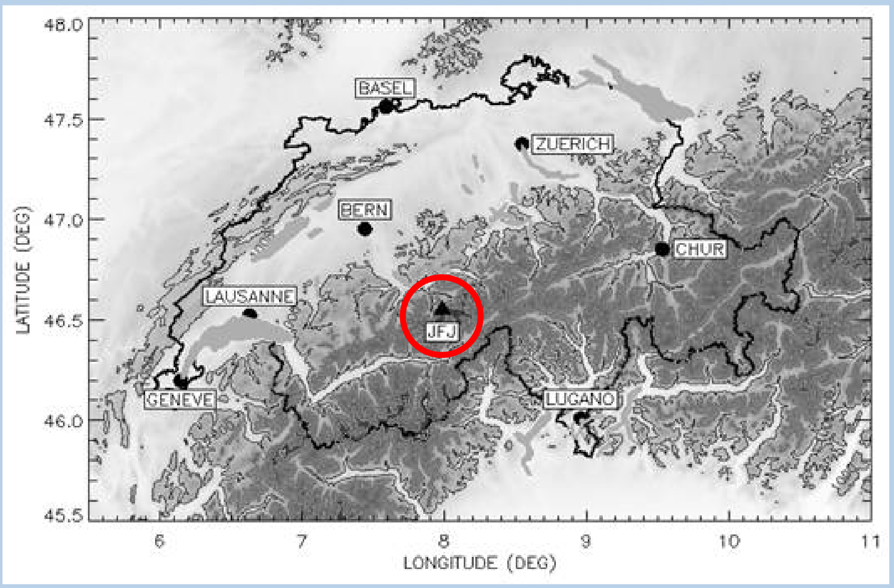


<u>Direct effect:</u> Absorption of incoming sunlight

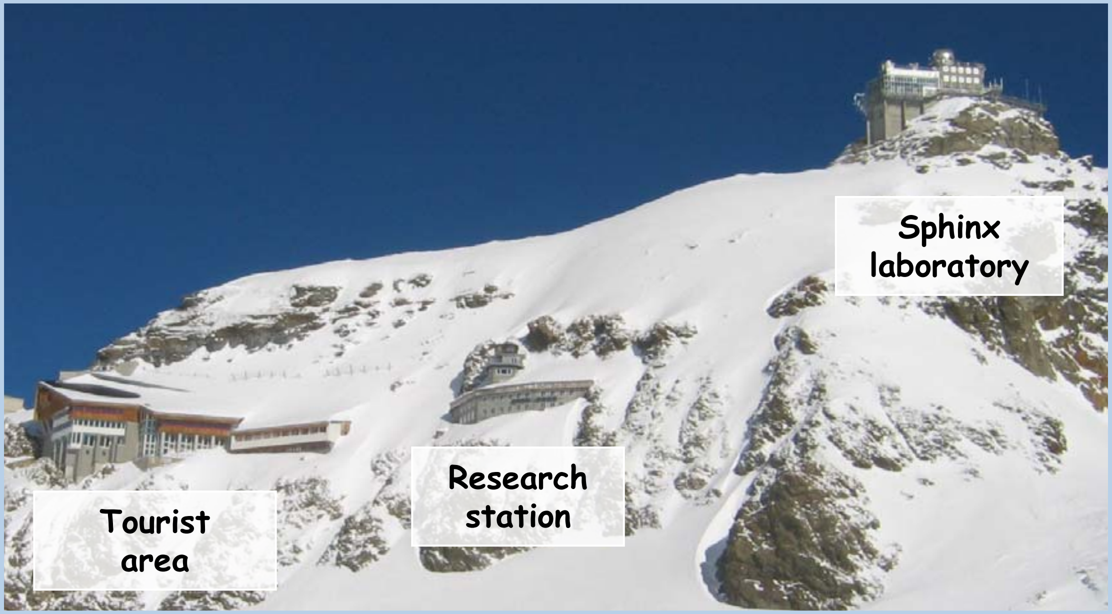
<u>Indirect effect:</u> Incorporation of BC into cloud droplets and ice crystals
(wet deposition of BC → decreasing absorption)
(modification of cloud optical properties)

<u>Semi direct effect:</u> Absorption of solar radiation by soot may cause evaporation of cloud droplets

Jungfraujoch 3580 m a.s.l.

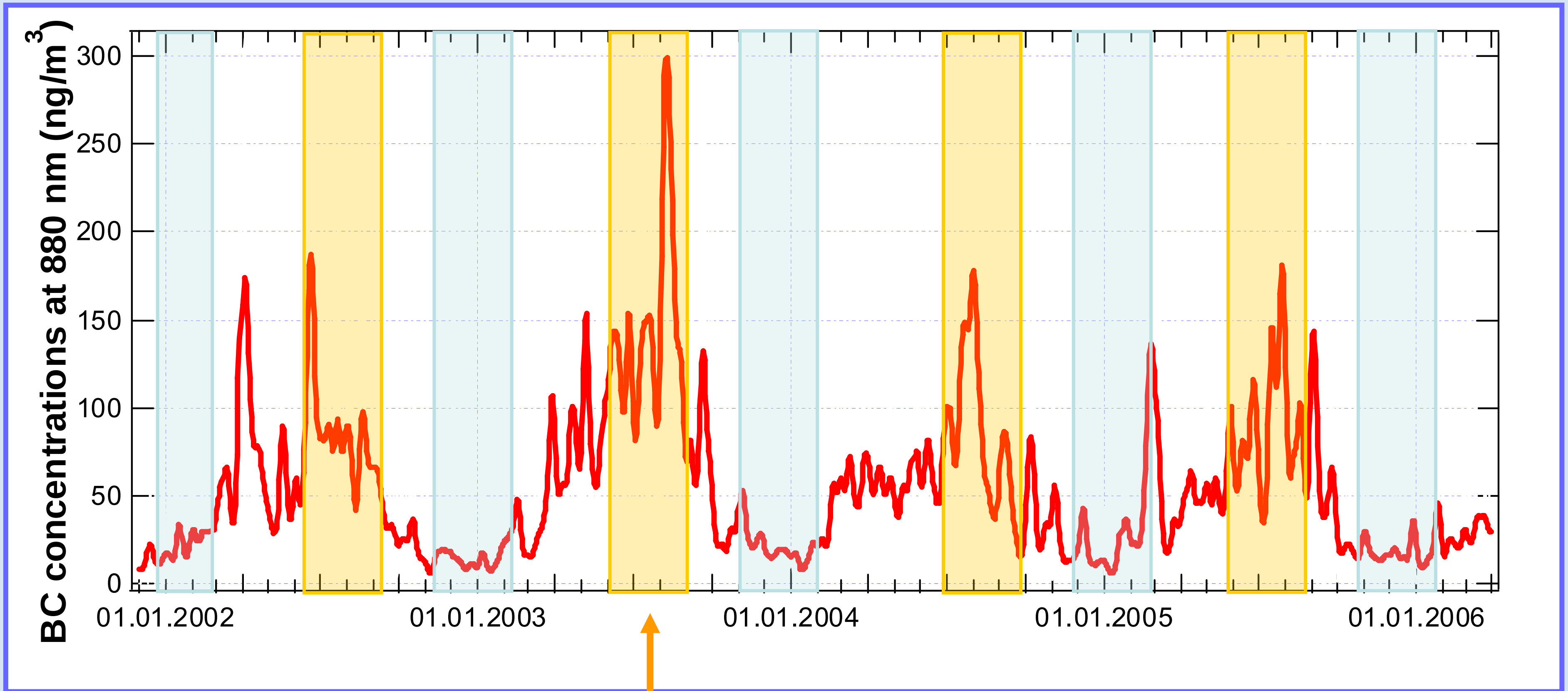


- GAW station
- Few local emissions
- Good infrastructure
- Free troposphere
- Aged aerosol
- 40% cloud occurrence



BC seasonality

Winter (November-December-January) Summer (June-July-August)



BC (Nov-Dec-Jan) = 18.9 ng/m3

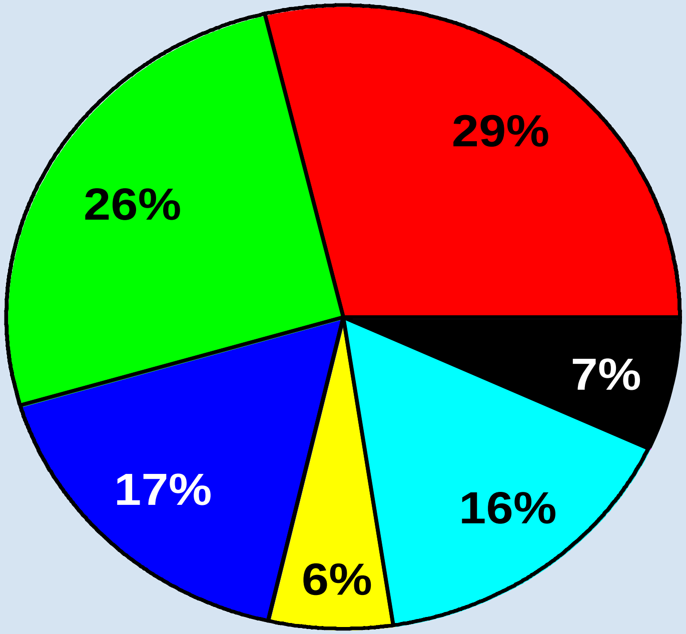
BC = 145 ng/m3

BC (Jun-Jul-Aug) = 101.7 ng/m^3

BC mass fraction in PM1

OM = 1.9 OC for summer and winter

Winter & Spring 2004 PM1



OM:
Predominantly from anthropogenic sources

SO4: Mainly anthropogenic
< 20% from natural sources

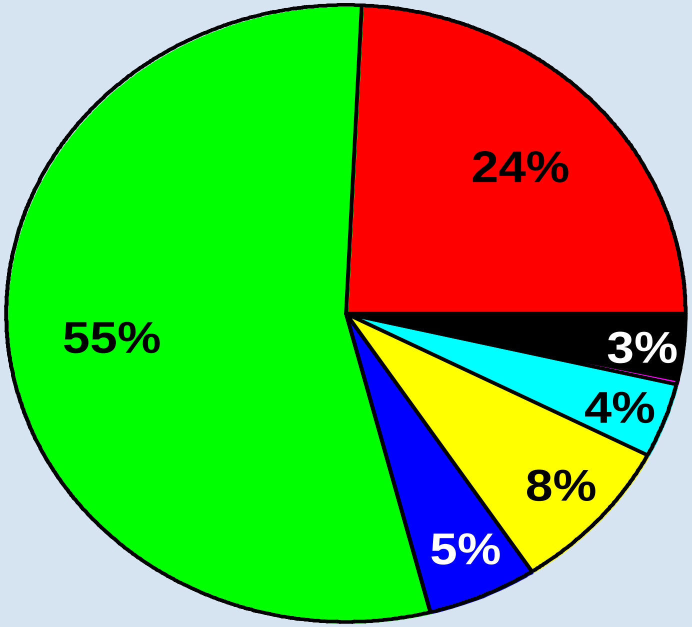
NO3: Mainly anthropogenic

NH_4: Anthropogenic

BC: Anthropogenic

None determined

Summer 2005 PM1

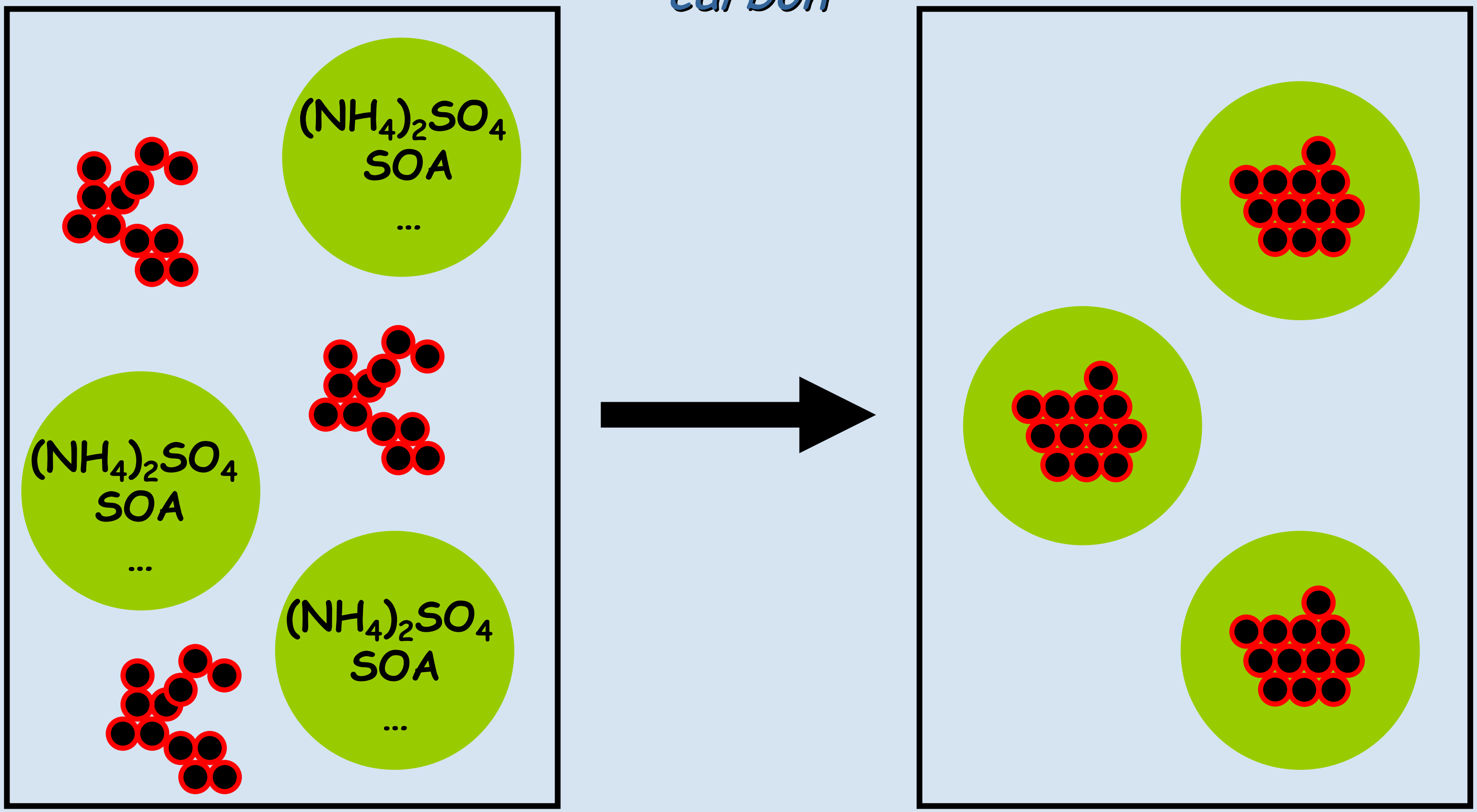


PM1 mass concentration = 1.4 µg/m3
BC mass concentration = 84 ng/m3

PM1 mass concentration = 3.4 µg/m3
BC mass concentration = 89 ng/m3

Atmospheric aging processes change the mixing state

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



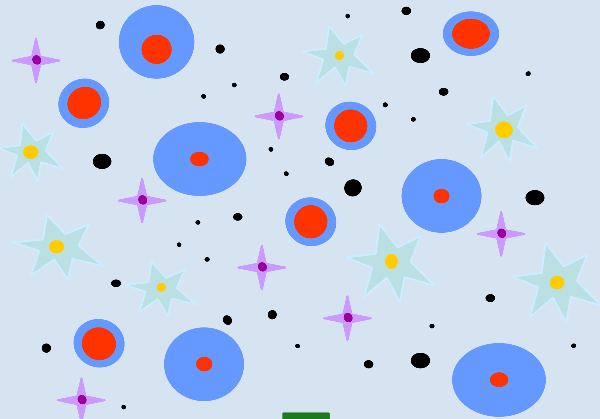
External Mixture

BC particles are separated from scattering particles

Coated Internal Mixture

BC particles are coated with scattering material

Inlets



Ice CVI inlet:

removes :

- droplets
- int. particles
- large ice crystals

(Size : 5-30 µm)

Interstitial inlet:

(no activated particles)

removes :

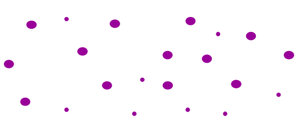
- droplets
- ice crystals

(Size < 2µm)

Total inlet :

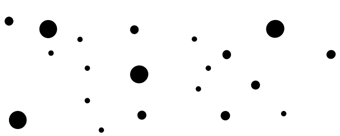
(all particles, including activated ones)

heated inlet

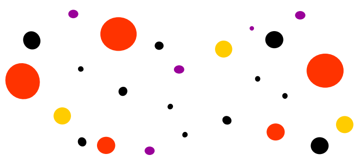


Ice residuals

Laboratory (dry aerosol)

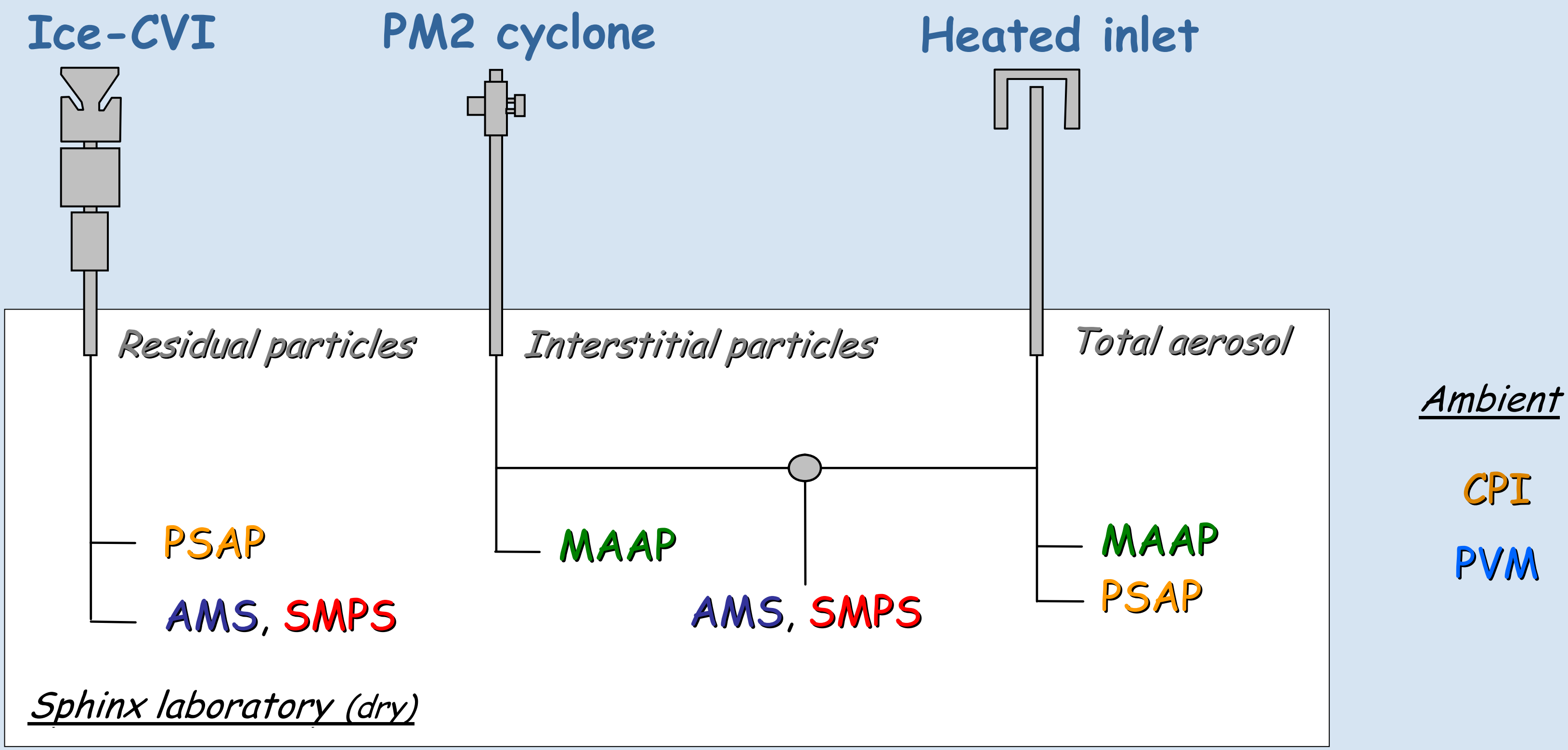


Free particles



All particles

CLACE instrumentation



BC measurements:

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

Chemical composition measurements:

- AMS = Aerosol Mass Spectrometer

Cloud microphysics:

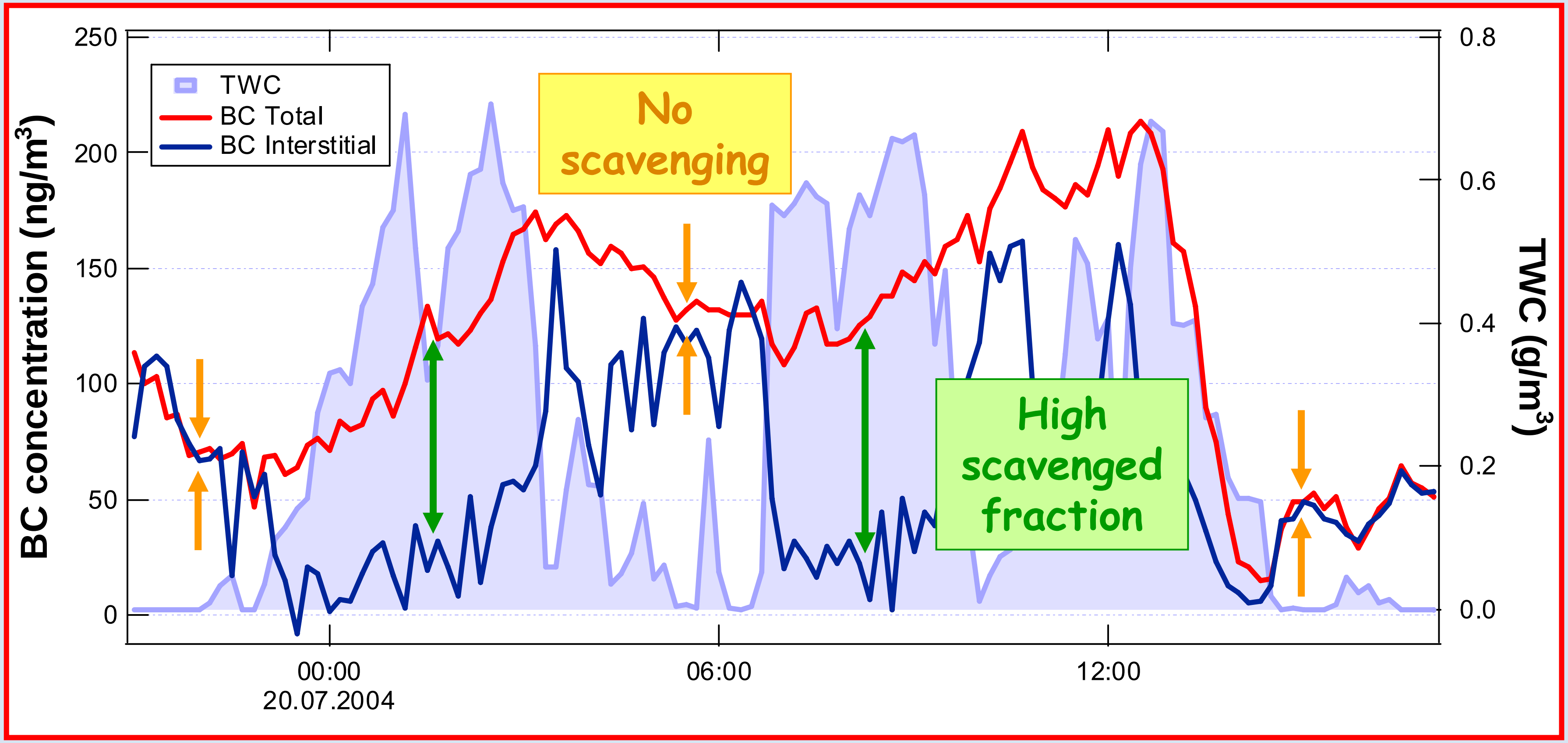
- PVM = Particulate Volume Monitor
- CPI = Cloud Particle Imager

Size distribution:

- SMPS = Scanning Mobility Particle Sizers

Scavenging of Black Carbon in liquid cloud

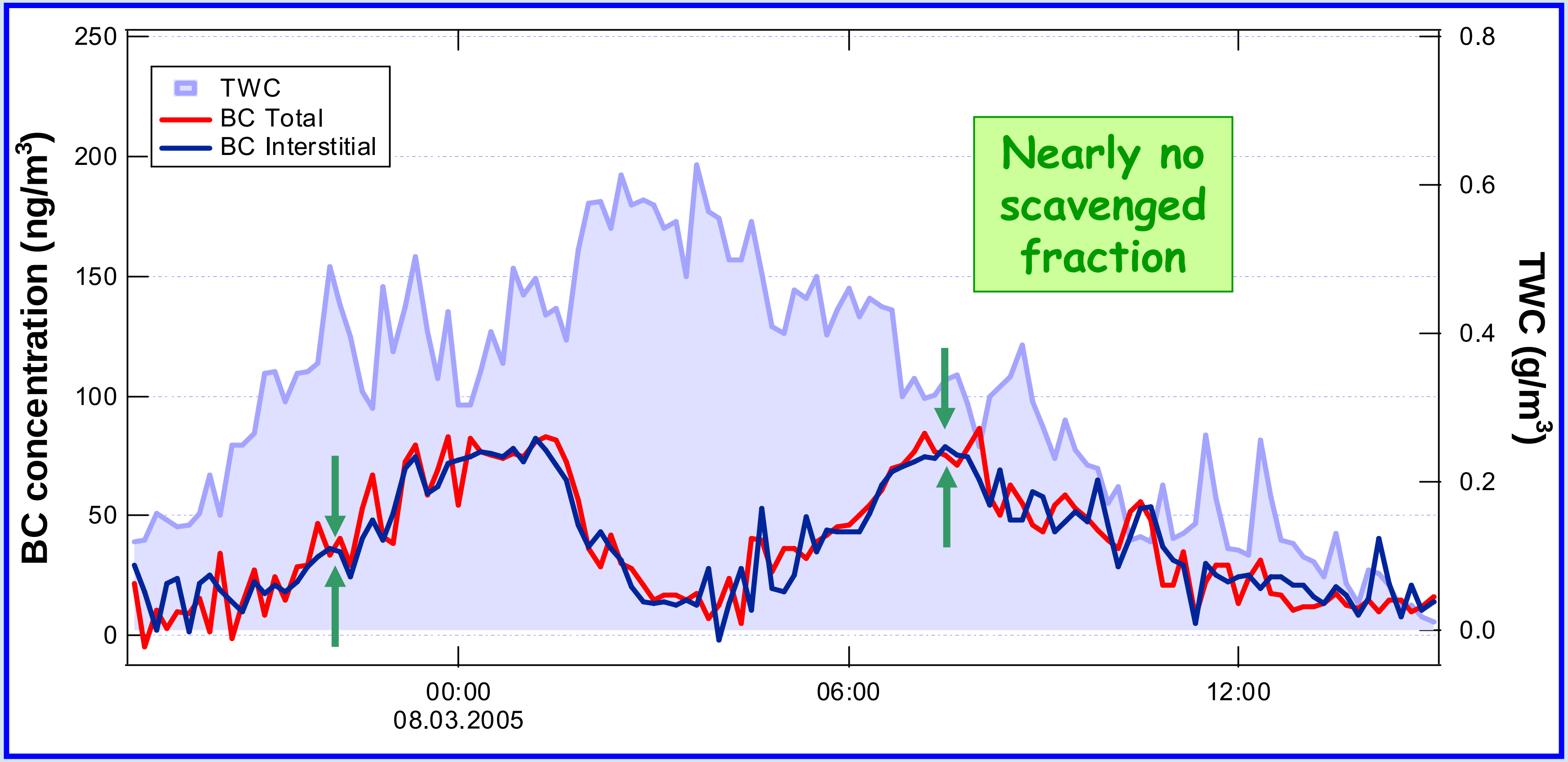
Fraction of BC aerosol that is incorporated into a cloud droplet or an ice crystal



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

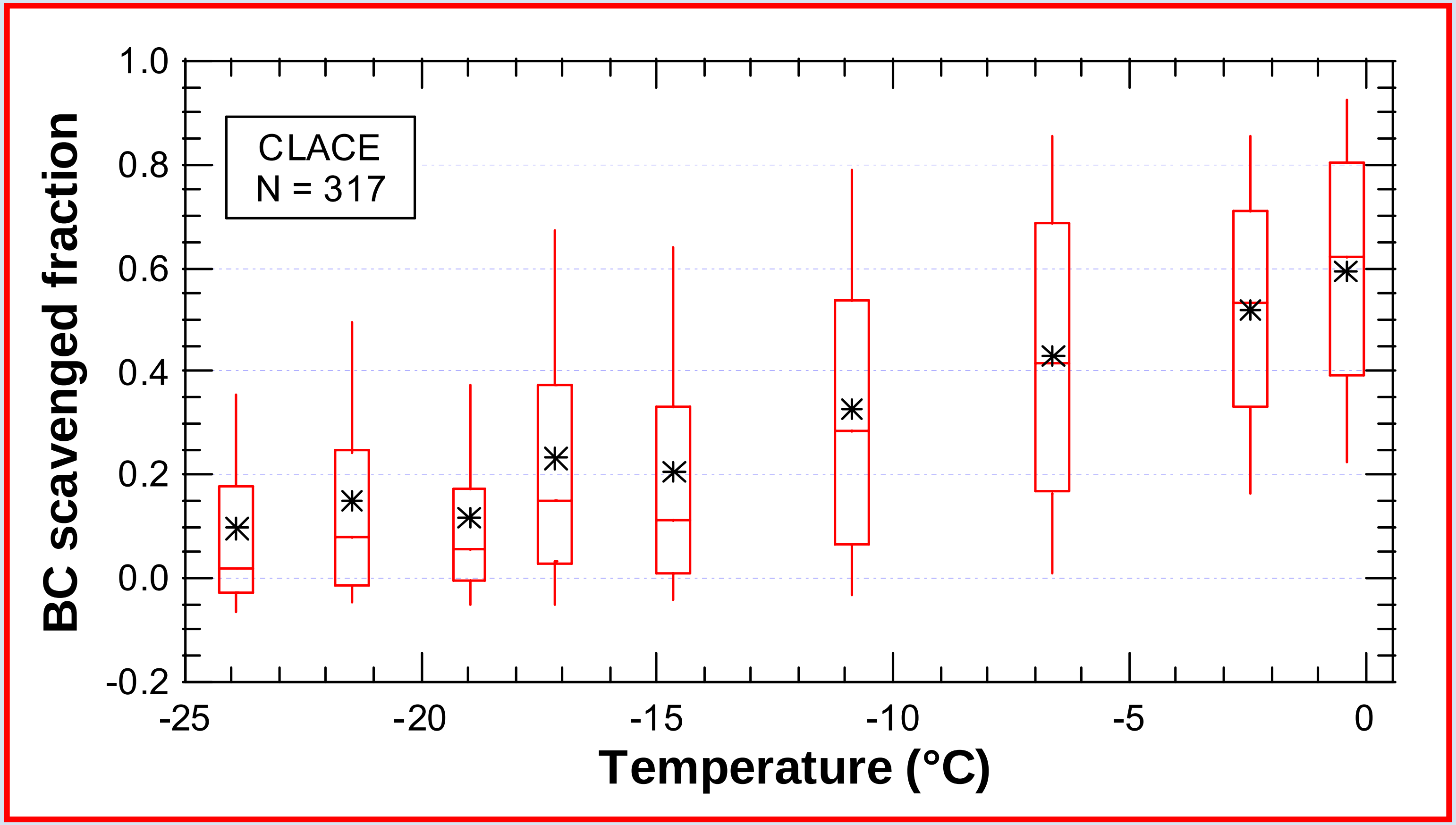
Scavenging of Black Carbon in *mixed phase cloud*

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



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

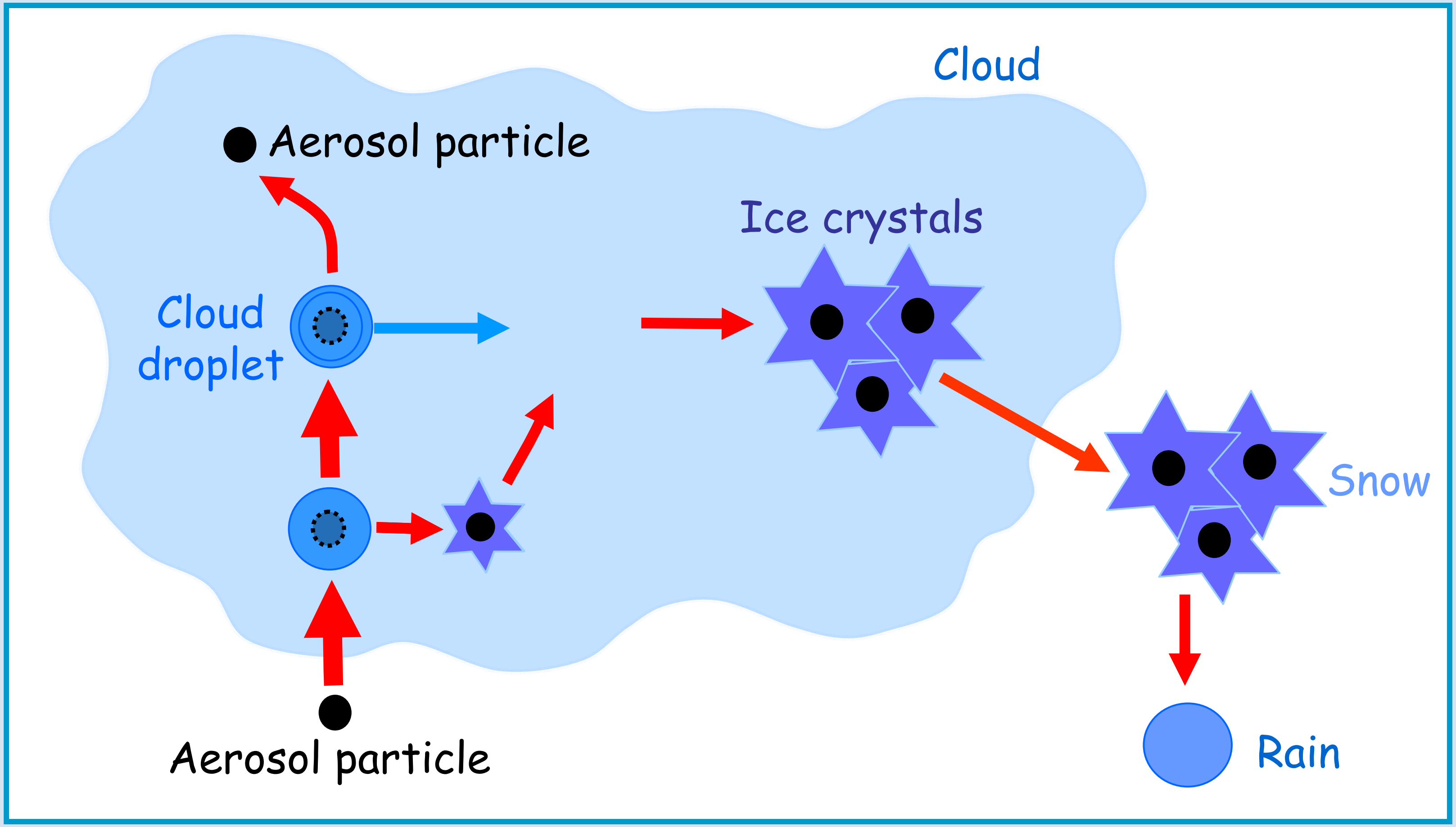
Scavenged BC fraction evolution with temperature



< -20°C: - cloud exists mainly of ice crystals (low scavenging)

> -20°C: - ↗ of liquid droplet number (↗of BC scavenging)
- BC scavenged fraction is 61% at T>-5°C

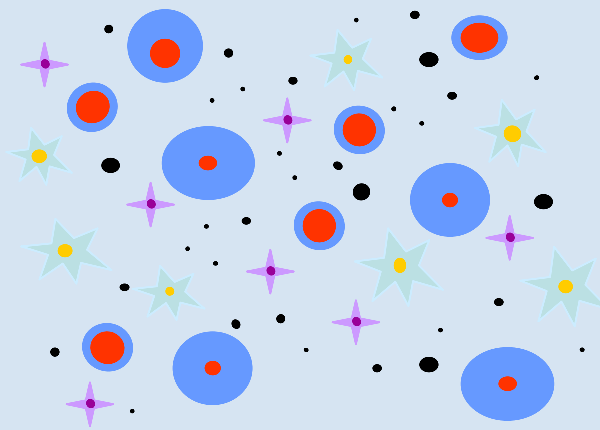
Evolution of particles in cloud : Bergeron-Findeisen process



Saturation Vapor Pressure (SVP) difference: SVP (**ice**) < SVP (**liquid**)

⇒ Flux of water vapor from liquid droplets to ice crystals

Inlets



Ice CVI inlet:

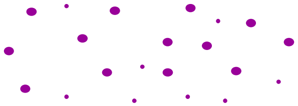
removes :
- droplets
- int. particles
- large ice crystals

(Size : 5-30 µm)

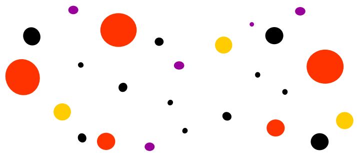
Total inlet :

(all particles, including activated ones)

heated inlet



Laboratory (dry aerosol)



Ice nuclei chemical composition

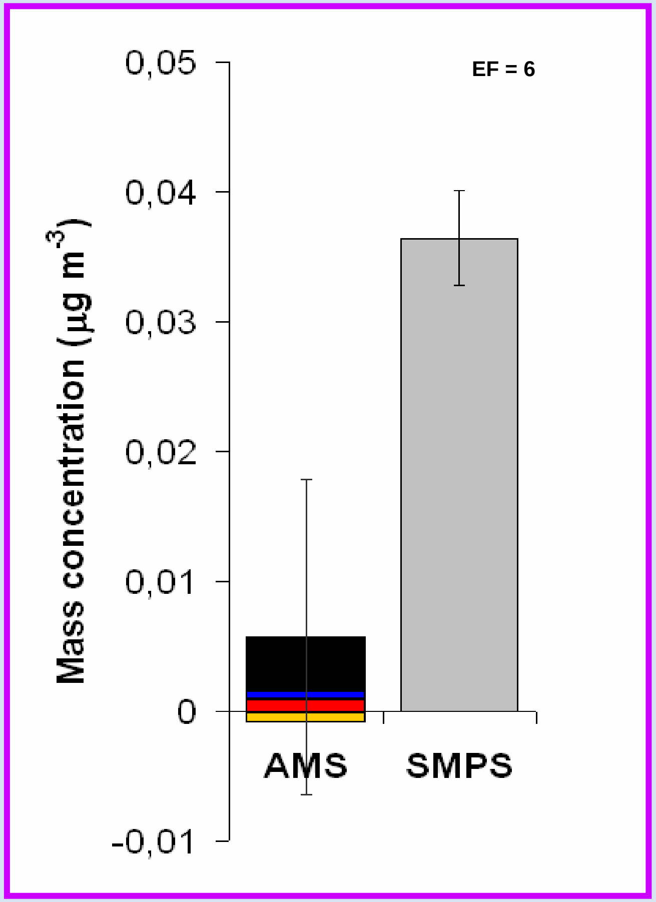
AMS data from Max-Planck Institut Mainz

MAX-PLANCK-GESELLSCHAFT

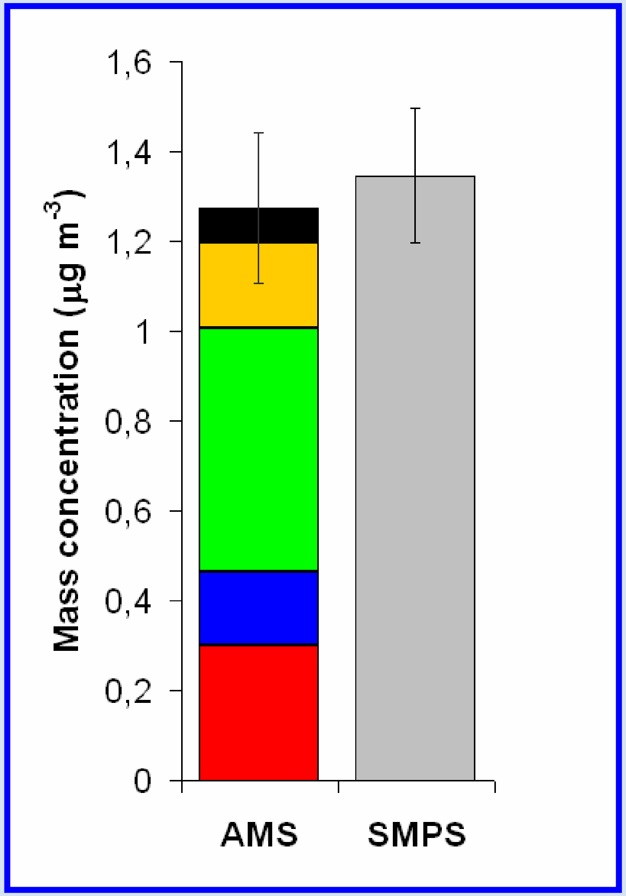
PAUL SCHERRER INSTITUT PSI

CLACE

Ice residuals



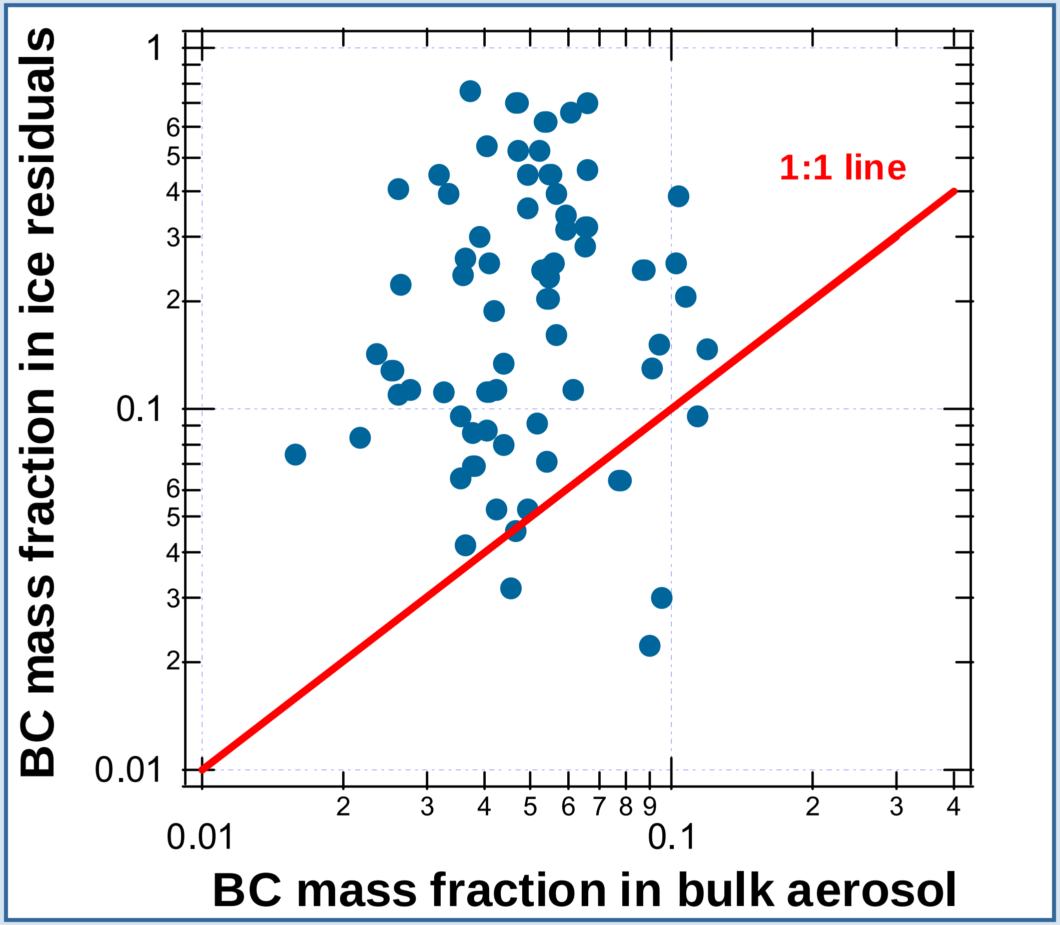
Total



Ammonium
Organics
Nitrate
Sulphate
BC

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

BC mass fraction in ice residuals and total aerosol



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

PAUL SCHERRER INSTITUT

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

Thank you for your attention

Acknowledgements:

PSI: Bart Verheggen
Ernest Weingartner
Urs Baltensperger

IFT: Stephan Mertes

MPI Mainz: Johannes Schneider
Saskia Walter

PAUL SCHERRER INSTITUT PSI

CLACE

LABORATORY OF ATMOSPHERIC CHEMISTRY