Aerosol light scattering at high relative humidity

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Atmospheric aerosols influence the Earth radiation budget both directly and indirectly. Particles with diameters between 0.1 and 1 µm are very efficient in scattering and, depending on their chemical composition, absorbing solar radiation. This is referred to as the direct aerosol effect. The direct effect can result in a negative or positive radiative forcing, depending on the chemical and microphysical properties of the involved aerosols and the surface albedo. The anthropogenic impact of the direct effect is estimated to be -0.5 Wm^{-2}, compared to the total anthropogenic radiative forcing of 1.6 Wm^{-2} (IPCC 2007). Aerosols can additionally act as cloud condensation nuclei and thus influence the radiative properties and the lifetime of clouds. The anthropogenic radiative forcing of these indirect effects results is -0.7 Wm^{-2}. Aerosols are still a great source of uncertainty in the estimation of the total anthropogenic influence on climate (Ramanathan, Crutzen et al. 2001; IPCC 2007).
An important parameter in the context of radiative forcing is the single scattering albedo (SSA) (Haywood and Shine 1995), which is the ratio of the scattering to the sum of absorption and scattering. The single scattering albedo of Black Carbon at visible wavelength is about 0.2, whereas for sulfate aerosols it is close to 1 (Haywood and Boucher 2000; IPCC 2001).
SSA can be determined by measurements of absorption and scattering coefficients. A convenient instrument to measure scattering is an integrating nephelometer. When ambient air enters into a housing its temperature (T) and relative humidity (RH) may change, and, this being the case, the measured aerosol properties will differ from the ambient - the climate relevant - ones. In order to ensure comparability between different locations the Global Atmosphere Watch aerosol monitoring network recommends measuring aerosol light scattering coefficients σ_s at RH below 40% (WMO/GAW 2003). The scattering coefficient depends on the particles chemical composition and much stronger on their size distribution. With increasing RH the particles take up water and increase their size (hygroscopic growth). Consequently for a correct estimation of the direct climate forcing by aerosol particles the influence of the RH on the scattering is needed (Charlson, Schwartz et al. 1992; Schwartz 1996). Both model calculations and measurements have shown that the aerosol light scattering is strongly dependent on RH (Nessler, Weingartner et al. 2005).

We built a humidification system for a nephelometer that allows for measurement of σ_s at a defined and controlled humidity in the range from 20 to 90%. The system is able to measure hygroscopic aerosol behavior and can be continuously operated.

Monodisperse ammonium sulfate and sodium chloride were measured at four different particle diameters (100 nm, 150 nm, 240 nm and 300 nm). Good agreement between measurement and calculations based on Mie theory were found for both σ_s and $f(RH)=\sigma_s(RH)/\sigma_s(dry)$ within the range of uncertainty.

In order to compare our humidified nephelometer to the one of NOAA it was running at a rural site in the Black Forest (Germany) from August 9-31, 2007. NOAA measured light scattering coefficients (σ_s) in a dry and humidified nephelometer plus other aerosol parameters in the DOE/ ARM (US Department of Energy Atmospheric Radiation Measurements) Mobile Facility. The two humidified nephelometers often detected different f(RH). One hypothesis for the difference in the f(RH) of the two instruments is that they probe the two different branches of a hysteresis curve, since the RH history in the two instruments is not the same.
We could prove that our humidified nephelometer agrees well with the instrument of NOAA and even provides more information about the measured aerosol. It is important to measure the dehydration branch since atmospheric aerosols are usually in this phase. For a reduction of the large uncertainties in the radiative forcing due to the direct aerosol effect the knowledge of ambient scattering coefficients adds one piece to the big puzzle.

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Haywood, J. M. and K. P. Shine (1995). "The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget." Geophysical Research Letters **22**(5): 603-606.

IPCC (2001). Climate Change 2001: The Scientific Basis. New York, Cambridge Univ. Press.

IPCC (2007). Climate Change 2007 - The Physical Science Basis. New York, Cambridge Univ. Press.

Nessler, R., E. Weingartner, et al. (2005). "Adaptation of Dry Nephelometer Measurements to Ambient Conditions at the Jungfraujoch." Environmental Science & Technology **39**: 2219-2228.

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Schwartz, S. E. (1996). "The whitehouse effect - shortwave radiative forcing of climate by anthropogenic aerosols: an overview " Journal of Aerosol Science **27**(3): 359-382.

WMO/GAW (2003). Aerosol Measurement Procedures Guidlines and Recommendations. W. M. O. G. A. Watch, WMO. **153**.

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Aerosol Light Scattering at High Relative Humidity

12th ETH Conference on Combustion Generated Nanoparticles

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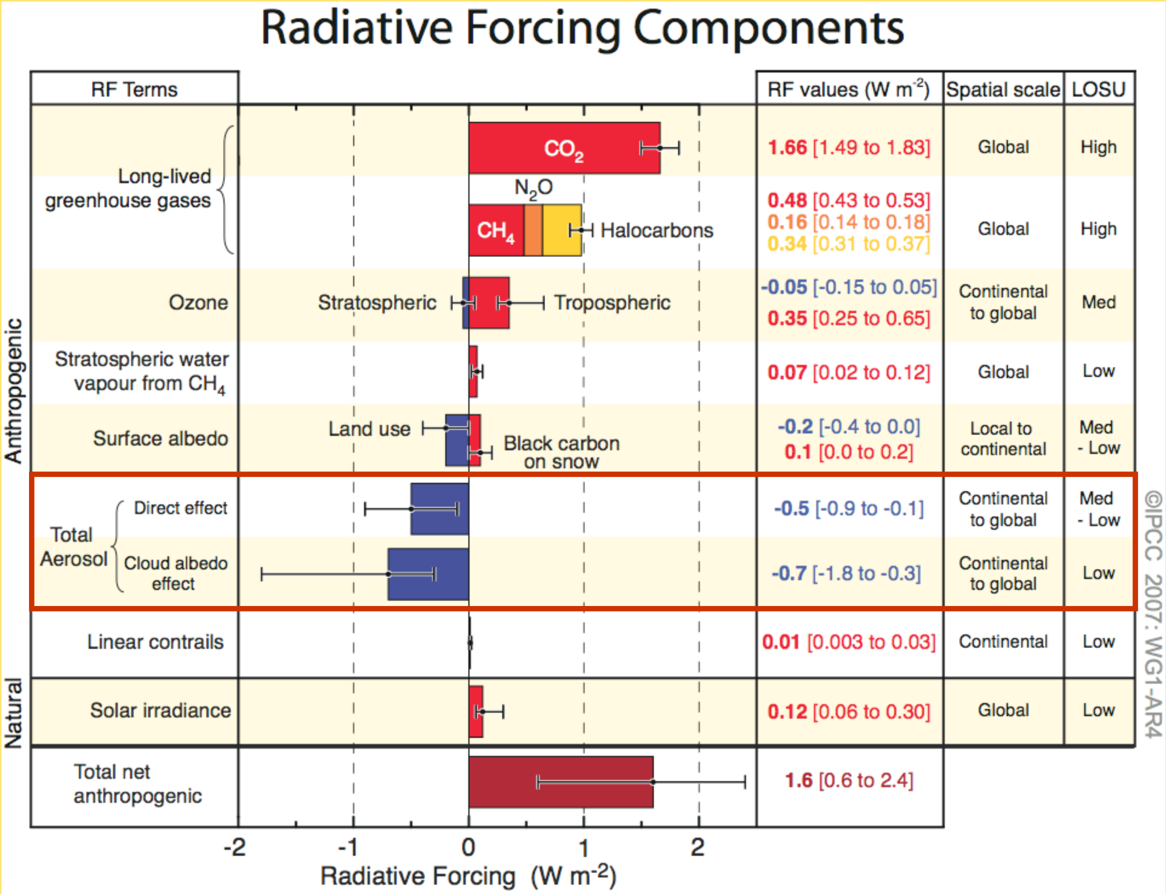
Outline of this talk

Aerosol light scattering at high relative humidity

- Importance of light scattering
- Importance of relative humidity
- Measurements

Why is scattering of aerosols important?

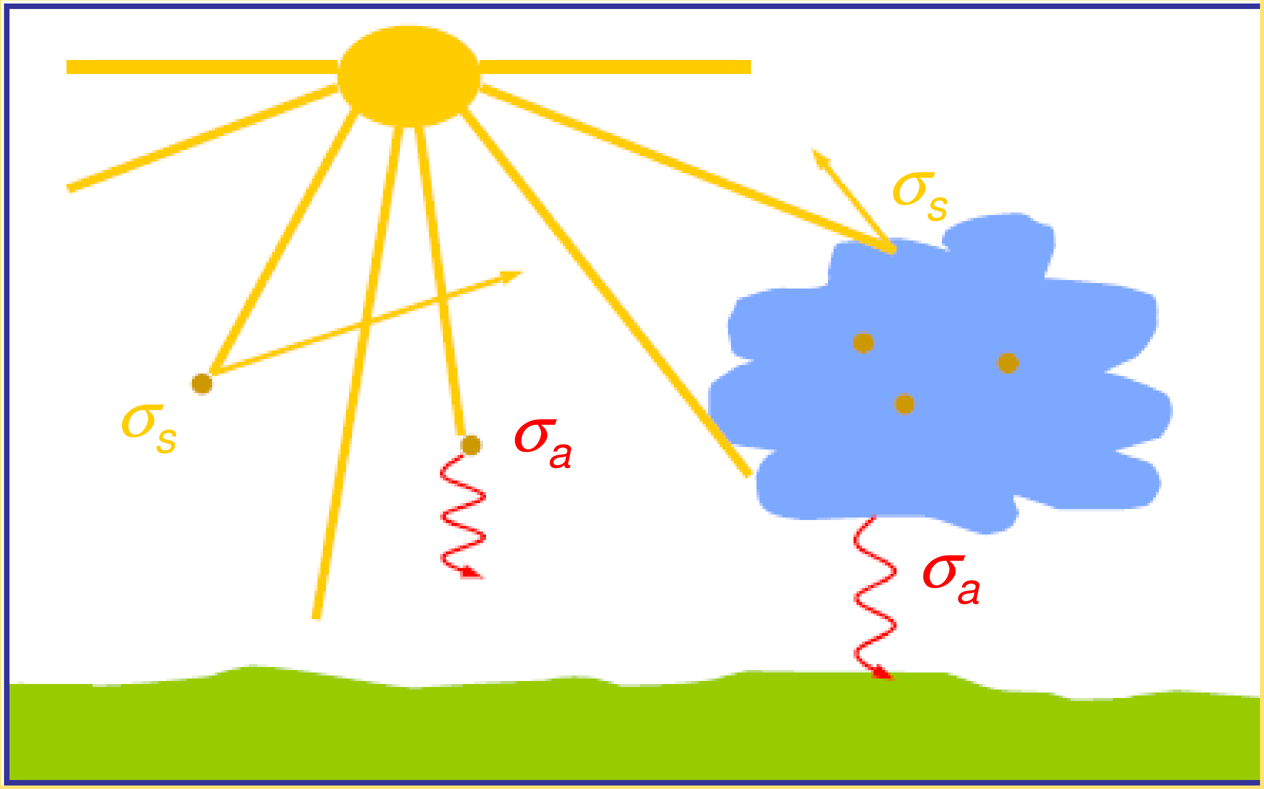
Global mean radiative forcing for the year 2000, relative to 1750



Source: IPCC, 2007

Why is scattering of aerosols important?

Direct and indirect - or cloud albedo - effect



scattering coefficient: σ_s, absorption coefficient: σ_a

Why is scattering of aerosols important?

Scattering + absorption = extinction coefficient

$$\sigma_s + \sigma_a = \sigma_{ext}$$

Beer-Lambert‘s Law:

$$I = I_0 \cdot e^{-\sigma_{ext} \cdot x}$$

I_0: Intensity of incident light

I: Intensity of remaining light

x: thickness of medium

Single Scattering Albedo SSA:

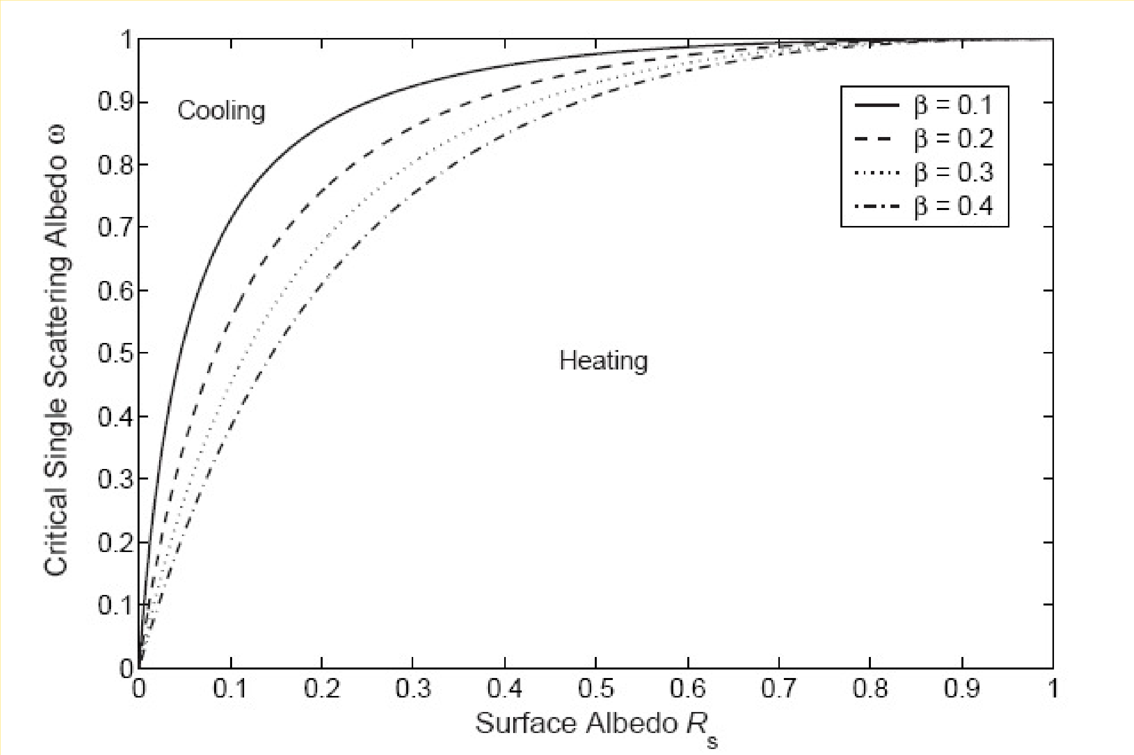
$$SSA = \frac{\sigma_s}{\sigma_s + \sigma_a}$$

In gereral: just scattering: 1
just absorbing: 0

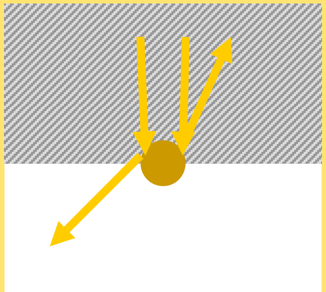
Soot: ~ 0.3
Aged soot: ~ 0.5
Mineral dust: ~ 0.8
NaCl: 1

Why is the single scattering albedo important?

$$SSA = \frac{\sigma_s}{\sigma_s + \sigma_a}$$



β: upscatter-fraction

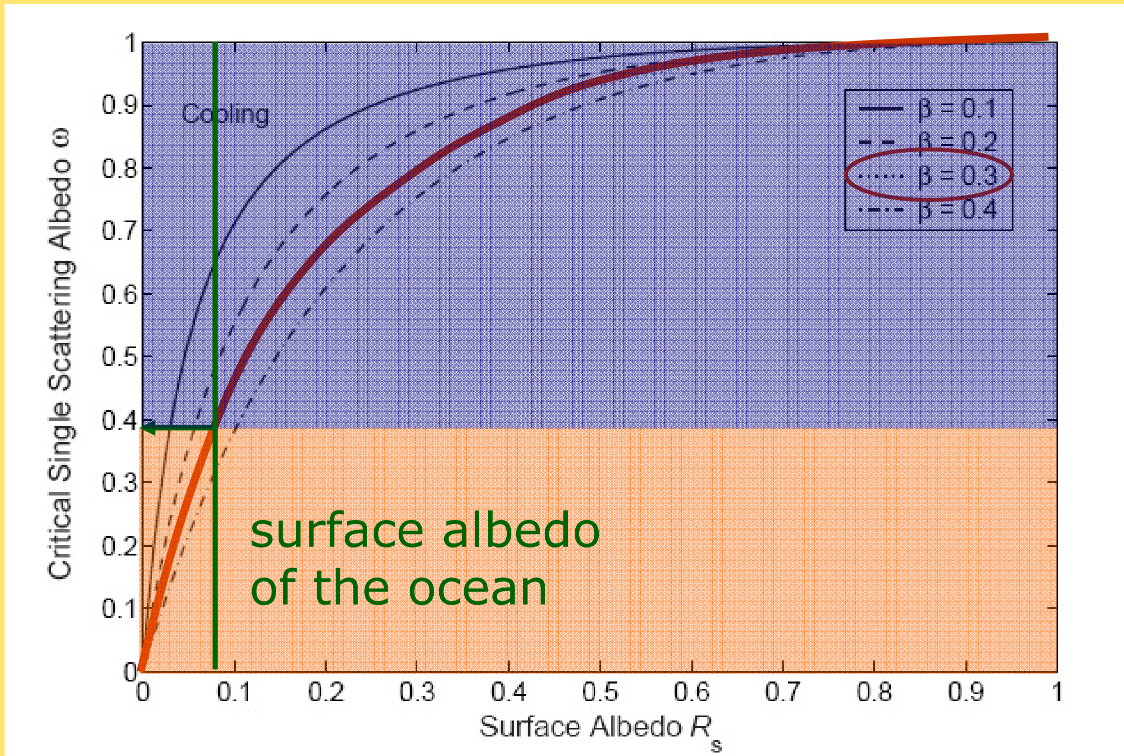


Source:
Seinfeld+Pandis, 1998

Surface albedo: the fraction of solar radiation reflected by a surface

Why is the single scattering albedo important?

$$SSA = \frac{\sigma_s}{\sigma_s + \sigma_a}$$



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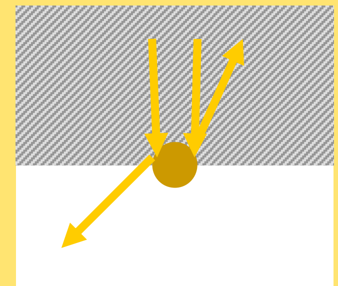

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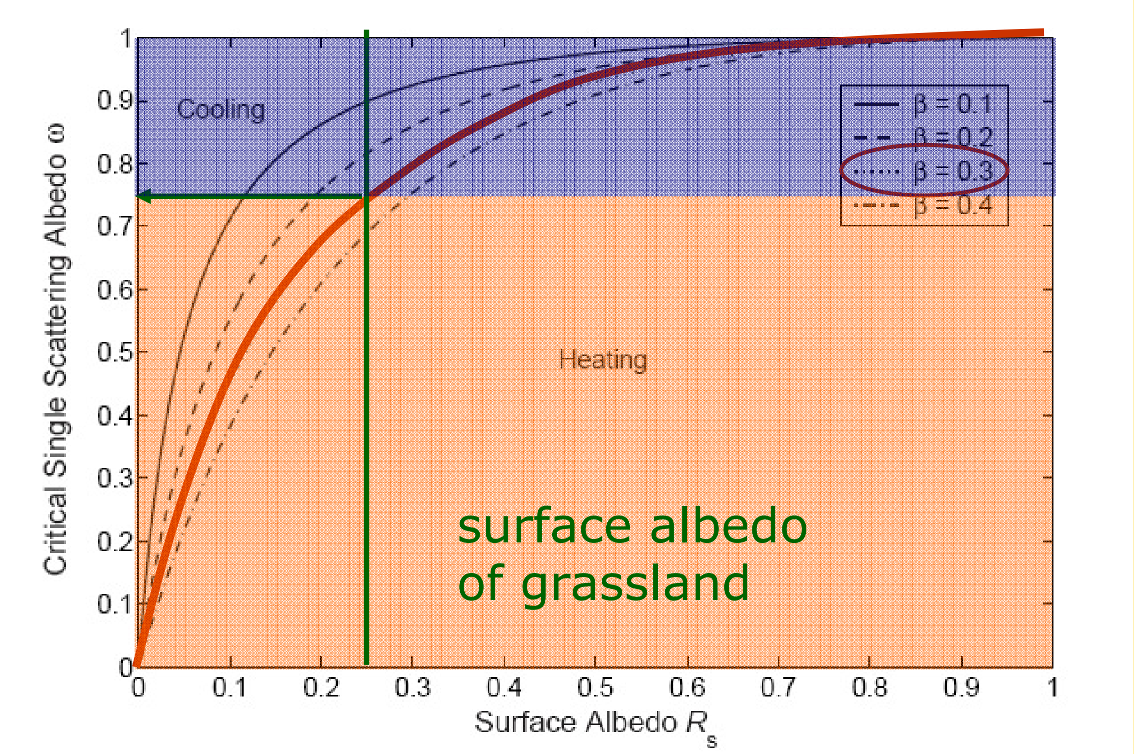

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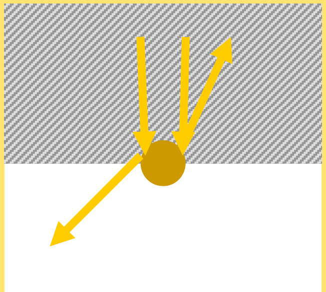

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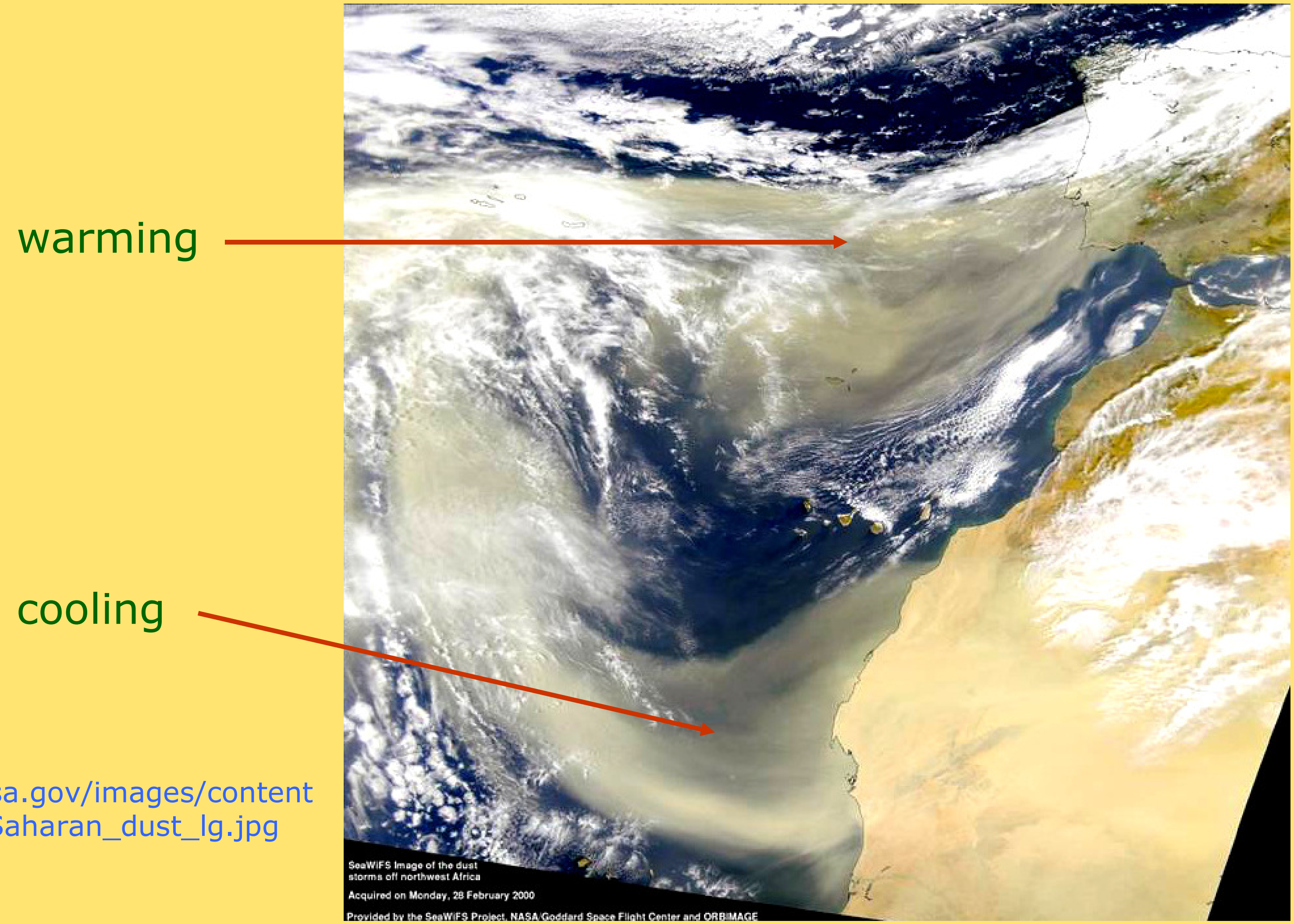
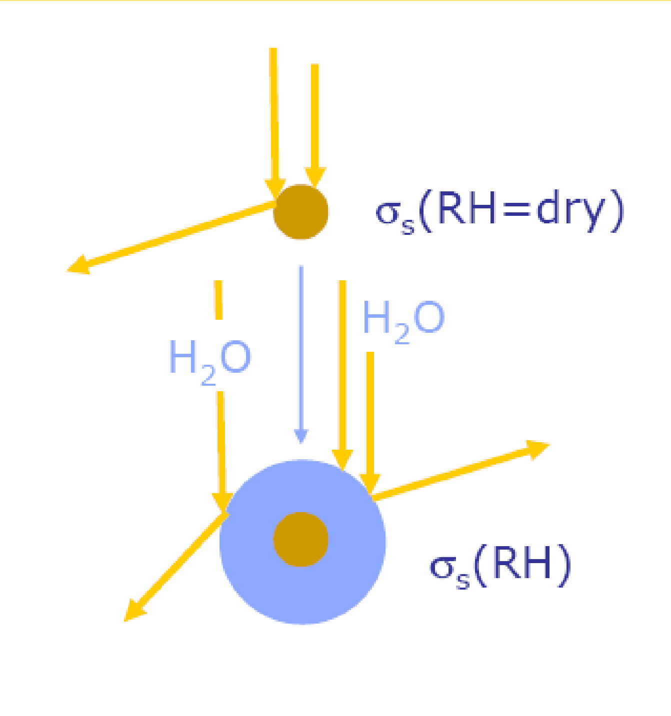


Source:
http://www.nasa.gov/images/content/105016main_Saharan_dust_lg.jpg

Why is the relative humidity important?



- Some particles grow in high relative humidity
- Scattering increases
- Light scattering enhancement factor f(RH):

$$f(RH) = \frac{\sigma_s(RH)}{\sigma_s(RH = dry)}$$

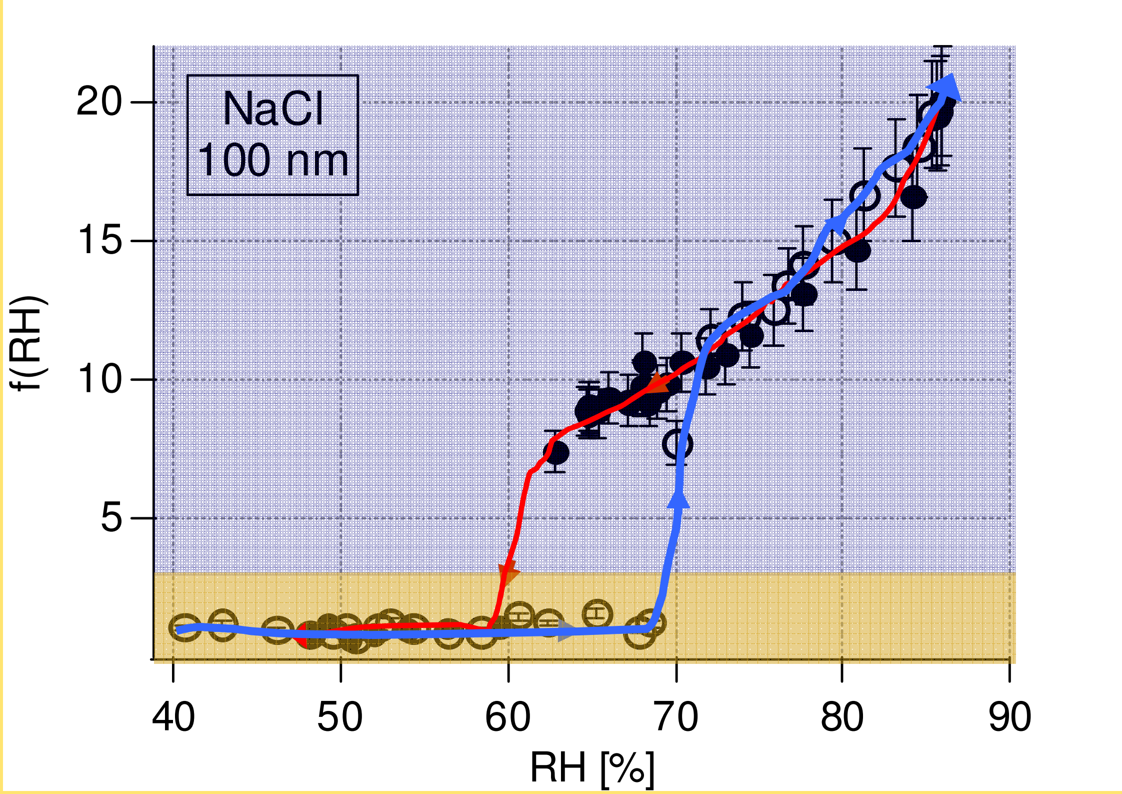
- f(RH) can be measured by PSI's humidified nephelometer

- Assumption: f(RH=80%)= 1.7

  → SSA increases by 10% (from 0.8 to 0.87)

Light scattering enhancement factor vs. RH

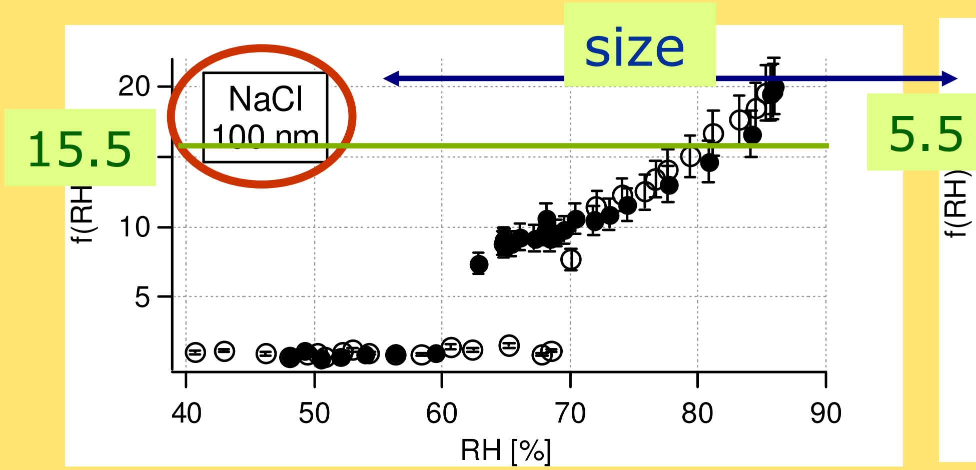
- Hydration: RH from dry to humid ○
- Dehydration: RH from humid to dry ●

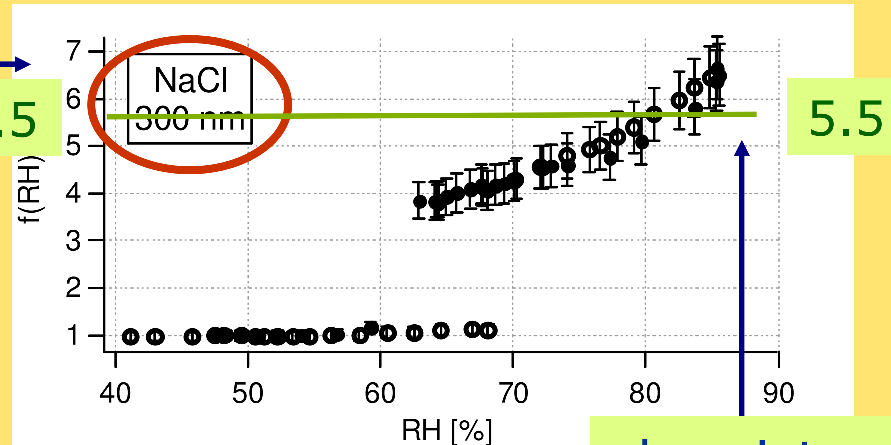


$$f(RH)=\frac{\sigma_s(RH)}{\sigma_s(RH=dry)}$$

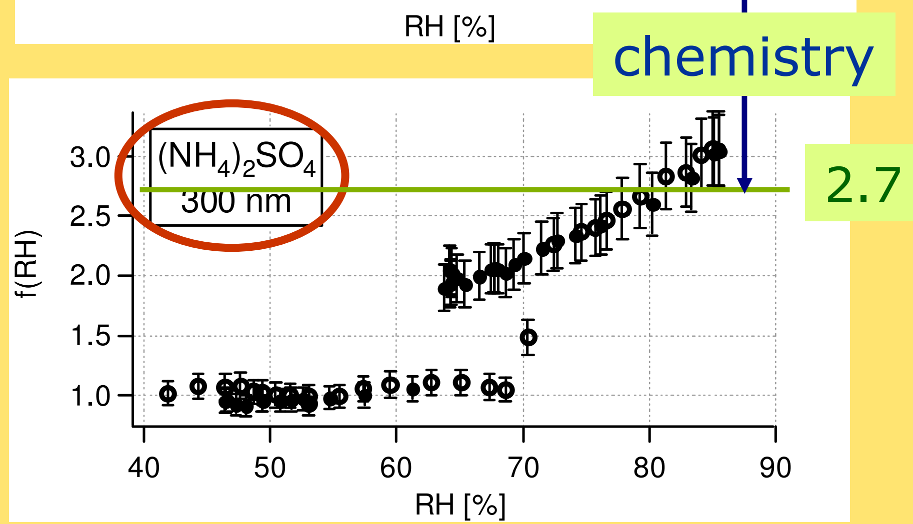
- Salts often experience phase transitions

What influences enhanced light scattering?





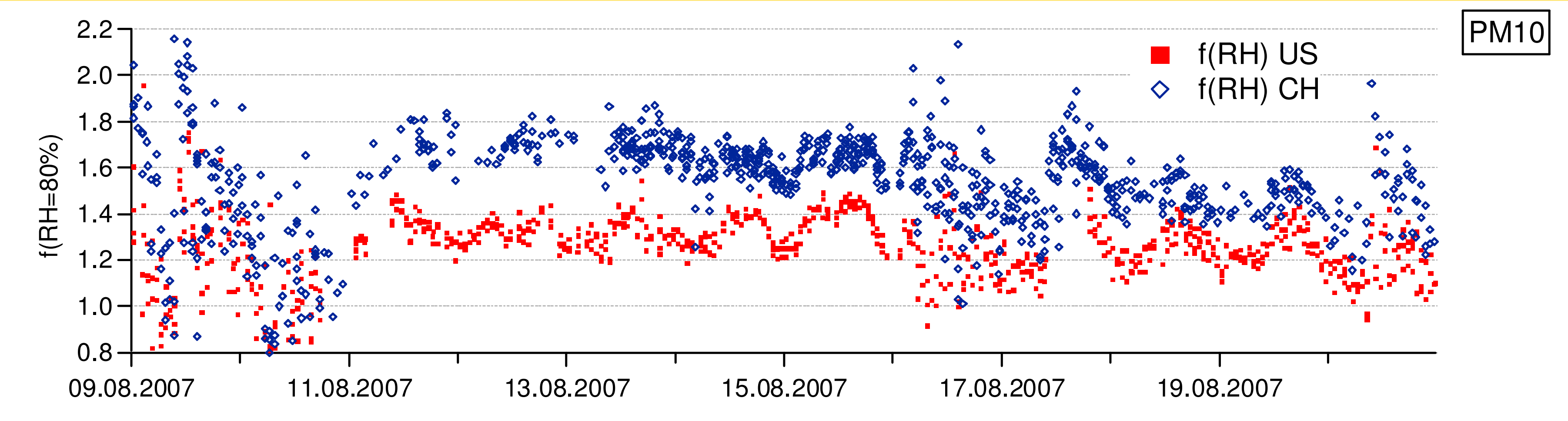
$$f(RH)=\frac{\sigma_s(RH)}{\sigma_s(RH=dry)}$$



- Size and Chemistry are important

Measurements in the Black Forest

- Comparison of two humidified nephelometers

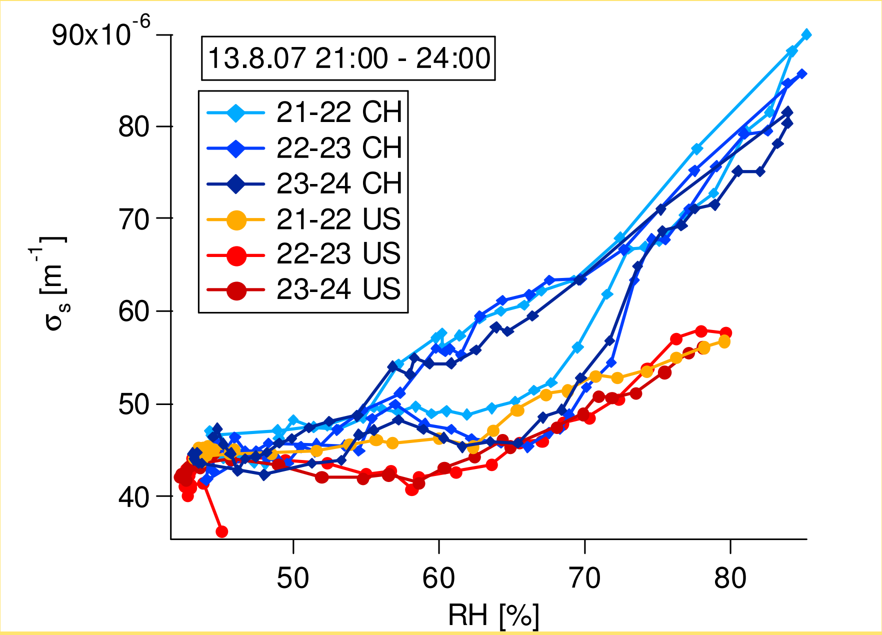


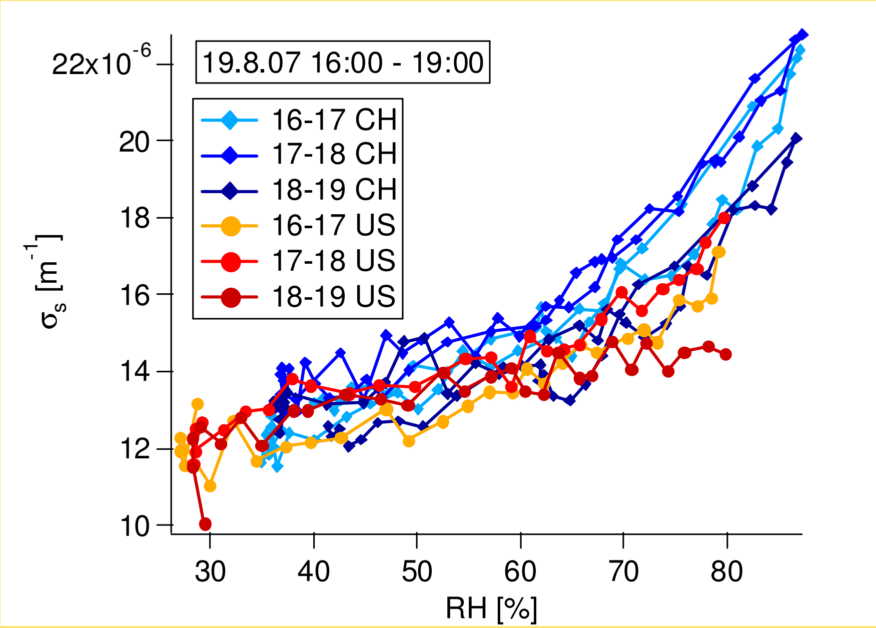
- f(RH) of (mixed) ambient aerosols are much lower compared to NaCl and $(NH_4)_2SO_4$

→ influence of OC (organic carbon), soot and the size distribution

Measurements in the Black Forest

- Phase transitions are occasionally also observed for ambient aerosol





- High fraction of ionic species (ammonium sulphate)

Conclusion

- The motivation behind the presented measurements is the reduction of the relatively large uncertainty of the aerosols climatic forcing

- Light scattering of ambient aerosols depends on RH
- RH dependent aerosol optical properties should be used as input in climate models
- The hydration state of aerosols should be taken into account

Thank you for your attention!