Influence of clouds on aerosol properties and implications for aerosol radiative forcing

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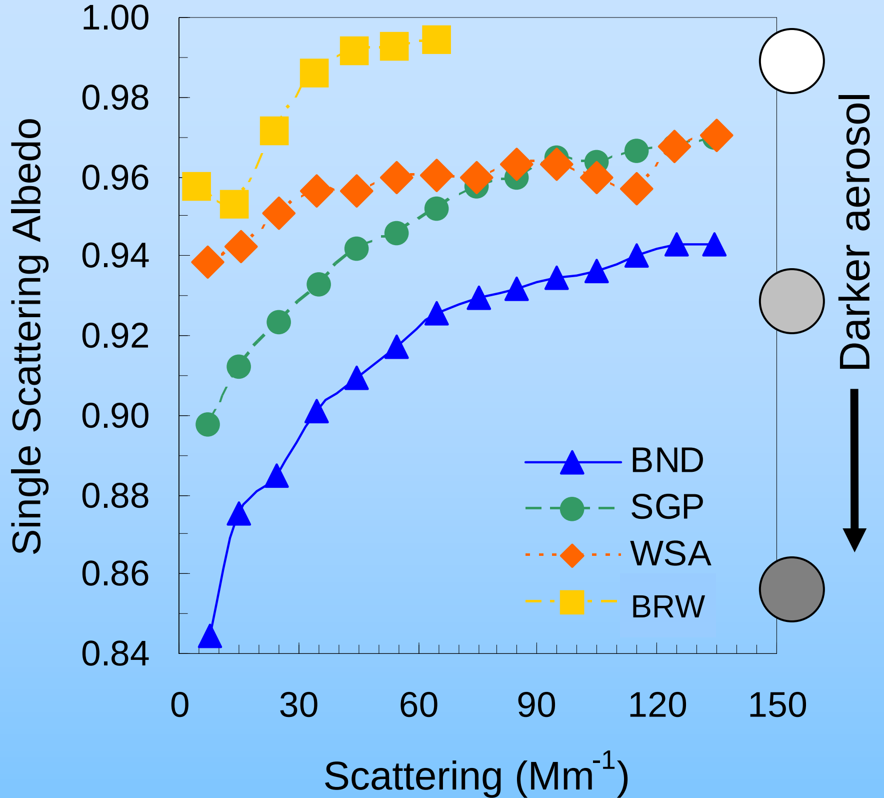
DOE/ASP program provided much of funding for this research

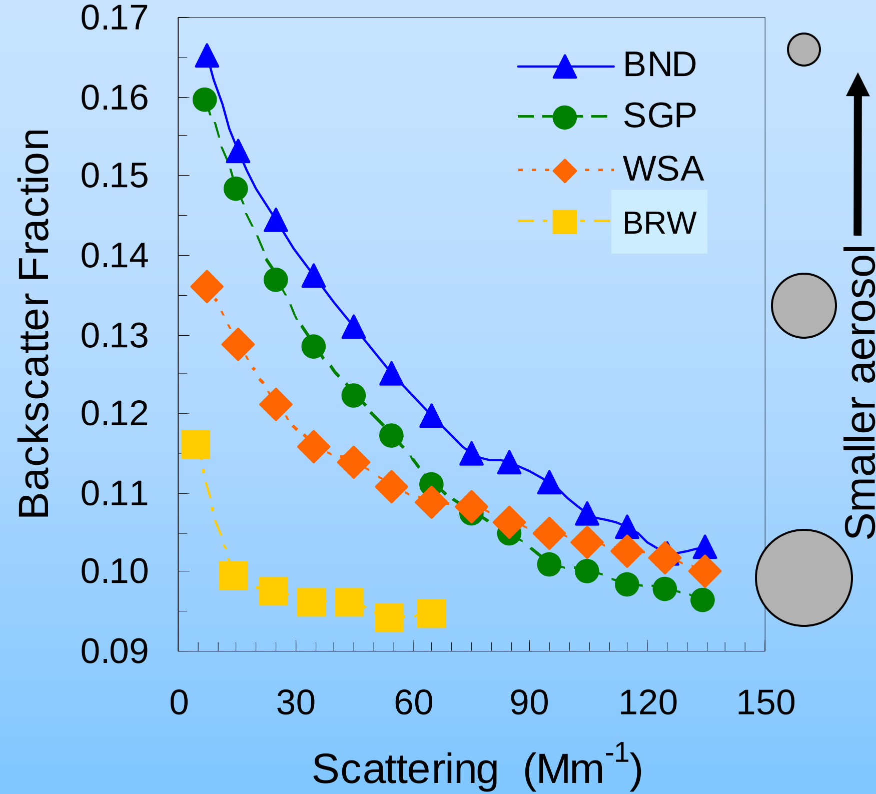
Outline

- Observations from long term measurements
- Hypotheses
- Results from process studies
- Conclusions
- Future work

Pt Reyes Lighthouse

Systematic Variability





Lower amounts of aerosol (less scattering) correspond to smaller, darker particles.

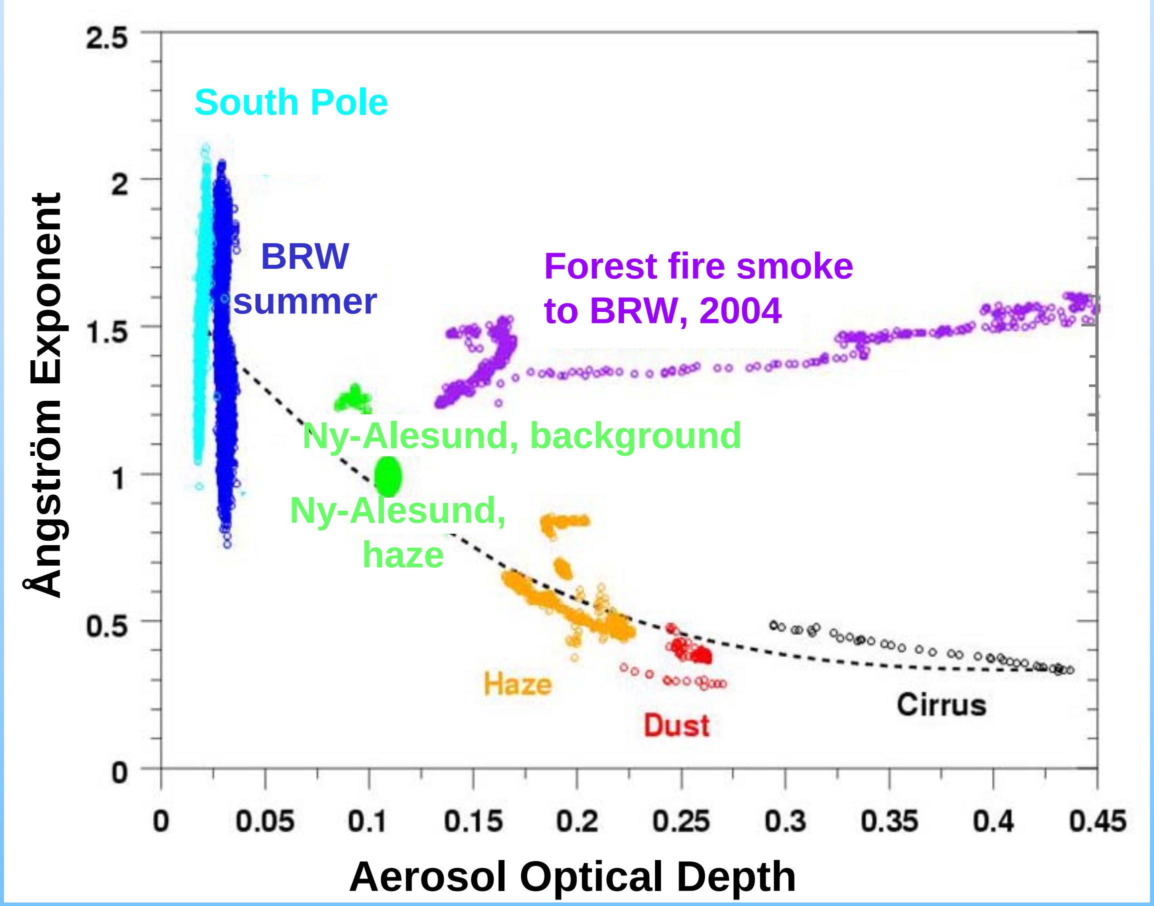
Single Scattering Albedo = scattering / extinction

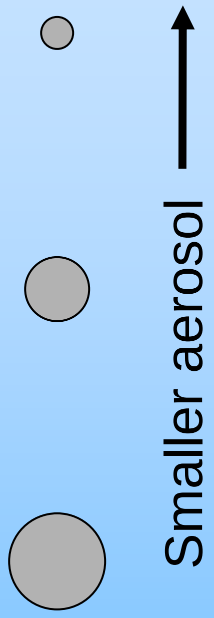
Back-scatter Fraction = back scattering / total scattering

extinction = scattering + absorption

From Delene and Ogren, 2002

Systematic variability and aerosol 'type'

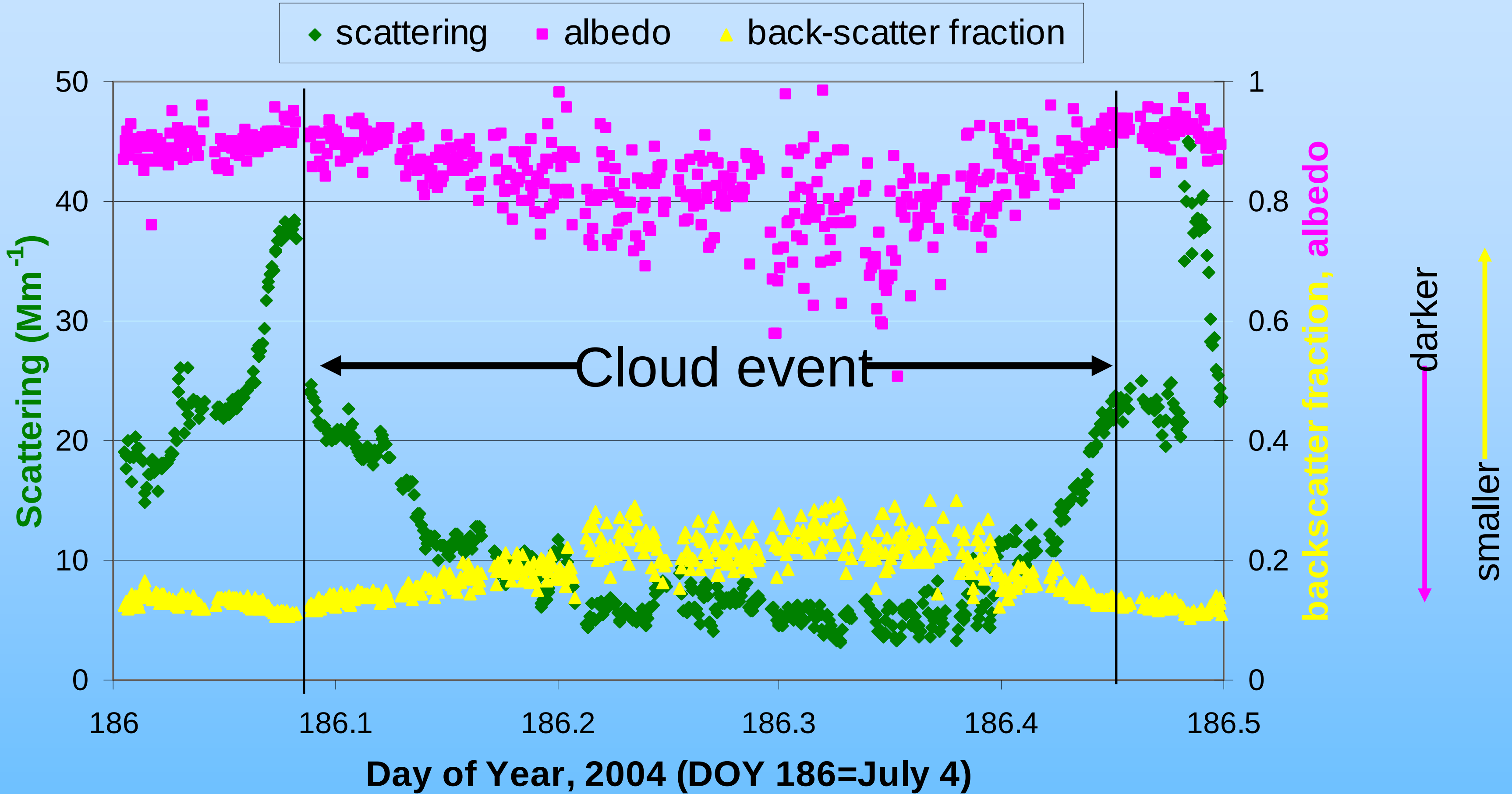




•Data acquired during cloud-free, sun-lit periods in Arctic and Antarctic
•Plot shows column aerosol properties at ambient conditions
•Dust and smoke data for specific events identified by satellite and other data

A version of this plot appeared in Tomasi et al., 2007

Effect of cloud on aerosol optical properties



Data from Chebogue Point, during ICARTT

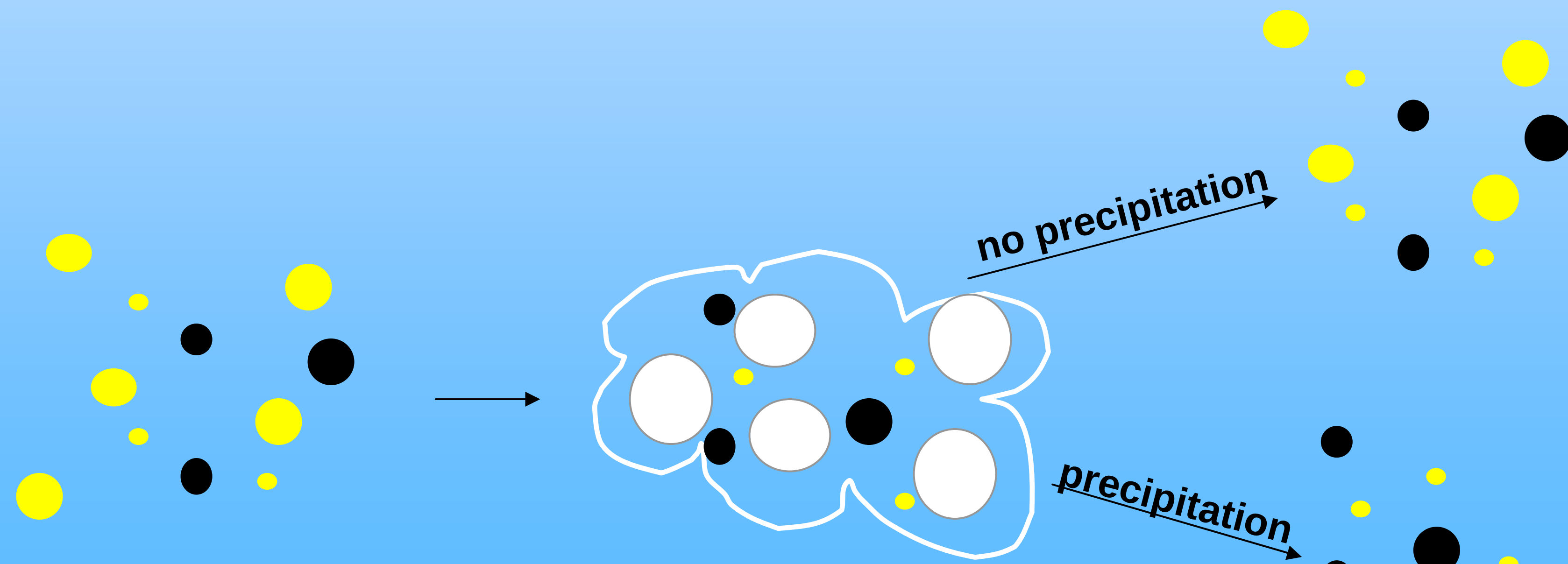
Onset of cloud causes:

- •Decrease in light scattering (less aerosol)
- •Decrease in single scattering albedo (darker aerosol)
- •Increase in backscatter fraction (smaller aerosol)

Hypotheses

- Aerosol light scattering is dominated by particles that are readily-scavenged by clouds, (e.g., sulfates)
- Aerosol light absorption is dominated by less readily-scavenged particles, (e.g., soot)
- Larger particles are more readily scavenged by clouds

→ **In a cloud, the unscavenged particles – interstitial aerosol - will be enriched in darker, smaller particles**

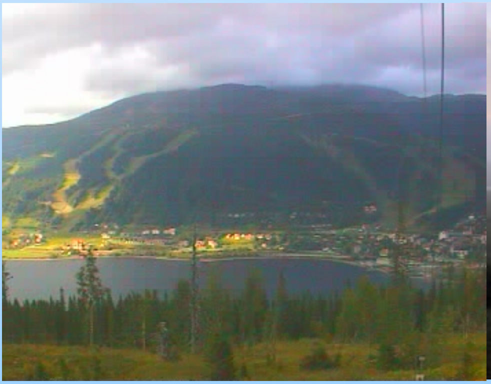


Absorbing aerosol (●)
Scattering aerosol (●)

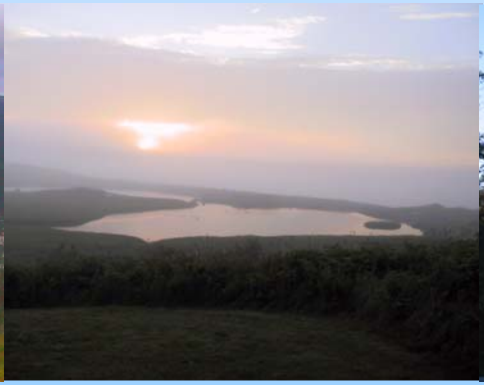
Large, scattering aerosol particles are scavenged.

Sampling Sites

Mt. Åreskutan, Sweden (ARE)



Chebogue Point, Nova Scotia (CBG)



Holme Moss, United Kingdom (HLM)

Point Reyes, California (PYE)

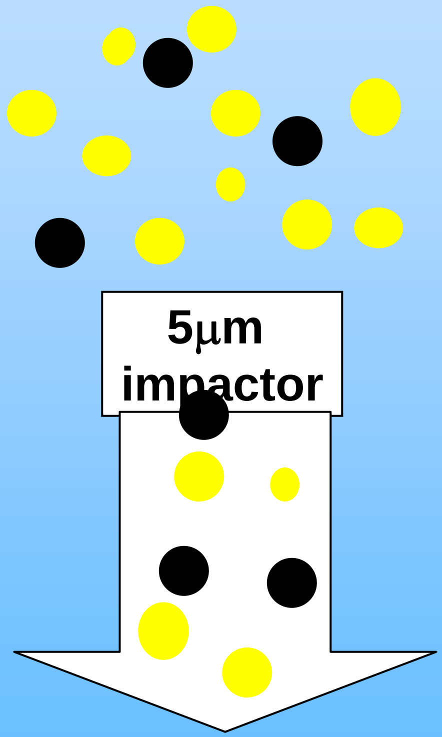
Site	Campaign	When	Aerosol type
ARE	SOACED	Summer, 2004	Arctic air, remote continental
CBG	ICARTT	Summer, 2004	Aged urban
HLM	n/a	Winter, 2006	Aged urban, biomass burning
PYE	MASRAD	Summer, 2005	Clean marine

- Sites had approximately same amount of aerosol - relatively clean
- Aerosol spanned range of size and darkness observed at other sites

Testing the Hypothesis

Assume cloud drops ( ) are larger than 5 µm in diameter.
Assume particles ( ) are smaller than 5 µm in diameter.

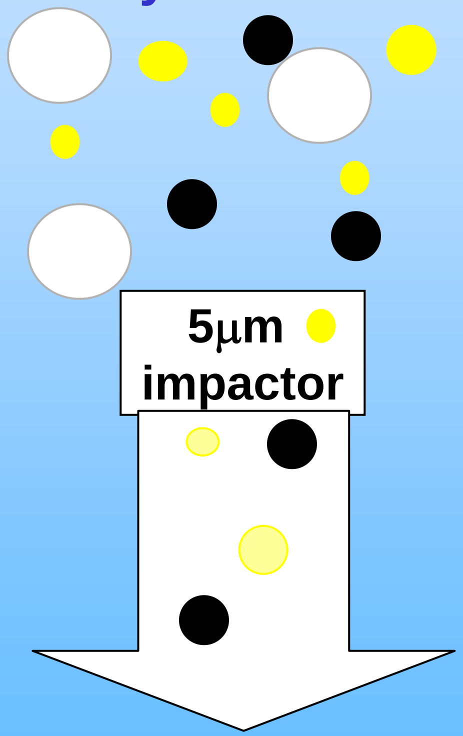
Clear conditions



INSTRUMENTS

Sampling inlet excludes cloud droplets
→ in-cloud measurements represent the particles that were not scavenged by the clouds - interstitial aerosol

Cloudy conditions



INSTRUMENTS

Note: air is at low RH (dry) when sampled by instruments

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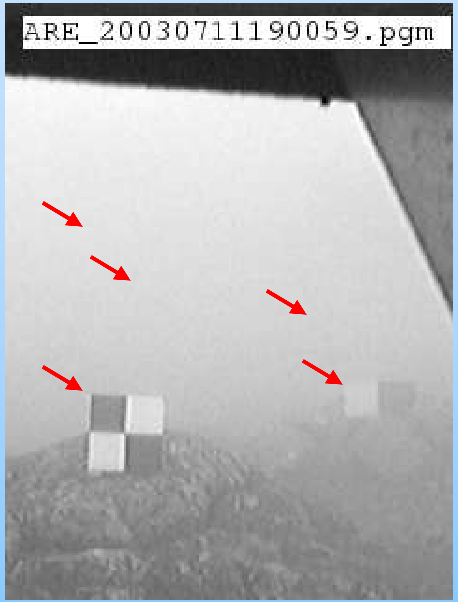
Identifying cloudy periods



Using a webcam

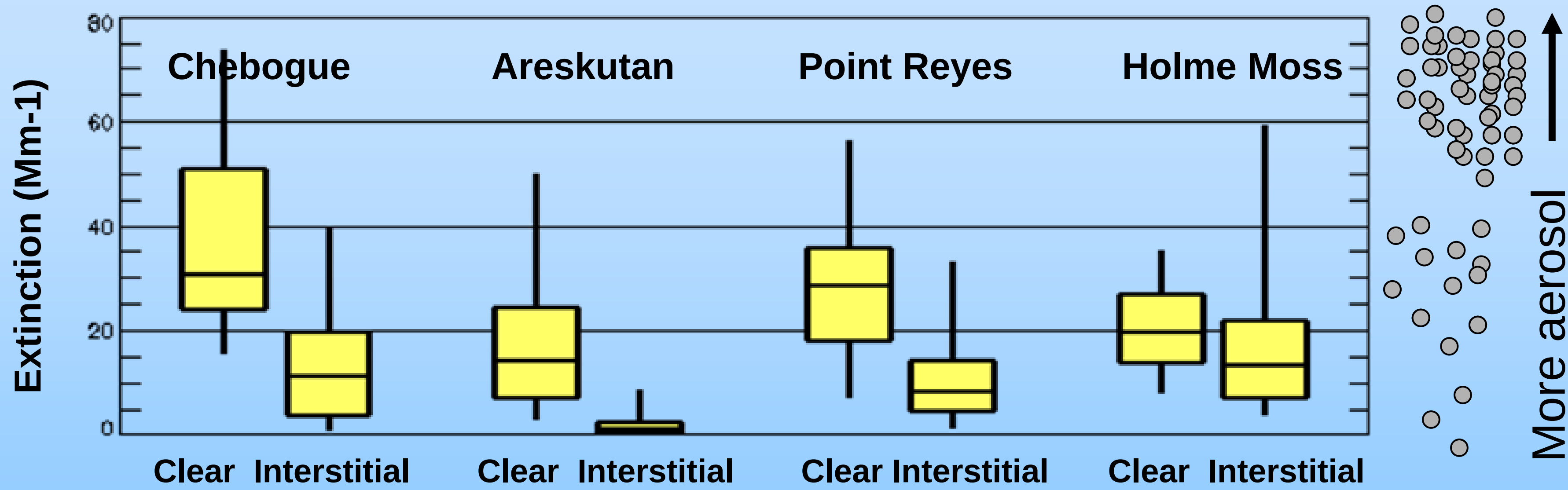
- Clouds decrease the contrast between black and white target areas
- Contrast reduction is a function of cloud extinction coefficient and distance from camera
- Works in places with LONG hours of daylight (Sweden in summer)

Other methods

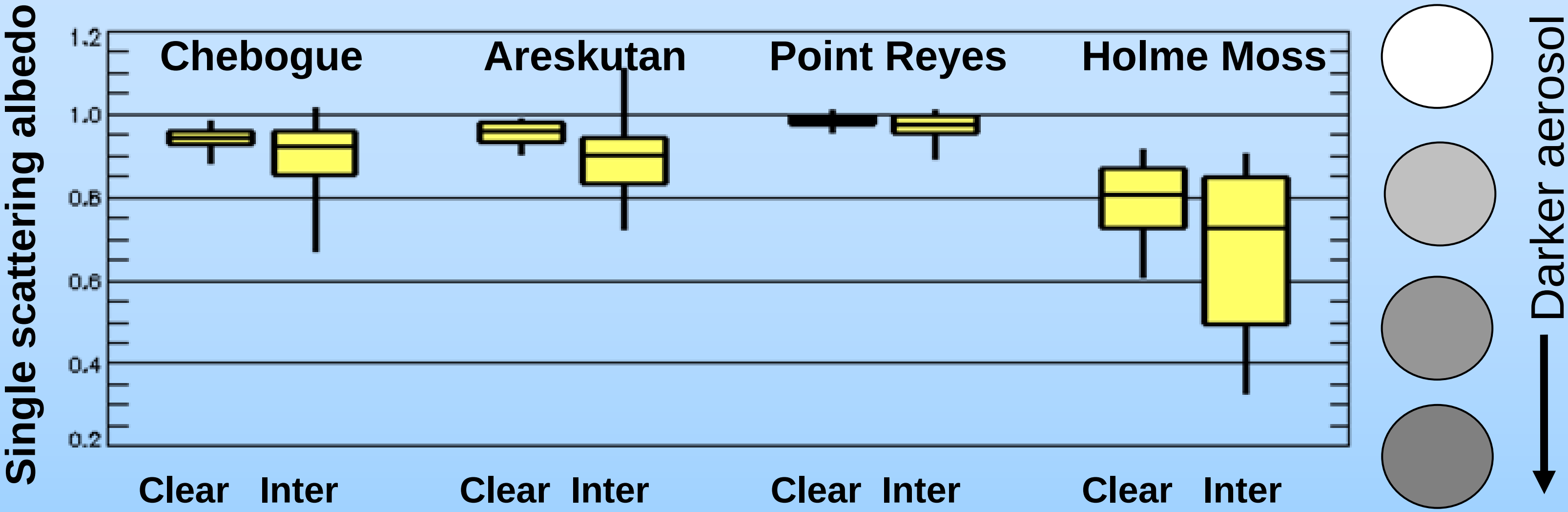
•IR flux ratios – Ratio of downwelling to upwelling IR flux > 0.99 →cloud

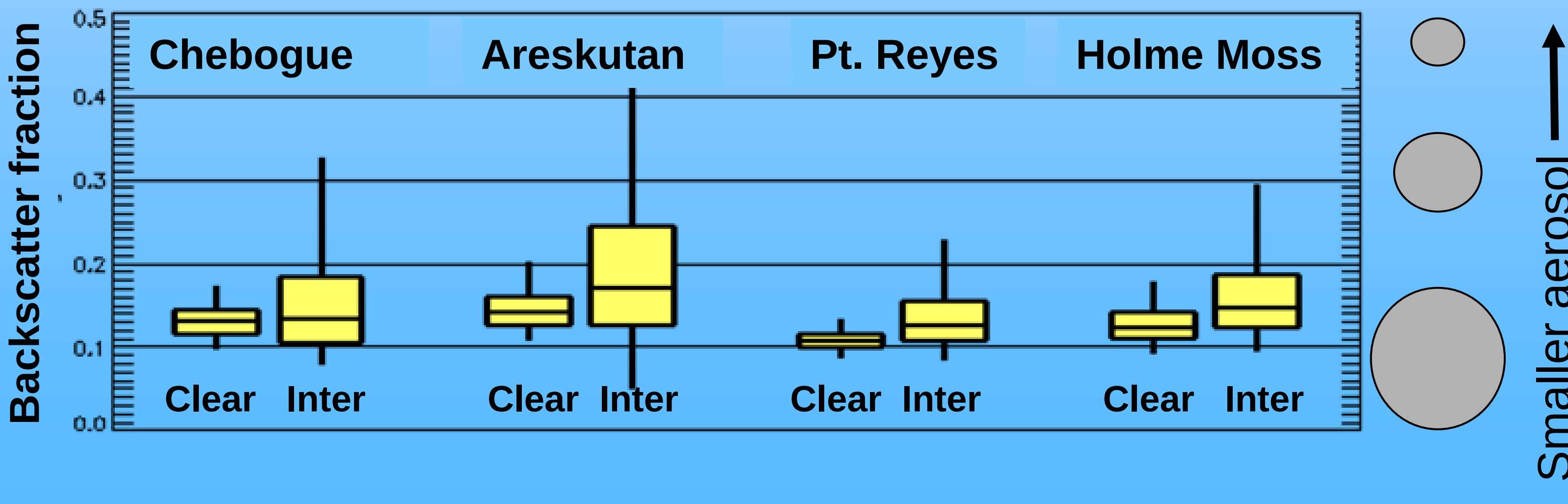
•Off the shelf instruments (e.g., Present Weather Sensor)

Effect of cloud on aerosol properties

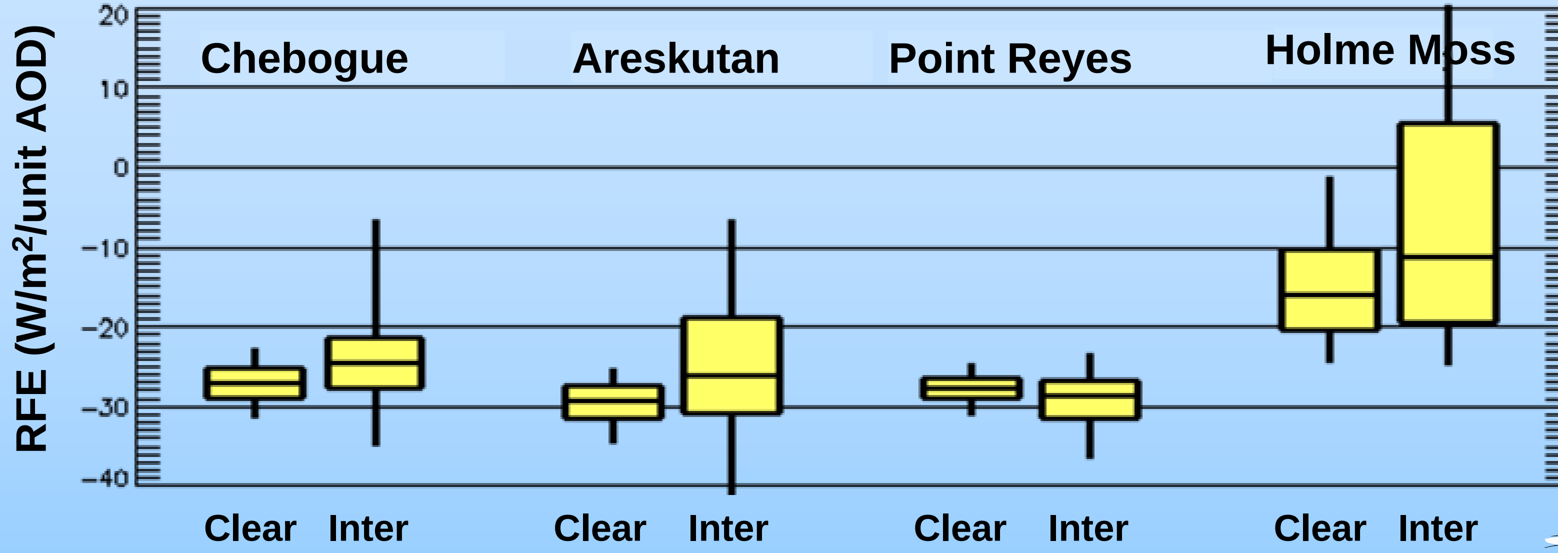


Effect of cloud on aerosol properties





Cloud processing and aerosol radiative forcing efficiency



→Cloud processing tends to reduce cooling <u>nature</u> of aerosol
→Changes in aerosol properties have opposite effects on RFE

- Darker aerosol → less cooling
- Smaller aerosol → more cooling

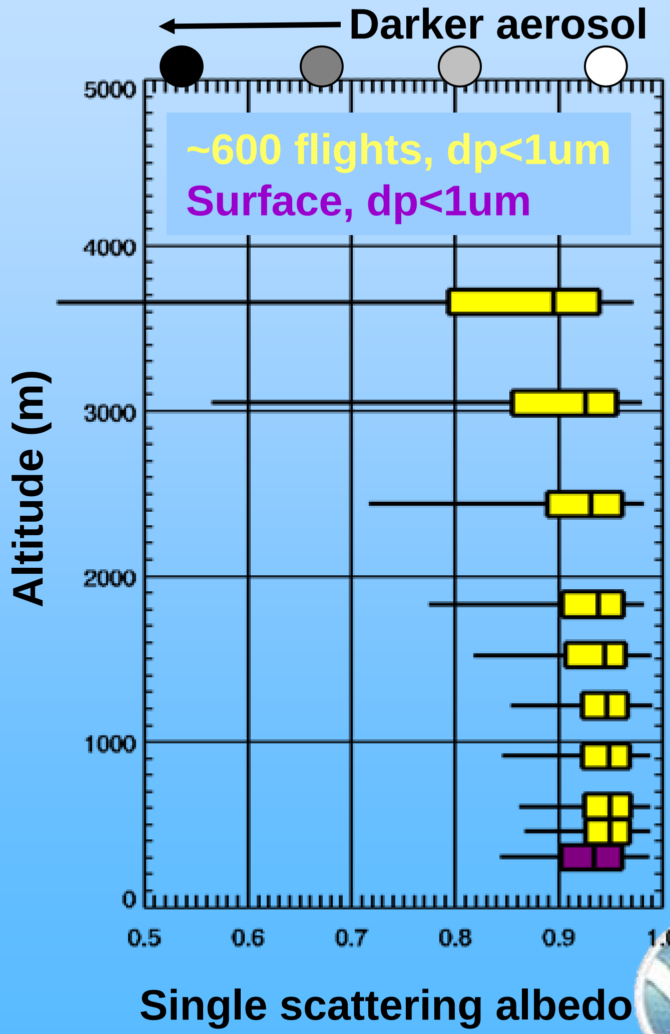
PYE: dominant aerosol change → increase in backscatter fraction → decrease RFE

Implications for aerosol lifetimes

The preferential scavenging of scattering aerosol by clouds may explain the relatively long lifetime and ubiquity of absorbing aerosol in the atmosphere.

→Smoke from the 2004 Alaskan forest fires was detected in Norway and Greenland a month after emission.

→Relatively dark aerosol (i.e., low single scattering albedo) is observed in some remote locations (e.g., aerosol aloft, Arctic haze).



Measurements made over DOE-ARM site in Oklahoma, March 2000-September 2005.

Conclusions

•Interaction with clouds changes the optical properties of aerosol.
→Less aerosol, darker aerosol, smaller aerosol

•Cloud-processed aerosol (as represented by interstitial aerosol) tends to be less cooling than aerosol measured in clear conditions.
→Cloud processing increases warming potential of aerosol

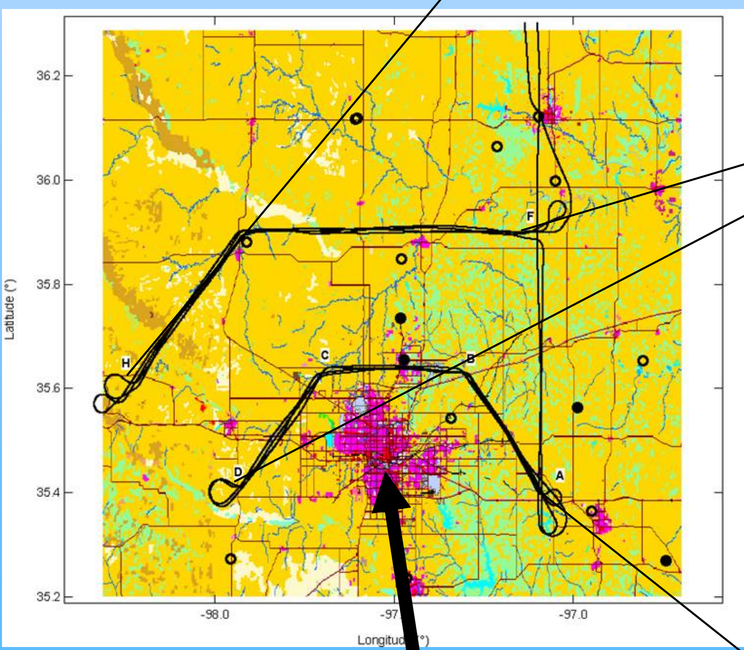
•Future work
→Investigate how differences in cloud processing are driven by other aerosol properties, e.g., hygroscopicity/composition, size, etc.

CHAPS flights, June 2007

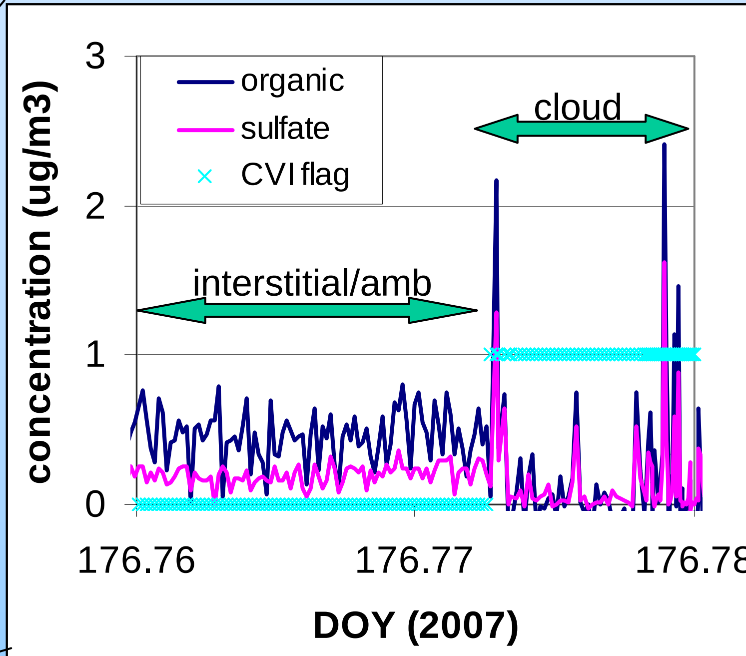
Preliminary AMS data, courtesy BNL

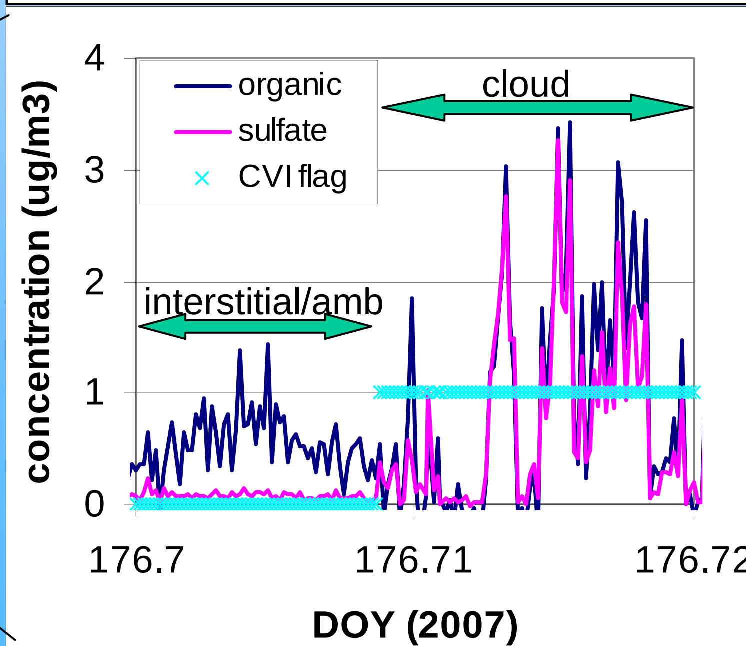
- **Flew within and outside clouds**
- **Measured chemical composition of cloud drop residuals, ambient, and interstitial aerosol**

Organic dominates interstitial and ambient aerosol.
Sulfate concentration increases relative to organic in cloud drop residuals

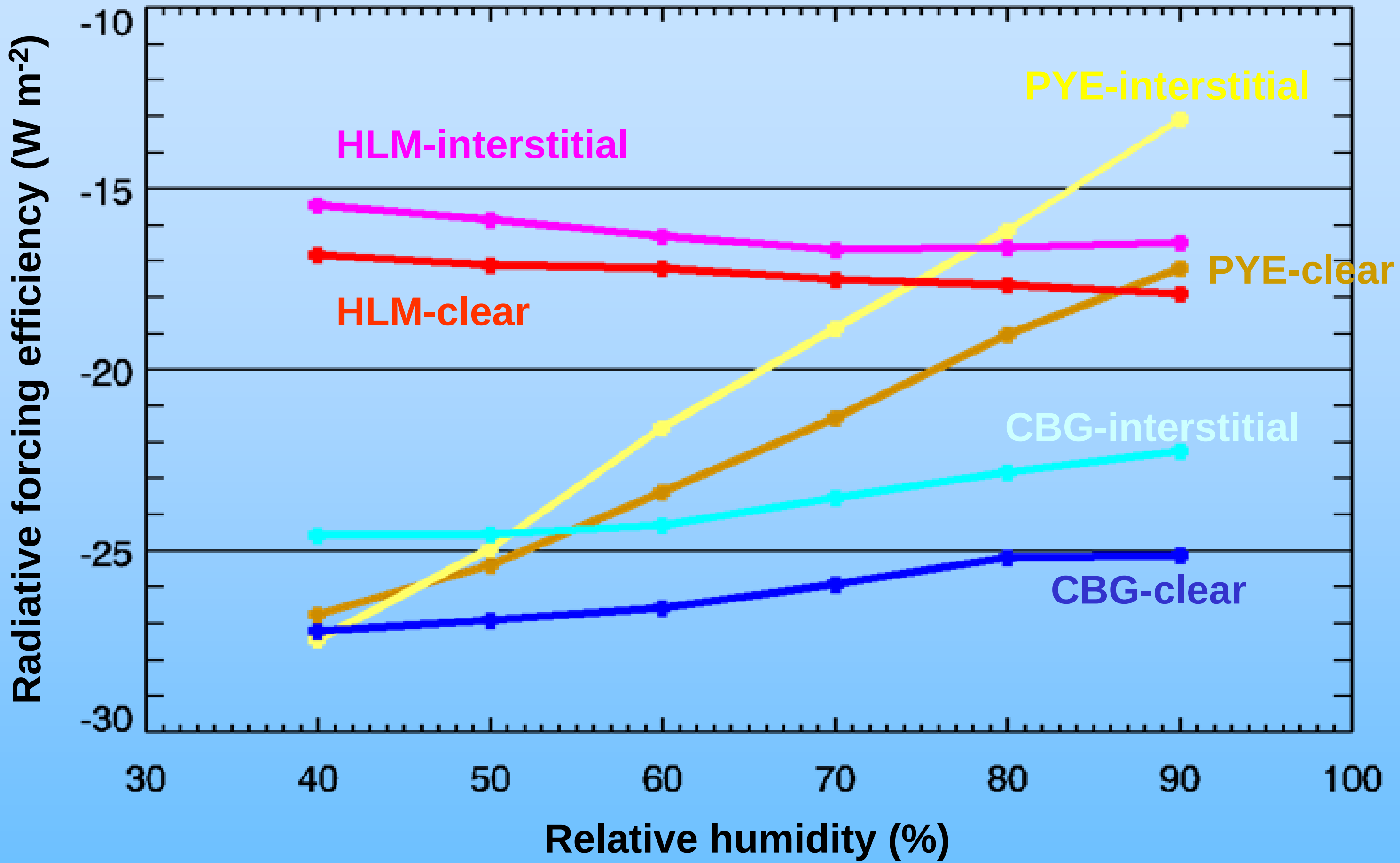


Oklahoma City





Forcing efficiency at ambient relative humidity



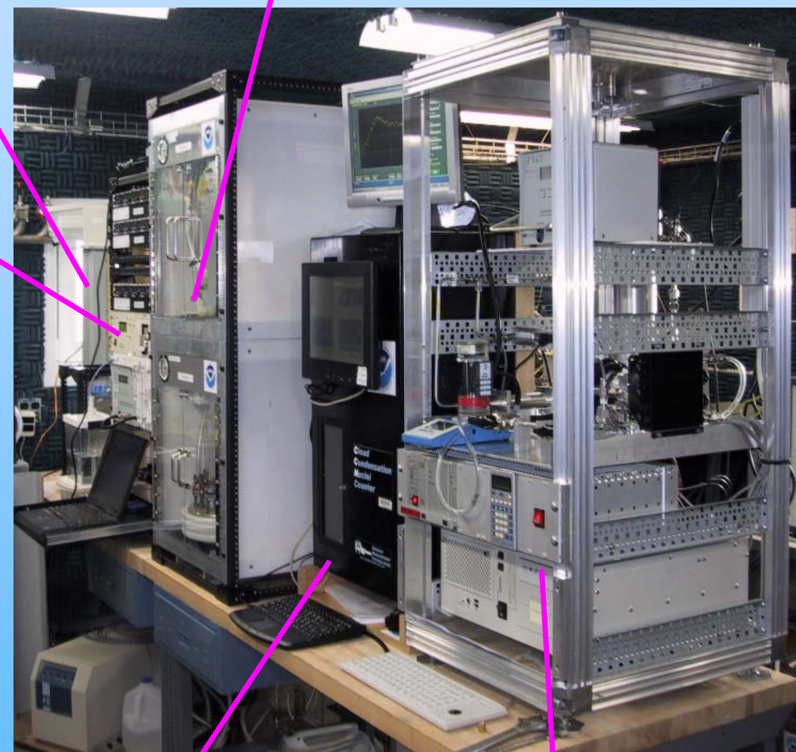
Water uptake by particles can have significant effect on forcing efficiency.
Interstitial aerosol still tends to be more warming than ambient aerosol when RH considered.
Difference is likely due to composition, microphysics or some combination.

Aerosol Sampling

Nephelometers and humidograph system
Scattering, backscattering, hygroscopicity (f(RH))

Aerosol chemistry
Inorganic ions and organics

PSAP and CPC
Absorption and N_{CN}



Cloud condensation nuclei counter
N_{CCN} as f(SS)

Aerosol size distribution

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Parameters controlling aerosol forcing

$$\Delta F \approx -DS_0 T_{at}^2 (1 - A_c)(1 - R_s)^2 \tilde{\omega}_0 \bar{\beta} \delta \left[1 - \frac{2R_s}{(1 - R_s)^2} \left(\frac{1 - \tilde{\omega}_0}{\tilde{\omega}_0 \bar{\beta}} \right) \right]$$

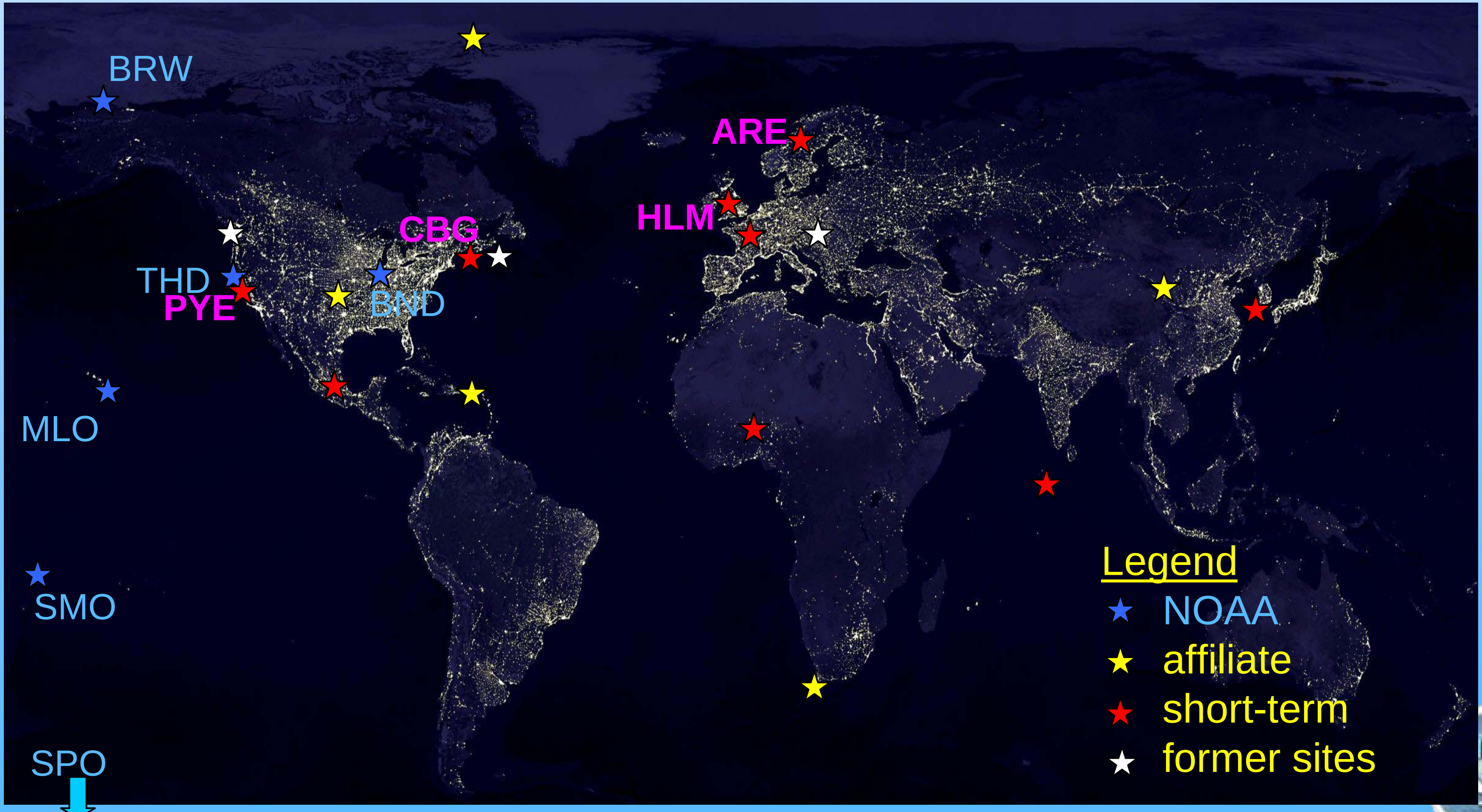
ΔF	average aerosol forcing at top of atmosphere (TOA)	D	daylight fraction
δ	aerosol optical depth	S_0	solar constant
$\tilde{\omega}_0$	aerosol single-scattering albedo	T_{at}	atmospheric transmission
$\bar{\beta}$	average aerosol up-scatter fraction	A_c	cloud fraction
		R_s	surface albedo

$\Delta F / \delta$ = Radiative Forcing Efficiency

Source: Haywood and Shine (1995)

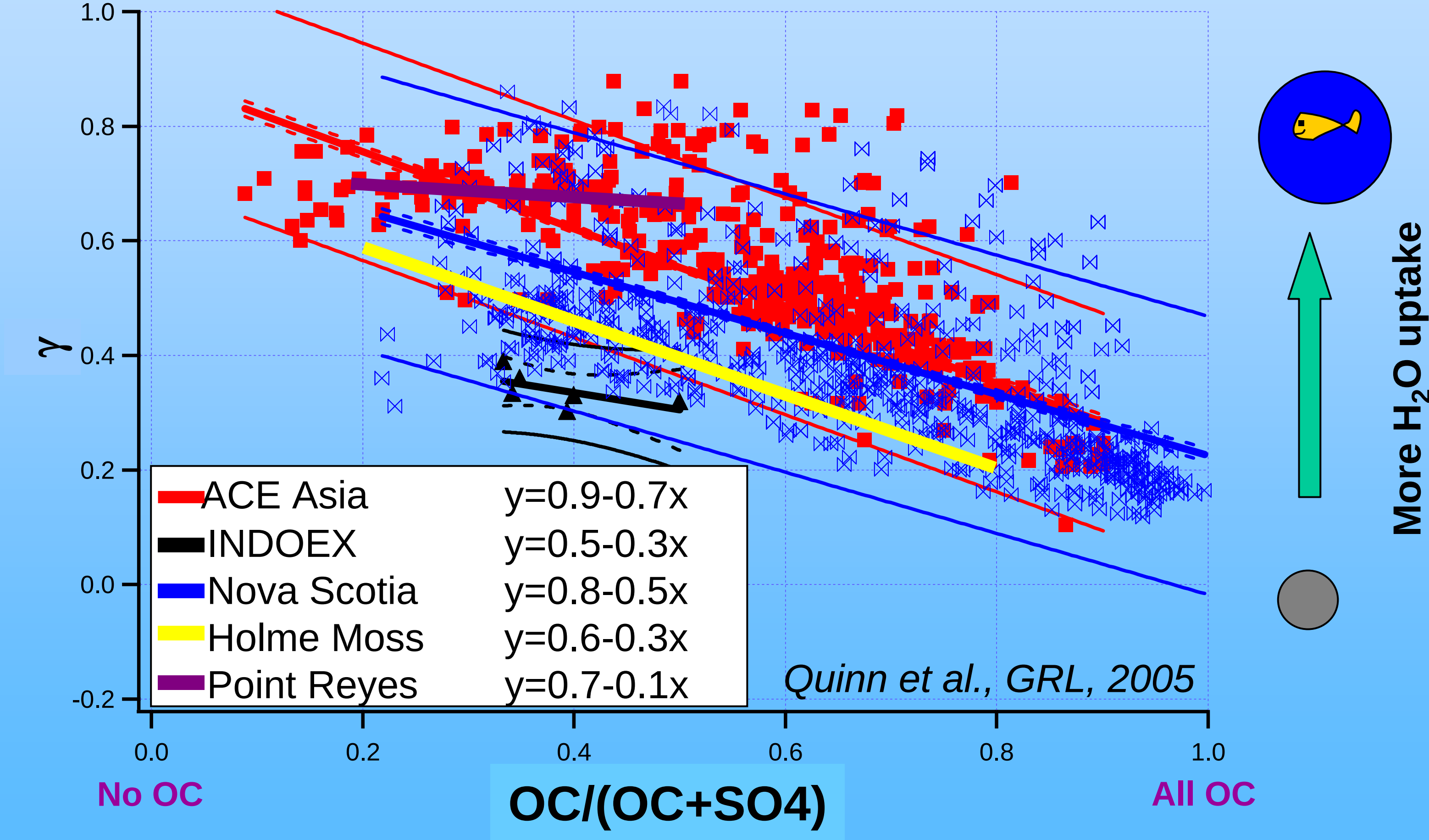
Scientific Questions

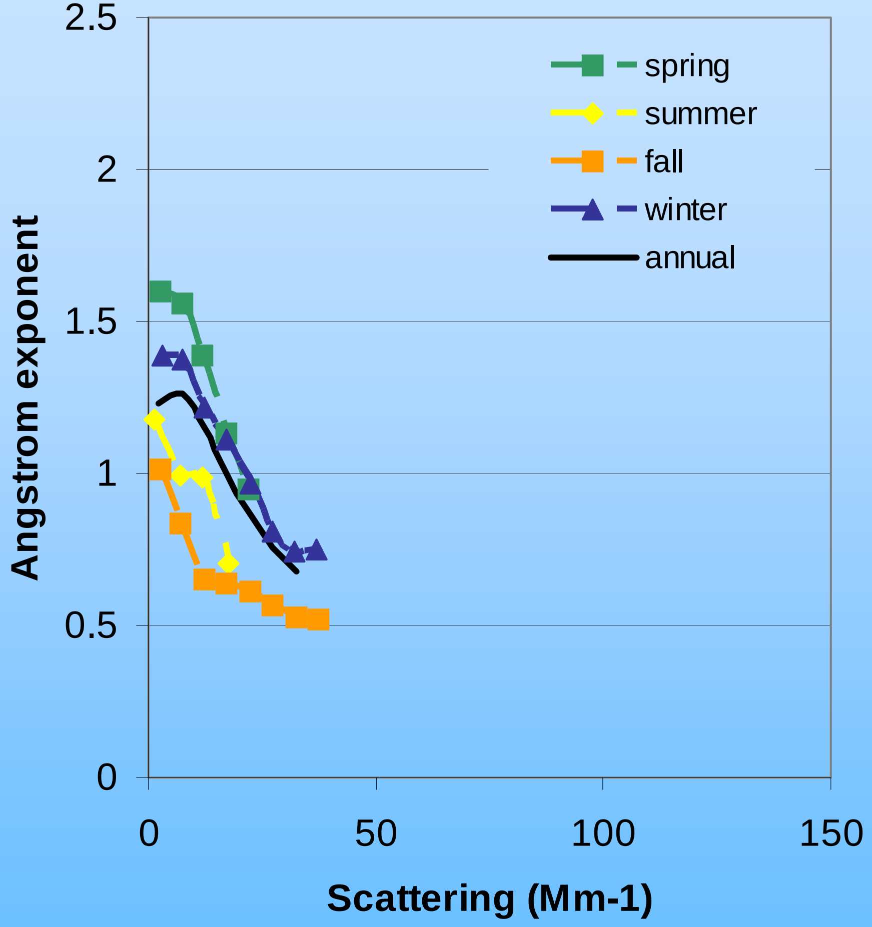
- How do clouds change the optical properties of the aerosol?
- What are the implications for radiative forcing?

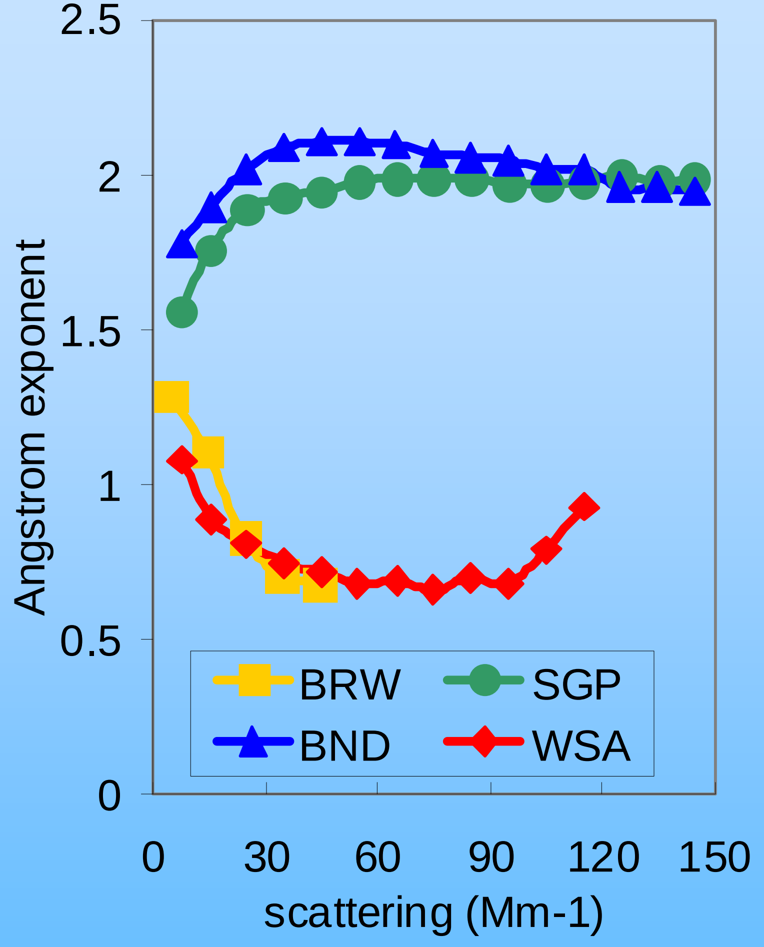


Aerosol Hygroscopicity f(RH)

$$f(RH) = \frac{\sigma_{sp,high}}{\sigma_{sp,low}} = \left[\frac{100-RH_{low}}{100-RH_{high}}\right]^{\gamma}$$

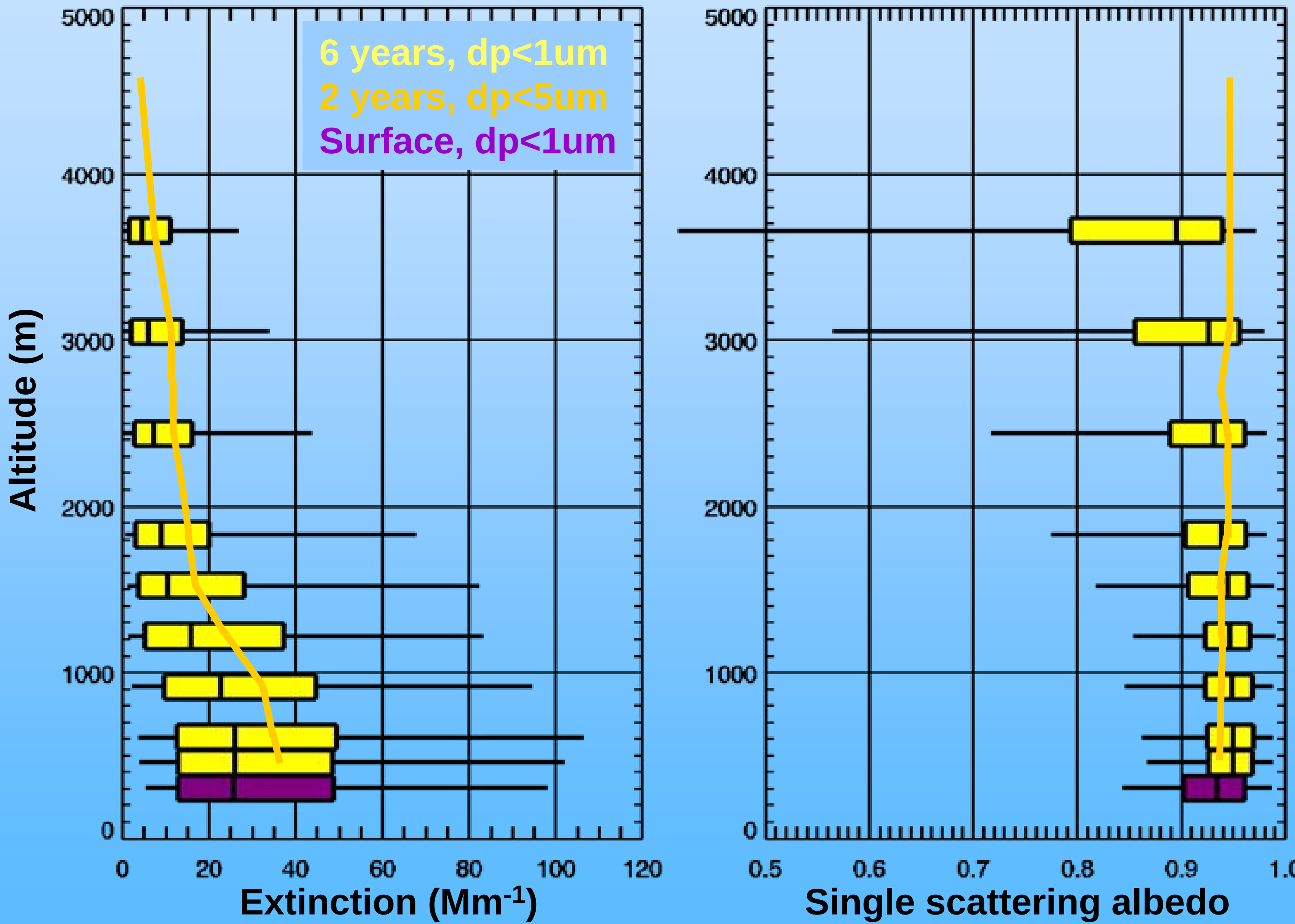




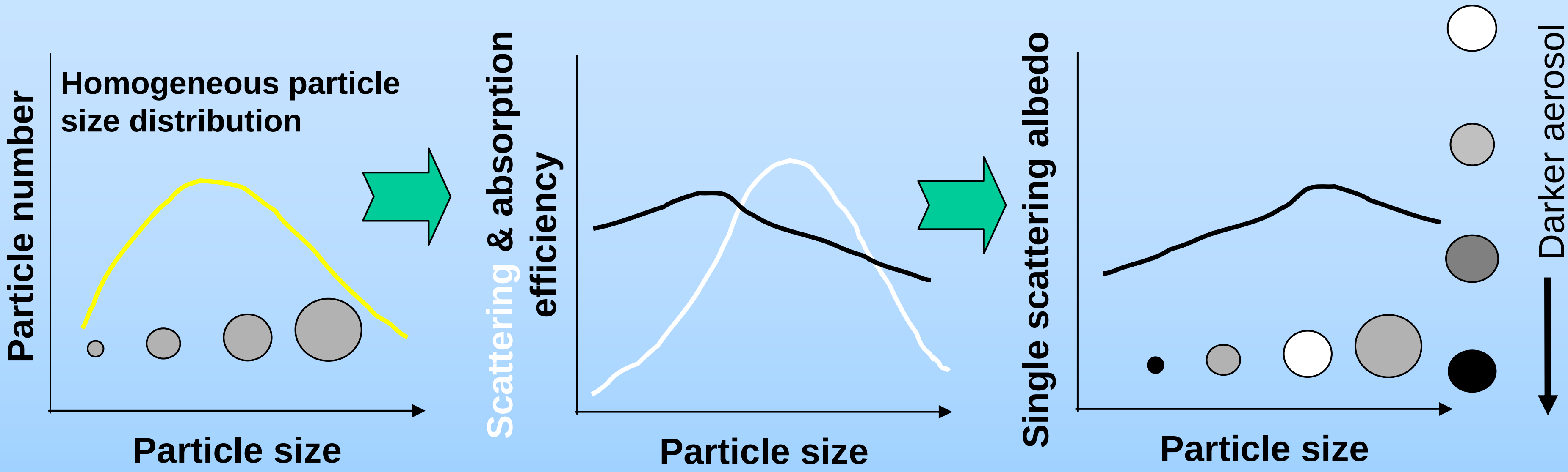


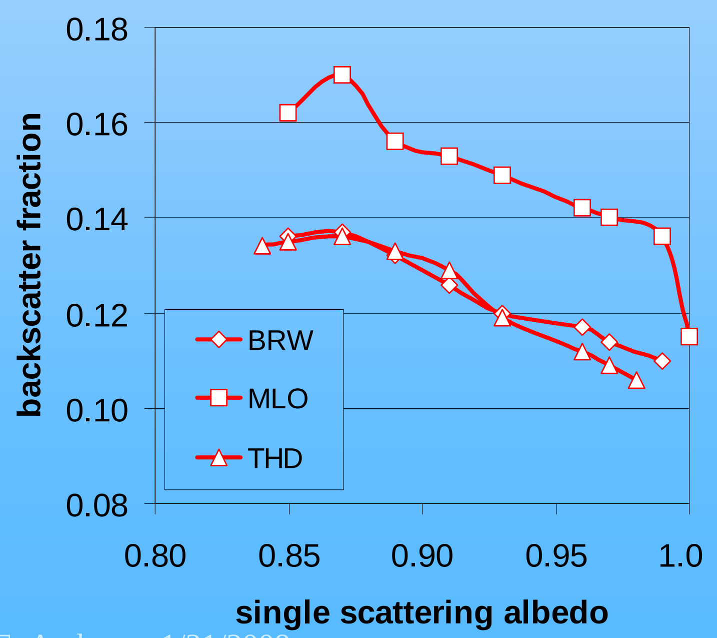
The systematic variability may be due to aerosol type

Airborne measurements over Oklahoma (March 2000-November 2007)



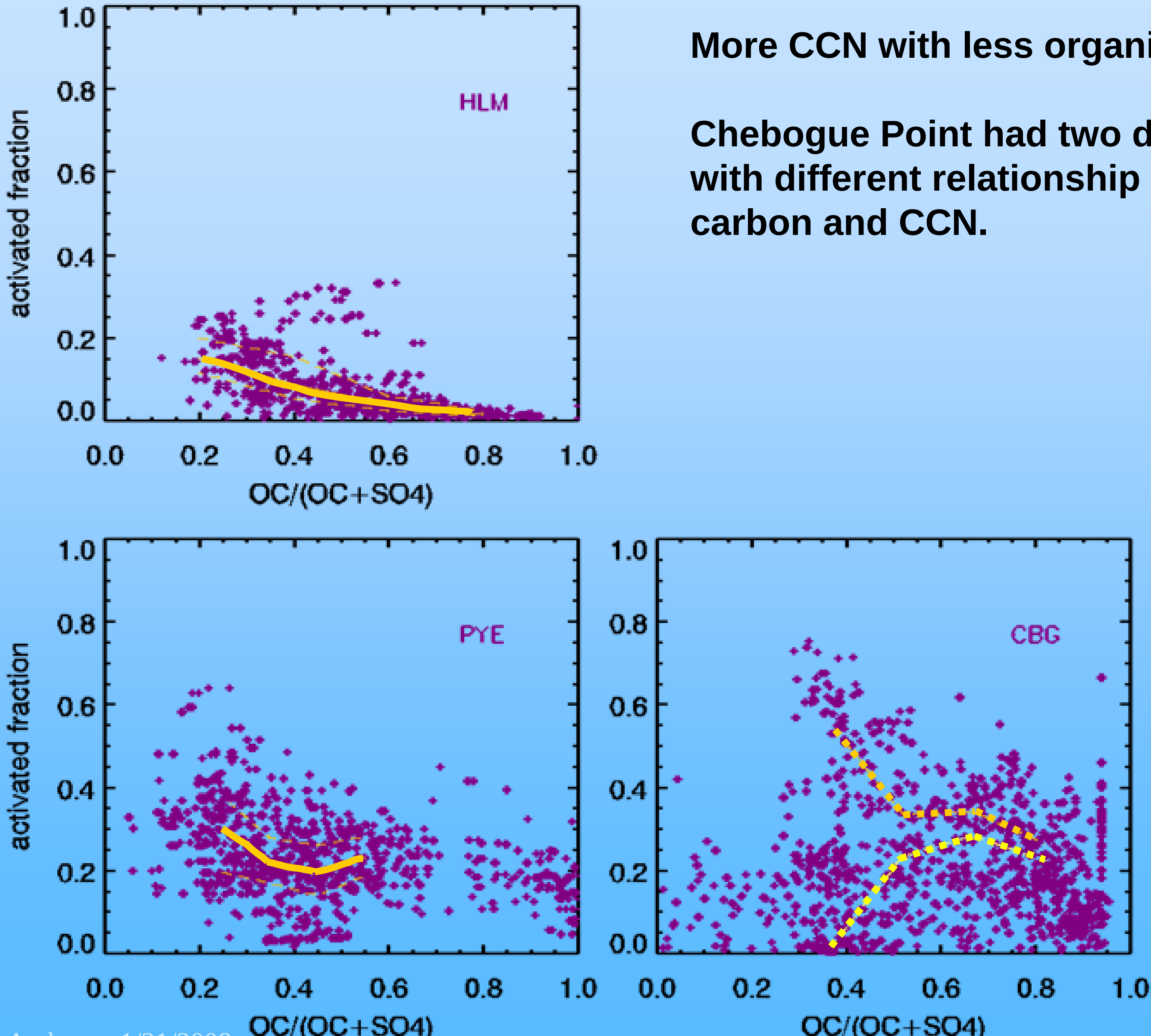
Interdependence of SSA and particle size





Scattering efficiency falls off faster with decreasing size than absorption efficiency.
→for a given composition, particle albedo is a function of size

Systematic relationship between SSA and BFR
→absorbing particles tend to be 'small' e.g., soot
→SSA related to proportion of smaller aerosol

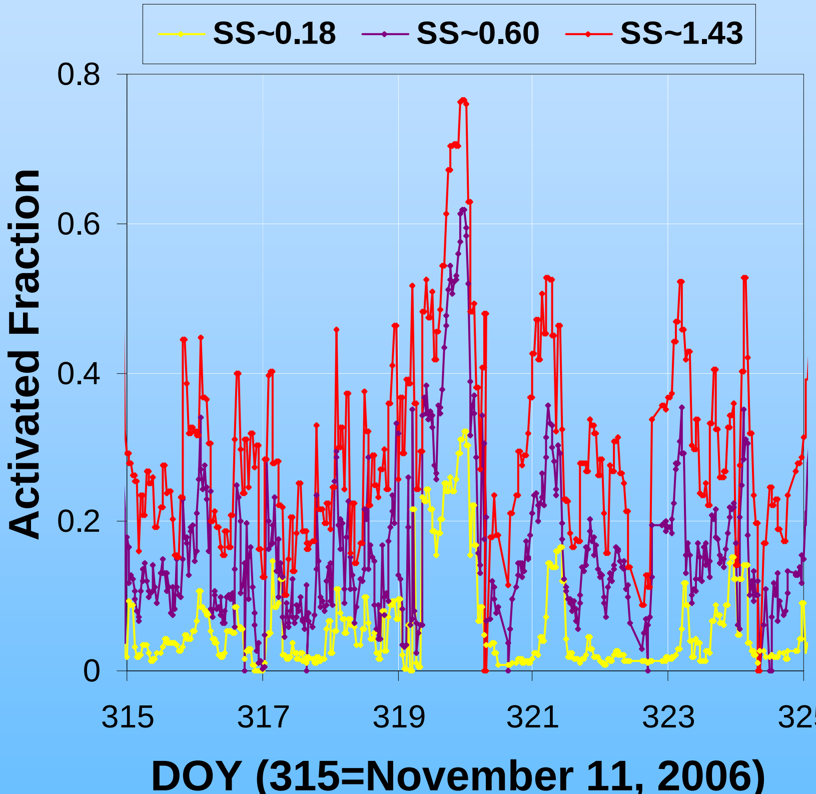


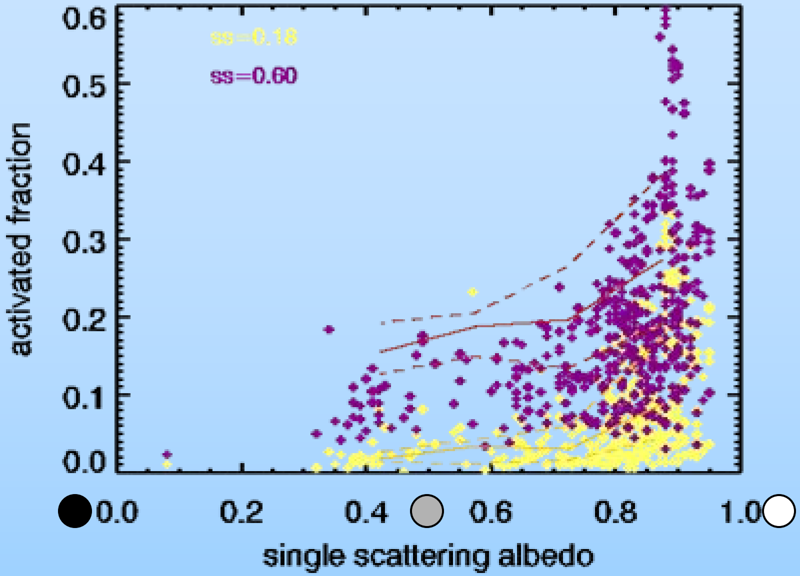
More CCN with less organic carbon…usually!

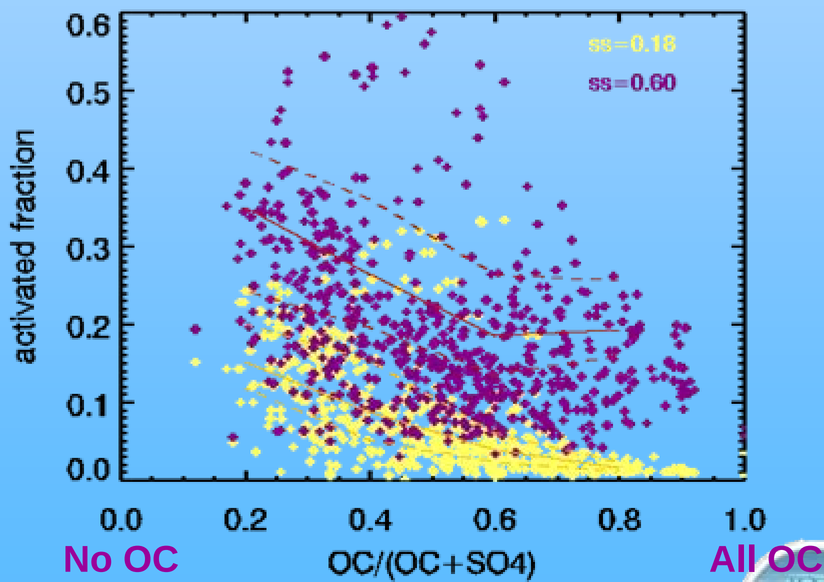
Chebogue Point had two different airmasses with different relationship between organic carbon and CCN.

CCN activation properties at Holme Moss

Activated fraction= $\frac{CCN}{CN}$







- activated fraction highly variable
- 100% activation never observed
- Compositional dependence?

Implications for global aerosol optical properties

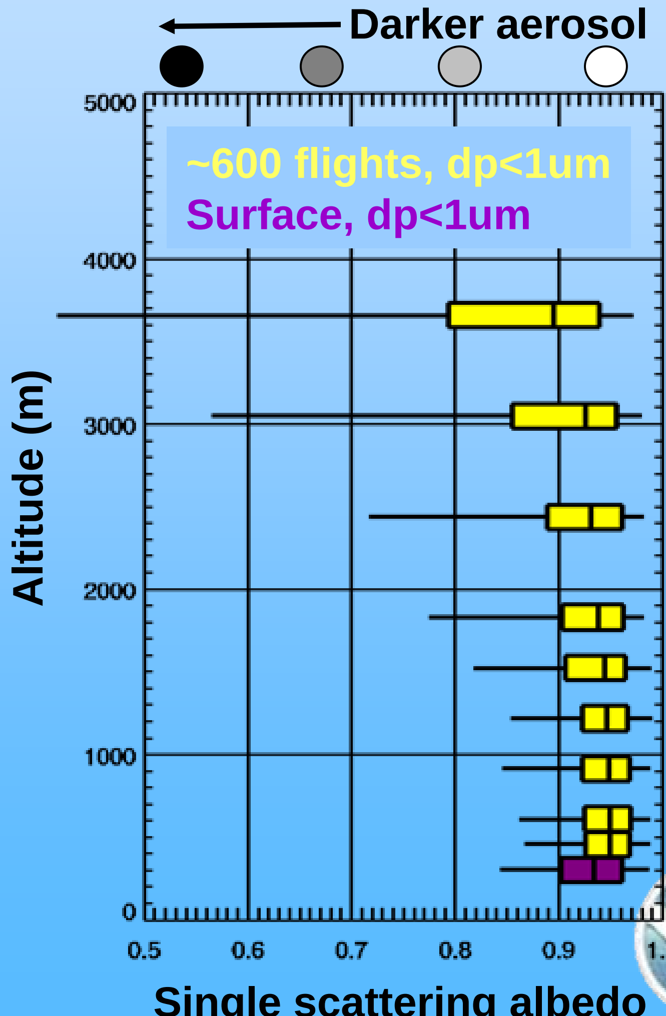
The preferential scavenging of scattering aerosol by clouds may explain the relatively long lifetime and ubiquity of absorbing aerosol in the atmosphere.

→Smoke from the 2004 Alaskan forest fires was detected in Norway and Greenland a month after emission:

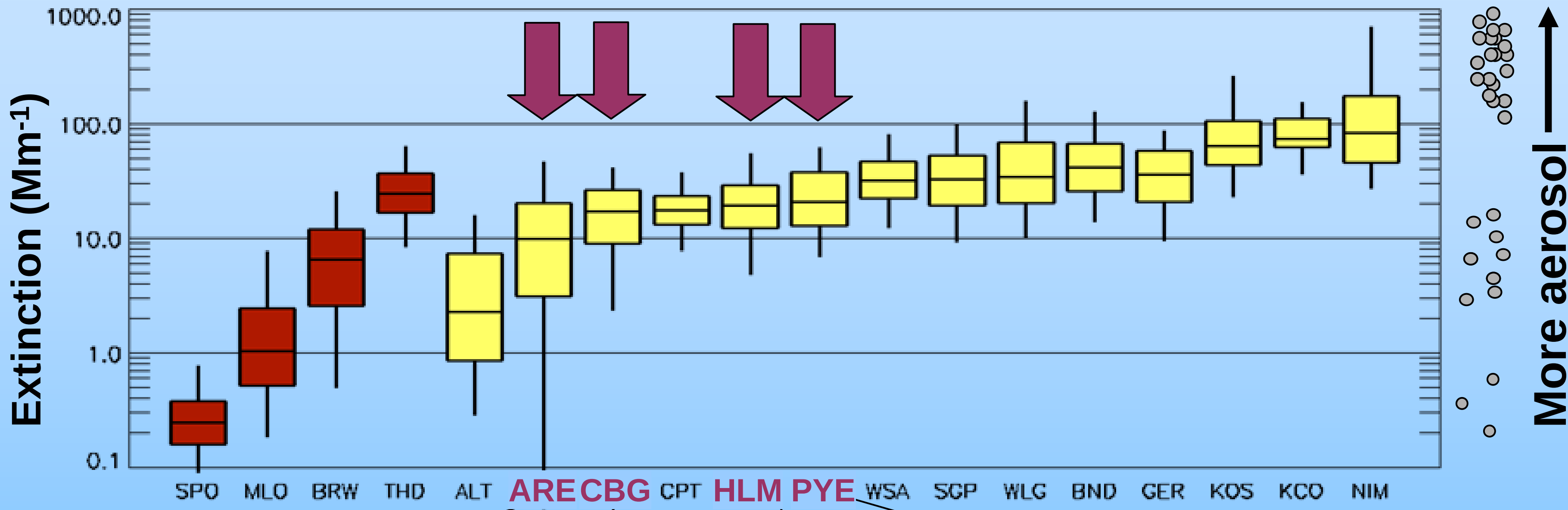
Station	Episode max.	Summer	Haze	Annual
Barrow abs	34.0	0.05	0.44	0.17
Alert abs	0.71	-	0.36	0.17
Norway abs	0.63	0.05	0.47	0.11
Greenl. EBC	828	20.2	12.8	14.5

From Stohl et al., 2006

→Relatively dark aerosol (i.e., low single scattering albedo) is observed in some remote locations (e.g., aerosol aloft, Arctic haze)



Light Extinction



Mt. Åreskutan, Sweden (ARE)

Chebogue Point, Nova Scotia (CBG)

Holme Moss, United Kingdom (HLM)

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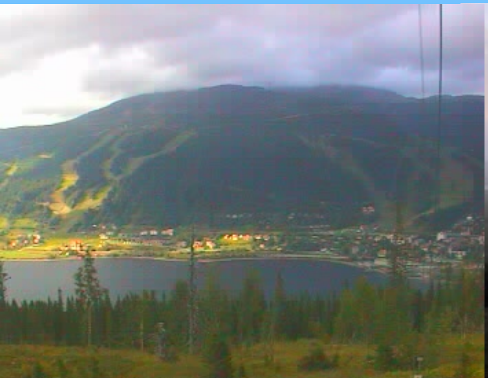
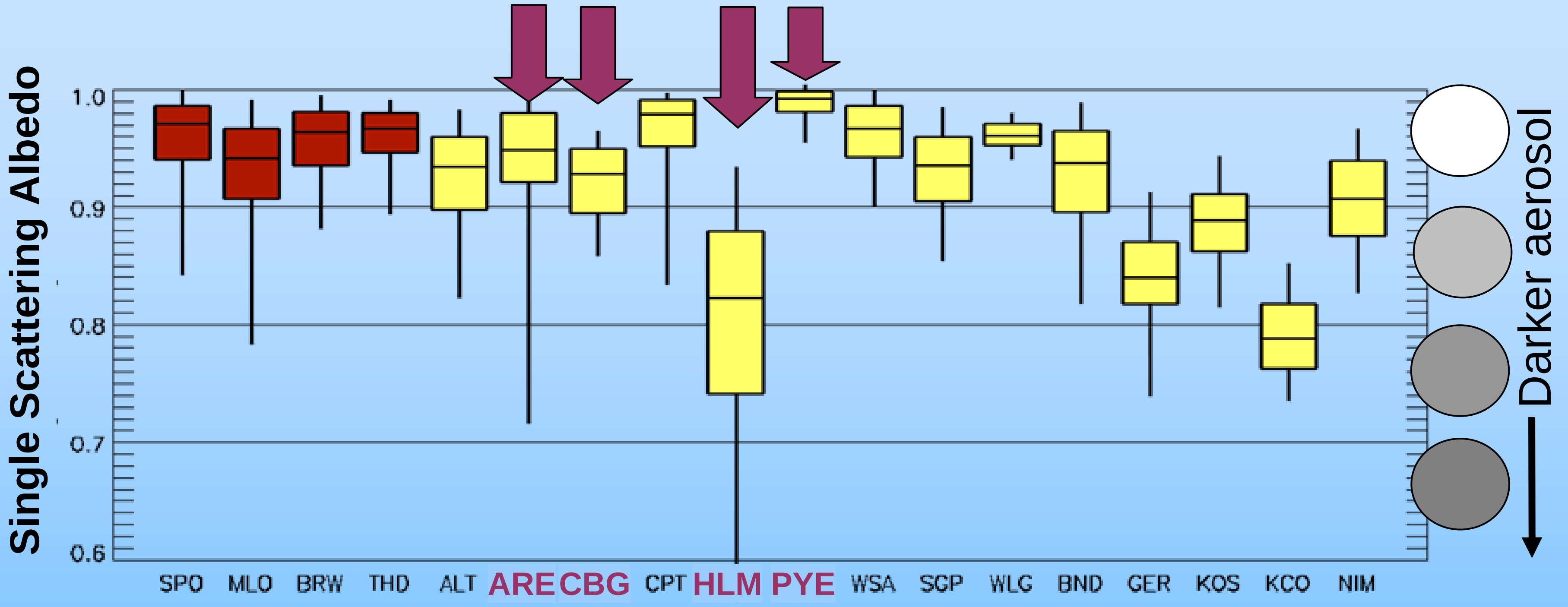


Single Scattering Albedo



Site	Campaign	When	Aerosol type
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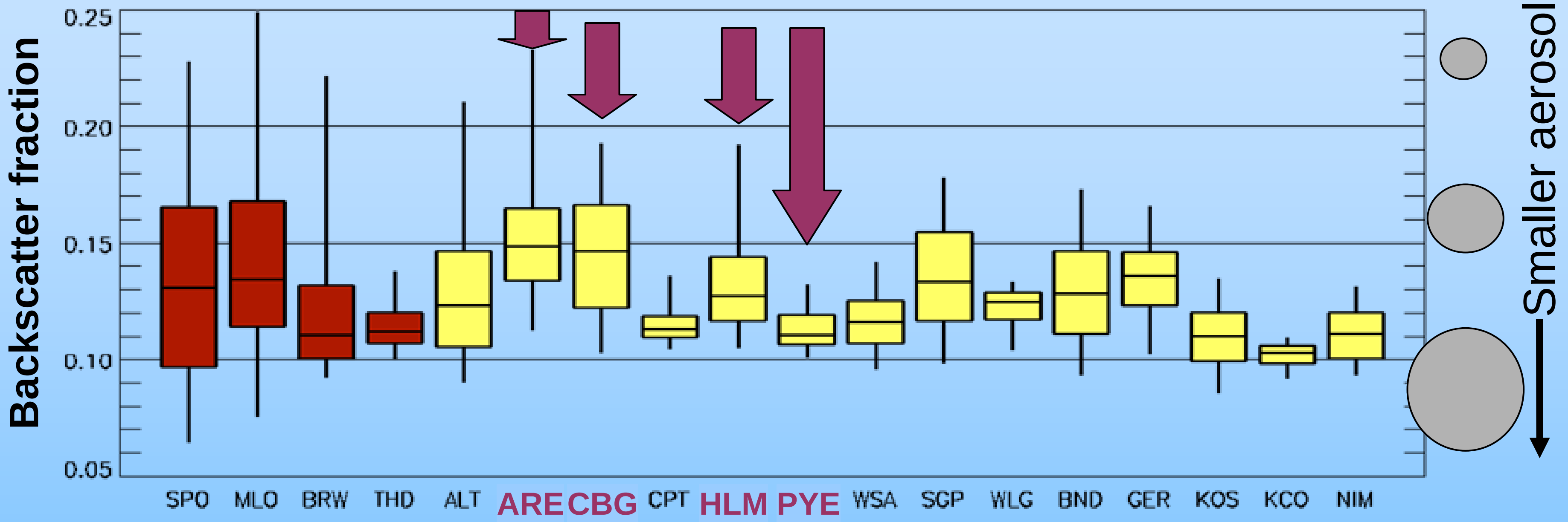
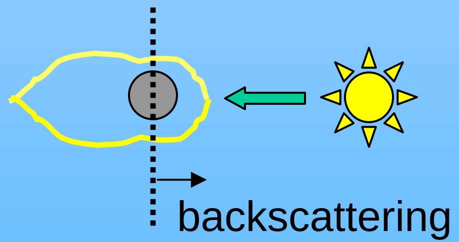
CBG	ICARTT	Summer, 2004	Aged urban
HLM	n/a	Winter, 2006	Aged urban, biomass burning
PYE	MASRAD	Summer, 2005	Clean marine

Backscatter fraction



Backscattering fraction = backscattering / total scattering



Backscatter fraction can be used to estimate other parameterizations of angular dependence of light scattering, e.g., upscatter fraction and asymmetry parameter.