# Using long-term atmospheric aerosol optical measurements to study climate


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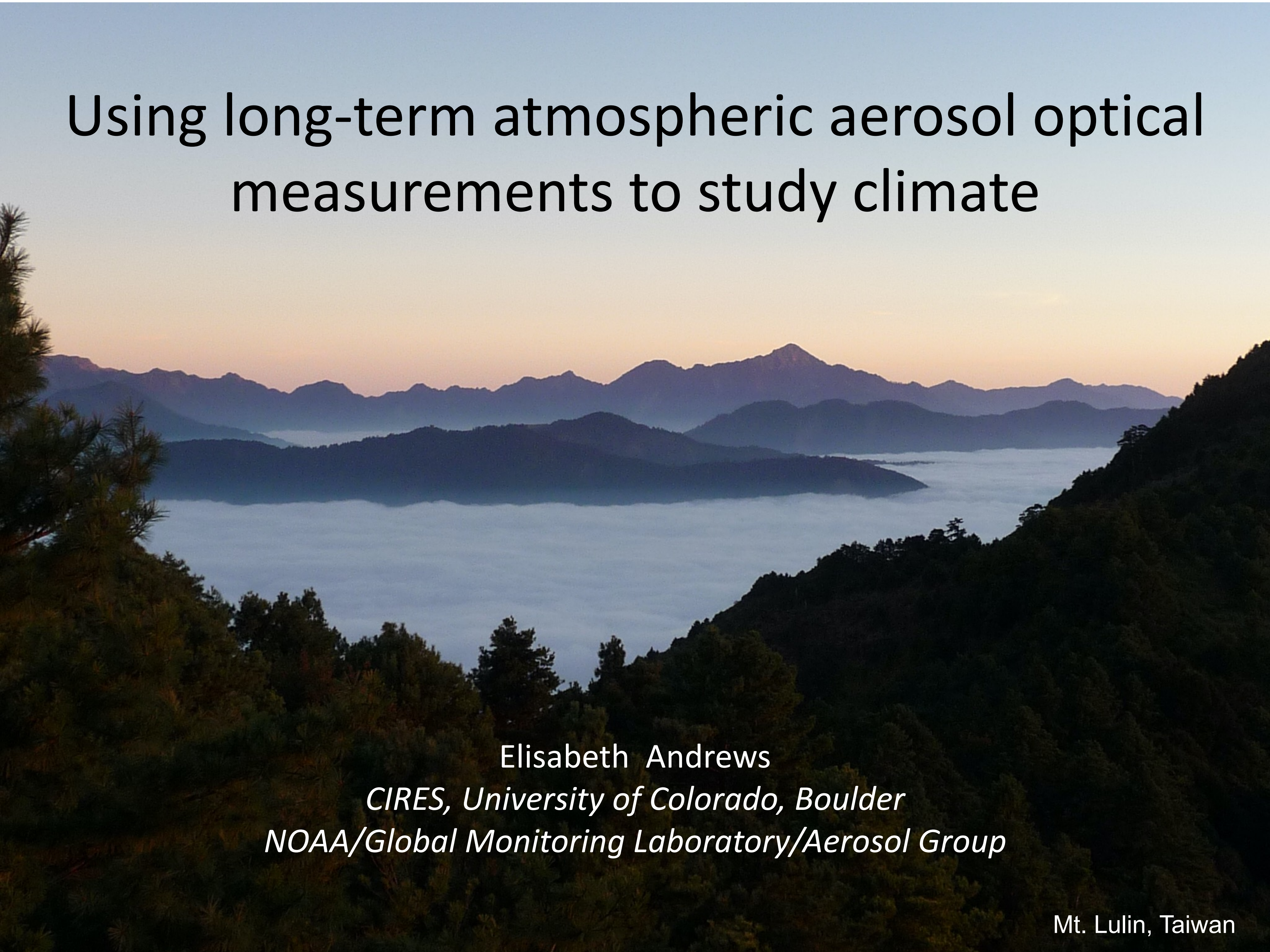


Mt. Lulin, Taiwan

**Measurement data**

J. Ogren, A. Jefferson, P. Sheridan, D. Hageman, J. Wendell, L. Schmeisser, S. Sharma, J., G. Loeschau, J. Sherman, P. Tunved, M. Sorribas, I. Kalapov, W. Aas, P. Tunved, N. Bukowiecki, W. Birmili, B. Henzing, M. Keywood, J. Nicolas, A. Marinoni, O. Mayol, C. Labuschagne, K. Eleftheriadis, Y. Toh, G. Kouvarakis, S. Kim, H. Flentje, J. Backman, J.-P. Putaud, S. Rodriguez, E. Cuevas, A. Hoffer, A. Wiedensohler, N-H. Lin, C. O'Dowd, A. Alastuey, M. Pandolfi, M. Aalo, R. Weller,K. Sellegri, V. Dudoitis, P. Bonasoni, J. Sciare, A.G.Hallar, E. Asmi, M. Fiebig, L. Arbolades, G. Titos, K. Stenstrom, J. Sun, L. Ries

**AeroCom coordination:**

M. Schulz, Jan Griesfelder

**Model data:**

T. van Noije (TM5),G. Curci (GeosChem), K. Zhang (CAM5), H. Kokkola (ECHAM-SALSA), H. Bian (GEOS5-Globase), V. Buchard (GEOS5-MERRAero), G. Myhre (OsloCTM2), M.Chin (GOCART), P. Stier (MPIHAM), T. Takemura (SPRINTARS), S. Tsyro (EMEP), A. Kirkevåg (OSLO-CAM5.3),

## Talk outline

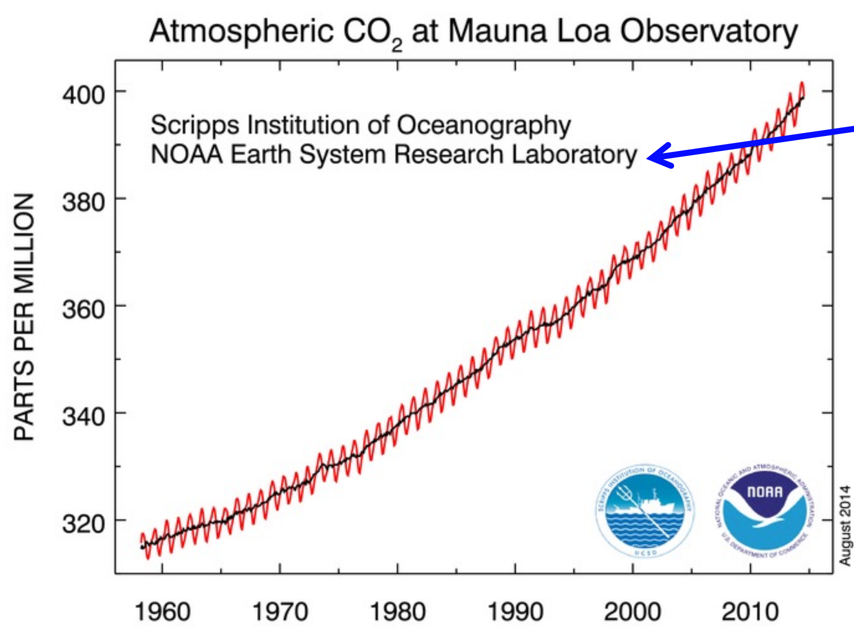
- Climate and NOAA/GML and NOAA's Federated Aerosol Network
- Instruments and 'Black Carbon'
- Studies using data from the NOAA Federated Aerosol Network

# NOAA Global Monitoring Laboratory (GML)

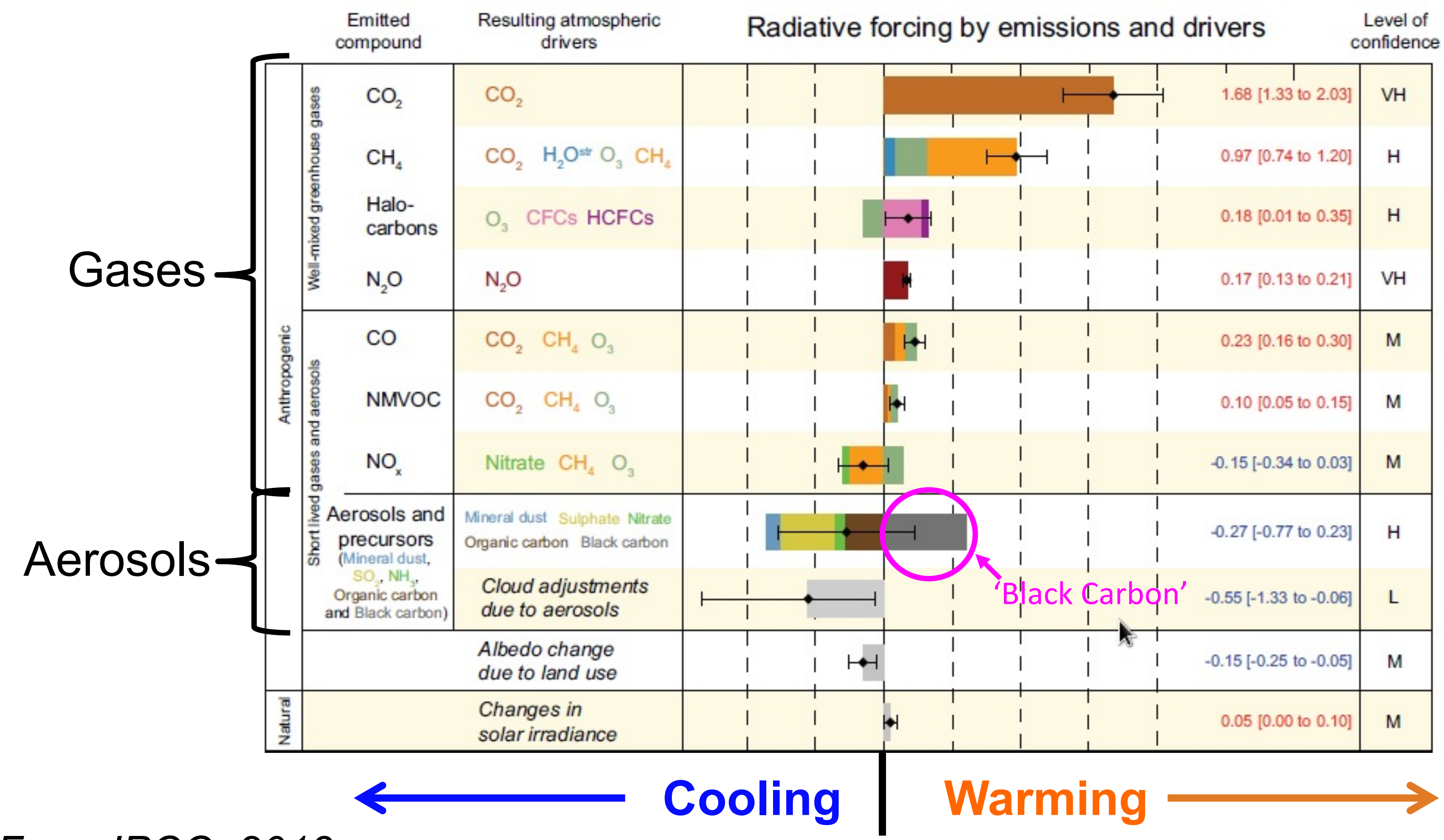
{Established 1973}

**Objective:** Long term measurements of background atmospheric constituents →climate change, ozone depletion and air quality.

**Measurements:**
- Greenhouse gases
- Ozone/ozone-depleting gases
- Solar/thermal radiation
- Aerosols




# Factors influencing climate change



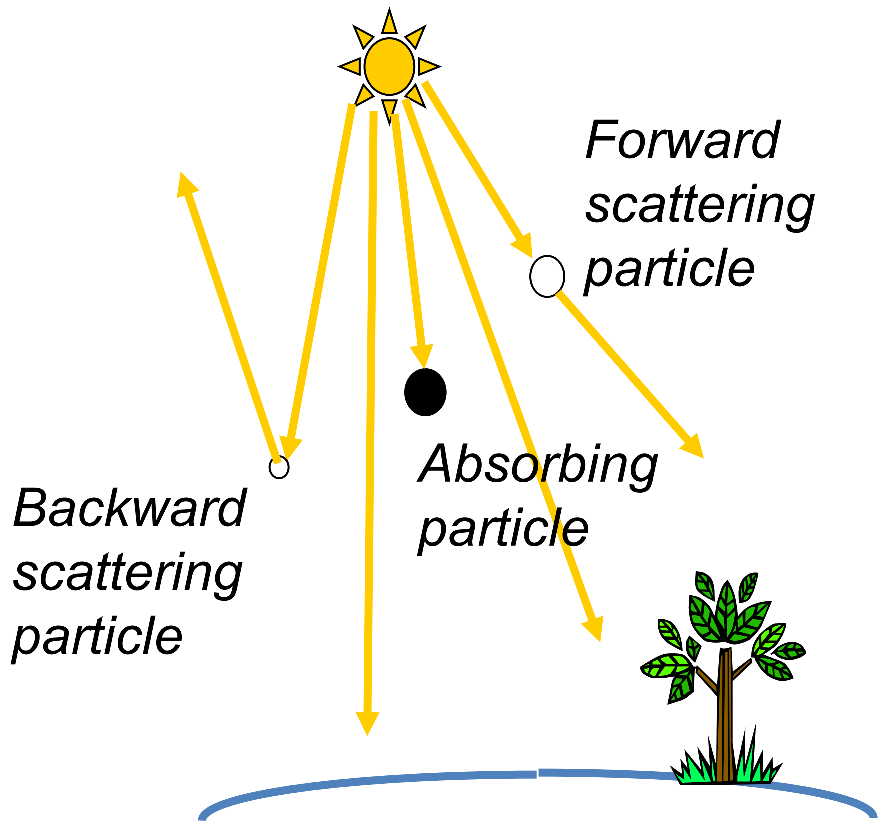

GML
GHG
Ozone
Radiation
Aerosols

*From IPCC, 2013*

Global averages based on models, **measurements** and theory.
Aerosols contribute the largest uncertainty to the total radiative forcing estimate.
'Black Carbon' is only aerosol with a warming effect.

# Direct Aerosol Effect on Climate

- *Surface cooling:* sunlight is prevented from reaching the Earth's surface
- *Atmospheric warming:* energy is transferred as heat by absorbing particles.




Aerosol particles can also indirectly affect climate through clouds.
Aerosol particles impact air quality (health, aesthetics, …).

# NOAA/GML Aerosol Group

## Global Monitoring Laboratory

Earth System Research Laboratories

Search GML...

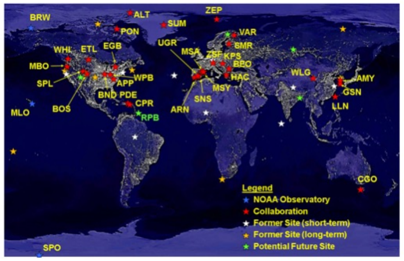


### Information



## NOAA Federated Aerosol Network (NFAN)

The NOAA Federated Aerosol Network (NFAN) monitors surface in-situ aerosol optical properties at field sites around the world. At a subset of NFAN sites, aerosol microphysical and chemical properties are also measured in collaboration with partner organizations.

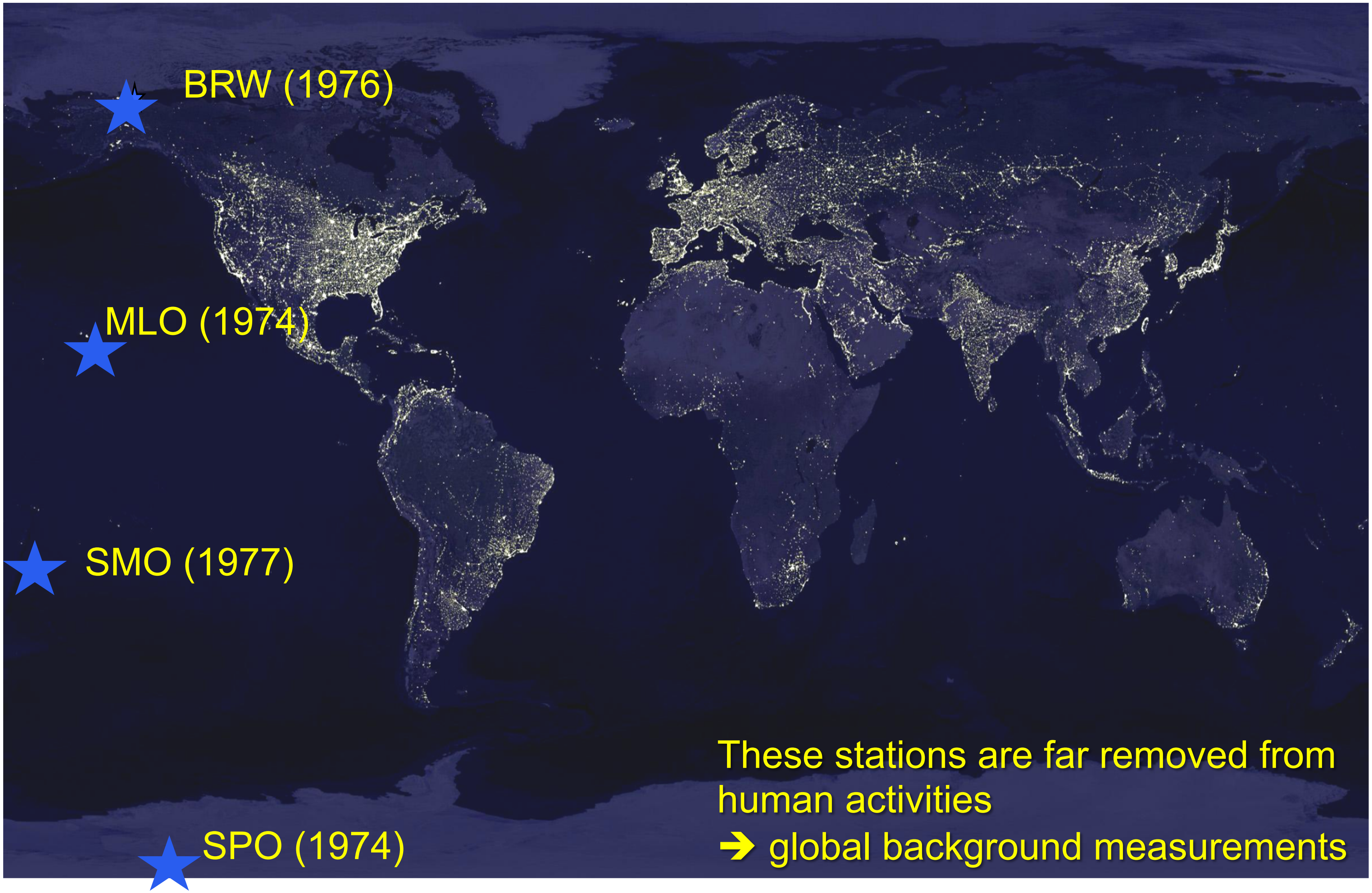



For an overview and information about the NFAN see:

Andrews, E., Sheridan, P.J., Ogren, J.A. et al. (2019), "Overview of the NOAA/ESRL Federated Aerosol Network" *Bulletin of the American Meteorological Society*, **100**,123-135, 2019.

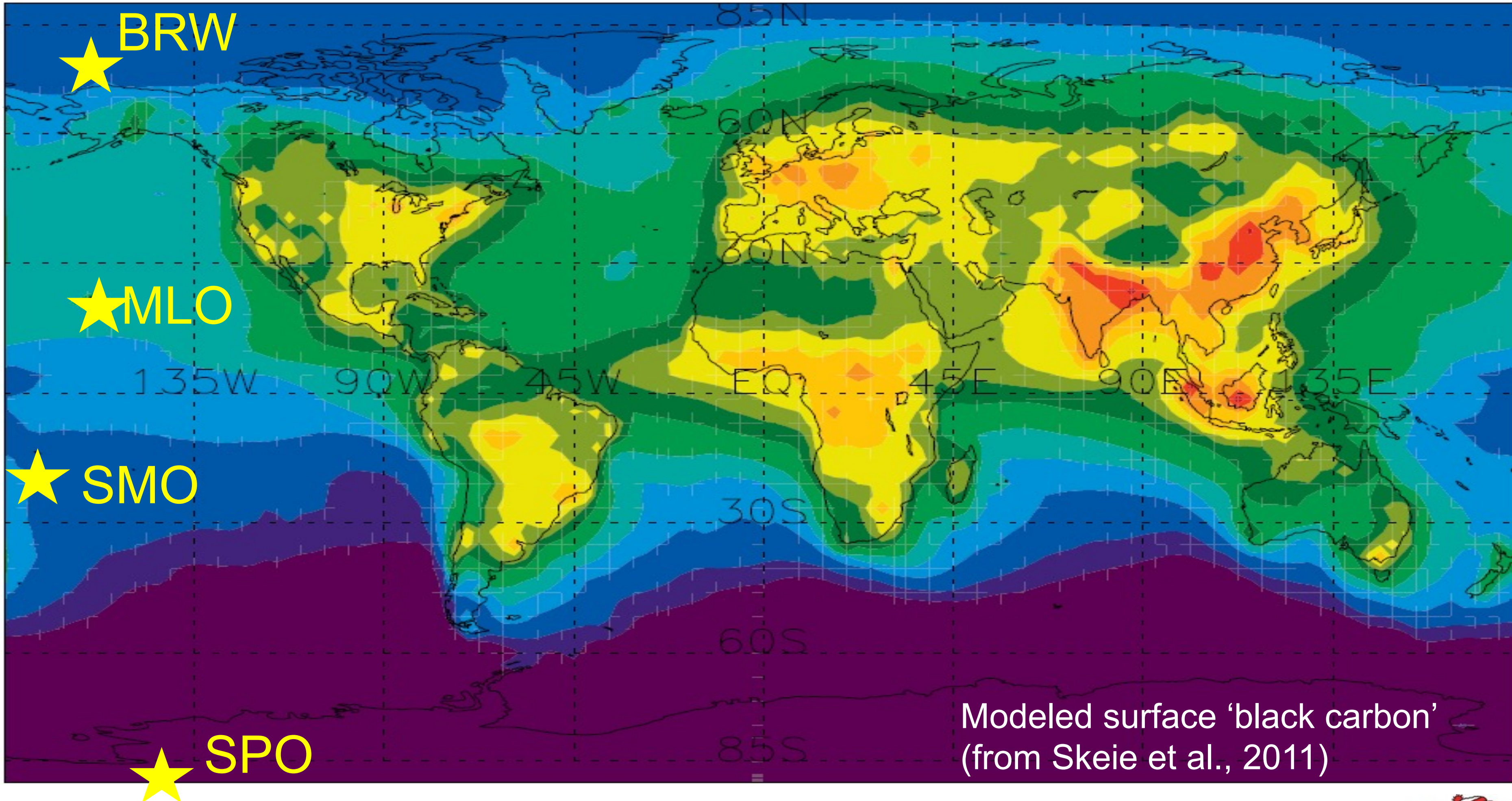
https://gml.noaa.gov/aero/

# GML's Original 4 'baseline' stations, 1970s-1990s




But…aerosol/climate effects happen where the particles are…

# Where’s the aerosol?

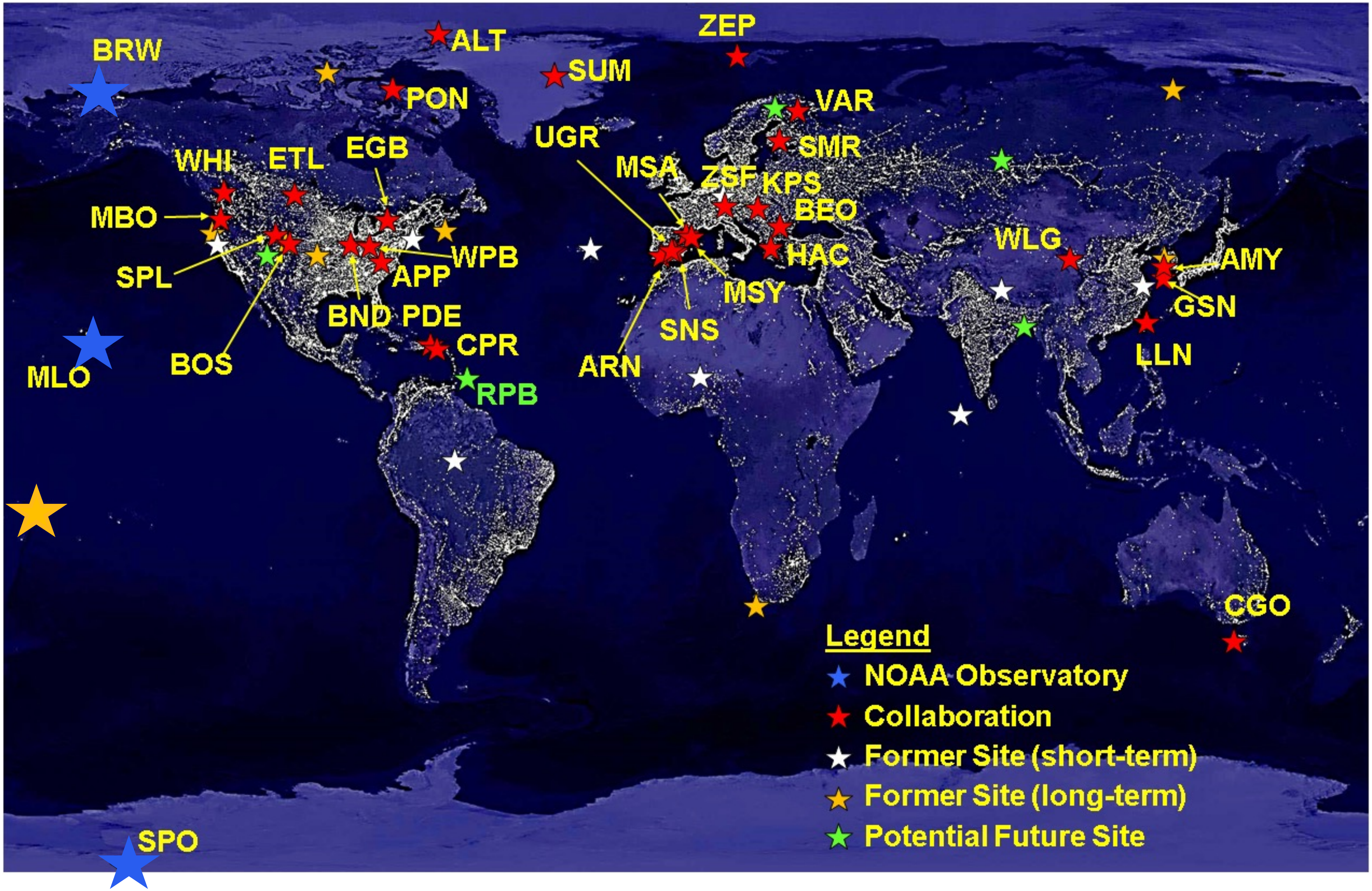



Modeled surface ‘black carbon’
(from Skeie et al., 2011)

→Not at NOAA’s baseline stations
Aerosol distribution is patchy due to source distribution and short residence times (order of 1 week)

# 40+ years later….

# NOAA Federated Aerosol Network, 2022




Currently 34 active sites, in wide variety of places
Lots of global gaps

# NOAA/GML/Aerosol Group

**Objective:**

- Characterize the means, variabilities, and trends of climate-forcing properties of different types of aerosols
- To understand the factors that control these properties.

**Our approach:**

→Collaborate collaborate collaborate!

→Standardized suite of measurements and protocols

→Standardized software

→Long-term permanent and shorter-term “mobile” sites

→Globally distributed network (pristine and polluted sites)

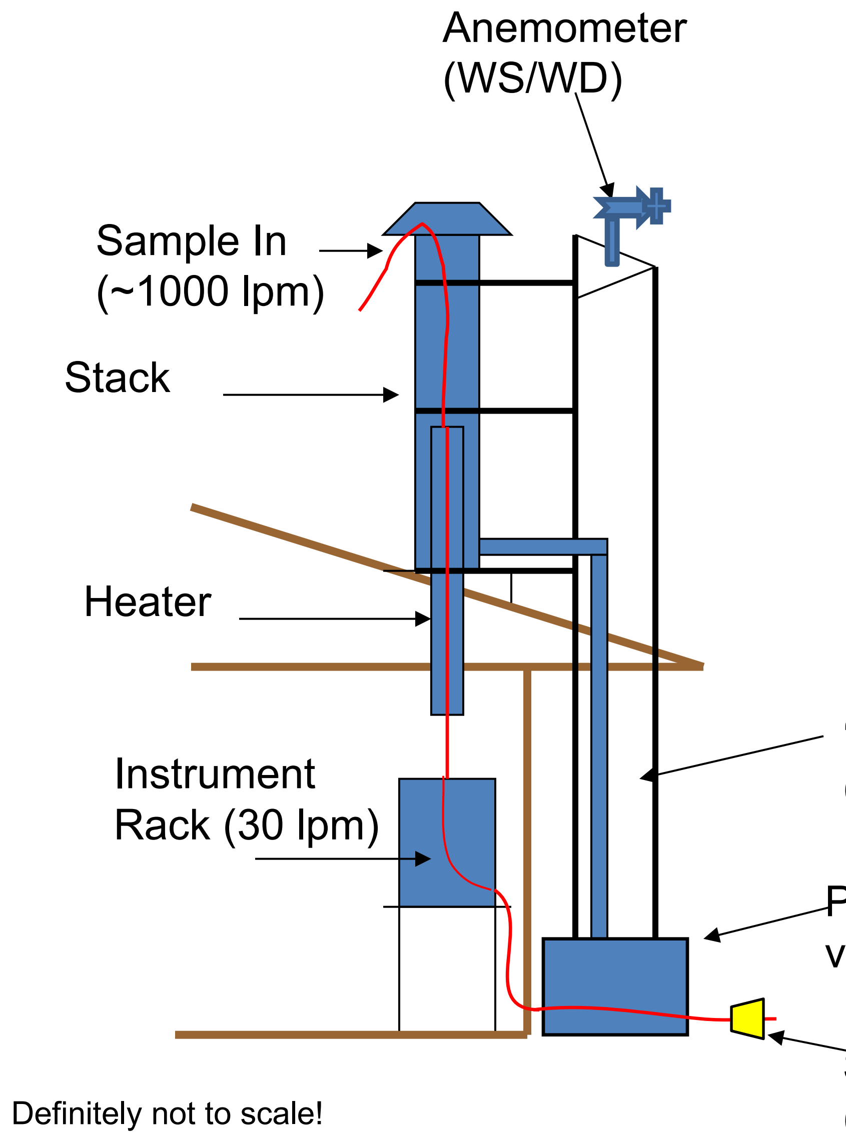

**Some additional applications:**

- Context for field campaigns
- Ground truth for remote sensing (e.g., satellites)
- Evaluate/constrain models

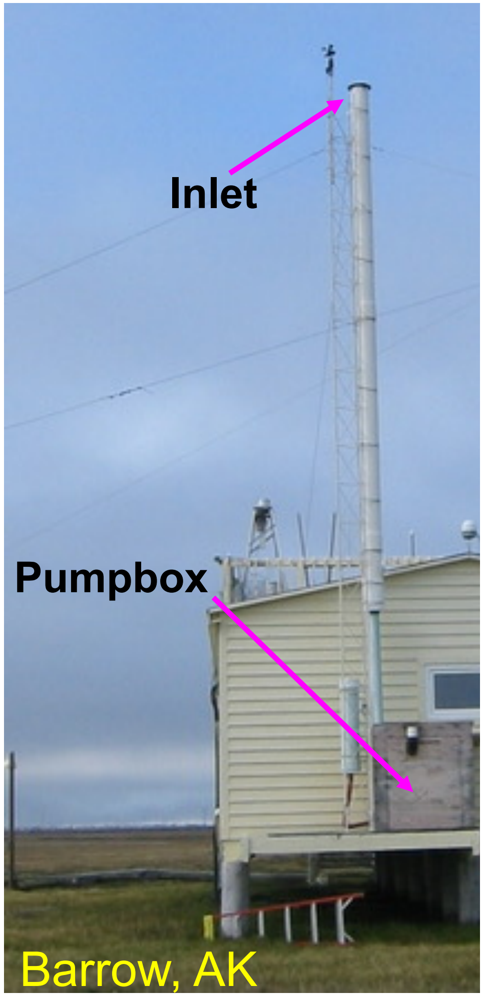
# In-situ surface Aerosol Sampling

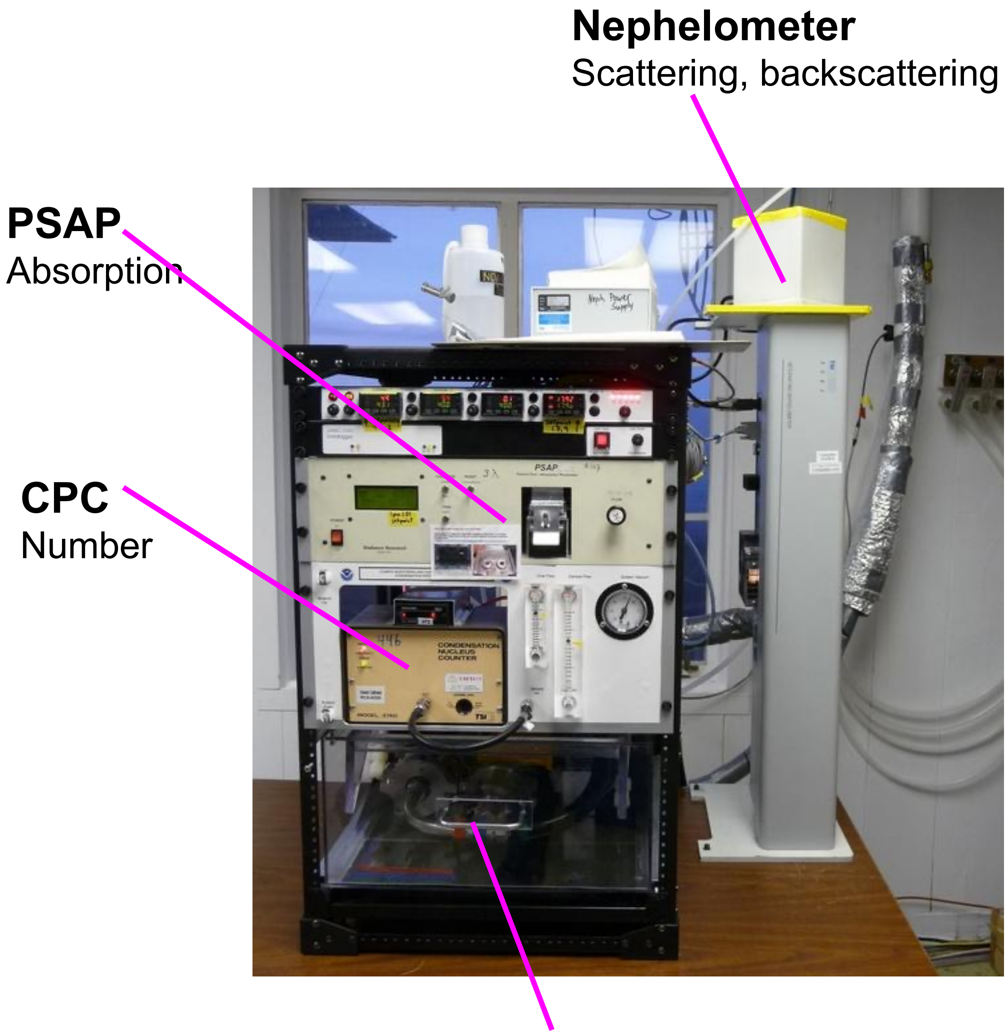
→Inlet design is important

- 10 meter high stack with rain hat
- Super-isokinetic→ particle losses are minimized (except ultrafines)
- Sample humidity <40% (heat and/or dilute)
- Pumps pull air through the stack and instruments




# In-situ surface Aerosol Sampling



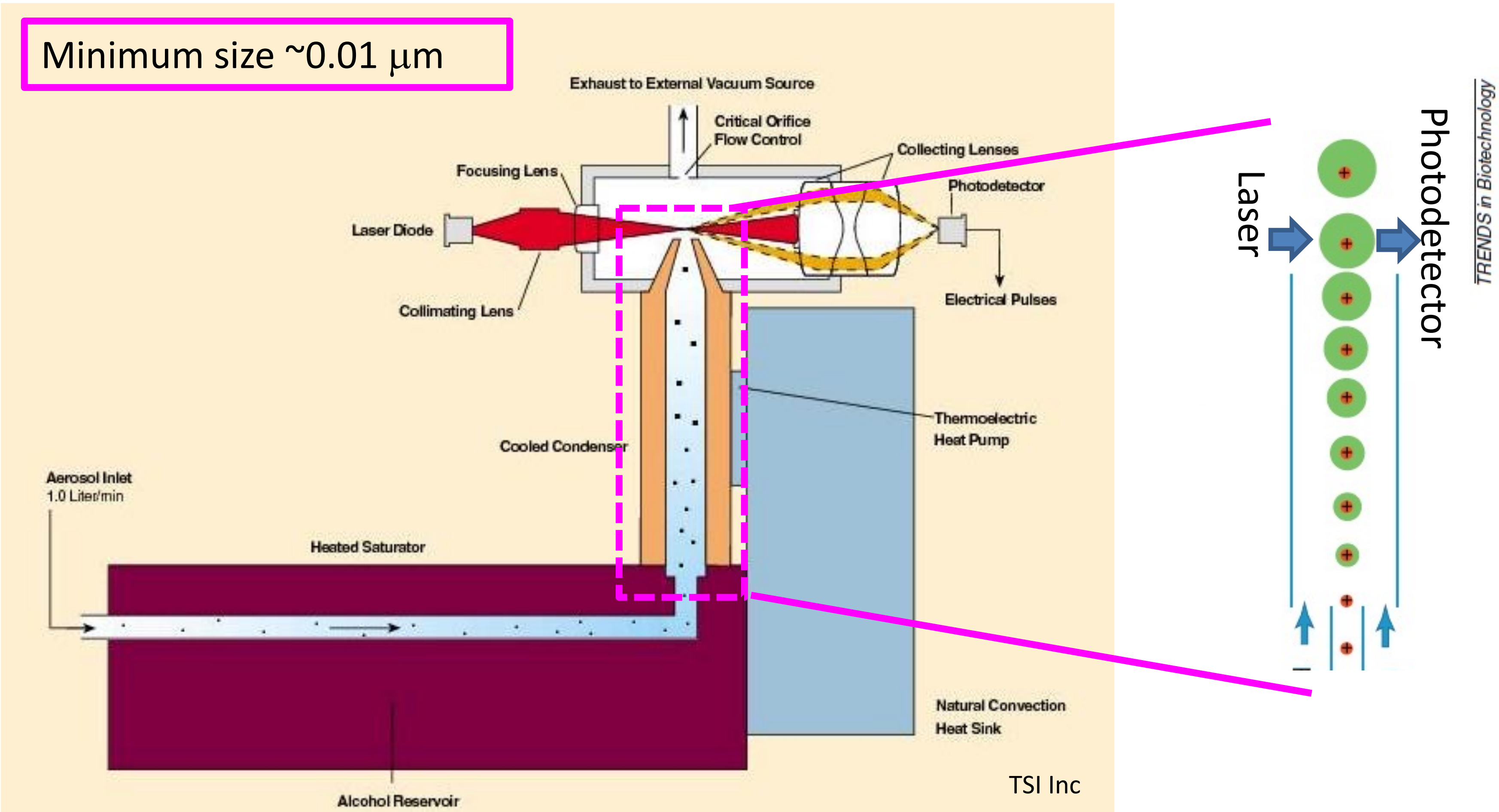




Measurements made at low RH (<40%) and 1 min frequency

# Aerosol Number Concentration

Condensation Particle Counter (CPC)

Minimum size ~0.01 μm

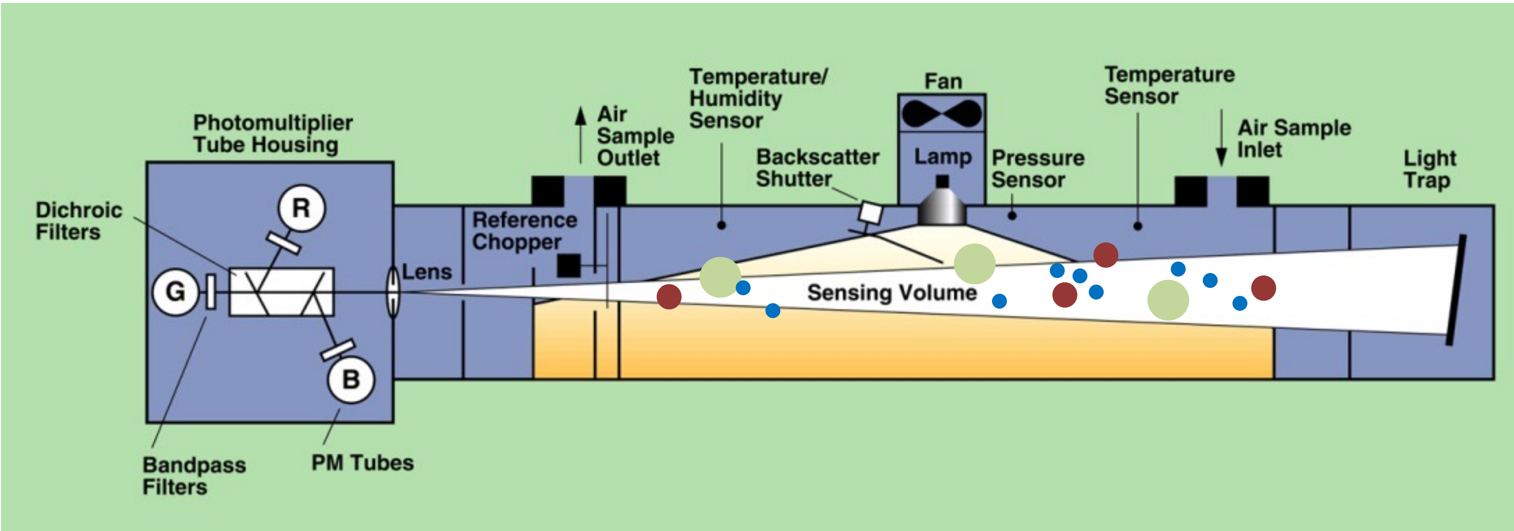



→Particles exposed to supersaturated vapor

→Particles grow to size that can be optically detected

Minimum detection size related to instrument temperatures/supersaturations

# Aerosol Optical Properties - Scattering



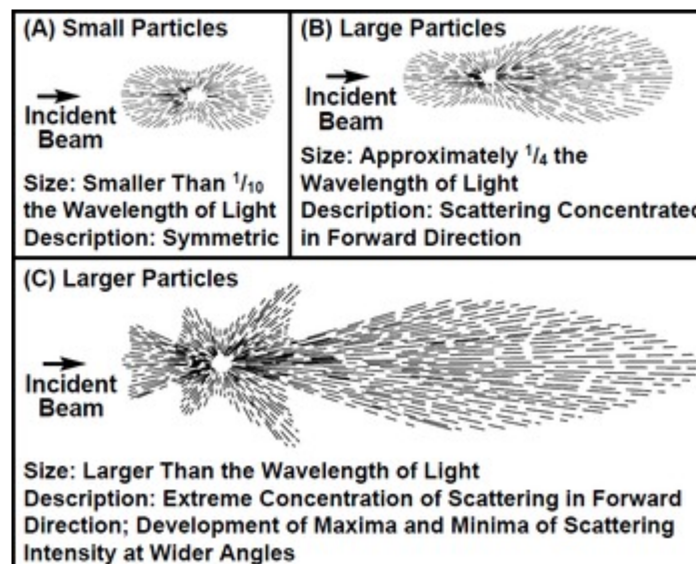

## Nephelometer

→particles scatter light in sensing volume
→photomultiplier tubes detect scattered light

Common options:

- Multiple wavelength
- Angular dependence of scattering

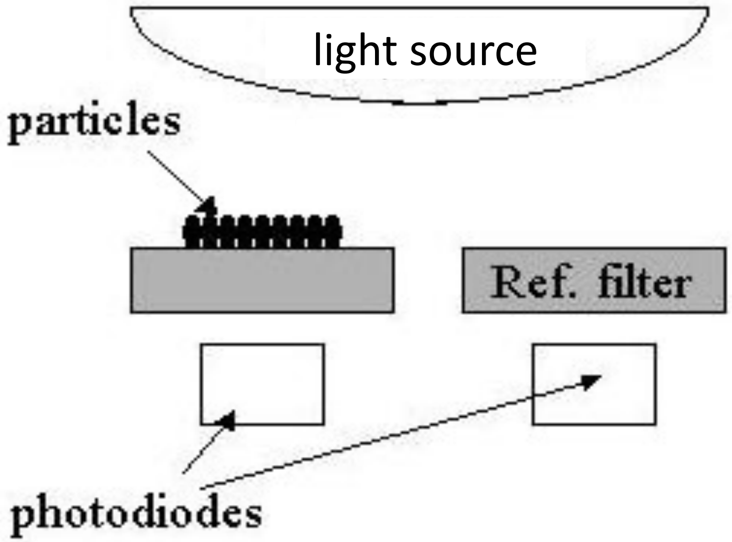
Calibrated with gas (e.g., $CO_2$)
Needs correction for angular non-idealities

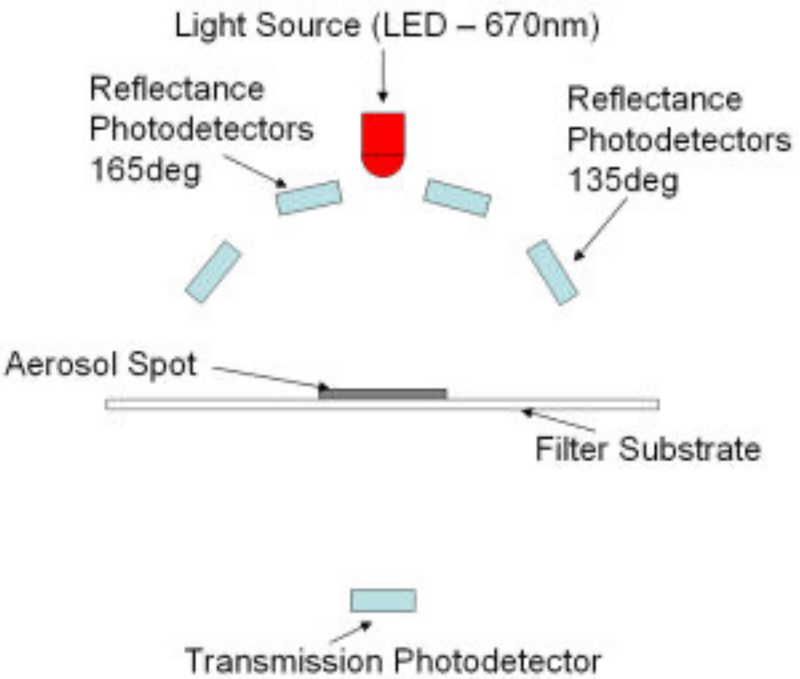


# Aerosol Optical Properties - Absorption


Filter-based absorption measurements:

- PSAP – Particle Soot Absorption Photometer
- TAP – Tri-color Absorption Photometer
- Aethalometer
- MAAP – Multi-Angle Absorption Photometer




Needs correction for attenuation due to non-absorbing particles

# NOAA-designed absorption instrument

Continuous Light Absorption Photometer (CLAP)
→ 8 spots on filter, spot automatically advances at set transmittance
→ Operates 8x longer than PSAP (1 filter spot)
→ ~1/5 size of PSAP (1/12 size of aethalometer or MAAP)
→ No moving parts (no tape reels), but lots of fiddly solenoid valves
→ Now manufactured by Brechtel Inc. as Tricolor absorption photometer (TAP)

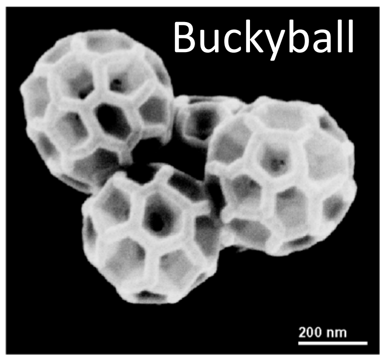

Ogren, J.A. et al.,"Continuous Light Absorption Photometer for Long-Term Studies," Atmos Meas. Tech., 2017.

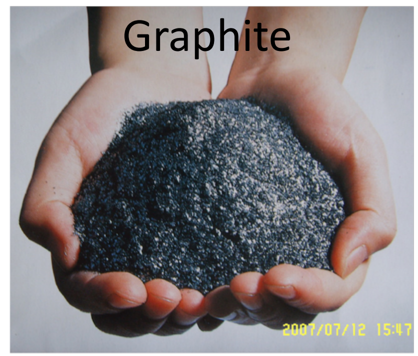
# Light absorption vs. 'Black Carbon'

**'BC' is poorly defined in the scientific literature**

- Carbonaceous matter doesn't appear in atmosphere as a pure substance
- Measurements may refer to same quantity with different names or different quantities with same name (BC, EC, soot, …)
- Models may use emissions based on one analytical method and verify with observations using different analytical method (apples and oranges/diamonds and soot!)

Examples of pure elemental carbon:

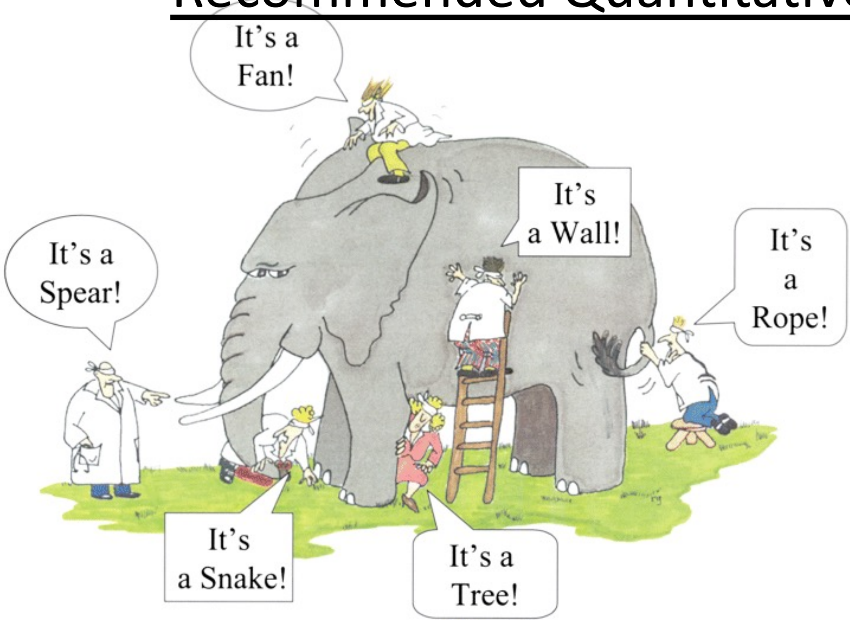



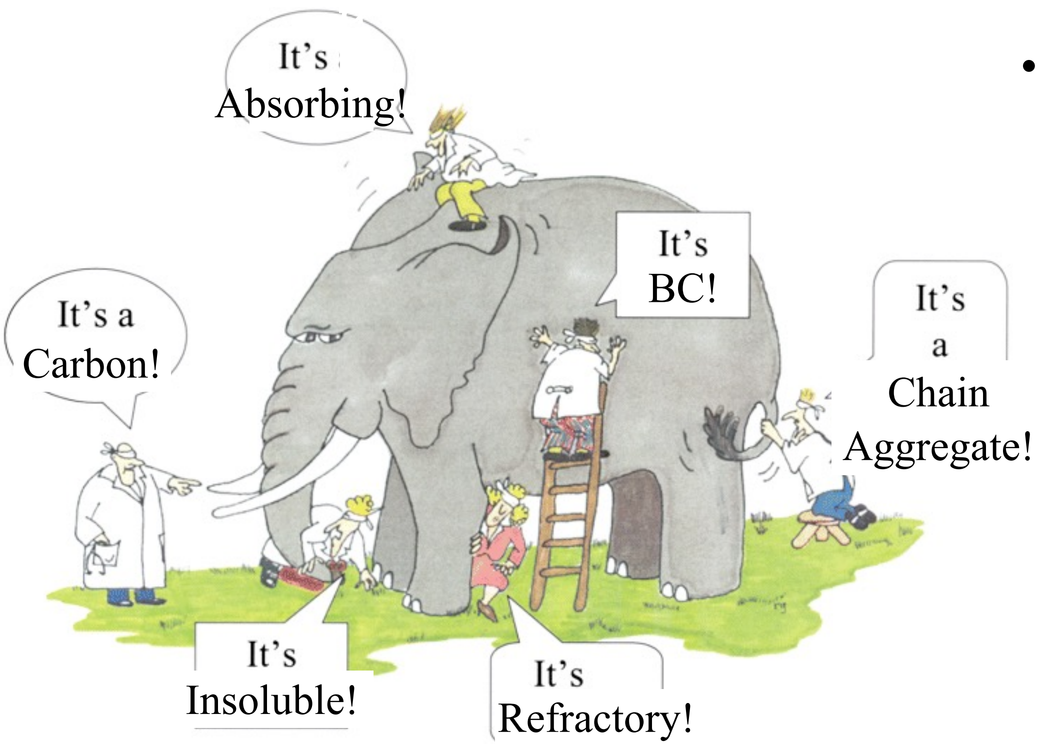

*"None of these pure forms are found in the atmosphere (or aerosol scientists would be considerably wealthier)."*
--Bond&Bergstrom 2006

**'BC' measurements depend on method used**

- Methods respond to different properties of BC
  - Insoluble in water
  - Refractory – thermally stable
  - Strongly **absorbs** light across all visible wavelengths
- Correlations between measurement methods tend to be high, but correlations can vary depending on site, season, aerosol type….

# Recommended Quantitative Terminology (Petzold et al., 2014)

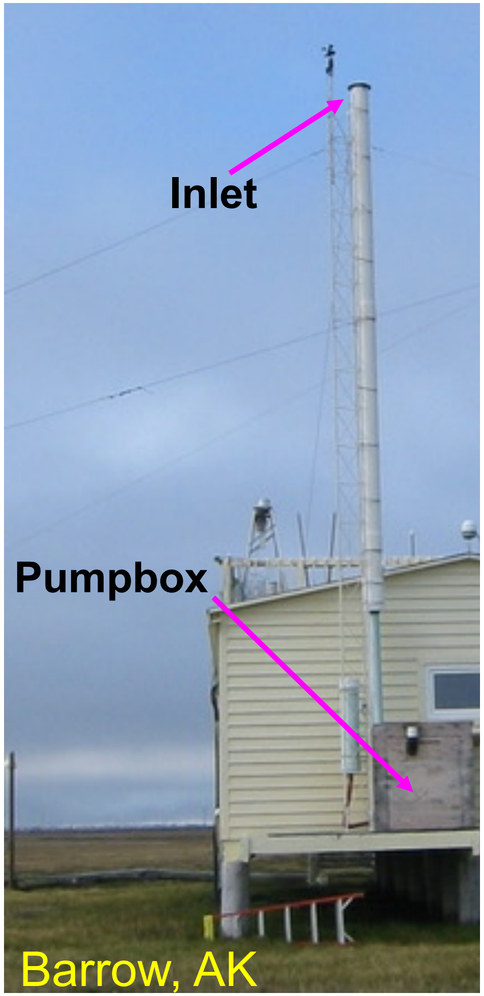



- **Light Absorption Coefficient ($\sigma_{ap}$) or EBC**
  - Derived from optical methods, e.g.,
    - Filter-based (e.g., PSAP)
    - Suspended particles (e.g., photo-acoustic)
- **Elemental Carbon (EC)**
  - Derived from measurement of $CO_2$ evolved from thermal or thermo-optical methods
- **Refractory Black Carbon (rBC)**
  - Derived from laser incandescence methods

→What you measure depends on how you measure it!

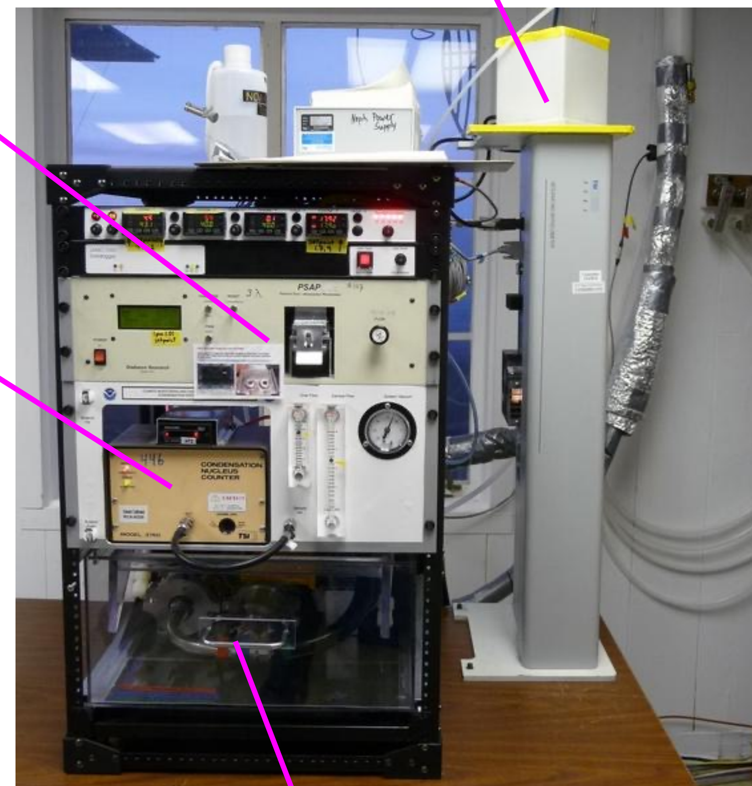
# In-situ surface Aerosol Sampling




**Nephelometer**
Scattering, backscattering

**PSAP**
Absorption

**CPC**
Number



**Switched Impactors**
Size cut (1 and 10 μm)

Measurements made at low RH (<40%) and 1 min frequency

# NFAN Measured Parameters

These properties depend on AMOUNT of Aerosol:

--Number concentration, $N_{CN}$

Spectral, optical properties

--Light absorption, $\sigma_{ap}$

--Light scattering, $\sigma_{sp}$

--Back-scattering, $\sigma_{bsp}$

Light extinction, $\sigma_{ext} = \sigma_{sp} + \sigma_{ap}$

Clean Region: 10 $Mm^{-1}$ < Light extinction

Polluted Region: Light extinction > 60 $Mm^{-1}$

South Pole: ~0.5 $Mm^{-1}$
Storm Peak, Colorado: ~10 $Mm^{-1}$
Jeju, South Korea ~ 100 $Mm^{-1}$

Aerosol absorption is typically ~10% of total aerosol extinction (rule of thumb)

# $Mm^{-1}$ - What are these weird units?

Megameter

Visibility → How far one can see (unit of distance)

1 km = 0.001 Mm

Light extinction inversely proportional to visibility →(unit of 1/distance)

$$\sigma_{ext} \sim 3.9/vis$$


220 km visibility = 0.22 Mm


20 km visibility = 0.02 Mm

Rocky Mountain National Park

http://vista.cira.colostate.edu/views/

# Derived aerosol optical properties

- Independent of amount of particles – dimensionless
- Useful for comparing different sites, events
- Used in climate forcing calculations
- Provide information about particle 'nature' (chemistry/microphysics)

Backscatter fraction } *Angular dependence of scattering*

Scattering Ångström exponent } *λ dependence of scattering*

} SIZE

Absorption Ångström exponent } *λ dependence of absorption*

Single scattering albedo } *Relative amount of absorption*

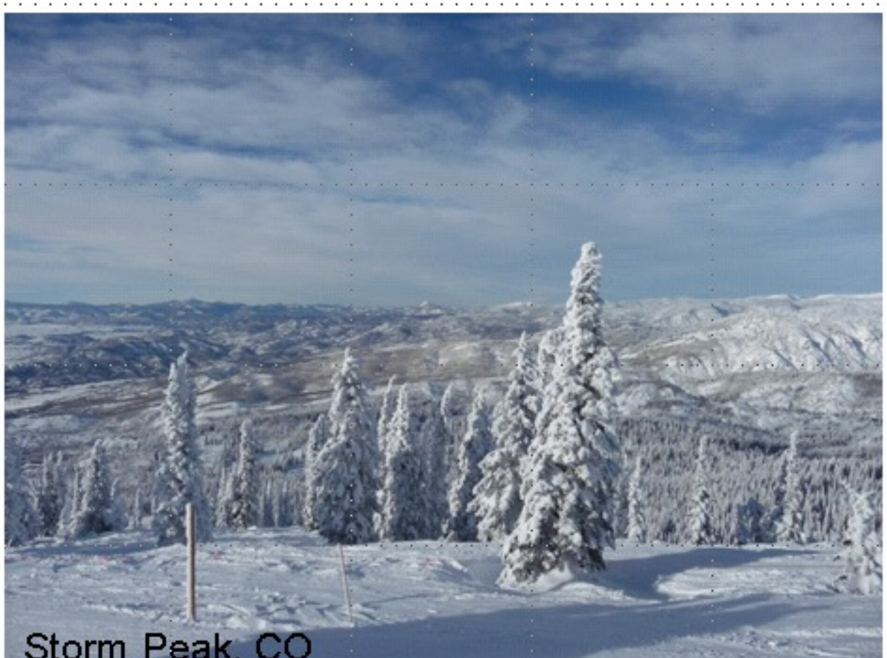
} COMPOSITION

Aerosol Radiative Forcing = f(extinction, backscatter fraction, single scattering albedo)

# It's showtime! (science)

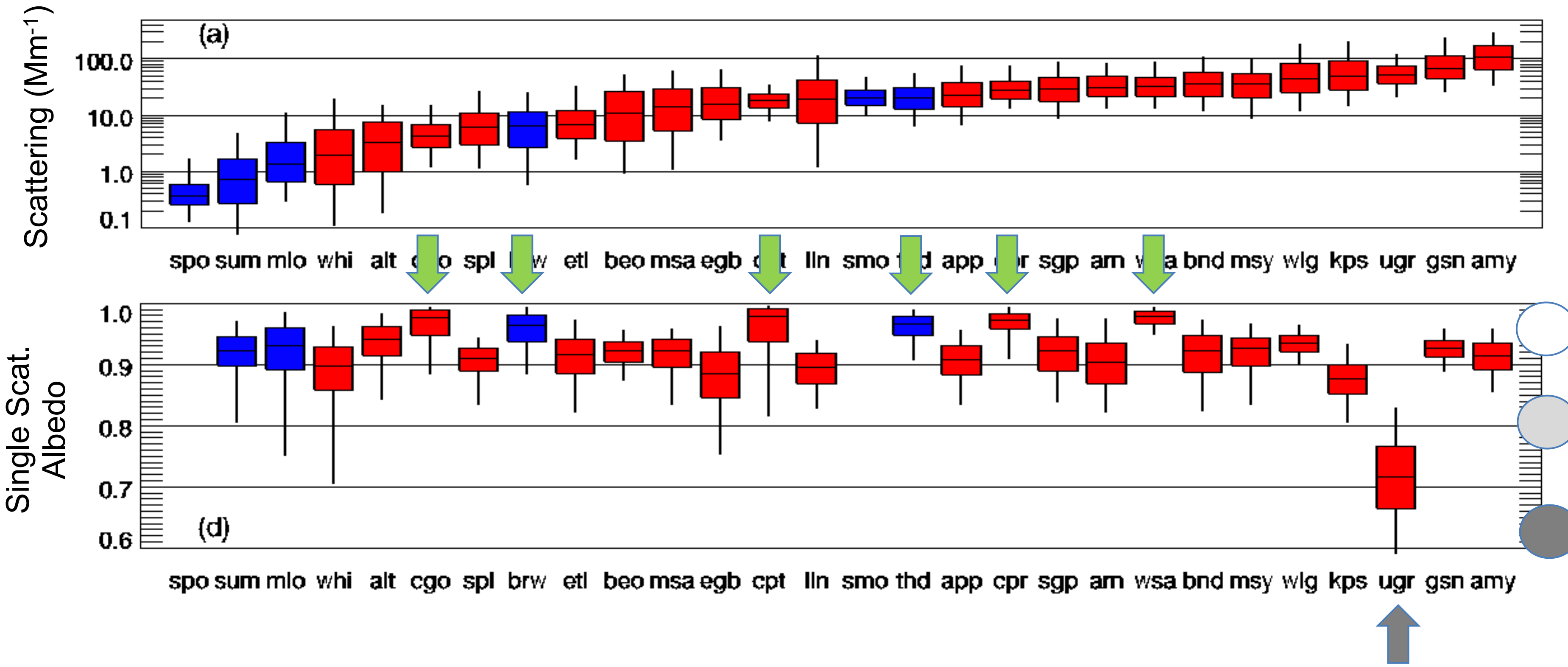
Long term data:
→consistent measurements (instruments and operations)
→good documentation (sampling and instrument changes)
→regionally representative - local contamination identified/removed

- Climatology and trends in aerosol optical properties
- Model-Measurement comparisons
- Satellite validation
- Process studies




# Annual climatology from NOAA Federated Aerosol Network

- Wide range in aerosol amount
- No relationship between amount and “nature” of aerosol




From Andrews et al., 2019

Clean marine sites have highest SSA – dominated by non-absorbing aerosol – sea salt

Granada is impacted by agricultural burning and home heating – low SSA – lots of ‘BC’

In general, SSA tends to be >0.85

# Annual climatology from NOAA Federated Aerosol Network

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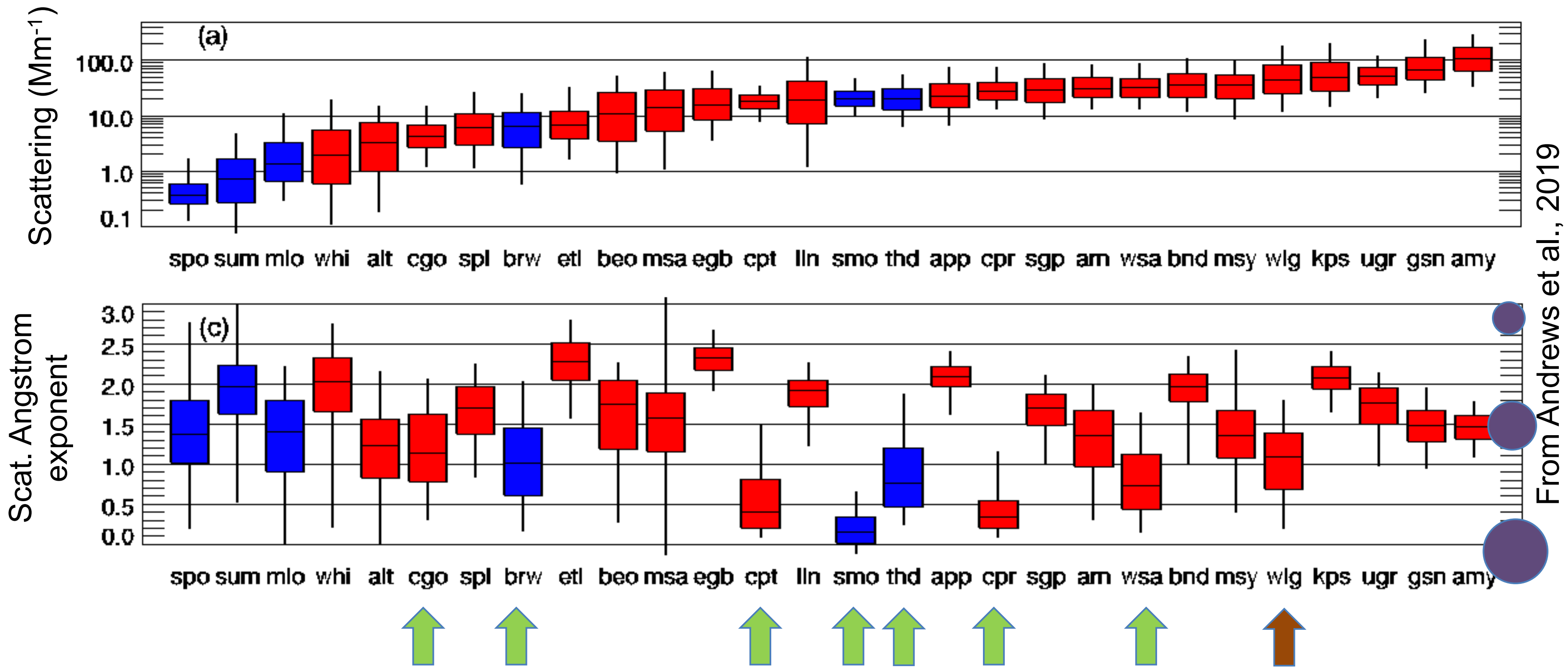
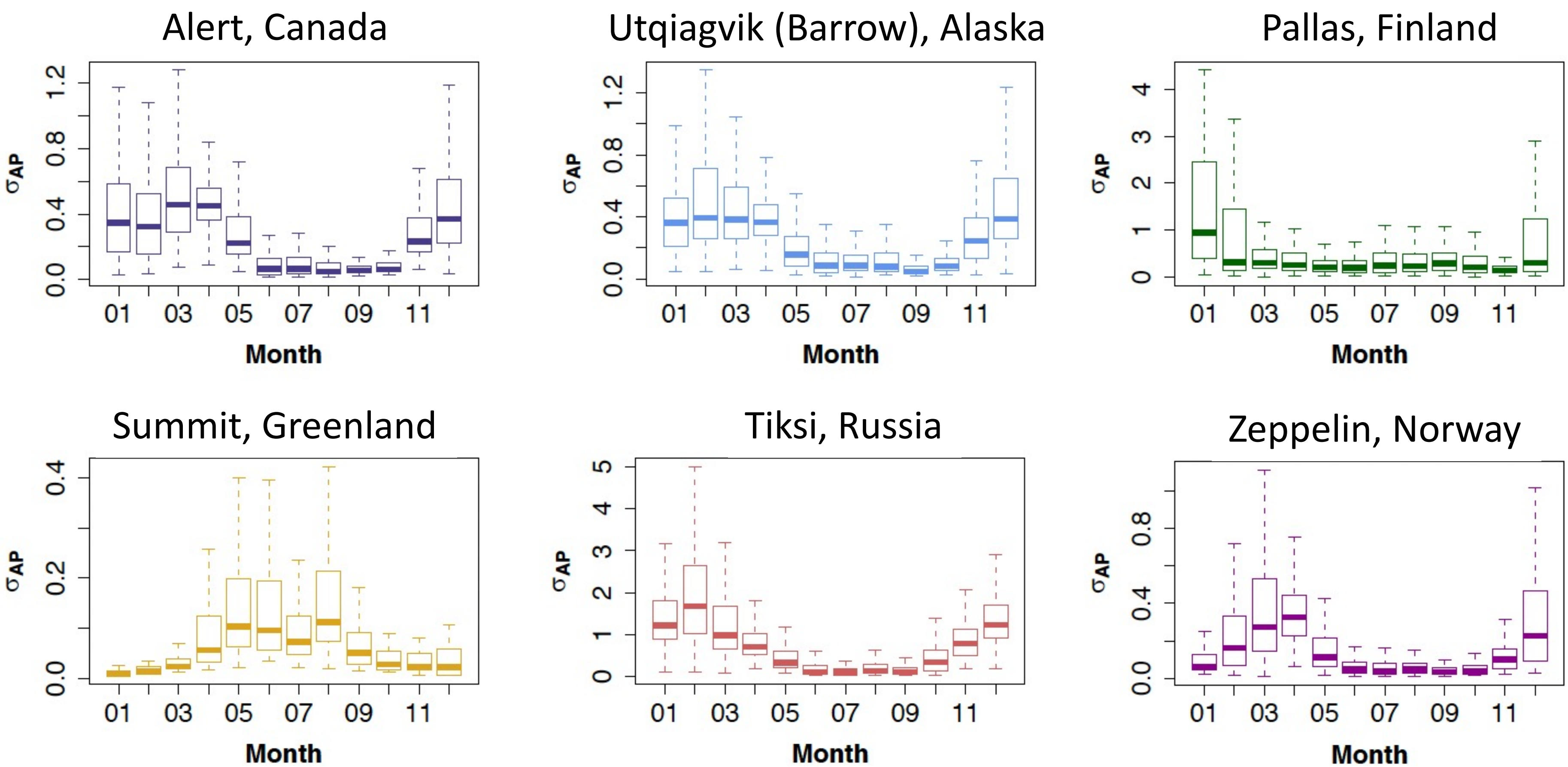



Clean marine sites have lowest Angstrom exponent

Mt Waliguan (China) is impacted by desert dust

# Arctic Aerosol Monthly Climatology



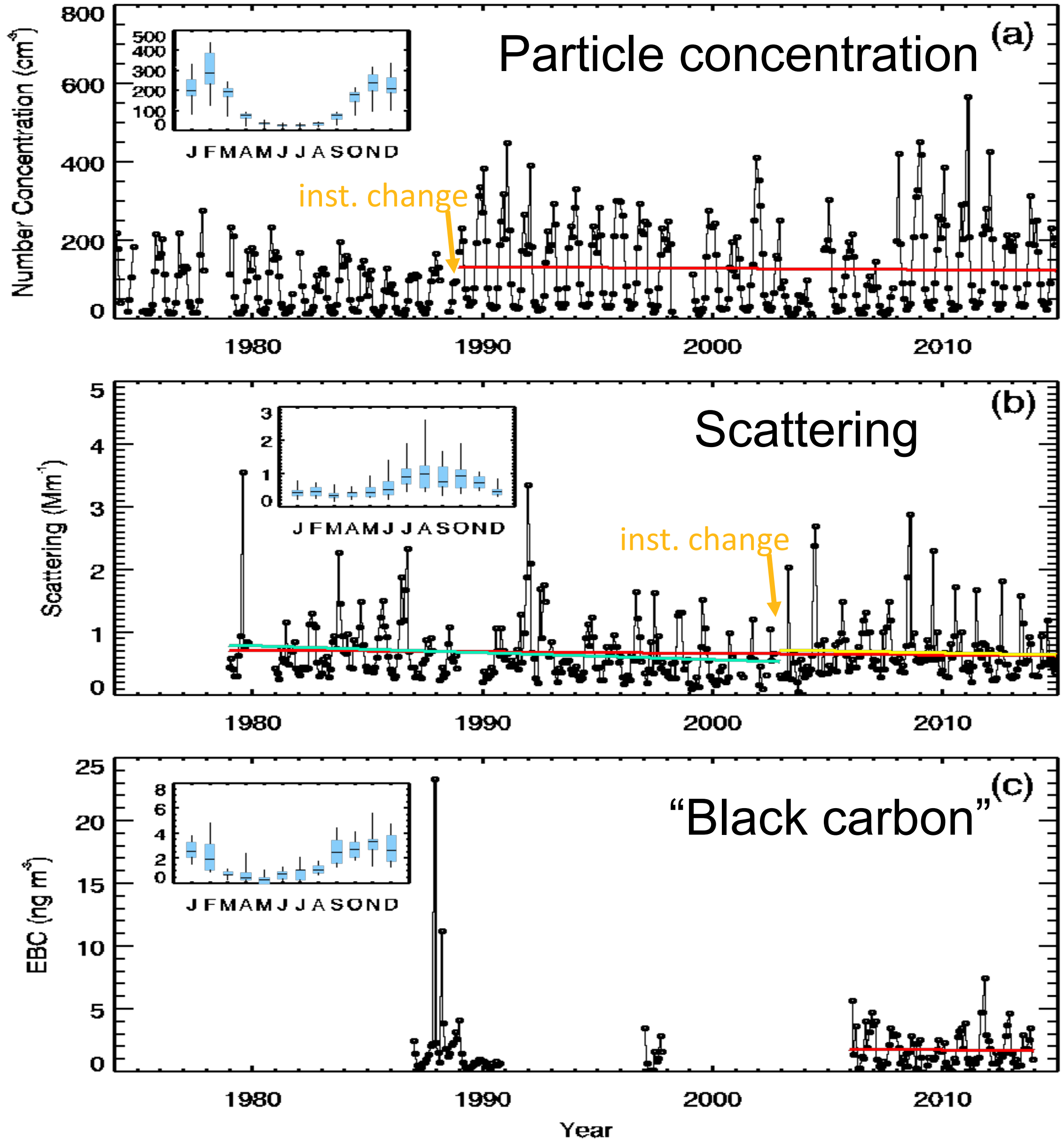

From Schmeisser et al., 2018

Different seasonal cycles and amounts of absorbing aerosol across the Arctic.

→Springtime haze phenomenon is not universal feature of the Arctic

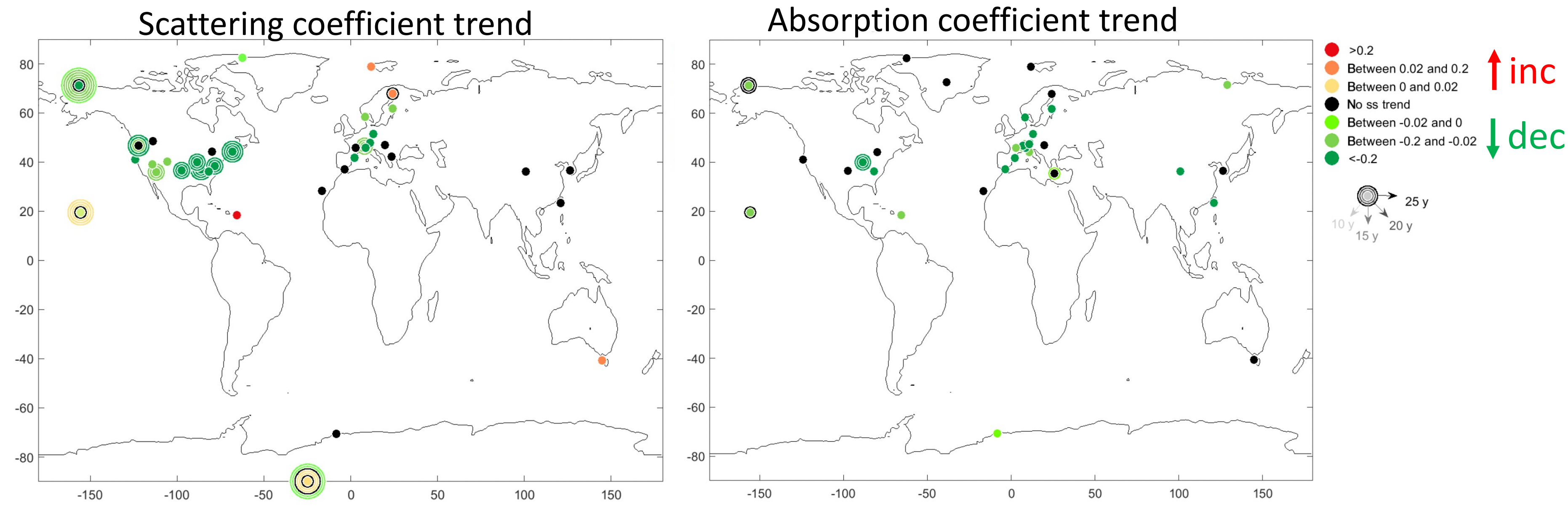
# South Pole Aerosol Climatology: 1974 - 2014

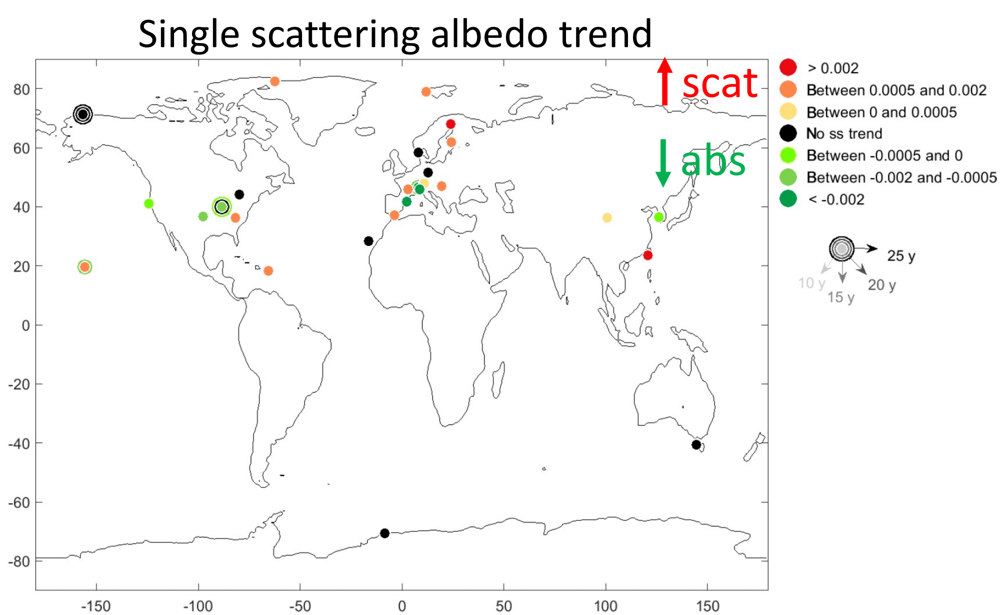
- No statistically significant trends
  → *need to track instrument changes*
- Annual cycles in the different aerosol properties
- Different parameters have different annual cycles → different sources/types of particles??




From Sheridan et al., 2015

# Long-term trends in aerosol properties



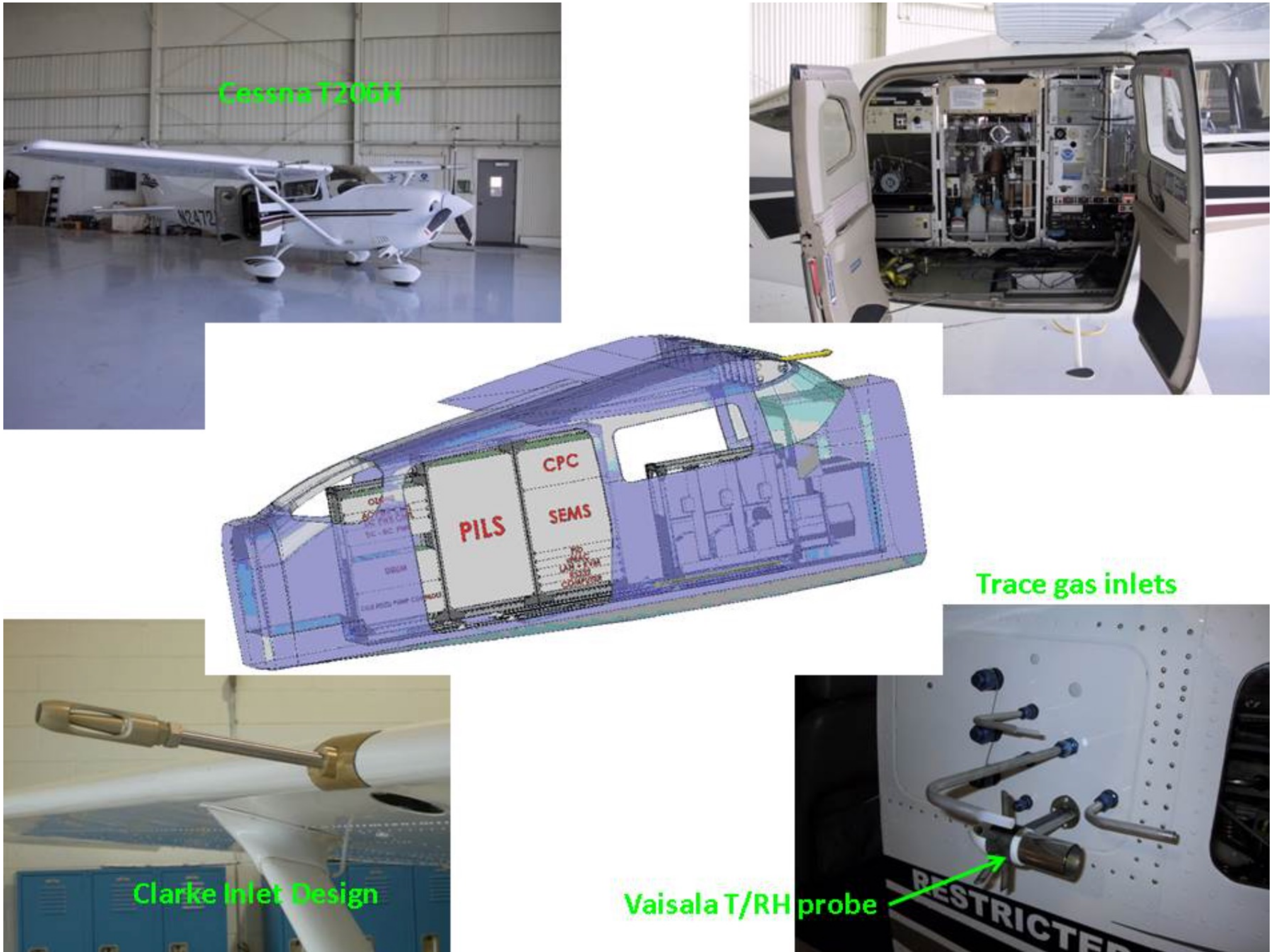

From Collaud Coen et al., 2021

Decreasing trends for scattering and absorption coefficients
→air pollution control policies work

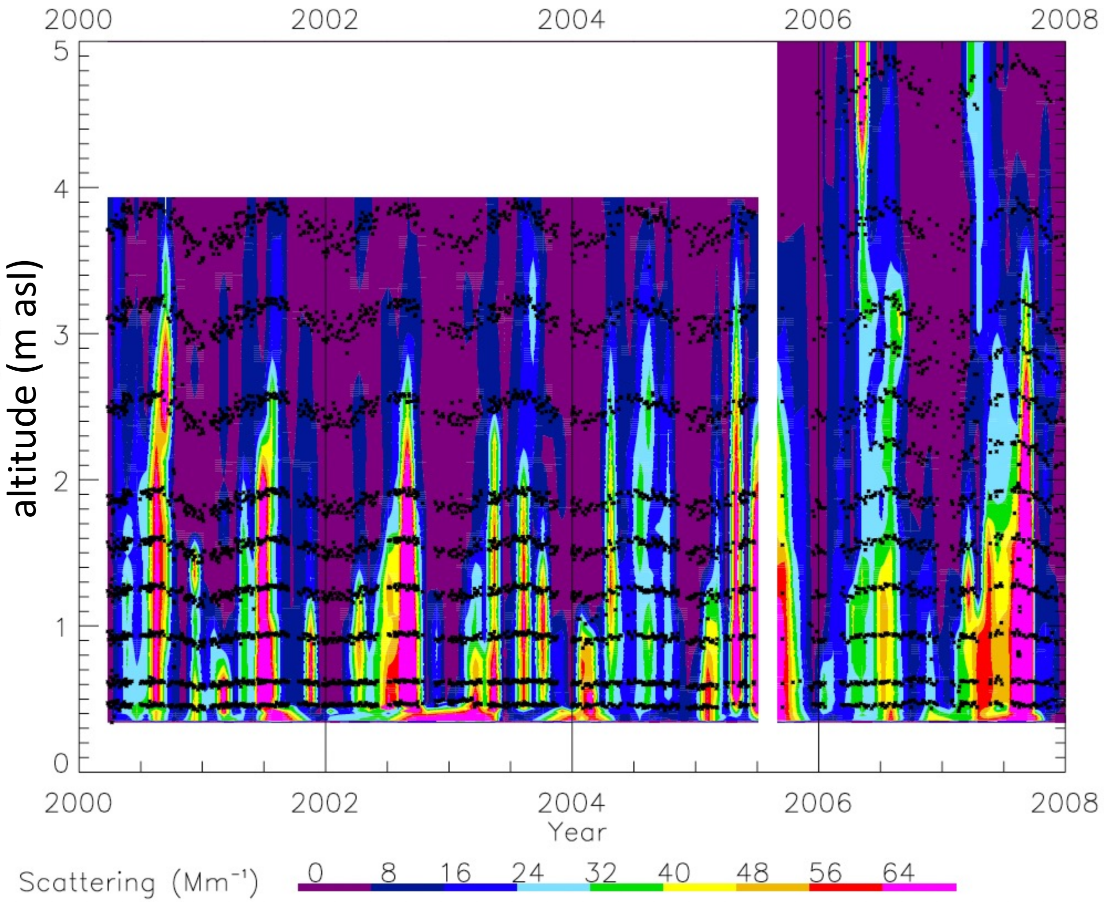
Increasing and decreasing trends for single scattering albedo
→Different sources/control strategies for scattering and absorbing particles – they decrease at different rates.

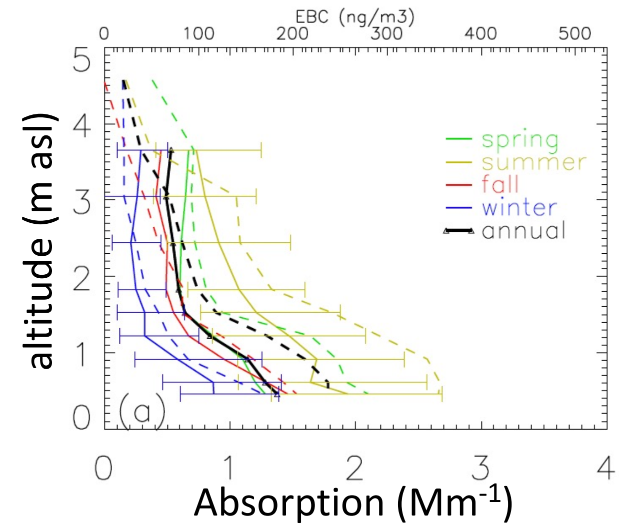
# Are surface measurements representative of atmosphere?

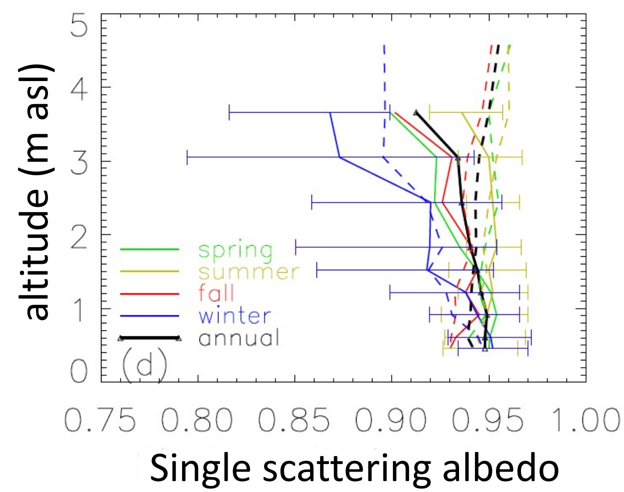
→ Fly an instrumented airplane over surface sites and find out!



# Are surface measurements representative of atmosphere?

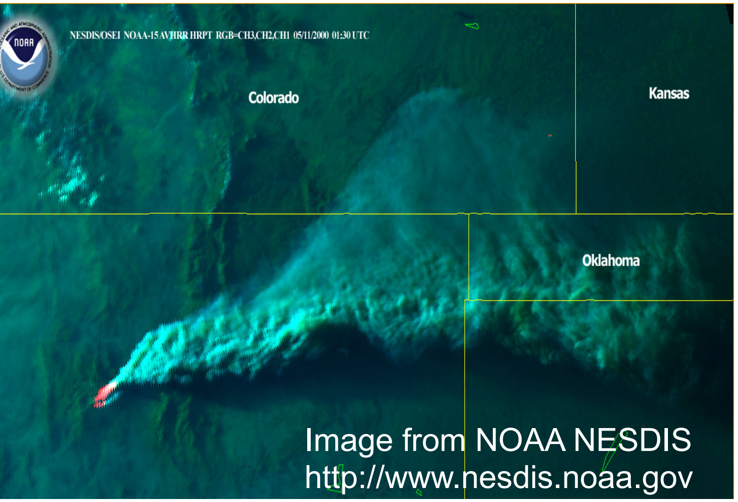


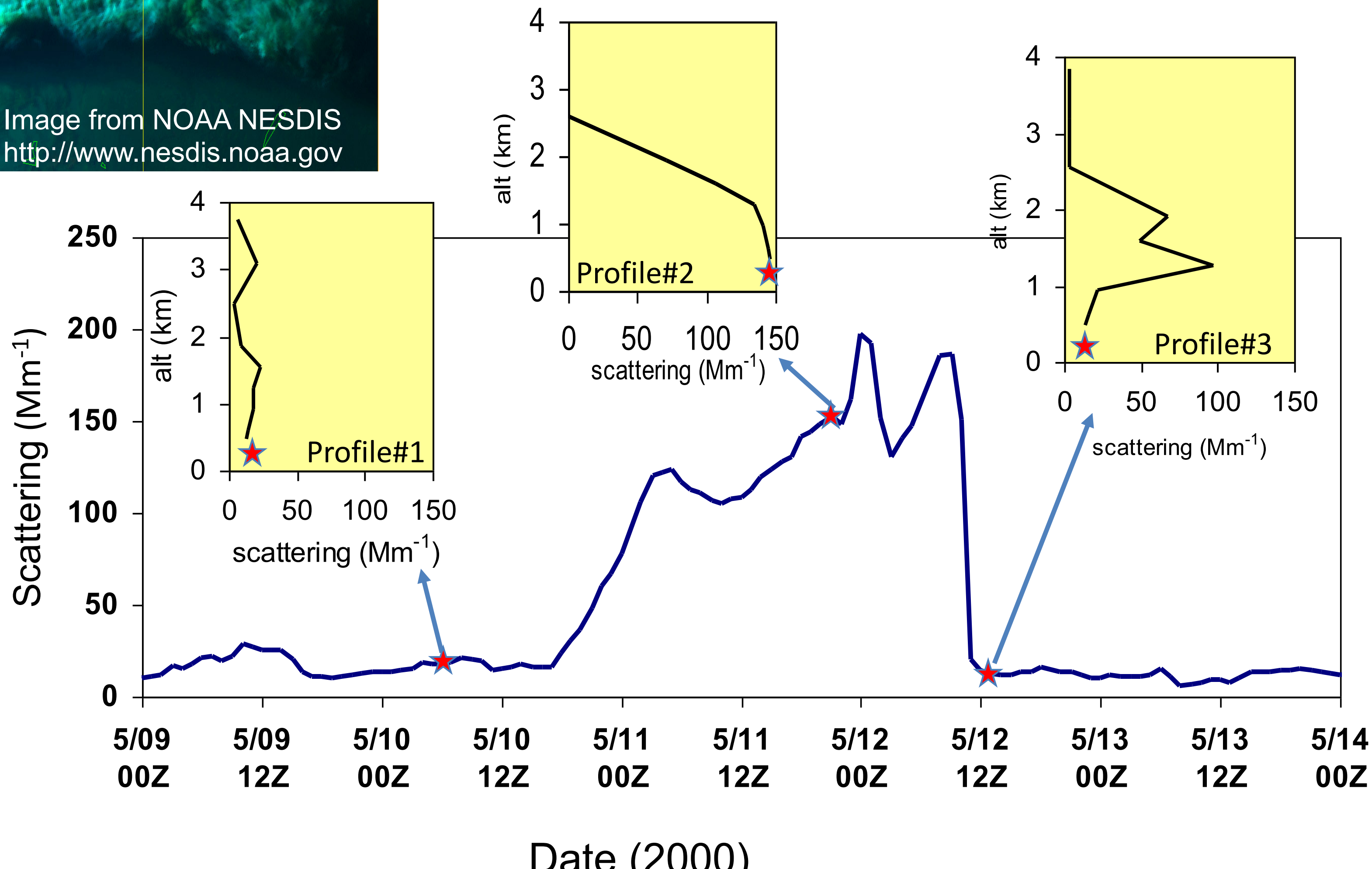




- Clear aerosol seasonality through atmospheric column.
- Measured properties decrease with altitude.
- Derived properties are relatively constant up to 2 km.

# Are surface measurements representative of atmosphere?

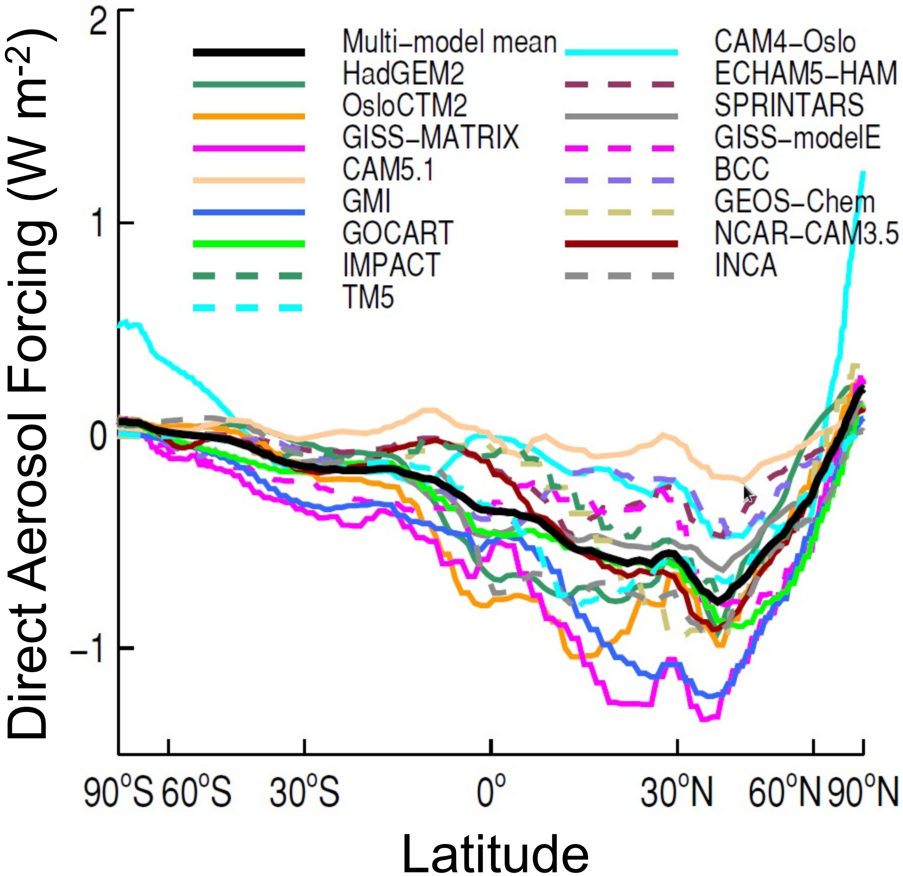


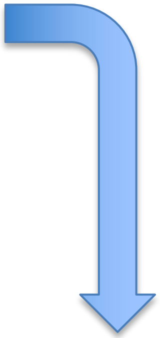

→There are clearly times when aerosol at surface is different than aloft!



# Why evaluate models?

- Models are used to predict climate forcing
- Models parameterize complex aerosol processes
- Aerosols are large source of model uncertainty



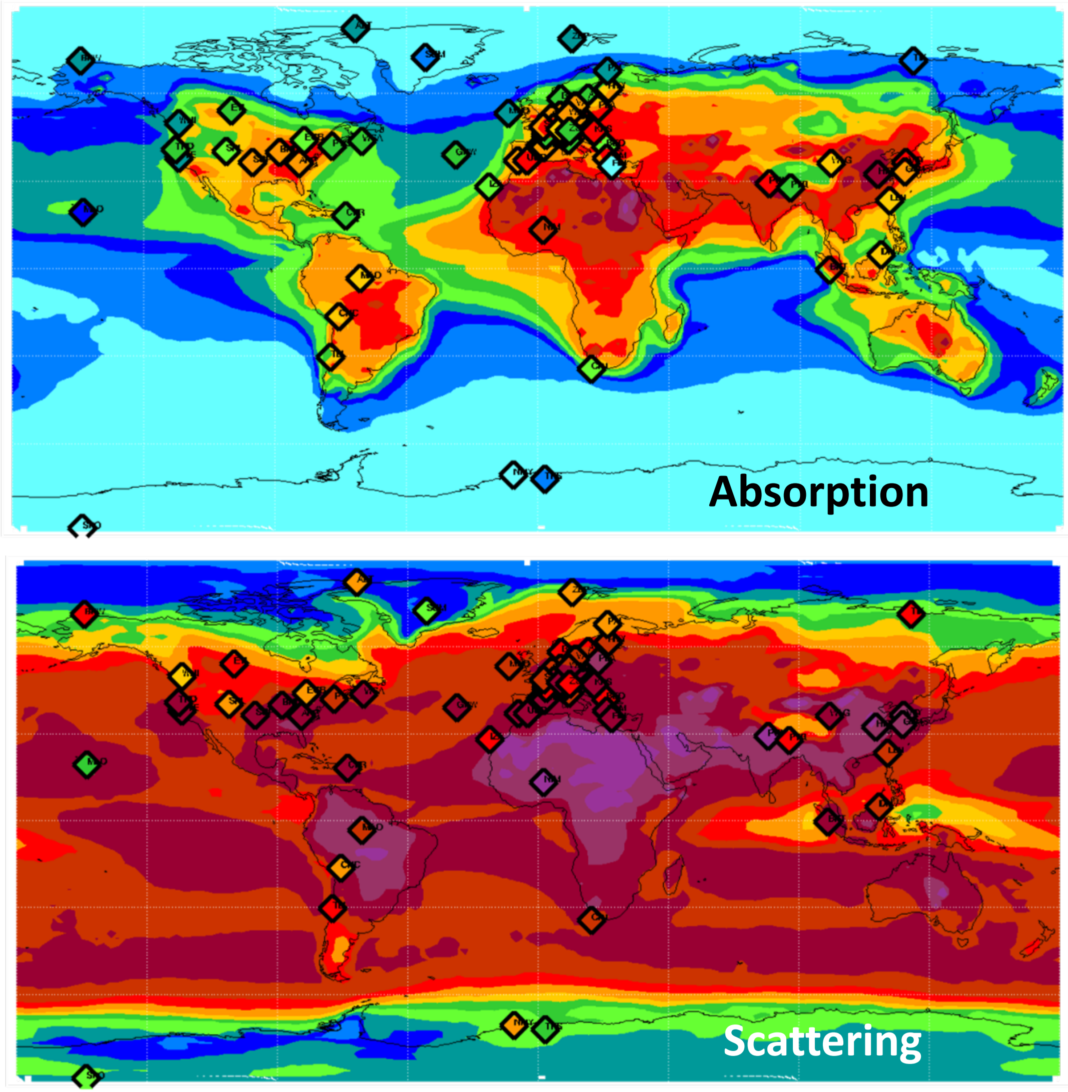
- cloud formation
- deposition (wet/dry)
- aerosols on snow
- transport (vert and horiz)
- new particle formation
- chemistry and aging
- emission strength

(from Myhre et al., 2013)

# Why the model variability?

--emissions
--transport
--parameterization of atmospheric processes
 →wet/dry deposition
 →chemical transformation
 →aerosol water interaction
 →aerosol size and physical processes
--spatial resolution and topography
--assumptions about converting from chemical to optical properties

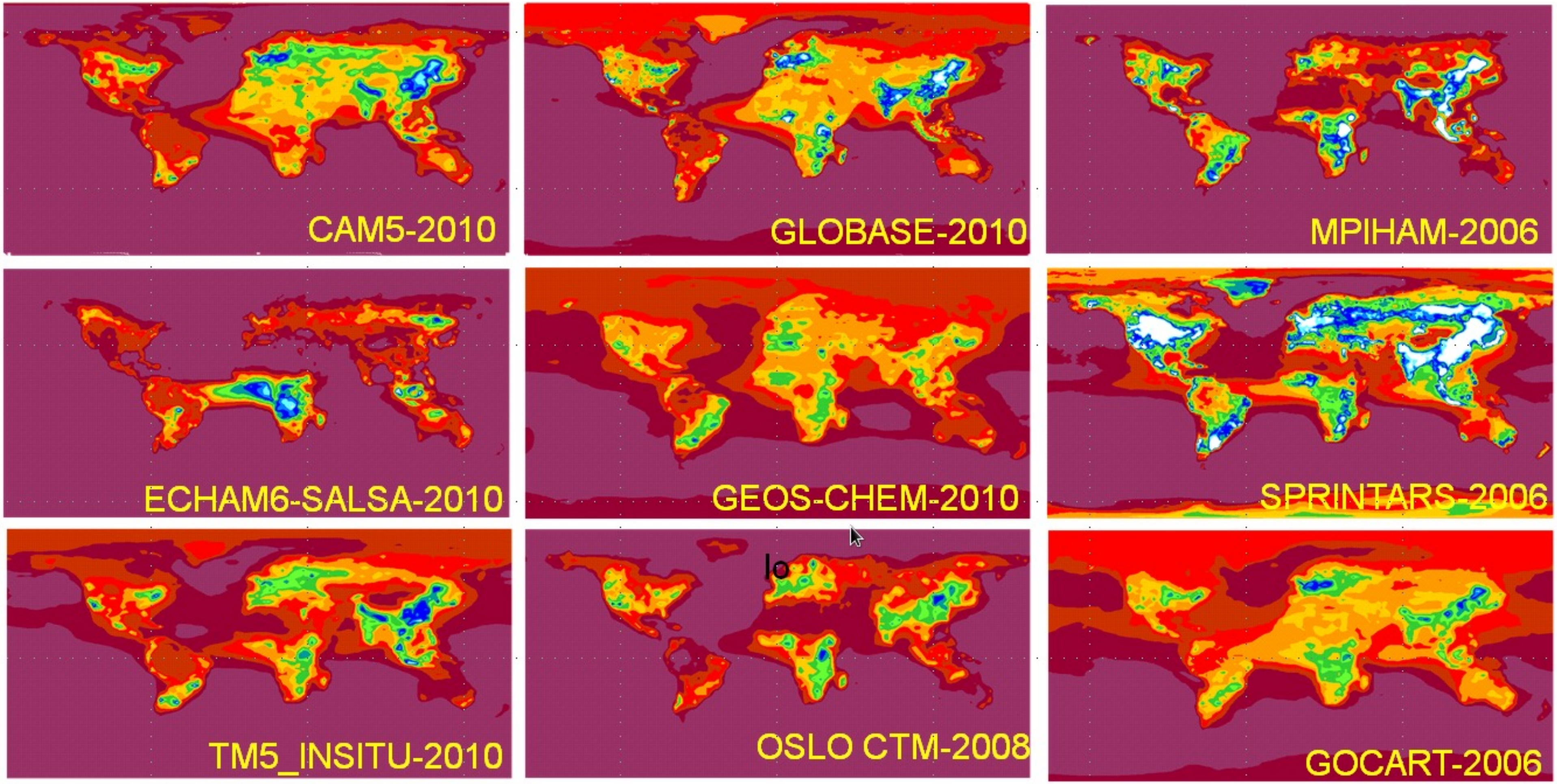
# Aerosol Climatology: **Big Picture**




- General pattern of absorption and scattering is similar for models and in-situ measurements
- Models simulate high aerosol amounts in source regions and low aerosol amounts in regions

CAM5 output for AEROCOM P3 INSITU project

# Aerosol Annual Climatology: **Single Scattering Albedo**

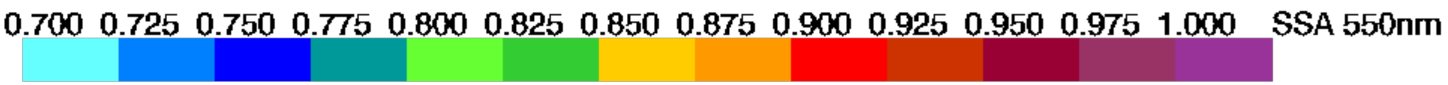



Some models see much darker aerosol in some source regions

→ Model emissions/processing?

→ Aerosol mass to optical property parameterization?

→ ???



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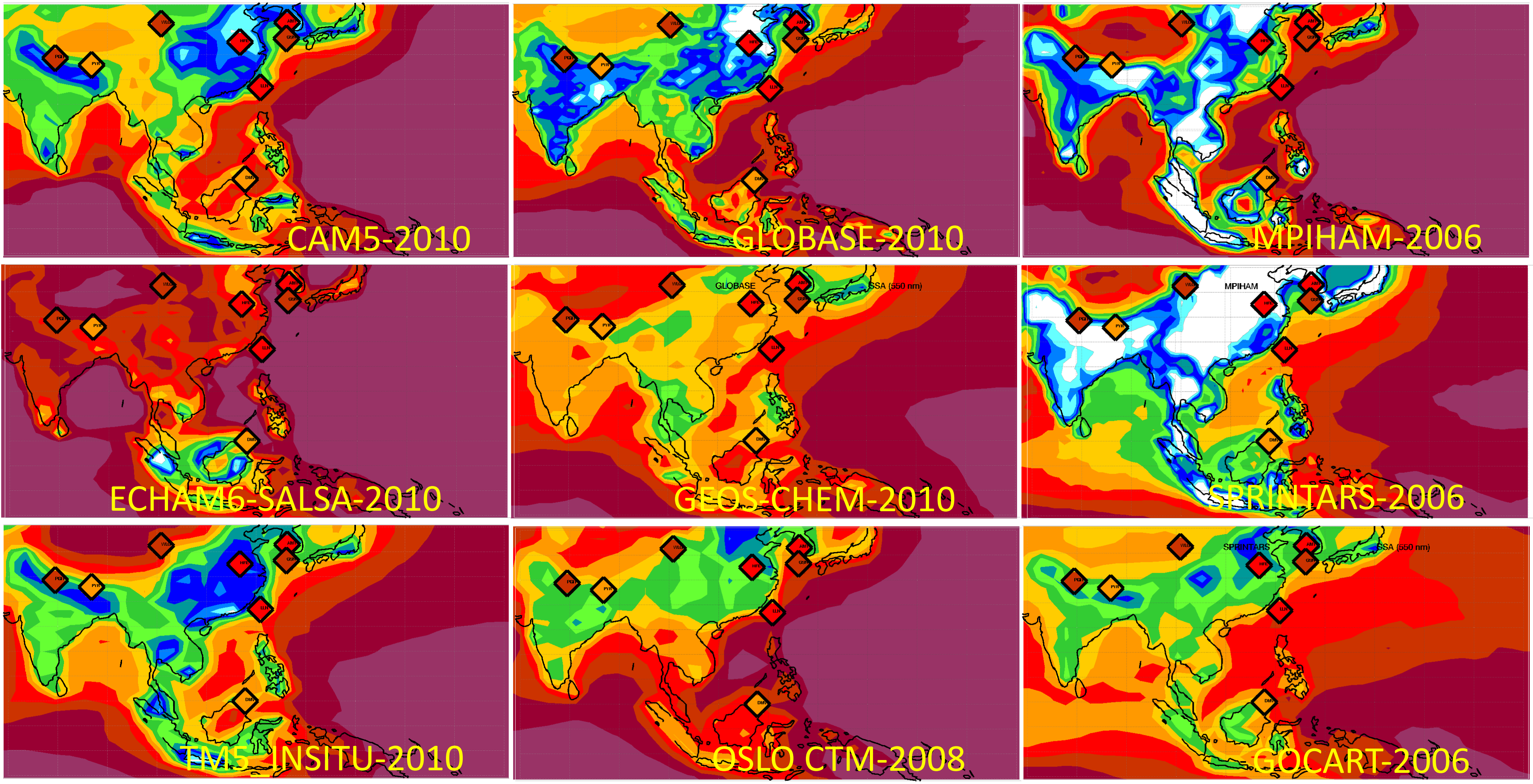
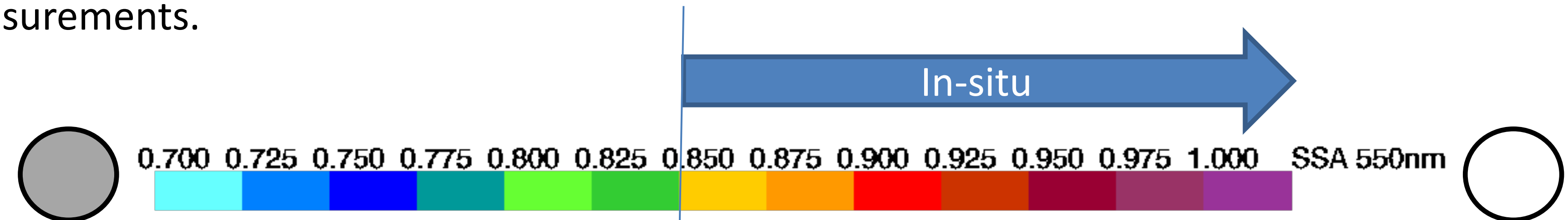
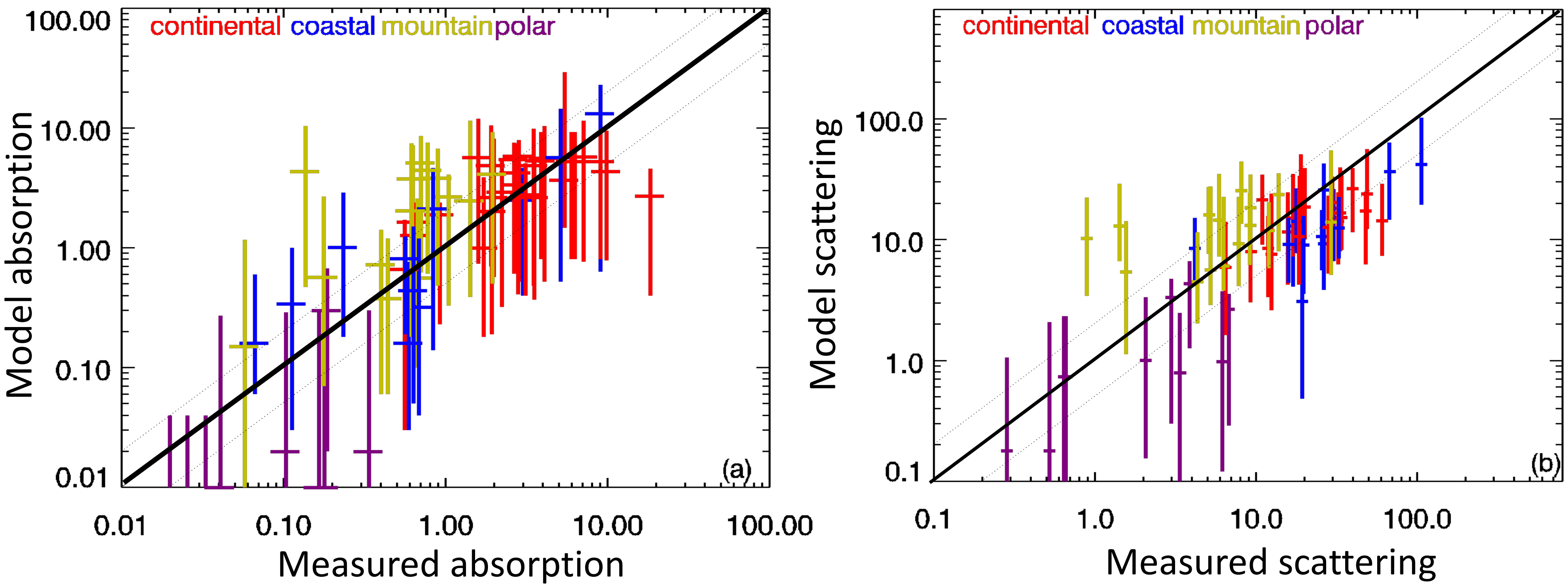



In Asia, most models simulate darker aerosol (lower SSA) than is observed by the in-situ measurements.



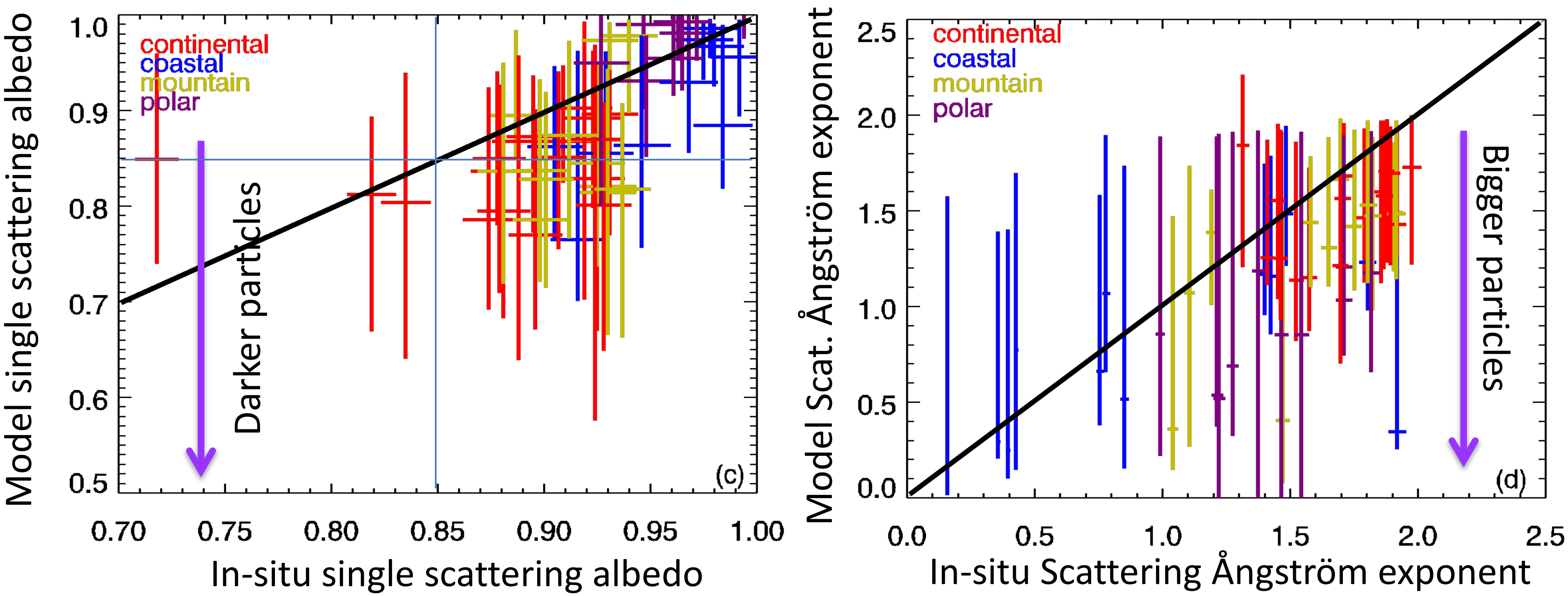
# Aerosol Annual Climatology: **Absorption and Scattering**




- Models tend to over-predict absorption and scattering at mountain sites
- Modeled absorption tends to be over-predicted
- Scattering tends to be under-predicted (except at mountain sites)
- More range (relatively) in model prediction of absorption than scattering

*Vertical bar shows range of models, horizontal bar is measurement uncertainty based on Sherman et al. (2015)*

# Aerosol Annual Climatology: **SSA and Ångström exponent**

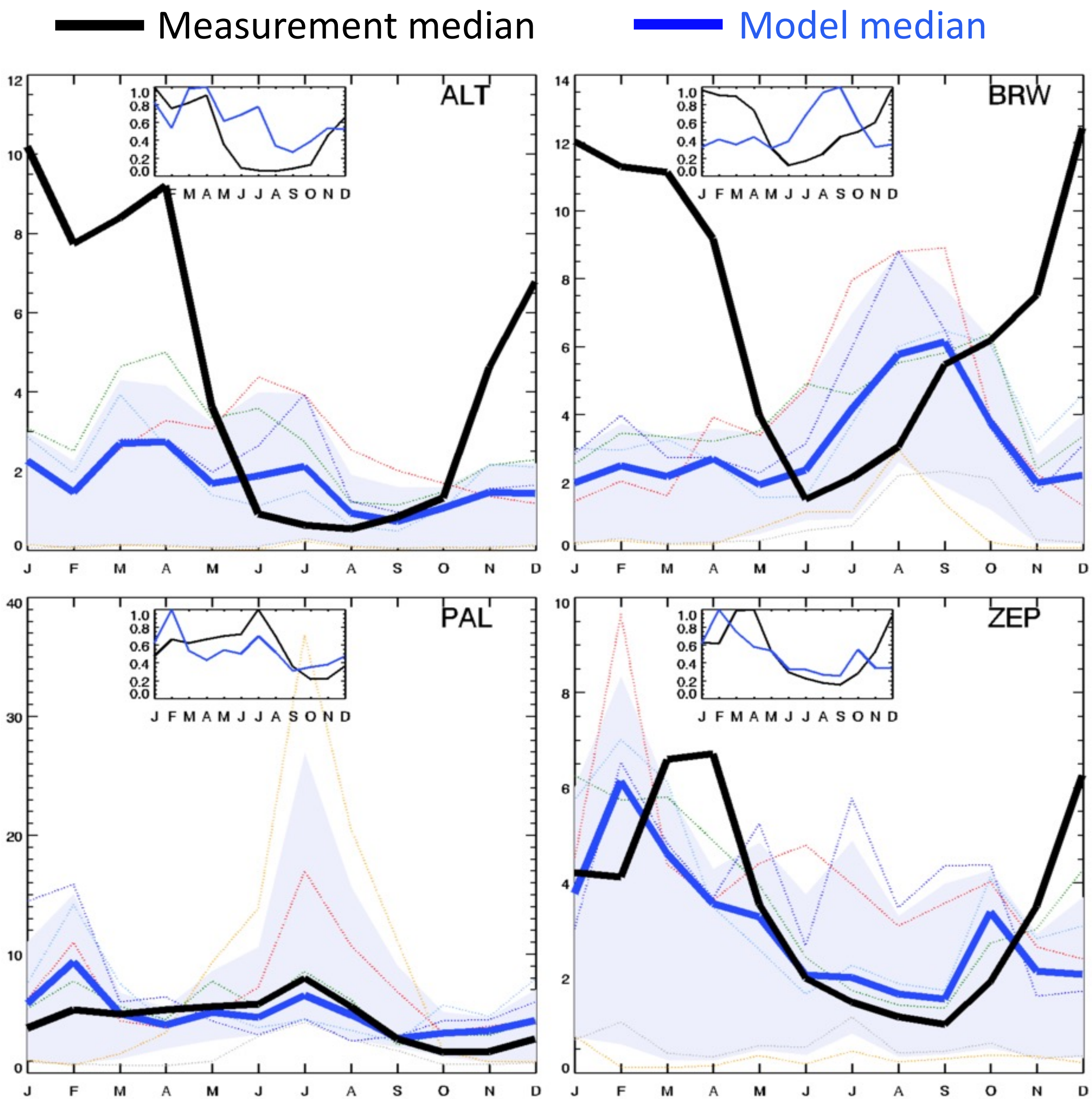



- Model SSA tends to be lower (more absorbing) than in-situ SSA → partly driven by model under-prediction of scattering

- Modelled Ångström exponents suggest larger particles than observed by in-situ measurements

*Vertical bar shows range of model medians, horizontal bar is measurement uncertainty based on Sherman et al. (2015)*

# Aerosol Seasonality: Scattering in the Arctic




Seasonal comparisons can suggest model processes to focus on.

For example: what is causing the model peak in summer at Barrow?
--Overestimating forest fire emissions?
--Underestimating removal processes?

Why is model/measurement agreement better in the European Arctic than the North American Arctic?

# Aerosol 'behavior': **Systematic variability**

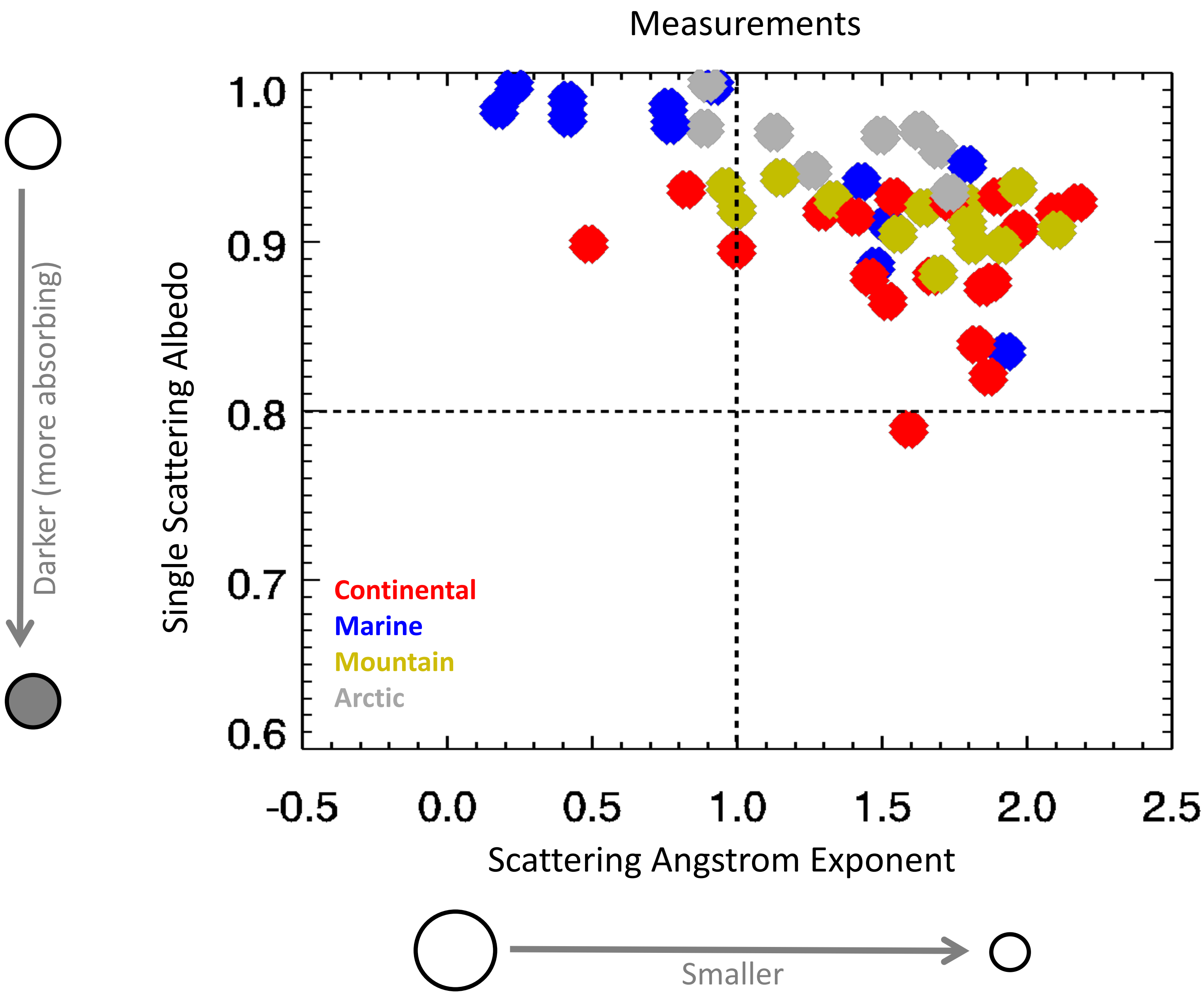
Systematic variability is the relationship between two parameters

→Can provide information about how well the model is simulating aerosol aging processes, chemistry, sources, transport, etc.

- If models correctly simulate how two parameters co-vary but have the overall values wrong, then maybe they just need to tweak the emissions.

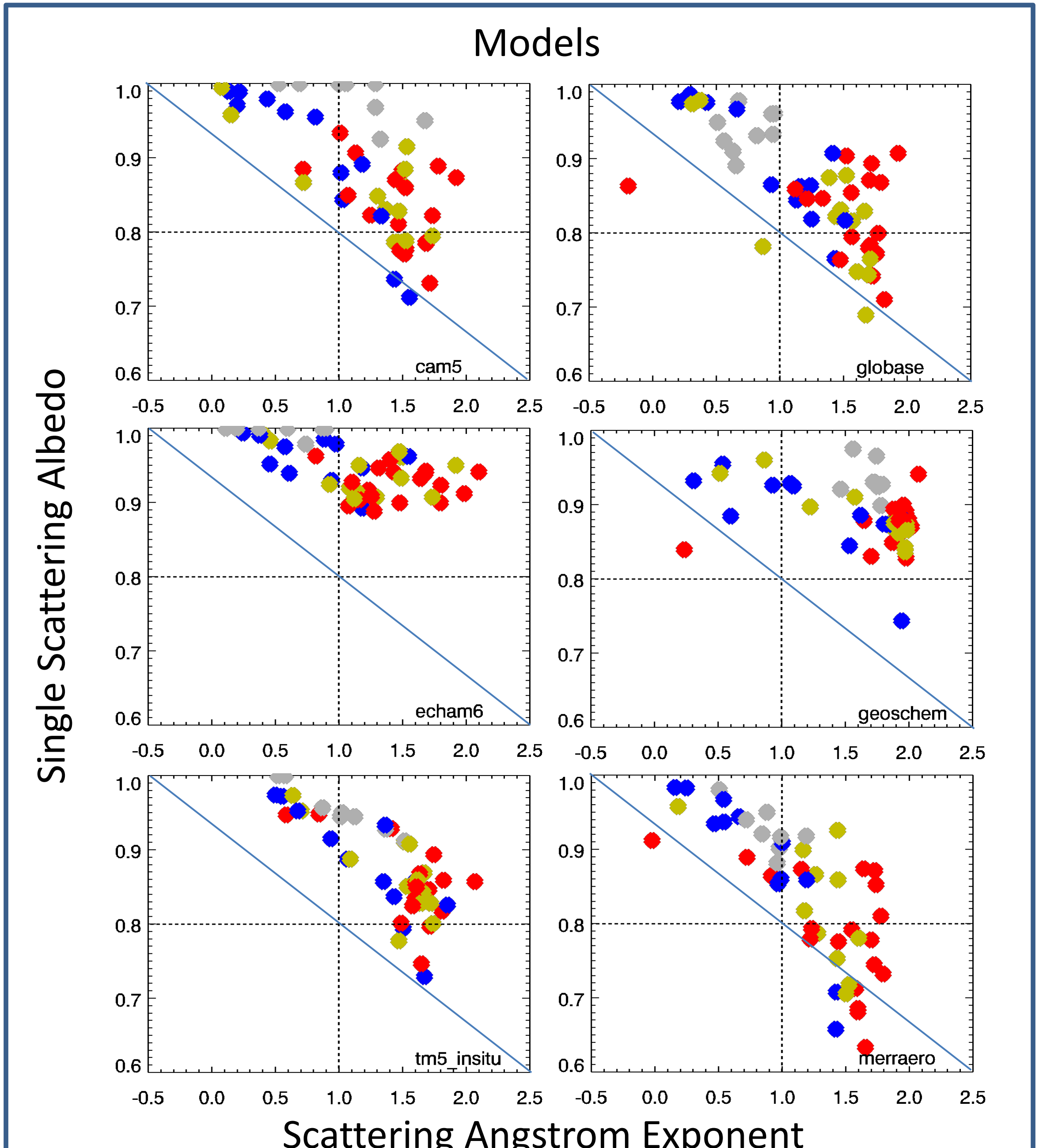
- If models cannot simulate the observed co-variance then perhaps an atmospheric process (e.g., wet removal) is incorrectly parameterized.

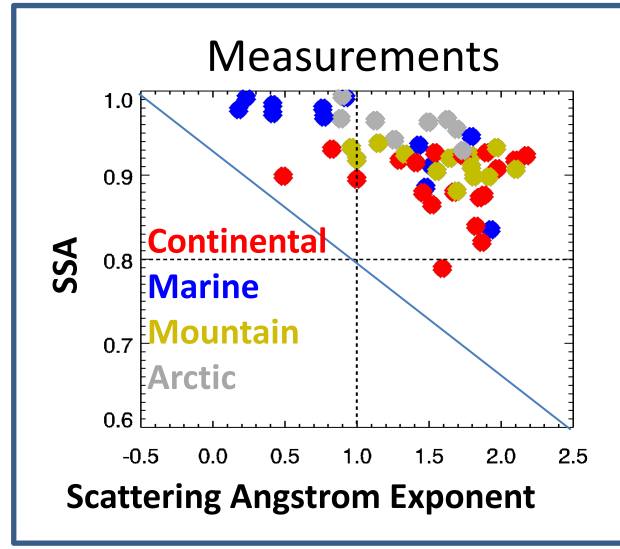
# Aerosol Behavior: **Systematic Variability**




Each point represents climatological median for one site

# Aerosol Behavior: Systematic Variability

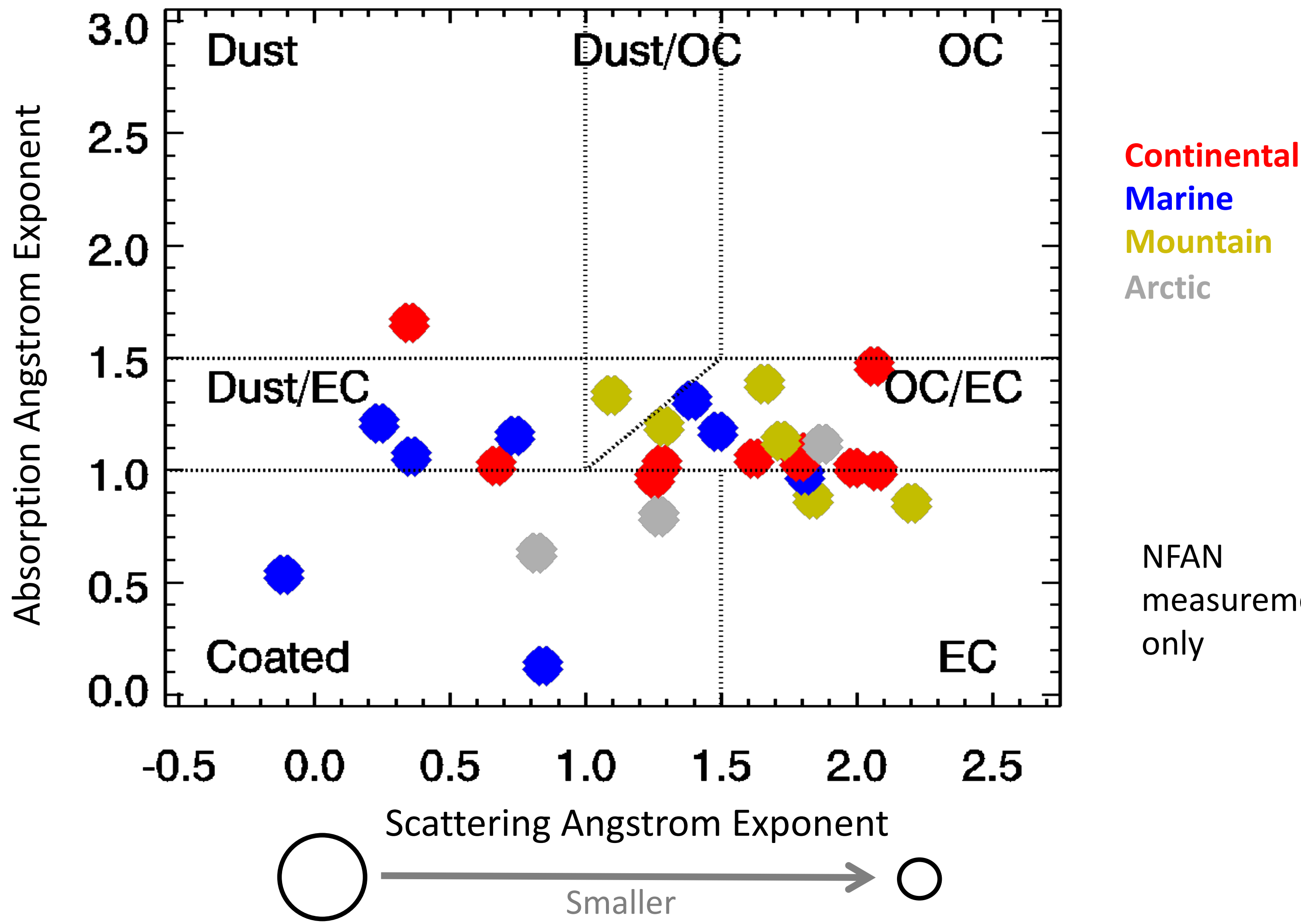



Model data exhibit similar overall relationships between SSA (chem) and SAE (size)

- general pattern of decreasing SSA with increasing SAE
- models tend to have darker, larger particles

# Aerosol Behavior: **Systematic Variability**

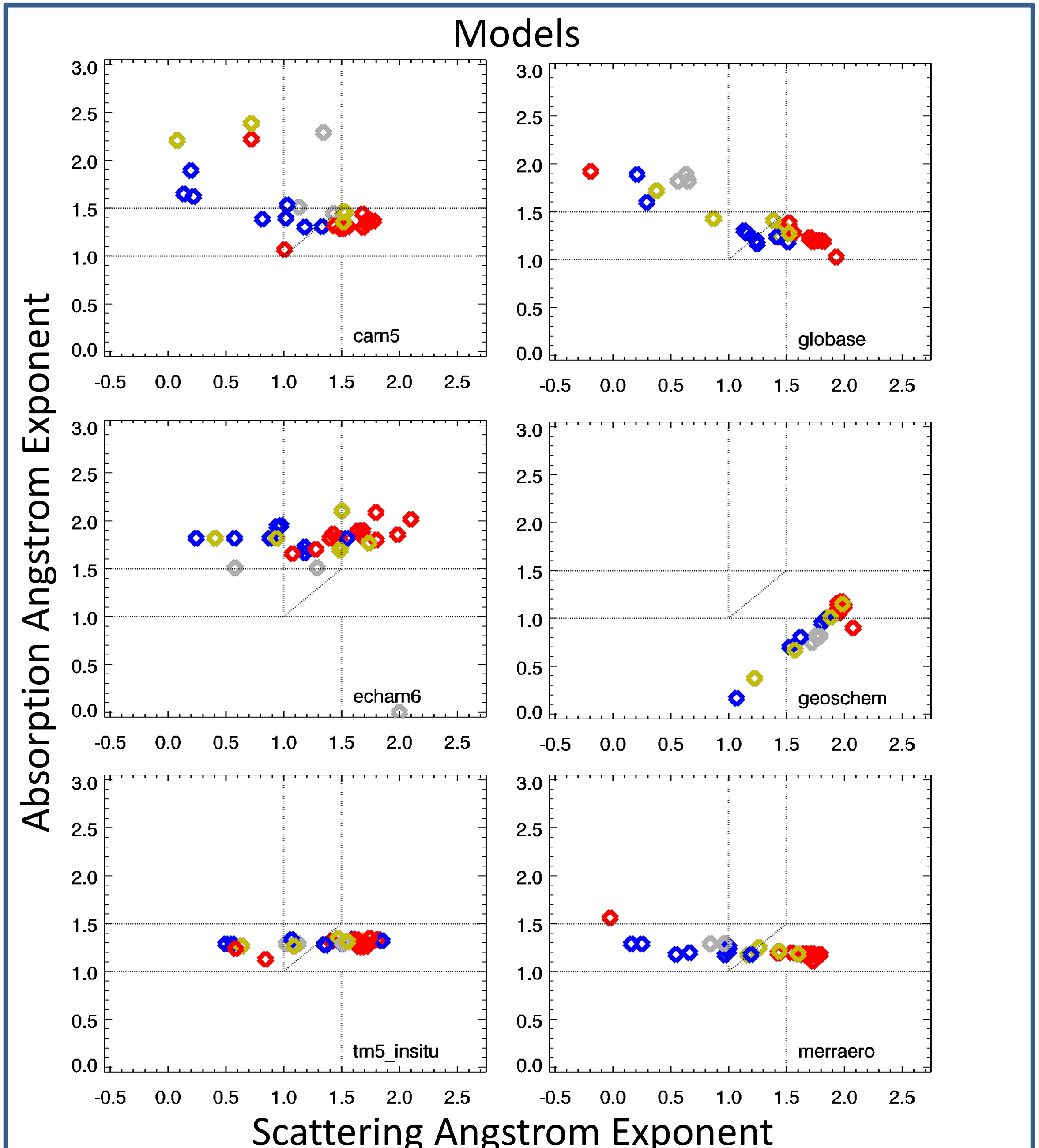


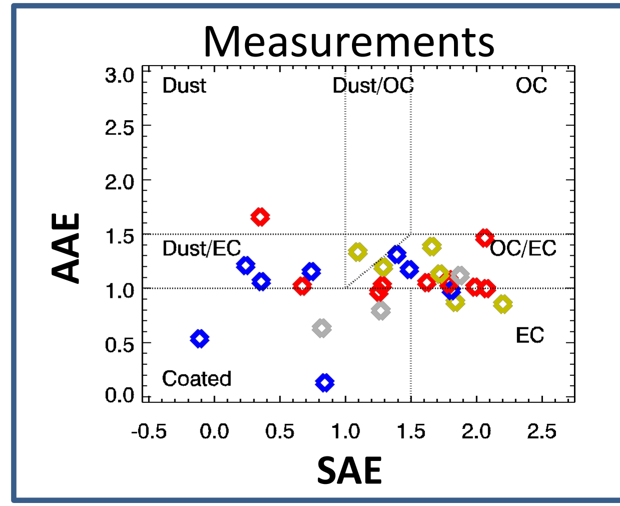

NFAN measurements only

Relationships between aerosol optical parameters may indicate aerosol type/composition.

*Cazorla et al., (2013); Schmeisser et al. (2017)*

# Aerosol Behavior: Systematic Variability




Model data exhibit very different relationships between AAE and SAE

- differences amongst models
- differences between models and insitu

# Take aways

General:

→Lots can be done with long-term data

- Climatologies, trends, remote sensing validation, model evaluation…

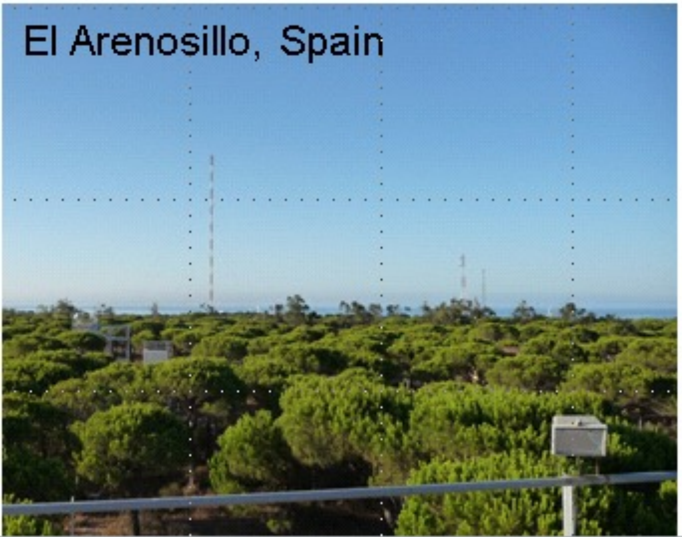
→Measurement data needs to be evaluated and instruments and measurement systems need to be characterized and calibrated.

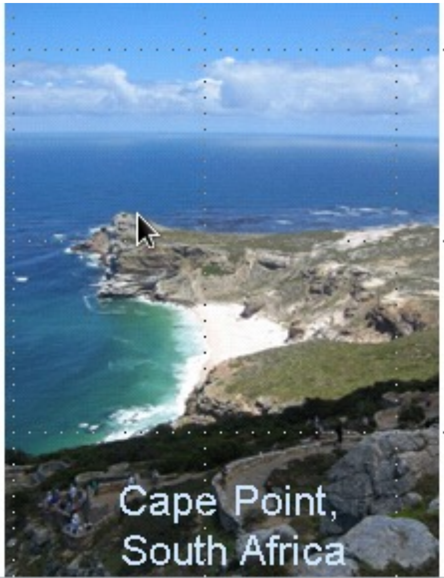
Model evaluation

→models tend to see lower scattering than observations

→models tend to see darker, smaller aerosol than observations

→models have varying success in reproducing observed seasonality and co-variance amongst aerosol optical properties



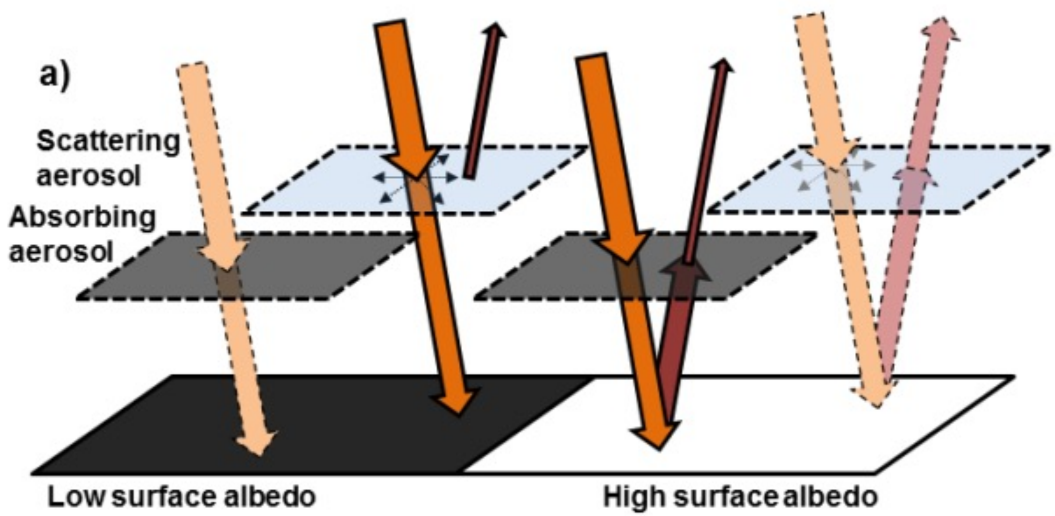




Thanks!

# Aerosol Location, Darkness and Surface Albedo

Dark surface absorbs a large portion of the solar radiation
→Absorbing aerosols will thus have a small effect.
→Scattering aerosols increase reflected solar radiation and have a cooling, since the solar radiation would otherwise be absorbed at the surface.

Bright surface reflects incoming solar radiation
→Scattering aerosols have a minimal effect.
→Absorbing aerosols reduce the outgoing radiation and have a warming effect




From Myhre et al 2013

# Recommended Quantitative Terminology (Petzold et al., 2014)

- No current method responds to all essential characteristics of BC
- Consequently, no current method can justifiably claim to provide a quantitative measurement of BC

**Equivalent black carbon (EBC)**

Data derived from *optical absorption methods*.

Report the optical measurements primarily as light absorption coefficient, and secondarily as EBC, along with the mass absorption efficiency used to convert absorption to EBC.

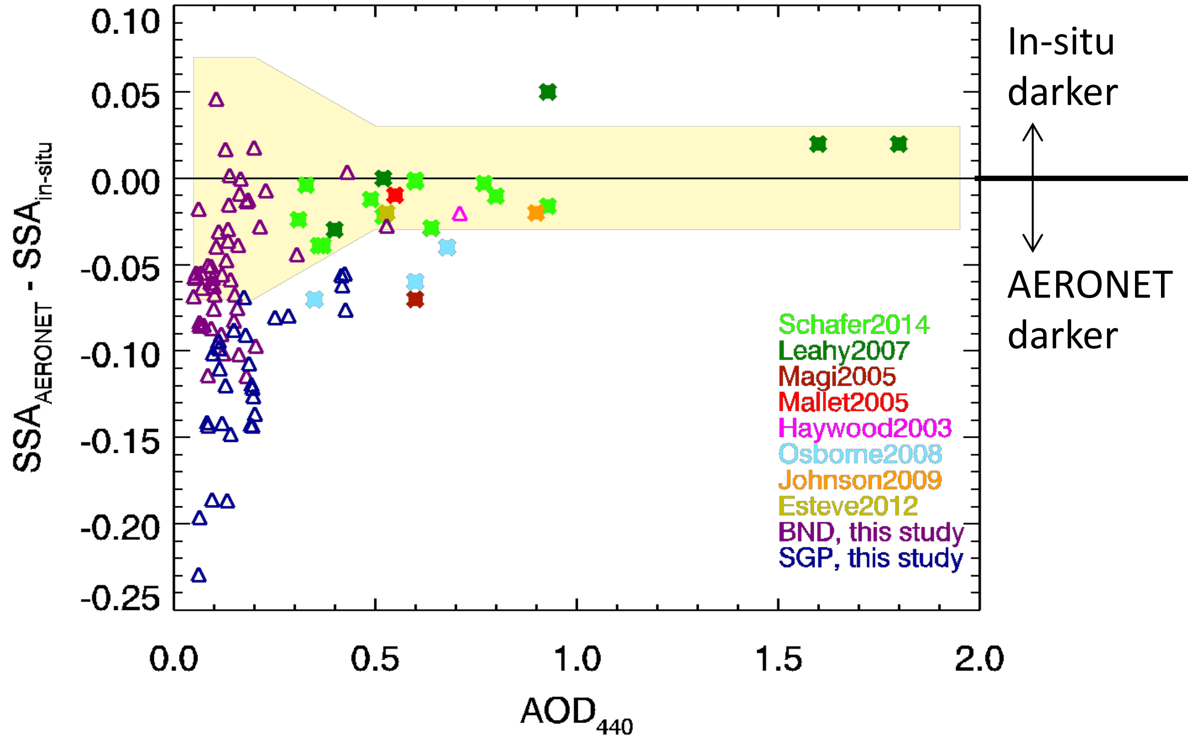
**Refractory black carbon (rBC)**

Data derived from *incandescence methods*.

**Elemental carbon (EC)**

Data derived from methods that are specific to the *carbon content* of carbonaceous matter (evolved carbon, aerosol mass spectrometry, Raman spectroscopy).

# Comparison with remote sensing observations




**Different methods give different results for single scattering albedo**

→AERONET SSA < in-situ SSA (even for $AOD_{440}$ > 0.4)

→Retrievals tend to be within SSA uncertainty

# AeroCom model evaluation – INSITU project

TIER I
Evaluation of dry, in-situ optical parameters

1. Collect data from long-term, surface, in-situ sites
2. Review and develop benchmark data set
3. Collect model output from AeroCom models
4. Compare models and measurements
5. Identify discrepancies and potential parametrization issues

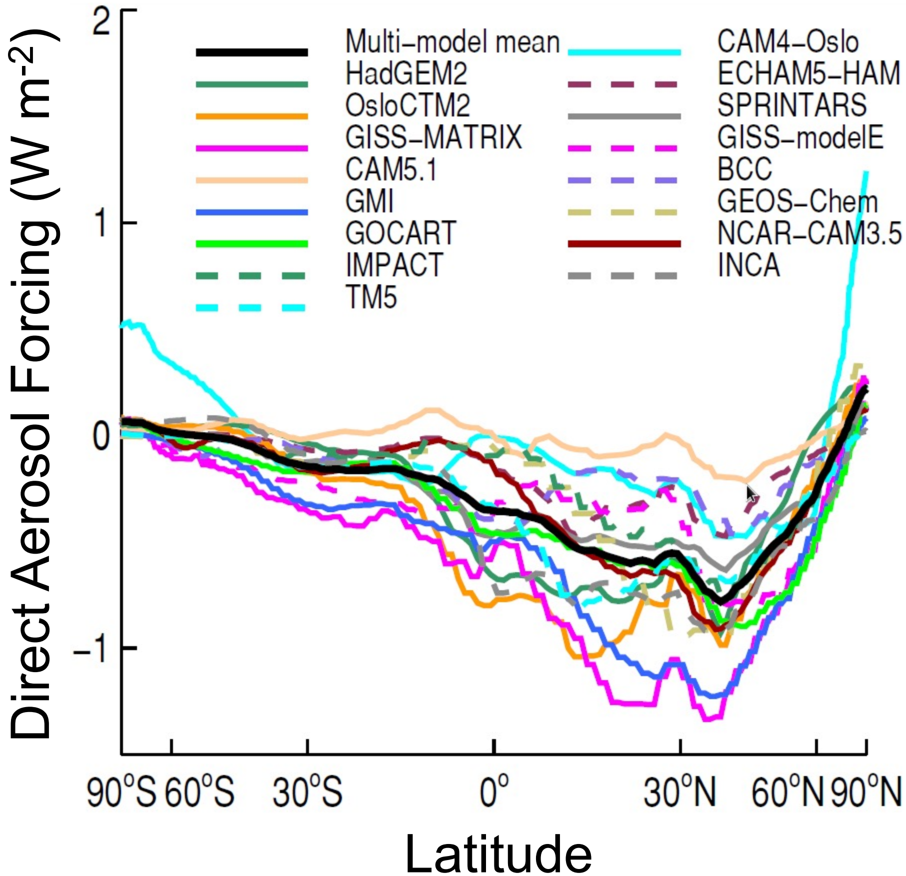
## Models Used in this Analysis

| Model name | Grid size | Output Yr |
|---|---|---|
| TM5 | 3.0° x 2.0° | 2010 |
| GEOS-Chem | 2.4° x 2.0° | 2010 |
| CAM5 | 2.4° x 1.9° | 2010 |
| ECHAM6-SALSA | 1.8° x 1.9° | 2010 |
| GEOS5-Globase | 1.25° x 1° | 2010 |
| GEOS5-MERRAero | 0.6° x 0.5° | 2010 |
| OsloCAM5 | 1° x 1° | 2010 |
| EMEP | 0.5° x 0.5° | 2010 |
| OsloCTM2 | 2.8° x 2.8° | 2008 |
| GOCART | 2.5° x 2.0° | 2006* |
| MPIHAM | 1.8° x 0.9° | 2006* |
| SPRINTARS | 1.1° x 1.1° | 2006* |

Models are all participants in 'AeroCom' project (http://aerocom.met.no/)
Models provide simulated dry optical properties at the surface at several wavelengths.

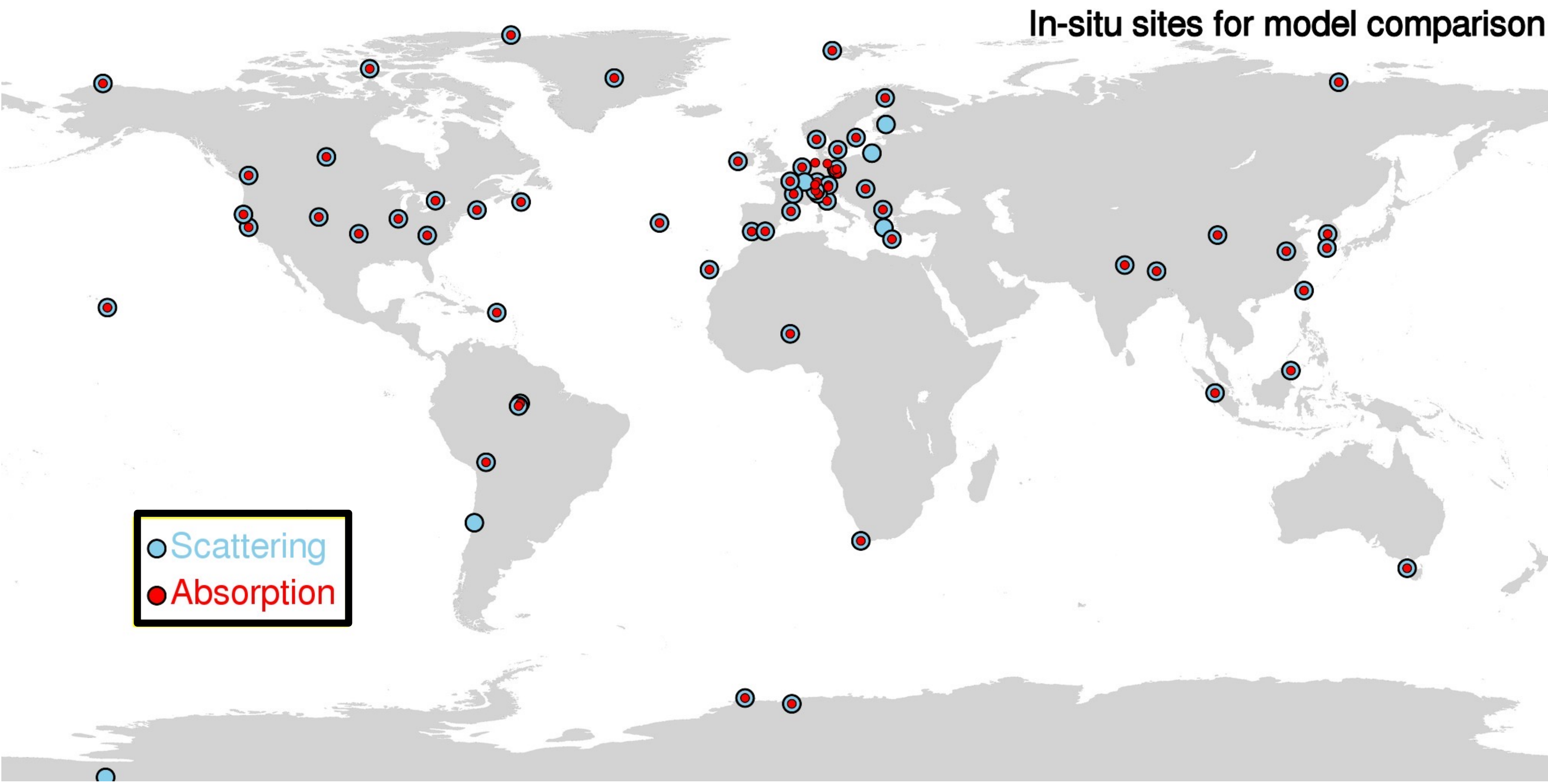
# Why evaluate models?

- Models are used to predict climate forcing
- Models parameterize complex aerosol processes
- Aerosols are large source of model uncertainty




(from Myhre et al., 2013)

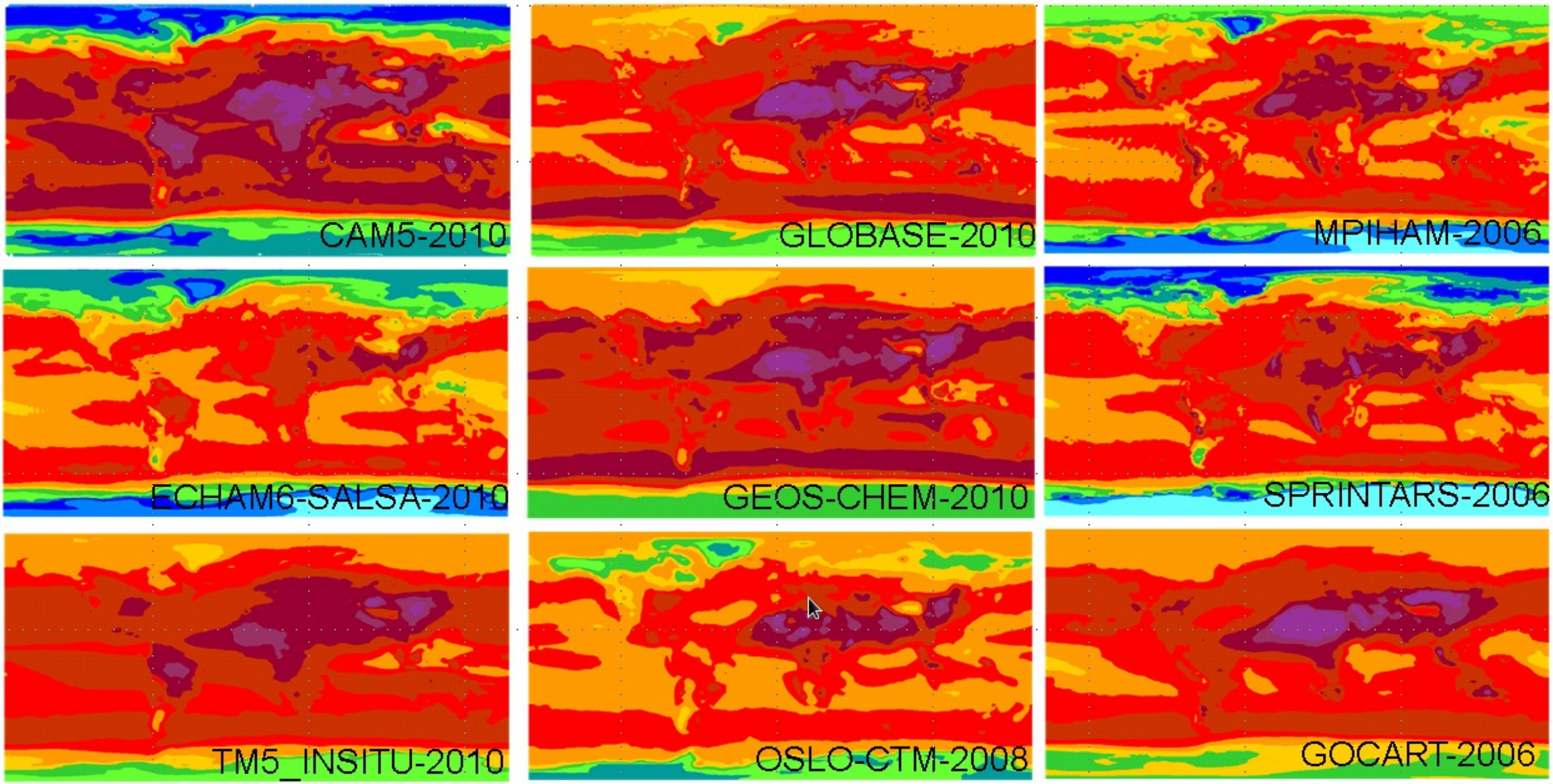
# In-situ Measurement Sites




- Sites with aerosol light scattering and/or absorption (~70 sites)
- Fewer sites than AERONET
- Gaps in S. America, Africa, Middle East, Russia, Asia

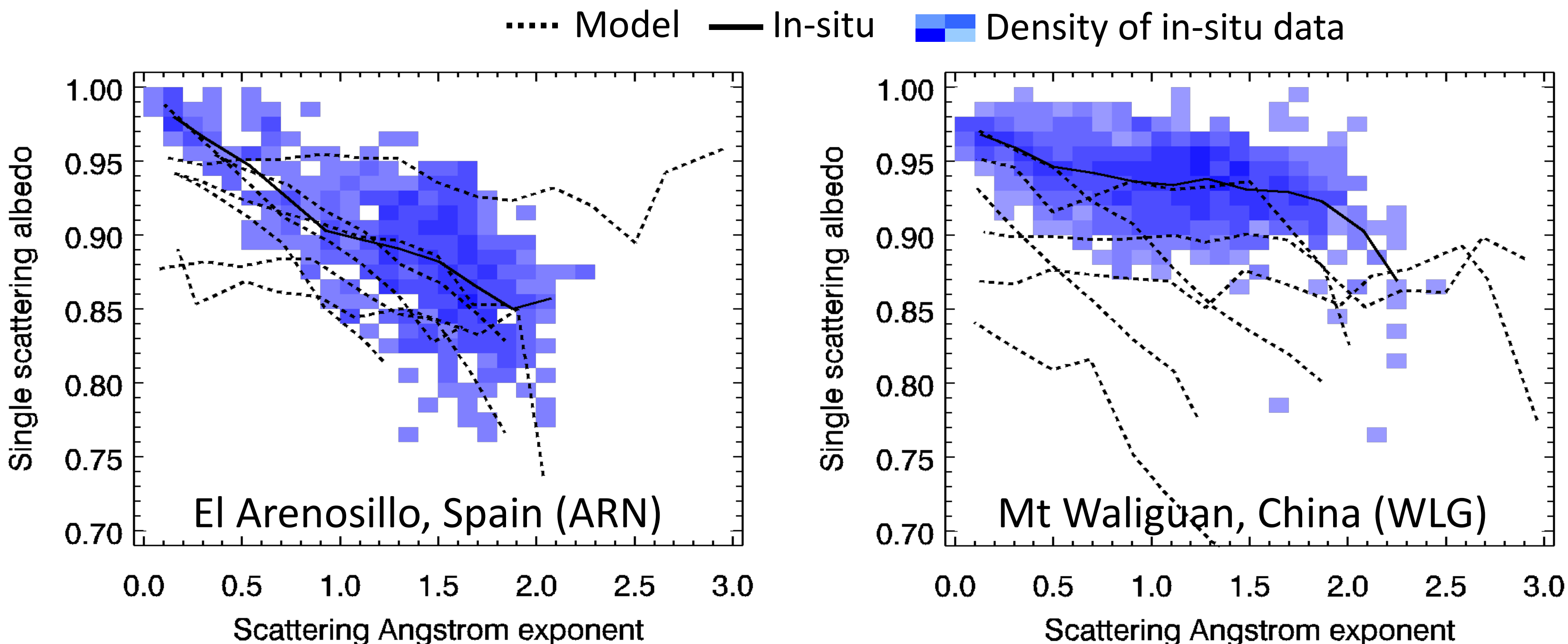
Currently updating data set with many additional sites

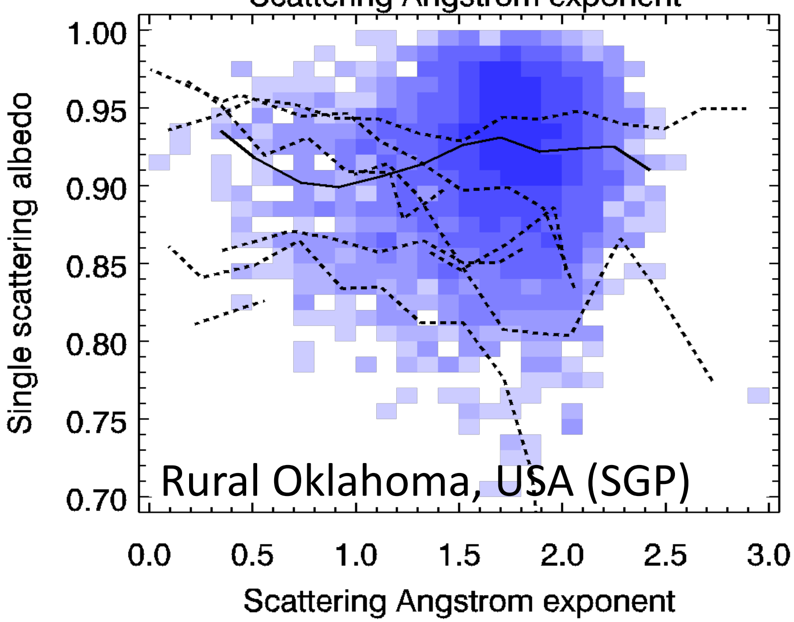
# Aerosol Annual Climatology: Aerosol Scattering




- General pattern of scattering similar for models
- Differences due to model input and conditions

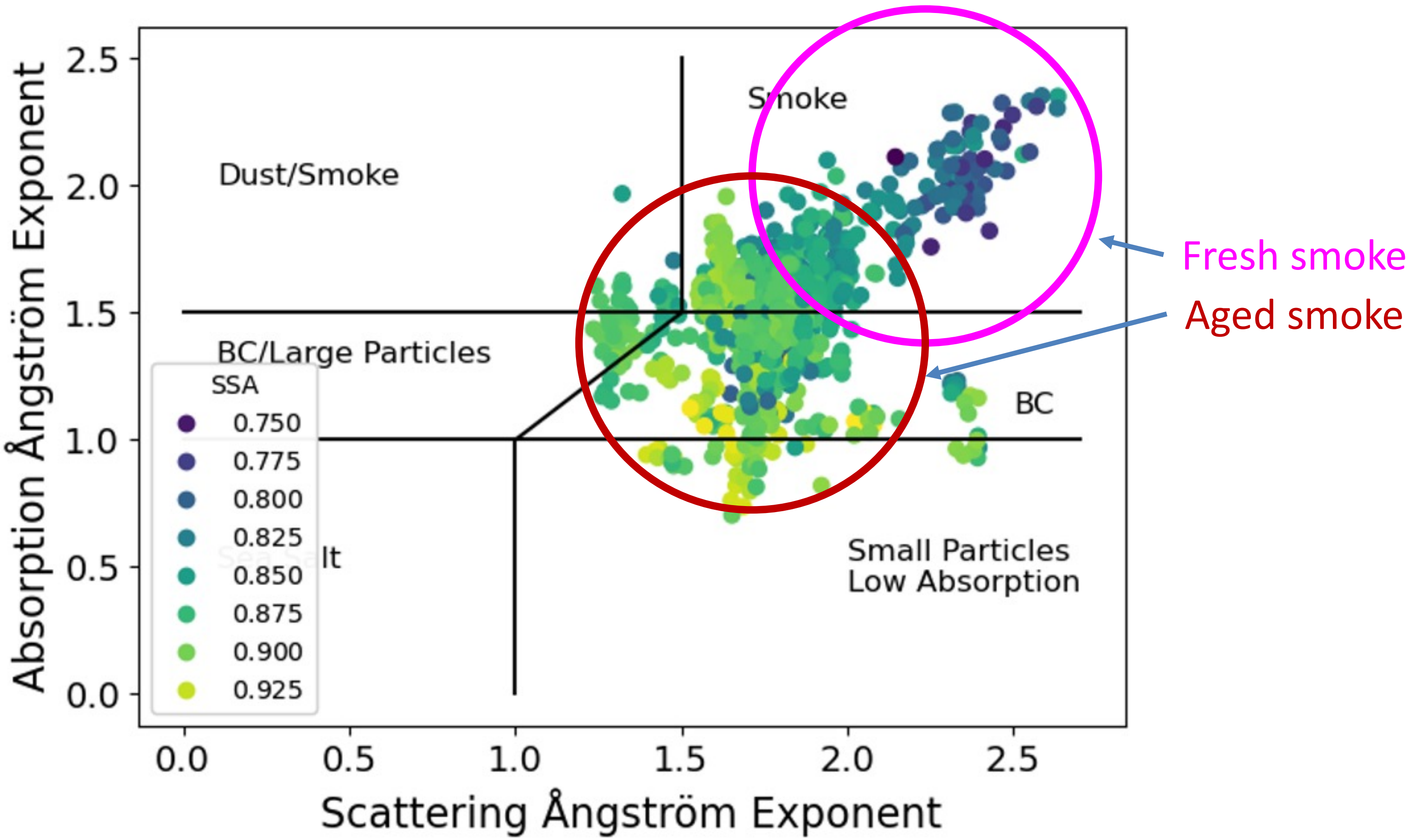
# Aerosol Behavior: **Systematic Variability**




- Models and in-situ tend to agree at coastal sites (ARN)
- Models tend to be darker than in-situ in Asia (WLG)
- Mid-continental, rural sites may be hard to characterize this way (SGP)

# Systematic variability of smoke optical properties




Colorado was impacted by both local and transported smoke in summers of 2020 and 2021

Fresh smoke tends to be small (high SAE), dark (low SSA) and organic-dominated (high AAE)
Aged smoke is not so different from typical background air – a little higher AAE value.

Schmeisser et al. (in prep)