Session O7 **Constrain aerosol properties / types**

Moderator: Thomas Popp / Rapporteur: Linlu Mei

1 12:00 Balkanski, Yves AeroCom-AeroSat Commission on Constraining Aerosol Properties
2 12:15 Schuster, Greg Tables of Aerosol Optics (TAO)
3 12:30 Kahn, Ralph Systematic sub-orbital aircraft measurements (SAM-CAAM)

12:45 Discussions (measurement priorities, additional needs, complementarity of activities)

13:15 15 min break

4 13:30 Schutgens, Nick Model evaluation with satellite data of AAOD and SSA
5 13:45 Sayer, Andrew All-sky vs. clear-sky AOD / partial cloudiness when comparing model and satellite aerosol fields
6 14:00 Tsikerdekis, Athanasios Aerosol data assimilation as a tool to detect model errors

14:15 Discussions (integration of model and data, best practices)

14:45 15 min break

AeroCom-AeroSat Commission on Constraining Aerosol Properties

Plenary O7 - Constrain

Yves Balkanski, Lucia Mona, Betsy Andrews, Nicolas Bellouin, Ken Carslaw, Mian Chin, Peter Colarco,

Ed Gryspeerdt, Paola Formenti, Stefan Kinne, Gerrit de Leew, Claudia Di Biagio, Roy Grainger,

Ralph Kahn, Pekka Kolmonen, Rob Levy, Tero Mielonen, Thanos Nenes, Thomas Popp, Adam Povey,

Claire Ryder, Andrew Sayer, Lauren Schmeisser, Michel Schulz, Greg Schuster, Nick Schutgens,

David Winker, Hongbin Yu, Ying Zhang.

Intent

- Setting up bounds and means on useful global aerosol property, to be revised annually.
- What should models and satellite retrievals be able to simulate/retrieve in relation to global aerosol loads and optical properties?
- What should be recommended to modellers/satellite scientists to test and document?
- Bounds and means on these properties and on aerosol radiative effects could suggest strategies for future observations

Means to do it

- Create a maintenance framework, e.g. a table that will be updated every year at AeroCom-Aerosat workshop, with revised property bounds (global averages or global pdf, vertical distribution, regional averages).

Where should we focus the effort on constraining aerosol properties? (1/3)
(Group discussion 27 Sept. 2021)

- Measurements, necessity to constrain aerosol intensive properties: ***hygroscopity, mass extinction efficiency, spectral light absorption and CCN properties*** to complement 20+yrs of satellite measurements with quantities not yet accessible (for ex: particle numbers <0.1um) see Kahn et al., BAMS, 2017

- *Water associated with the aerosol* (→ Lauren Schmeisser talk on INSITU project Tue. Afternoon). The two sources of uncertainties for water associated with aerosols are:
  - *HG* in the *models*, and how model capture *RH*.

  Of the two, HG is probably the largest and it needs to be quantified:

  → Representativeness of the measuring sites,

  → Could we run several models with the same assumptions for HG?

  → Suggestion to use of the stations above 1000m elevation which are generally excluded from the comparisons

Where should we focus the effort on constraining aerosol properties? (2/3)
(Group discussion 27 Sept. 2021)

- *S. Kinne* presented an analysis ***aerosol trends of both the fine and the coarse mode*** *(see poster or talk)* using the 20-year MODIS record.

  - Anomalies for fine mode can be traced to wildfires and pollution, coarse mode anomalies dominated by dust.

  - Regions mostly affected: India, China, South-East Asia and the Arctic region.

  - How these anomalies translate into TOA forcing was computed separating the SW and LW.

- ***Optical properties*** (Greg's initiative TAO) This initiative aims at updating OPAC dataset on optical properties. The refractive index of BC is way too low compared to observation/lab. OPAC uses NaCl rather for sea salt characteristics of actual sea salt aerosol (see discussion section 4.3 of *Burgos et al 2020, ACP*).

- Properties of dust (*H. Yu, C. Ryder, C. Di Biagio*). We know that dust from different source regions has very different ***optical properties linked to the mineralogy and to particle size***. We lack information on the variations of these properties within the same region. We will show in a few slides how the dust cycle was constrained using both observations and a subset of models.

- *The* database from Don Grainger's group: http://eodg.atm.ox.ac.uk/ARIA/index.html **includes optical data for *volcanic ash aerosol***. This database also includes acids and minerals among other constituents. *B. Reed* infers the refractive indices for several eruptions (see *Reed et al.. JGR, 2018).*

Where should we focus the effort on constraining aerosol properties? (3/3)
(Group discussion 27 Sept. 2021)

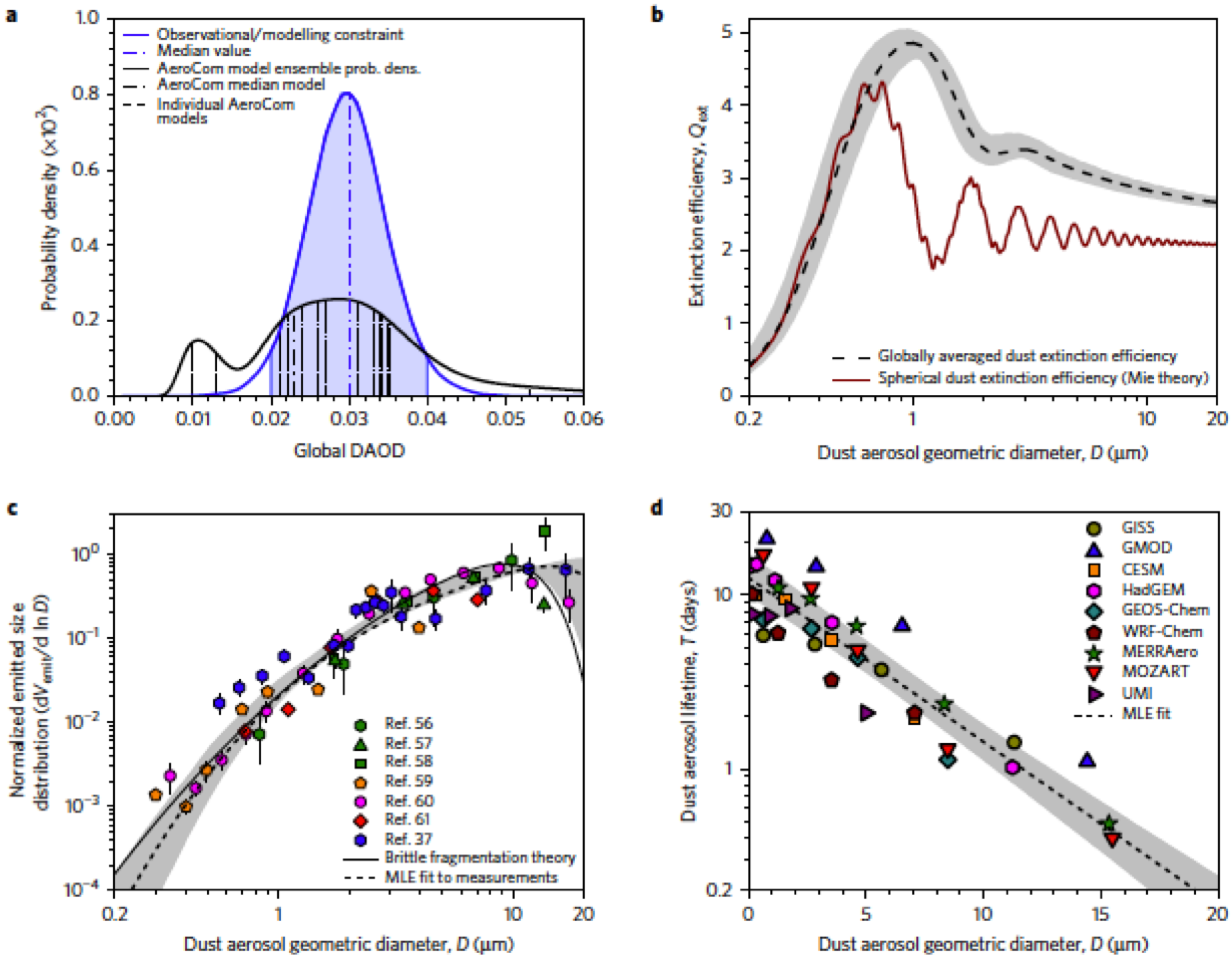
- Nicolas Bellouin suggests that a top-down approach that focuses upon extinction optical depth, AAOD, size distribution as well as fine-mode AOD should be looked into.
- Finally the approach taken in Leeds is presented by Ken Carslaw. First an ensemble of measurements is selected to build a pdf. The next step is to perturb the main factors that influence the variable, the ensemble of what is observationally plausible is determined (see *Johnson et al., ACP, 2020)*
- We now review significant advances that have been done to understand aerosols through three examples.
- Question "What sites would be good for long-term observations", see papers:

Li, J., et al., 2016. Reducing Multi-sensor Monthly Mean Aerosol Optical Depth Uncertainty Part I: Objective Assessment of Current AERONET Locations. J. Geophys. Res. 122, doi:10.1002/2016JD026308.

Li, J., et al., 2017. Reducing Multi-sensor Monthly Mean Aerosol Optical Depth Uncertainty Part II: Optimal Locations for Potential Ground Observation Deployments. J. Geophys. Res. 122, doi:10.1002/2016JD026308.

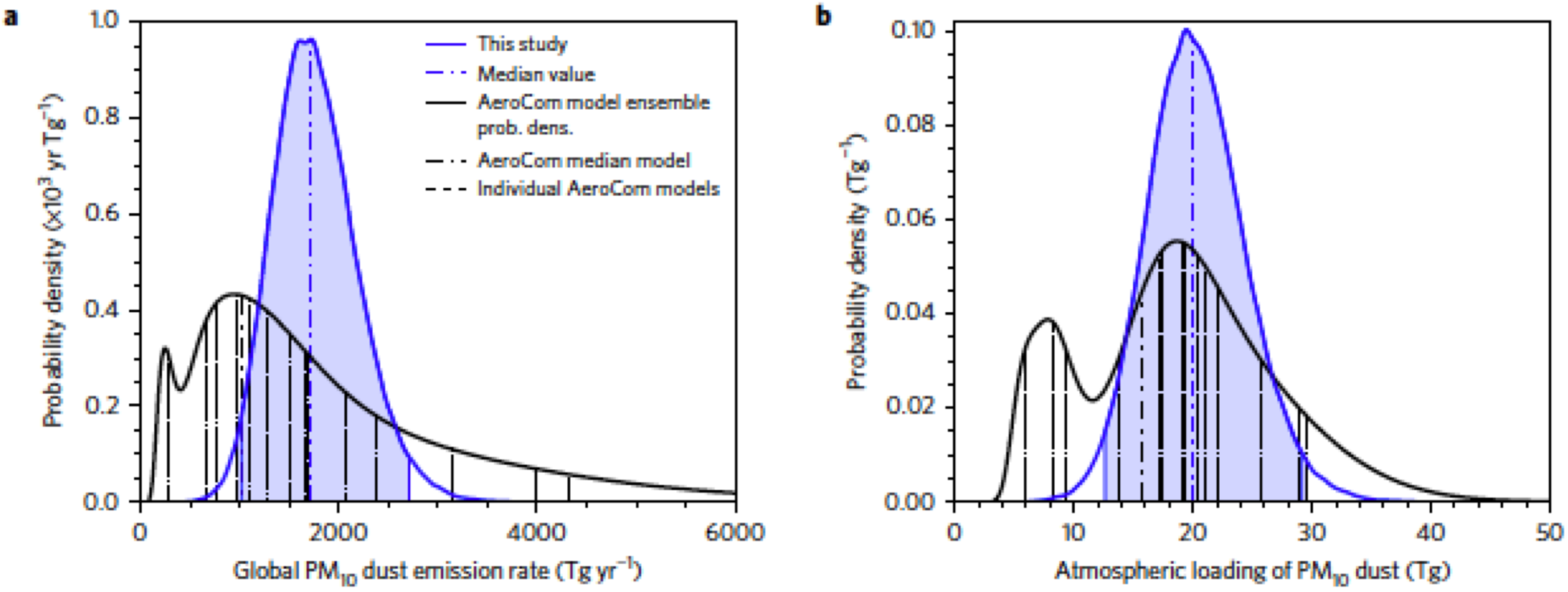
Example 1: Constraining the dust aerosol cycle

Kok et al. 2017



Constraints apply to both emission rates and atmospheric load (PM10)

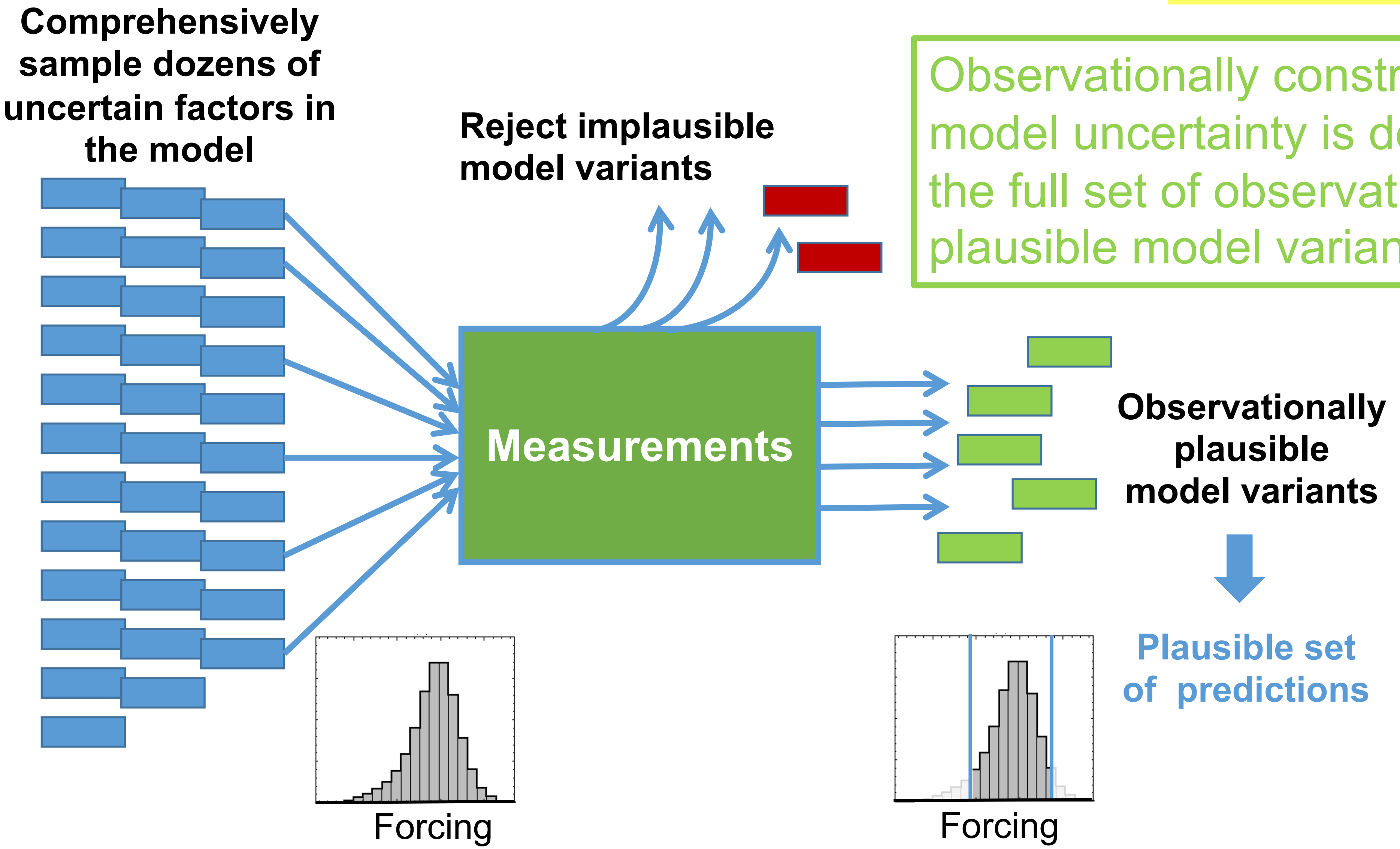
Kok et al. (2017)



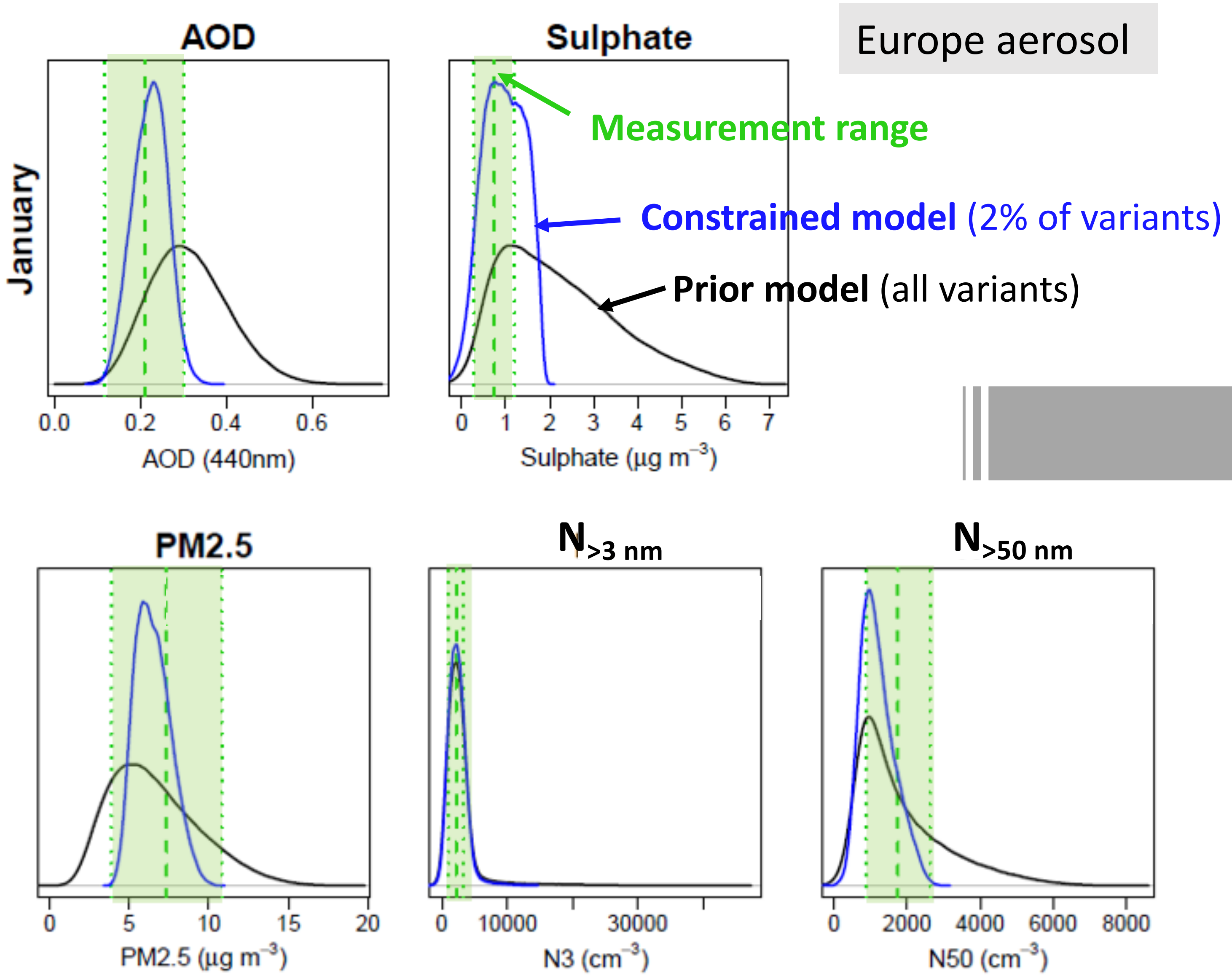
Example 2: Observational constraint

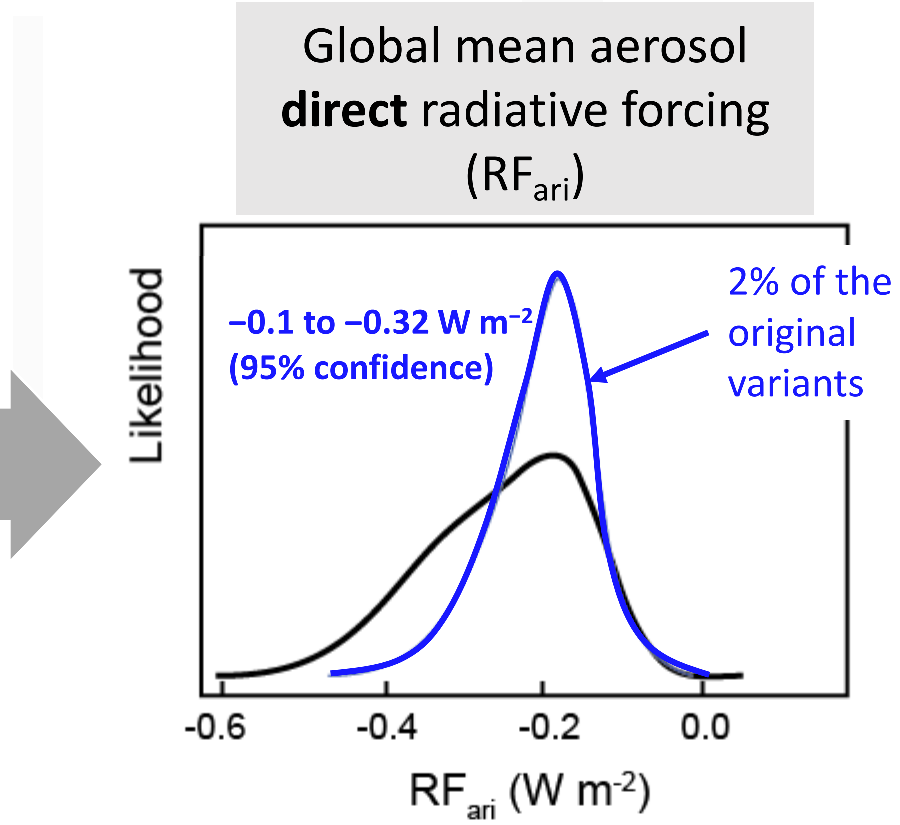
Johnson et al. ACP (2018)

Observationally constrained model uncertainty is defined by the full set of observationally plausible model variants



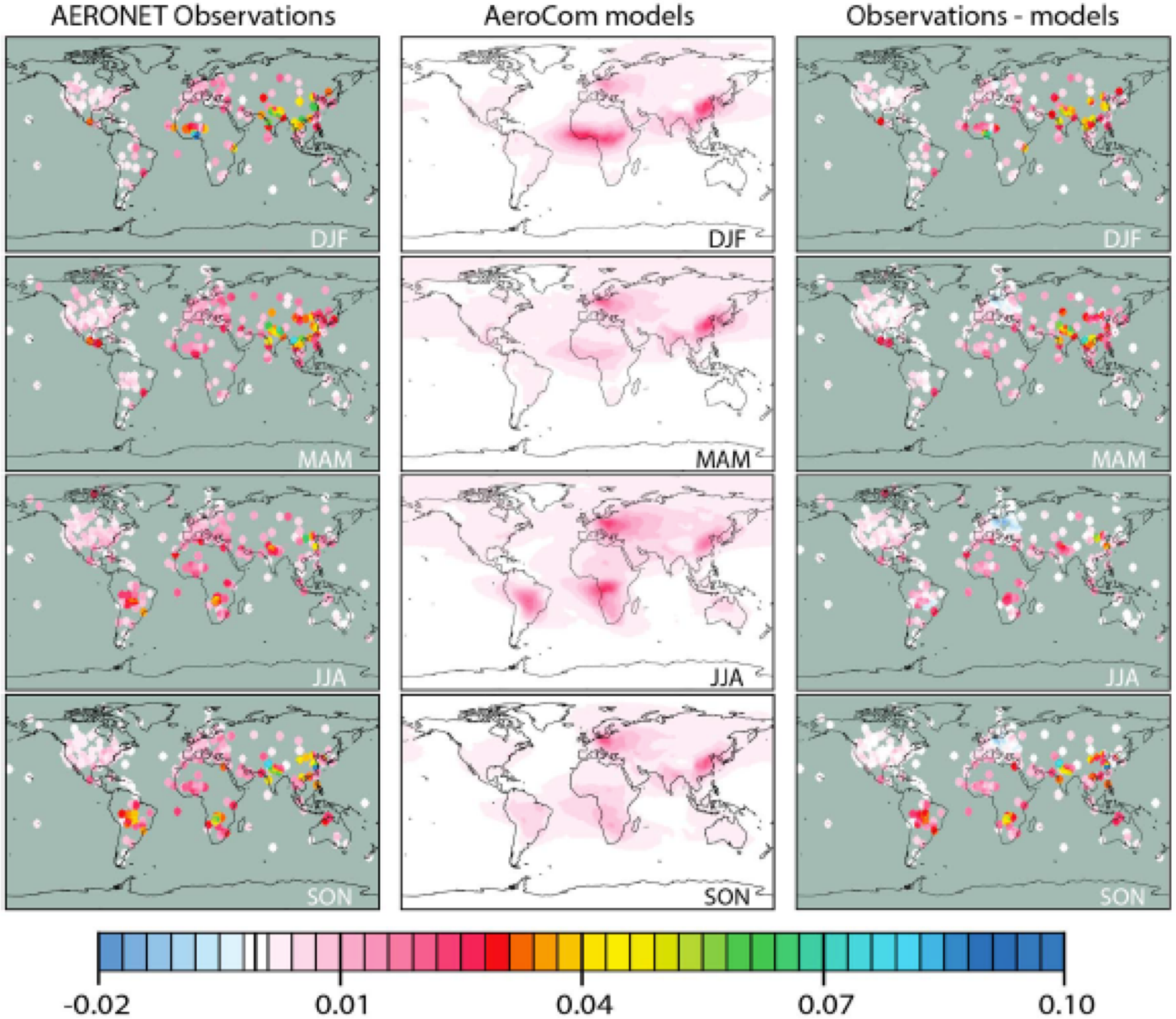
Observationally constrained direct forcing





Johnson et al. (ACP 2019) Robust observational constraint of uncertain aerosol processes and emissions in a climate model…

Example 3: Seasonal Distribution of BC Absorption Aerosol Optical Depth

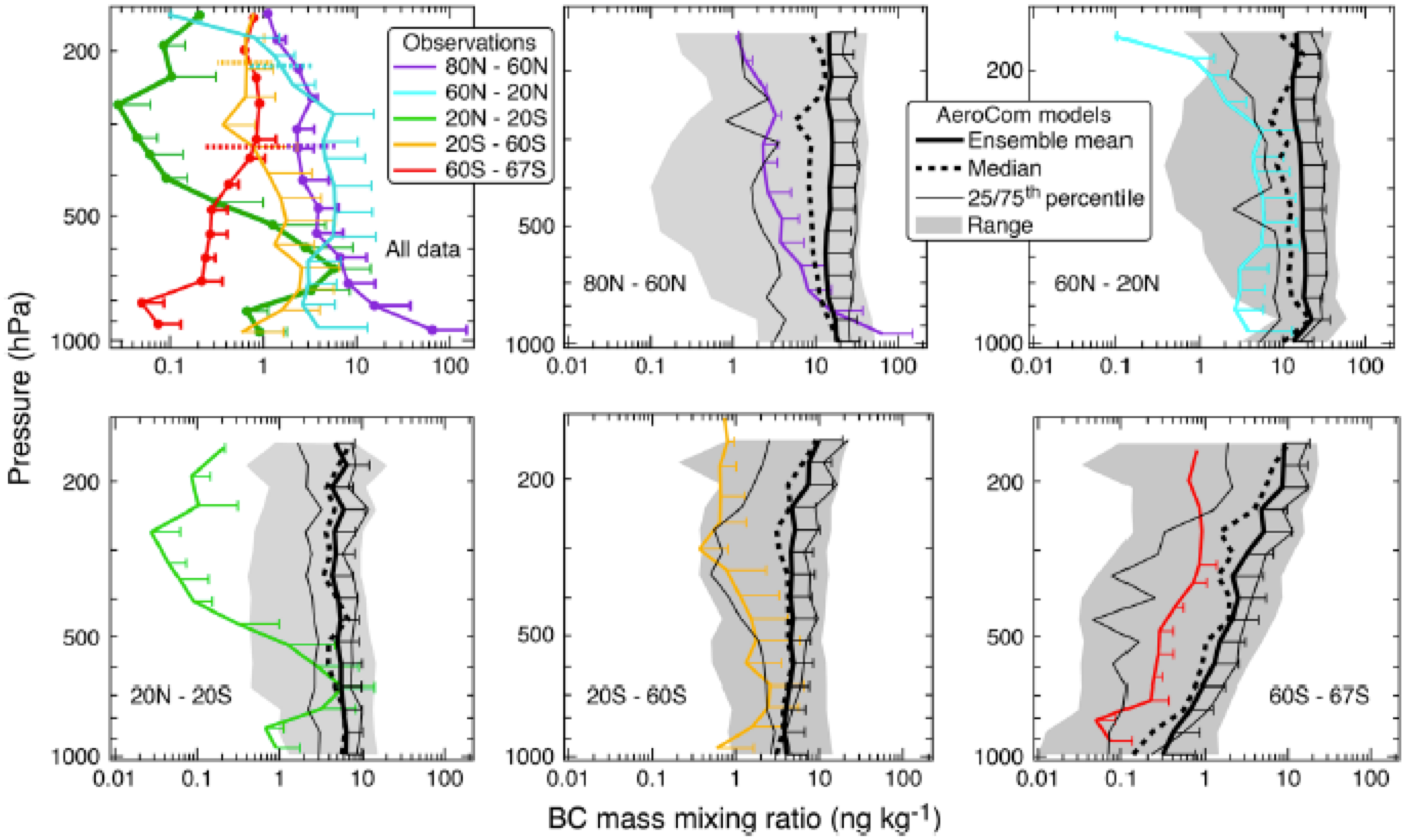


BC absorption aerosol optical depth (AAOD) and AAOD differences

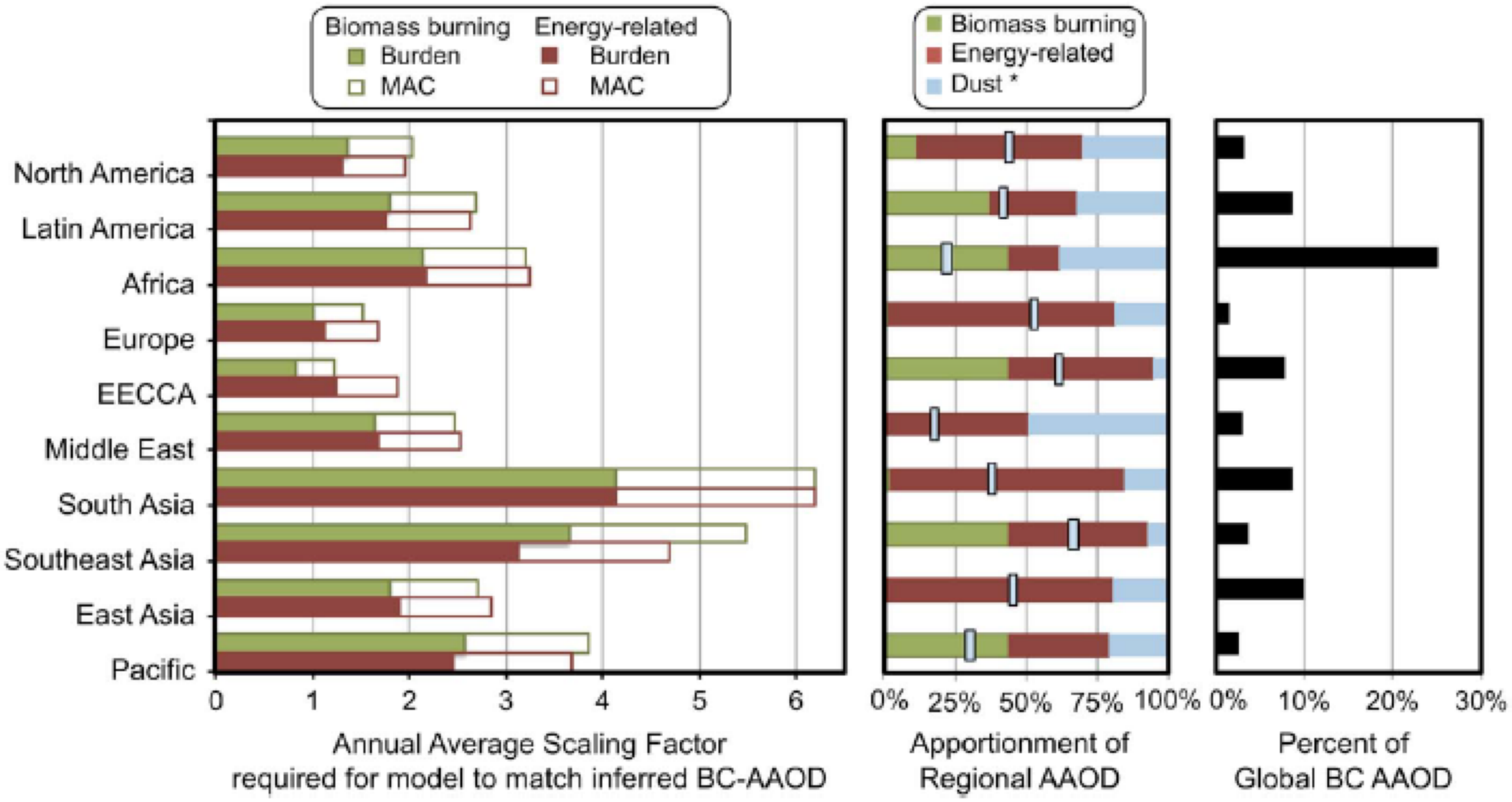
Bond et al., 2013

Comparison of BC Vertical Profiles with Aerocom Models

Schwartz et al., 2010



Model scaling and apportionment of BC-AAOD by region



Conclusions (what needs to be done)

- Create a Table of aerosol properties including not only global means and pdfs but also documenting key regions/ key long-term measuring sites
- Better account for/constrain water associated with the aerosol, examine how model represent HG, gather information from field/lab measuements in a single document
- Fine mode aerosol: optical properties of BC and sea salt should revised from the ones listed in the OPAC database. Trends, absorption and ageing of the components in the fine mode need more attention
- Large mode aerosol: Recent progress has been made in constraining the role of the large (> 10 μm) dust mode. It plays a large role in the SW and LW that few models have yet included; it will also affects heterogeneous chemistry. More information on mineralogical varations is needed (this could come from the EMIT mission / aerosol chamber measurements).
- Long-term observations at sites are not enough looked at by modellers. The representativeness of these sites could be a hot topic.

Tables of Aerosol Optics (TAO)

TAO Committee (largely assembled by Arlindo da Silva):

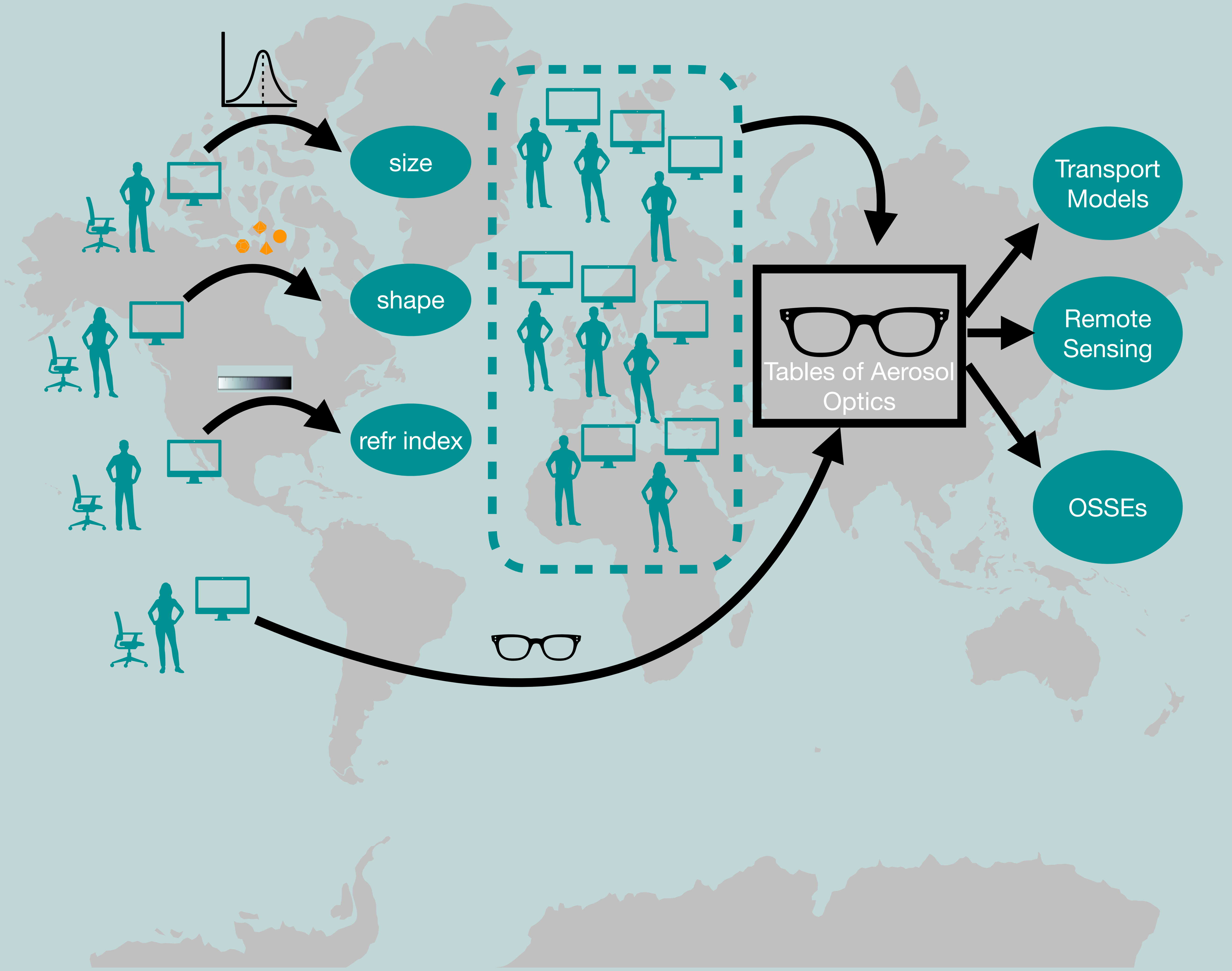
Greg Schuster
ARLINDO DA SILVA
Stefan Kinne
Mian Chin
Michael Schulz
Peter Colarco
Thomas Popp
Meloe Kacenelenbogen
Tero Mielonen
Yves Balkanski
Richard Moore
Jason Tackett
Osku Kemppinen
Masa Saito
Betsy Andrews

Send email to aerosol-optics-join@lists.nasa.gov to join:

- allows access to NASA Google Drive that hosts TAO
- email updates (infrequent)
- permission to add new tables to TAO

TAO Scheme

- The **Table of Aerosol Optics** is a community repository of optics computations that are useful for models and remote sensing (extinction, absorption, SSA, Lidar Ratio, etc).
- Expands upon historical efforts (Shettle and Fenn, d'Almeida, GADS, OPAC, etc) by building a database that includes recent measurements and new computational techniques for non-spherical particles.
- Originally conceived by Arlindo da Silva, TAO is meant to be a community repository where scientists can put their computations.
- Quality control will occur through corresponding literature.
- TAO will also provide links to other repositories, e.g.:
  - http://aram.ess.sunysb.edu/tglotch/optical_constants.html,
  - http://www.astro.uni-jena.de/Laboratory/OCDB/carbon.html
- It is expected that TAO will include computations for traditional aerosol species (amm sulfate, amm nitrate, organics, etc.), but TAO will also accept computations for aerosol 'type.'
- Presently, TAO is highly fluid and located on my NASA google drive. This is not permanent. TAO will eventually establish a home (e.g., GitHub)
- TAO is also part of the new Models, In situ, and Remote sensing of Aerosols (MIRA) working group (more on that later).



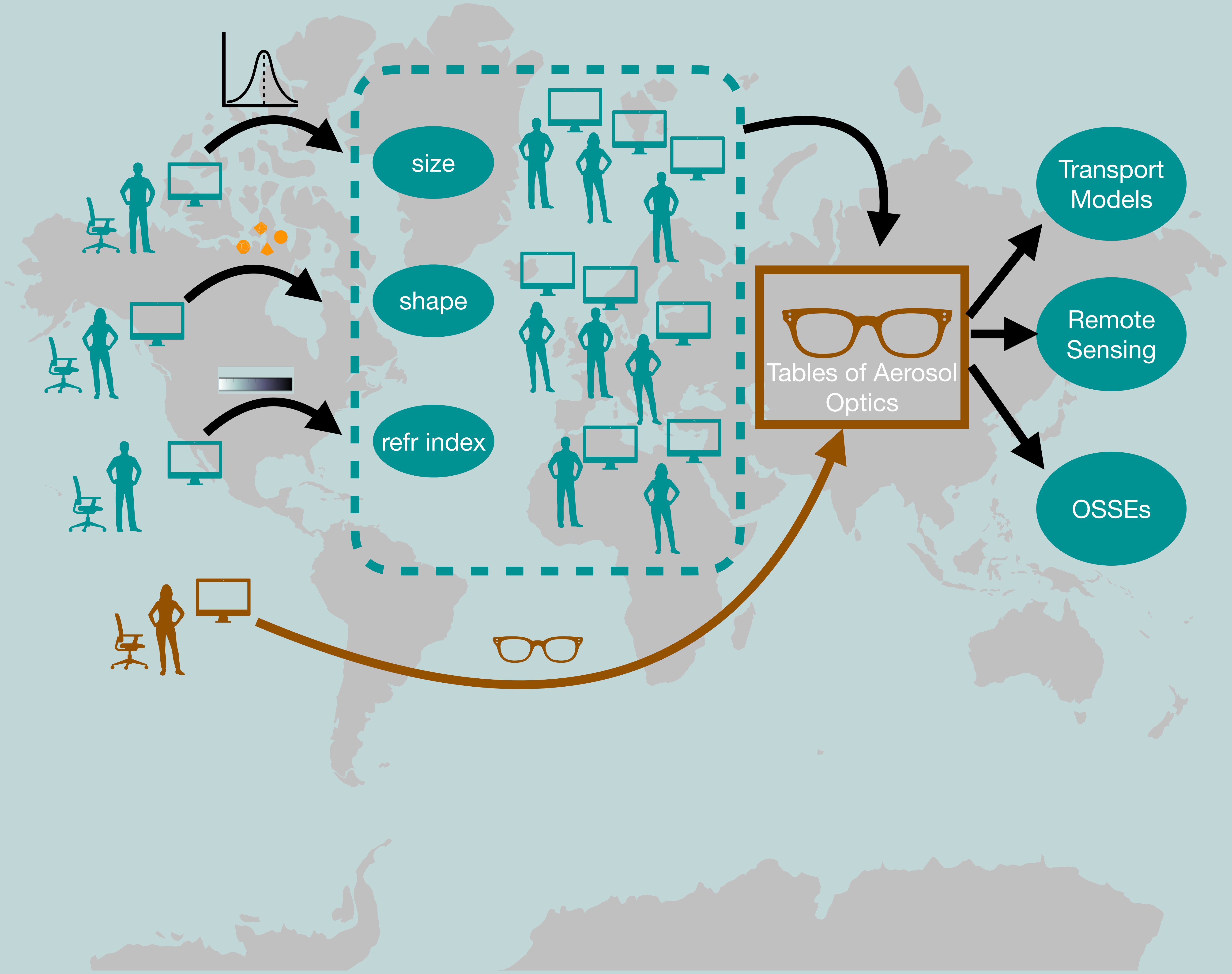
Send email to aerosol-optics-join@lists.nasa.gov to join:

- allows access to NASA Google Drive that hosts TAO
- email updates (infrequent)
- permission to add new tables to TAO

TAO Scheme

TAO is a community database.
What we seek from the community:

- Existing tables that modelers are using
- Additional tables (especially non-spheres)
- Measurements (firsthand or from the literature)
- Single-scatter computations (spheres, irregular dust, fractal BC, internal mixtures, etc.).
- Customers and "Special orders."



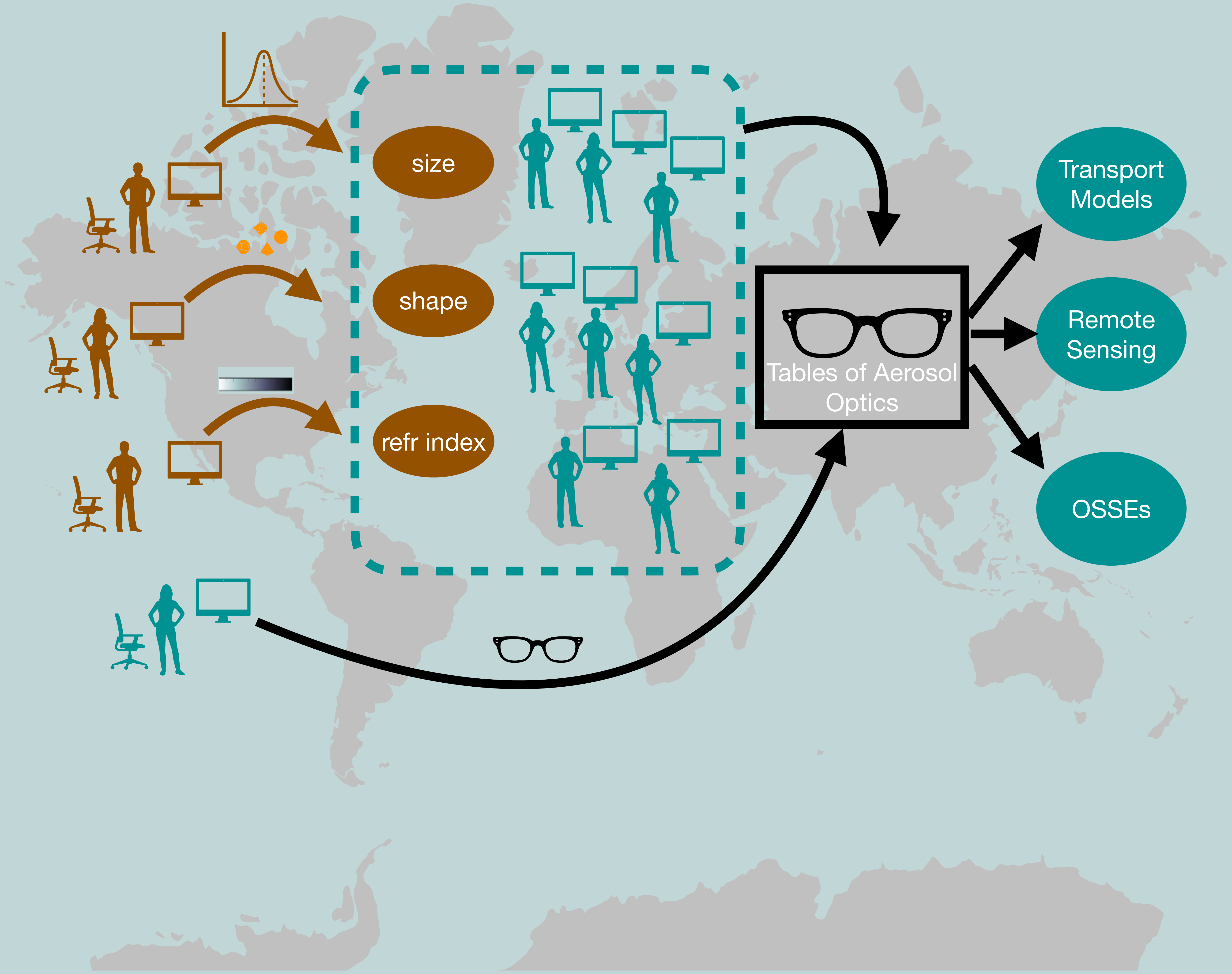
Send email to aerosol-optics-join@lists.nasa.gov **to join:**

- allows access to NASA Google Drive that hosts TAO
- email updates (infrequent)
- permission to add new tables to TAO

TAO Scheme

TAO is a community database.
What we seek from the community:

- Existing tables that modelers are using
- Additional tables (especially non-spheres)
- Measurements (firsthand or from the literature)
- Single-scatter computations (spheres, irregular dust, fractal BC, internal mixtures, etc.).
- Customers and "Special orders."



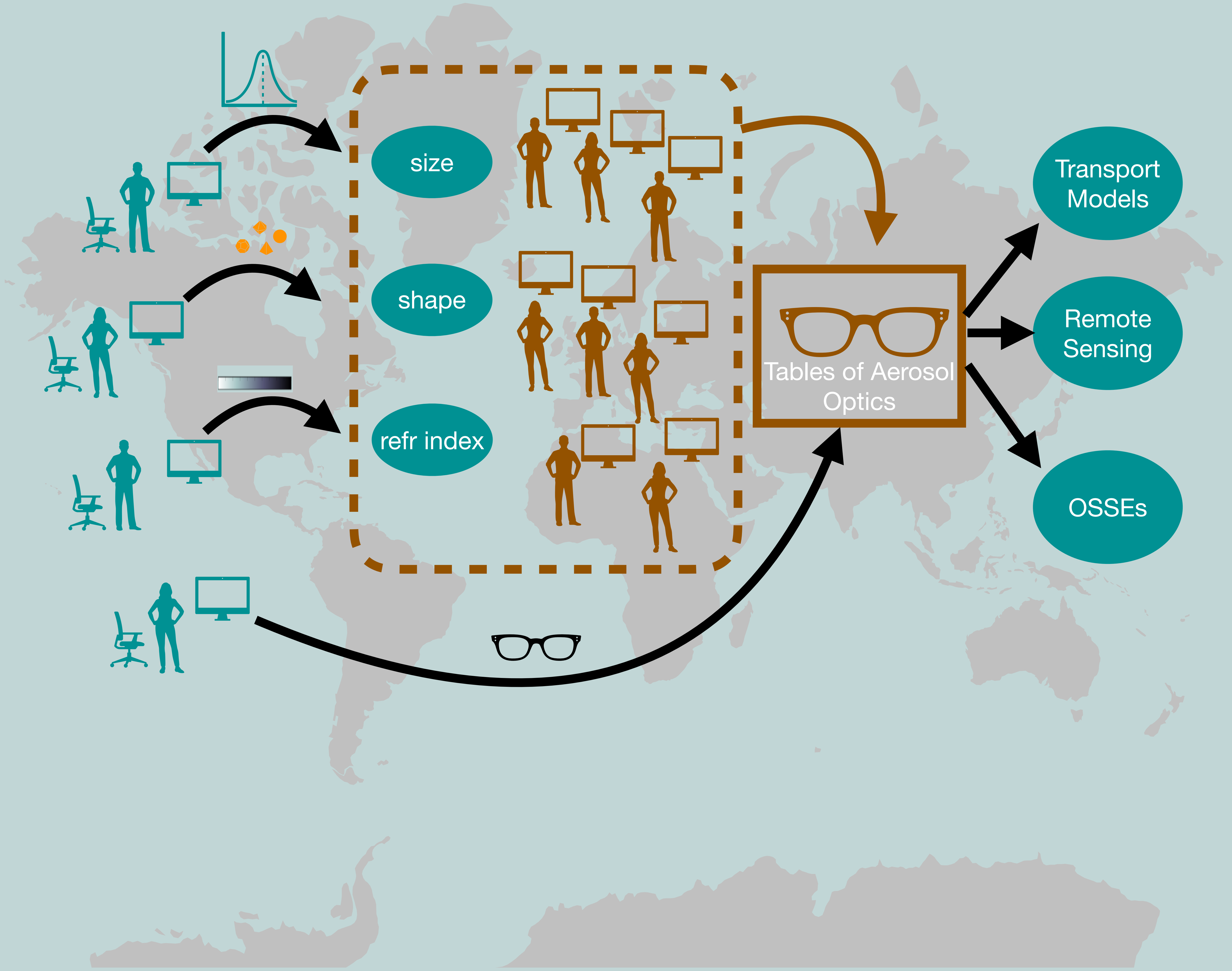
Send email to aerosol-optics-join@lists.nasa.gov **to join:**

- allows access to NASA Google Drive that hosts TAO
- email updates (infrequent)
- permission to add new tables to TAO

TAO Scheme

TAO is a community database.
What we seek from the community:

- Existing tables that modelers are using
- Additional tables (especially non-spheres)
- Measurements (firsthand or from the literature)
- Single-scatter computations (spheres, irregular dust, fractal BC, internal mixtures, etc.).
- Customers and "Special orders."



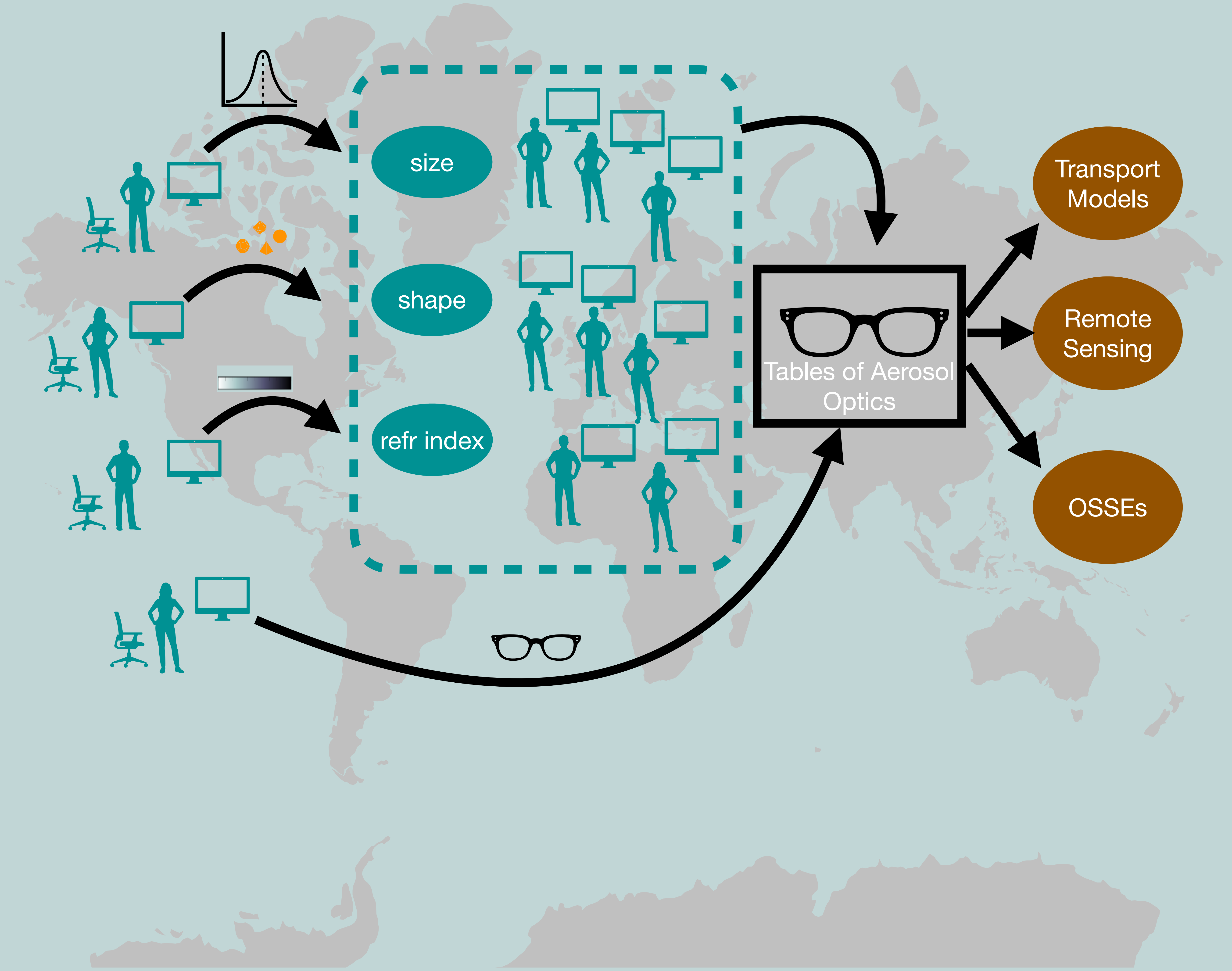
Send email to aerosol-optics-join@lists.nasa.gov to join:

- allows access to NASA Google Drive that hosts TAO
- email updates (infrequent)
- permission to add new tables to TAO

TAO Scheme

TAO is a community database.
What we seek from the community:

- Existing tables that modelers are using
- Additional tables (especially non-spheres)
- Measurements (firsthand or from the literature)
- Single-scatter computations (spheres, irregular dust, fractal BC, internal mixtures, etc.).
- Customers and "Special orders."



Send email to aerosol-optics-join@lists.nasa.gov **to join:**

- allows access to NASA Google Drive that hosts TAO
- email updates (infrequent)
- permission to add new tables to TAO

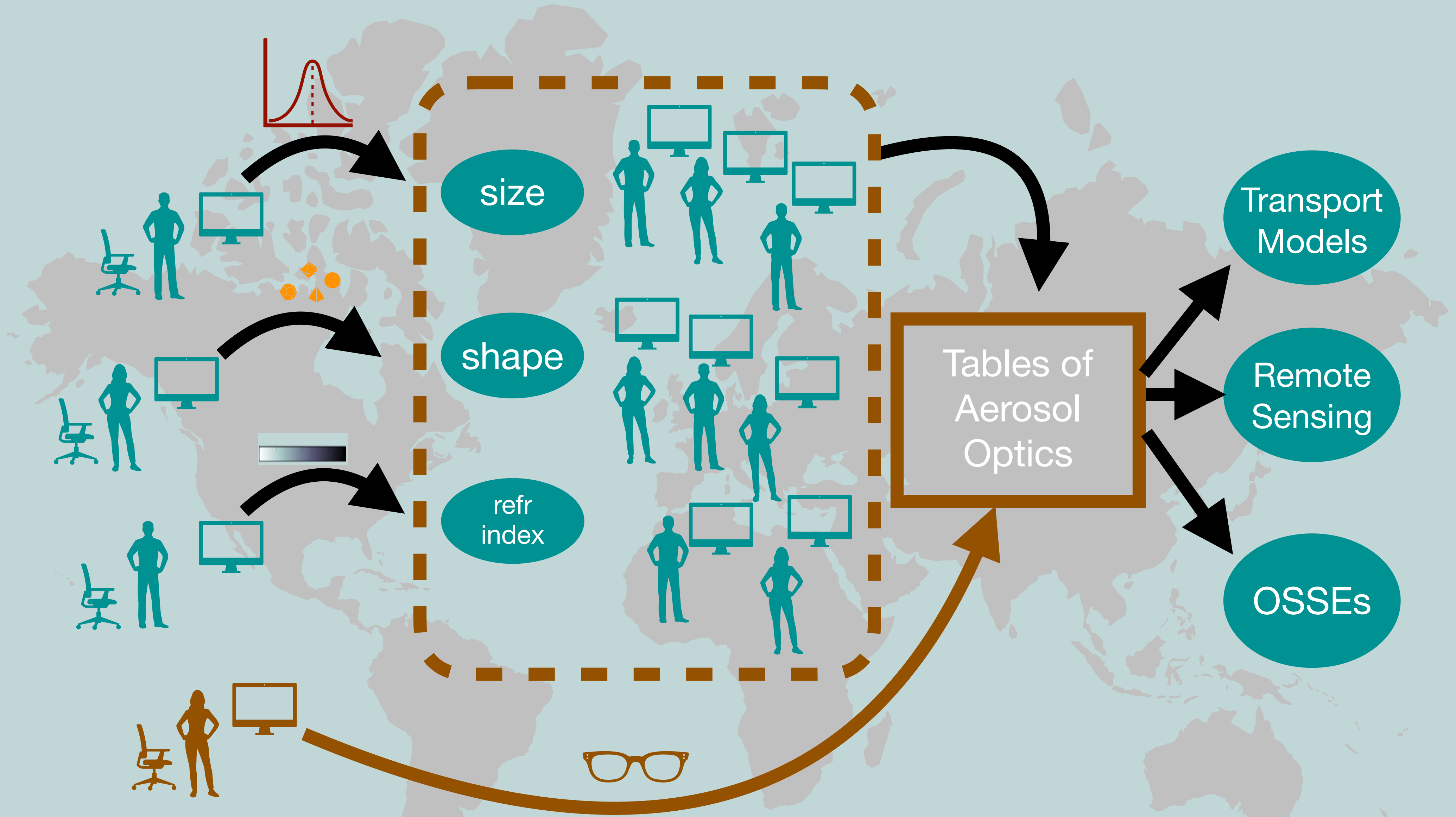
Example: Mie Theory

Processed 36 lognormals so far:

- We have created aerosol optical tables that include mass extinction, absorption, and backscatter coefficients, single-scatter albedos, etc.,
- Based upon Rissler (ACP, 2006), kappa growth, and Mie Theory.

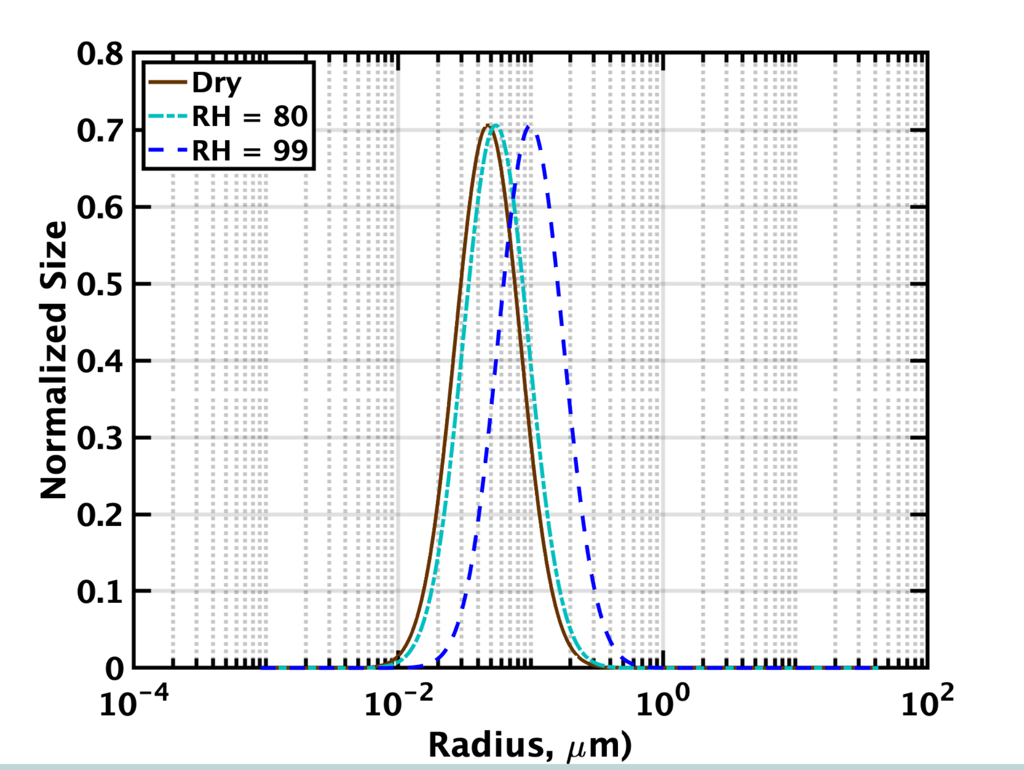
✓Water-insoluble Brown Carbon
✓Water-soluble Brown Carbon
✓Water-insoluble "White" Carbon
✓Water-soluble "White" Carbon
} 36 SDs, CRIs, & kappas Amazon (Rissler, ACP, 2006)

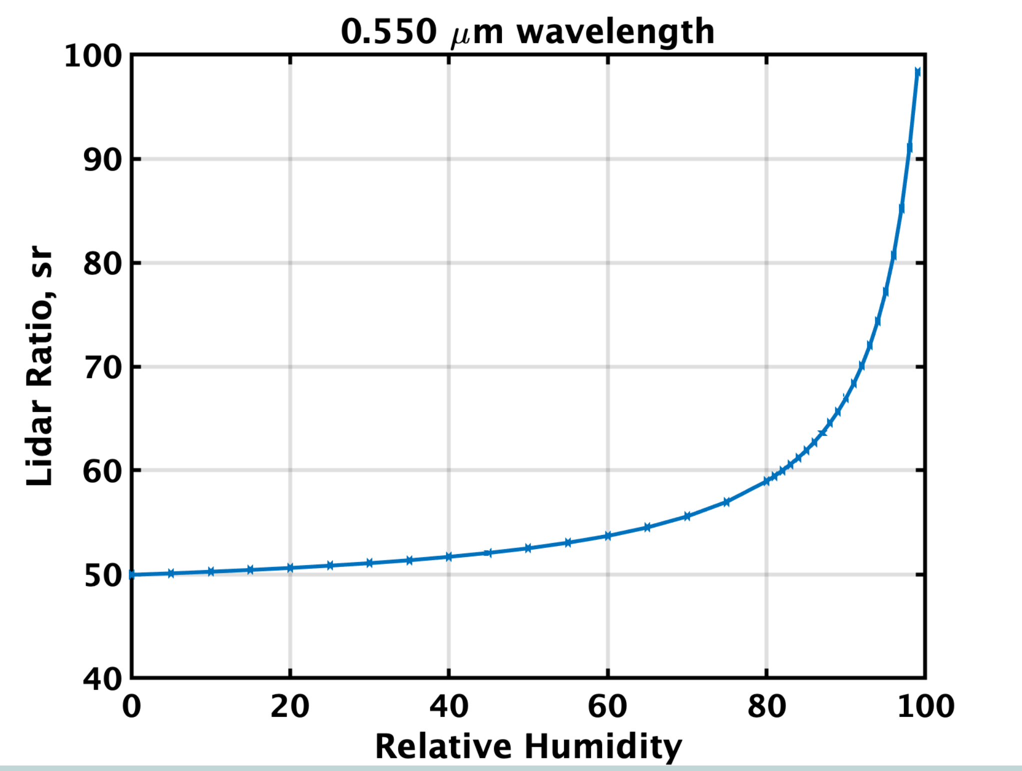
- Externally-mixed Black Carbon
- Internally-mixed Black Carbon
- Multi-mineral dust mixtures (non-spheres) SAMUM, AERONET
- Sulfates
- Nitrates
- Sea salt (some RRI issues) Brock (ACPD, 2021)

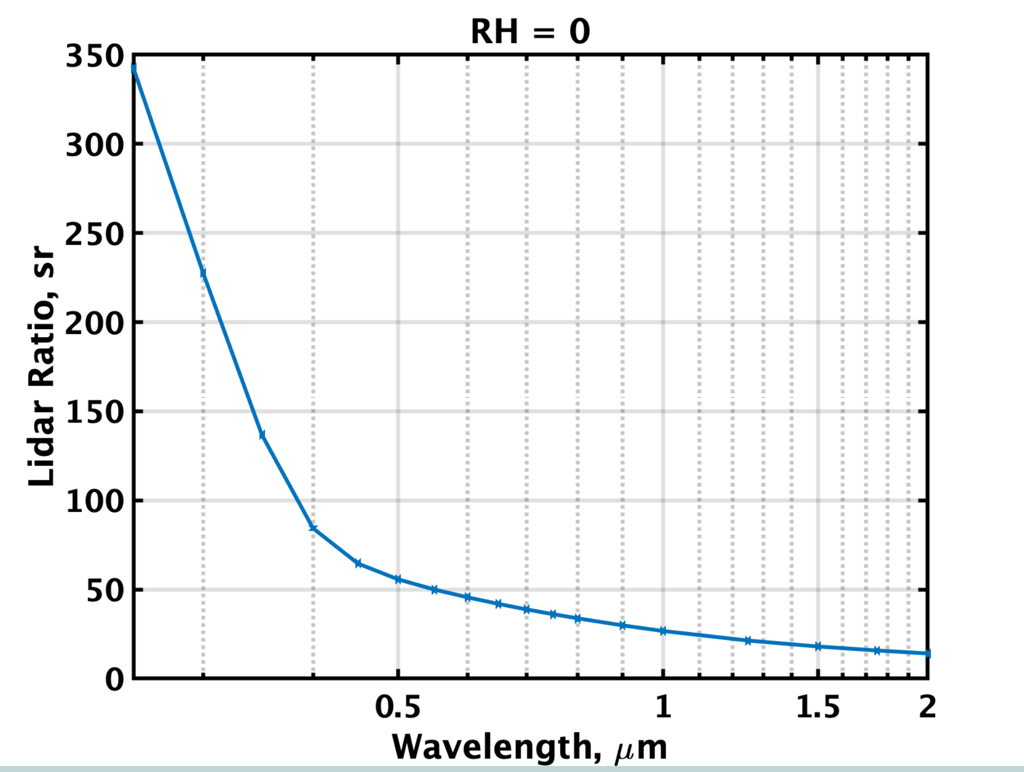


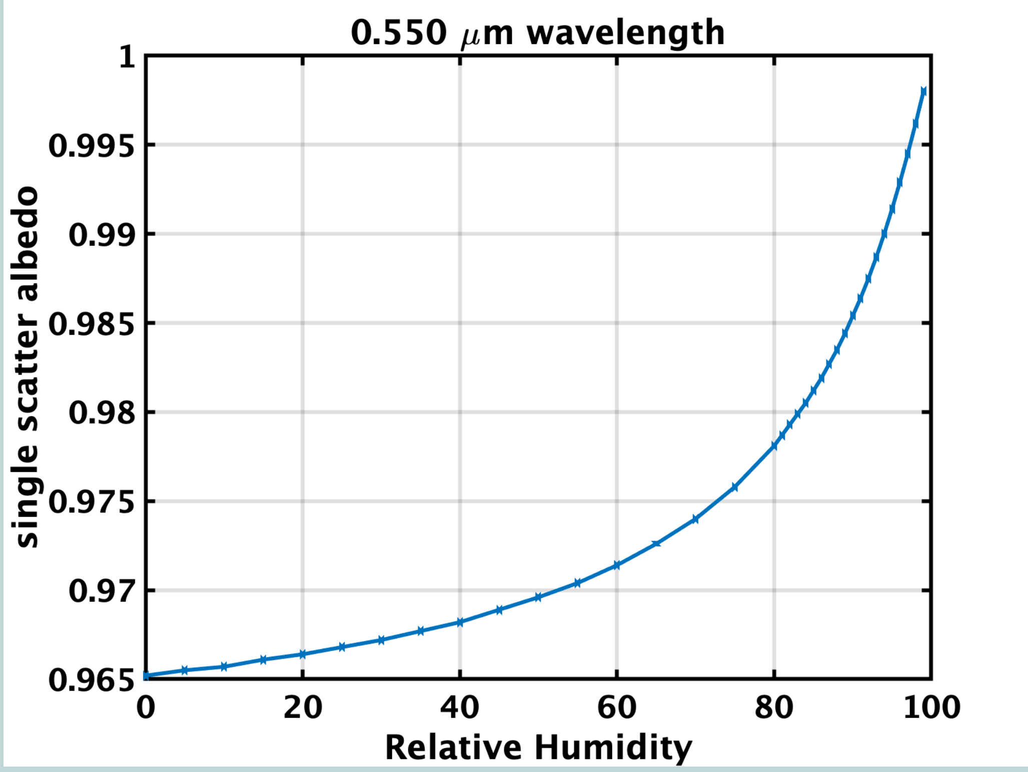
Example

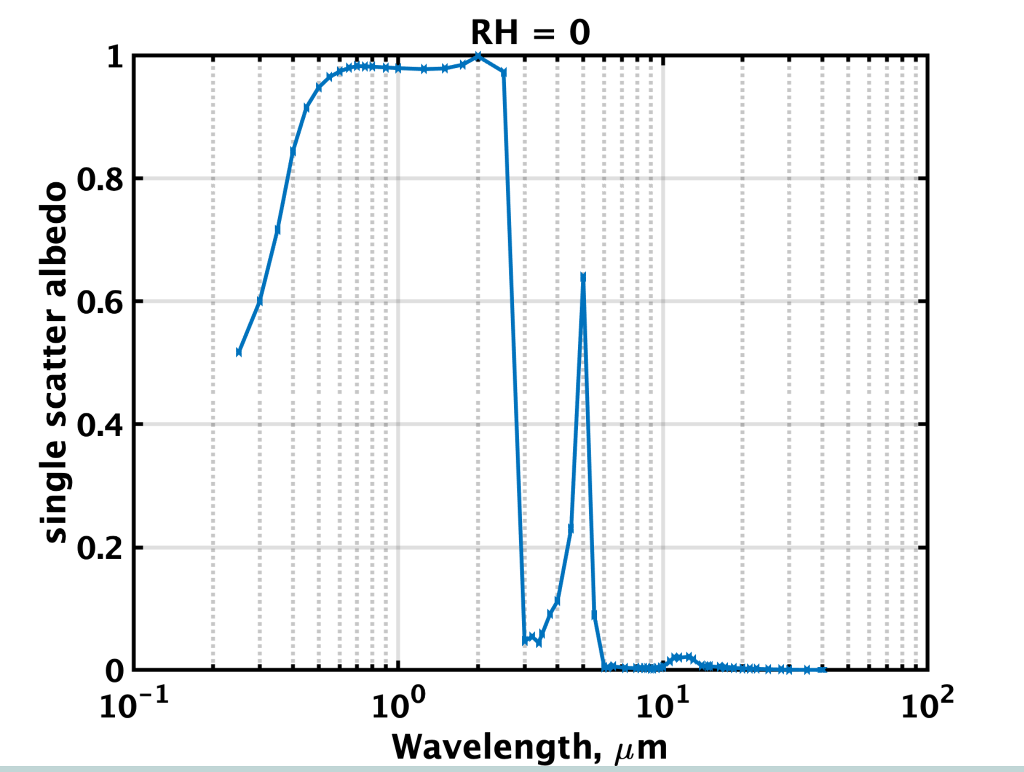
Water-soluble Brown Carbon



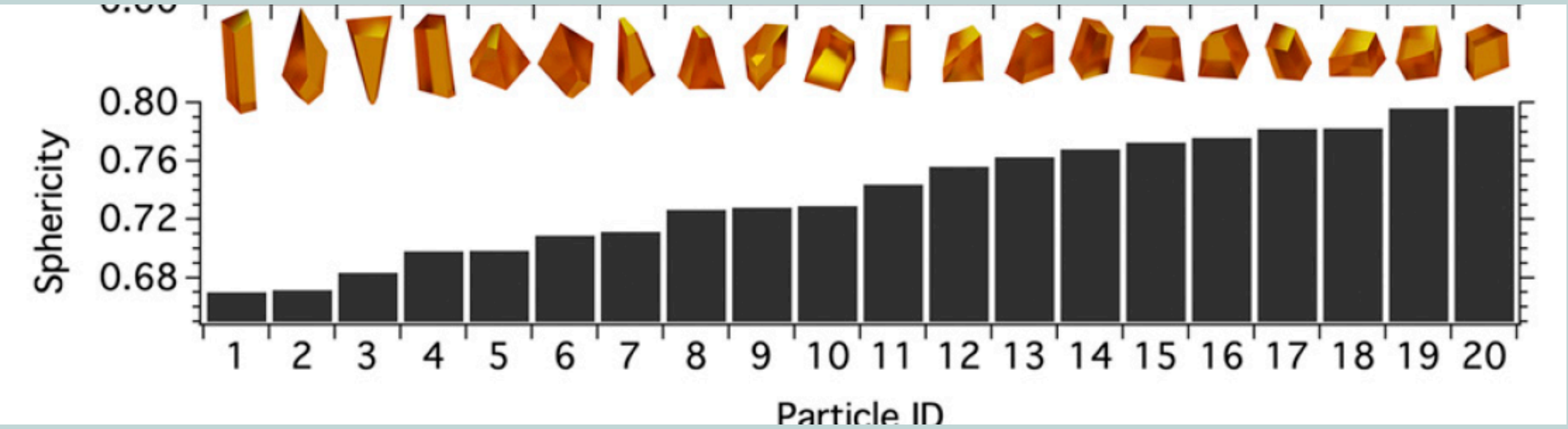








Texas A&M Hexahedrals for Mineral Dust - First Irregular Shapes for TAO!



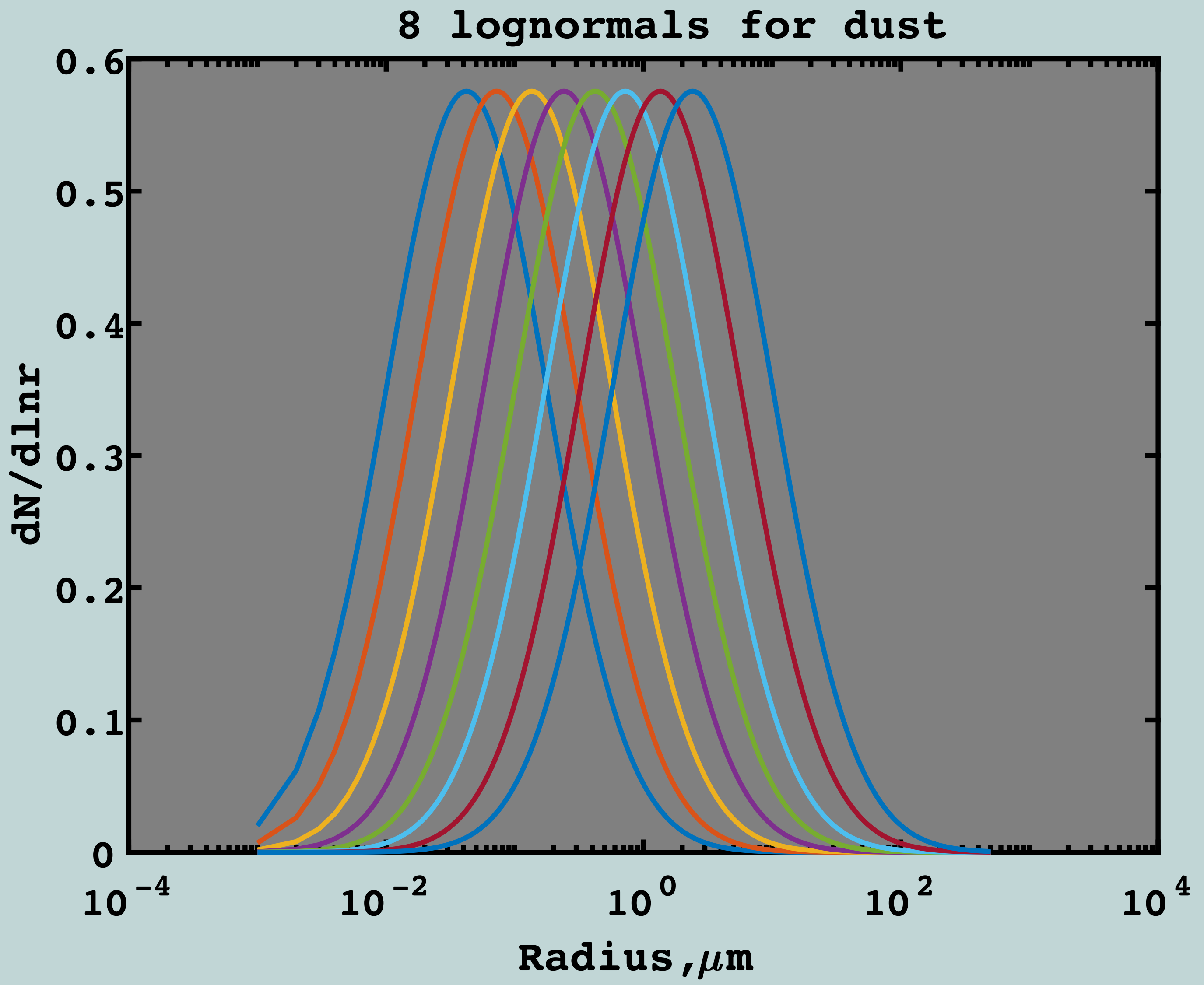
Saito, M., Yang, P., Ding, J., and Liu, X.:
A Comprehensive Database of the Optical Properties of Irregular Aerosol Particles for Radiative Transfer Simulations, J. Atmos. Sci., 78, https://doi.org/10.1175/JAS- D-20-0338.1, 2021.

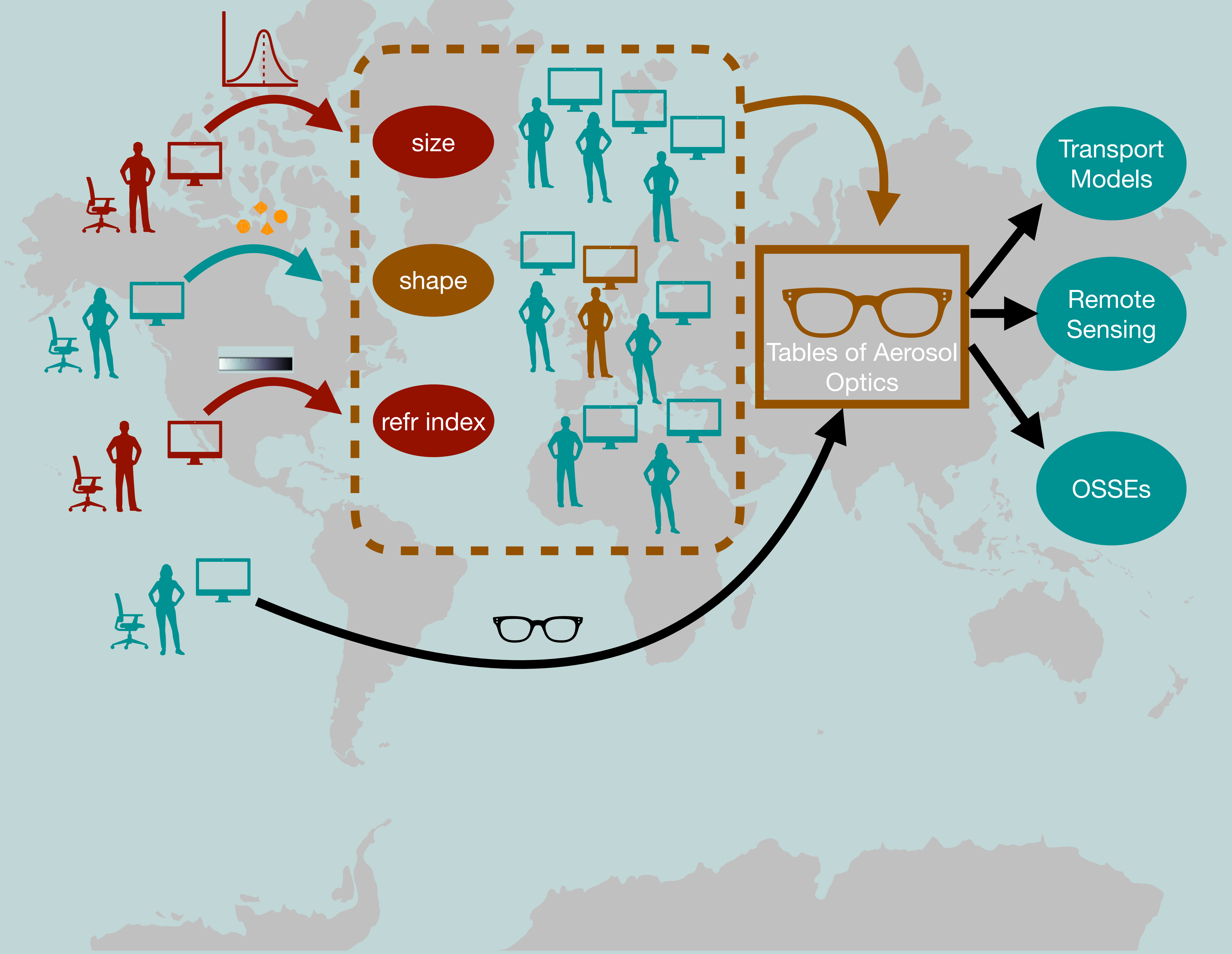
Saito, M. and Yang, P.:
Advanced Bulk Optical Models Linking the Backscattering and Microphysical Properties of Mineral Dust Aerosol, GRL, https://doi.org/10.1029/2021GL095121, 2021.

Mode parameters from Chin(2009):

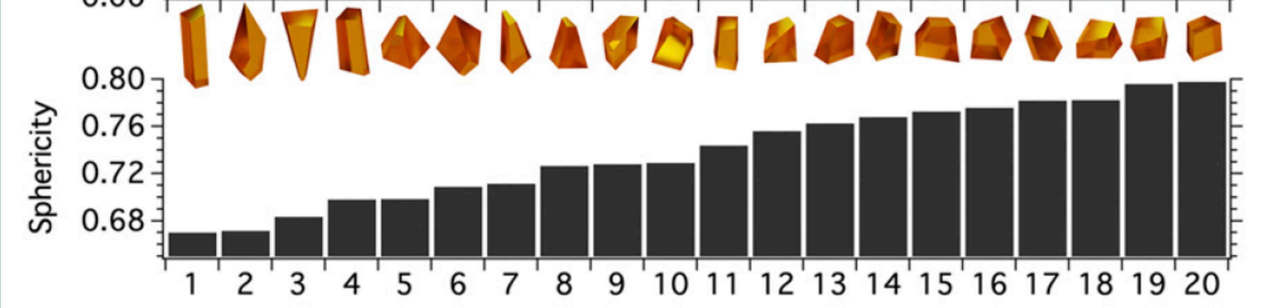
$$\sigma_g = 2.0$$

R_mdn	R_eff
0.0421	0.14
0.0722	0.24
0.1354	0.45
0.2407	0.80
0.4212	1.40
0.7220	2.40
1.3540	4.50
2.4070	8.00





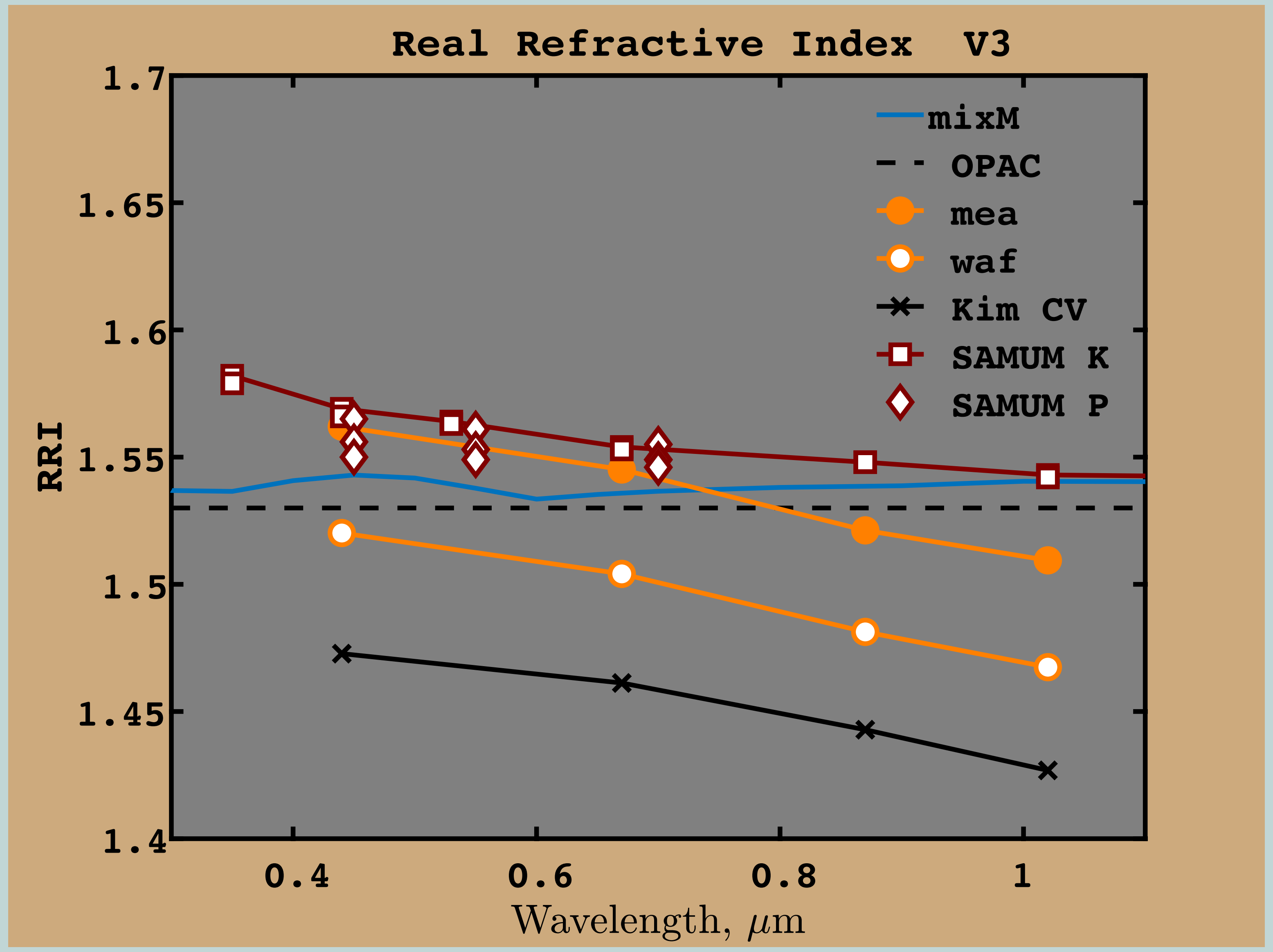
$$\frac{dN}{d\ln r} = \frac{N}{\sqrt{2\pi}\ln\sigma_g}\exp\left(-\frac{(\ln r - \ln r_m)^2}{2\ln^2\sigma_g}\right).$$

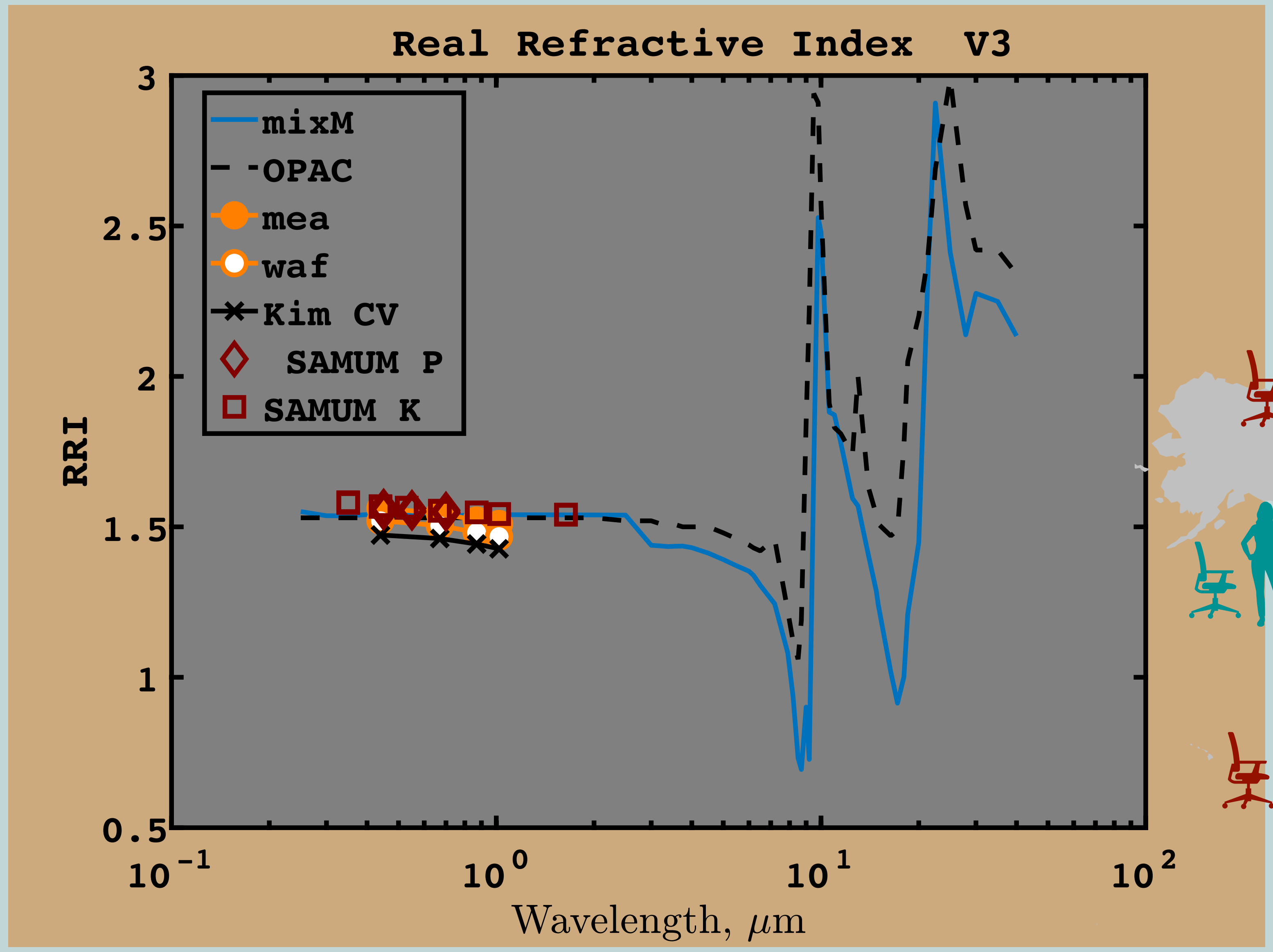


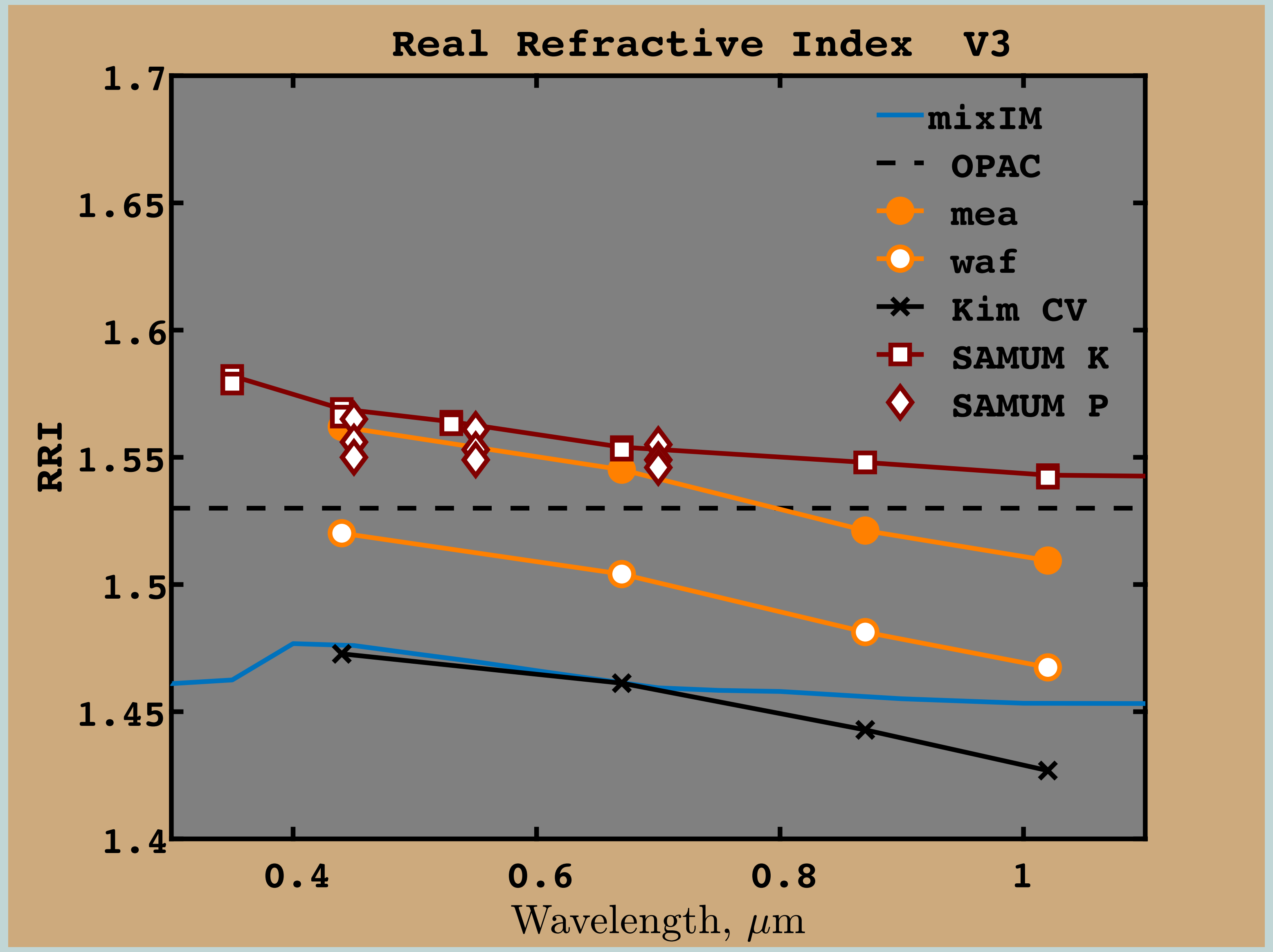
Saito, M., Yang, P., Ding, J., and Liu, X. (JAS 2021)
Saito, M. and Yang, P. (GRL 2021)

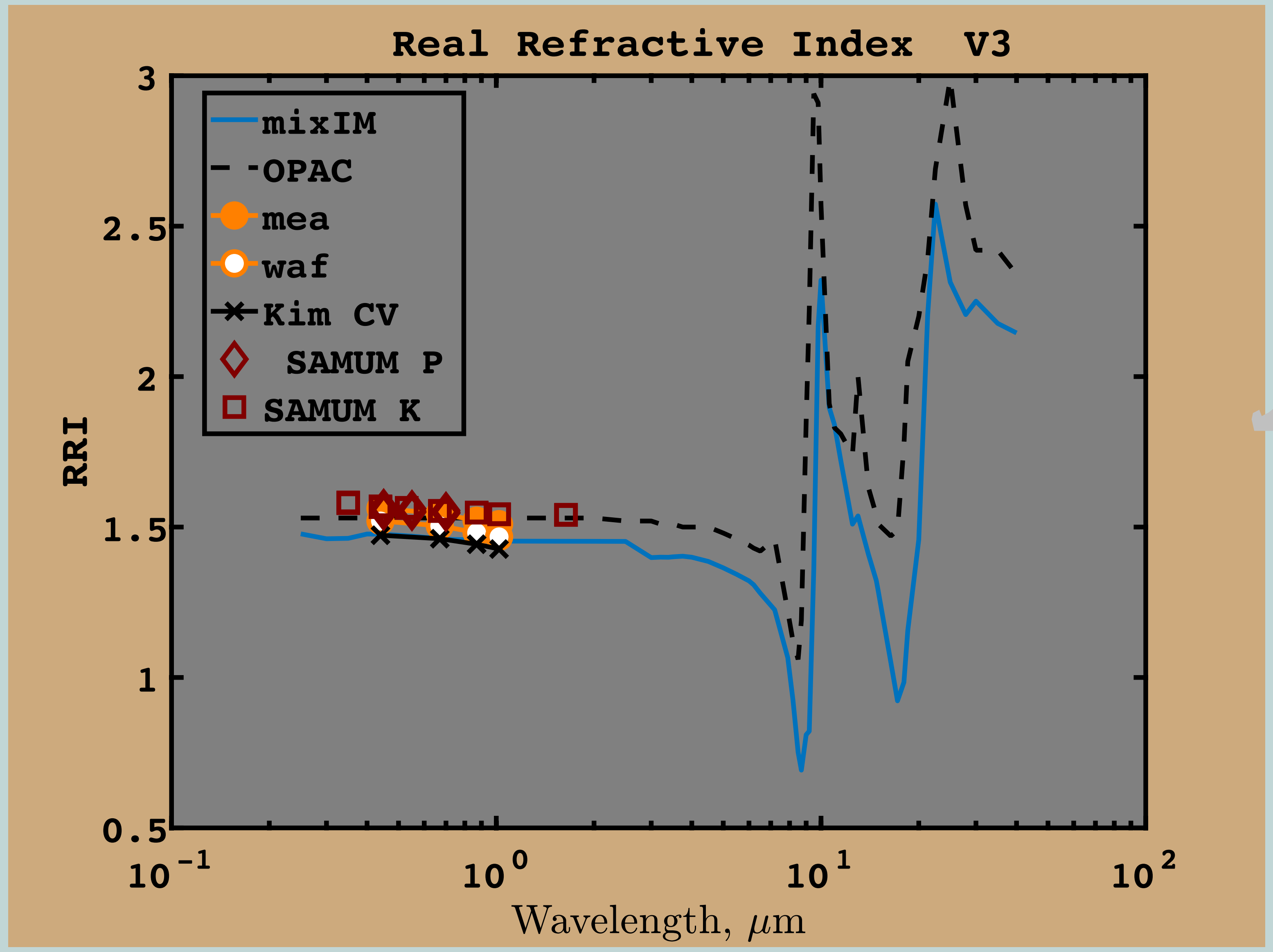
Texas A&M Hexahedrals

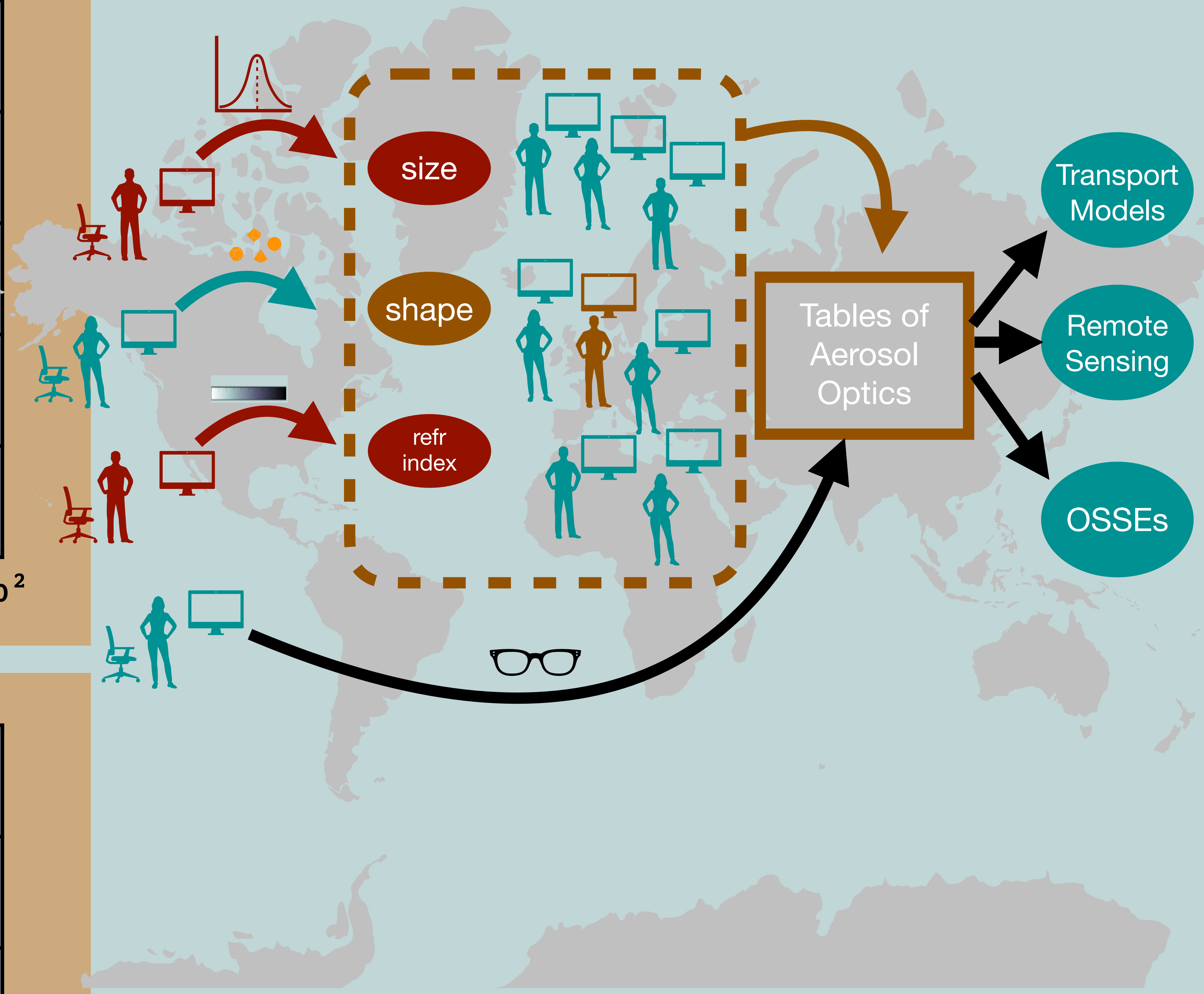
First Irregular Shapes for TAO

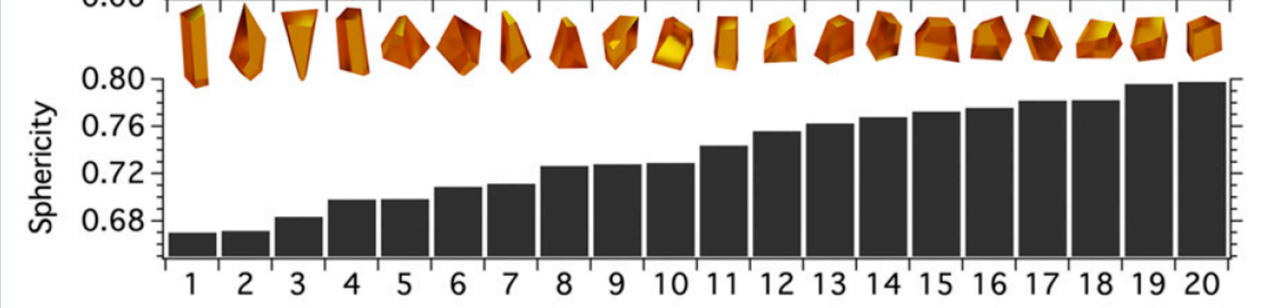








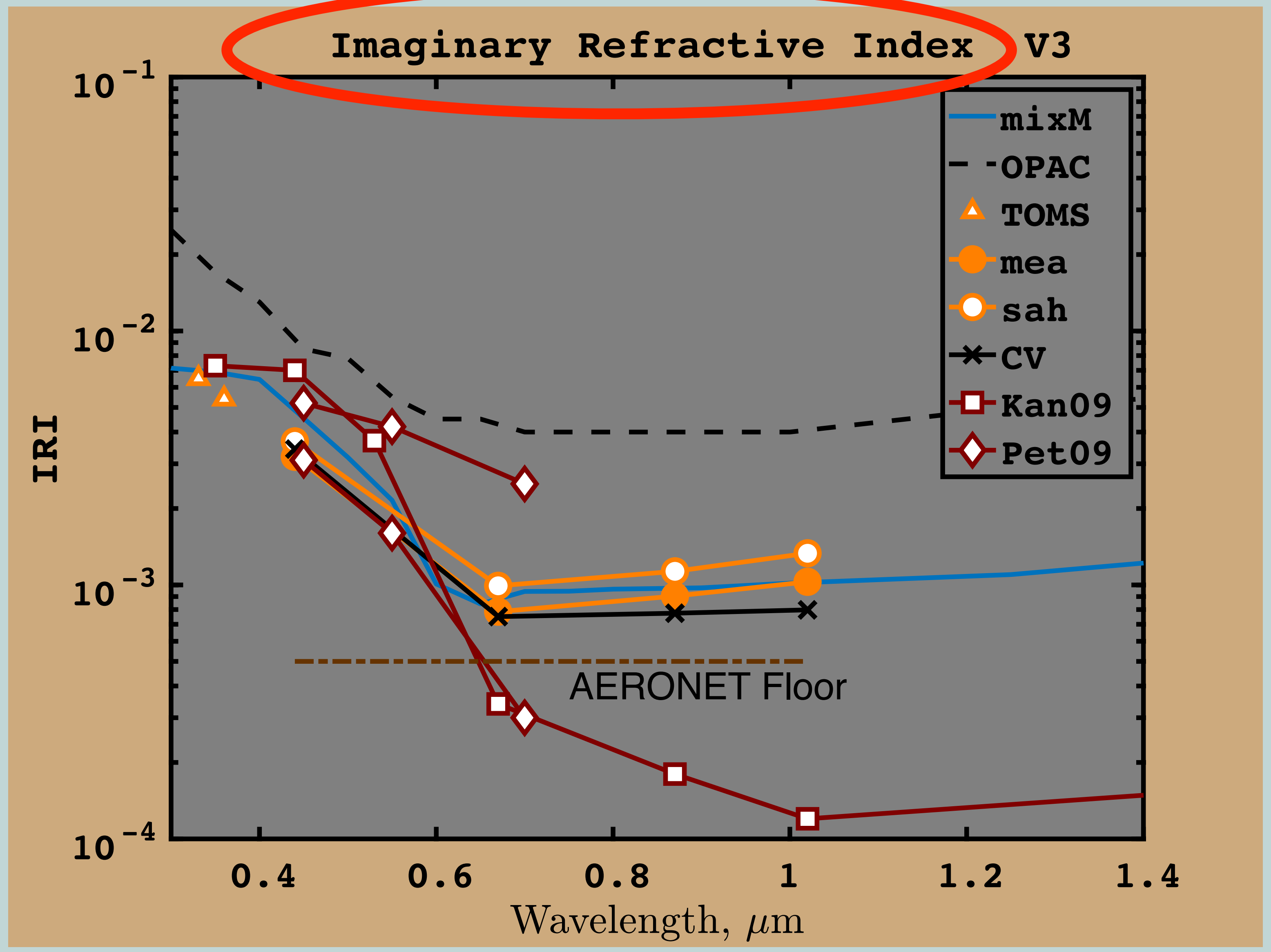


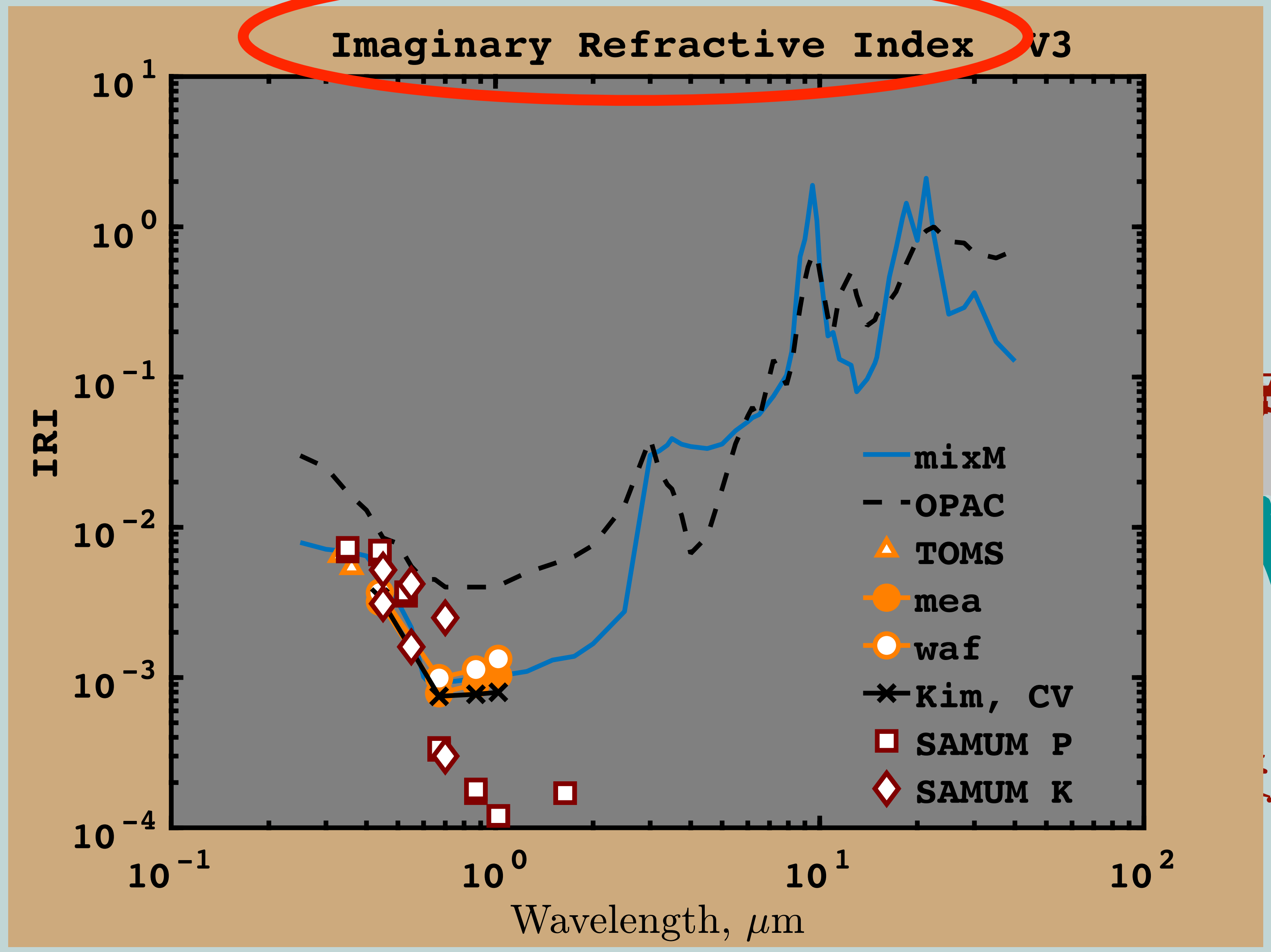


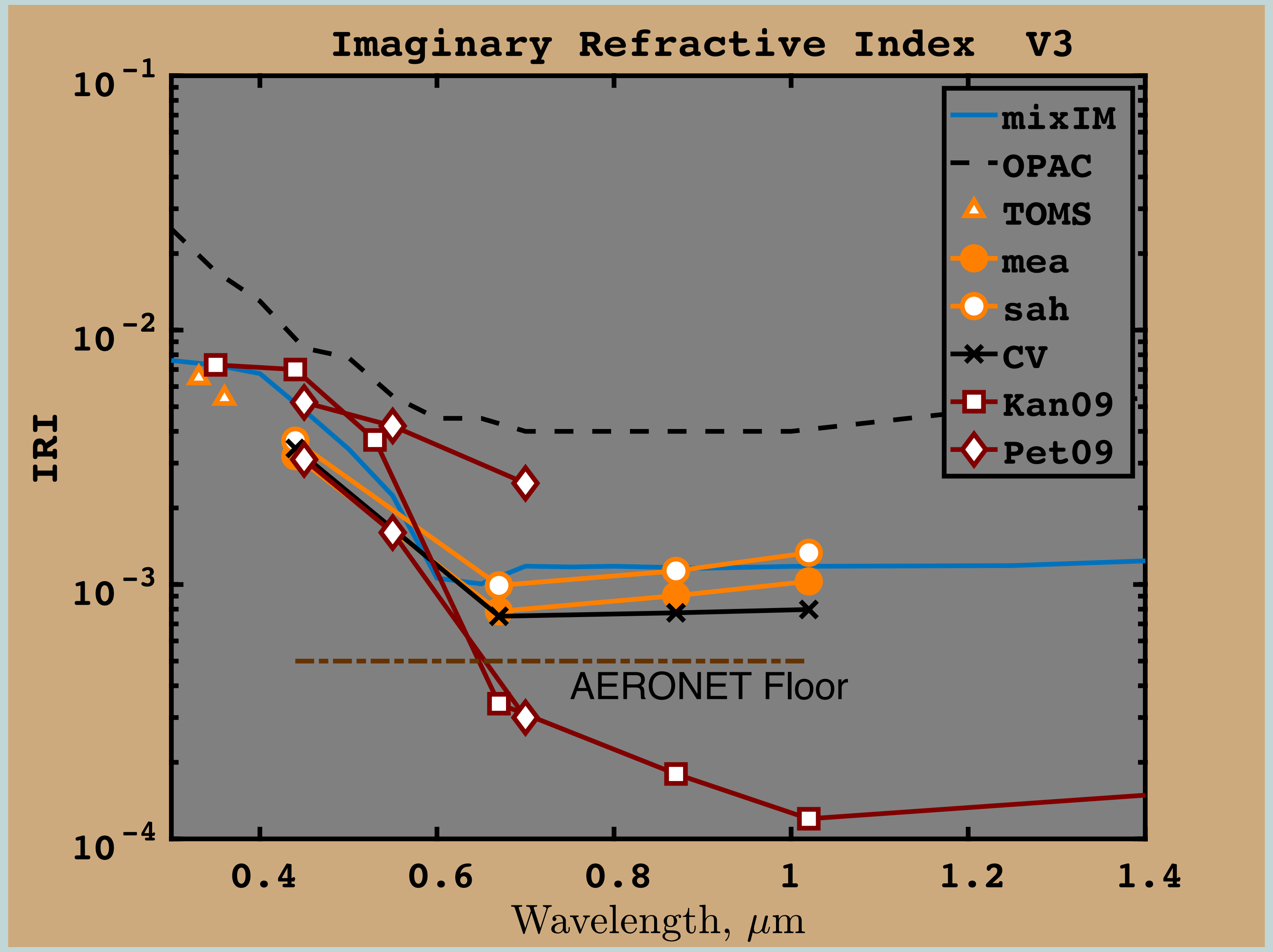
Saito, M., Yang, P., Ding, J., and Liu, X. (JAS 2021)
Saito, M. and Yang, P. (GRL 2021)

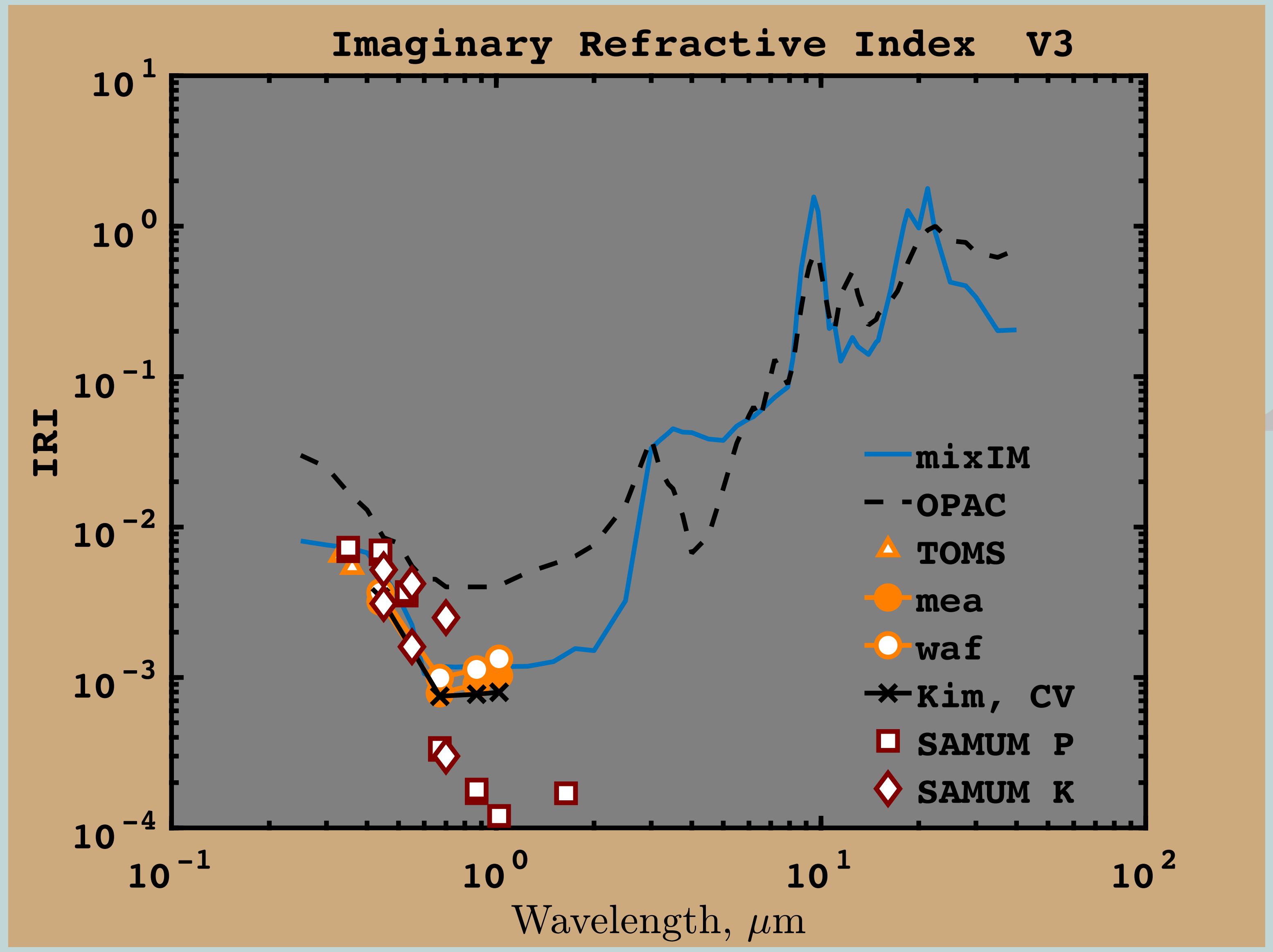
Texas A&M Hexahedrals

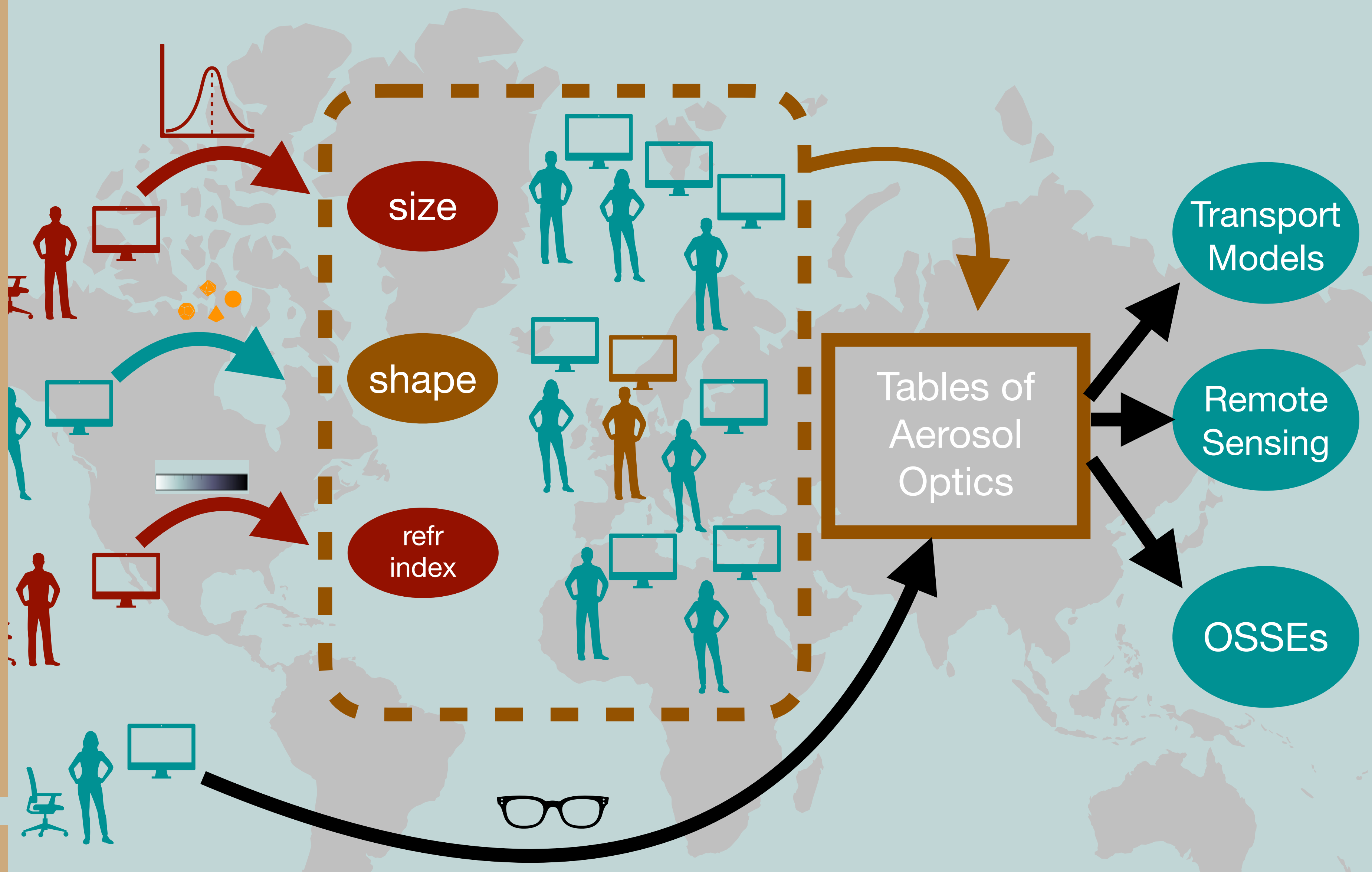
First Irregular Shapes for TAO

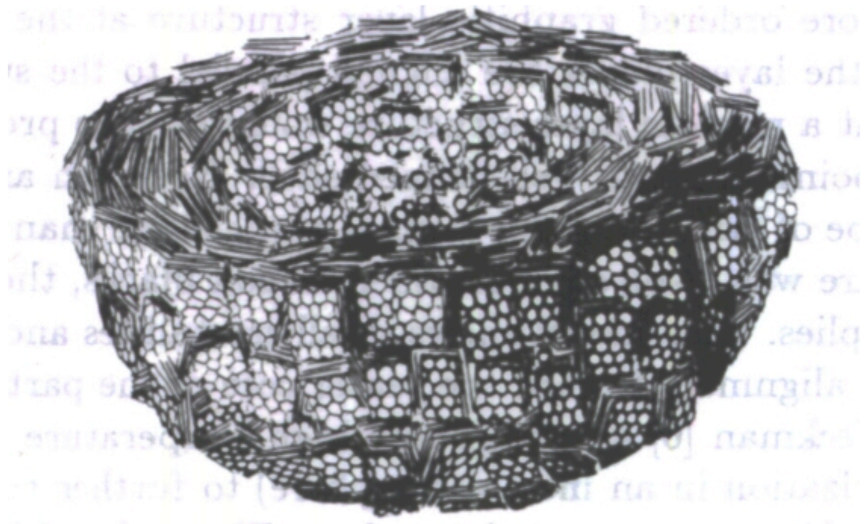












Hess and Herd, Carbon Black (1993)

What is Black Carbon?

- Byproduct of incomplete combustion
  - fossil fuel burning
  - biomass burning
- Graphitized
- Other names:

carbon blacks	produced in controlled conditions
soot	atmospheric; contains impurities
elemental carbon	measured by thermal analysis
black carbon	measured by optical absorption

Schuster, Dubovik, Holben, First AERONET Workshop, Spain, 2004

F 20 - 6 LI ET AL.: COMPOSITIONS AND AGING OF INORGANIC PARTICLES

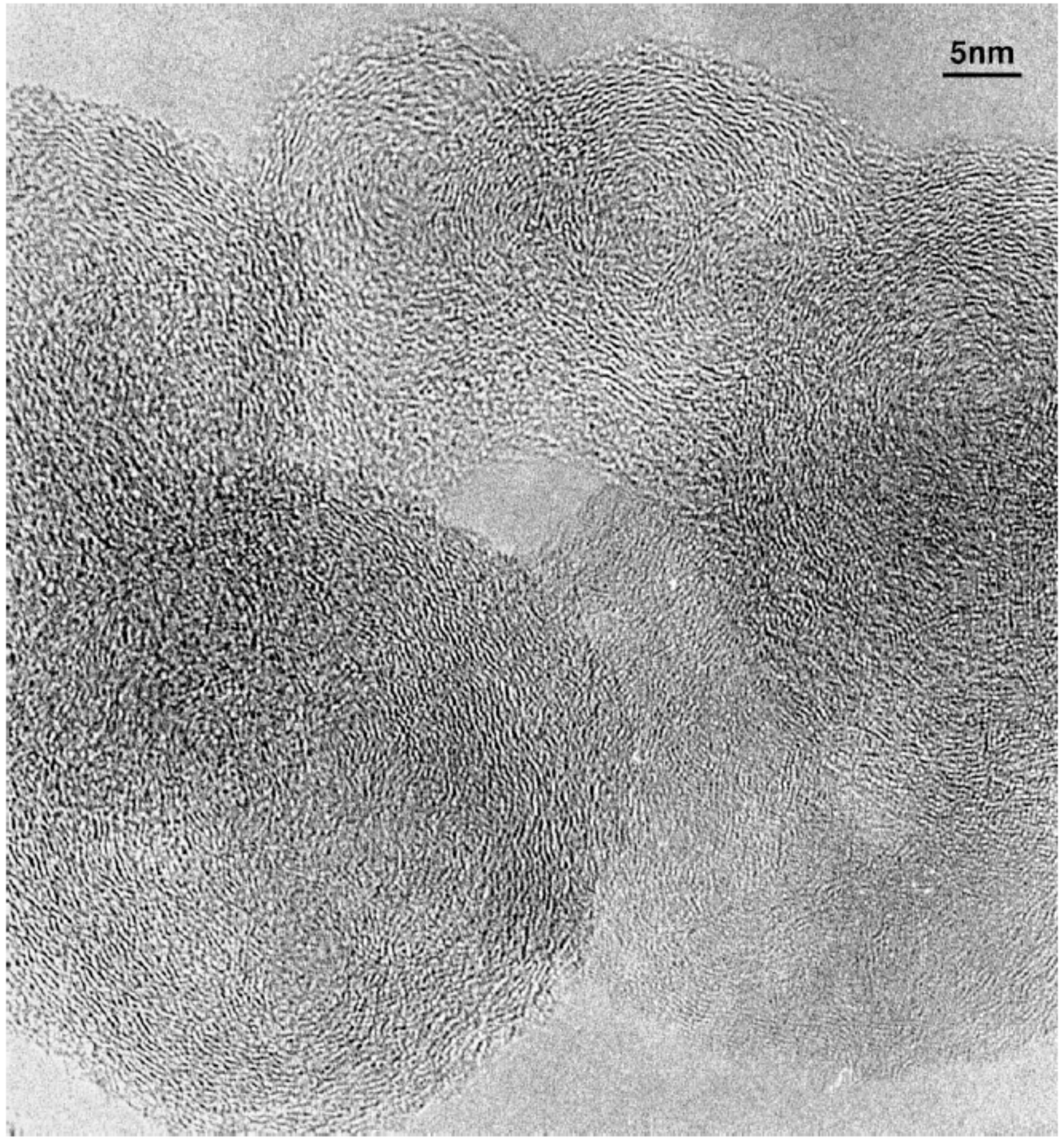
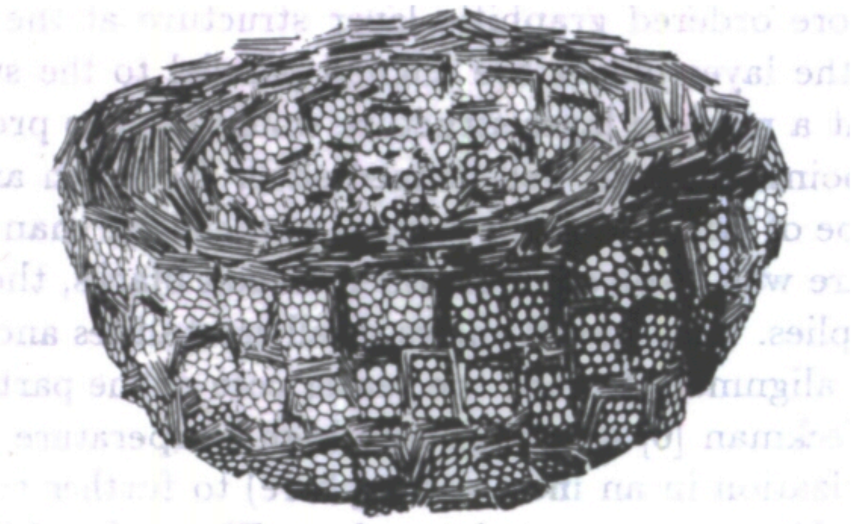


Figure 5. High-resolution TEM image of a soot aggregate in smoke from the Madikwe Game Reserve fire, South Africa, on 20 August 2000. The soot spheres show structures with onion-like curved, disordered graphitic (graphene) layers.

Li (JGR, 2003)

What is Black Carbon?

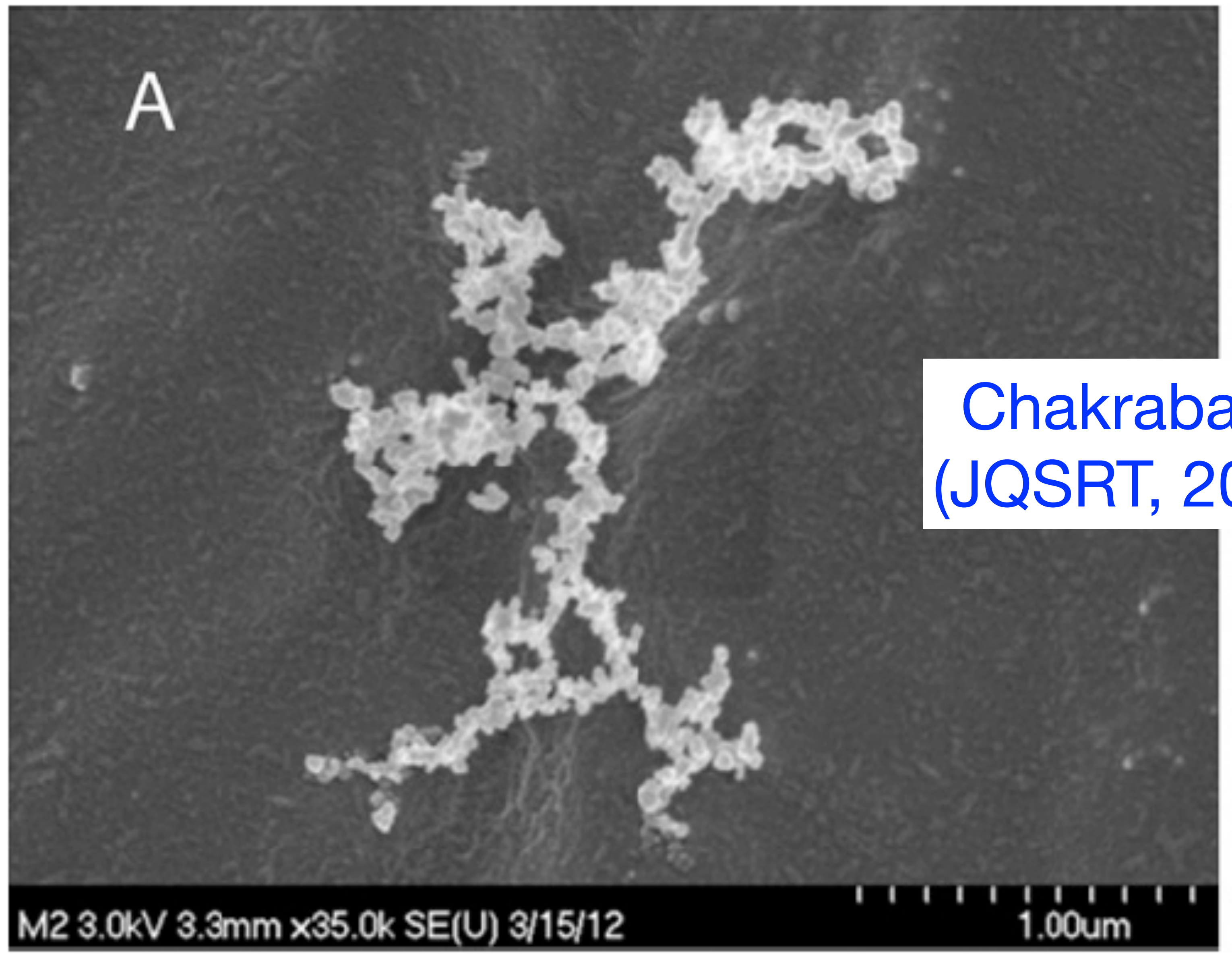


Hess and Herd, Carbon Black (1993)

- Byproduct of incomplete combustion
  - fossil fuel burning
  - biomass burning
- Graphitized
- Other names:

carbon blacks	produced in controlled conditions
soot	atmospheric; contains impurities
elemental carbon	measured by thermal analysis
black carbon	measured by optical absorption

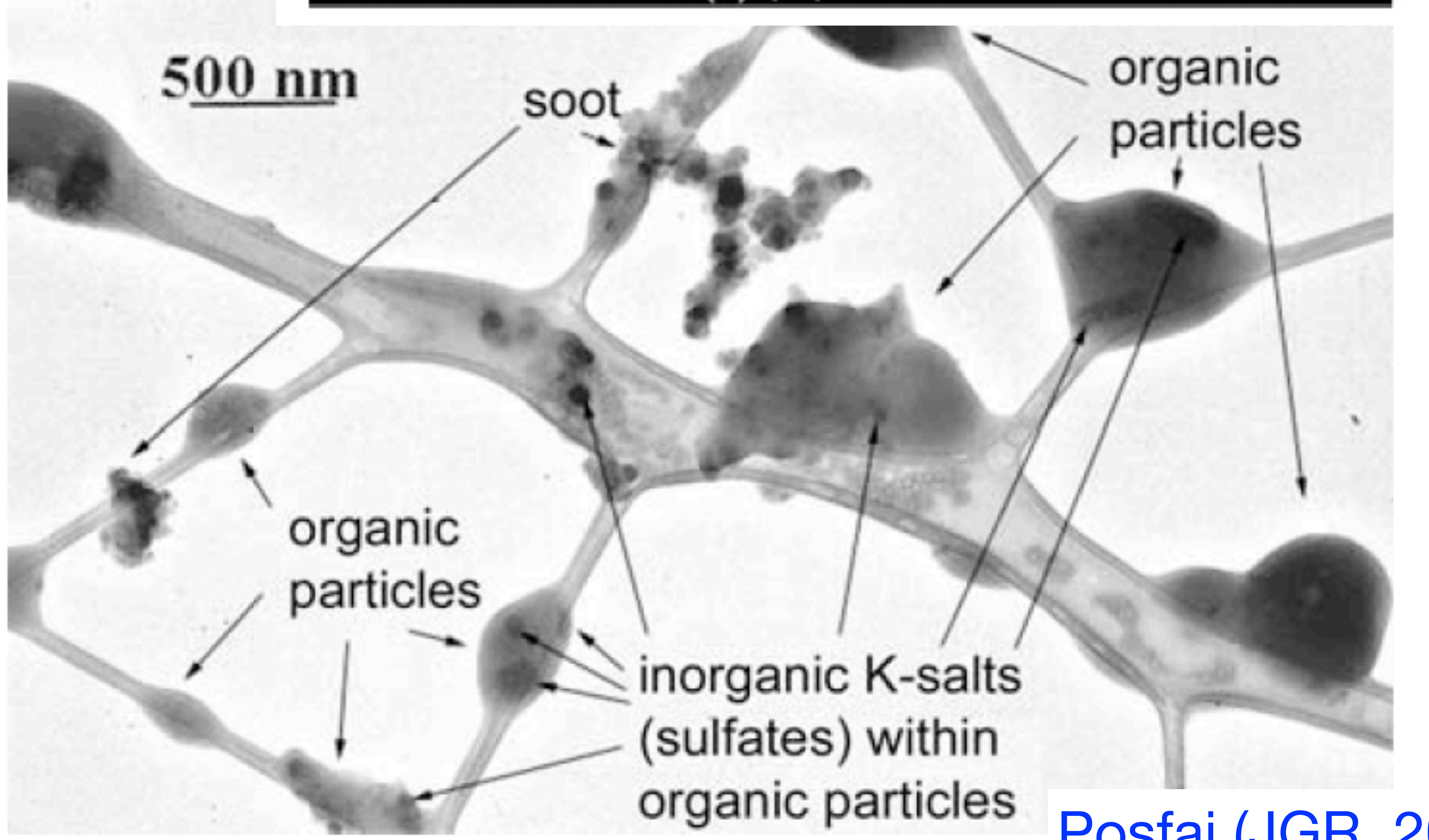
Schuster, Dubovik, Holben, First AERONET Workshop, Spain, 2004



Chakrabarty (JQSRT, 2013)

BC is highly graphitized, so the refractive index that we use has to reflect this.

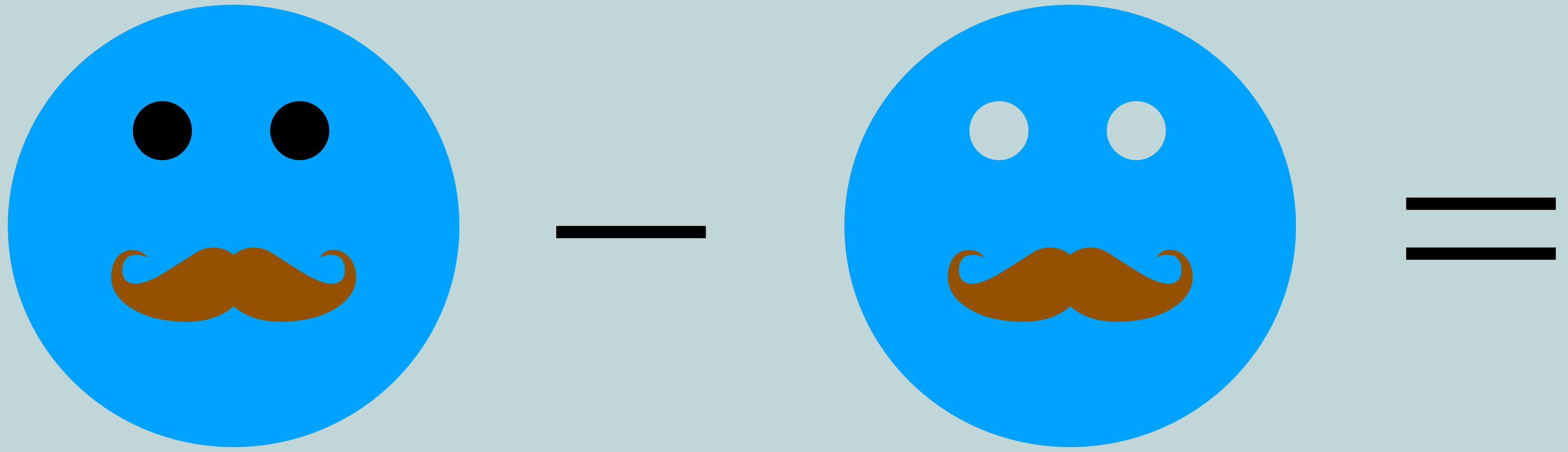
Source	Density (g/cm³)	*n - ki*	Comment
Borghesi and Guizzetti (1991)	2.26	2.67 - 1.34i	Graphite
Moteki et al (2010)	1.8	2.26 - 1.26i	Measured CRI at 1064 nm, but applied here at 550 nm
Janzen (1979)	1.7	2 - 1i	Carbon Blacks
Bond and Bergstrom (2006)	1.7	1.96 - 0.79i	Literature review.
Hess et al. (1998)	**1.0**	**1.75 - 0.44i**	OPAC; density is unrealistic
Chang and Charalampopoulos (1990)	1.7	1.63 - 0.48i	

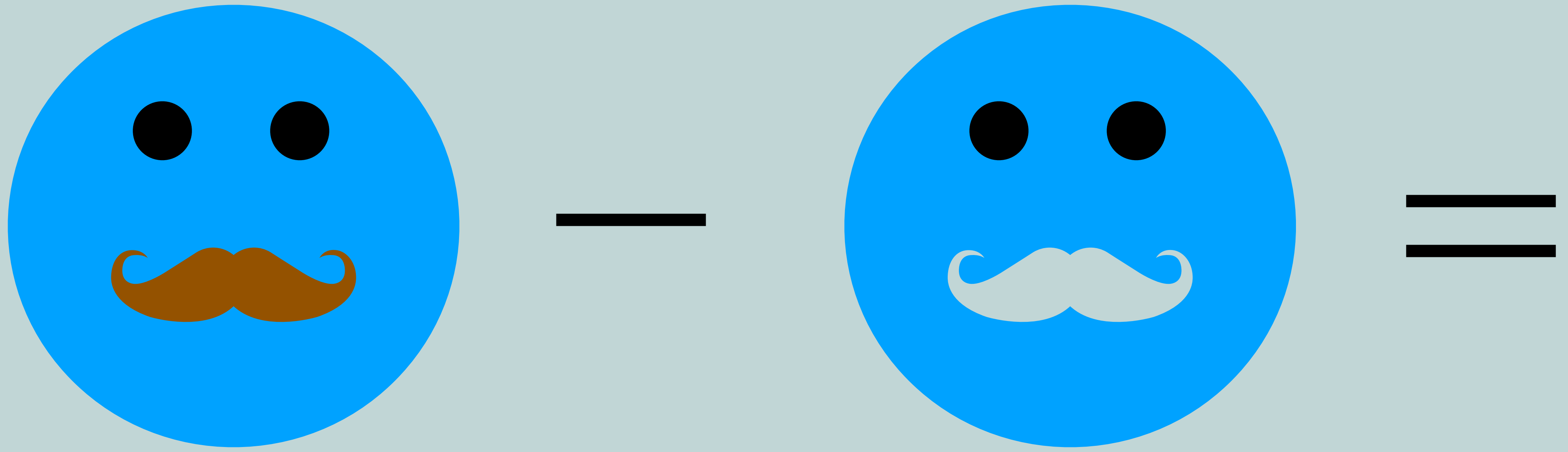


Posfai (JGR, 2003)

Figure 1. A typical portion of a sample of young smoke from a smoldering and flaming fire. Particles are attached to the lacey support film; most particles are carbonaceous (organic) with inorganic K-sulfate inclusions. As a result of diffraction contrast, the inclusions appear darker than the

Component AAODs for Internal Mixtures of BC, BrC, and a non-absorbing host

1.)  *BC AAOD*

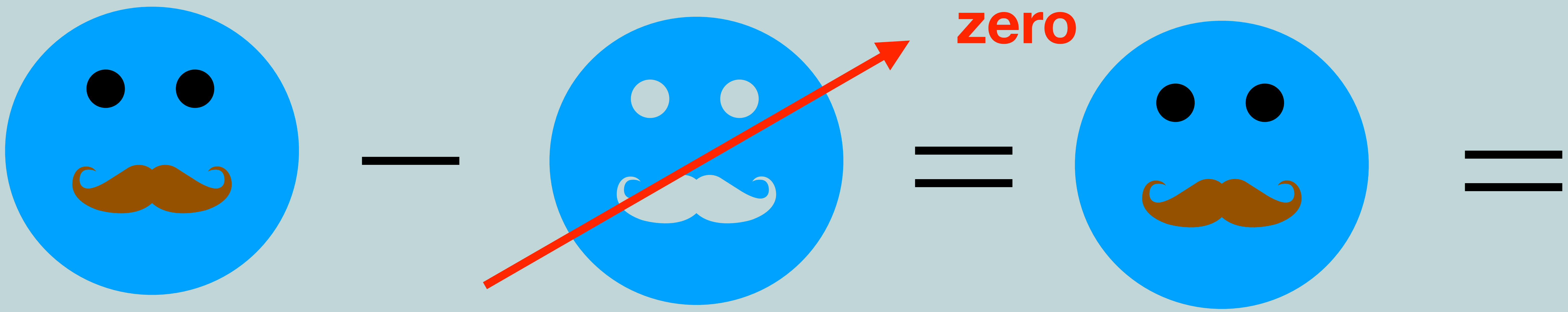
2.)  *BrC AAOD*

But we can not sum these components to get the total effect of the two absorbers (since both absorbers are enlarged by the host aerosol). That is,

(BC + BrC) AAOD < BC AAOD + BrC AAOD!!

This is OK. It just means that we have to use Model 3 to determine the effective AAOD of two absorbers (not the sum of Models 1 & 2).

The effective AAOD of multiple components is computed in the same way:

3.) 

= *(BC + BrC) AAOD*

Models, In situ, and Remote sensing of Aerosols (MIRA) Working Group

What is MIRA:

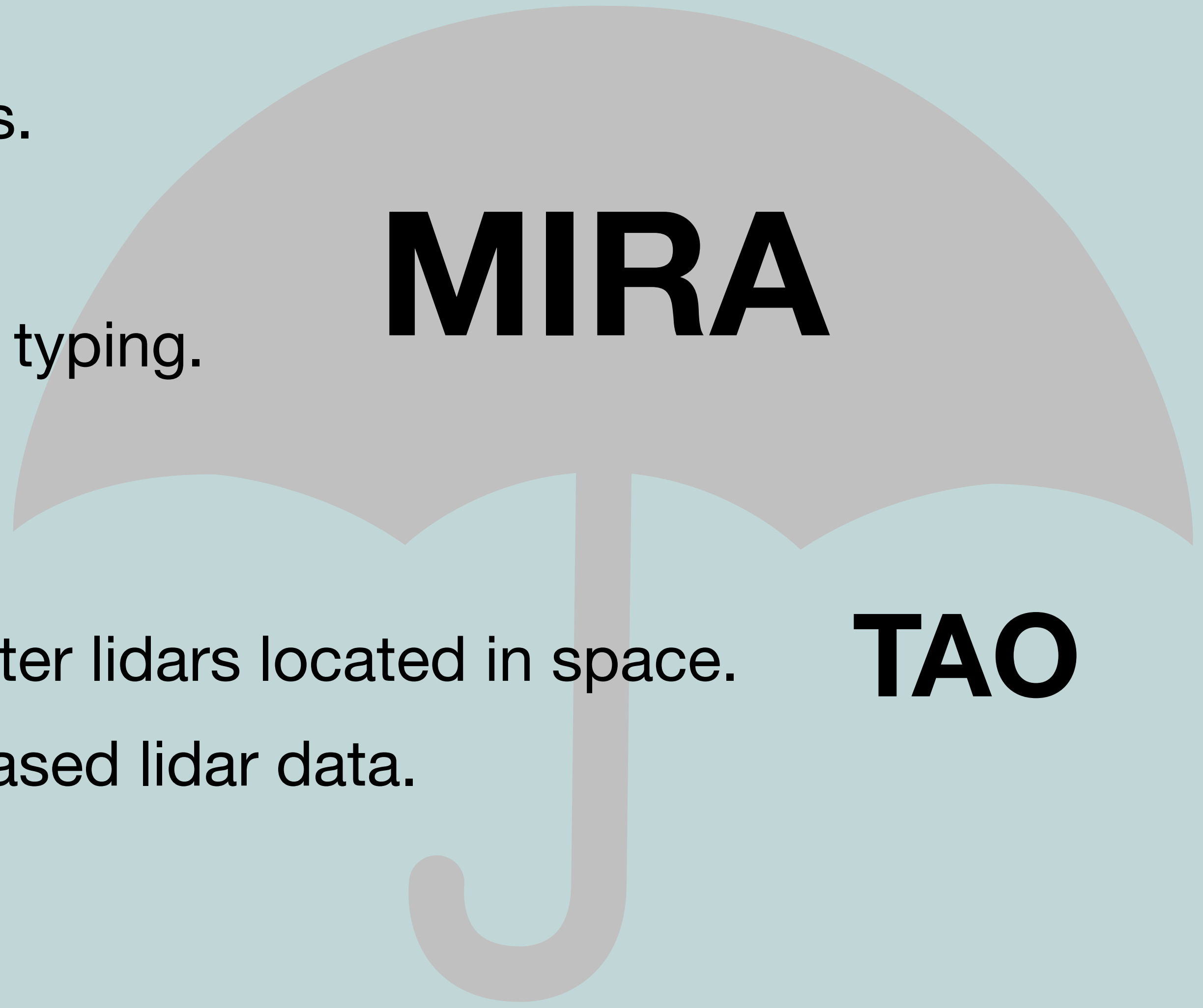
- The new MIRA working group establishes a forum for identifying collaborations and improving discussions amongst specialties and across regional boundaries.
- MIRA overlaps with Aerocom/AeroSat, but MIRA emphasizes lidar ratio and aerosol type; it is not a sub-group of AeroCom/AeroSat.
- MIRA is targeting IGAC and Gordon Research Conferences as venues; thus, leadership will change every few years in accordance with the protocol of those meetings.

Overall purpose:

- The purpose of MIRA is to foster international discussions between aerosol science specialties, and it follows an effort that was started with the CALIPSO Version 5 Aerosol Lidar Ratio Virtual Workshop that was held March 9-11, 2021.
- The first meeting under the MIRA banner was held as a virtual side meeting at IGAC on September 17, 2021.
- MIRA is aimed at bringing aerosol specialists together to expand our knowledge base while also accomplishing specific goals.

The near-term purpose of the MIRA working group is to:

- Characterize regional aerosol lidar ratios to support improvements of aerosol extinction profiles and understanding of aerosol typing.
- Create TAO -- a community cooperative of aerosol optical tables.
- Facilitate international communications between aerosol measurement and modeling groups.
- Encourage the use of regional knowledge to develop and improve remote sensing techniques for current and future backscatter lidars located in space.
- Enable and foster communication between the scientists who run global aerosol models and scientists who analyze space-based lidar data.



Near-term activities

- Bi-annual side meetings at IGAC.
- Bi-annual Gordon Conference meetings in the non-IGAC years
- Additional virtual meetings as appropriate.

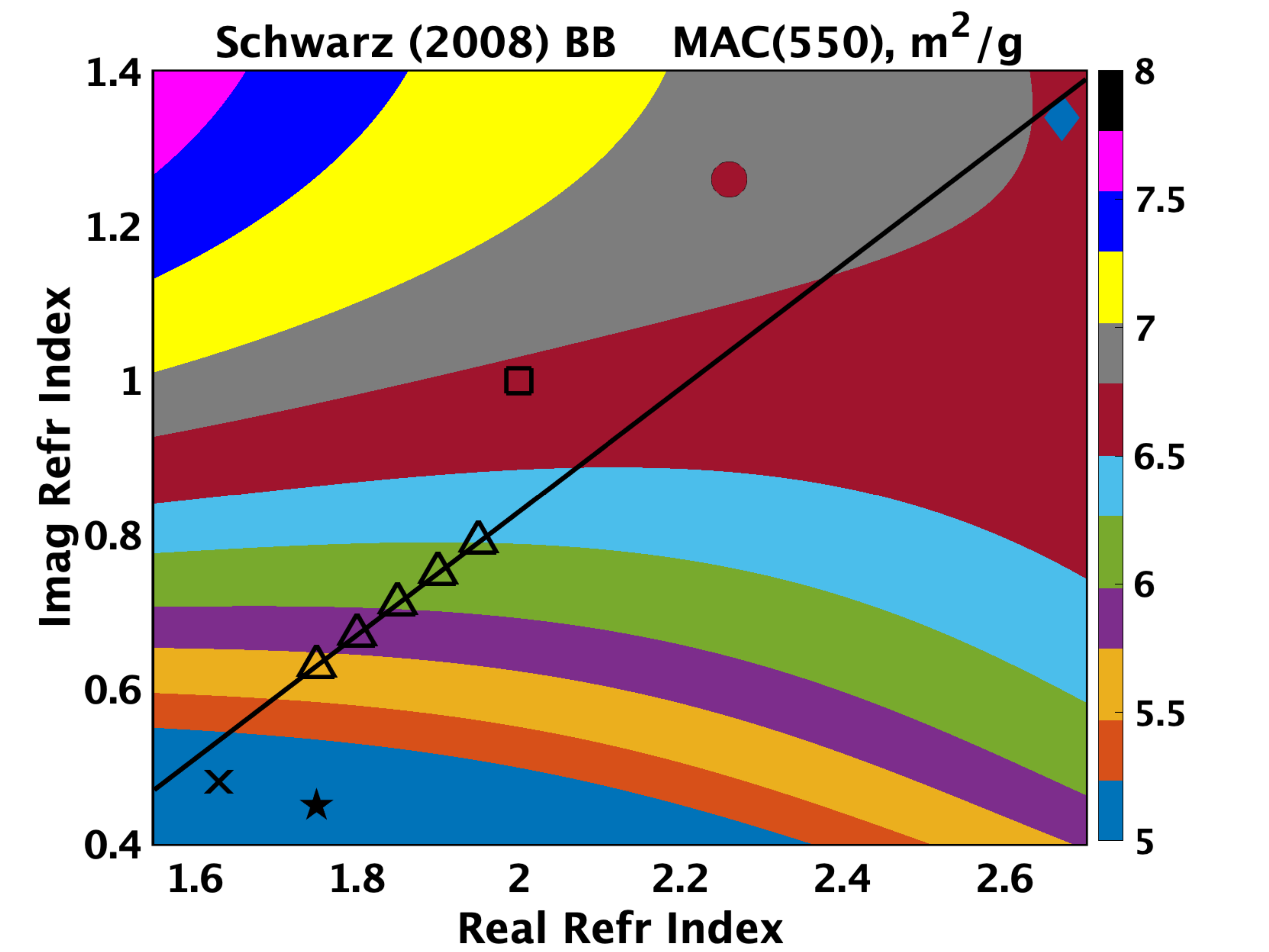
• The MIRA working group is open to all interested aerosol scientists, and those who are interested can register for the MIRA email list server at https://forms.gle/qdbCnngzNJc5YJi57.

• Registered participants will receive a MIRA newsletter and a short survey (soon).

QUESTIONS?

Recommended BB06 MAC (6.5 - 8.5 m2/g) can not be achieved with recommended BB06 CRI and Mie Theory.

Count Median Radius (CMR) = 0.072, Geometric Std. Dev = 1.430
BC density = 1.7 g/cm³ for contours



Bond and Bergstrom (2006):
Measured MAC ranges from 6.5-8.5 m2/g

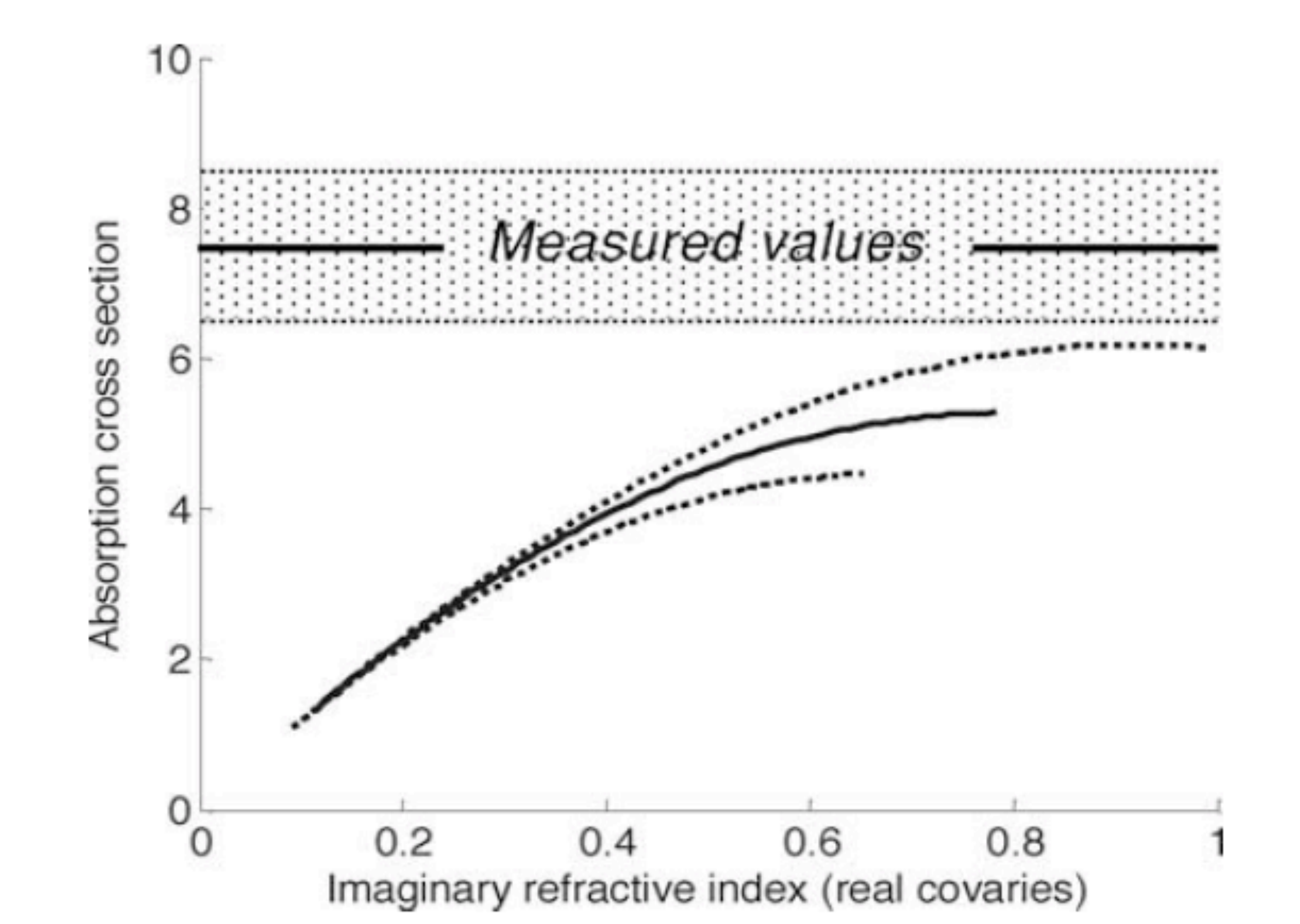


FIG. 9. Mass absorption cross sections m^2/g for fresh LAC: measured values from Figure 8(b) (shaded region, with central estimate marked by a thick line), and calculated values using the best guess of refractive index for fully-graphitized carbon; see Figure 7. Central curve shows "void-fraction" line in Figure 7 (approximately 1.8–0.74i); higher and lower curves are similar "void-fraction" lines assuming that pure LAC has refractive indices of 2 – 1i and 1.96 – 0.66i. The latter two values are not expected to represent atmospheric LAC, as discussed in text. Assumed density is 1.8 g /cm³.

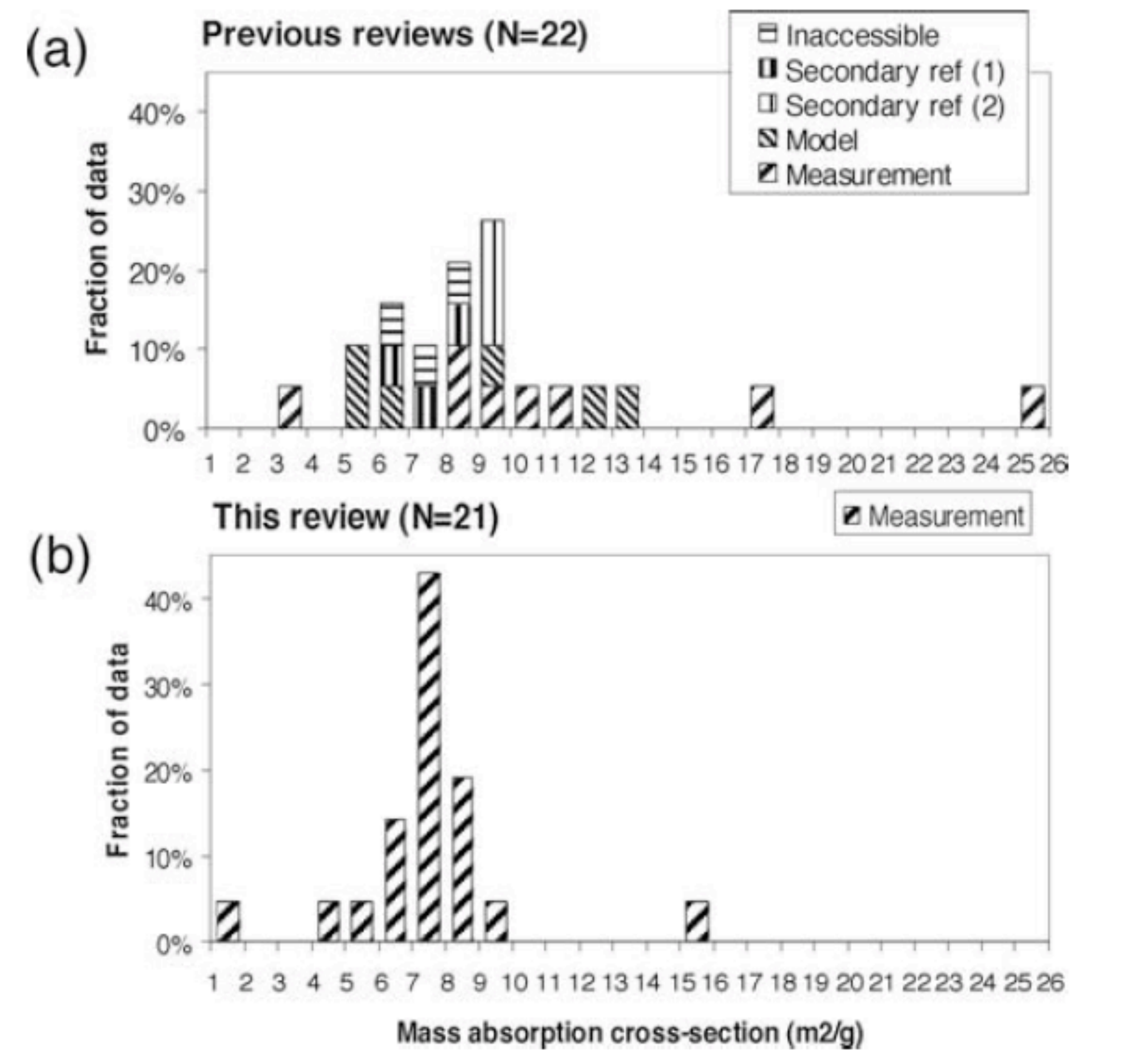


FIG. 8. Comparison of mass absorption cross sections tabulated (a) in previous reviews, showing large variability, and (b) in this review for near-source measurements, with much smaller variability. Legend in Figure 8a: (1) Traceable to value reported. (2) Not traceable to value reported.

Symbol	Source	Density (g/cm³)	*n - ki*	Comment
◇	Borghesi and Guizzetti (1991)	2.26	2.67 - 1.34i	Graphite
○	Moteki et al (2010)	1.8	2.26 - 1.26i	Technique sensitive to density. Measured CRI at 1064 nm, but applied here at 550 nm
□	Janzen (1979)	1.7	2 - 1i	Carbon Blacks
△	Bond and Bergstrom (2006)	1.7	1.96 - 0.79i	Literature review.
⋆	Hess et al. (1998)	1.0	1.75 - 0.44i	OPAC; density is unrealistic
x	Chang and Charalampopoulos (1990)	1.7	1.63 - 0.48i	

Reasons for Updating OPAC

1.) It was a great run, but retirement is overdue for OPAC BC

Bond and Bergstrom (Aerosol Sci. Technol., 2006) reported this in their assessment of BC refractive indices:

"The value commonly used by climate modelers (m = 1.74-0.44i at 550 nm) represents none of the possible refractive indices and should be retired."

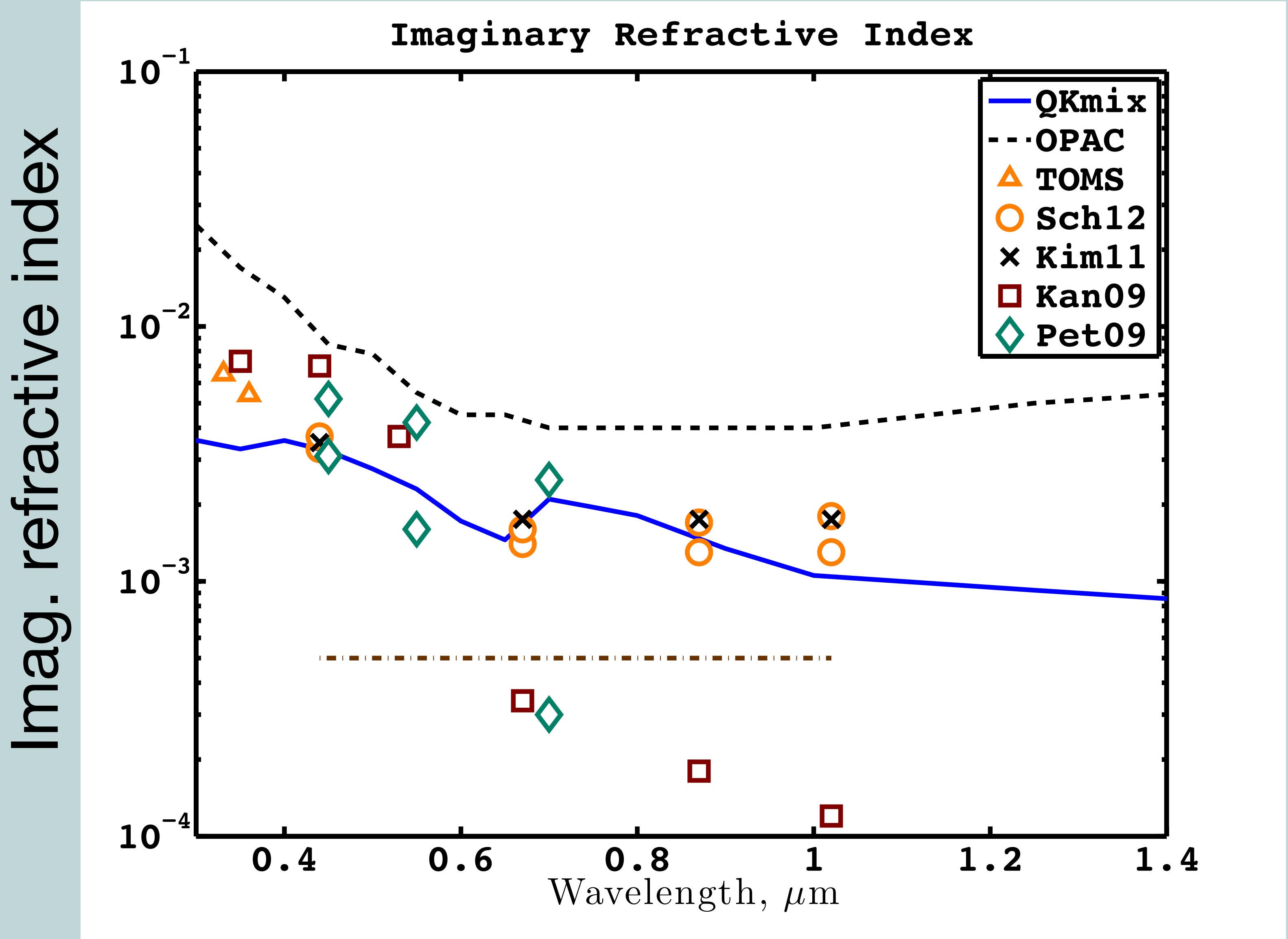
They also said:

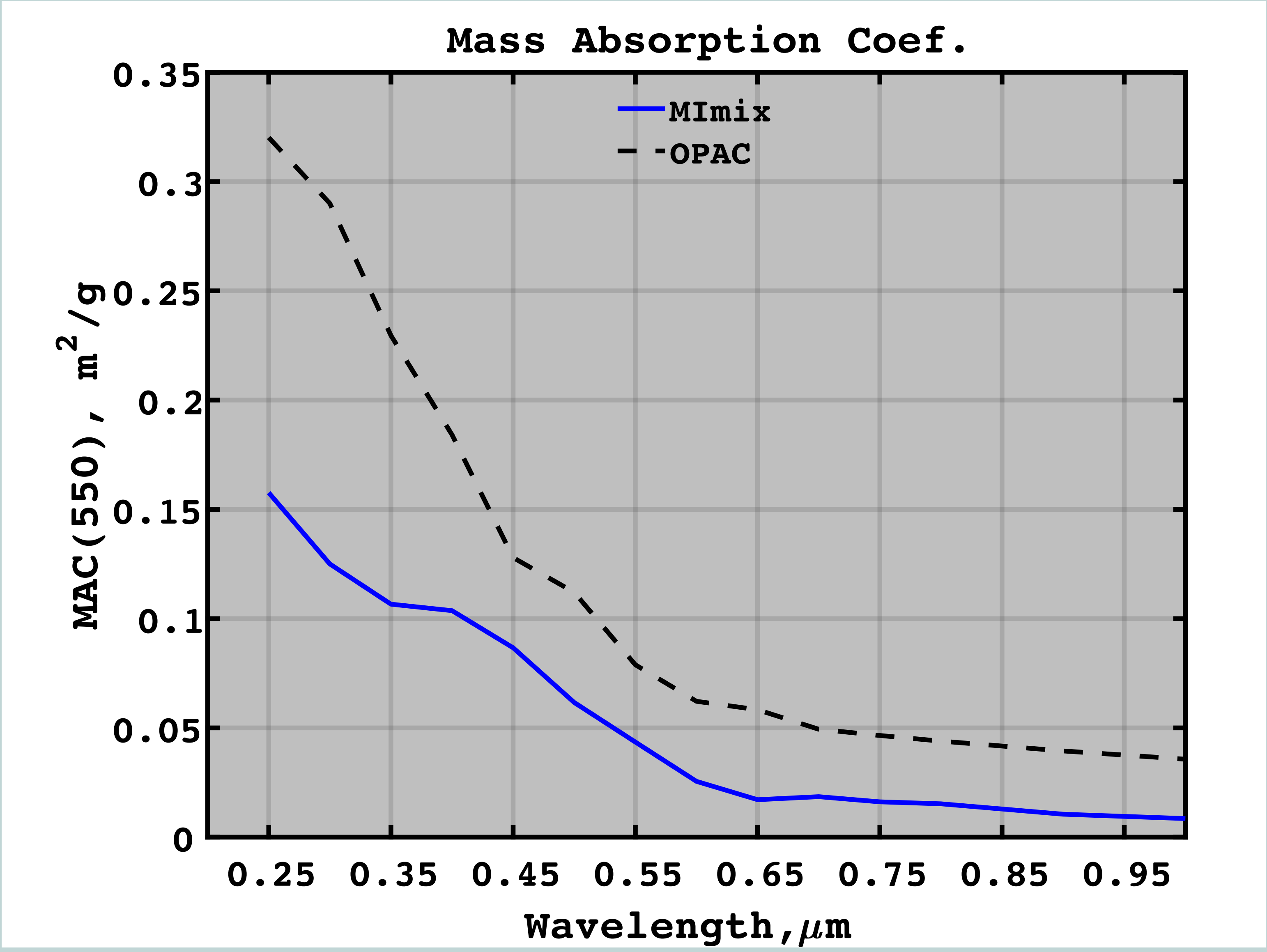
"The history of refractive index values tabulated by Shettle and Fenn (1976, 1979) is worth special mention. These are by far the most prevalent values for use in climate modeling, and have been incorporated into widely-cited literature, including a book by d'Almeida et al. (1991), and the Optical Properties of Aerosols and Clouds (OPAC) program (Hess et al. 1998). The original work by Shettle and Fenn (1976) averaged values from an earlier review by Twitty and Weinman (1971). In turn, the averaged data are taken from McCartney et al. (1965), who measured three coals, and Senftleben and Benedict (1918), who reported soot generated from an arc lamp. The review does not incorporate most of the findings on soot in the combustion literature, and indeed was written before most of that work was available. The precision of both n and k provided in OPAC values (three decimal places) is unwarranted, given this history. The OPAC value of 1.74-0.44i is drawn from incompletely graphitized carbon and has a lower value of k than most soot…

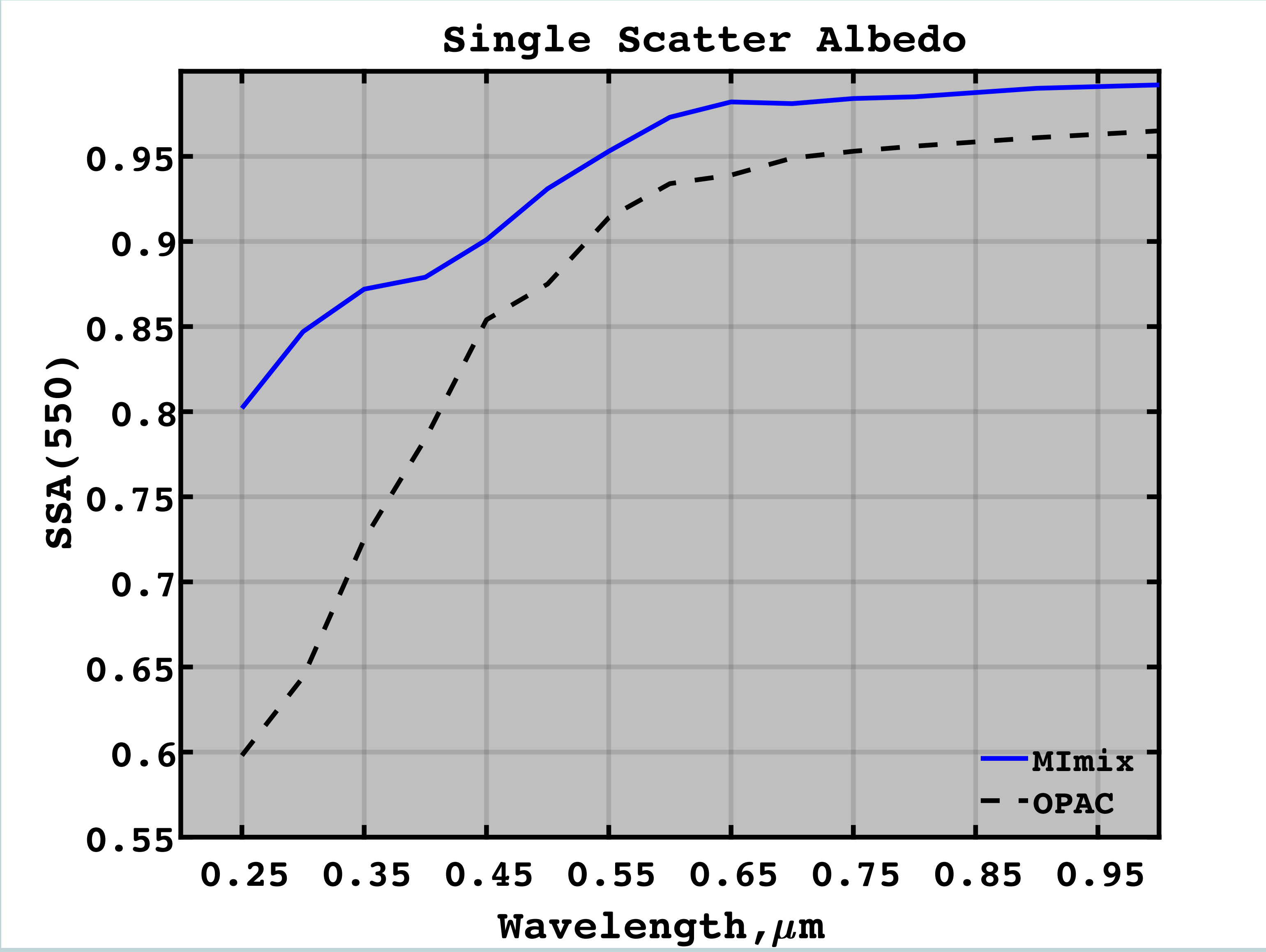
…The optical and physical data for LAC propagated by d'Almeida et al. (1991) have some interesting properties. Along with an imaginary refractive index that is too low, these authors recommend: (1) a particle size that is far too small (23 nm is the approximate size of primary spherules, not aggregates); (2) a geometric standard deviation that is somewhat too large (2.0); and (3) a density that is far too low (a density of 1.0 is never observed; Fuller et al. (1999) tabulate measurements indicating densities of about 1.8 g/cm3). Despite returning to the string of citations that led to d'Almeida et al. (1991), we have been unable to unearth the sources of these values. When compared with measured values, each of the individual assumptions above may lead to an error of 50-75% in calculated properties that affect climate forcing."

Reasons for Updating OPAC

2.) OPAC dust is too absorbing wrt recent measurements







OPAC / MImix at 0.55 um = 1.81

OPAC - MImix at 0.55 um = 0.04

MAC and SSA computed with Mie Theory (Wiscombe, Appl, Opt., 1980).

Dust lognormal parameters from SAMUM (Kandler, 2009): $R_n = 0.370,\ \sigma_g = 1.75$

K. Adachi and P. R Buseck: Internally mixed soot particles

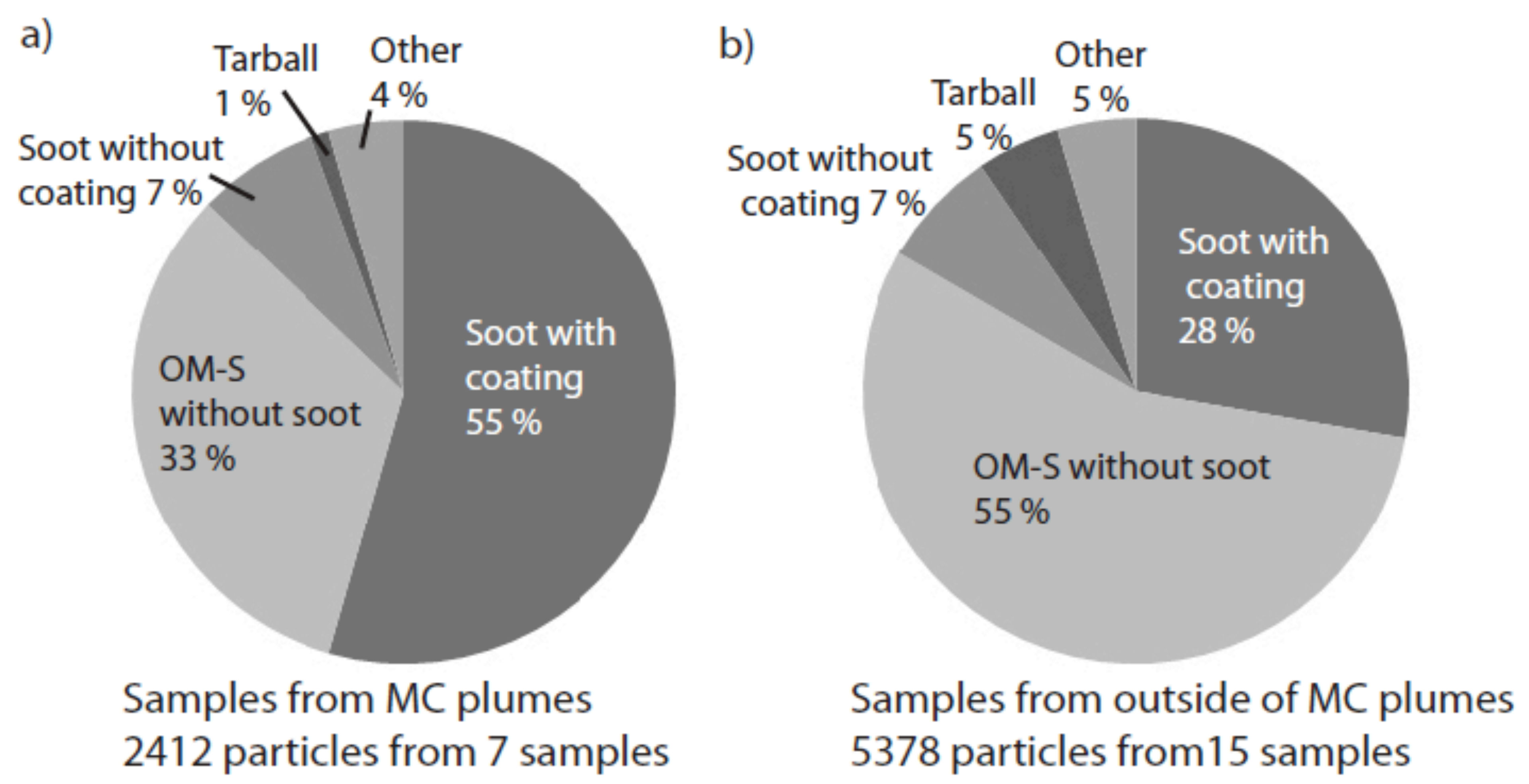
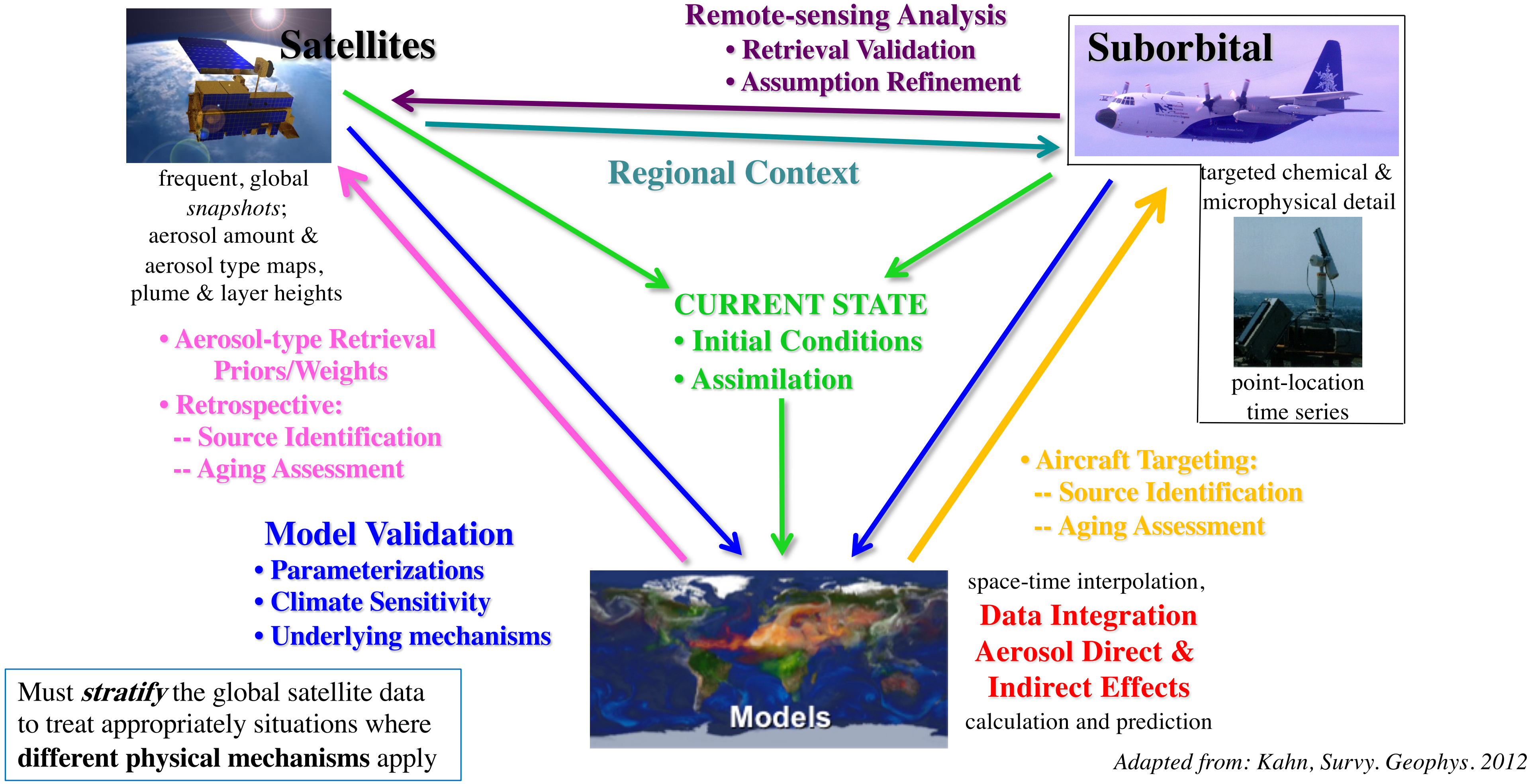


Fig. 6. Number fraction of particles.

Atmos. Chem. Phys., 8, 6469–6481, 2008

Closing the Aerosol Forcing Uncertainty Gap



Highlights the essential & unique role of each community in completing the aerosol forcing picture…

The SAM-CAAM Example
Systematic Aircraft Measurements to Characterize Aerosol Air Masses

The SAM-CAAM Science Definition Team

SAM-CAAM
A Concept for Acquiring Systematic Aircraft Measurements to Characterize Aerosol Air Masses

RALPH A. KAHN, TIM A. BERKOFF, CHARLES BROCK, GAO CHEN, RICHARD A. FERRARE, STEVEN GHAN, THOMAS F. HANSICO, DEAN A. HEGG, J. VANDERLEI MARTINS, CAMERON S. MCNAUGHTON, DANIEL M. MURPHY, JOHN A. OGREN, JOYCE E. PENNER, PETER PILEWSKIE, JOHN H. SEINFELD, AND DOUGLAS R. WORSNOP

SAM-CAAM aims to characterize particle properties statistically with systematic, aircraft in situ measurements of major aerosol air masses, to refine satellite data products and to improve climate and air quality modeling.

AMERICAN METEOROLOGICAL SOCIETY OCTOBER 2017 BAMS | 2215

SAM-CAAM *Concept*

[Systematic Aircraft Measurements to Characterize Aerosol Air Masses]

[This is currently a ***concept-development effort***, not yet a project]

Primary Goal:

• *Characterize statistically particle properties* for major aerosol types globally, to provide detail unobtainable from space, adding value to models & satellite aerosol data, offering *improved aerosol property assumptions for:*

-- *Modeling* aerosol direct forcing and aerosol-cloud interactions

-- *Satellite retrieval algorithm* climatology options or priors

Plus: More robust & consistent *translation between satellite-retrieved aerosol optical properties and* species-specific aerosol mass and size tracked in *aerosol transport, climate, & air quality models*

Helps enhance the value of 20+ years of satellite aerosol retrieval products

Kahn et al., BAMS 2017

Suborbital *In Situ* Required for PDFs of Particle Microphysical Properties

Aerosol intensive properties required for key aerosol science objectives, but ***cannot be retrieved adequately*** or are ***entirely unobtainable from remote sensing***

- ***Hygroscopicity**** – Ambient ***particle hydration***, ***aerosol-cloud interactions***
- ***Mass Extinction Efficiency*** – Translate between retrieved ***optical properties*** from remote sensing ***& aerosol mass*** book-kept in models
- ***Spectral Light-Absorption*** – Aerosol ***direct & semi-direct forcing***, atmospheric stability structure & circulation
- ***CCN Properties**** – At least part of the CCN size spectrum is ***too small to be retrieved*** by remote-sensing

Acquiring such data is feasible because:
Unlike aerosol amount, ***aerosol microphysical properties tend to be repeatable*** from year to year, for a given source in a given season

*Under special conditions, hygroscopicity (*Dawson et al. 2020*) and CCN # (*Rosenfeld et al. 2016*) can be derived from remote sensing; however: (*Stier*, ACP 2016)

SAM-CAAM ***Required Variables***

[Systematic Aircraft Measurements to Characterize Aerosol Air Masses]

1. AEROSOL PROPERTIES FROM *IN SITU* MEASUREMENTS & INTEGRATED ANALYSIS

	Abbrev.	Required Variable
1	EXT	Spectral Extinction
2	ABS	Spectral Absorption
3	GRO	Hygroscopic Growth
4	SIZ	Particle Size
5	CMP	Particle Type (a composition constraint)
6	PHA	Single-scattering Phase Function
7	MEE	Mass Extinction Efficiency
8	RRI	Real Refractive Index

Kahn et al., BAMS 2017

SAM-CAAM *Required Variables*

[Systematic Aircraft Measurements to Characterize Aerosol Air Masses]

2. METEOROLOGICAL CONTEXT

	Abbrev.	Required Variable
9	CO	Ambient Gases ($CO + O_3 + NO_2$)
10	T; P; RH	Standard Ambient Meteorological Variables
11	LOC	Geographic Location

3. AMBIENT REMOTE-SENSING CONTEXT

	Abbrev.	Required Variable
12	A-EXT & A-ABS	Ambient Spectral Extinction & Absorption
13	A-PHA	Ambient Particle Phase Function
14	A-CLD	Ambient Cloud & Large-Particle Size/Type
15	HTS	Aerosol Layer Heights

Open-path instruments

Kahn et al., BAMS 2017

Systematic Aerosol *Measurement Requirements*

• Aircraft *in situ* aerosol measurements have rarely been made with the aim of obtaining ******statistical sampling***** of specific aerosol types. For climate modeling and other applications, the distribution of values (e.g., the PDF) associated with key variables for major aerosol air mass types is needed. Few individual measurements are the best we currently have in most cases, whereas the PDF of a quantity is obtained only once subsequent measurements reproduce the distribution of values already acquired.

• Estimates of measurement ******uncertainty***** are difficult to obtain for many in situ instruments. However, uncertainty characterization can be considerably improved with techniques that are often unavailable for past field campaigns. For example, open-path nephelometers, developed in recent years, can now provide an uncertainty estimate for the particle hygroscopicity derived from in-aircraft measurements, and can also provide an uncertainty estimate (or even a correction) for directly measured inlet efficiency, which is especially important for super-micron particles.

• A ******suite of specific quantities*****: physical, chemical, and optical, is needed to adequately characterize an aerosol type for climate and air quality applications. All the required types of measurements have been made in the past, but rarely have they all been made together for the same aerosol air mass.

• The properties of many aerosol types change significantly as they age. As such, particle properties of major aerosol types need to be characterized systematically ******from the source downwind***** (or upwind toward the source) to adequately capture the aging process. Further, the ******source, the age, and the associated environmental conditions***** need to be recorded for the samples. With the help of aerosol transport modeling and/or synoptic-scale satellite observations this can be done, but again, this has not been done for much of the program of record.

-- The **program of record** has provided information used by both the satellite and modeling communities.

-- However, the ***diversity of assumed aerosol properties*** in remote-sensing retrieval algorithms, and the even greater diversity among model assumptions, give some indication of what is yet needed by way of systematic *in situ* aerosol measurements.

SAM-CAAM *Implementation*

[Systematic Aircraft Measurements to Characterize Aerosol Air Masses]

- ***Dedicated Operational Aircraft*** – routine flights, 2-3 x/week, on a continuing basis
- ***Sample Aerosol Air Masses*** accessible from a given base-of-operations, then move base
- Sample aerosol air masses ***Upwind & Downwind of Sources***, until ***subsequent measurements replicate the PDFs*** of the key variable measurements already made
- Sampling can be done even under ***non-precipitating Cloudy Conditions***, provided the aerosols are not concentrated within a cloud layer
- ***Process Data Routinely*** at central site; instrument PIs develop & deliver algorithms, upgrade as needed; data distributed via central web site, ***as with EOS satellite data***
- ***Project Science Team*** to set ***Deployment Schedule*** and ***fixed***, ***pre-determined Flight Plan Options***, accounting for model- and satellite-based ***major aerosol airmass locations*** & ***seasonality***, with cognizance of laboratory & suborbital measurement ***Program-of-Record*** (review studies*)
- Where possible, overfly ***Surface Sampling Stations,*** participate in ***Intensive Field Campaigns*** and under-fly ***Satellites***
- ***Models*** would (a) determine aircraft sampling ***Targets***, (b) assess ***Source***, ***Aging*** of actual measurements
- ***Payload Options***: A) Minimal; B) Fine-mode only; **C)** ***Fine + Coarse***; D) "Great to have"

**e.g., Burgos et al., ACP 2020; GASSP: Reddington et al., BAMS 2017; Di Biagio et al. ACP 2017*

Kahn et al., BAMS 2017

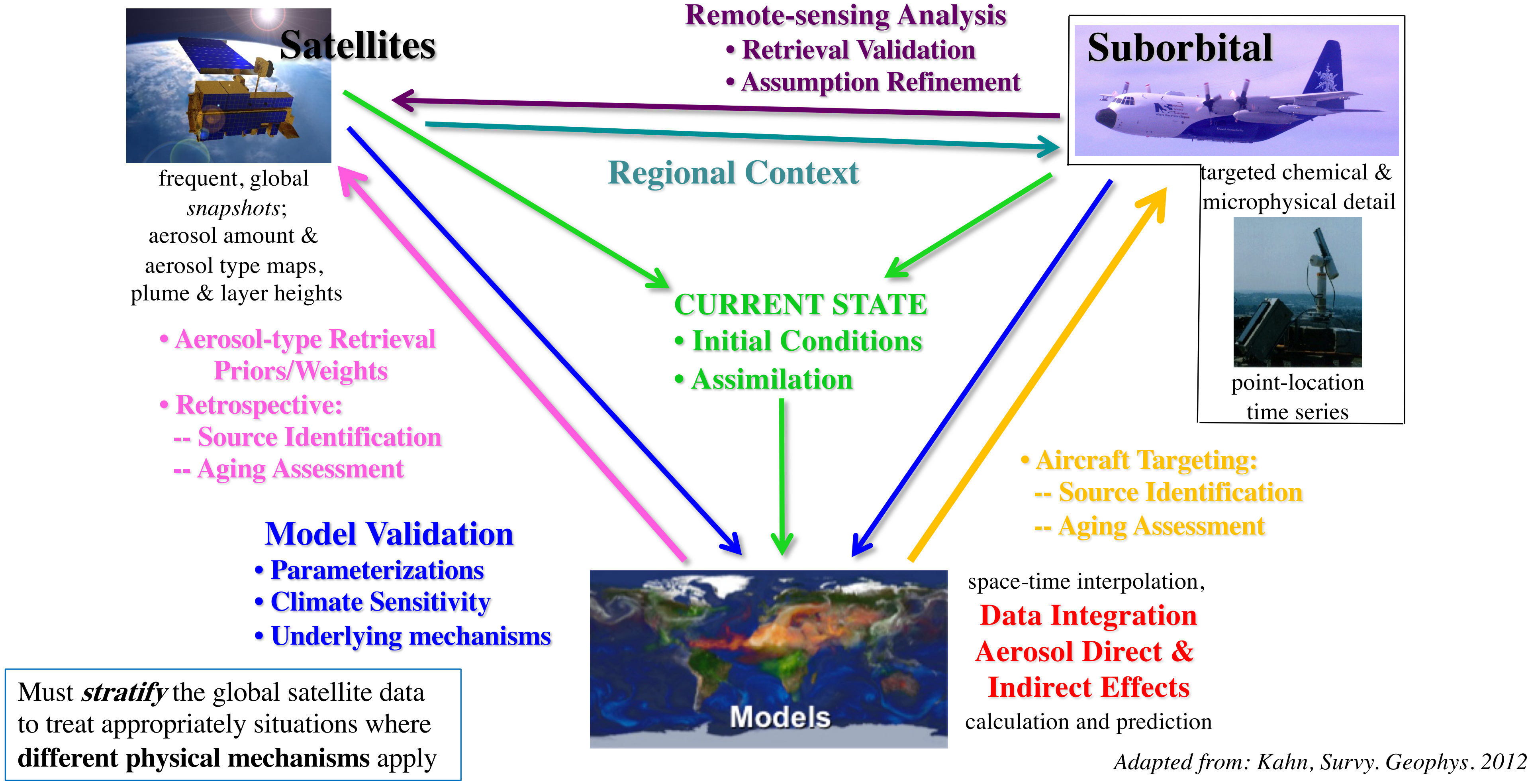
<u>SAM-CAAM</u> ***Operational Context*** & ***Heritage***

[Systematic Aircraft Measurements to Characterize Aerosol Air Masses]

- All the ***required measurements*** have been made at least *somewhere* in the past, with available technologies, but ***rarely all together***, ***and not with Systematic Sampling***
- ***Systematic Aircraft Aerosol Measurements*** have also been made in the past, but with different objectives (e.g., CARIBIC: *Nguyen et al., 2016*, doi:10.1080/02786820600767807; ARM: *Sheridan et al., 2012*, doi:10.5194/acp-12-11695-2012; ACTIVATE: *Sorooshian et al., 2019*, doi:10.1175/BAMS-D-18-0100.1)
- Parallels the relationship between ***AERONET and MODIS / MISR*** during the EOS era
- Notional payload *similar in some respects to* 2016-2018 ***ATom*** aircraft mission aerosol instrument suite
- Fills gaps in satellite remote-sensing, as ***IceBridge*** did for the cryosphere
- Peer-reviewed paper with notional payload ***demonstrating feasibility*** with a small aircraft (Sherpa); actual instrument selections would be based on agency buy-in and available resources, including newer technologies, as appropriate
- ***Addresses Decadal Survey & IPCC*** goals; ***can tap a range of international resources***

SAM-CAAM is feasible because:
Unlike aerosol amount, ***aerosol microphysical properties tend to be repeatable*** from year to year, for a given source in a given season

The Three-Way Street



Adapted from: Kahn, Survy. Geophys. 2012

Highlights the essential & unique role of each community in completing the aerosol forcing picture…

How to derive global/regional averages from sparsely sampled data

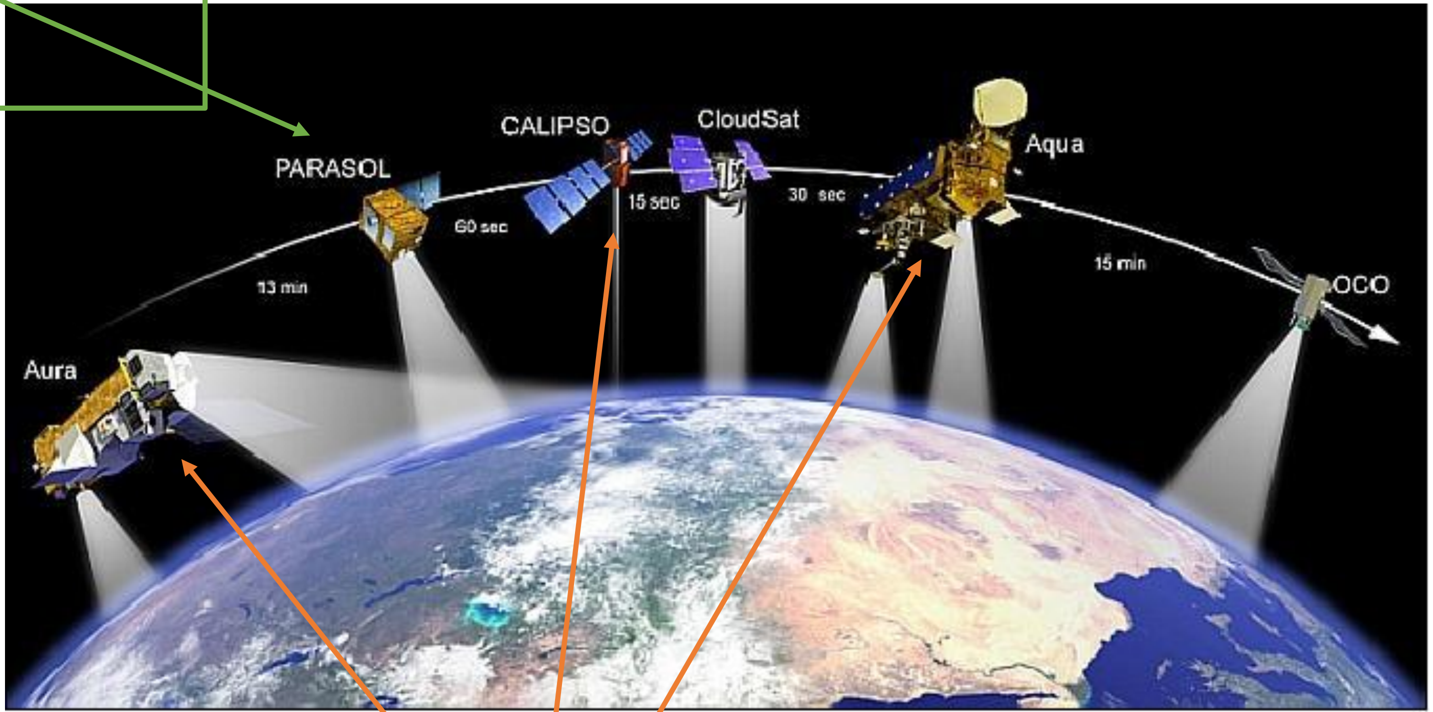
Nick Schutgens

Vrije Universiteit Amsterdam

Satellites

POLDER:
(POLarization and Directionality of the Earth's Reflectances)

GRASP & SRON:
Use multi-angle polarization measurements



OMI:
(Ozone Monitoring Instrument)
High spectral resolution spectrometer

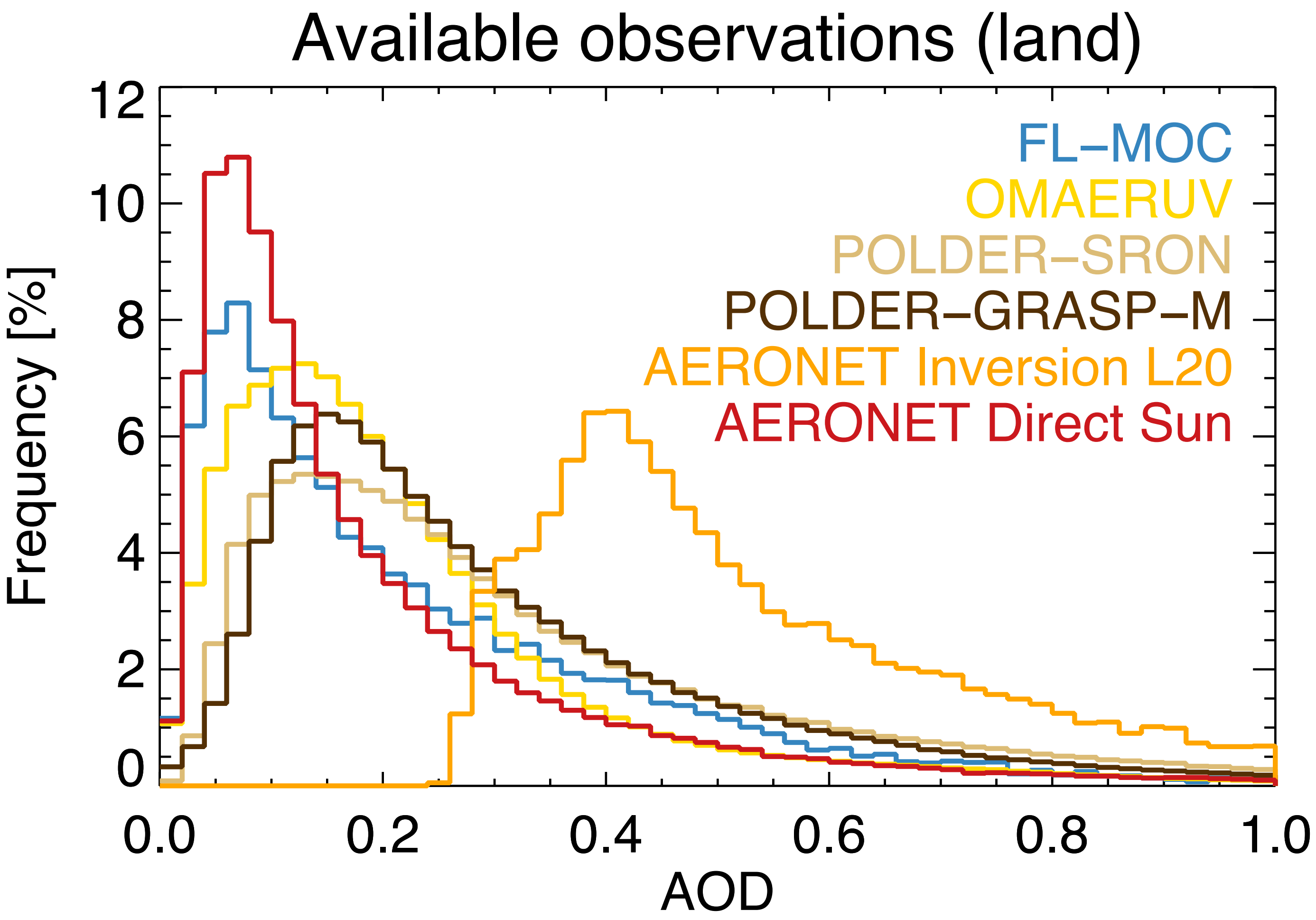
OMAERUV:
Uses UV wavelengths

MODIS-OMI-CALIOP:

FL-MOC:
Reinterprets existing measurements

Schutgens et al. *ACP* 2021
Schutgens et al. *ACP* 2020

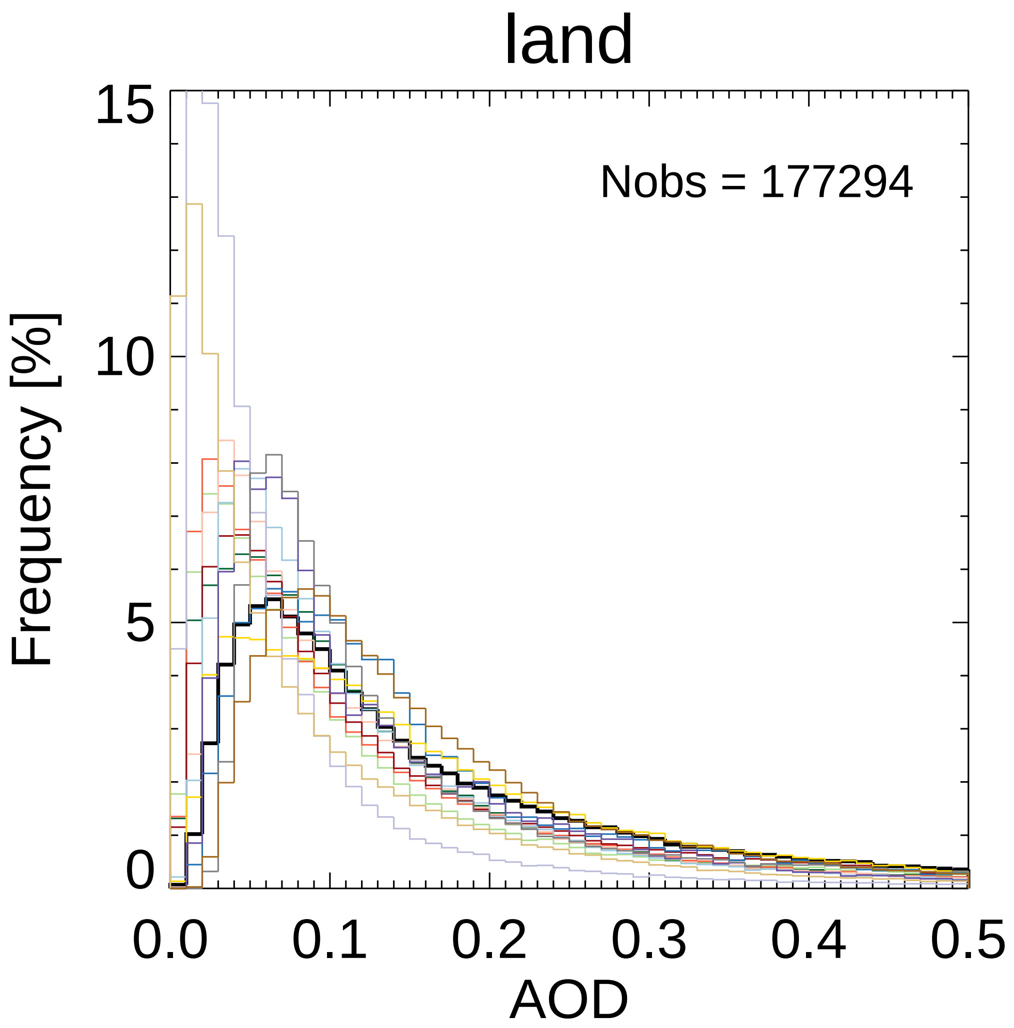
Sampling of satellite observations

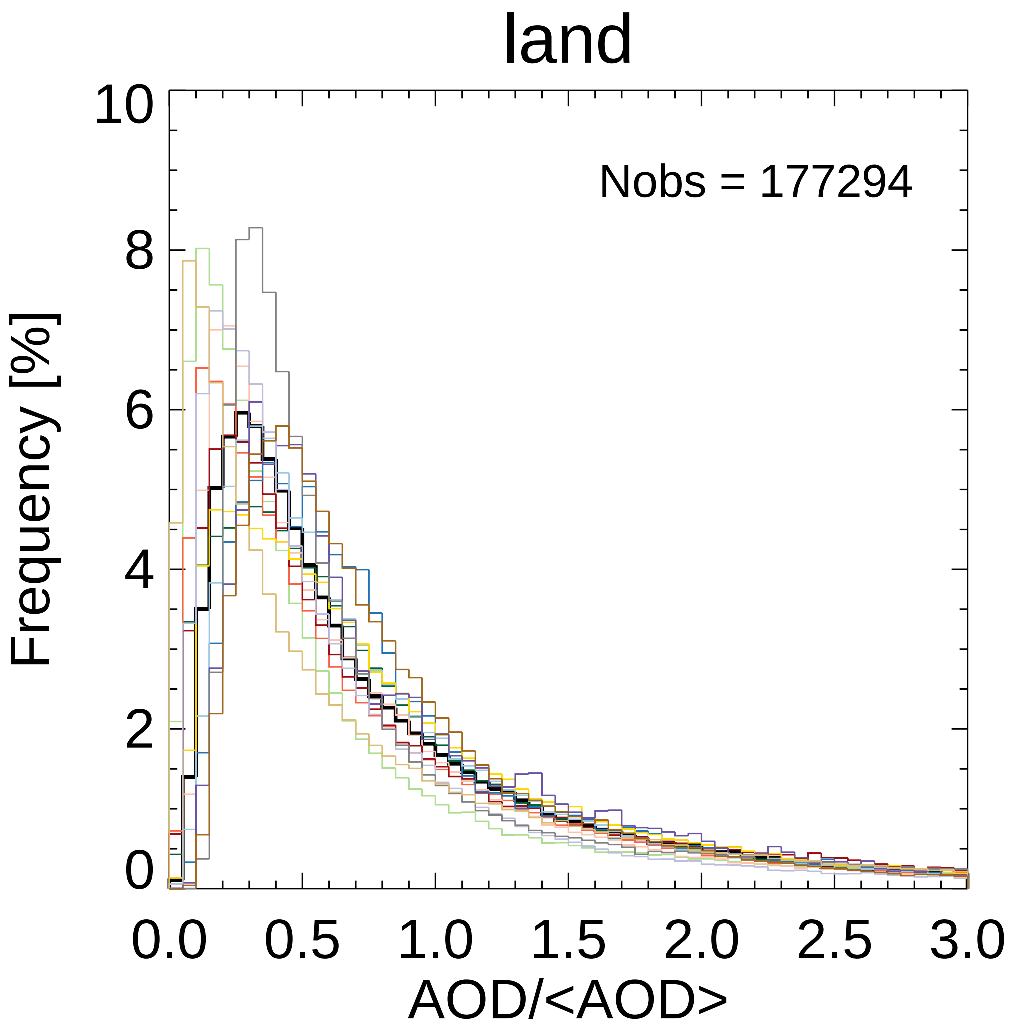


Note: these data are *uncollocated*

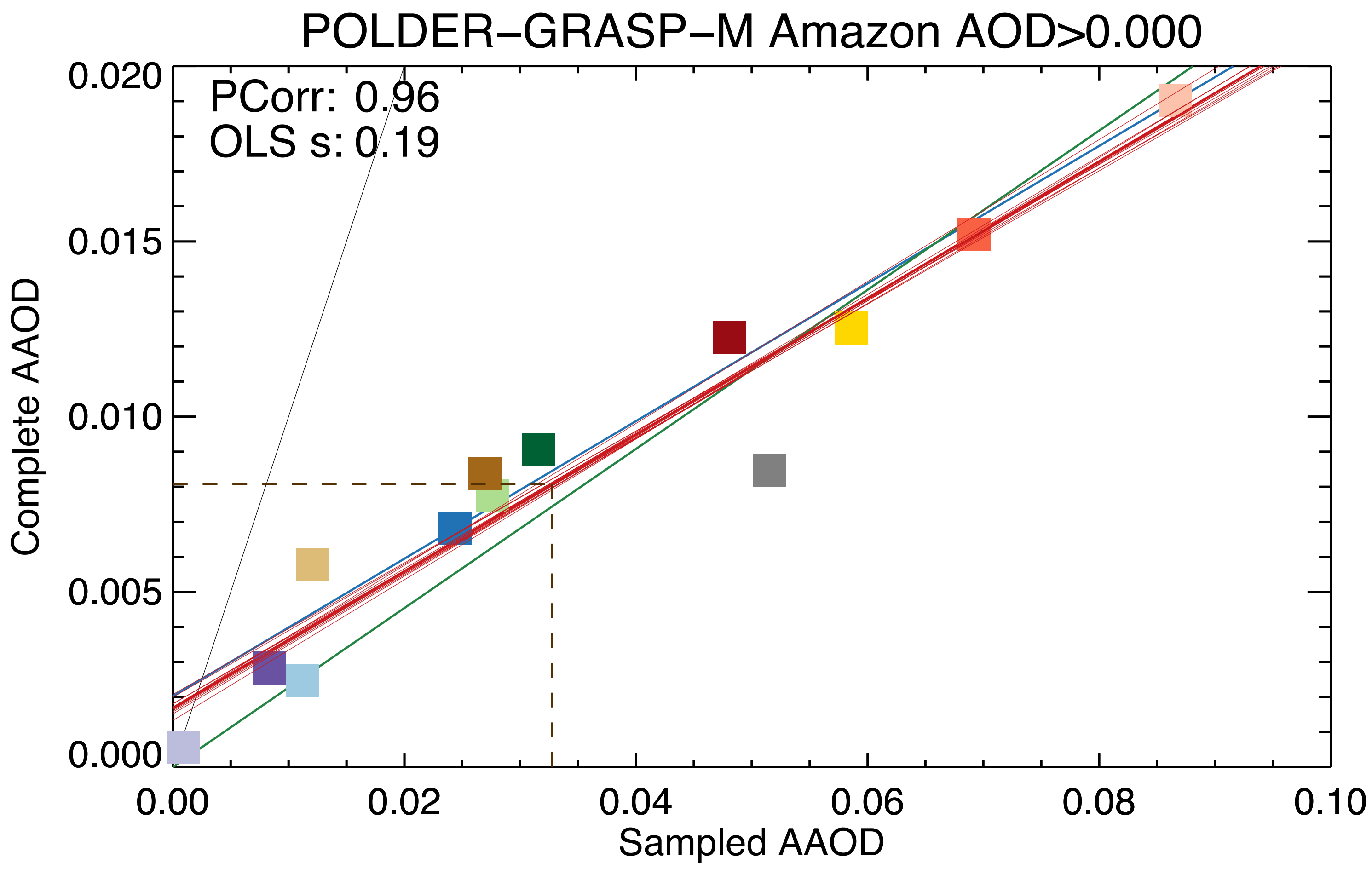
Note: these data are *collocated*

Model vs AERONET AOD distributions



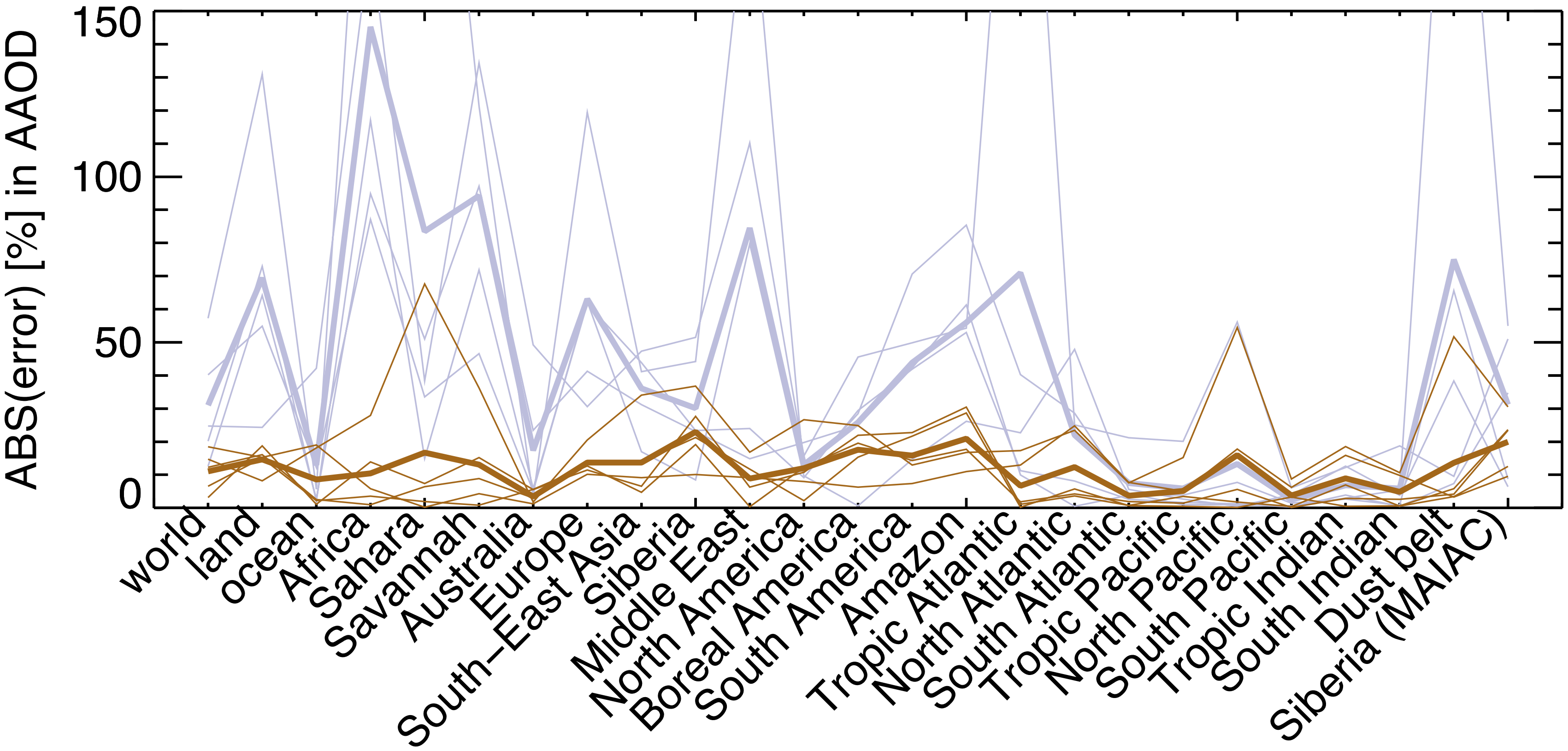


Using model data to homogenize observations



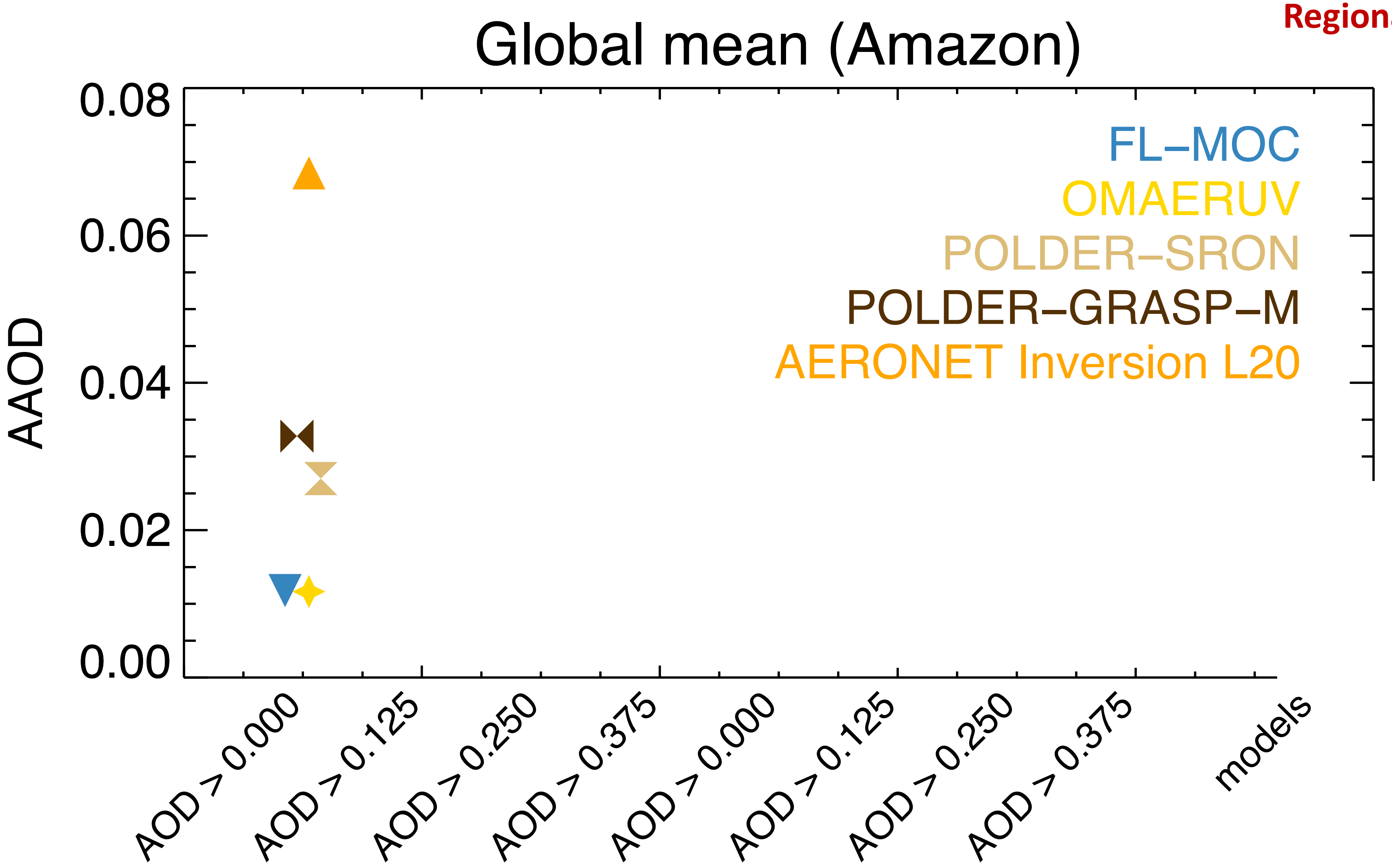
Based on 3 years of data over the Amazon

Predicted homogenization errors according to 2 models

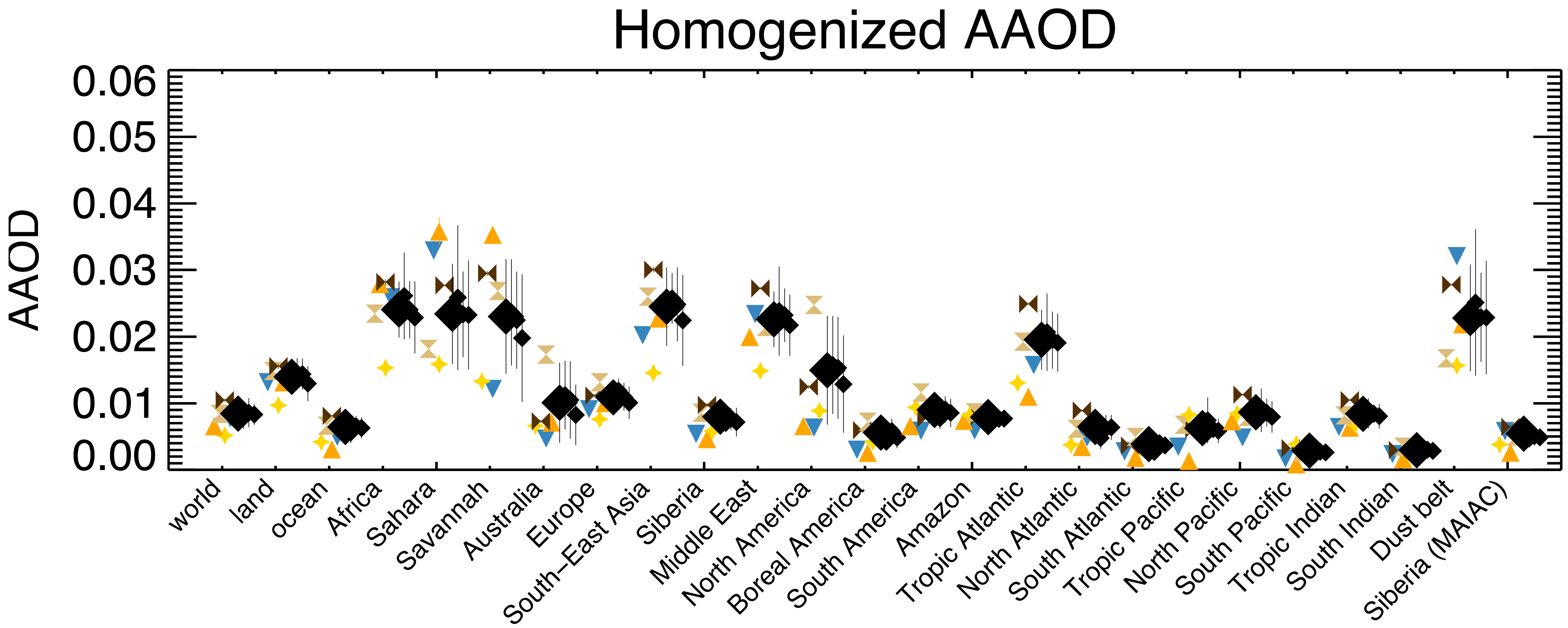


Using real observations:

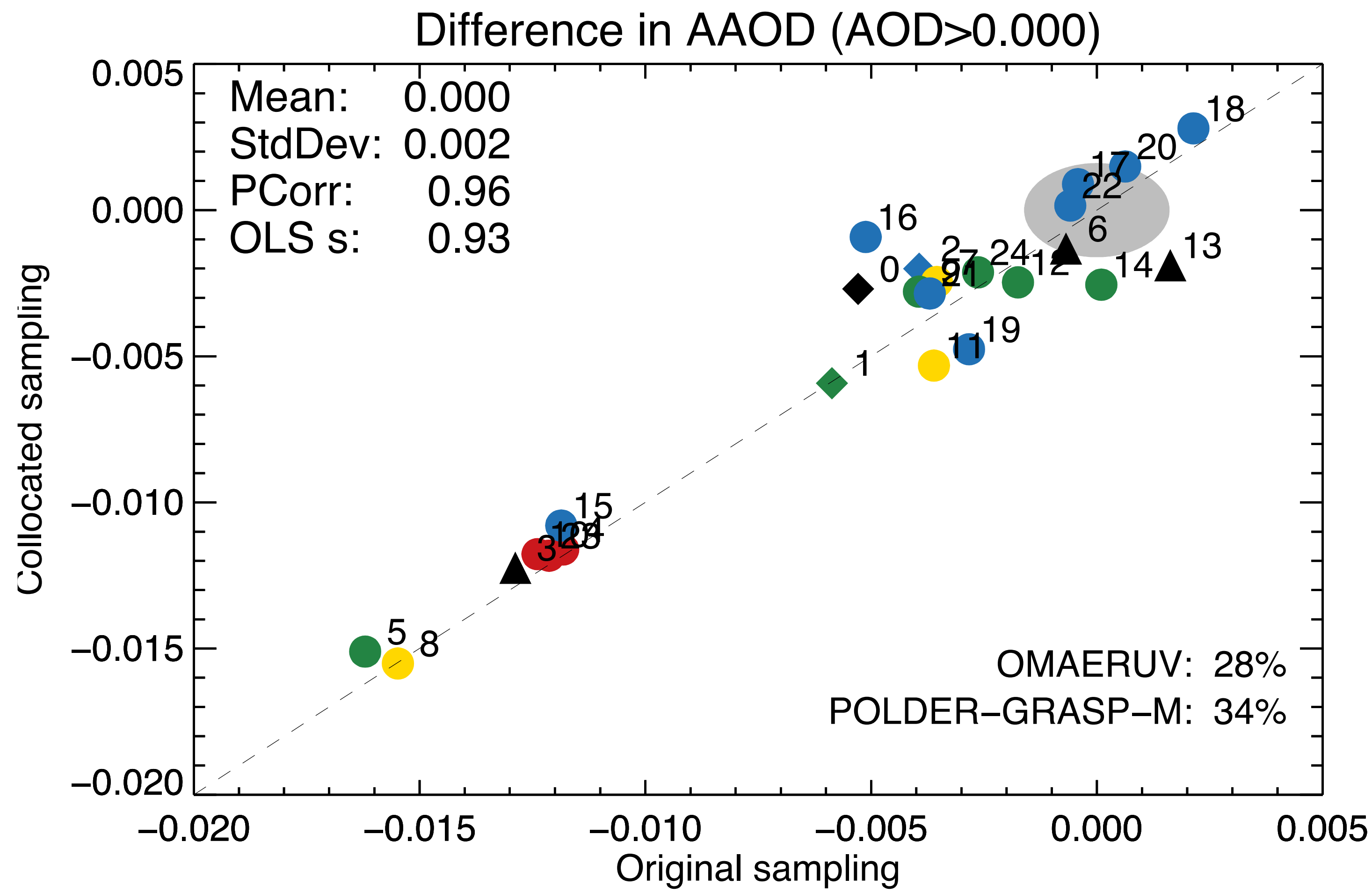
Regional, 3 year estimates



For different regions



Impact of retrieval biases



Difference due to retrieval errors only

Difference due to homogenization and retrieval errors

Global AOD & AAOD & SSA

Region	AOD
world	0.153 ± 0.008

AAOD	Region
0.0085 ± 0.0017	world

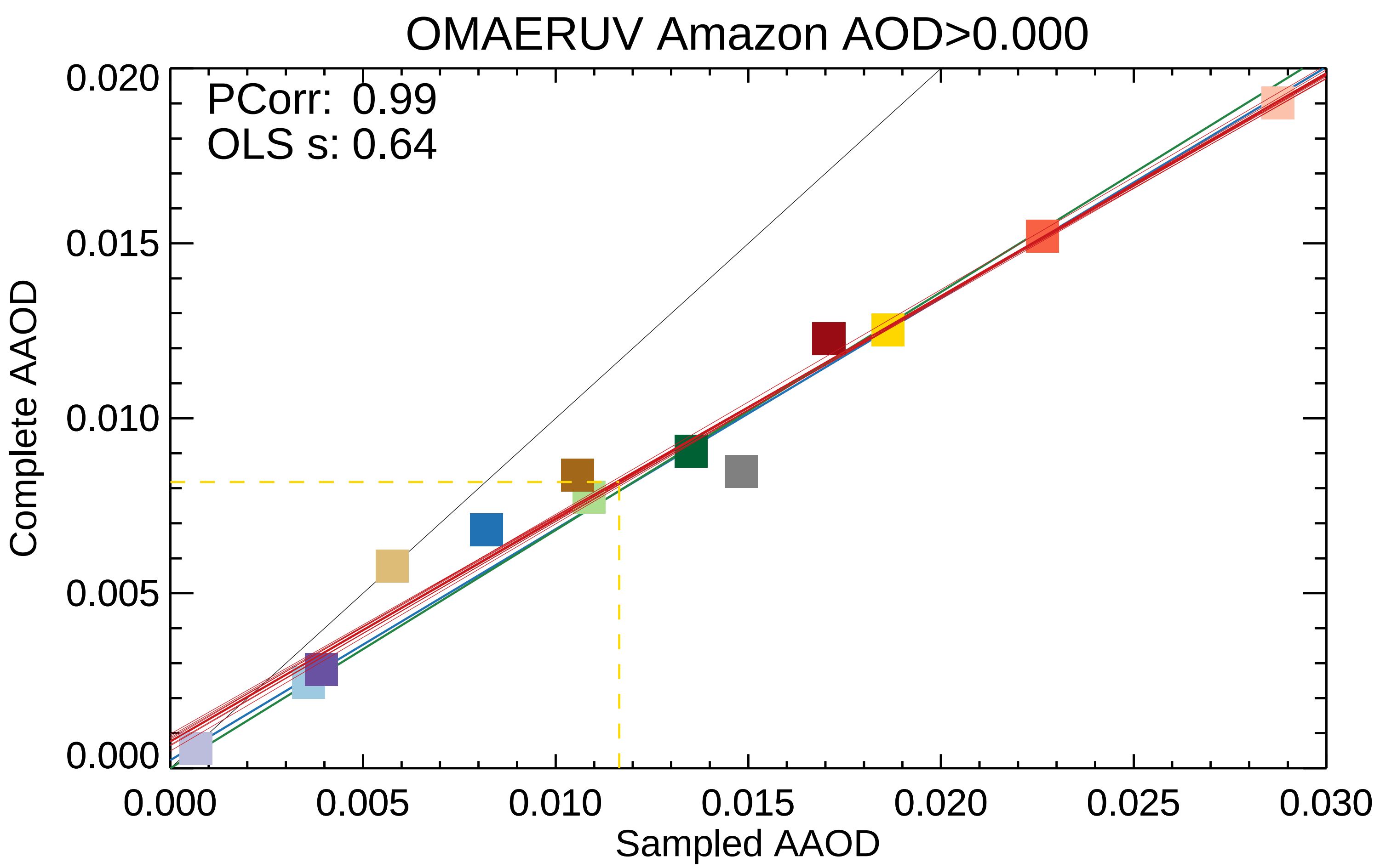
Region	SSA
world	0.944 ± 0.011

Summary

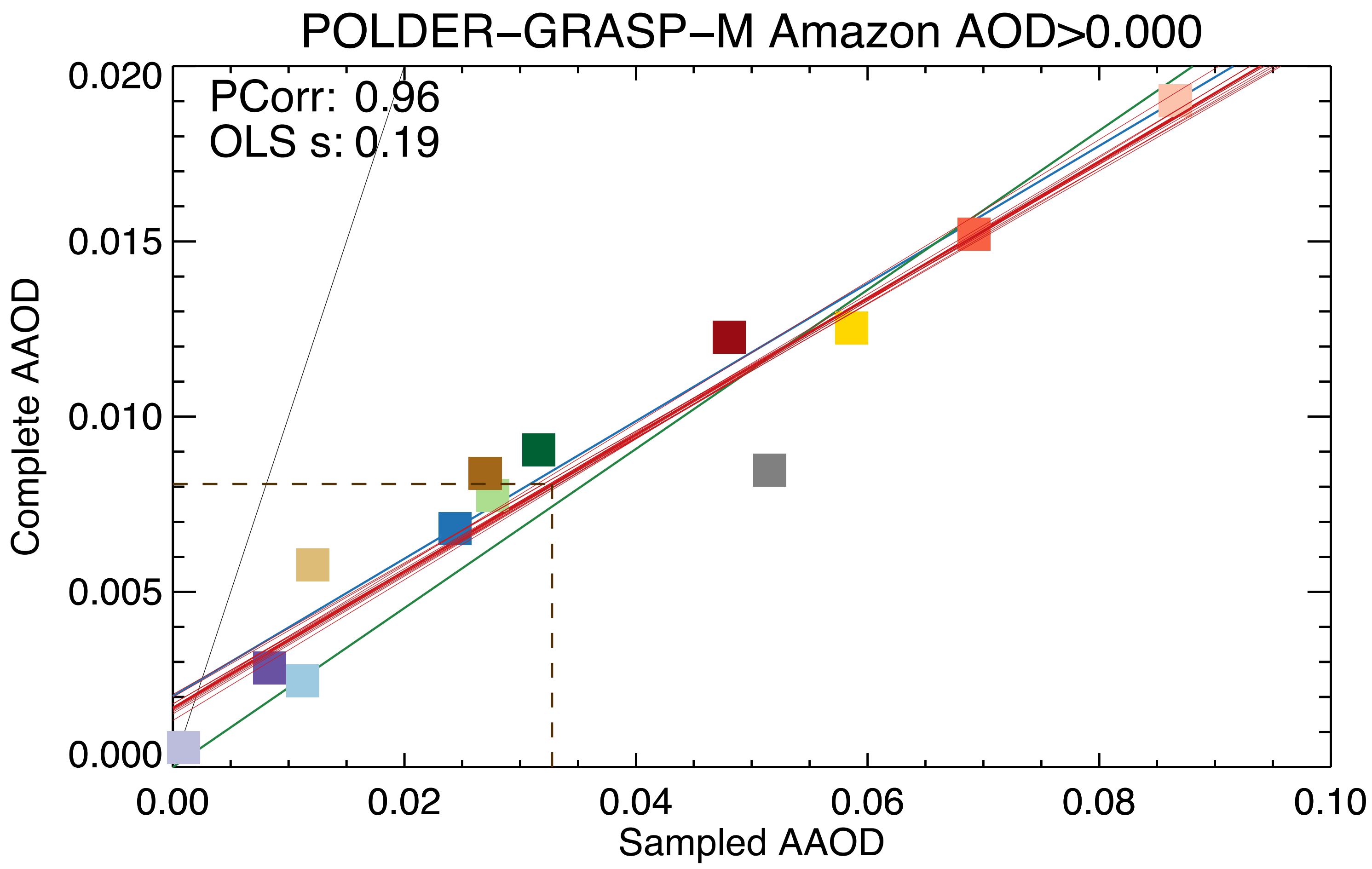
- It seems possible to “homogenize” sparsely sampled observations
- Model studies suggest uncertainties of less than 20%, often even 10%
- Real observations suggest that retrieval biases are leading cause of remaining differences between products
- Homogenization is technically simple and allows conceptually easy comparison of models to observations

- Note: at present, this works at regional and seasonal scales
  THIS STILL ALLOWS PROCESS STUDIES, SEE Q. ZHONG TALK TODAY

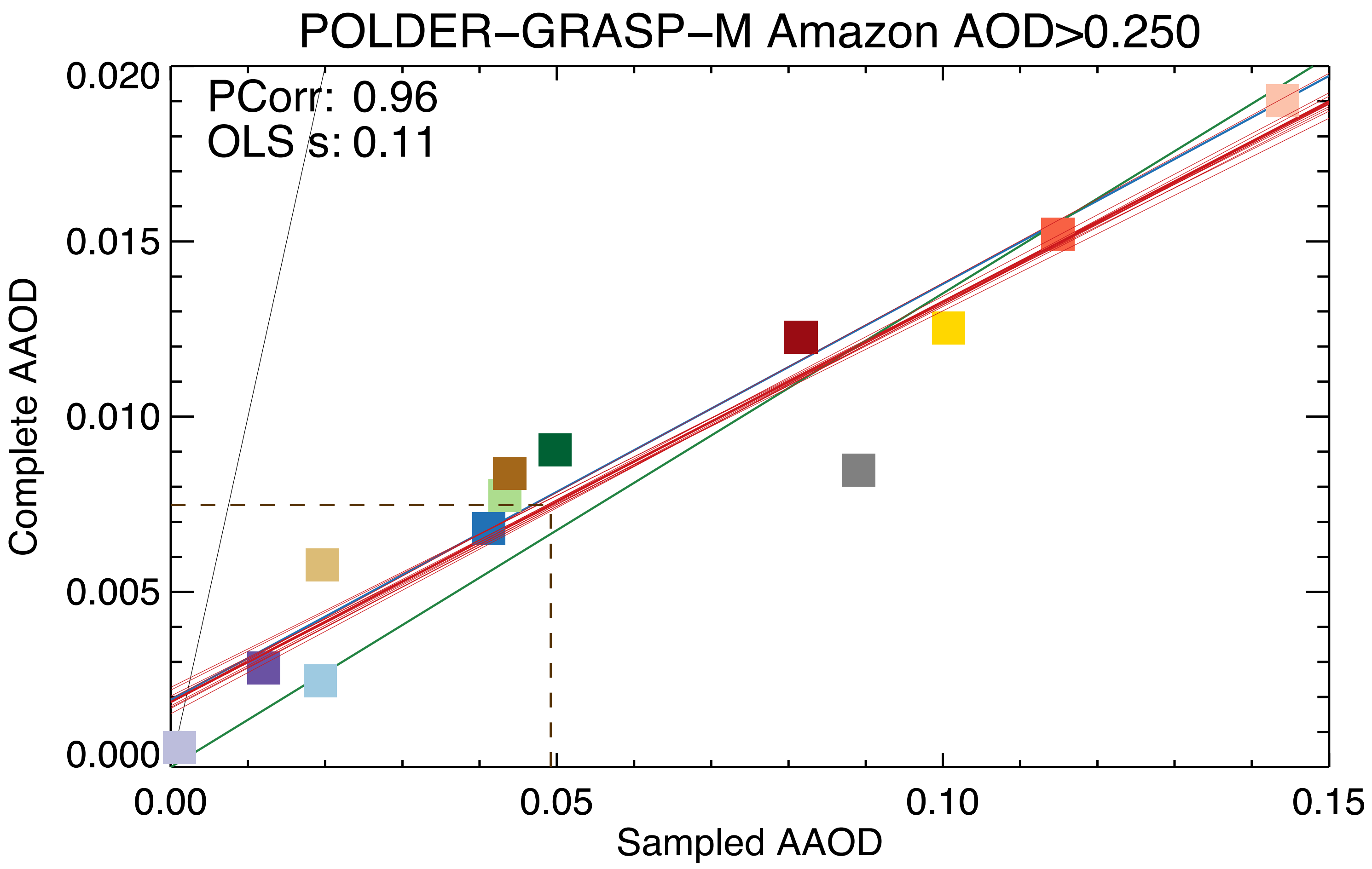
Using model data to homogenize observations



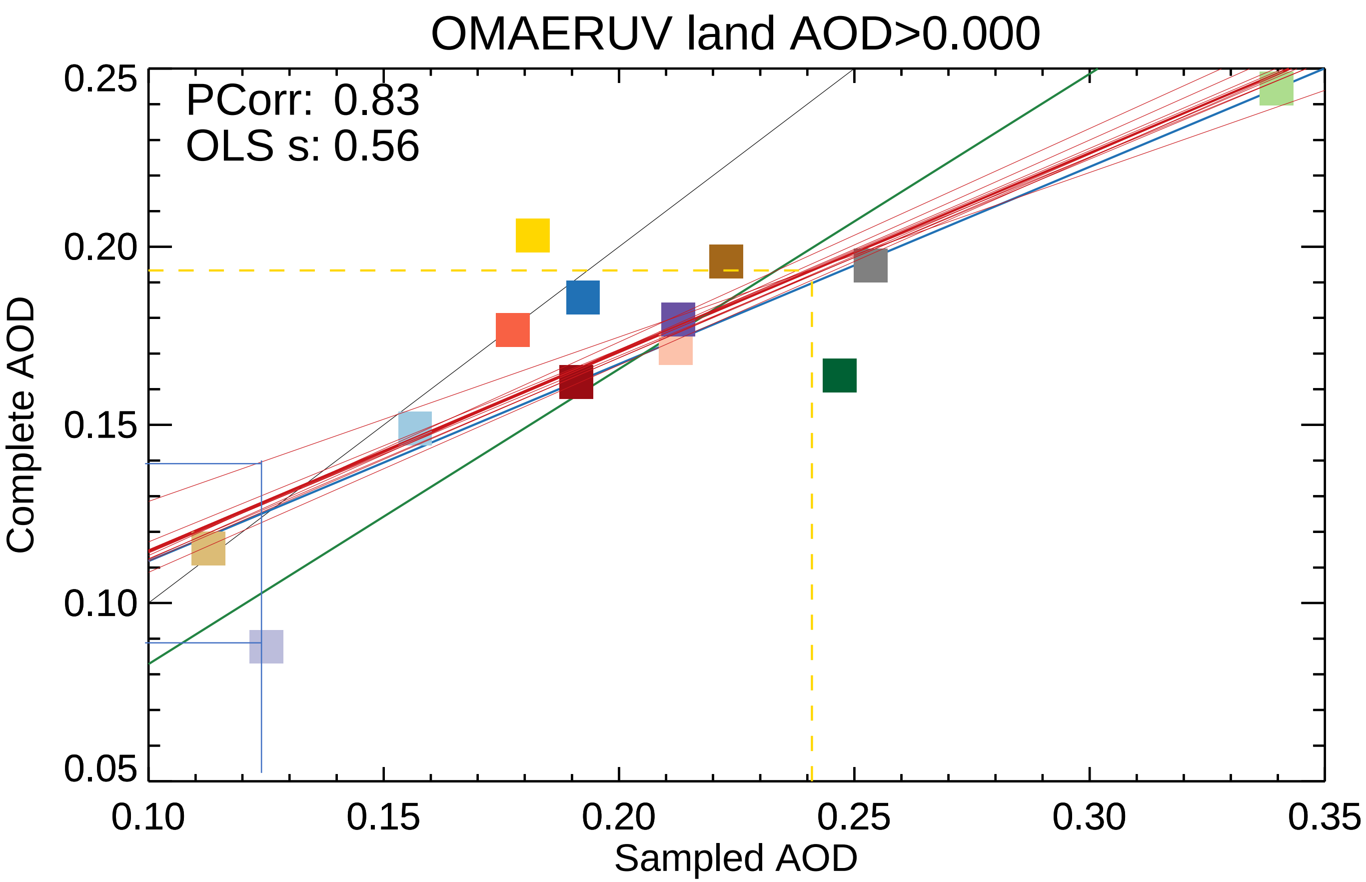
Using model data to homogenize observations



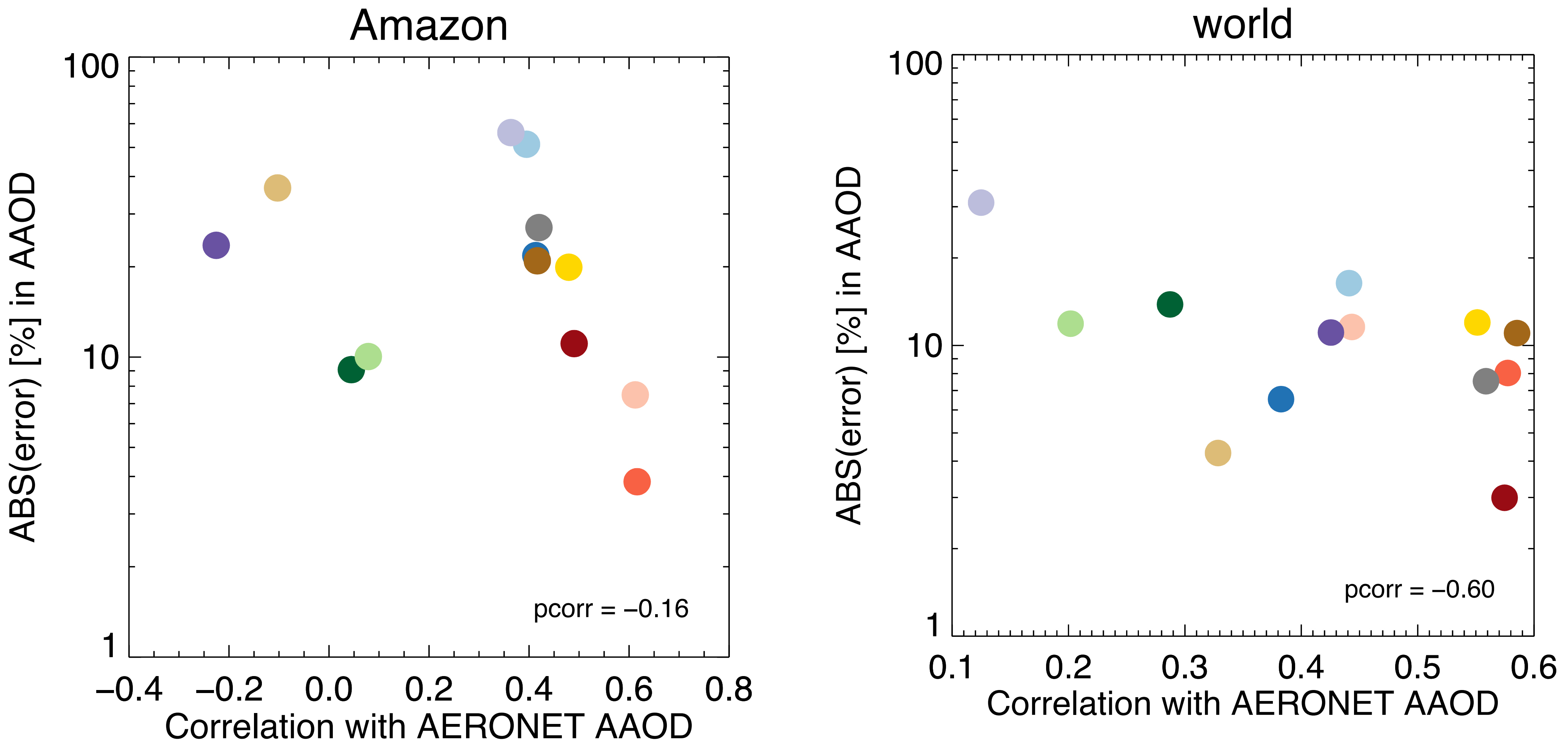
Impact of lower AOD threshold



Using model data to estimate homogenization errors

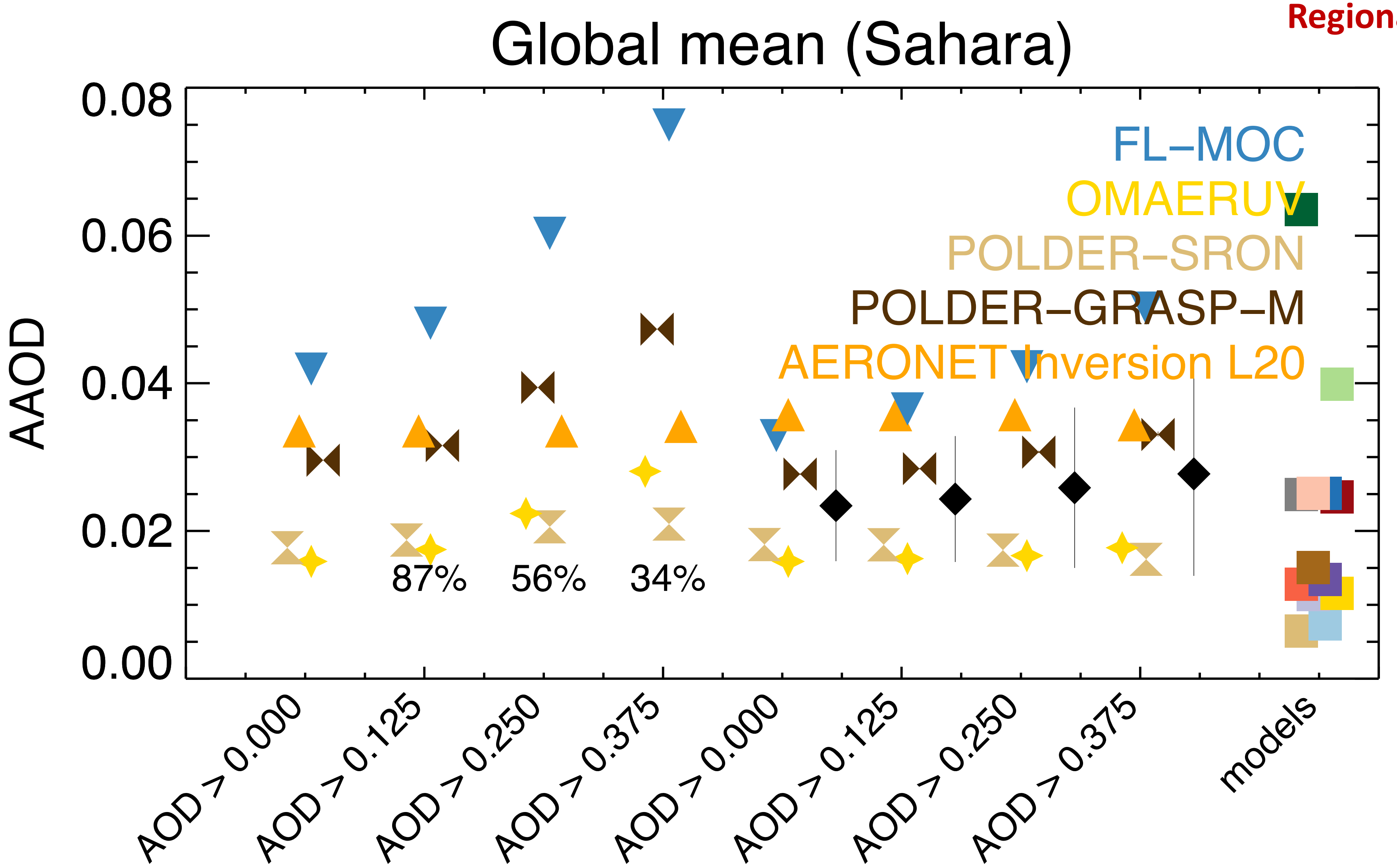


Predicted homogenization errors vs model performance



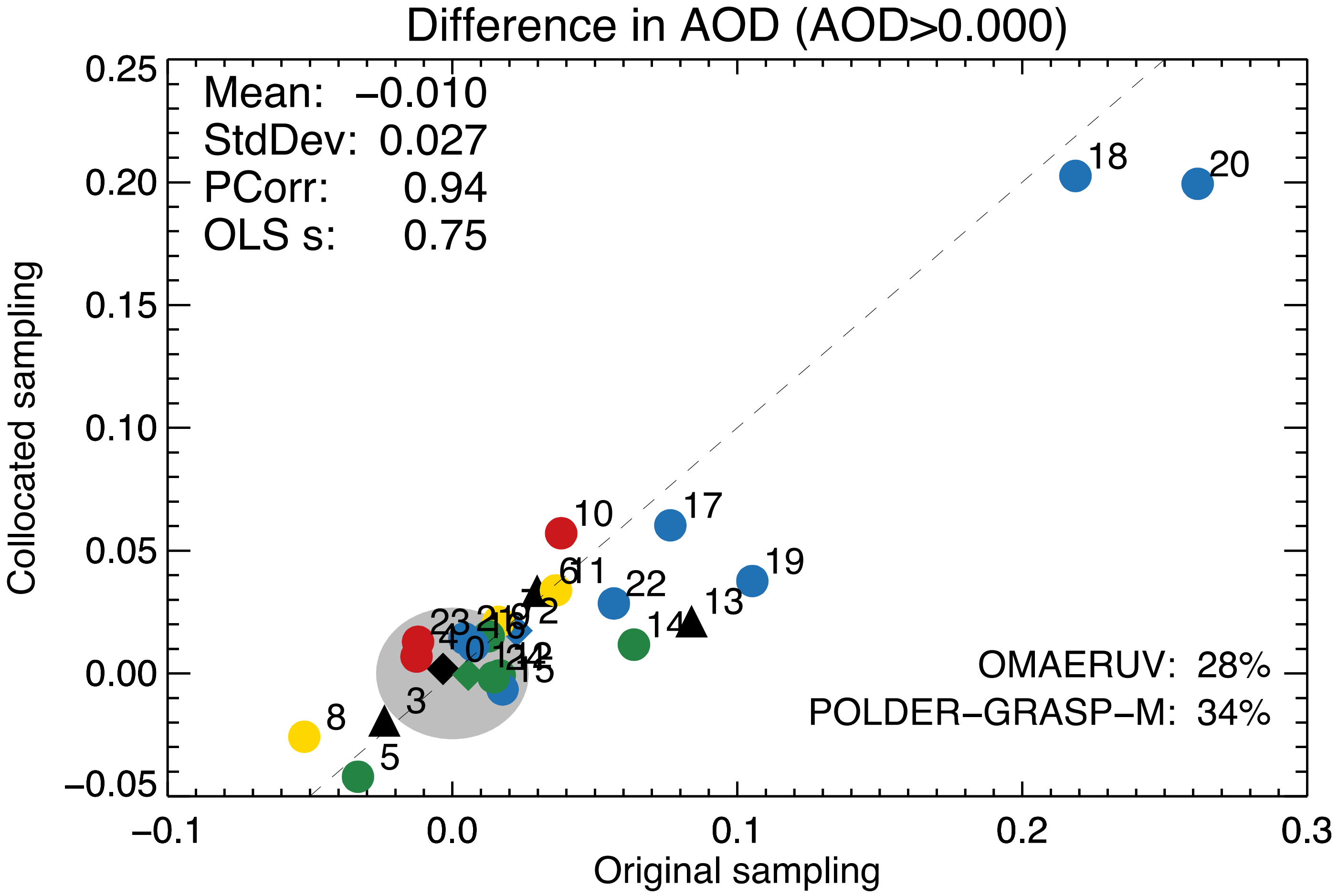
Using real observations:

Regional, 3 year estimates



Impact of retrieval biases

Difference due to retrieval errors only



Difference due to homogenization and retrieval errors

Global AOD & AAOD over land and ocean

Does aerosol life-time and extent of outflow have an impact?

Region	AOD
world	0.153 ± 0.008
land	0.188 ± 0.012
ocean	0.143 ± 0.014
land + ocean	0.157

Only 2.5 % difference in AOD

AAOD	Region
0.0085 ± 0.0017	world
0.0140 ± 0.002	land
0.0065 ± 0.0016	ocean
0.0088	land + ocean

Only 3.5 % difference in AAOD

Global AOD from different ensembles

Global AOD	Products	Datasets	paper
0.156 ± 0.014	AOD	14 (incl. MODIS, AATSR)	Schutgens et al. *ACP* 2020
0.153 ± 0.008	AOD & AAOD	4 (incl. POLDER)	Schutgens et al. *ACP* 2021

There is a single dataset (OMAERUV) that is present in both ensembles

Note: the 1st ensemble contains 7 datasets that mostly/only contain land data

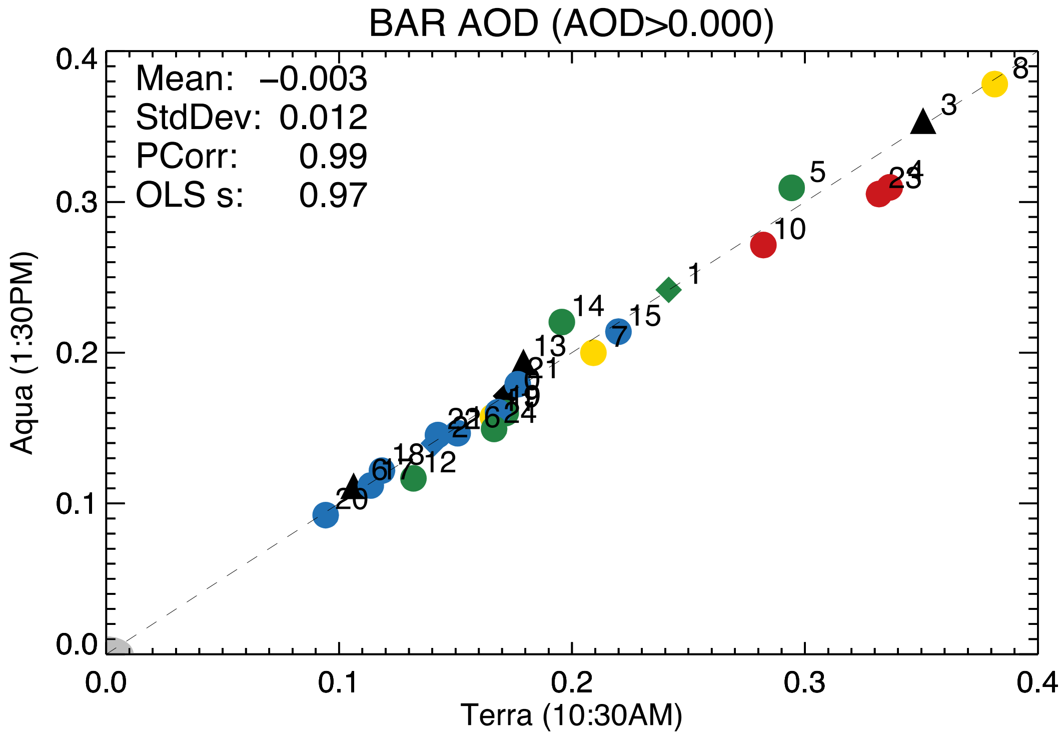
Note: in the 2nd ensemble, a weighted mean is calculated based on AERONET evaluations. The weights for the POLDER products are twice as high as for OMAERUV and FL-MOC.

Simpler estimate on smaller ensemble (7 datasets): 0.155 ± 0.018

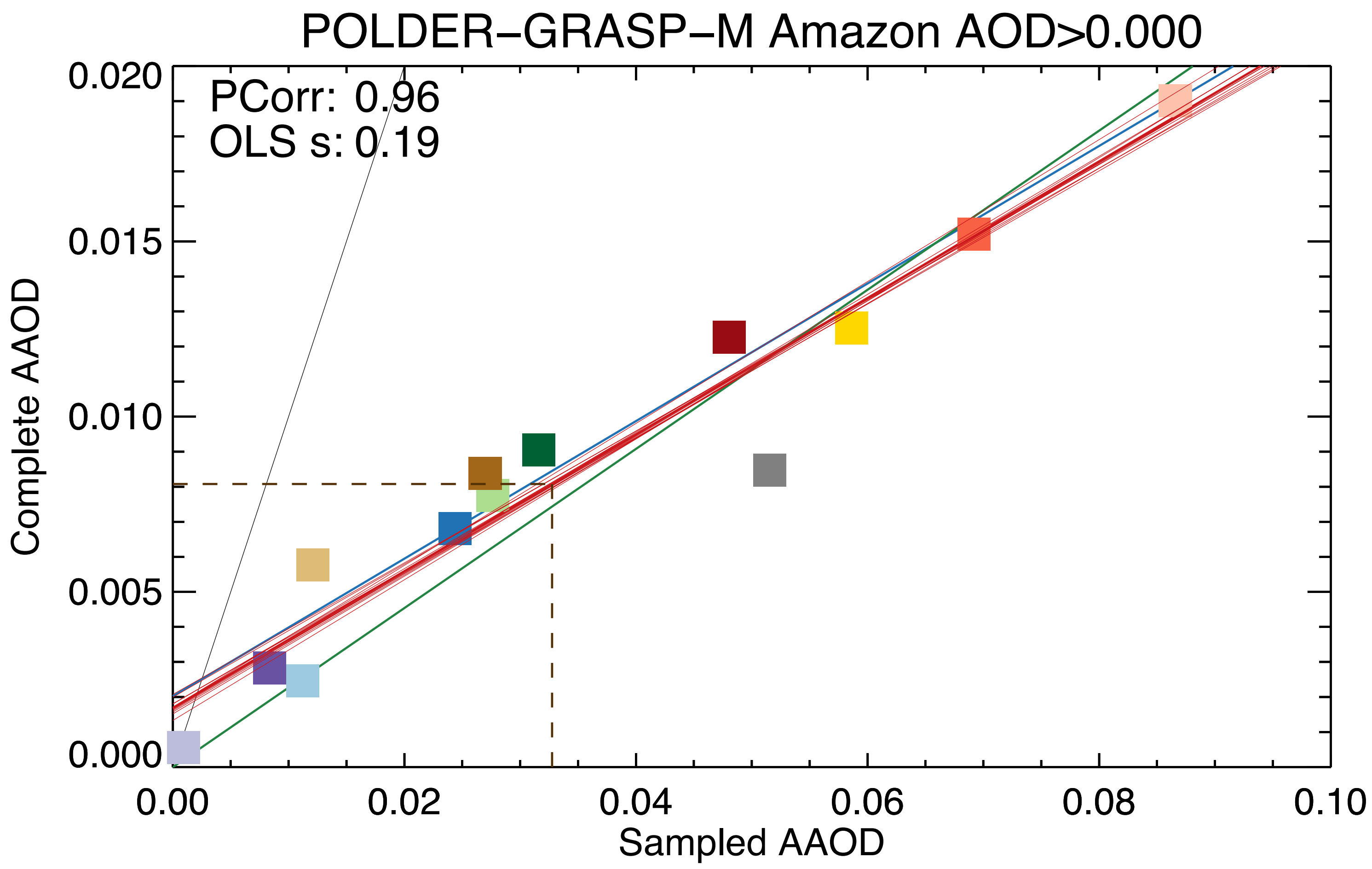
(Watson-Parris et al. GRL 2020)

Global AOD from morning & afternoon platforms

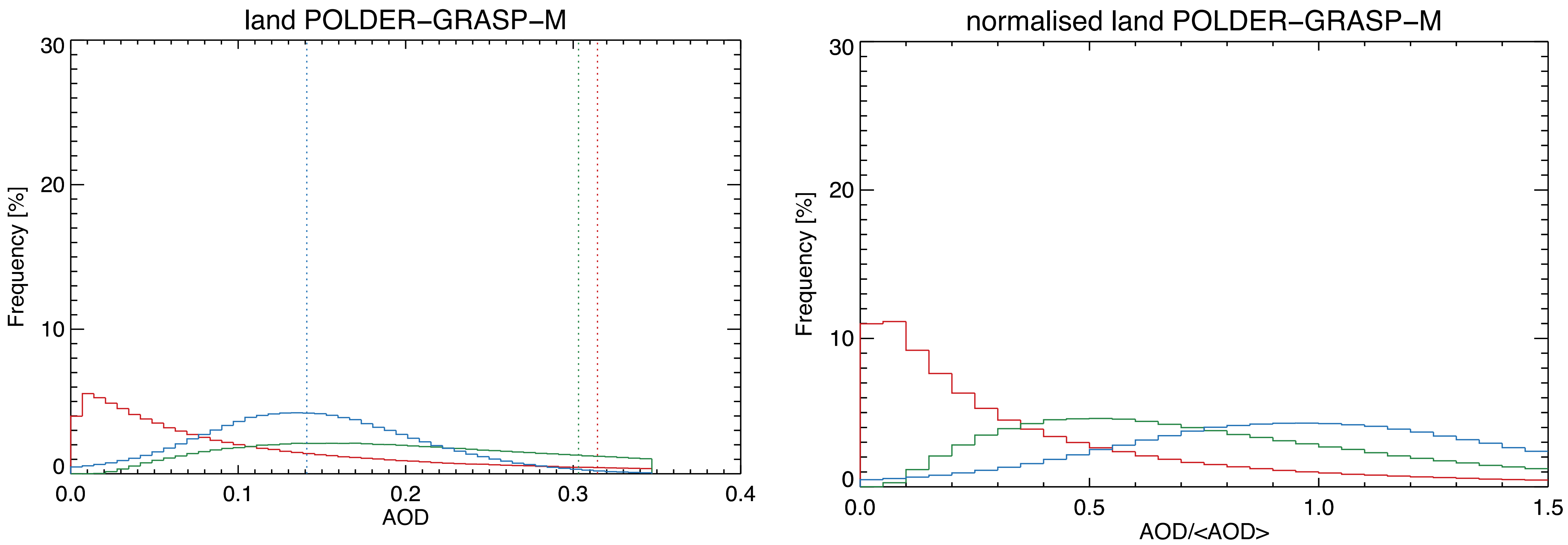
Land AOD	Products	Datasets	Platform (LCT)
0.216 ± 0.042	AOD	DeepBlue, MAIAC, BAR	Terra (10:30AM)
0.217 ± 0.040	AOD	DeepBlue, MAIAC, BAR	Aqua (01:30PM)



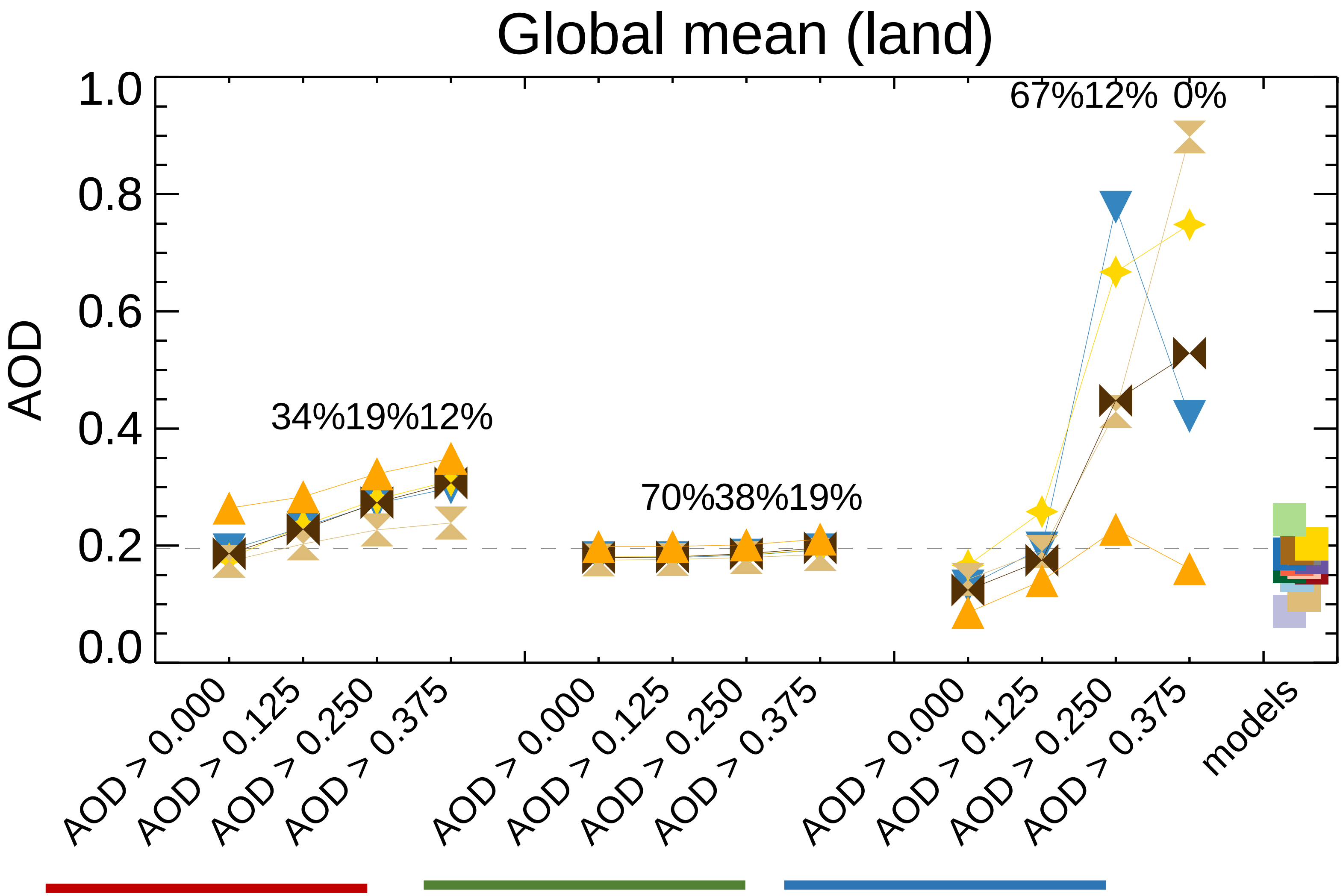
Using model data to homogenize observations



Model errors result in changes to distribution



Sampling allows probing the distribution



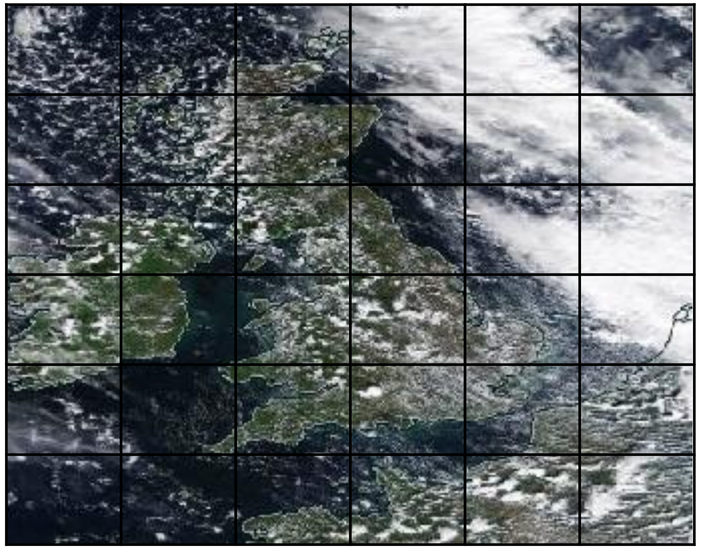
All-sky vs. clear-sky AOD

The problem of partial cloudiness
when comparing model and satellite aerosol fields

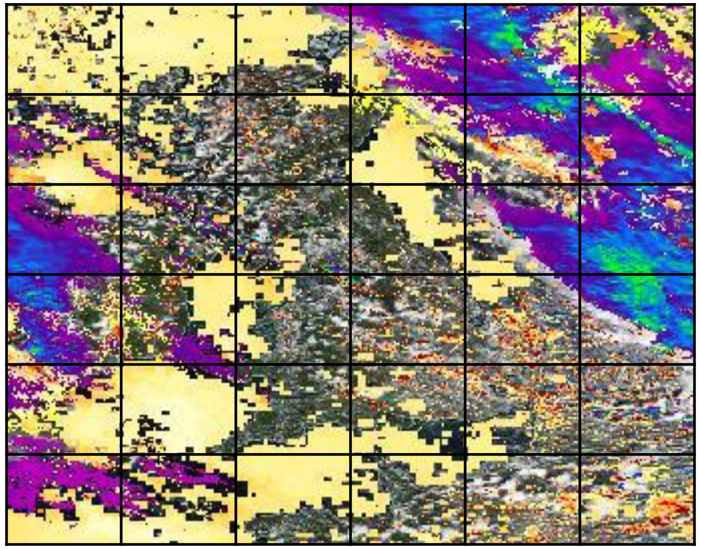
Andrew M Sayer (USRA/NASA GSFC) and Kostas Tsigaridis (Columbia University/NASA GISS)

Problem statement

Satellites (generally)	Climate models (generally)
Observe along a swath with some revisit frequency	Simulate globally with fixed output cadence
Pixels of order ~0.1-10 km	Grid of order ~10s-100s km
Retrieve aerosol properties in the absence of clouds	Model aerosols everywhere
Retrieve cloud properties with some lower detection limit	Parameterize sub-grid cloudiness
Some pixels are not retrieved by either discipline	Water needs to be partitioned between cloudy and cloud-free grid cell partitions, and many aerosols are hygroscopic

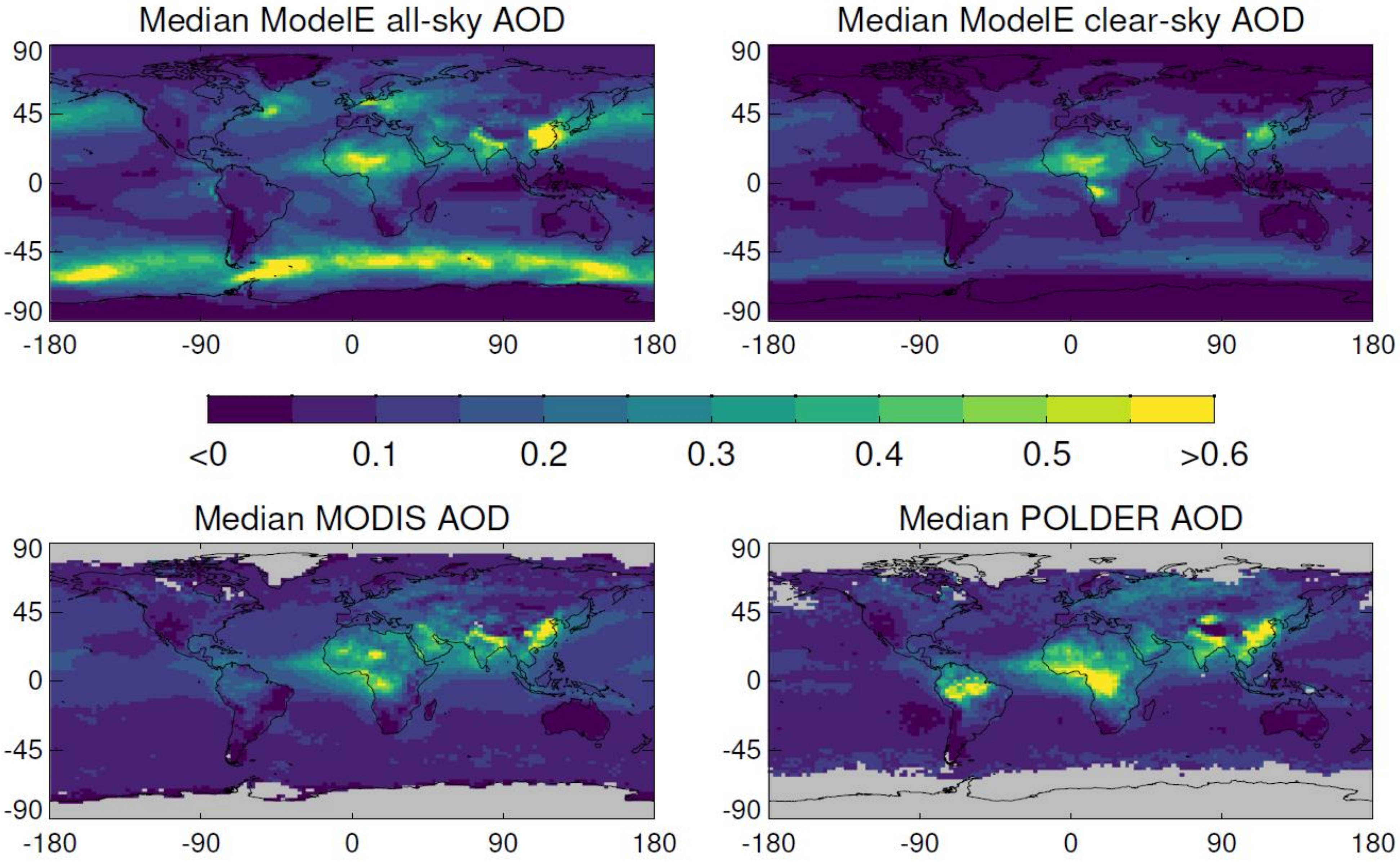


MODIS Aqua image over ~6x6 GISS ModelE grid cells



MODIS Aqua aerosol (light yellow) and cloud (orange-red and cold colours) optical depth

How different are modeled all-sky and clear-sky AOD?



ModelE, MODIS, and POLDER AOD at 550 nm Median of year 2010, gridded to ModelE resolution.

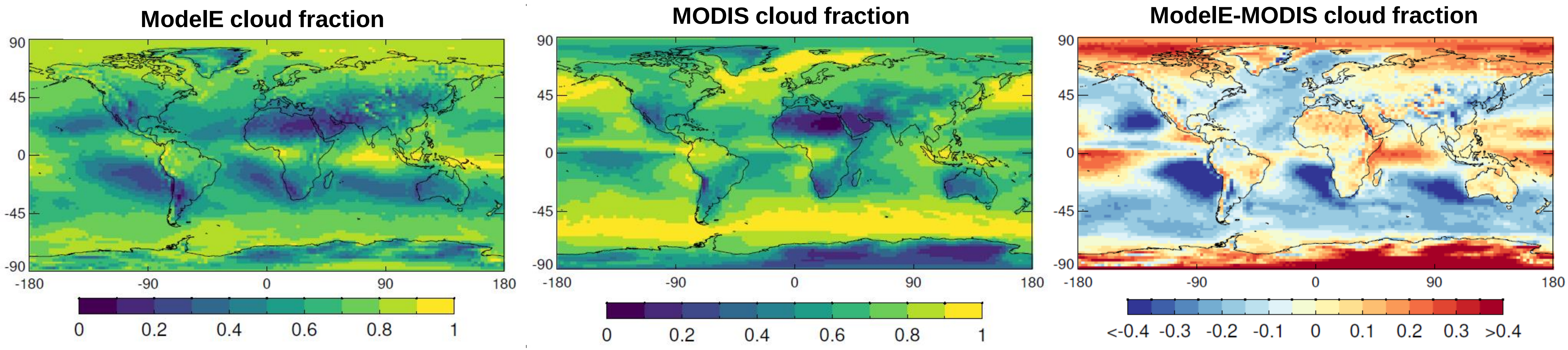
Only shown for satellites for cells with 10+ days.

Data mapped prior to colocation. The sampling of the four are quite different!

Finding some data…

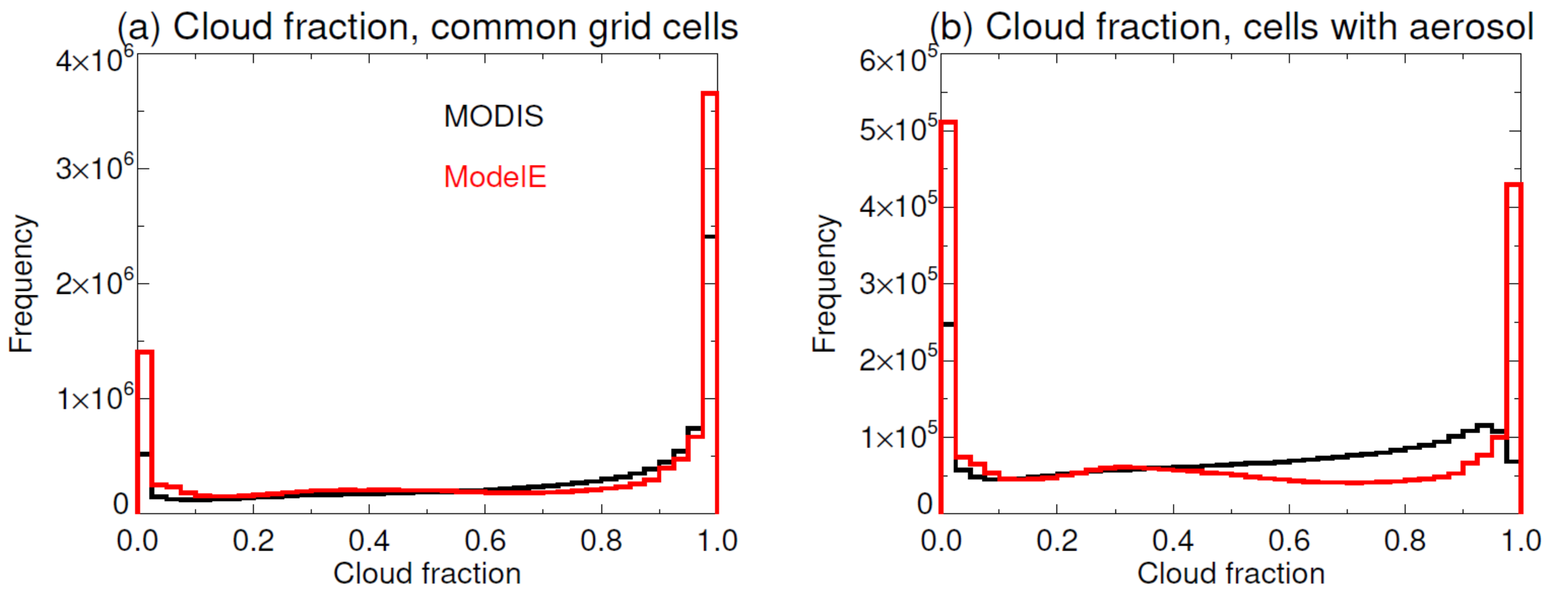
- **GISS ModelE** simulations for year 2010, 2° (lat) x 2.5° (lon) grid, 30 min timestep
  - All-sky and clear-sky AOD at 550 nm
  - Prognostic cloud fraction
- Custom satellite L3 on *model space/time scale* for year 2010
  - **MODIS cloud mask**, Terra+Aqua c6.1
  - **MODIS DB/DT aerosol** optical depth at 550 nm, Terra+Aqua c6.1
- Define a **matchup** if MODIS overflew at least 75% of model grid box, and only consider aerosol in cells with >15 retrievals

Are clouds in the same places?



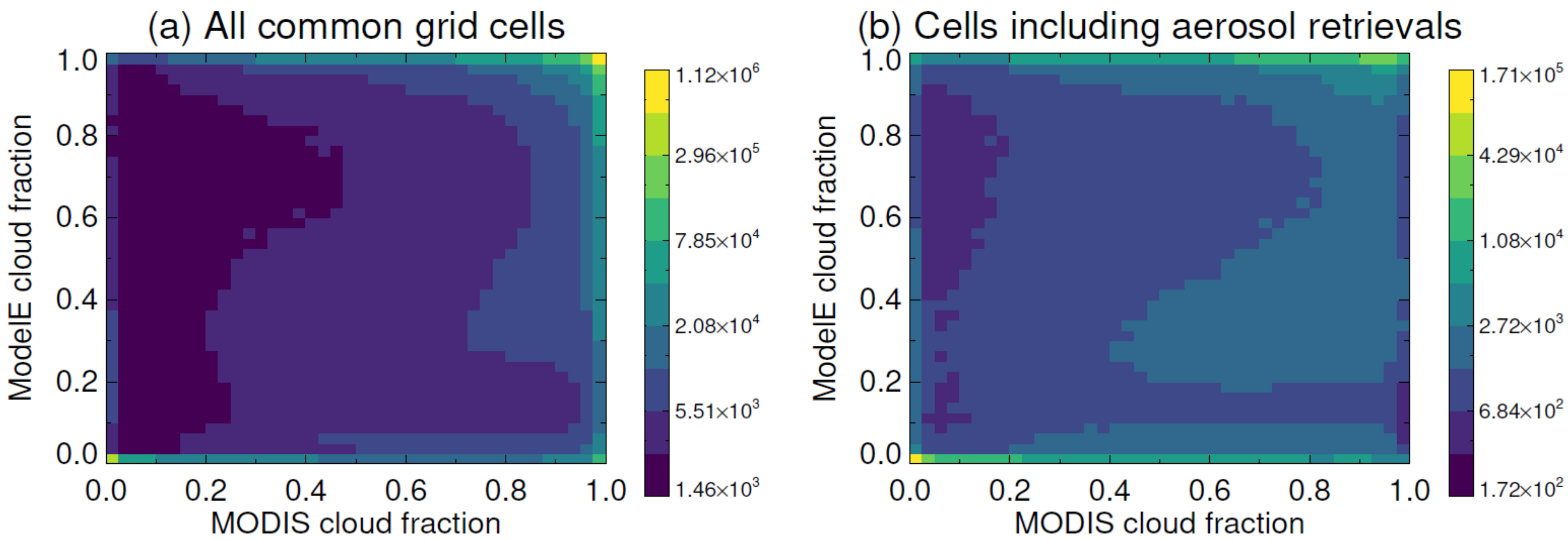
- **Systematic offsets** in matched data between ModelE and MODIS cloud fraction
  - **More** cloud around ITCZ and poles
  - **Less** cloud in stratocumulus decks
  - **Mixed** over land
- Remember satellites aren't perfect

What about sub-grid cloudiness?



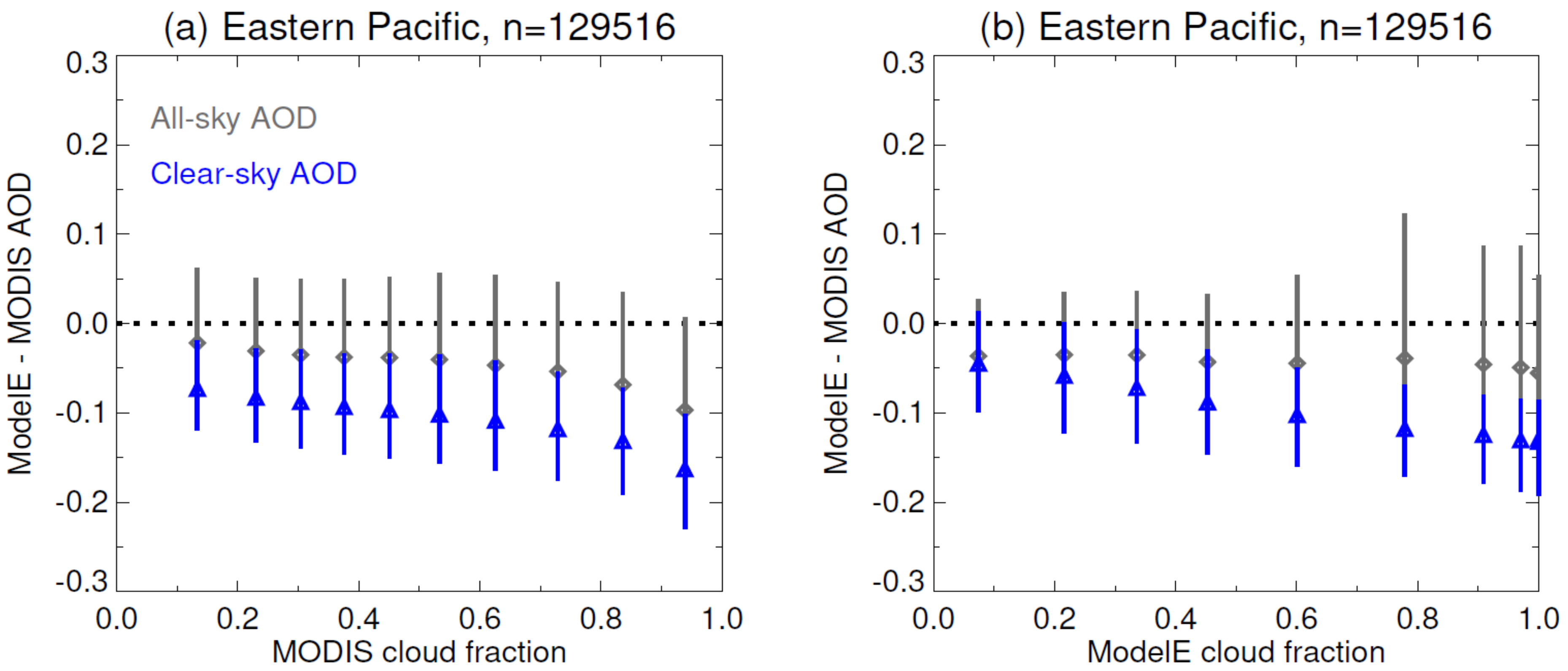
- Globally, most grid cells are either **pretty cloud-free** or **pretty cloudy**
- This is true for **both MODIS and ModelE**
- ModelE has more fully clear/fully overcast cells
- Reminder: “with aerosol” = 15+ MODIS aerosol retrievals

What about sub-grid cloudiness?



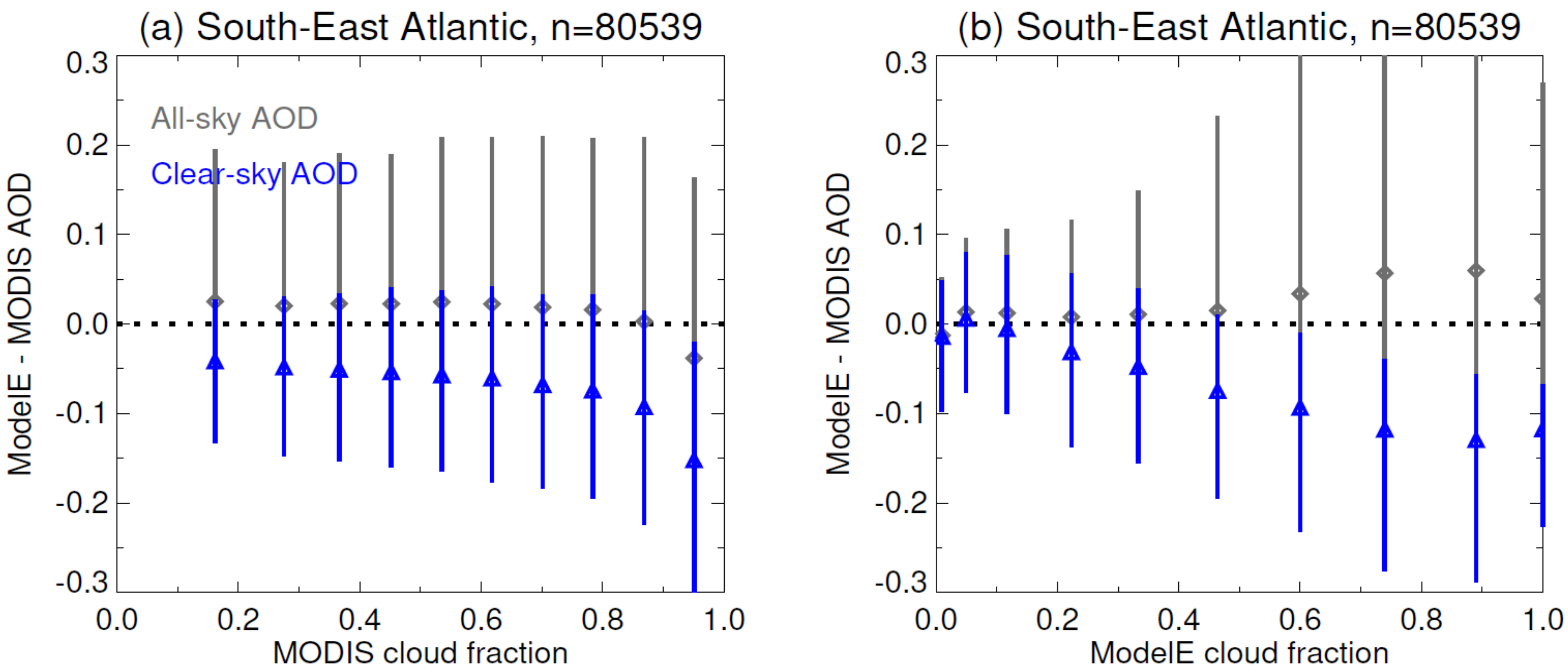
- A 2d version of the previous slide
- **Bottom left/top right corners** remind us that most 2° x 2.5° grid cells are near **fully clear** or **fully overcast**
- **Not much agreement** on sub-grid cloudiness for **partially cloudy grid cells**
- So, look at aerosol patterns vs. both sets of cloud fraction

A look at the tropical eastern Pacific Ocean



- ModelE AOD is **lower** than MODIS; offset **more negative** as cloud fraction increases
- Clear vs. all-sky offset **constant** vs. MODIS cloud fraction, **diverges** (as expected) vs. ModelE cloud fraction
- Large variability, even in this fairly clean ocean region

A look at the south-eastern Atlantic Ocean (ORACLES)



- **Less negative offset** but otherwise **similar trends** to Eastern Pacific
- Even **larger variability** likely due to more complicated aerosol systems

Questions/plans

- **How can we use satellite retrievals to interrogate potential model shortcomings of sub-grid cloudiness and partitioning of aerosol water?**
  - How can we tell the relative importance of these two factors?
  - What other confounders should we be controlling for?
- Expand to include GRASP POLDER retrievals and model/satellite fine mode AOD
- Focus on alternative/additional regions
- Add additional models (please talk to us)

Project: AEROSOURCE
Aerosol data assimilation as a tool to detect model errors

Athanasios Tsikerdekis[1,2]

Nick Schutgens[2], Otto Hasekamp[1]

1. Netherlands Institute of Space Research (SRON)
2. Free University of Amsterdam (VU)

A.Tsikerdekis@sron.nl

AEROCOM / AEROSAT 2021
14 October 2021

Emission estimation using AOD & AE & SSA

OBSERVATIONS
Measures light intensity and polarization properties at multiple viewing angles (up to 16) and multiple wavelengths (0.44 to 1.02μm)

UNCERTAINTIES
Estimated by evaluating with AERONET

POLDER (OBSERVATIONS)

Retrieved

ECHAM-HAM (MODEL)

Simulated

MODEL
Simulates different aerosol species (dust/DU, sea salt/SS, organic carbon/OC, black carbon/BC, sulfates/SO_4, water/WAT) for 7 size/hygroscopicity modes (M7 scheme)

UNCERTAINTIES
Ensemble (32 members) where emissions were perturbed.

Aerosol Optical Depth (AOD) ~ Aerosol Amount
Angstrom Exponent (AE) ~ Aerosol Size
Single Scattering Albedo(SSA) ~ Aerosol Absorption

Importance of assimilating aerosol absorption observations
Tsikerdekis et al. 2021 ACP

STATE VECTOR
Emissions factors distinctive for each aerosol species, each grid cell, every 2 days.

LETKF-Smoother

- Spatial localization
- Emission estimated iteratively in time
- Prior correction which takes care of biases

Improved Model Aerosol Emission

Emission estimation method under idealized experiments for SPEXone
Tsikerdekis et al. Submitted GMD

Emissions estimation under idealized experiments (OSSEs)

Emission estimation under idealized experiments (OSSEs) with a “SUPER” sensor

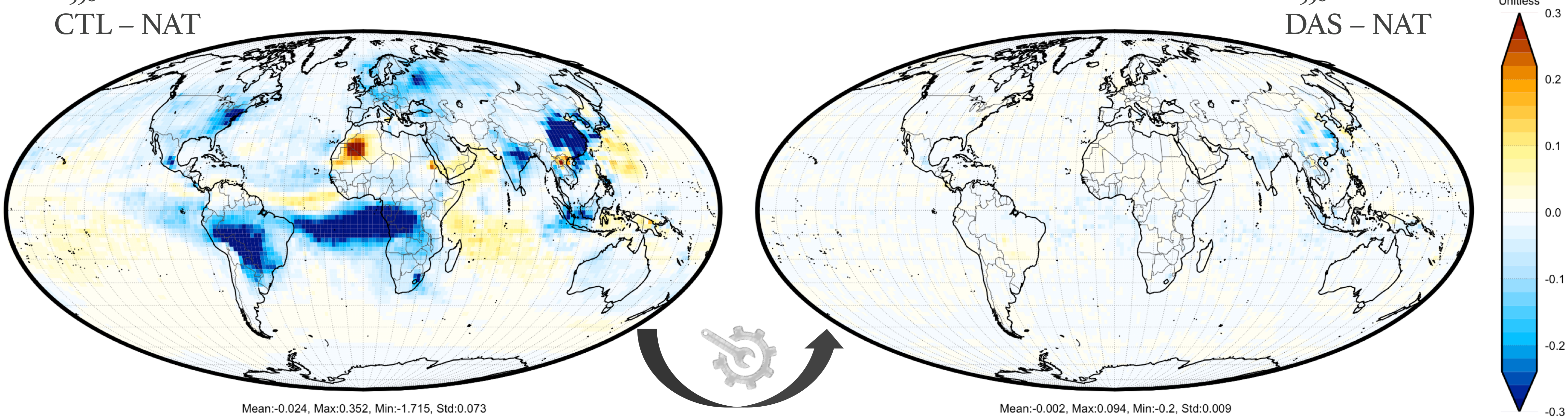
Observation everywhere anytime

Low observation uncertainty

Emissions Errors → Observables Errors

AOD$_{550}$ Differences
CTL – NAT

AOD$_{550}$ Differences
DAS – NAT



Emissions estimation using POLDER observations

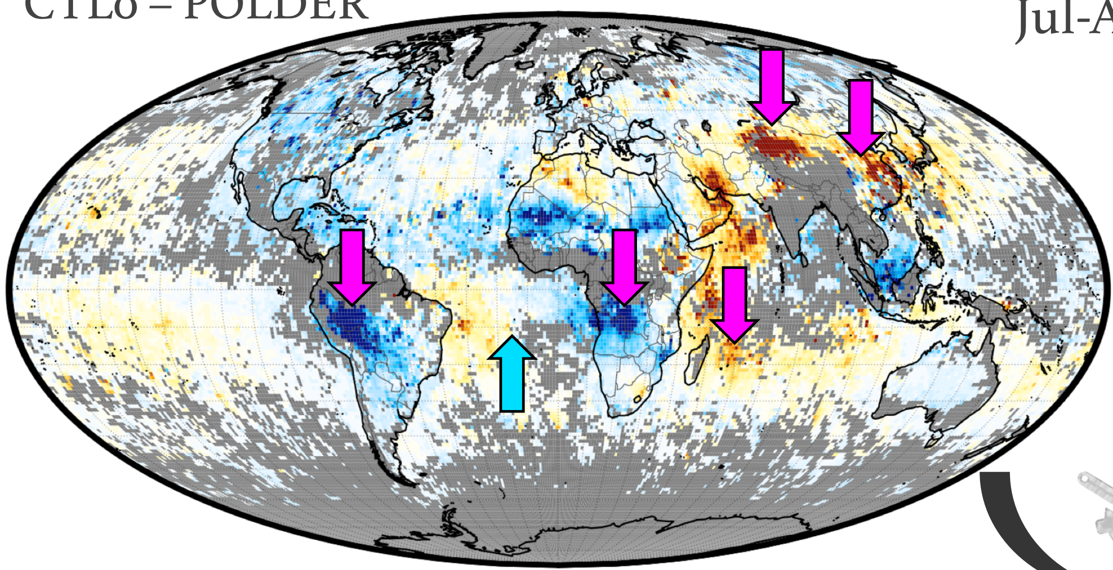
Emissions estimation with POLDER observations it works, but it is more challenging

Spatiotemporal gaps

Higher observation uncertainty (based on AERONET evaluation)

<u>Emission Errors + Meteorology Errors</u> → <u>Observables Errors</u>

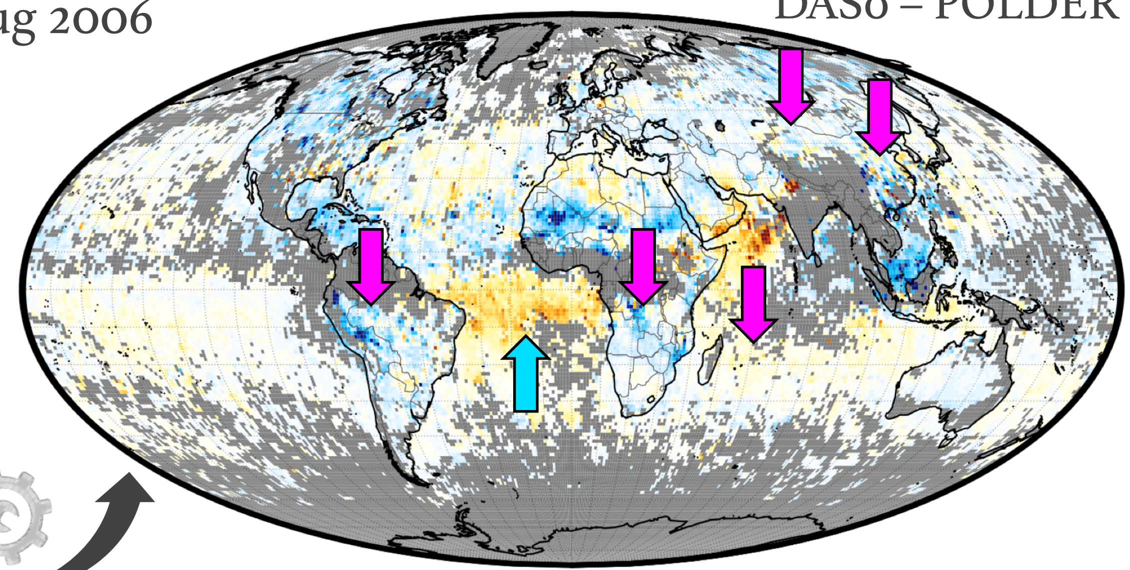
AOD_{550} Differences
CTL0 – POLDER



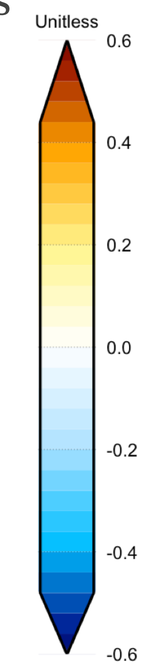
Mean:-0.026, Max:1.997, Min:-1.203, Std:0.178

Jul-Aug 2006

AOD_{550} Differences
DAS0 – POLDER

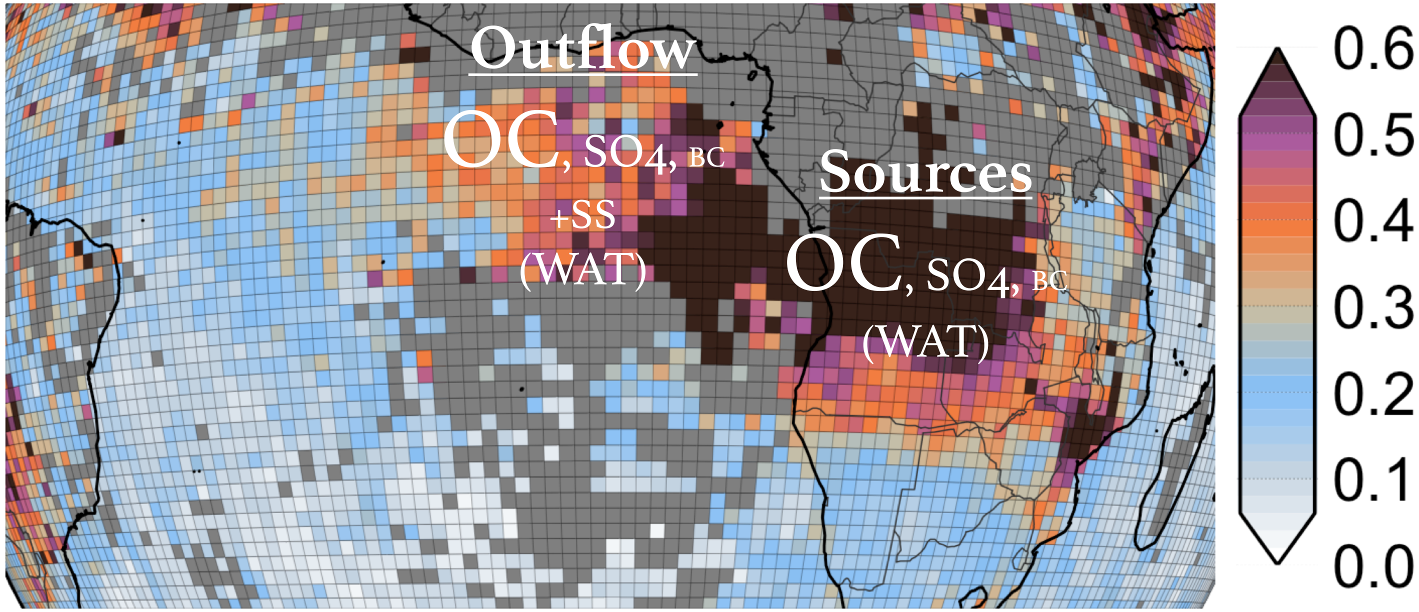


Mean:-0.022, Max:1.167, Min:-1.199, Std:0.123



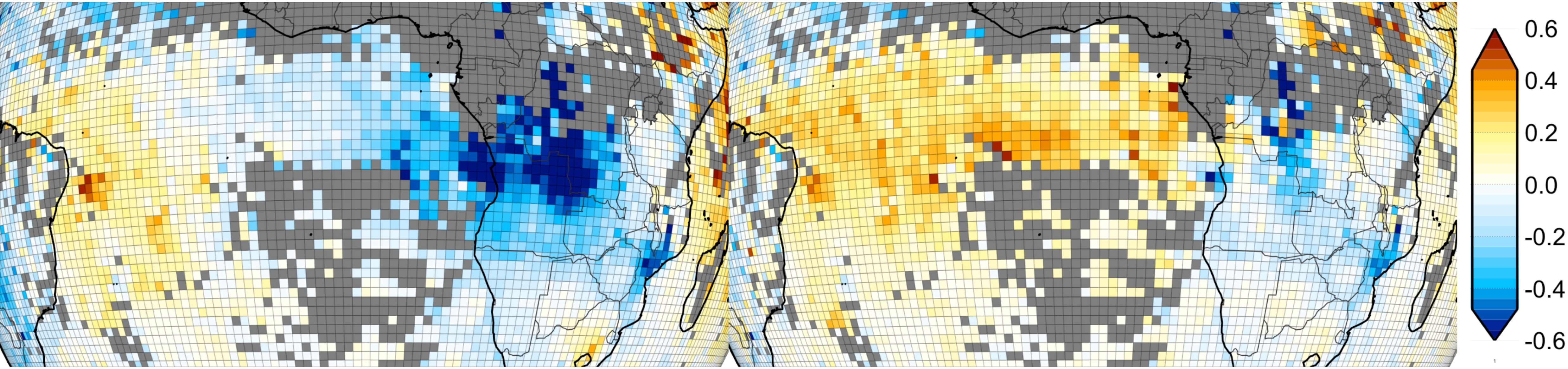
Aerosol outflow in tropical Atlantic

POLDER AOD_{550}

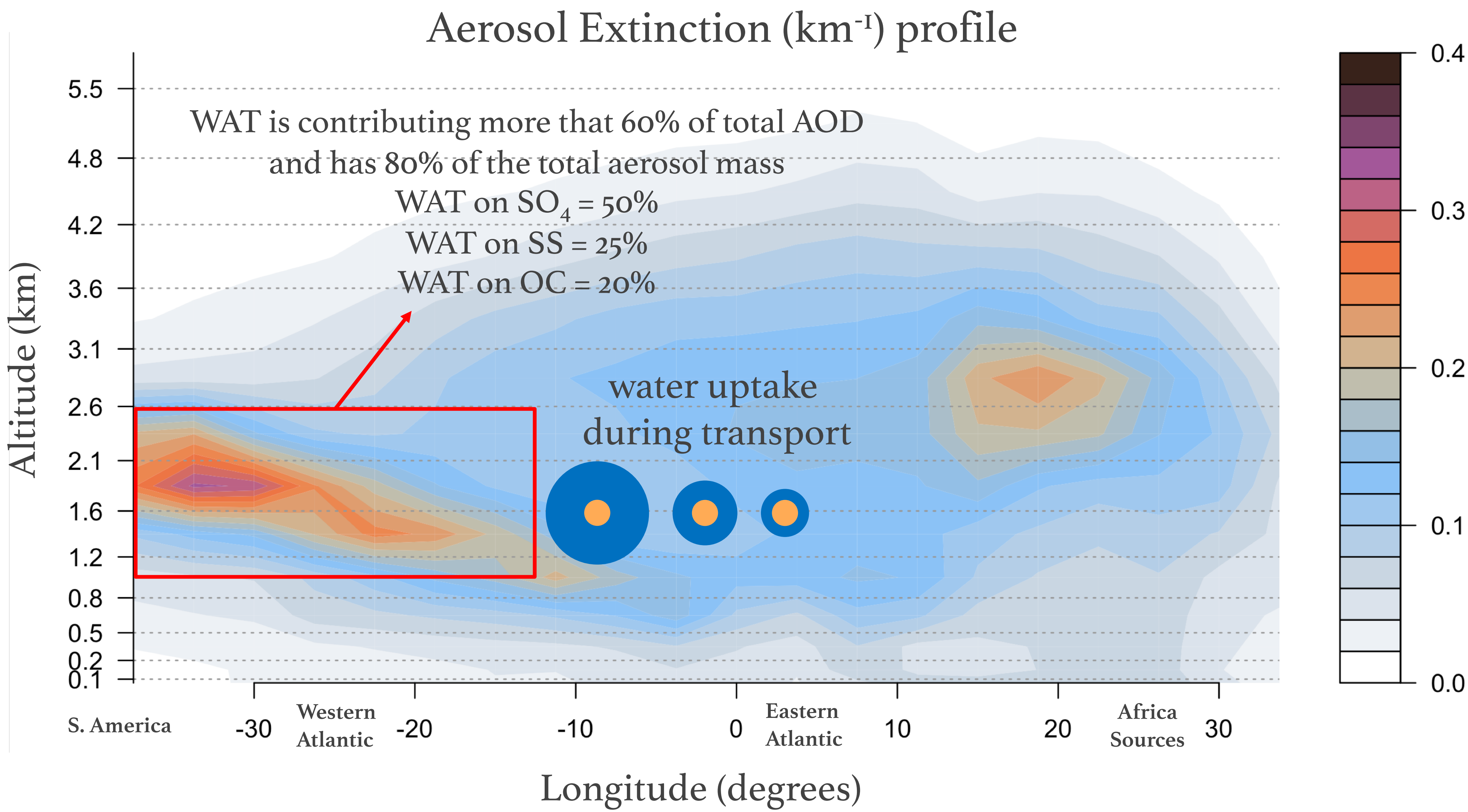


AOD_{550} differences CTL0 – POLDER

AOD_{550} differences DAS0 – POLDER

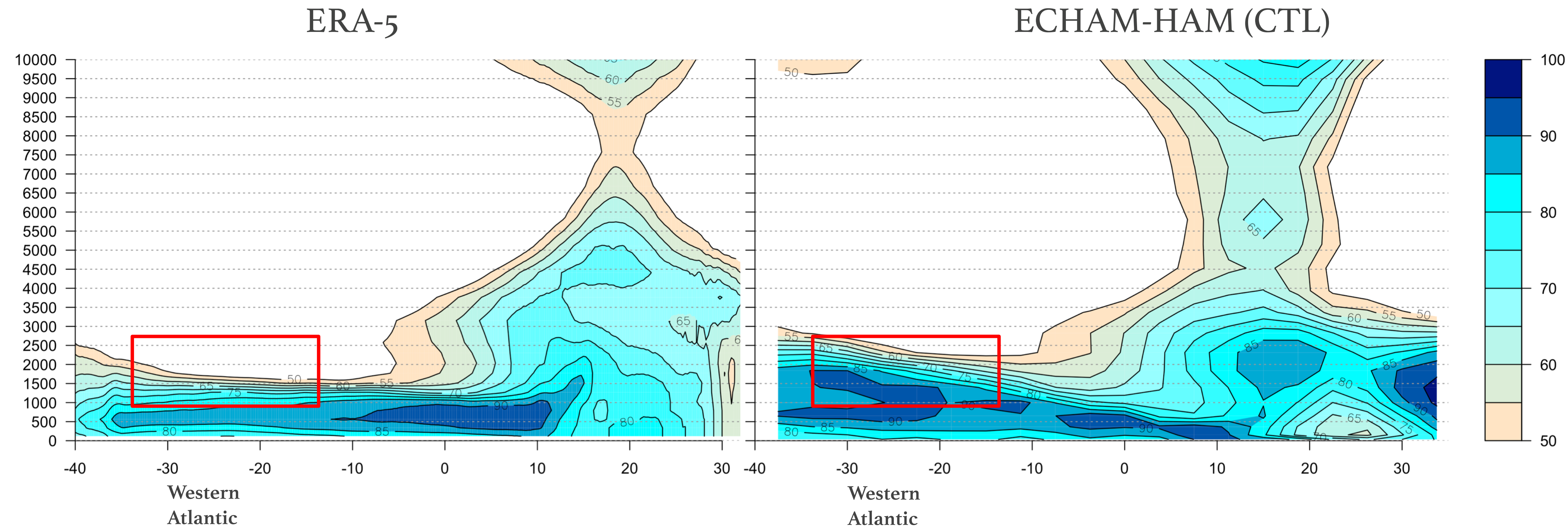


The role of water uptake (I)



The role of water uptake (2)

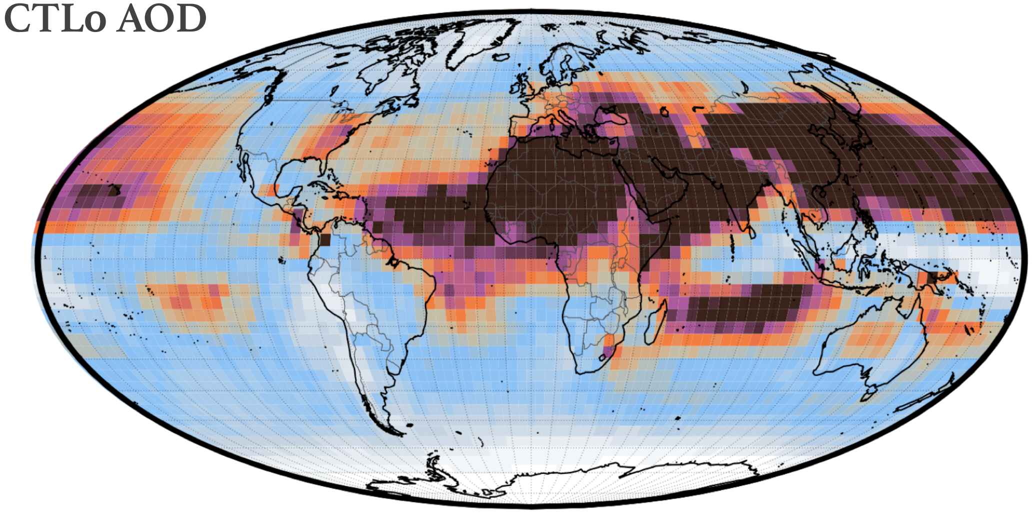
Relative Humidity (%) profiles



Higher relative humidity by the model,
Especially in the western part of the Atlantic

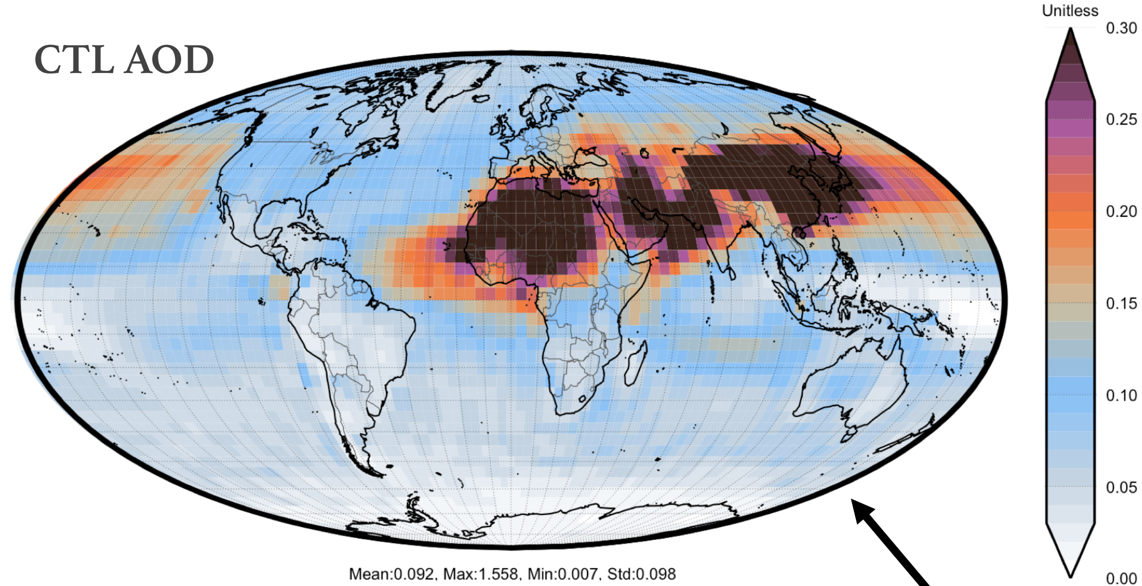
The role of water uptake on AOD

CTLo AOD



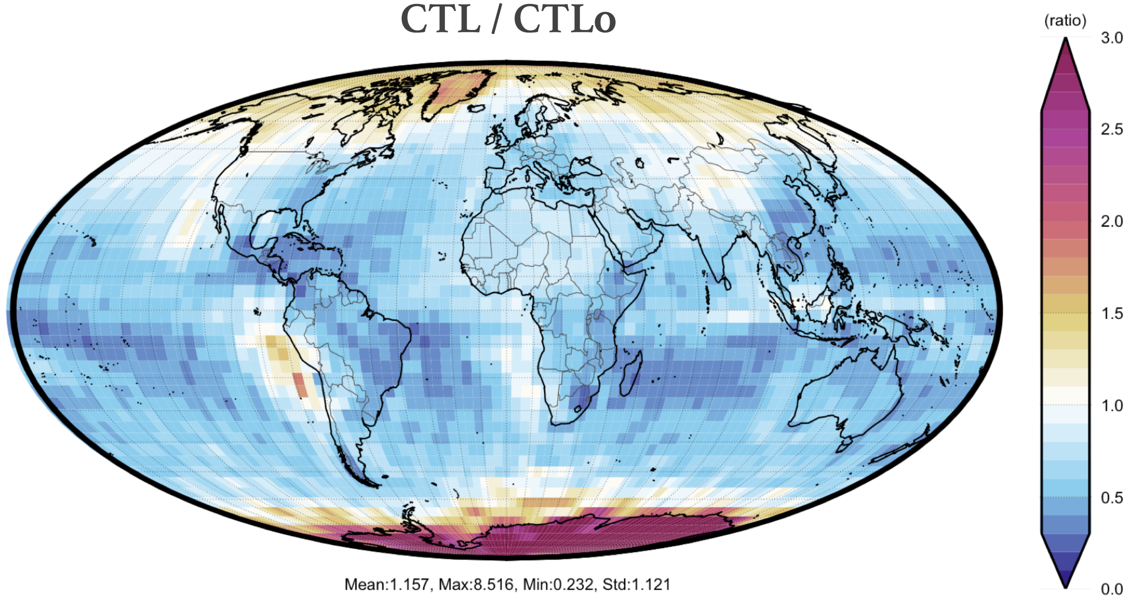
Mean:0.129, Max:1.659, Min:0.003, Std:0.148

CTL AOD



Mean:0.092, Max:1.558, Min:0.007, Std:0.098

CTL / CTLo

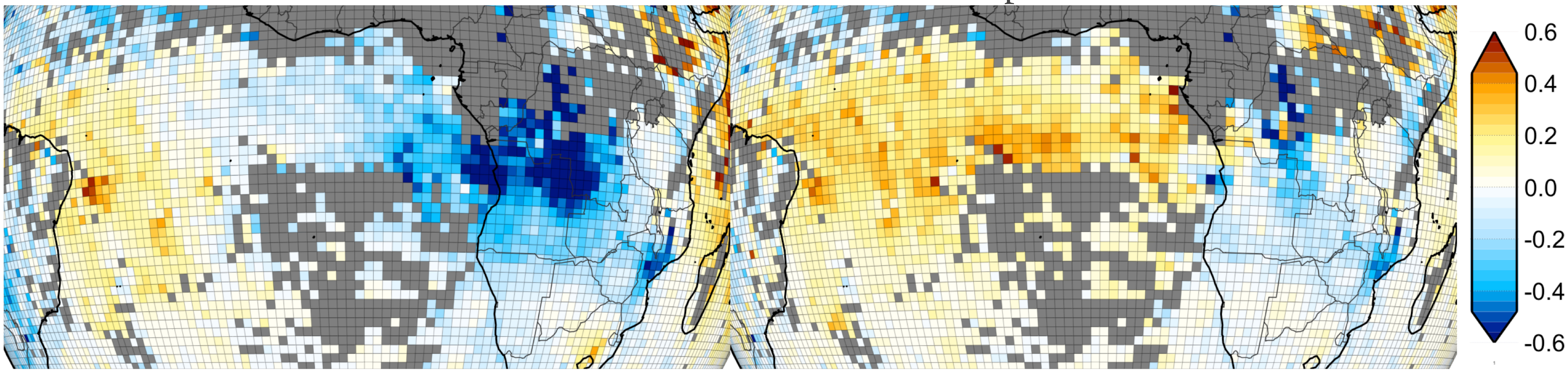


Mean:1.157, Max:8.516, Min:0.232, Std:1.121

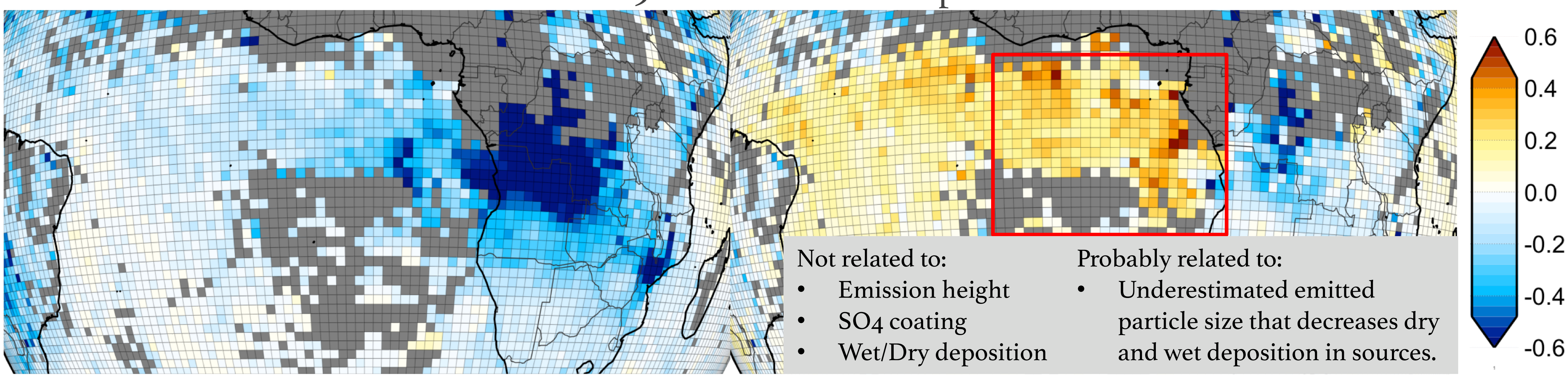
Use ERA-5 RH
for the water uptake only!

The role of water uptake on AOD (POLDER)

CTL0 – POLDER | ECHAM-HAM RH for the water uptake | DAS0 – POLDER



CTL – POLDER | ERA-5 RH for the water uptake | DAS – POLDER



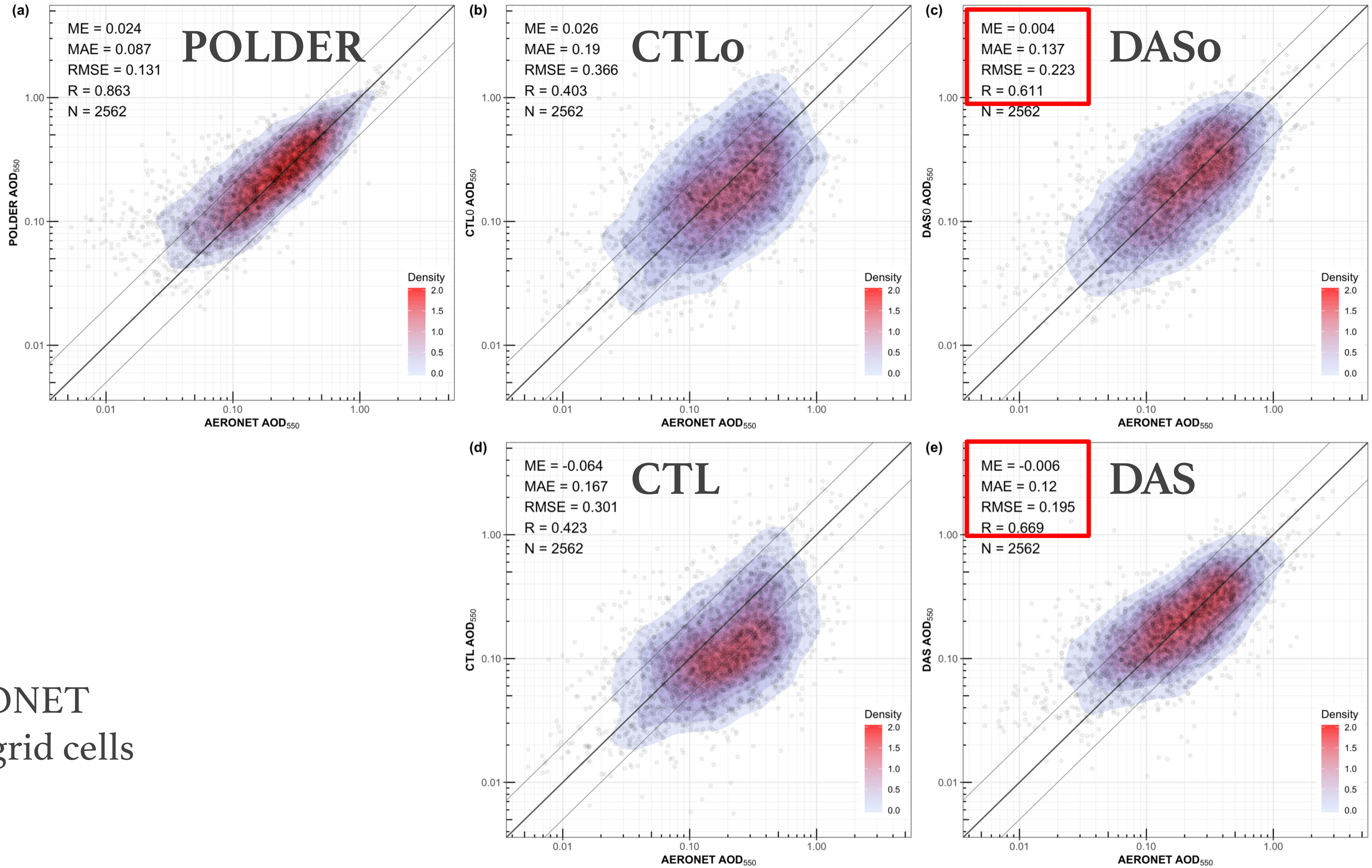
Not related to:

- Emission height
- SO4 coating
- Wet/Dry deposition

Probably related to:

- Underestimated emitted particle size that decreases dry and wet deposition in sources.

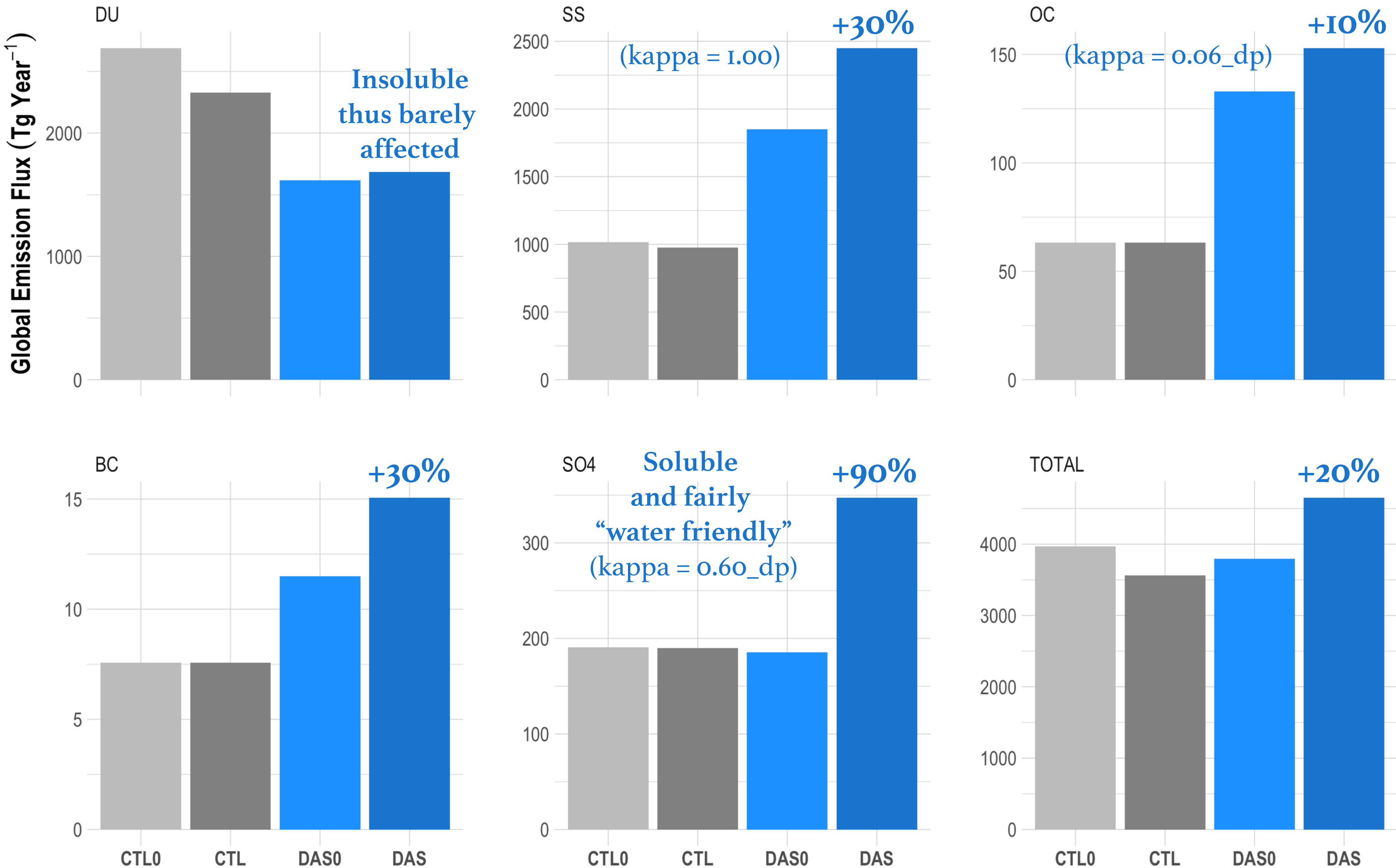
The role of water uptake on AOD (AERONET)



Evaluation with AERONET over retrieved POLDER grid cells for 2006

The role of water uptake on emissions

Global emissions for 2006



Conclusions

Conducted a data assimilation experiment that corrected aerosol emissions and used it as a tool to detect model errors.

- Linked an existing ensemble Kalman filter code with ECHAM-HAM and developed a system that estimates aerosol emissions based on POLDER retrievals.

- In the outflow over tropical Atlantic AOD_{550} errors increase because ***relative humidity*** is overestimated by the model which leads to an overestimation of water uptake.

- Using relative humidity of ERA5 for water uptake in ECHAM-HAM and conducting a second data assimilation experiment with that set up:
  - Improves the AOD in the long-range transport over the Atlantic (Summer)
  - Improves the AOD (absolute error and correlation) against AERONET (2006)
  - Leads to an increase of total emissions by 20% with an almost doubling of SO_4 emissions

A.Tsikerdekis@sron.nl