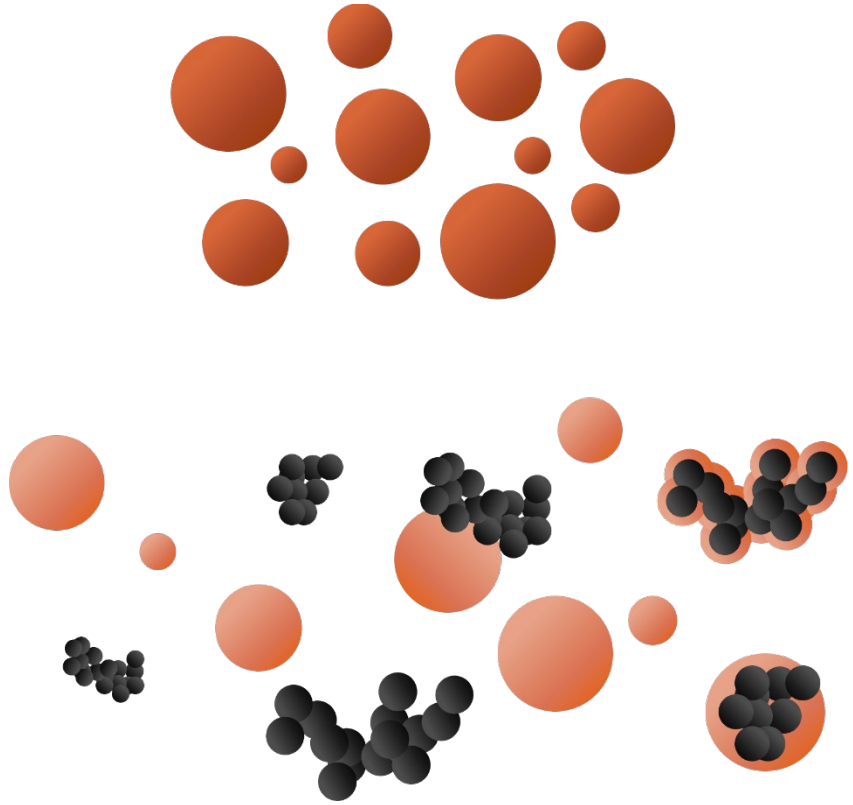
PSI Center for Energy and Environmental Sciences

Labor für Atmosphären-Chemie

Accurate retrieval of pure black carbon aerosol properties including light absorption from polarization-resolved in situ measurements of light scattering



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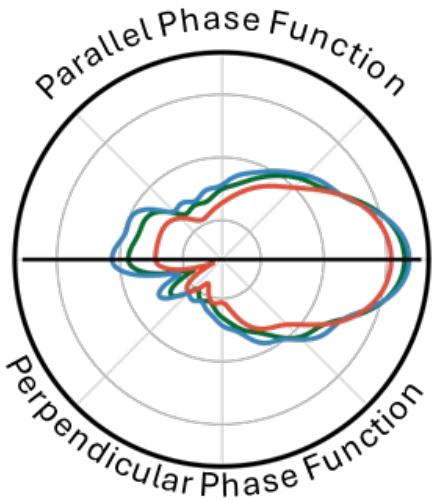
From light scattering to aerosol properties

Spatial and temporal variability on global scale?

Remote sensing
based on light scattering

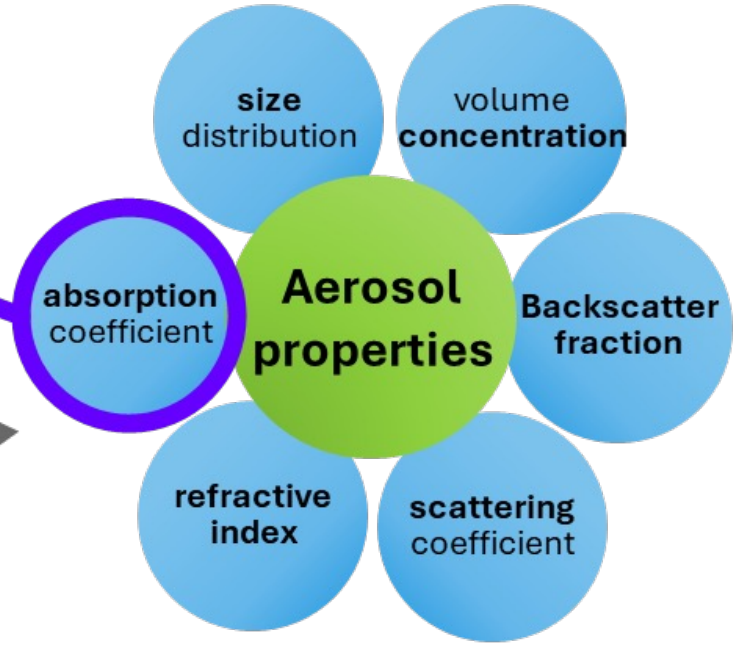
Polarimetric
measurements



Light absorption:

- Major challenge!
- Plays peculiar role in aerosol-radiation interactions & semi-direct climate effects

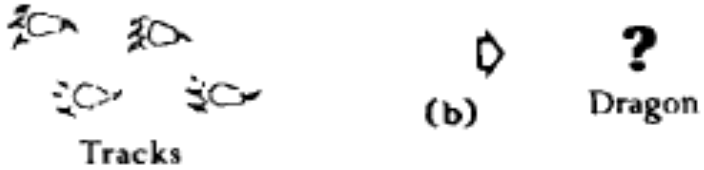
Solving the
inverse problem
of polarimetry

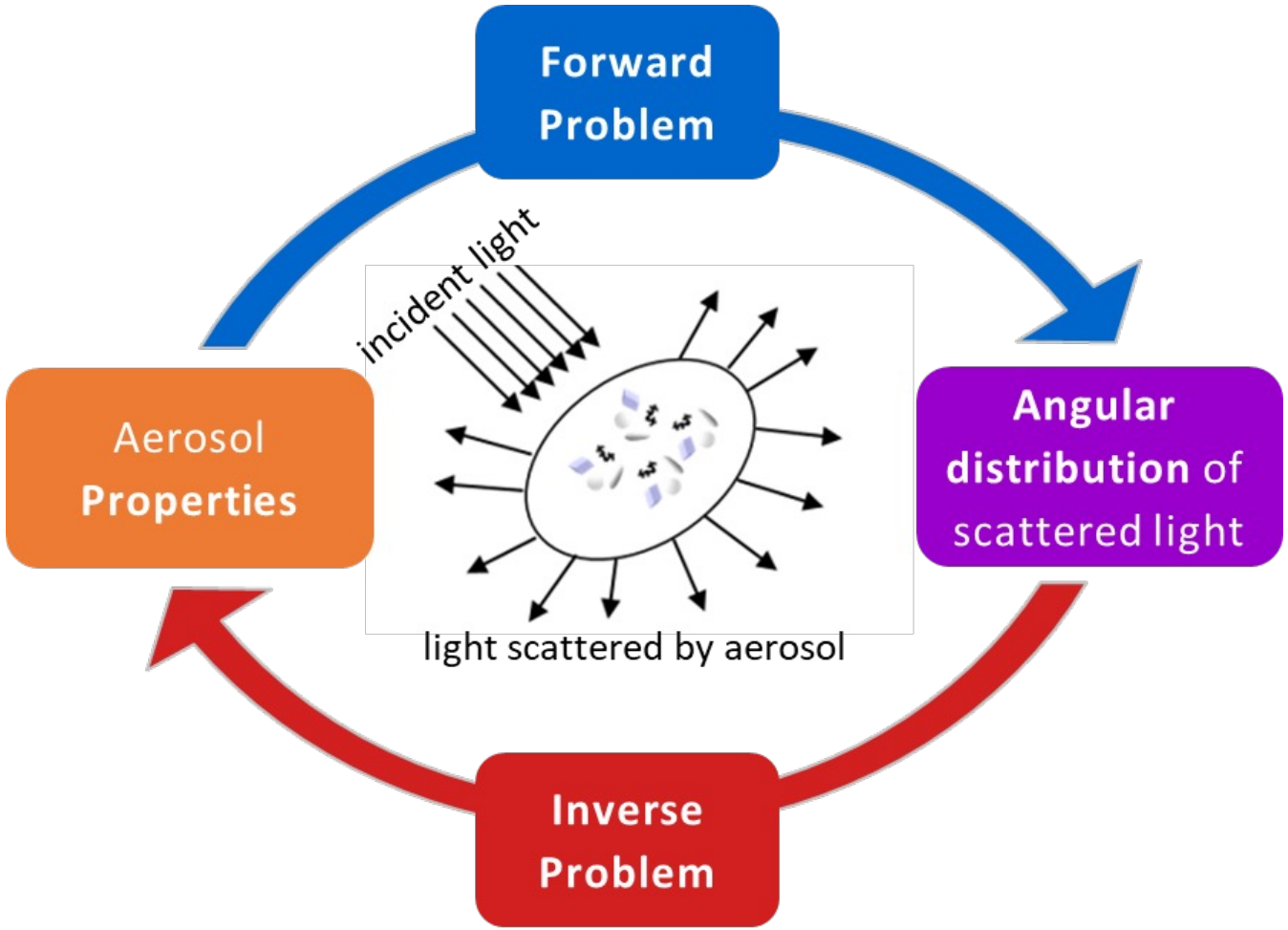


Forward and Inverse Problem of Aerosol Polarimetry

The forward problem
Describe the tracks of a given dragon

The inverse problem
Describe a dragon from its tracks





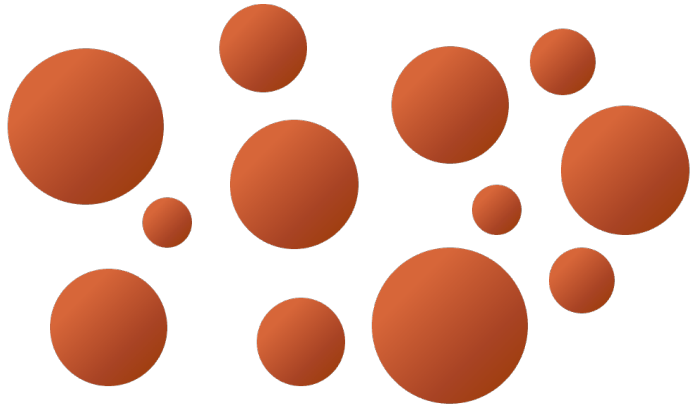
Bohren and Huffmann, 2004

Some problems with retrieval of light absorption

Aerosol model and optical forward kernel in **standard retrieval algorithms** (e.g. GRASP*):

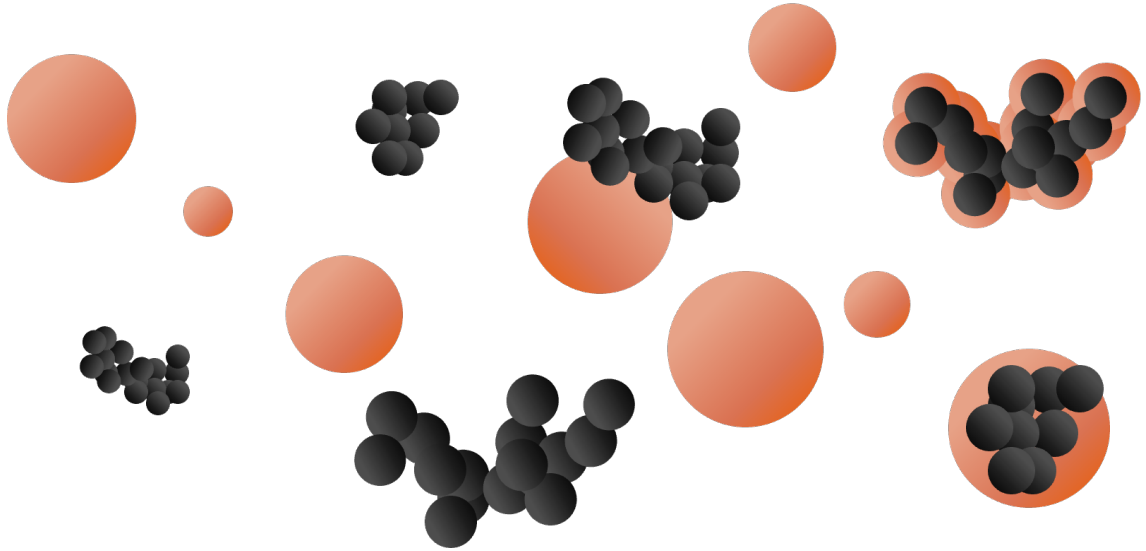
- Kernel is based on ***Mie theory for spherical particles***
- Identical optical material properties assumed for all particles, i.e. ***"homogenous internal mixture"***

*Dubovik et al., 2014



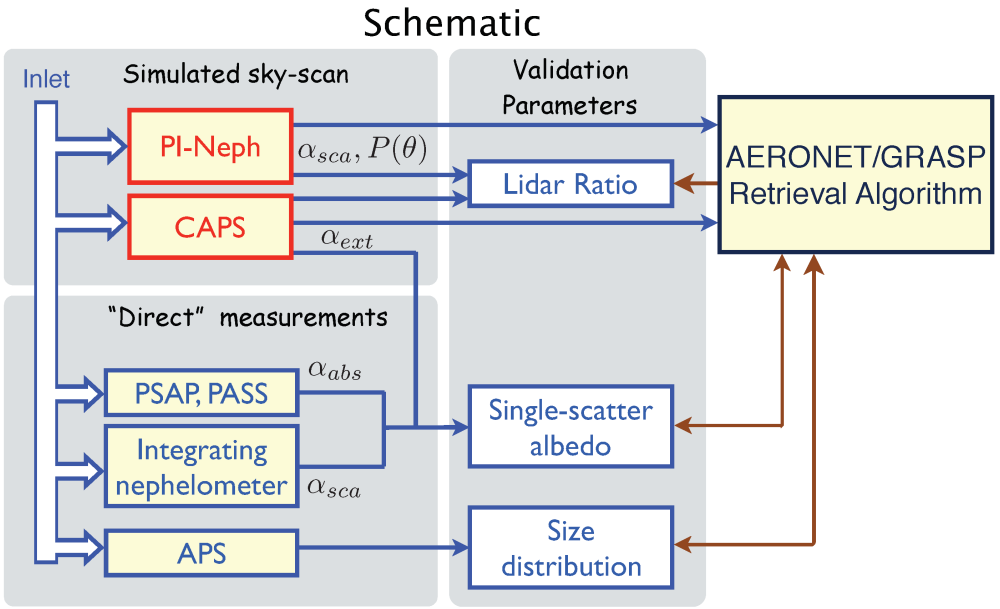
However, **black carbon** particles

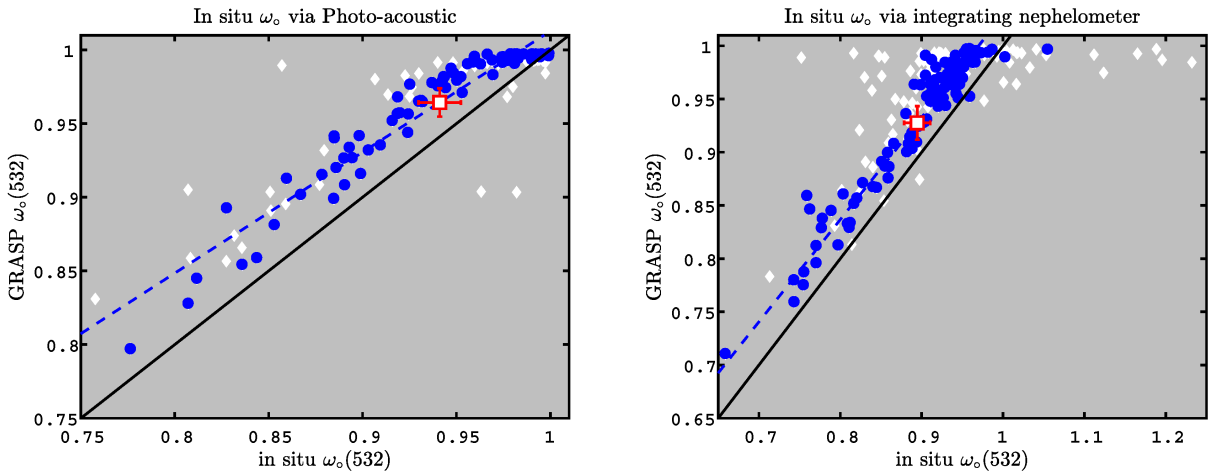
- have ***non-spherical shape***
- are ***not homogeneous***
- are to some extent ***externally mixed***



➔ Mismatch of aerosol model and optical kernel with real aerosol properties can cause systematic retrieval bias.

Previous laboratory study using in situ polar nephelometer





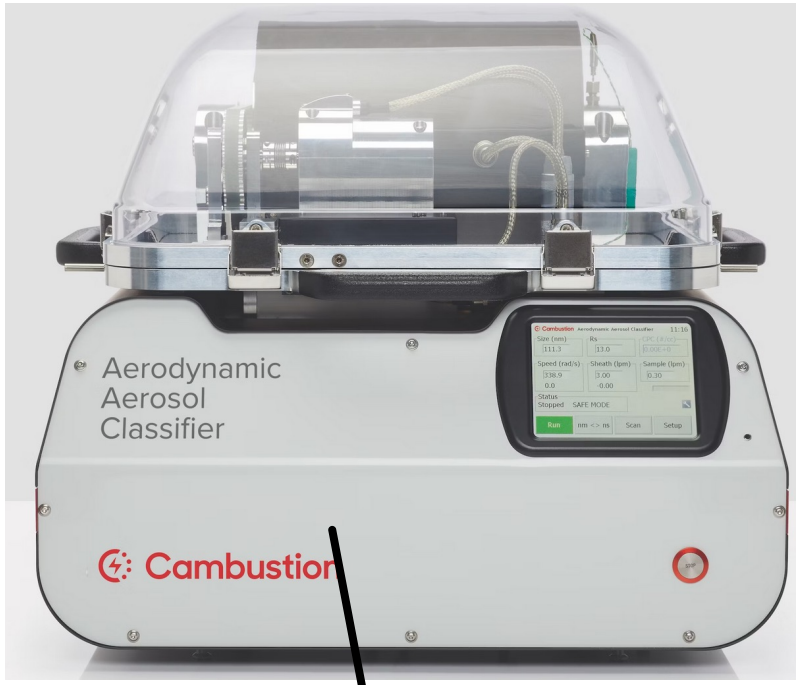
➔**Systematic bias in single scattering albedo** found for a wide range of absorbing aerosols tested in laboratory experiments

This work:

- **Assessing accuracy of multi-sphere T-matrix simulations for black carbon aggregates**
- **Feasibility of using more complex aerosol representation without introducing ambiguity**

Schuster et al.,
Remote Sensing, 2019

Aerosol samples



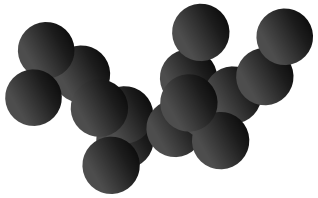
AAC used as size selector to provide truly unimodal aerosol

Laboratory generated aerosol:

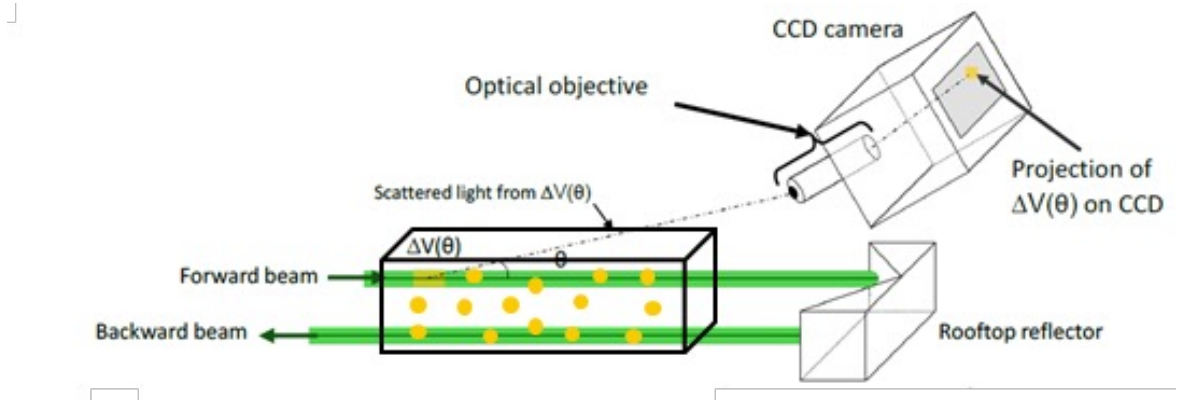
- Polystyrene latex (**PSL**) size standards
  - Spherical
  - Homogeneous
  - Non-absorbing

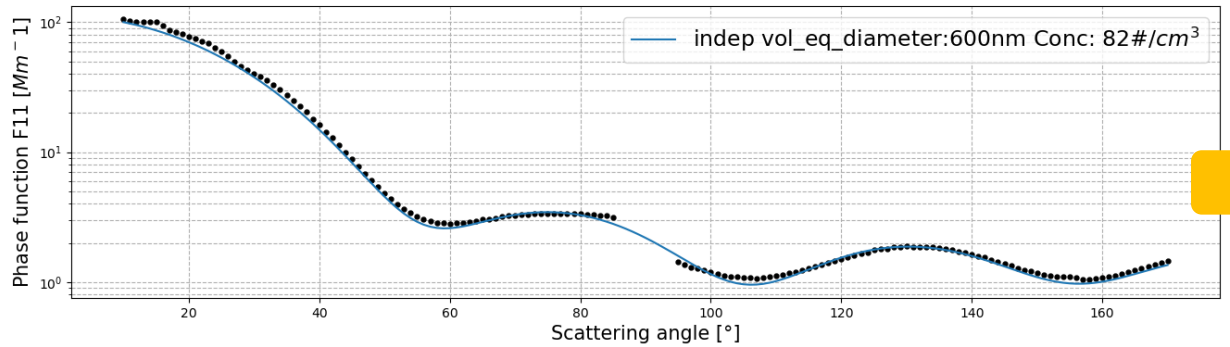
- **Nigrosin**
  - Spherical
  - Homogeneous
  - Light-absorbing

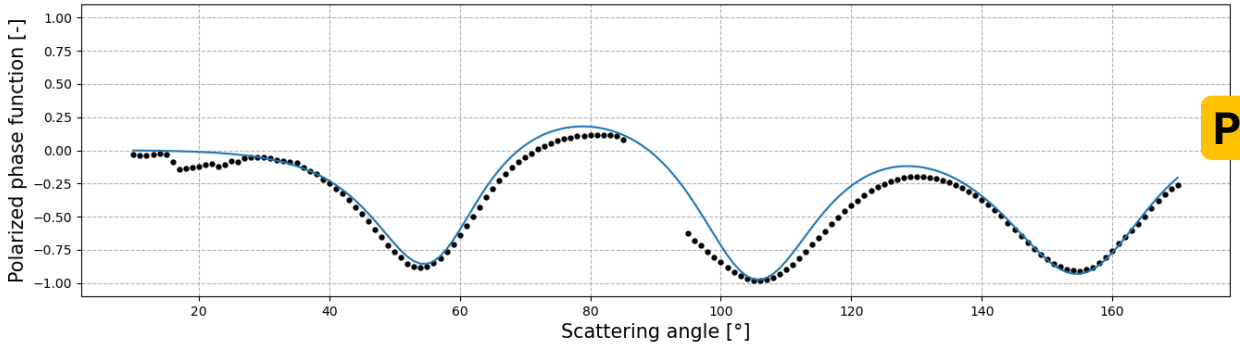
- Pure **black carbon aggregates**
  - Non-spherical – “fractal-like”
  - Homogeneous material properties
  - Strongly light-absorbing



Laser Imaging Polar Nephelometer – «uNeph»







Laser Imaging Polar Nephelometer – "uNeph"

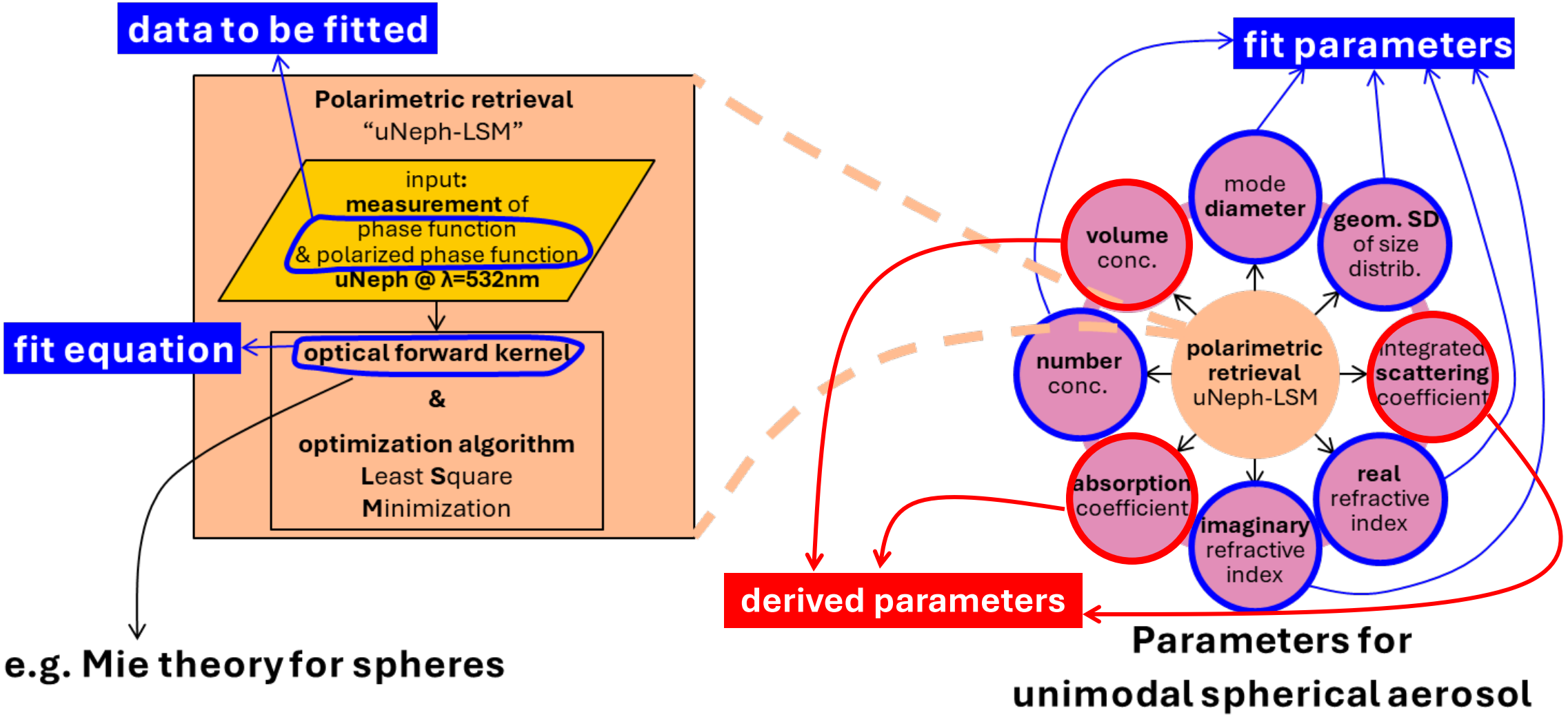
- High angular resolution
- Polarization dependent measurement
- Small truncation angle
- 532 nm wavelength

Phase Function, F_{11}

Polarized Phase Function, $-F_{12}/F_{11}$

Measurement principle: Dolgos et al., Opt. Express, 2014
Our instrument: Moallemi et al., Atmos. Meas. Tech., 2023

Polarimetric retrieval of aerosol properties



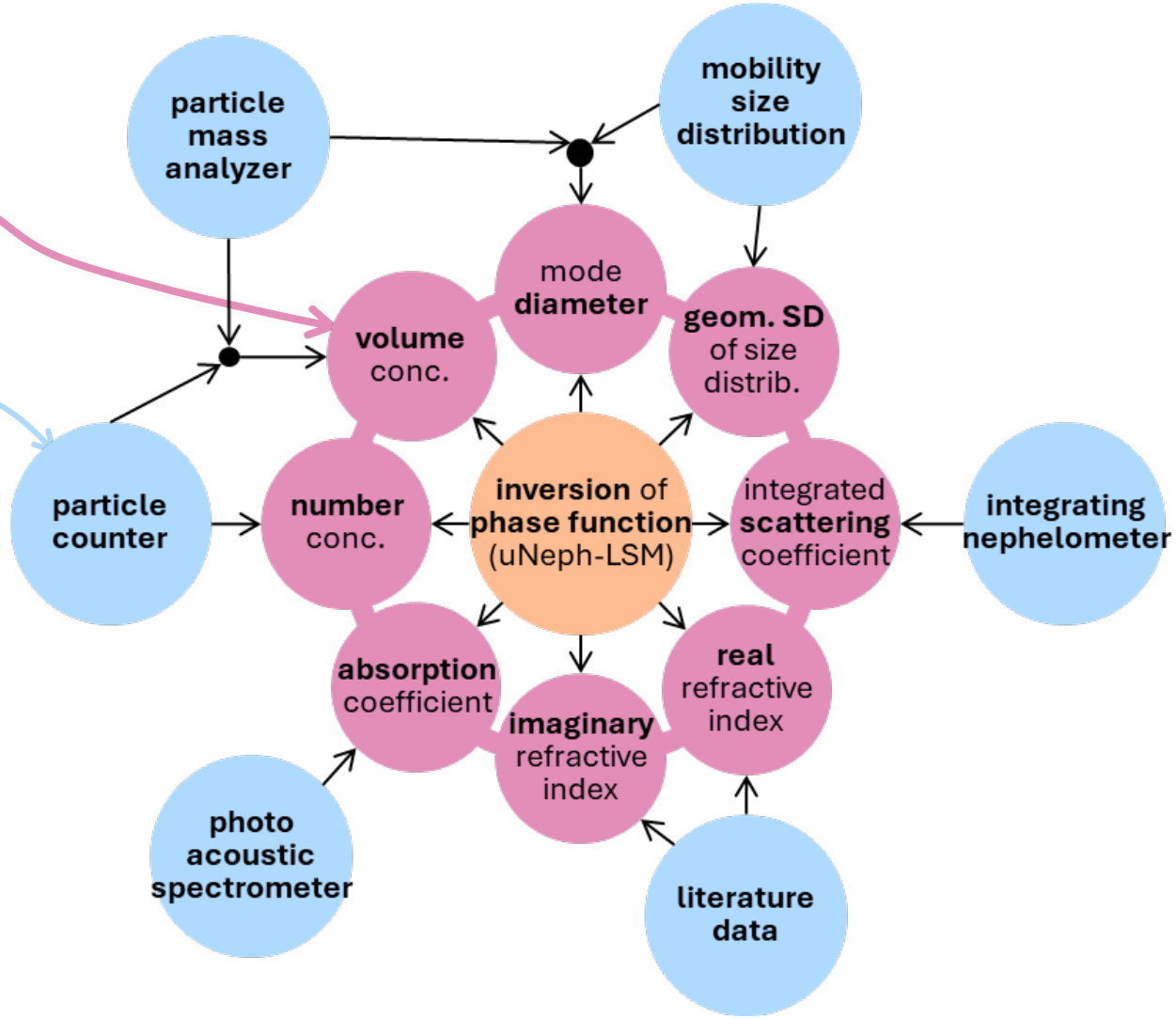
Independent validation of retrieved aerosol properties

PSI

uNeph-LSM retrieval result

VERSUS

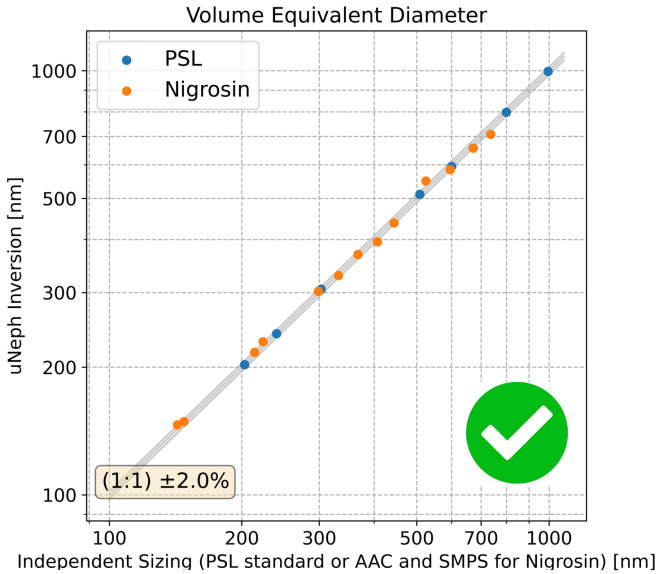
Independent measurements serving validation

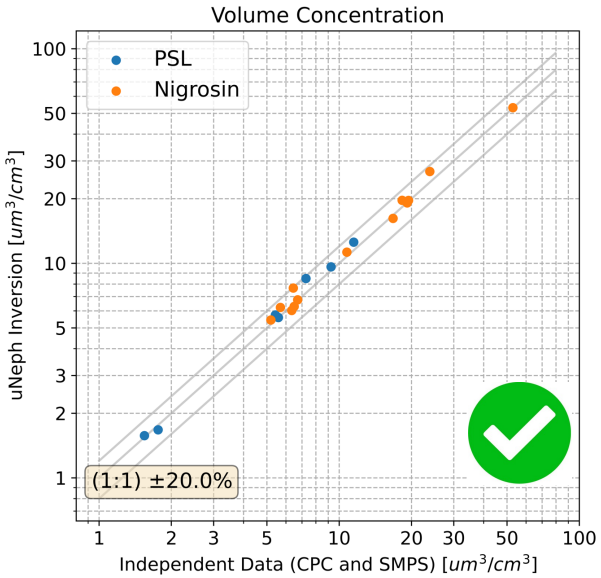


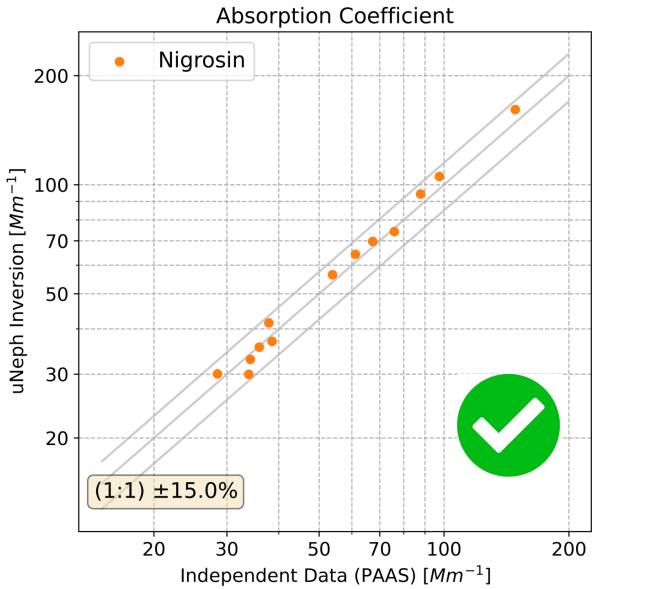
Research questions:

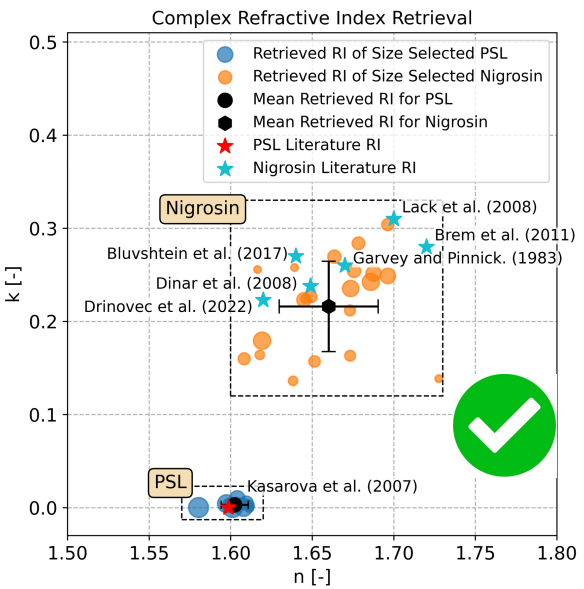
- How accurate is the polarimetric retrieval?
- Specific focus:
  - **Light absorption** coefficient & complex refractive index
  - ***Importance of considering non-spherical shape*** *of black carbon particles*

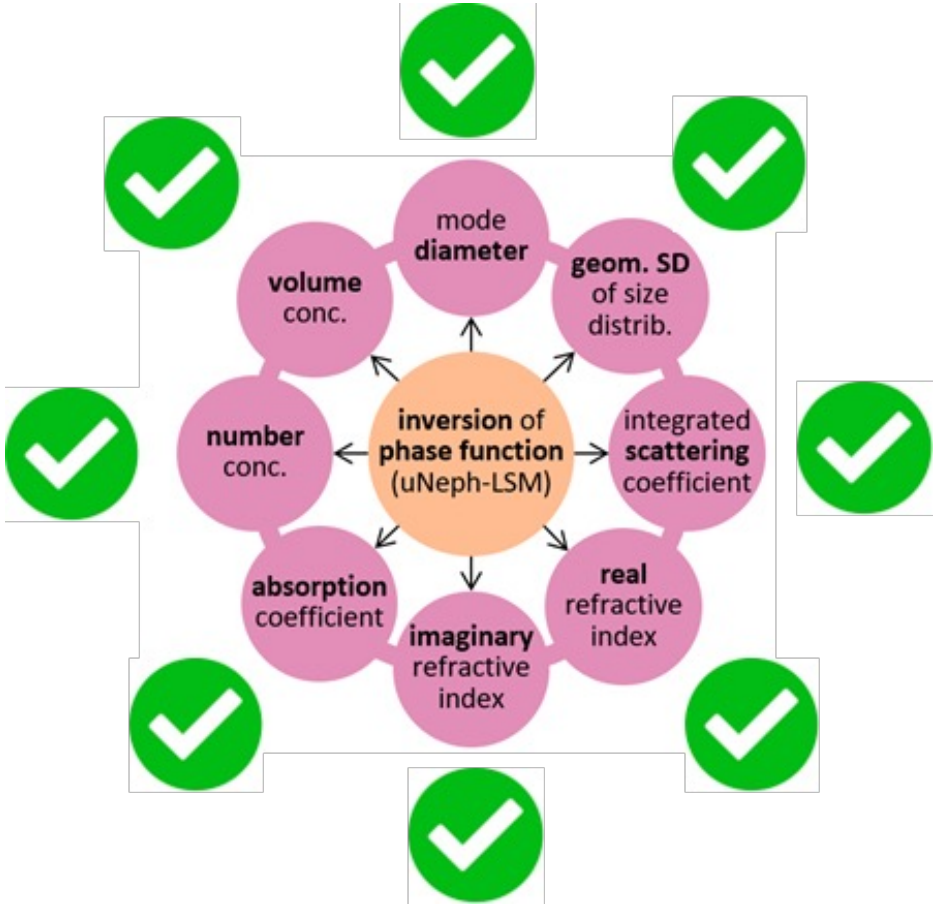
Polarimetric retrieval for spherical particles







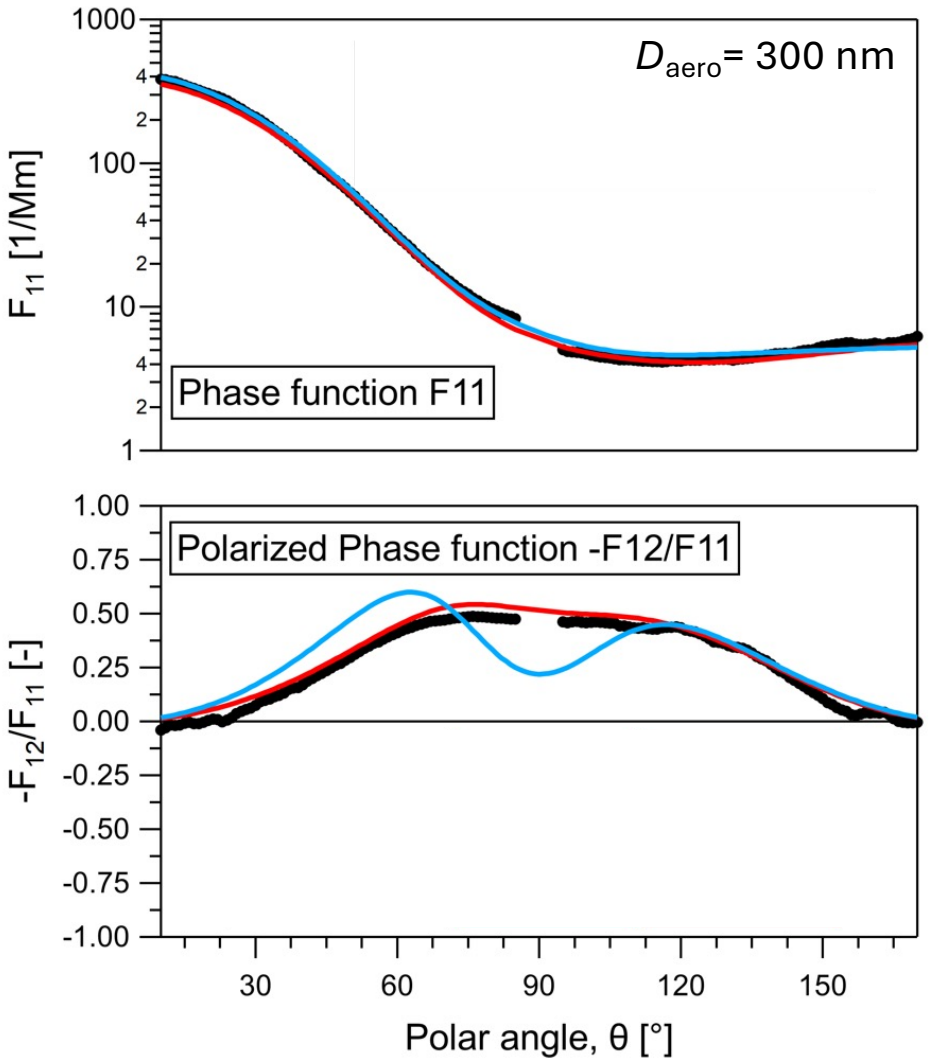




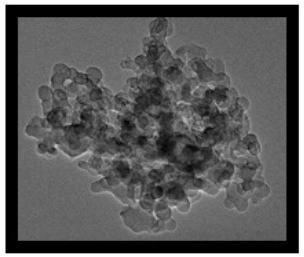
Excellent retrieval performance
including absorption coefficient
for **spherical** unimodal homogeneous aerosol samples

High accuracy of retrieved parameters is essentially consistent with actual information content of phase function data (see Moallemi et al., 2022).

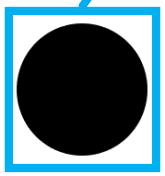
Polarized phase function of aggregates: example measurement & fit



Aerosol sample:
black carbon aggregate



Measurement (uNeph)
Fit: MSTM (aggregates)
Fit: Mie (spherical)



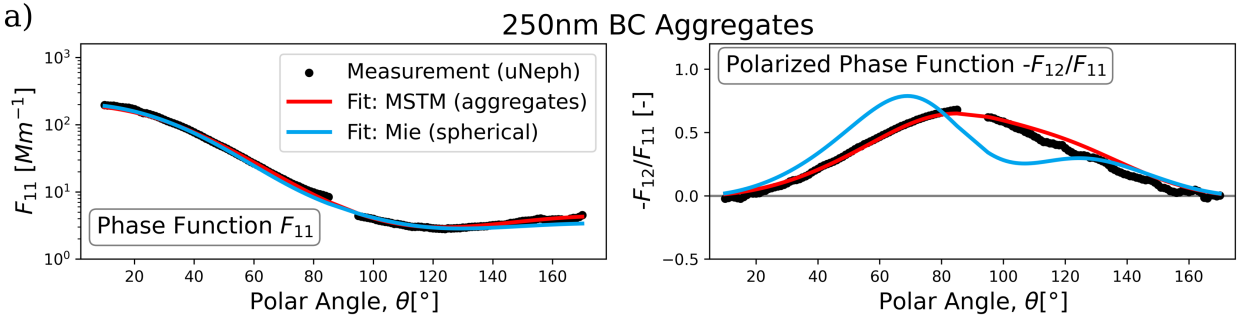
Result:
Mie ➔ poor fit
MSTM ➔ good fit

Laser imaging type polar nephelometer
Moallemi et al., Atmos. Meas. Tech., 2023

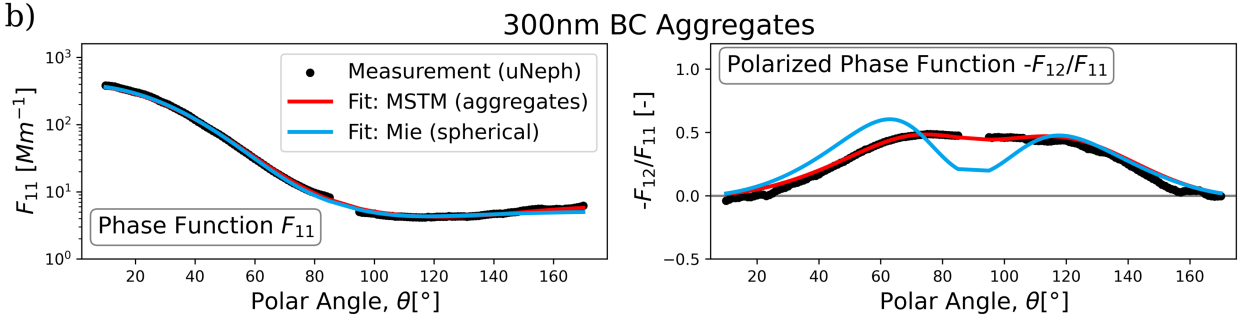
Multi-Sphere-T-Matrix (MSTM) optical kernel applied to aggregates generated using a tuneable diffusion-limited aggregation (DLA) software.
Romshoo et al., 2021
Mackowski and Mishchenko, 2011
Woźniak et al. 2012

Probing 6 different diameters

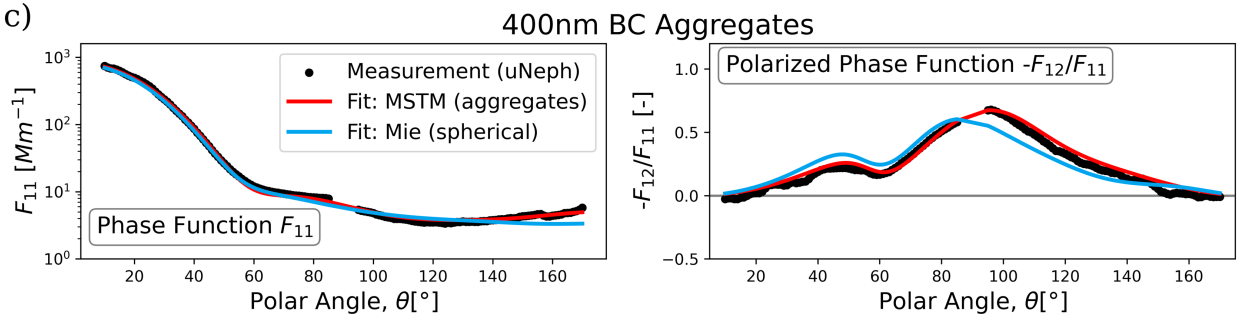
a)



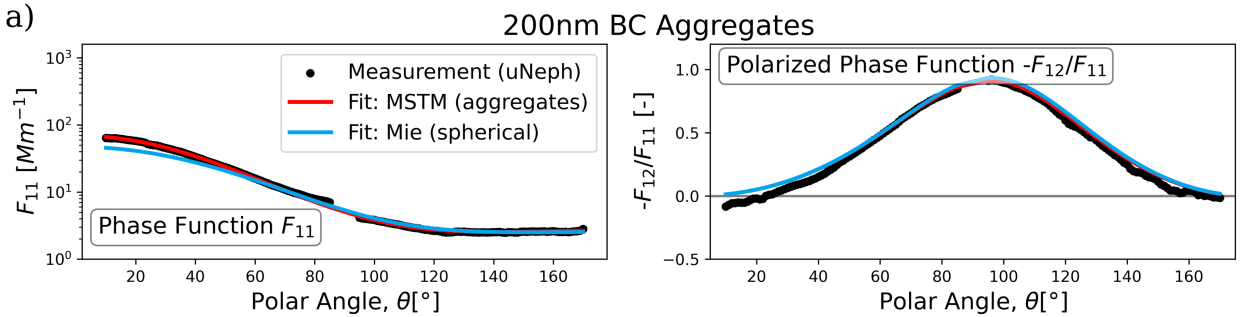
b)



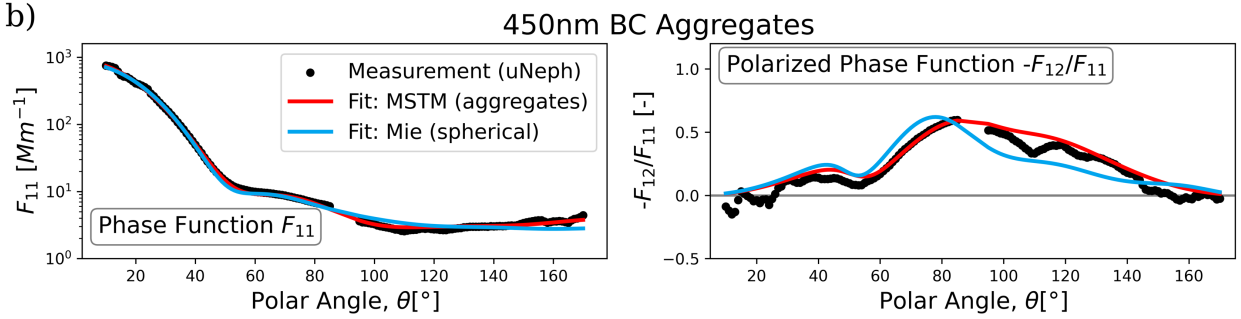
c)



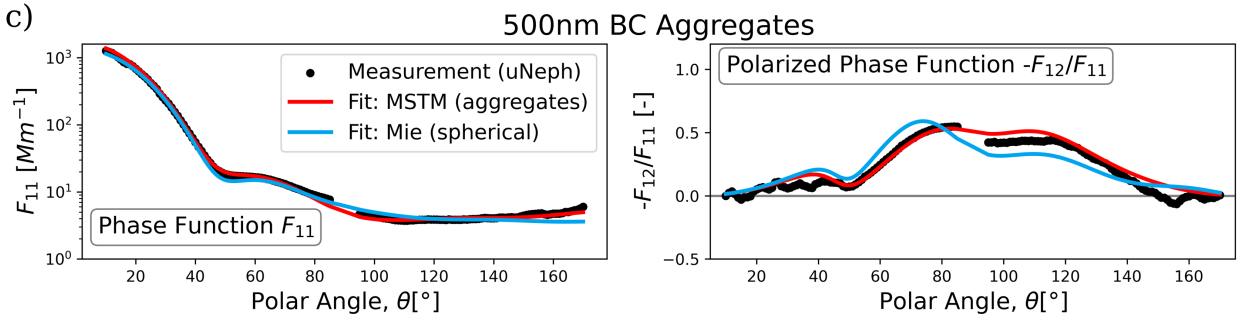
a)



b)



c)

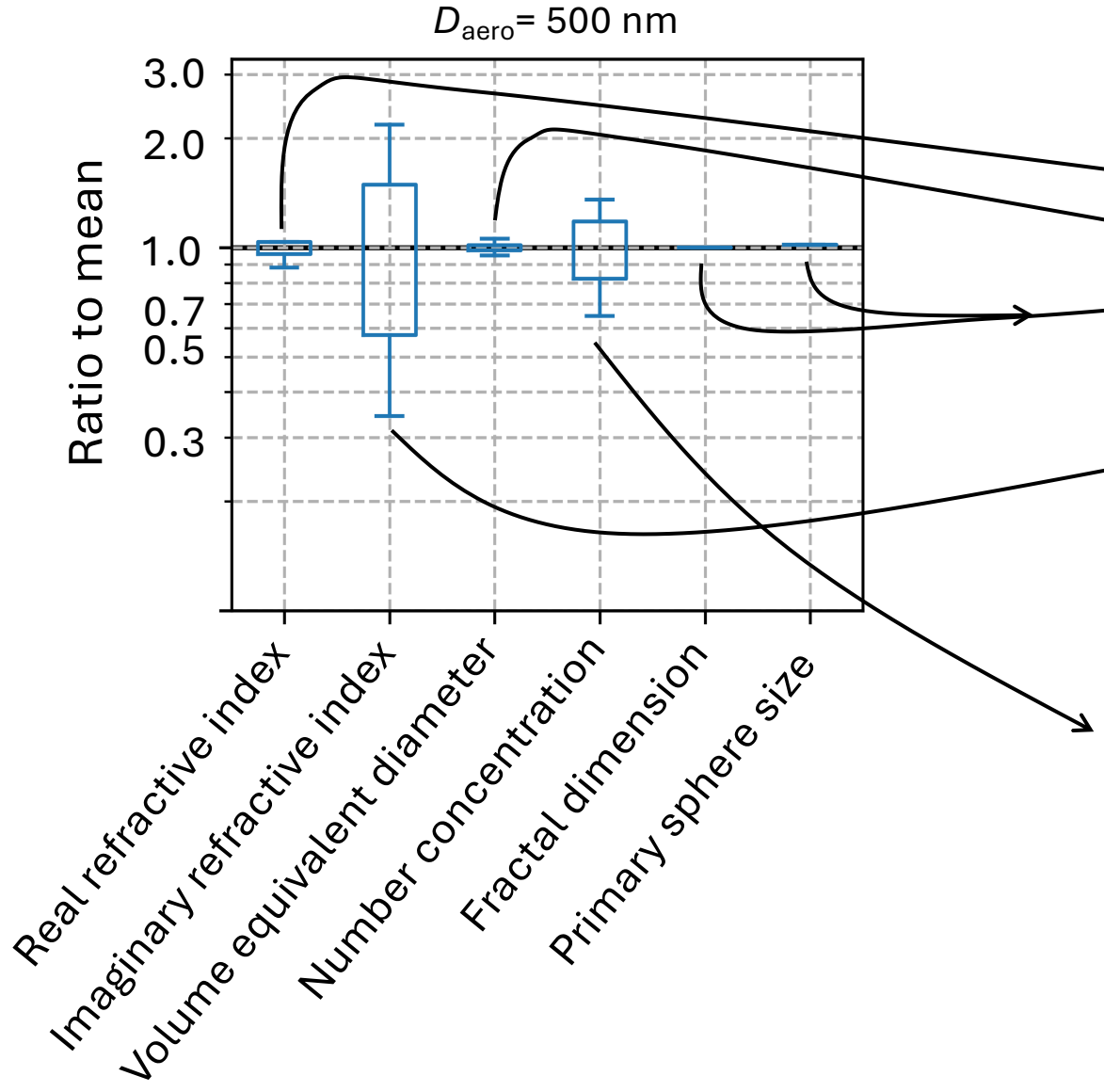


General result: Mie → poor fit MSTM → good fit

Note: Sphere & aggregate are indistinguishable on the basis of F_{11} at a single wavelength

→ Does better fit result in better aerosol property retrieval?

Uncertainty of retrieved parameters («precision»)



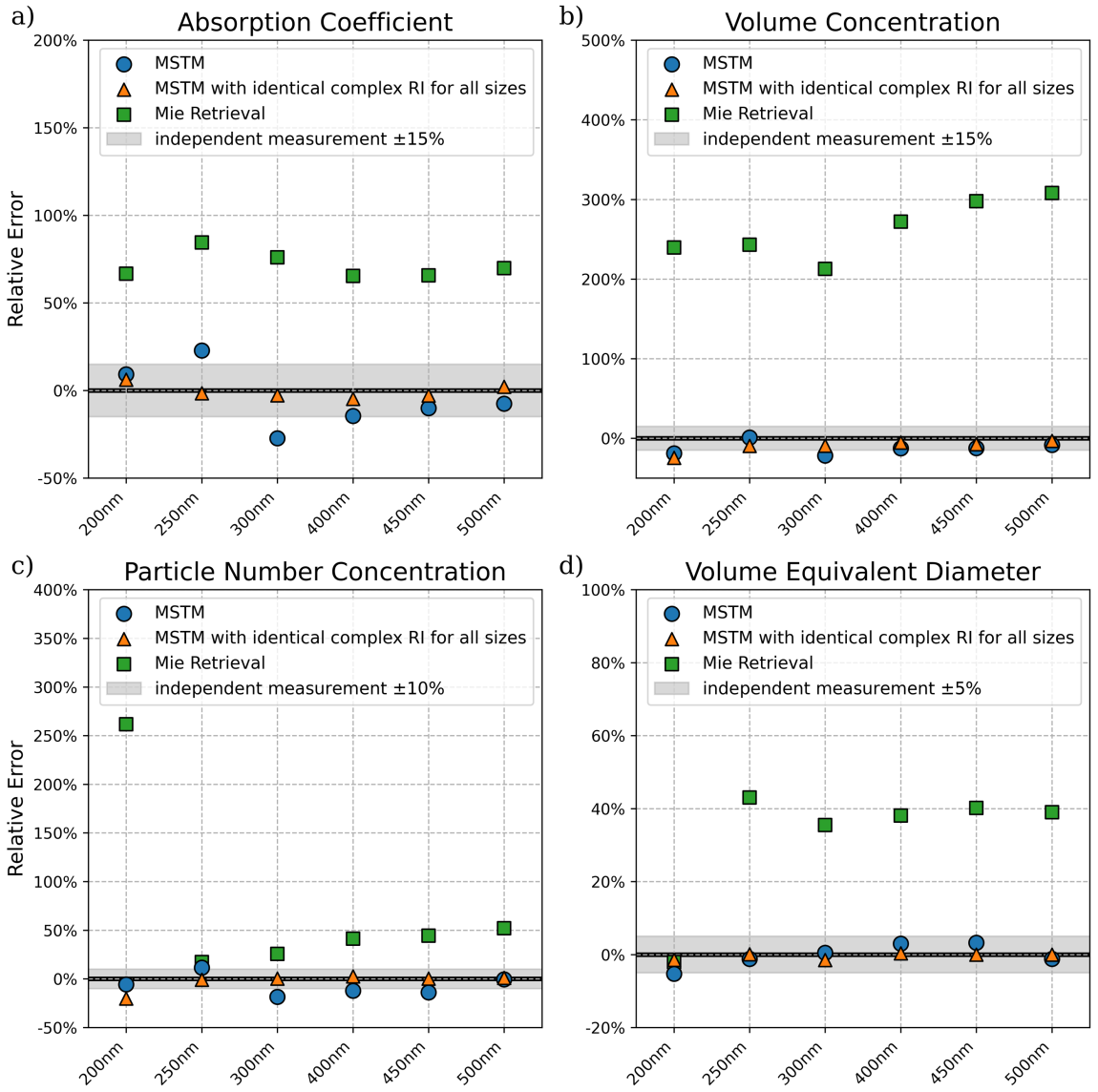
- High precision of retrieved
  - Real refractive index
  - Particle volume
  - Shape parameters
- Only moderate precision for “absorption” (imaginary refractive index), consistent with actual information content of phase function data (see Moallemi et al., 2022)
- Uncertainty of imaginary refractive index causes some uncertainty in retrieved number & volume concentration

Performance of different optical kernels: MSTM versus Mie

MSTM optical kernel → *Some random noise*

MSTM with identical complex RI for all sizes → *Accurate and precise* when combining info from multiple sizes

Mie optical kernel → *Large systematic bias*



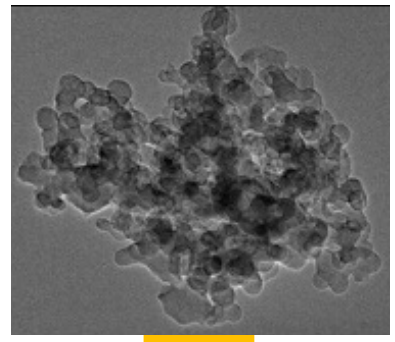
It is possible to keep the shape representation simple

These 5 parameters are also required
for unimodal spherical aerosol

Aerosol properties considered in the MSTM retrieval
1. Number concentration
2. Real refractive index
3. Imaginary refractive index
4. Volume equivalent diameter
5. Variability of particle volume
6. Primary sphere size
7. Fractal dimension

Only 2 parameters needed to describe the shape

➔ Simplified representation of aggregate geometry
is sufficient for achieving accurate aerosol property retrieval
(Warning: retrieved shape parameters are “effective” rather than exact values!)



Simplify:
- Spherical primary particles
- No variability of primary sphere size
- No necking
- …

Conclusions and outlook for polarimetric aerosol retrievals