Light scattering by nonspherical atmospheric particles

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Scattered radiation = Scattering event * Incident radiation

$$\begin{bmatrix} E_{\parallel s} \\ E_{\perp s} \end{bmatrix} \propto \underbrace{\begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix}}_{\text{“Amplitude scattering matrix”}} \begin{bmatrix} E_{\parallel i} \\ E_{\perp i} \end{bmatrix}$$

More practical approach:
Stokes vector and the phase matrix

$$
\begin{aligned}
I &\propto \left\langle E_{\parallel}E_{\parallel}^{*} + E_{\perp}E_{\perp}^{*} \right\rangle \\
Q &\propto \left\langle E_{\parallel}E_{\parallel}^{*} - E_{\perp}E_{\perp}^{*} \right\rangle \\
U &\propto \left\langle E_{\parallel}E_{\perp}^{*} + E_{\perp}E_{\parallel}^{*} \right\rangle \\
V &\propto i\left\langle E_{\parallel}E_{\perp}^{*} - E_{\perp}E_{\parallel}^{*} \right\rangle
\end{aligned}
$$

$$
\begin{bmatrix} I_s \\ Q_s \\ U_s \\ V_s \end{bmatrix} = \frac{C_{sca}}{4\pi r^2} \underbrace{\begin{bmatrix} P_{11} & P_{12} & P_{13} & P_{14} \\ P_{21} & P_{22} & P_{23} & P_{24} \\ P_{31} & P_{32} & P_{33} & P_{34} \\ P_{41} & P_{42} & P_{43} & P_{44} \end{bmatrix}}_{} \begin{bmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{bmatrix}
$$

Phase matrix depends on particle size, shape, composition, orientation, wavelength of light, illumination geometry

Phase matrix is simplified in special cases:

Rayleigh scattering

$$\begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{11} & 0 & 0 \\ 0 & 0 & P_{33} & 0 \\ 0 & 0 & 0 & P_{33} \end{bmatrix}$$

Mie scattering (= spheres)

$$\begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{11} & 0 & 0 \\ 0 & 0 & P_{33} & P_{34} \\ 0 & 0 & P_{-34} & P_{33} \end{bmatrix}$$

$$\begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{22} & 0 & 0 \\ 0 & 0 & P_{33} & P_{34} \\ 0 & 0 & P_{-34} & P_{44} \end{bmatrix}$$

More general case. Randomly oriented nonspherical particles, equal amount of particles and their mirror particles.

If incident light is *unpolarized* (*Q,U,V* = 0):

Intensity of scattered light $I_s \propto P_{11}(\Theta)$

Degree of linear polarization $-\dfrac{P_{12}(\Theta)}{P_{11}(\Theta)}$

Solving light scattering

- Maxwell's equations
- Constitutive relations
- Boundary conditions

- Analytical solution exists for certain special cases:
  - Homogeneous sphere, coated sphere
  - Infinite cylinder

- In a general case lead to a set of equations that cannot be solved analytically → numerical methods and/or approximations must be used.
  - E.g., *T*-matrix method, discrete-dipole approximation, ray tracing

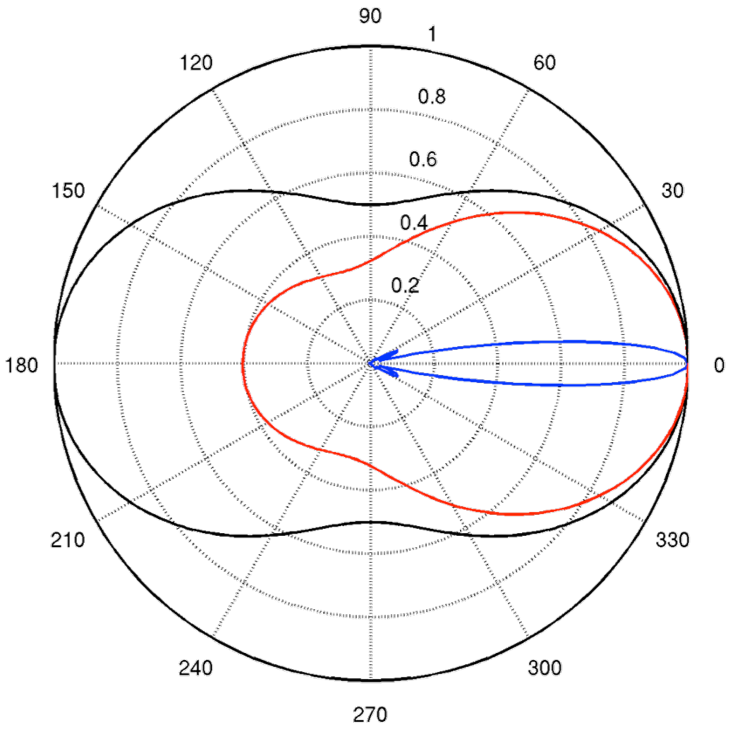
Size parameter *x*

- The relative size of the particle with respect to the wavelength (*a* = radius of the particle):

$$x = \frac{2\pi a}{\lambda} = ka$$

- Nonspherical particles: *a* = radius of a volume-eqv. sphere or an area-eqv. sphere

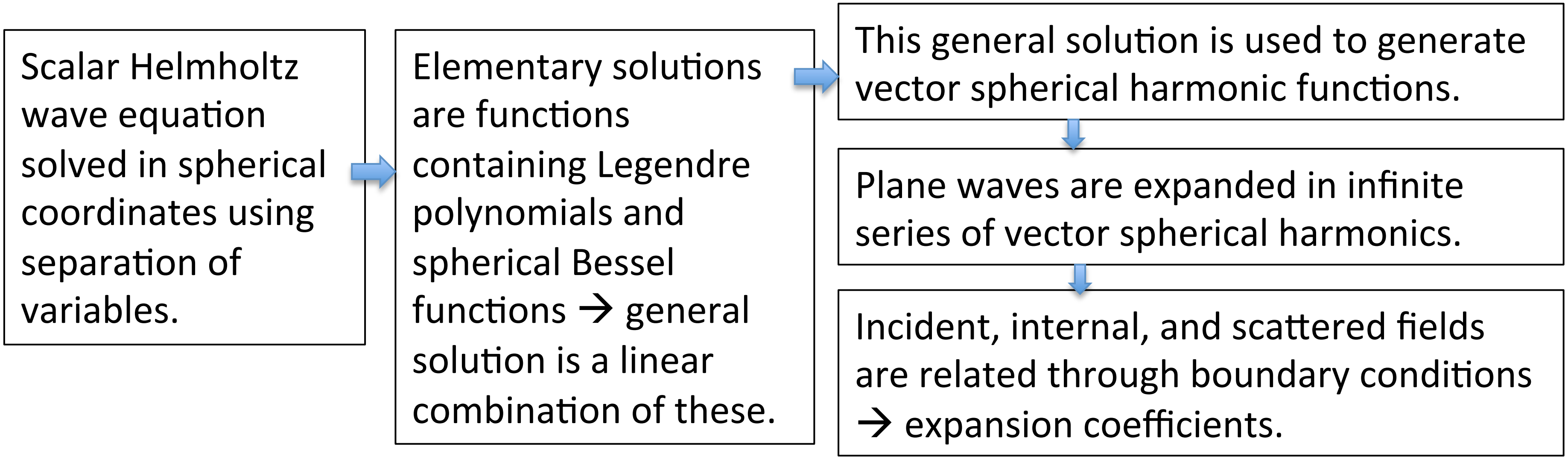
- Three different regions based on *x*:
  - Rayleigh domain ($x \ll 1$, $|m|x \ll 1$)
  - Resonance domain ($x \approx 1 \ldots 30$)
  - Ray optics domain ($x \gg 1$)



The angular dependence of scattering by spherical water droplets of varying size in visible λ:
$x = 0.01$ $x = 1$ $x = 10$

Mie solution

- The Mie solution refers to light scattering by *spherical particles.*
  - Mie theory, Lorenz-Mie theory, Lorenz-Mie-Debye theory,…
- Exact solution; valid for all size parameters.
  - In practice, the computation slows down at large x.



Scalar Helmholtz wave equation solved in spherical coordinates using separation of variables.

Elementary solutions are functions containing Legendre polynomials and spherical Bessel functions → general solution is a linear combination of these.

This general solution is used to generate vector spherical harmonic functions.

Plane waves are expanded in infinite series of vector spherical harmonics.

Incident, internal, and scattered fields are related through boundary conditions → expansion coefficients.

Mie solution; the result

$$S_1 = \sum_n \frac{2n+1}{n(n+1)}(a_n\pi_n + b_n\tau_n)$$

$$S_2 = \sum_n \frac{2n+1}{n(n+1)}(a_n\tau_n + b_n\pi_n)$$

$$\Rightarrow \quad P_{11} = \frac{1}{2}\left(|S_1|^2 + |S_2|^2\right)$$

Expansion coefficients of the scattered field:

$$a_n = \frac{m\psi_n(mx)\psi_n'(x) - \psi_n(x)\psi_n'(mx)}{m\psi_n(mx)\xi_n'(x) - \xi_n(x)\psi_n'(mx)}$$

$$b_n = \frac{\psi_n(mx)\psi_n'(x) - m\psi_n(x)\psi_n'(mx)}{\psi_n(mx)\xi_n'(x) - m\xi_n(x)\psi_n'(mx)}$$

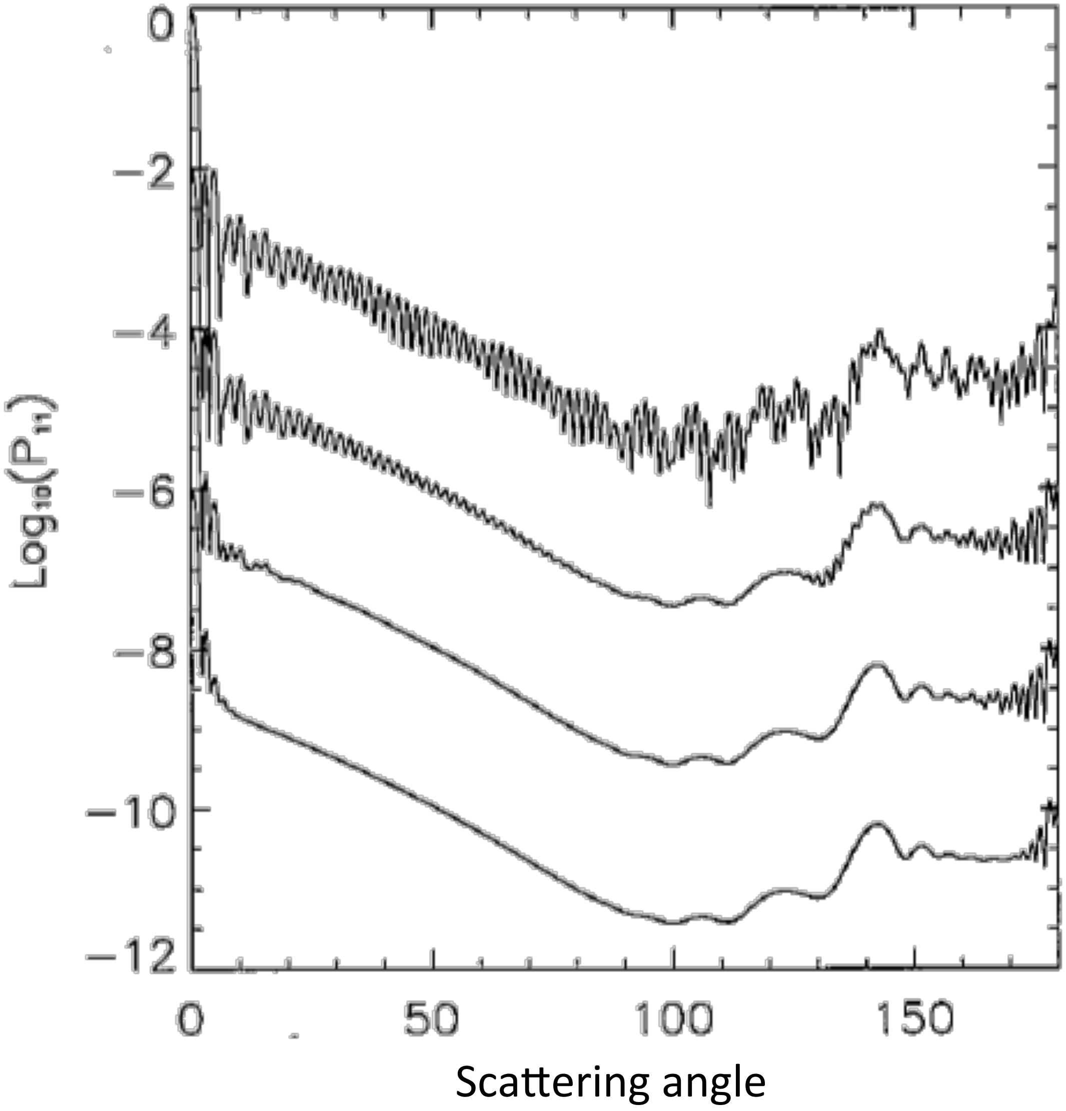
The series expansions converge at large *n* and are, in practice, truncated at some *n* ≈ *x* … 2*x*.

Here *ψ*, *ξ* are Riccati-Bessel functions and *π*, *τ* are functions of Legendre polynomials, *x* is the size parameter and *m* the complex refractive index.

Mie codes and their input / output

- Publicly available Mie codes:
  - http://www.giss.nasa.gov/staff/mmishchenko/t_matrix.html (f77)
  - http://www.scattport.org/index.php/light-scattering-software/mie-type-codes (Fortran, Python, Matlab, C++, …)
  - Later on this course, we'll use a Mie code provided by Chris.
- Typical inputs: wavelength, radius of the sphere, complex refractive index
- Example outputs: phase matrix elements, ϖ, g, coefficients of the scattered field (*a*,*b*)

Example Mie results



Top: Phase function for a spherical water particle (x = 100).

Others: Phase functions integrated over a narrow size distribution of spheres (top to bottom: relative σ = 0.01, 0.03 and 0.1). The curves have been shifted downwards by 2, 4, and 6 for illustrational purposes.

Assumption of spherical particles

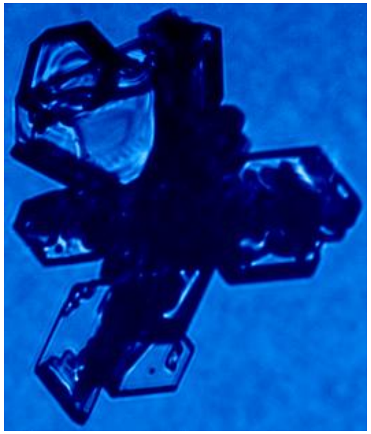
- Excellent for: cloud droplets, small liquid-phase aerosol particles (e.g., sulphuric acid), or other spherical or nearly-spherical particles.

- Too often used for all kinds of particles without thorough testing.
  - Exact solution but to a wrong problem!

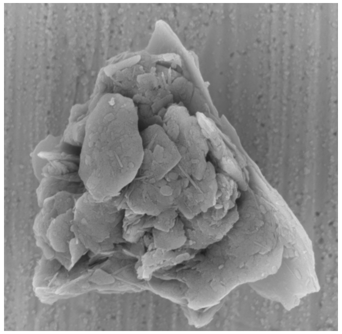
- Known to produce considerable errors.
  - Sometimes, one error may partly cancel another.

Nonspherical particles in the atmosphere

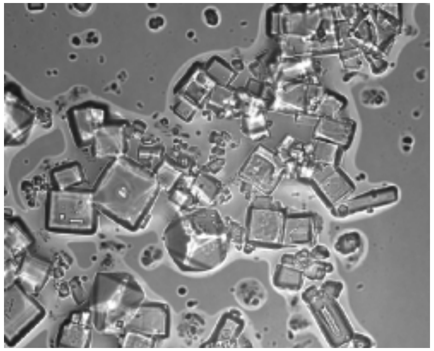
Ice crystal



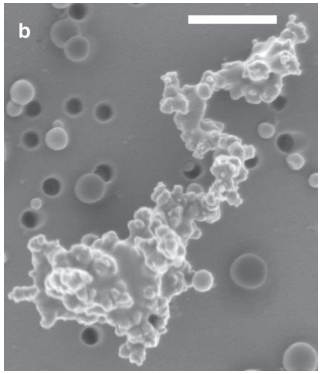
Mineral dust



Sea salt



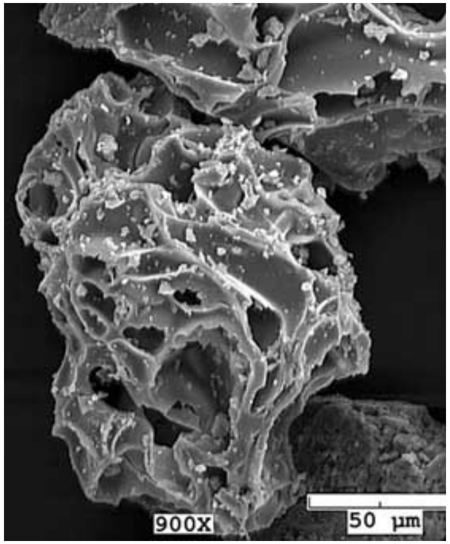
Soot



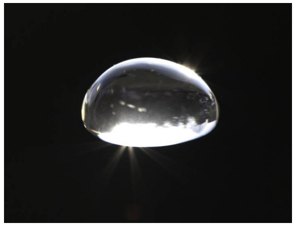
Snowflake

Graupel

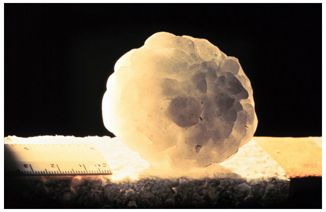
Volcanic ash



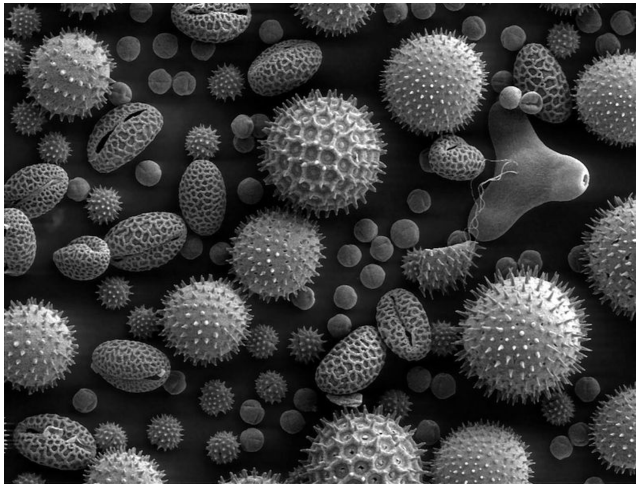
Rain drop

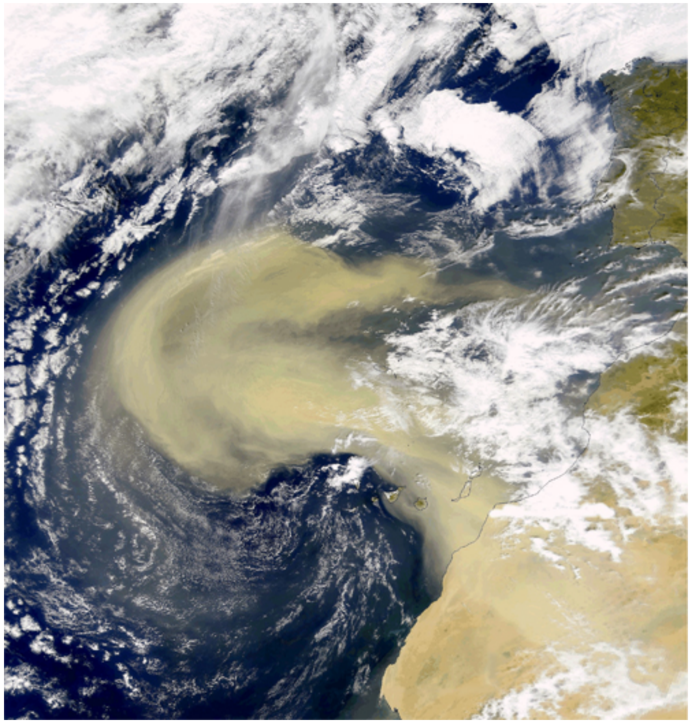


Hail



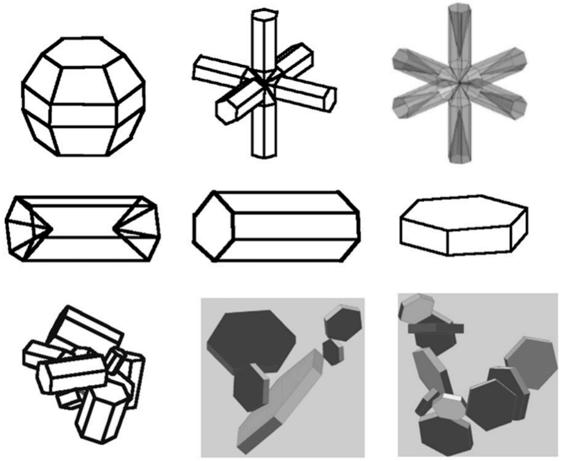
Pollen





Does the shape really matter?

Example: Surface roughness of ice crystals



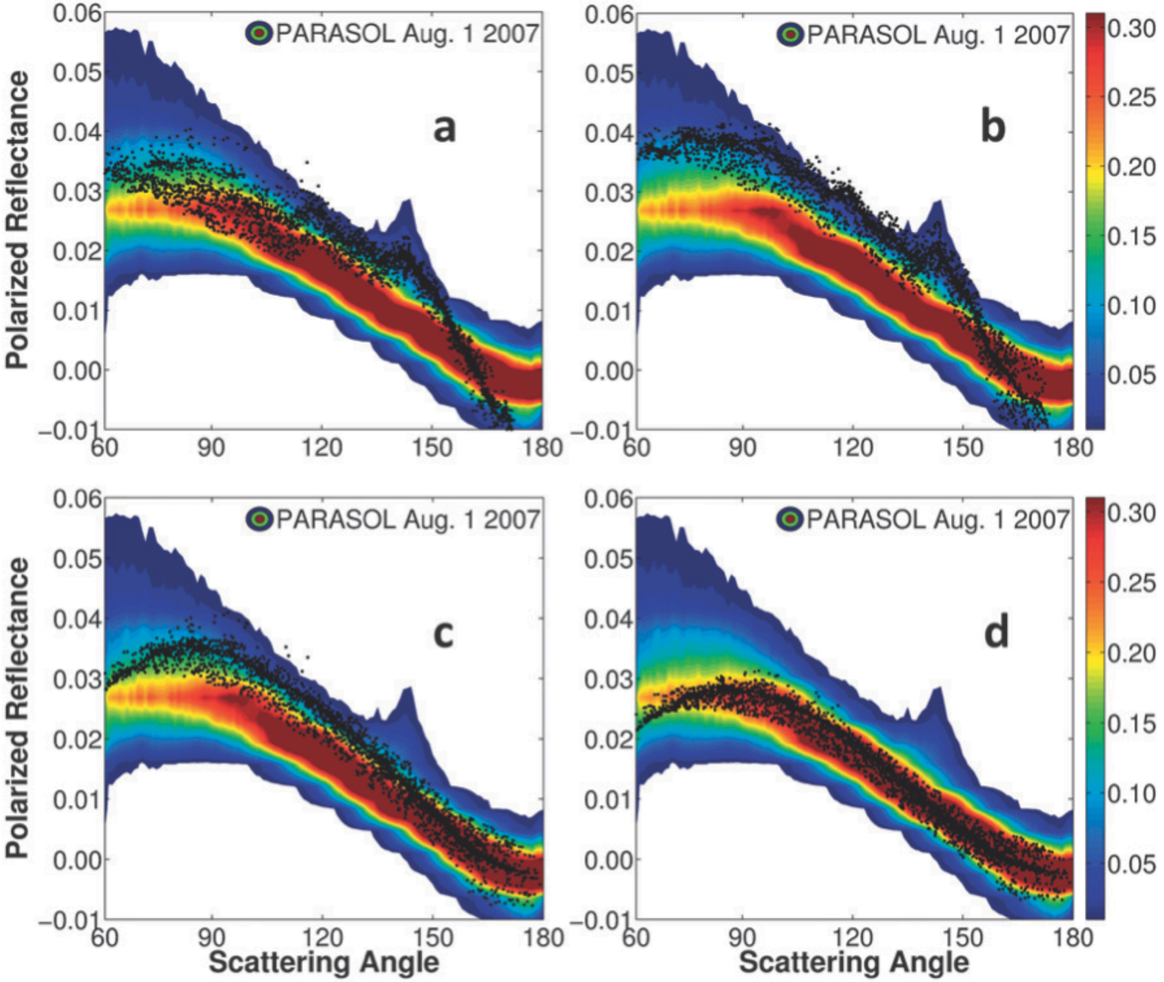
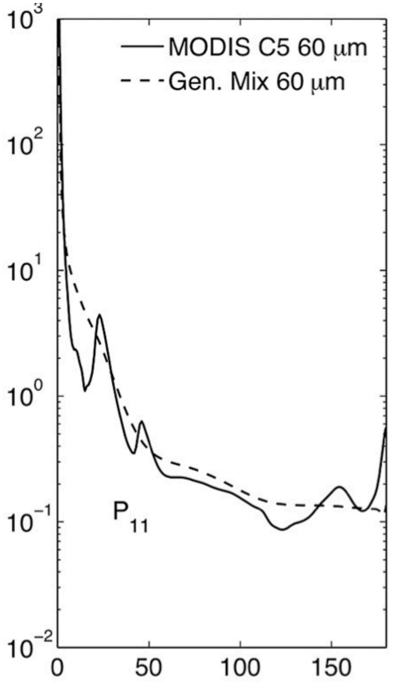
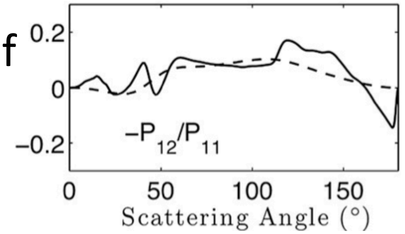


FIG. 3. Simulated polarized reflectance calculated at an effective diameter of 60 μm with an optically thick ($\tau = 5$) ice cloud for (a) the MODIS collection-5 model, (b) a general habit mix with smooth ice particles, (c) a general habit mix with moderately rough ice particles, and (d) a general habit mix with severely rough ice particles. Color contours show the density of the *PARASOL* polarized reflectance data from 1 Aug 2007, and black dots are simulations.

Phase function

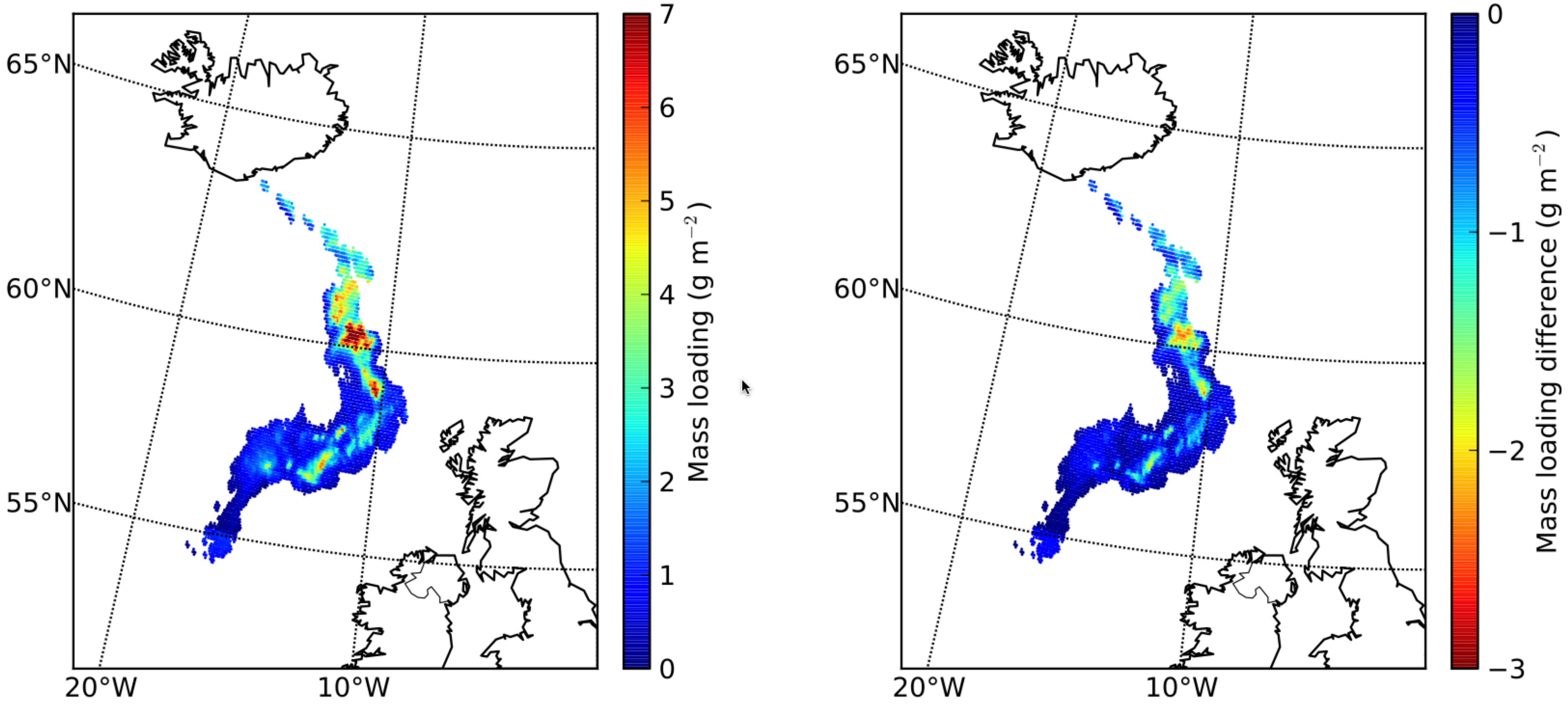


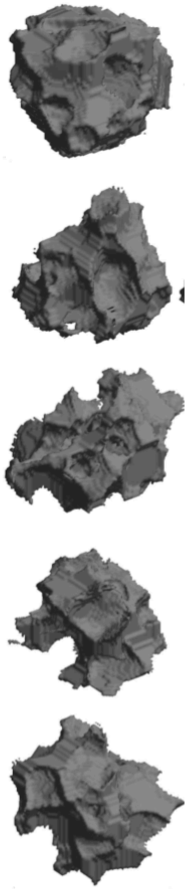
Degree of lin. pol.



[Cole et al., J. Appl. Meteor. Climatol., 2013]

Example: Shape of volcanic ash particles



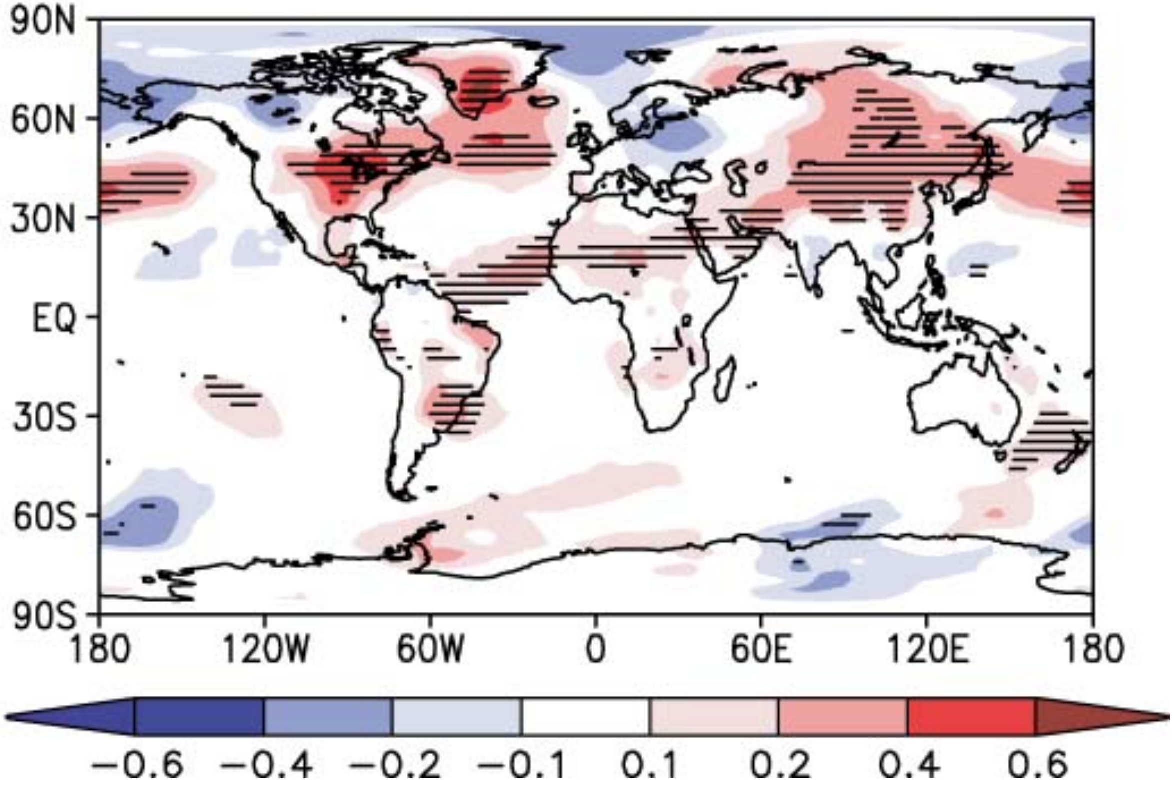


Left: Example SEVIRI ash mass loading retrieval of the volcanic ash plume of 2010 Eyjafjallajokull eruption in Iceland, assuming porous, nonspherical particles.

Right: The difference in ash mass loading between retrievals using mass-equivalent spheres and porous, nonspherical ash particles.

[Kylling et al., 2014, AMT, revised]

Example: dust shape in climate modeling



Difference between spheroidal and spherical dust particles in simulated average 2 m air temperature in ECHAM5.5-HAM2 global aerosol-climate model, 80 years. Global mean temperature was only 0.04 K higher when assuming spheroidal dust particles. [Raisanen et al., 2012, QJRMS]

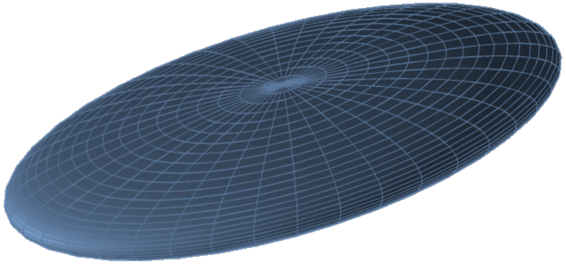
How to deal with nonspherical particles

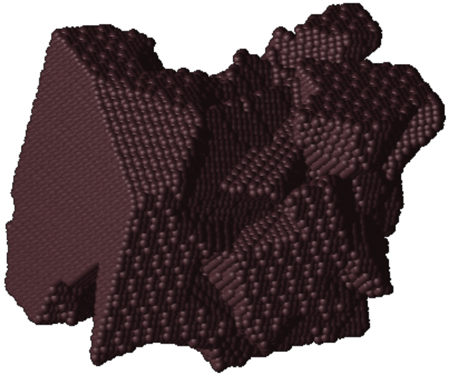
- Necessary: model for the shape + a suitable scattering method.

- Compute scattering properties using publicly available codes.
  - In the resonance domain ($x \approx 1 \dots 30$): *T*-matrix method, discrete-dipole approximation, …
  - In the ray optics domain ($x >> 1$): ray tracing + diffraction

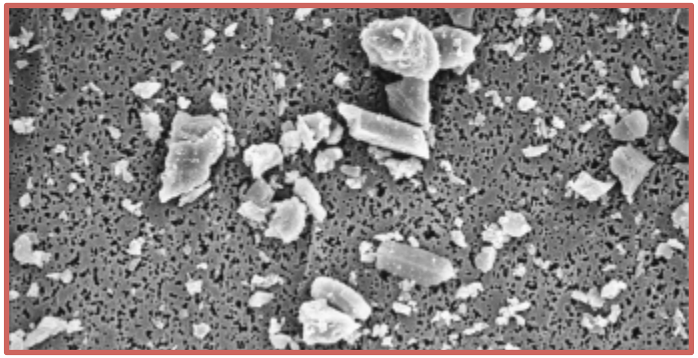
- Utilize existing, pre-calculated databases of scattering properties.

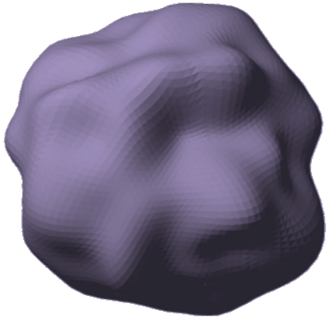
- At specific wavelengths, also measured scattering properties (of aerosols) are available.

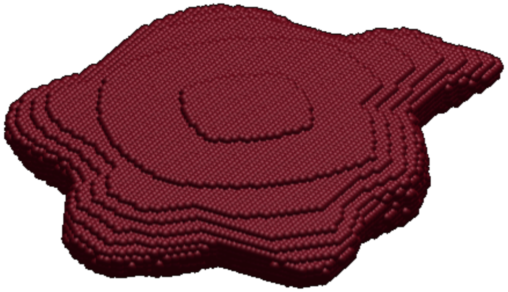


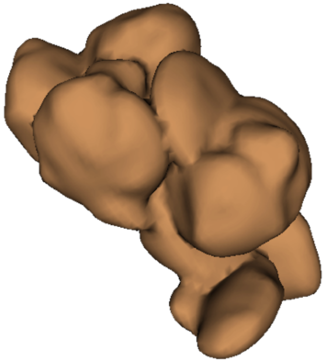


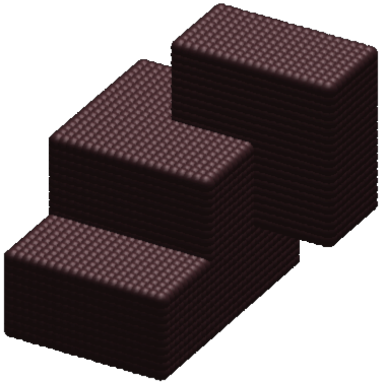
Examples of mathematical shape models suggested for mineral dust particles

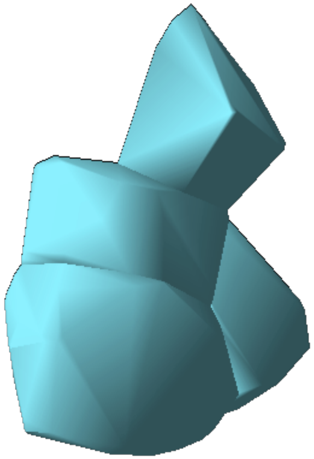












T-matrix method (TMM)

- Extension of Mie theory to particles without spherical symmetry.
  - Electric fields are expanded in vector spherical harmonic functions.
  - The *T*-matrix transforms the incident field expansion coefficients to the scattered field expansion coefficients through surface integrals accounting for boundary conditions.

- Shapes: spheroid, finite (circular) cylinder, other rotationally symmetric particles, also general star-like shapes.

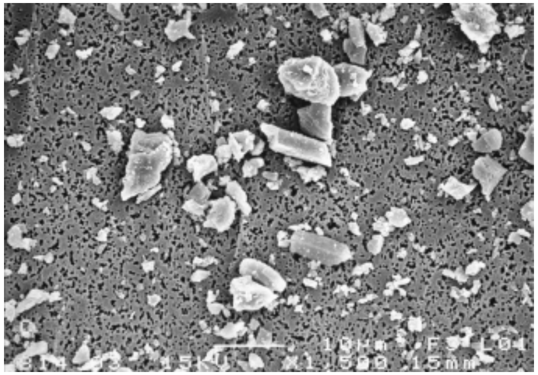
+ Analytical orientation averaging

+ Fast if particle symmetries are used

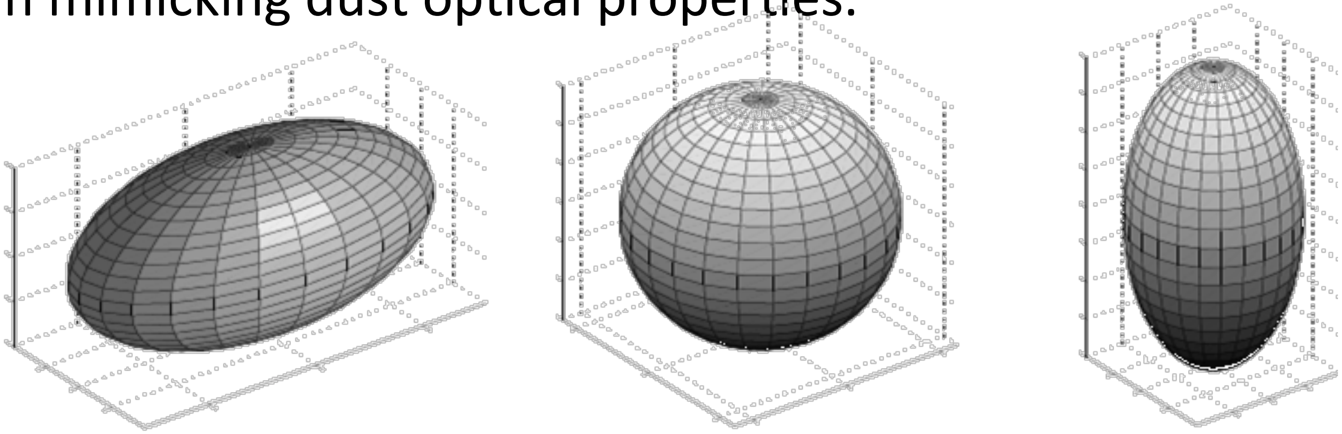
\- Homogeneous composition only (though could be generalized to layered structures)

\- Limitations in *x* and aspect ratio

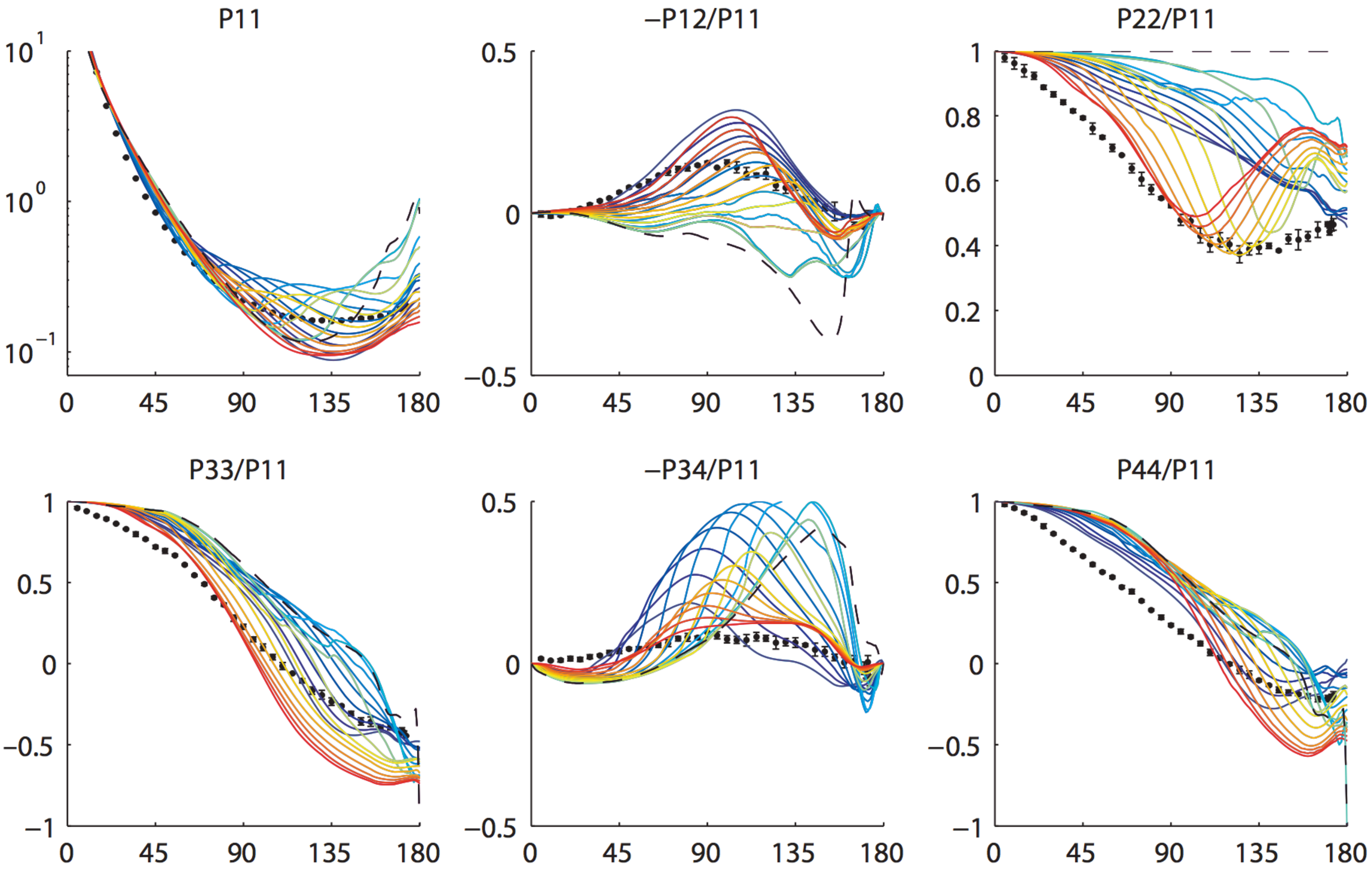
Assumption of spheroidal particles



- One of the simplest nonspherical shapes.
- There are two types of spheroids:
  - Oblate (two long semiaxes and one short)
  - Prolate (one long semiaxis and two short)
- Recently, spheroids have replaced spheres as model shapes for dust in many applications.
- Despite their simplicity, they have proven very successful in mimicking dust optical properties.



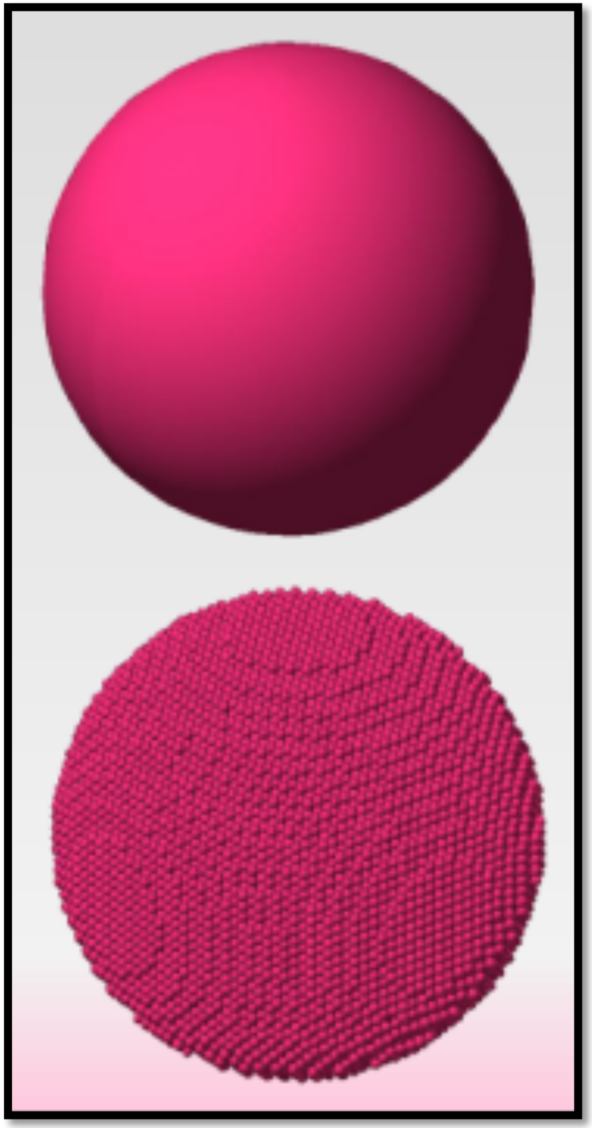
Example: Scattering by different spheroids



Colored lines correspond to different spheroidal aspect ratios, and dashed line is for a sphere (m = 1.55+0.001i). Black dots with error bars are measurements for loess sample at 632.8 nm.

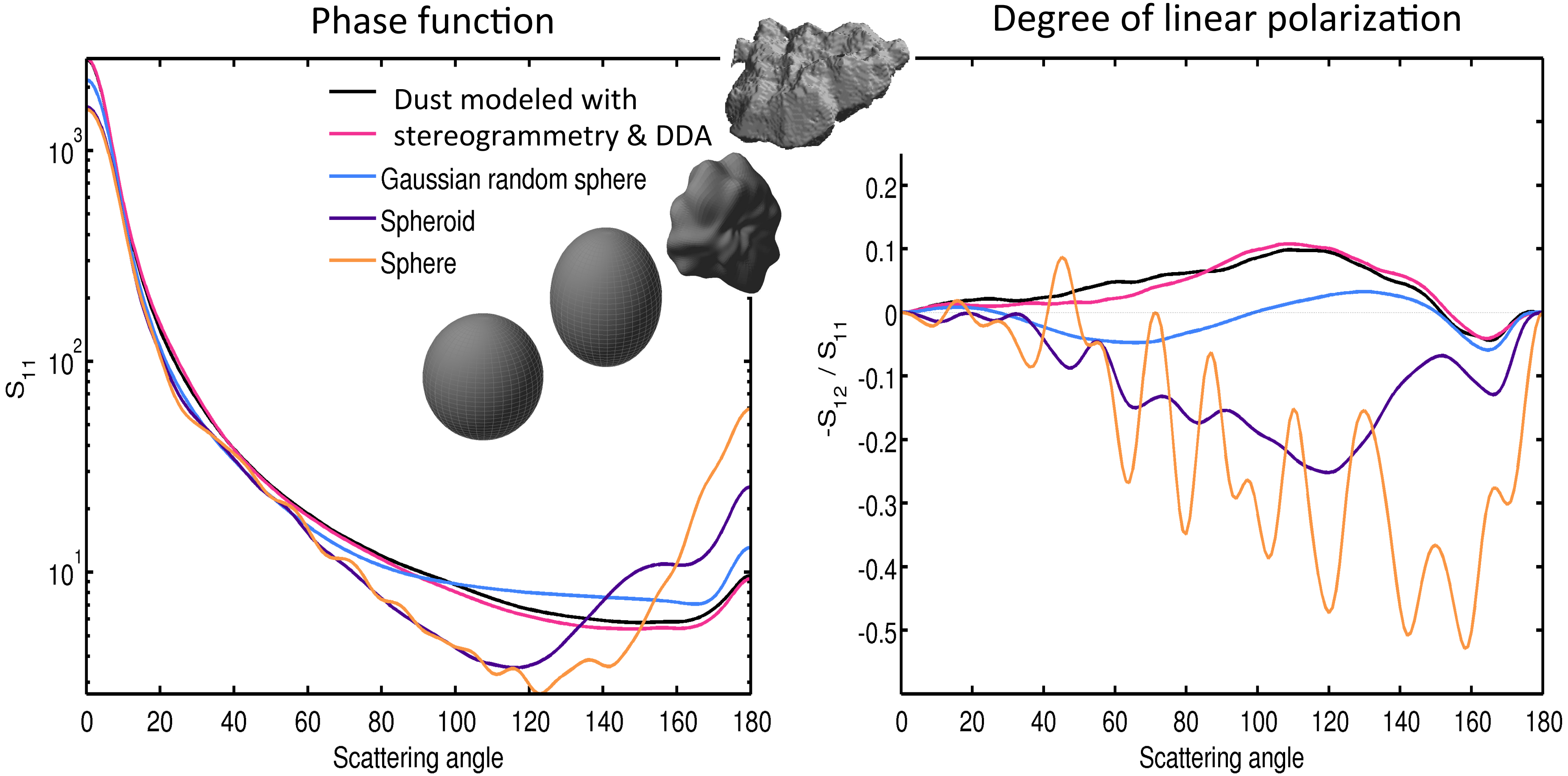
Discrete-dipole approximation (DDA)

- Particle is discretized into small volume elements, "dipoles".
- Electromagnetic field inside the particle is obtained by integrating over the fields induced in the dipoles.
- Publicly available codes: ADDA and DDSCAT.



+ Arbitrary shapes (shape model!)
+ Inhomogeneous composition
- Computations can be very time-consuming
- Computations can require a lot of memory
- In practice, limitations in *x*

Example: Model comparison for a single dust particle at 550 nm



Phase function

Degree of linear polarization

Example: Comparison of ice particle shapes in the microwave

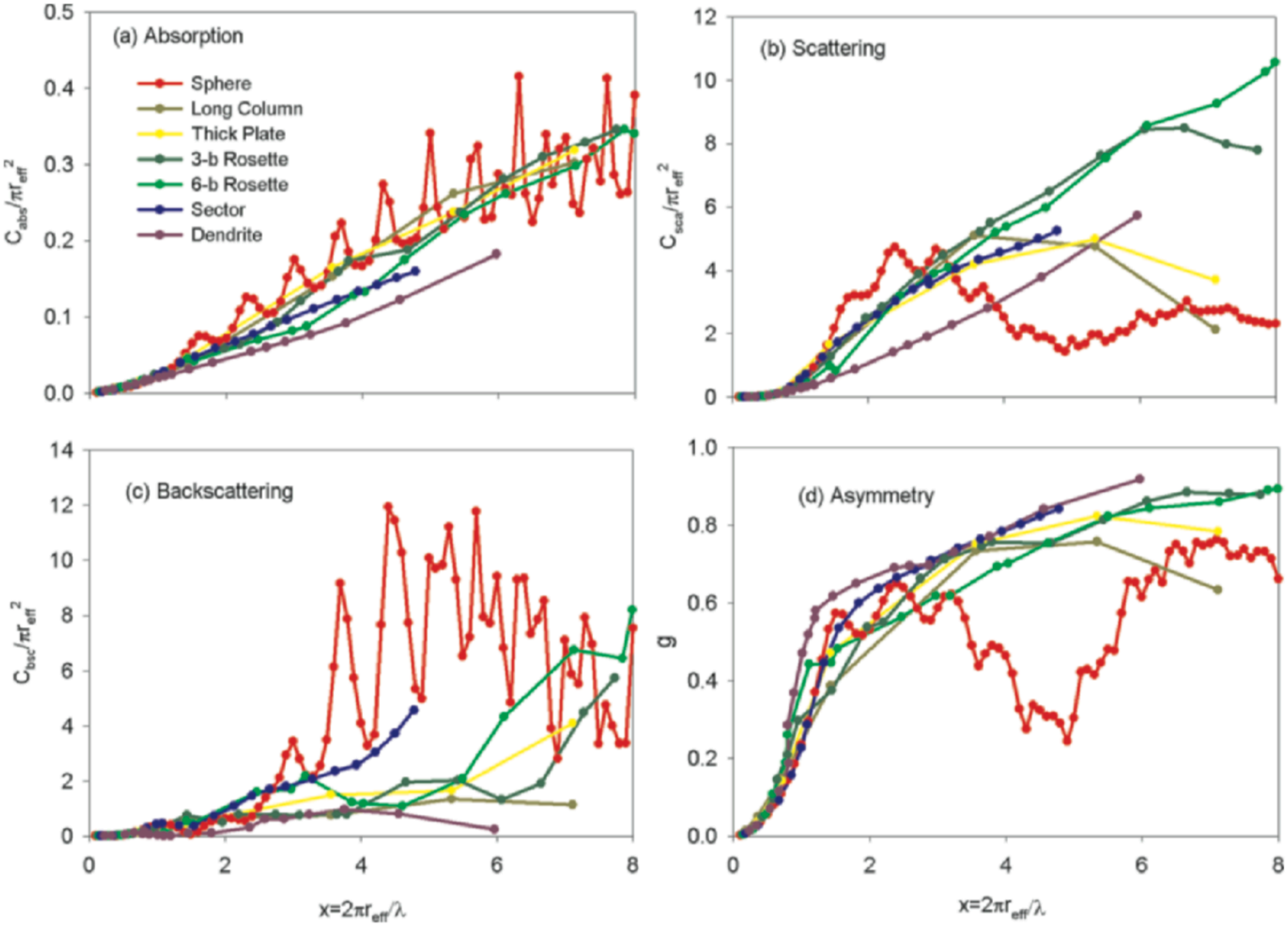
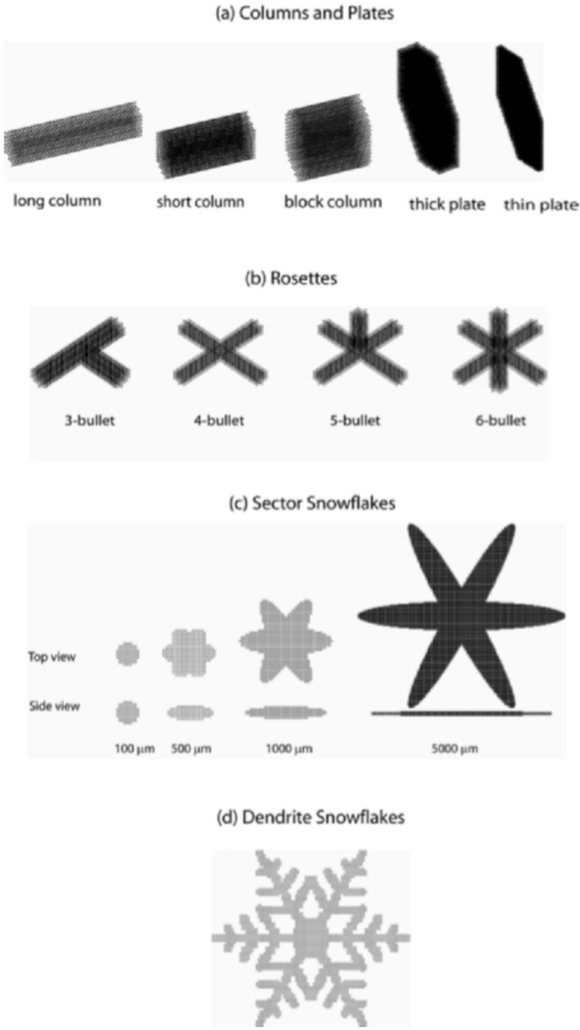


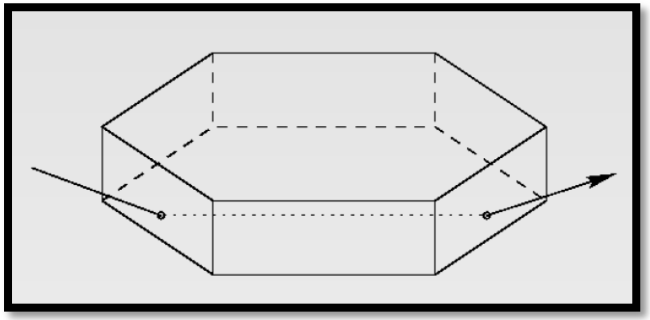
Fig. 2. Normalized (a) absorption, (b) scattering and (c) backscattering cross sections, and (d) asymmetry parameters at 340 GHz and −10°C for selected particles. Parameters of equal-mass spheres are also shown.



[Liu, 2008, BAMS]

Ray tracing

- Widely used method for solving scattering by particles large compared to wavelength.
- Radiation is treated as rays. Interaction at surfaces with the law of specular reflection, Snell's law, and the Fresnel equations.
- For a full solution, diffraction in the forward direction also needs to be considered.

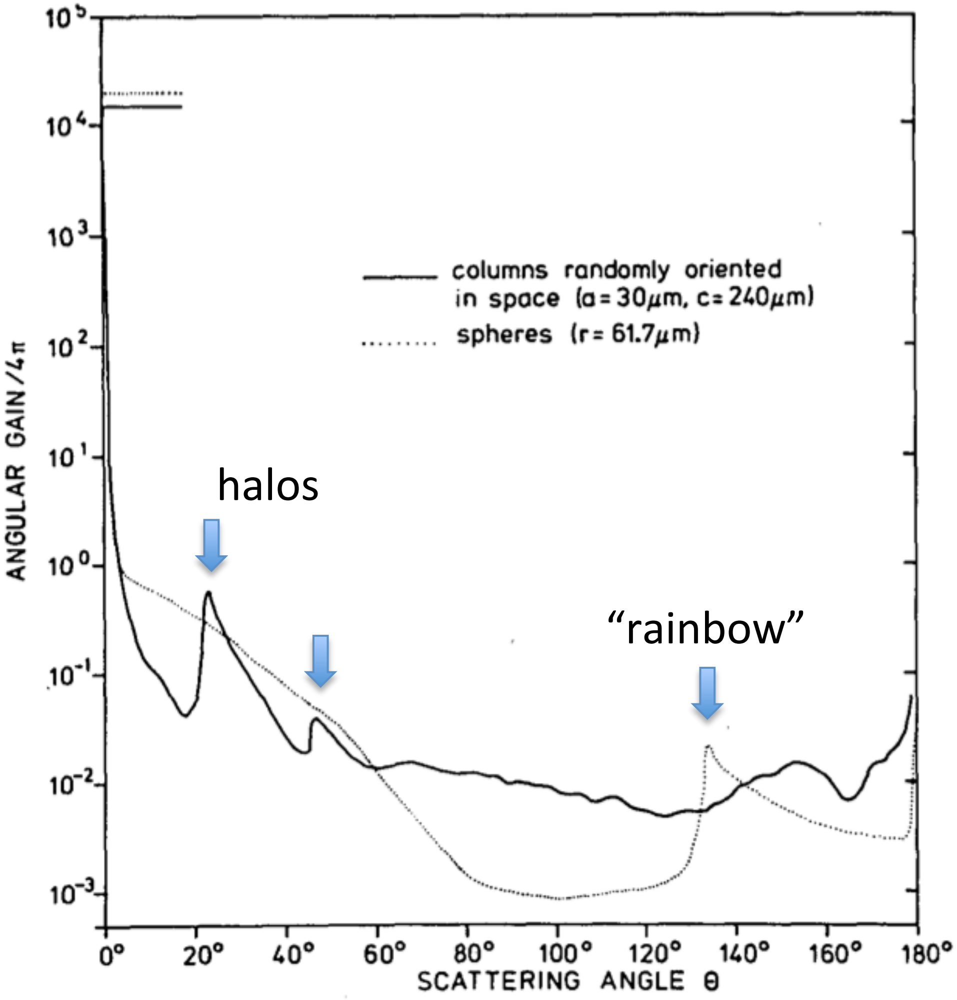


+ Almost arbitrary shapes possible

- Cannot produce phase-involving optical phenomena
- Small-scale surface structures ignored
- Homogeneous composition only
- Applicable for large *x* only

One of the largest problems in light scattering research is how to cover the range $20 < x < 50\text{-}100$ in a physically correct way!

Example: Hexagonal column and spherical ice particles at 550 nm



Can differ more than an order of magnitude!

[Wendling et al., 1979, Appl. Opt.]

Pre-calculated databases of scattering properties

- Guoshen Liu’s database for ice particles
  - Frequency range 15-340 GHz with temperatures from 0° to −40°C
  - Particle sizes (maximum dimension) from 50 to 12,500 μm
  - 11 particle shapes
  - Available online http://cirrus.met.fsu.edu/research/scatdb.html

- Bryan Baum’s and Ping Yang’s (et al.) database for ice crystals
  - 9 shapes, 3 alternatives for surface roughness
  - Wavelength range 0.2-100 µm
  - Available online http://www.ssec.wisc.edu/ice_models/

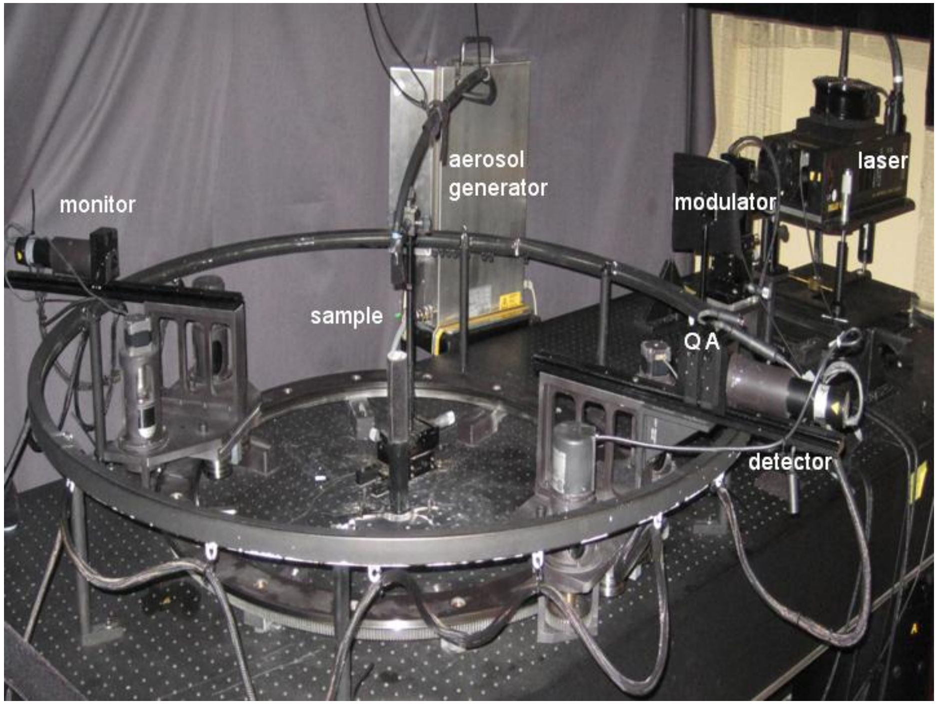
- Oleg Dubovik’s database for spheroidal dust particles
  - Based on *T*-matrix computations
  - Used in retrievals in AERONET, MODIS, SEVIRI, PARASOL, …
  - Available on request from Dr. Oleg Dubovik; Oleg.Dubovik@univ-lille1.fr

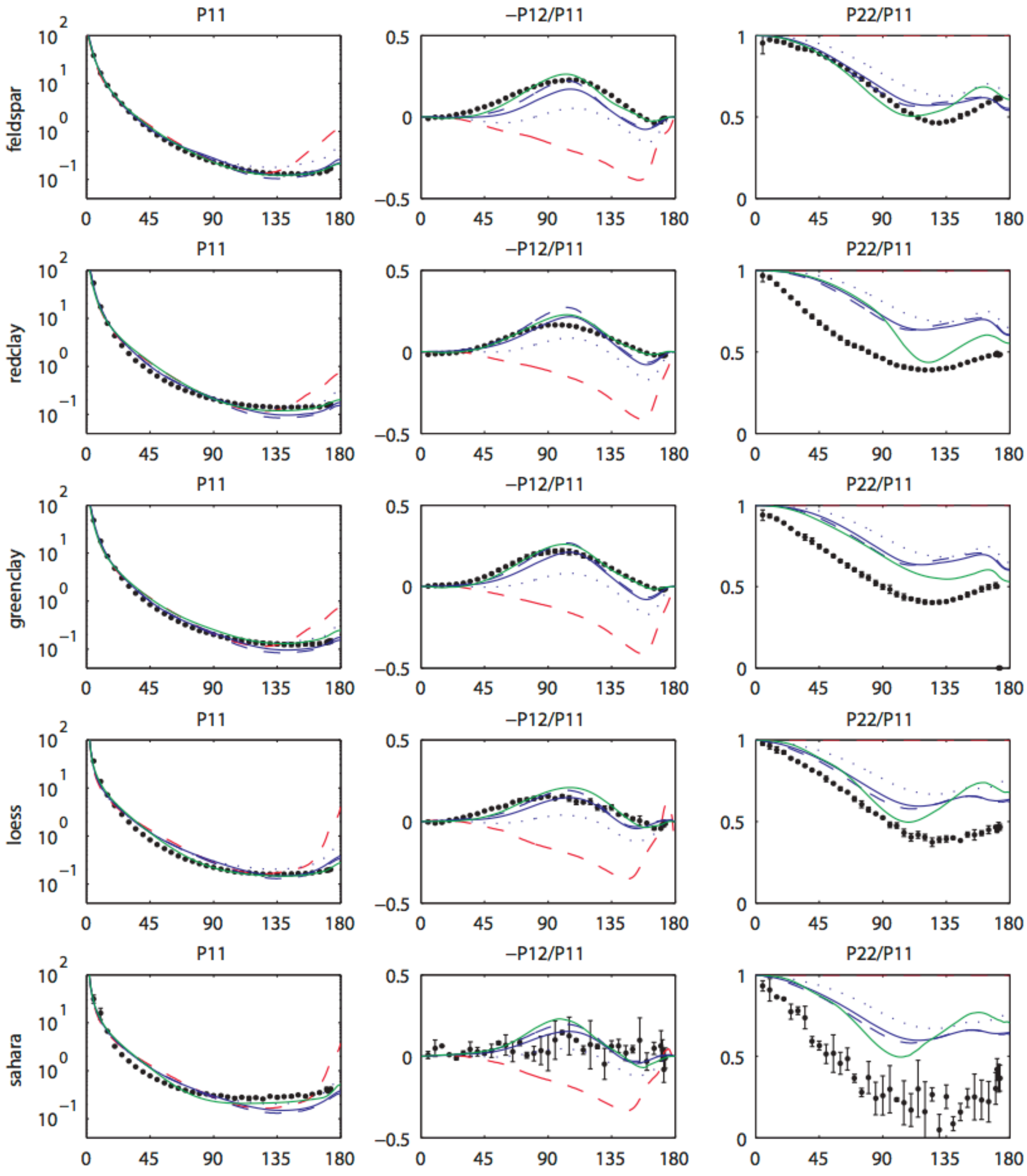
Single-scattering measurements

- Amsterdam-Granada light scattering database
  - http://www.iaa.es/scattering/

- Laboratory-measured full scattering matrices of various aerosols mainly at wavelengths 632.8 nm and 441.6 nm.

- Also average and synthetic scattering matrices available (e.g., volcanic ash, mineral dust).





Example: Measured light scattering properties of 5 different mineral dust species (black dots, error bars).

Blue and green curves are for different shape distributions of spheroids and red is for spherical particles.