# Determining Airborne Particle Properties from Polarimetric Light Scattering Measurements

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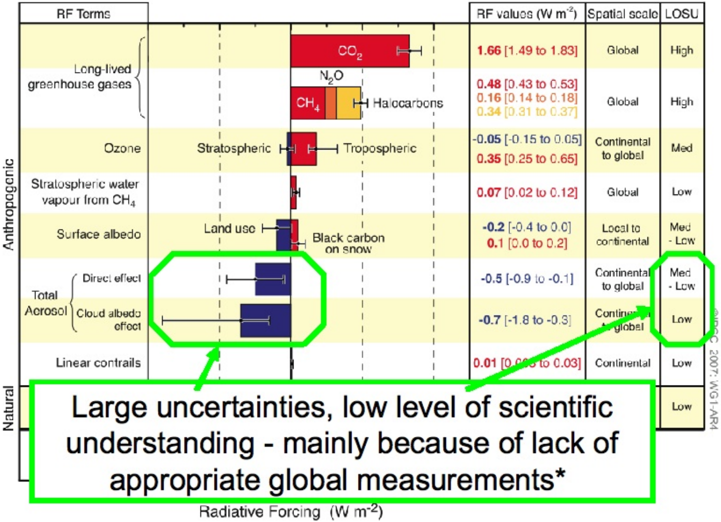
March 9, 2012

## Clouds and Aerosols

Clouds, aerosols, and cloud aerosol interactions are important for the radiation budget and climate change.

# Uncertainty in aerosol radiative effects




[IPCC 2007]

# Bio-mass burning

Figure: California

Figure: Oregon

# Volcanic eruption

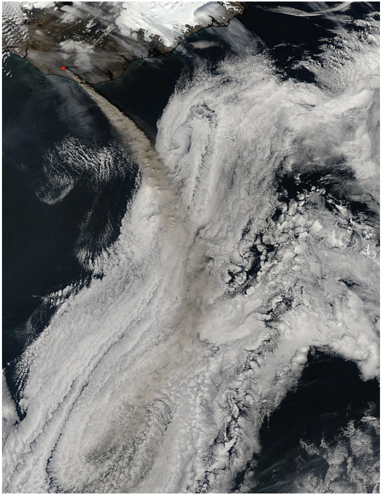


Figure: Iceland

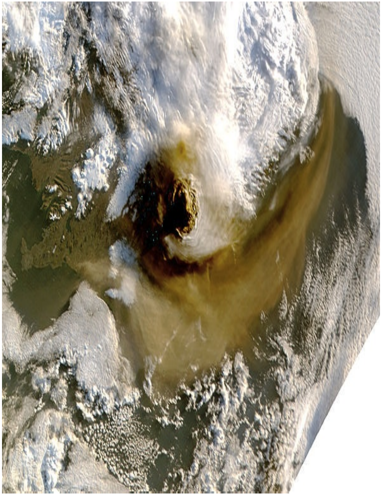


Figure: Iceland

# Desert dust

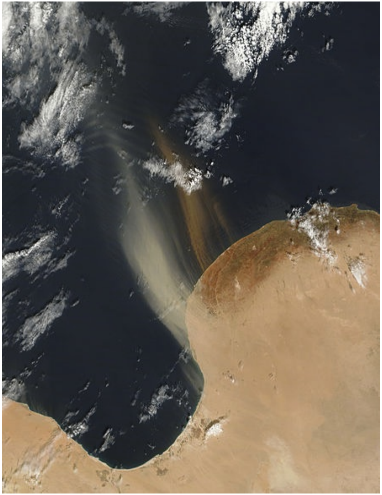


Figure: Libya

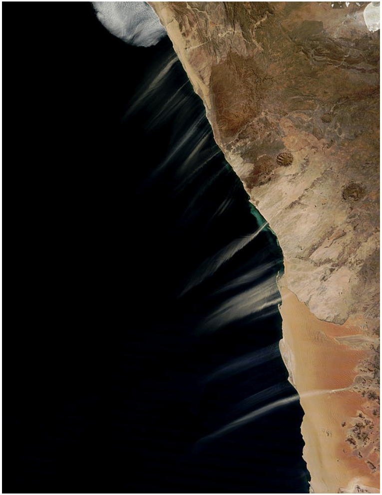
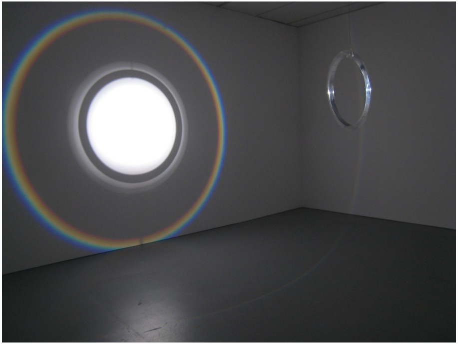


Figure: Namibia

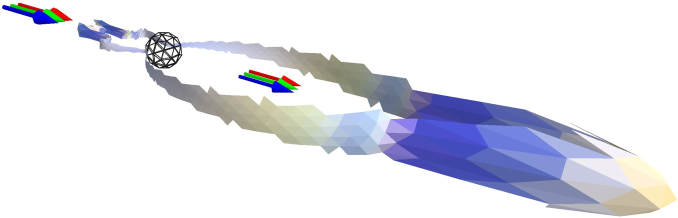
# Macroscopic particle

Particle: connected bit of matter with size, shape, and constant refractive index. Represented by state variable $\boldsymbol{\xi} \in \Omega$, where $\Omega$ is the space of all possible particles.

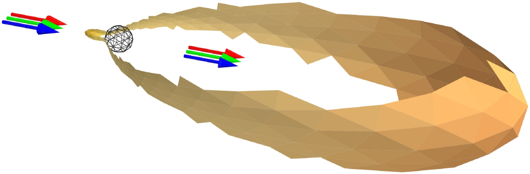


- Particle scattering calculated from the macroscopic Maxwell's equations
- All scattering info contained in the extinction crossection $C^{\text{ext}}(\boldsymbol{\xi})$ and the scattering matrix $\mathbf{F}(\Theta; \boldsymbol{\xi})$

# Water droplets

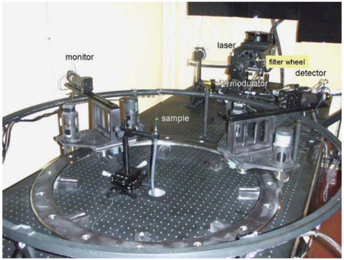


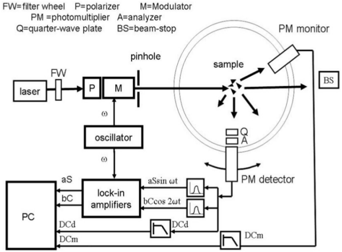
# Brown carbon



# Measurement Setup

*O. Muñoz et al. / Journal of Quantitative Spectroscopy & Radiative Transfer 111 (2010) 187–196*



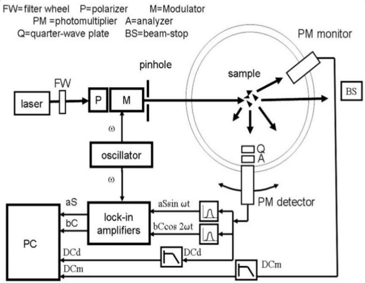




## Specifications

- $\lambda$ : 483, 488, 520, 568, and 647 nm (tunable argon-krypton laser)
- $\Theta$ : angles from $3^\circ$ to $177^\circ$ with steps of $5^\circ$ or $1^\circ$ or less.
- **F** : independent measurements of all 16 elements of the scattering matrix

**Measurement sequence:**

1. polarize and modulate phase $\phi(t) = \phi_0 \sin(\omega t)$
2. scatter off particles emitted from nozzle
3. quarter-wave plate, analyzer, flux detector
4. Lock-in amplifiers for 3 Fourier-Bessel expansion coefficients
5. Process $\left( \frac{F_{11}(\Theta)}{F_{11}(\Theta{=}30^\circ)} \quad \cdots \quad \frac{F_{ij}(\Theta)}{F_{11}(\Theta)} \quad \cdots \right)$
6. Validate using Cloude coherency tests

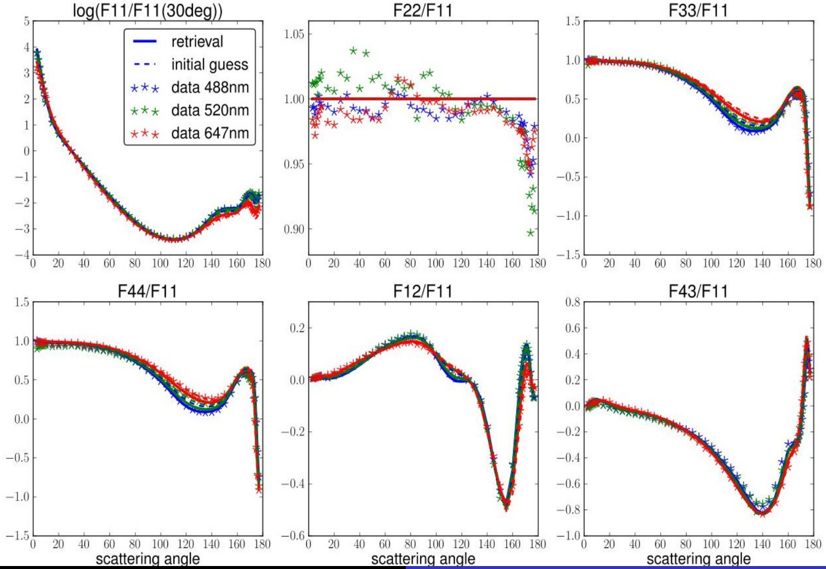



**Measurement operator:**

$$\boldsymbol{\Phi}_{\text{out}}(\lambda,\Theta) = c_1\, \mathbf{A}_{\gamma_a}\, \mathbf{Q}_{\gamma_q} \begin{pmatrix} F_{11}(\Theta) & F_{12}(\Theta) & F_{13}(\Theta) & F_{14}(\Theta) \\ F_{21}(\Theta) & F_{22}(\Theta) & F_{23}(\Theta) & F_{24}(\Theta) \\ F_{31}(\Theta) & F_{32}(\Theta) & F_{33}(\Theta) & F_{34}(\Theta) \\ F_{41}(\Theta) & F_{42}(\Theta) & F_{43}(\Theta) & F_{44}(\Theta) \end{pmatrix} \mathbf{M}_{\gamma_m}(\phi(t))\, \mathbf{P}_{\gamma_p}\, \boldsymbol{\Phi}_{\text{in}}(\lambda,\Theta)$$

| Configuration | $\gamma_P$ (deg) | $\gamma_M$ (deg) | $\gamma_Q$ (deg) | $\gamma_A$ (deg) | $DC(\theta)$ | $S(\theta)$ | $C(\theta)$ |
|---|---|---|---|---|---|---|---|
| 1 | 90 | −45 | – | – | $F_{11}$ | $F_{14}$ | $-F_{12}$ |
| 2 | 90 | −45 | – | 0 | $F_{11}+F_{21}$ | $F_{14}+F_{24}$ | $-F_{12}-F_{22}$ |
| 3 | 90 | −45 | – | 45 | $F_{11}+F_{31}$ | $F_{14}+F_{34}$ | $-F_{12}-F_{32}$ |
| 4 | 90 | −45 | 0 | 45 | $F_{11}+F_{41}$ | $F_{14}+F_{44}$ | $-F_{12}-F_{42}$ |
| 5 | 45 | 0 | – | – | $F_{11}$ | $-F_{14}$ | $F_{13}$ |
| 6 | 45 | 0 | – | 0 | $F_{11}+F_{21}$ | $-F_{14}-F_{24}$ | $F_{13}+F_{23}$ |
| 7 | 45 | 0 | – | 45 | $F_{11}+F_{31}$ | $-F_{14}-F_{34}$ | $F_{13}+F_{33}$ |
| 8 | 45 | 0 | 0 | 45 | $F_{11}+F_{41}$ | $-F_{14}-F_{44}$ | $F_{13}+F_{43}$ |

# Grenada Water Droplet test case



# Mapping particles to measurements

**Forward Model**

Particle state space $\longmapsto$ Vector Radiative Transfer $\longmapsto$ Scattering measurements

1. Airborne particles are bits of matter with some size, shape and refractive index.
2. Maxwell's equations are solved for each particle to get scattering behavior.
3. Average scattering gives coefficients for the vector radiative transfer equation.
4. Single scattering predicts measurements on the polar-scattering nephelometer.
5. Synthetic measurements have quantization noise at the pixel level of the camera.

# Writing software for particle characterization **SCATering atMOsphere**

**SCATMO is . . .**

- light scattering codes for optical characterization of cloud and aerosol particles
- a *Python* interface to FORTRAN Mie scattering routines
- like NASA's Aerosol Robotic Network, and with mode dependent refractive index

# Modeling framework

A **forward model** in SCATMO is built around pre-computed scattering properties for a mode-bin decomposition of particle state space.

$$\mathbf{y}_{\text{data}} = \mathbf{f}\left(\mathbf{x}\right) + \epsilon_{\text{noise}} \tag{1}$$

$$\mathbf{f}\left(\mathbf{x}\right) = \mathbf{g}\left(\sum_{0\leq m<\#\text{m}} \alpha_m\left(\mathbf{a}_m\left(\mathbf{x}\right),\ \mathbf{b}_m\left(\mathbf{x}\right)\right)\right) \tag{2}$$

**Ingredients** to the forward model:

- **g:** GSF single scattering $(\alpha) \longmapsto$ measurable quantities
- $\alpha_m$**:** mode refractive indices $(\mathbf{b}_m)$ and bin coefficients $(\mathbf{a}_m) \longmapsto$ GSF single scattering
- $\mathbf{a}_m$, $\mathbf{b}_m$**:** single vector of log parameters $(\mathbf{x}) \longmapsto$ mode parameters $\left(\mathbf{a}_m, \mathbf{b}_m\right)_{0\leq m<\#\text{m}}$

# Average single scattering properties

Intending to average single scattering properties ($\boldsymbol{\alpha}$) over particle state space ($\boldsymbol{\xi} \in \Omega$):

$$N = \int_\Omega n_\Omega(\boldsymbol{\xi})\, d\boldsymbol{\xi} \qquad \text{number concentration } \frac{\text{particles}}{\text{volume}} \text{ from density function } n(\boldsymbol{\xi})$$

Mono disperse computations from Mie code give,

$$\boldsymbol{\alpha}_{\text{mono}}(\boldsymbol{\xi}) = C^{\text{sca}}(\boldsymbol{\xi}) \begin{pmatrix} \alpha_1^0 & & \cdots & & \alpha_1^{\ell_{\max}} \\ 0 & 0 & \alpha_2^2 & \cdots & \alpha_2^{\ell_{\max}} \\ 0 & 0 & \alpha_3^2 & \cdots & \alpha_3^{\ell_{\max}} \\ \alpha_4^0 & & \cdots & & \alpha_4^{\ell_{\max}} \\ 0 & 0 & \beta_1^2 & \cdots & \beta_1^{\ell_{\max}} \\ \frac{C^{\text{ext}}(\boldsymbol{\xi})}{C^{\text{sca}}(\boldsymbol{\xi})} & 0 & \beta_2^2 & \cdots & \beta_2^{\ell_{\max}} \end{pmatrix} \quad \text{units of } \left[\frac{\text{area}}{\text{particle}}\right]$$

Single scattering properties by integrating over all particles per unit volume

$$\boldsymbol{\alpha} = \int_\Omega \boldsymbol{\alpha}_{\text{mono}}(\boldsymbol{\xi})\, n(\boldsymbol{\xi})\, d\boldsymbol{\xi} \qquad \text{units of } \left[\frac{\text{area}}{\text{volume}}\right]$$

# Generalized spherical function representation

**Single scattering array** representation,

$$\boldsymbol{\alpha} = \begin{pmatrix} \alpha^{00} & & \dots & & \alpha^{0\ell_{\max}} \\ 0 & 0 & \alpha^{12} & \dots & \alpha^{1\ell_{\max}} \\ 0 & 0 & \alpha^{22} & \dots & \alpha^{2\ell_{\max}} \\ \alpha^{30} & & \dots & & \alpha^{3\ell_{\max}} \\ 0 & 0 & \alpha^{4} & \dots & \alpha^{4\ell_{\max}} \\ \sigma^{\text{ext}} & 0 & \alpha^{5} & \dots & \alpha^{5\ell_{\max}} \end{pmatrix}$$

**Standard single scattering**

$$\sigma^{\text{ext}} \equiv N \left\langle C^{\text{ext}} \right\rangle_{\xi} = (\,\boldsymbol{\alpha}\,)^{50}$$

$$\sigma^{\text{sca}} \equiv N \left\langle C^{\text{sca}} \right\rangle_{\xi} = (\,\boldsymbol{\alpha}\,)^{00}$$

$$\tilde{\mathbf{F}}\,(\,\Theta\,) = \frac{1}{\sigma^{\text{sca}}} \begin{pmatrix} f^0 & f^4 & & \\ f^4 & f^1 & & \\ & & f^2 & f^5 \\ & & -f^5 & f^3 \end{pmatrix}$$

Benefits to storing with units $[\frac{\text{area}}{\text{volume}}]$ :

- Well defined addition
- Exploit linearity
  $$\boldsymbol{\alpha}_1 + \boldsymbol{\alpha}_2 = \int_{\Omega} \boldsymbol{\alpha}_{\text{mono}}(\boldsymbol{\xi})\,(\,n_1 + n_2\,)(\boldsymbol{\xi})\,\mathrm{d}\boldsymbol{\xi}$$
- Total scattering is the sum of modes, without mode fraction pre-factor

**Generalized Spherical expansion (Using Wigner-d)**

$$\begin{aligned} f^0(\Theta) &= \sum_{\ell} d^{\ell}_{00}(\Theta)\ \alpha^{0\ell} \\ (f^1 + f^2)(\Theta) &= \sum_{\ell} d^{\ell}_{22}(\Theta)\ (\alpha^{1\ell} + \alpha^{2\ell}) \\ (f^1 - f^2)(\Theta) &= \sum_{\ell} d^{\ell}_{2-2}(\Theta)\ (\alpha^{1\ell} - \alpha^{2\ell}) \\ f^3(\Theta) &= \sum_{\ell} d^{\ell}_{00}(\Theta)\ \alpha^{3\ell} \\ f^4(\Theta) &= -\sum_{\ell} d^{\ell}_{02}(\Theta)\ \alpha^{4\ell} \\ f^5(\Theta) &= -\sum_{\ell} d^{\ell}_{02}(\Theta)\ \alpha^{5\ell} \end{aligned}$$

# Mode-bin decomposition

Assume the **number concentration density** function $n_\Omega(\xi)$ is a sum of modes with fixed refractive index and size density function in the span of bins $\phi_{mk}(r)$.

$$n_\Omega\left(\xi;(\mathbf{a}_m,\mathbf{b}_m)_{0\le m<\#\mathrm{m}}\right)=\sum_{0\le m<\#\mathrm{m}} a_m^k\,\phi_{mk}(r)\;\delta\Big((m_\Re(\lambda),m_\Im(\lambda))-\mathbf{b}_m\Big)$$

Integrate to get total scattering properties,

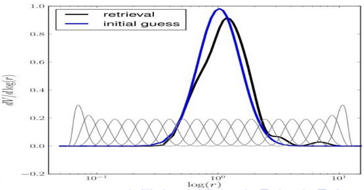
$$\begin{aligned}\boldsymbol{\alpha}\left((\mathbf{a}_m,\mathbf{b}_m)_{0\le m<\#\mathrm{m}}\right) &= \int_\Omega \boldsymbol{\alpha}_{\mathrm{mono}}(\xi)\,n_\Omega\left(\xi;(\mathbf{a}_m,\mathbf{b}_m)_{0\le m<\#\mathrm{m}}\right)d\xi\\ &= \sum_{0\le m<\#\mathrm{m}} a_m^k \underbrace{\int_{\mathbb{R}} \phi_{mk}(r)\;\boldsymbol{\alpha}_{\mathrm{mono}}(r;\,\mathbf{b}_m)\,d\xi}_{\equiv(\mathbf{M}_m(\mathbf{b}_m))_k}\end{aligned}$$

Gives a **semi-linear model** for scattering by a multi-modal distribution of spheres

$$\boldsymbol{\alpha}\Big((\mathbf{a}_m,\mathbf{b}_m)_{0\le m<\#\mathrm{m}}\Big)=\sum_{0\le m<\#\mathrm{m}}\mathbf{M}_m(\mathbf{b}_m)\cdot\mathbf{a}_m$$

$$\nabla_{\mathbf{a}_m}\boldsymbol{\alpha}=\mathbf{M}_m(\mathbf{b}_m)$$

$$\nabla_{\mathbf{b}_m}\boldsymbol{\alpha}=\nabla\mathbf{M}_m(\mathbf{b}_m)\cdot\mathbf{a}_m$$



# Semi-linear lookup

**Combined single scattering array** due to all modes,

$$\boldsymbol{\alpha}\left(\left(\mathbf{a}_m,\ \mathbf{b}_m\right)_{0\leq m<\#\mathrm{m}}\right) = \sum_{0\leq m<\#\mathrm{m}} \boldsymbol{\alpha}_m\left(\mathbf{a}_m,\ \mathbf{b}_m\right) \tag{3}$$

$$= \sum_{0\leq m<\#\mathrm{m}} \mathbf{M}_m\left(\mathbf{b}_m\right)\cdot\mathbf{a}_m, \tag{4}$$

The **parameter derivative**,

$$\mathrm{d}\boldsymbol{\alpha} = \sum_{0\leq m<\#\mathrm{m}} \mathbf{M}_m\left(\mathbf{b}_m\right)\cdot\mathrm{d}\mathbf{a}_m + \left(\nabla\mathbf{M}_m\left(\mathbf{b}_m\right)\cdot\mathbf{a}_m\right)\cdot\mathrm{d}\mathbf{b}_m \tag{5}$$

The arrays $\mathbf{M}_m\left(\mathbf{b}_m\right)$ and $\nabla\mathbf{M}_m\left(\mathbf{b}_m\right)$ interpolated from values on a grid.

# Computing **measurables** for real data

The water droplet test case from *Muñoz et.al. 2010* has measurables,

$$\mathbf{y}_{\text{data}} \approx \left( \log\left(\frac{F_{11}}{F_{11}(\,\Theta{=}30^\circ\,)}\right) \quad \frac{F_{12}}{F_{11}} \quad \frac{F_{22}}{F_{11}} \quad \frac{F_{33}}{F_{11}} \quad \frac{F_{34}}{F_{11}} \quad \frac{F_{44}}{F_{11}} \right)^t$$

Giving the map, $\mathbf{g} : \boldsymbol{\alpha} \longmapsto \mathbf{y}_{\text{model}}$

$$\mathbf{y}_{\text{model}} = \mathbf{g}\,(\boldsymbol{\alpha})^{(\,ij\lambda\,)} = \begin{cases} \log\left(\dfrac{\bar{A}(\,\boldsymbol{\theta}\,)^{(\,0j\,)}_{(\,0\ell'\,)}\,\alpha^{(\,0\ell'\lambda\,)}}{\bar{A}(\,\theta_0\,)^{(\,00\,)}_{(\,0\ell''\,)}\,\alpha^{(\,0\ell''\lambda\,)}}\right) & \text{for} \quad i = 0 \\ \dfrac{\bar{A}(\,\boldsymbol{\theta}\,)^{(\,ij\,)}_{(\,i'\ell'\,)}\,\alpha^{(\,i'\ell'\lambda\,)}}{\bar{A}(\,\boldsymbol{\theta}\,)^{(\,0j\,)}_{(\,0\ell''\,)}\,\alpha^{(\,0\ell''\lambda\,)}} & \text{for} \quad 1 \leq i < 6 \end{cases}$$

| | |
|---|---|
| $i$ | data type |
| $j$ | data angle |
| $\lambda$ | data wavelength |
| $i'$ | GSF coefficient |
| $\ell'$ | GSF sum term |
| $\lambda'$ | model wavelength |

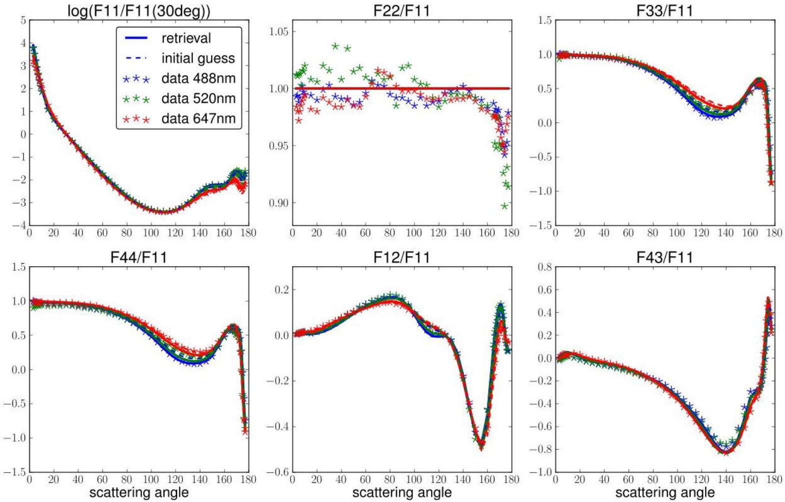
With derivative from chain rule.

$$\mathbf{d}\mathbf{y}_{\text{model}} = \nabla \mathbf{g}\,(\,\boldsymbol{\alpha}\,)^{(\,ij\lambda\,)}_{(\,i\ell\lambda\,)'} \cdot \mathbf{d}\boldsymbol{\alpha}^{(\,i\ell\lambda\,)'}$$

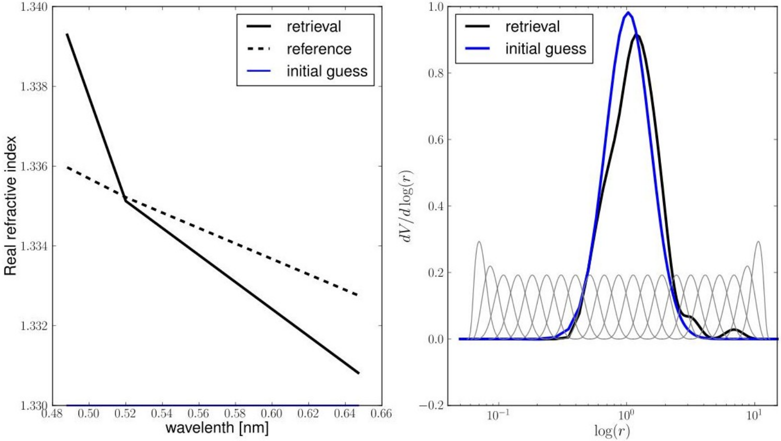
**Forward model**,

$$\mathbf{y}_{\text{model}}(\mathbf{x}) = \mathbf{g}\left( \sum_{0 \leq m < \#\text{m}} \mathbf{M}_m(\mathbf{b}_m(\mathbf{x})) \cdot \mathbf{a}_m(\mathbf{x}) \right)$$

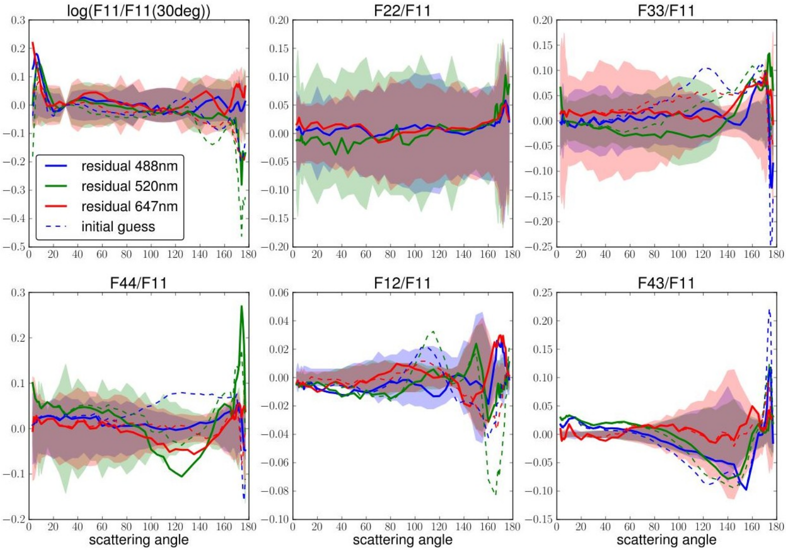
# Grenada water droplet test case



# Water droplet retrieval



# Water droplet retrieval



# Writing software for particle characterization **SCATering atMOsphere**

**SCATMO is . . .**

- a *Python* interface to FORTRAN Mie scattering code
- inspired NASA's Aerosol Robotic Network, and with mode dependent refractive index

**SCATMO is useful for . . .**

- parametrising (spherical) airborne particle populations
- computing single scattering properties
  - generalized spherical function (GSF) coefficients in units of cross section per volume
- constructing forward models to map particle parameters to measurables
- modeling data from the Amsterdam/Grenada light scattering group

# Future directions

Scattering atmosphere software **development track**:

- Retrieve particle microphysical properties from single scattering measurements
  - polar-scattering nephelometry
  - photo-acoustic-integrating nephelometry
- Integrate with radiative transfer simulations to model measurements of multiple scattering:
  - aircraft and satellite polarimeters
  - aircraft and satellite LIDAR

SCATMO features flexible logic for defining multi-modal particle populations and computing single scattering from lookup tables. Combined with rules for mapping the GSF single scattering array to measurables, it is a powerful tool for Nephelometry and atmospheric remote sensing.

**THANKS!!!**