Particle Complex Refractive Index From $3\beta+2\alpha$ HSRL/Raman Lidar Measurements: Conditions of Accurate Retrieval, Uncertainties and Constraints Provided by Information About RH

Alexei Kolgotin[1], Detlef Müller[2], Igor Veselovskii[1], Mikhail Korenskiy[1]

[1]A.M. Prokhorov General Physics Institute, Moscow, Russia, kolgotin@pic.troitsk.ru
[2]University of Hertfordshire, Hatfield, Hertfordshire, AL10 9AB, UK, d.mueller@herts.ac.uk

03 - Atmospheric aerosol and clouds properties

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Methodology

Problem to be solved

$$\boxed{\int_0^\infty K_g(\lambda,m,r)f(r)dr = g(\lambda)}$$

where:

- $g(\lambda)$ – the extinction ($g=\alpha$) and backscatter ($g=\beta$) coefficients which are measured with lidar at wavelength λ_1=355 nm, λ_2=532 nm and λ_1=355 nm, λ_2=532 nm, λ_3=1064 nm respectively ($3\beta+2\alpha$ dataset) so that 5 coefficients $\alpha_{\text{inp}}(355)$, $\alpha_{\text{inp}}(532)$, $\beta_{\text{inp}}(355)$, $\beta_{\text{inp}}(532)$ and $\beta_{\text{inp}}(1064)$ are known as well as 4 independent ratios $\{G_{\text{inp},j}\}=\{\frac{\alpha(355)}{\alpha(532)},\frac{\beta(355)}{\beta(532)},\frac{\beta(532)}{\beta(1064)},\frac{\alpha(355)}{\beta(355)}\}$;
- $K_g(\lambda,m,r)$ – the kernels described by Mie theory in case of spherical particles of radius r;
- $f(r)$ – monomodal particle size distribution (PSD) which is unknown and should be found;
- $m=m_R-im_I$ – spectrally independent complex refractive index (CRI) which is unknown and should be found on domain $m_R\in[1.3;1.8]$ and $m_I\in[0; 0.1]$

Approximation

$$\boxed{f(r) = nLN(\mu,\sigma,r) = \frac{n}{\sqrt{2\pi}\, r\ln\sigma}\exp\left[-\frac{(\ln r-\ln\mu)^2}{2(\ln\sigma)^2}\right]}$$

where

- μ – particle mean radius which is unknown and should be found on domain [0.01;0.5] μm;
- σ – particle standard deviation which is unknown and should be found on domain [1.3;2.5];
- n – particle number concentration which is unknown and should be found

Minimization procedure [1]

$$\boxed{\frac{1}{4}\sum_{j=1}^{4}\left(\frac{G_{\text{inp},j}-G_j(\mathbf{x})}{G_{\text{inp},j}}\right)^2 \to \min_{\mathbf{x}}}$$

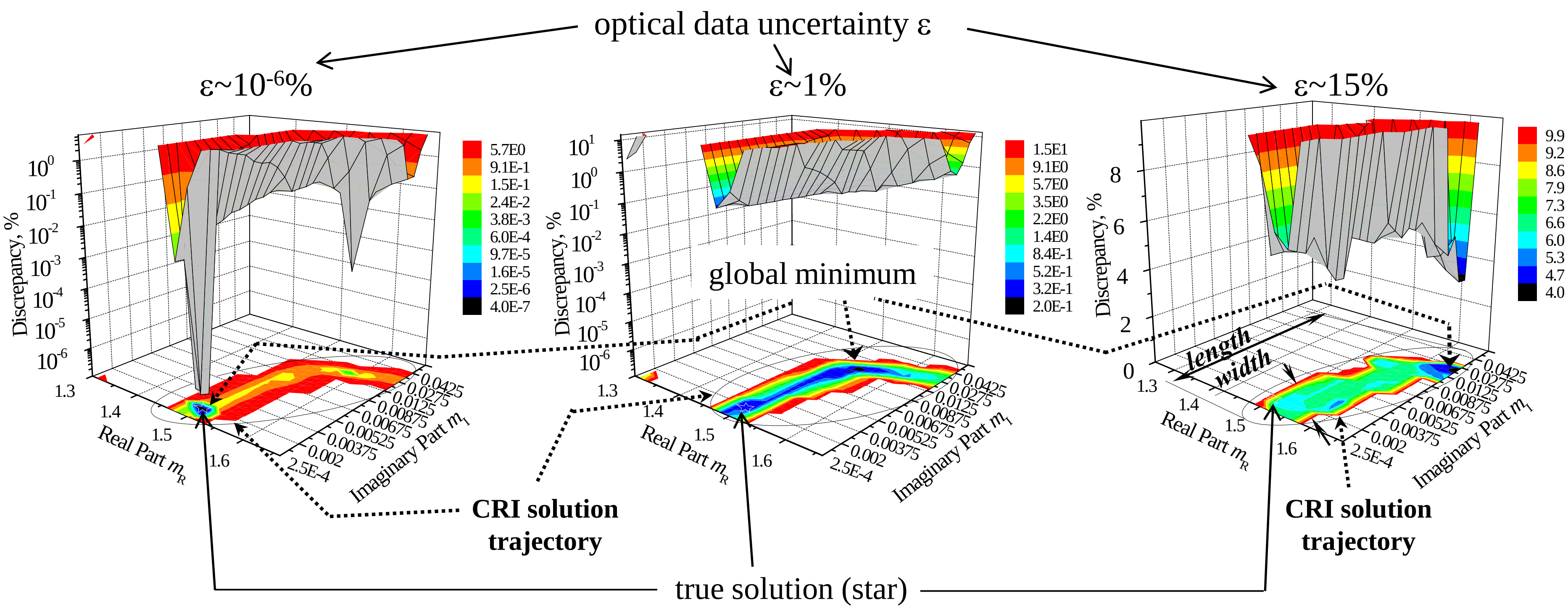
to find with preset accuracy the unknown vector $\mathbf{x}=\{x_i\}=\{m_R,m_I,\mu,\sigma\}$ that provides the best agreement (i.e. minimal discrepancy) between ratios $G_{\text{inp},j}$ and $G_j(\mathbf{x})$. The ratios $G_j(\mathbf{x})$ are determined similar to $G_{\text{inp},j}$ but from the respective coefficients $g(\lambda,\mathbf{x})$ that are directly computed with the equations

$$\boxed{g(\lambda_l,\mathbf{x}) = \int_0^\infty K_g(\lambda_l,x_1-ix_2,r)LN(x_3,x_4,r)dr \quad g=\alpha,\beta; \quad l=1,2,3}$$

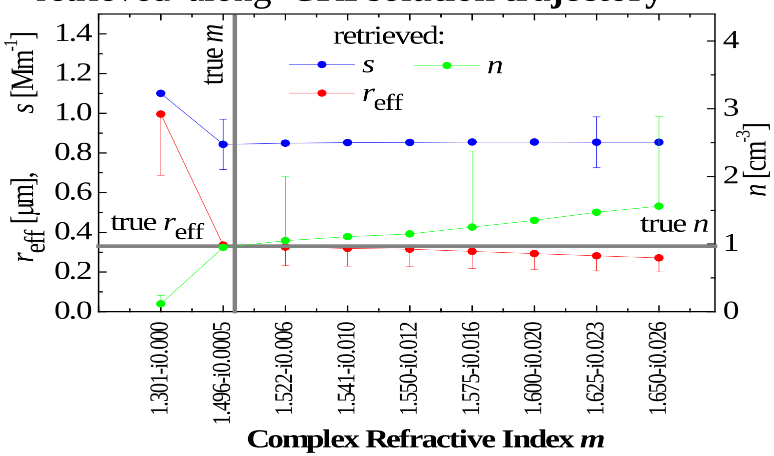
$$\boxed{n = \frac{g_{\text{inp}}(\lambda)}{g(\lambda,\mathbf{x})}}$$

[1] Kolgotin, A. et al.: Perspectives of the explicit retrieval of the complex refractive index of aerosols from optical data taken with lidar. In: Proceedings of ILRC 27, July 2015, New York, PS-B5-17

Application of the minimization procedure in numerical simulation



Particle microphysical parameters retrieved along **CRI solution trajectory**



The more CRI the less effective radius. Interdependency resembles features of size growth of hygroscopic particles.

Properties of **CRI solution trajectory**

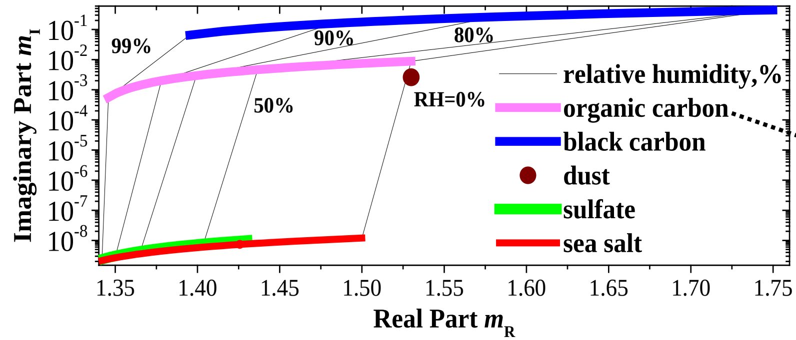
1. CRI solution trajectory describes a retrieval uncertainty of the CRI.
2. CRI solation trajectory may contain few minima.
3. Global minimum on CRI solution trajectory coincides with true solution only if (1) discrepancy magnitude ~ ε and (2) optical data uncertainty ε is less than 10^{-6}%, i.e. optical data are known to at least 8 significant digits.
4. Measurement uncertainties of ε~1%-3% of 3β+2α optical data result in a considerable spread of the CRI solution space described by CRI solution trajectory the local and global minima of which may not coincide with true solution (see star).
5. CRI solution trajectory crosses the complete (search) domain of the CRI (m_R, m_I)-plane from its lower-left corner to the top-right corner and always contains a true solution regardless measurement uncertainty ε. The trajectory *length* and *width* are determined by the CRI domain and the measurement uncertainty ε of the input optical data, respectively.

MERRA-2 data bank for major aerosol types [2]

Aerosol types, respective CRI at 532 nm and growth factors (GF) for some relative humidity (RH) intervals

Aerosol Types	RH<5%		20%≤RH≤25%		RH>90%	
	GF	CRI	GF	CRI	GF	CRI
organic carbon (OC)	1	1.53-*i*0.009	1.10	1.49-*i*0.007	2.5	1.35-*i*0.001
black carbon (BC)	1	1.75-*i*0.44	1	1.75-*i*0.44	1.9	1.43-*i*0.100
sea salt (SS)	1	1.50-*i*0.000	1.15	1.44-*i*0.000	2.5	1.35-*i*0.000
sulfate (SLFT)	1	1.43-*i*0.000	1.18	1.40-*i*0.000	2.0	1.35-*i*0.000
dust	1	1.53-*i*0.0026	1	1.53-*i*0.0026	1	1.53-*i*0.0026

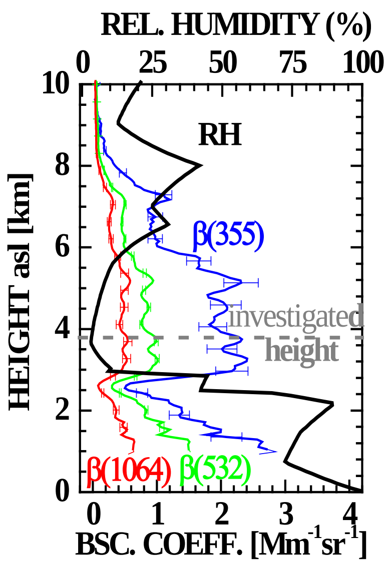
CRI trajectory versus relative humidity (RH)



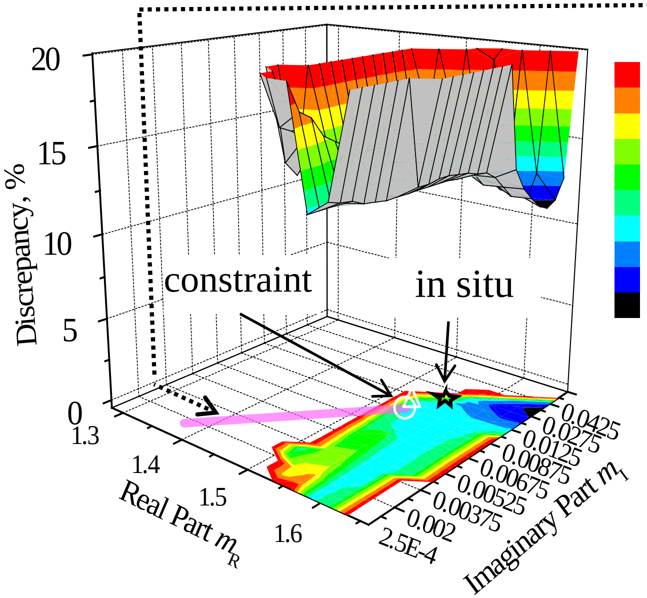
OC trajectory acts as constant on solution space at RH<5%

[2] Veselovskii, I. et al.: Characterization of smoke/dust episode over West Africa: Comparison of MERRA-2 modeling with multiwavelength Mie-Raman lidar observations, Atmos. Meas. Tech., 11, 949-969 (2018)

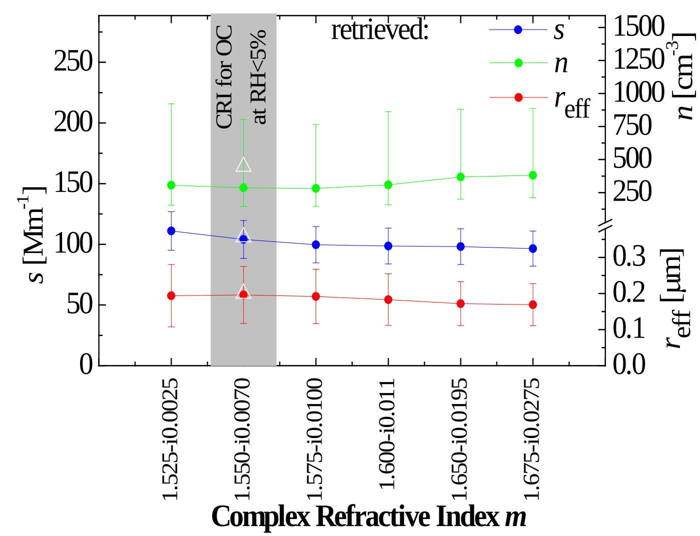
Application of the minimization procedure in smoke case study from 22 Jul 2004 at 3.8 km [3]



Profiles of backscatter coefficients measured with Raman lidar and RH taken with a radiosonde



CRI solution trajectory describing the large spread of possible solutions but final solution (circle) can be localized if information about RH is available.



Particle microphysical parameters retrieved along **CRI solution trajectory**. Final solution (circle, bullet from grey box) agrees with results derived with 2-dim. regularization (triangle) and in situ (star)

[3] Müller, D. et al.: Vertical profiles of microphysical particle properties derived from inversion with two-dimensional regularization of multiwavelength Raman lidar data: experiment, Appl. Opt., Vol. 50, No 14, 2069-2079 (2011)