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# Retrieval of complex refractive index and size distribution of spherical particles from Dual-Polarization Polar Nephelometer data

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ABSTRACT

An algorithm to retrieve the complex refractive index and size distribution of spherical particles from Dual-Polarization Polar Nephelometer data are presented. The assessment of the refractive index is based on the lookup-table approach, which is safe against converging to a local minimum, i.e. false solutions. The results of tests of the algorithm against real experimental data are outlined. The retrieved characteristics are in the expected ranges and in good agreement with the nature of particles and the specifications of the aerosol generators. It is demonstrated that, when a population of small particles, e.g., atmospheric aerosols, is dealt with and a gradient algorithm is employed to estimate the complex refractive index, either the algorithm must be able to overcome the problem of local minima or one should have some *a priori* information about credible ranges of the real and imaginary parts.

## 1. Introduction

Aerosols have been identified as a major uncertainty in predicting the global energy budget [1, Section 2.4]. The knowledge of microphysical and optical characteristics of aerosols is of importance for modelling the radiative balance of Earth's atmosphere, understanding the cloud life cycle, and remote sensing of tropospheric aerosols.

The knowledge of aerosol properties is extended first of all by means of *in situ* and laboratory measurements. And, nephelometers contribute significantly to that extension. For example, a considerable amount of experimental single scattering matrices as functions of the scattering angle (from about $3^\circ$ to $174^\circ$) have been obtained with the light scattering facility in Amsterdam. The measured data are available at the Amsterdam Light Scattering Database [2].

The long term objective of the Dual-Polarization Polar Nephelometer (D2PN) [3], designed at Laboratoire de Météorologie Physique, is to develop a database of optical and microphysical characteristics of aerosols and to test inverse codes against it. The D2PN is a device to measure the parallel and perpendicular polarized components of light scattered by an ensemble of aerosol particles. Measurements are carried out at the single wavelength $\lambda = 800\,\text{nm}$ in the interval of scattering angles from $10^\circ$ to $169^\circ$ with the step of $1^\circ$.

On the base of sensitivity tests of D2PN data to optical and microphysical parameters of ensembles of spherical particles, we showed [3] that D2PN data enable to retrieve some microphysical parameters along with the assessment of the complex refractive index. At the same time, there are limitations on retrieval of complex refractive index that are related to the

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degree of absorption of particles [3]. It is of importance to test those conclusions and our retrieval approach against real experimental data. The aim of this paper is to present results of tests we performed. Our retrieval approach is outlined first. Thereafter the experimental data and retrieval results are presented. Section 4 is devoted to the discussion.

## 2. Retrieval approach

As for atmospheric aerosols, most of the retrieval methods are based on the Lorenz–Mie theory (see, e.g., [4]), that is, aerosols are modelled as spherical particles. Due to the progress in modelling of optical properties of single particles with diverse geometrical shapes, spheroid models come into play to account for aerosol particle non-sphericity. For example, the Version 2 AERONET (AErosol RObotic NETwork) retrieval [5] provides a number of aerosol parameters (i.e., size distribution, complex refractive index and partition of spherical/non-spherical particles). Nevertheless, homogeneous spherical particles remain the most common model for aerosol inverse problems. And, our retrieval approach is also based on that model.

Broadly speaking, the retrieval of the complex refractive index $m = n$+i$\chi$ and the size distribution of particles consists of two tasks: (i) the retrieval of the size distribution when values of $n$ and $\chi$ are preassigned, and (ii) the assessment of the complex refractive index. When a value of the refractive index is preassigned, we employ a code that relies on the method developed by Dubovik and colleagues (see, e.g., [5–7]), hereafter D&C's method. There are several reasons to prefer that method. The most important reasons are discussed in the following.

First of all, D&C's method provides non-negative solutions. The problem of positivity of a retrieved particle size distribution is known for a long time (see, e.g., [8,9]). It concerns not only the contradiction that negative values are allowed as a solution for fundamentally positive parameters. Usually, negative values appear at the size range of large particles. Compared to the total number concentration their percentage can be sufficiently small. But, the contribution of the negative values to optical characteristics cannot be neglected because it is proportional to the cross section of a particle [4]. A number of algorithms, which employ the non-negativity constraint, were proposed (see, e.g., [8–12]). As it concerns the non-negativity constraint, we prefer D&C's method because, in our opinion, retrieval of logarithms of physical characteristics is the most natural and smart way to avoid negative values.

The second reason is the flexibility of D&C's method, i.e., there are a number of alternatives for implementing the inversion so that the scheme can be easily used with other applications [5–7]. The last but not least reason is that the AERONET operational code is based on that method. In other words, its high quality is proven through successful processing of huge sets of real experimental data. We note in passing that we performed a large number of simulations, including multimodal size distributions and different levels of measurement errors. Our simulations confirmed high quality of D&C's method when applied to D2PN data.

The details of D&C's method are beyond the scope of the present work; interested readers are referred to the papers [5–7]. For clarity sake, we point out that retrieval of particle size distribution from D2PN data is related to a system of Fredholm integral equations (or an equation) of the first kind (see, e.g., [5,7])

$$I_{//,\perp,0}(\theta) = \int_{r_{\min}}^{r_{\max}} K_{//,\perp,0}(\theta, n, \chi, r, )f(r)\,\mathrm{d}r, \tag{1}$$

where $I_{//}(\theta)$ and $I_{\perp}(\theta)$ are the parallel and perpendicular polarized scattered components when the sampling volume is illuminated by the unpolarized light; $I_0(\theta)$ is the unnormalized phase function; $\theta$ is the scattering angle; $K_{//,\perp,0}(\theta, n, \chi, r, )$ denote the corresponding cross sections; $r$ is the radius of a particle; $f(r)$ is the number size distribution of particles. The total number $N(r_{\min}, r_{\max})$ of particles with radii between $r_{\min}$ and $r_{\max}$ per unit of volume is

$$N(r_{\min}, r_{\max}) = \int_{r_{\min}}^{r_{\max}} f(r)\,\mathrm{d}r. \tag{2}$$

The components $I_{//}(\theta)$ and $I_{\perp}(\theta)$ can be expressed as follows:

$$I_{//}(\theta) = \frac{C_{sc}}{8\pi}[F_{11}(\theta) + F_{12}(\theta)], \quad I_{\perp}(\theta) = \frac{C_{sc}}{8\pi}[F_{11}(\theta) - F_{12}(\theta)], \tag{3}$$

where $C_{sc}[m^{-1}]$ is the scattering coefficient, $F_{11}(\theta)$ and $F_{12}(\theta)[sr^{-1}]$ are the elements of the scattering matrix (see, e.g., [13,14, Section 4.2]). The element $F_{11}(\theta)$ is called the scattering phase function and satisfies the following normalization condition:

$$\frac{1}{4\pi}\int_{4\pi} F_{11}(\theta)\,\mathrm{d}\Omega = \frac{1}{2}\int_0^{\pi} F_{11}(\theta)\sin(\theta)\,\mathrm{d}\theta = 1. \tag{4}$$

The values of the unnormalized phase function can be deduced from the D2PN data by means of the formula

$$I(\theta) = I_{//}(\theta) + I_{\perp}(\theta) = \frac{C_{sc}}{4\pi}F_{11}(\theta). \tag{5}$$

The parameter $P(\theta) = -F_{12}(\theta)/F_{11}(\theta)$ is called the degree of linear polarization for incident unpolarized light (see, e.g., [15]) and can be computed as follows:

$$P(\theta) = \frac{I_{\perp}(\theta) - I_{//}(\theta)}{I_{\perp}(\theta) + I_{//}(\theta)} \tag{6}$$