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Short communication

Aspect ratio as a function of particle radius: Inversion of extinction and scattering data

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HIGHLIGHTS

- A novel method for inversion of scattering and extinction data is developed.
- Size distribution and aspect ratio of nonspherical particles can be retrieved.
- Aerosol aspect ratio is determined iteratively if refractive index is *a-priori* known.
- The method is effective for large scattering angles including side and back scatter.

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ABSTRACT

The sun and sky photometry made concurrently or, alternatively, simultaneous measurements of extinction and scattering data both represent a valuable tool for gathering the information on aerosol particles. Most typically the size distribution and/or refractive index of aerosol particles can be inferred from multispectral and/or multiangle optical data. Extraction of size-dependent aspect ratio of aerosol particles from optical data is a highly non-trivial task since the kernel of the particular integral equation is a non-linear function of the sought solution. The iterative solution to this problem is introduced and demonstrated on synthetically generated data. It is shown that retrieval of size-dependent aspect ratio is possible even for complex morphologies, most typically for irregularly shaped dust particles.

1. Introduction

The long-term observations in the Earth's atmosphere have proven that aerosol particles of different origin coexist in diverse multicomponent populations. Such particles interact with solar radiation in a complex way forming the diffuse component of ground reaching radiation. There is no doubt that measuring the parameters of scattered light is a valuable tool in optical characterization of atmospheric aerosols because of high information content of spectral sky radiance data (Kokhanovsky, 2009; Dubovik and King, 2000).

It is well recognized that aerosol particles of different sizes have different residence times in the atmosphere. For instance, submicrometer sized aerosols can remain suspended for up to several weeks in the air (Lata et al., 2003) while other particles can be readily removed by rain. The airborne particles can be of different origin and transported at long distances by wind, but most typically they are produced by local sources of pollution (Swietlicki et al., 1996), like industry, fossil fuel combustion, or other anthropogenic activities (Almeida et al., 2005). Basically, urban pollution can be increasingly raised by motor vehicles that emit particles with sizes between 50 and 200 nm which could have a residence time of about 1 week in the atmosphere. Due to processes of formation these particles can be non-spherical.

In urban areas the dust load is directly related to changes in particle size distribution (Keller and Lamprecht, 1995). Even if the prevailing composition of local particles can be monitored chemically (by analyzing the filtered samples) or inferred indirectly (based on the information on local sources of pollution), the

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morphology as a function of particle size is usually poorly known or rather completely unknown. Therefore any direct or indirect method to detect the dominant shapes of aerosol particles is always of great demand.

It is well recognized that the simple approximation of a complex morphology by a volume equivalent sphere often fails in reproducing the realistic features of observed optical data, e.g. the negative polarization. This is because the natural and man-made aerosols are neither ideally spherical nor homogeneous objects (Mishchenko, 2009). A good statistics of particle shapes can make the theoretical and experimental data more consistent, but it is only hypothetically possible to characterize the particles independent of each other. Such a separation is difficult or rather unrealizable. Although novel optical methods allow for rapid signal processing and can provide information on single particles, such techniques are scarcely used in routine practice. A more convenient approach is to characterize the polydisperse systems from their bulk optical properties by measuring the collective optical effects like scattering by arbitrarily oriented, sized and shaped particles. This is possible with nephelometers (Müller et al., 2009) that measure singly-scattered radiation and can operate in the multi-wavelength regime (McCrowey et al., 2013). Similarly, the sky radiance under low turbidity conditions can serve as a source of information on ambient particles.

One of traditional concepts in treatment of non-spherical particles is to simulate them by oblate or prolate spheroids. The prevailing morphology can be then interpreted in terms of mean aspect ratio a (Voshchinnikov et al., 2000) which relates the largest and smallest size of a dust particle. Mishchenko et al. (1995) adopted the moderate aspect ratio of 1.7 as an appropriate average, but reported values of a can vary over a wide range from 1.1 to 2.2. In general, the aspect ratio has to be considered as a function of particle radius r (Zubko et al., 2013). For a non-spherical particle, r is defined as a radius of a sphere equivalent in volume.

In the Section 2 we are introducing a simple iterative concept for extracting the aspect ratio of aerosols particles using extinction and scattering data. The test on synthetically generated data is presented in Section 3.

2. Iterative concept for the aspect ratio retrieval

The inverse problems of the atmospheric optics usually transform to the Fredholm integral equations of the first kind in which the kernel maps a sought function to the data function. Assuming O_i is a measured optical quantity (e.g. the aerosol extinction coefficient or the scattering phase function) the inverse problem can be formulated as follows

$$O_i(\xi) = \int_0^\infty K_i(r,\xi,a)\, f(r)\, a(r)\, dr\,, \tag{1}$$

where K_i is the kernel of the integral equation, r is the mean particle radius (i.e. the radius of a volume equivalent sphere), $f(r)$ is the function interpreted as a size distribution, and $a(r)$ is used to characterize the size-dependent aspect ratio. If O_i is the aerosol extinction coefficient, then the parameter ξ is usually the wavelength ($\xi \equiv \lambda$). For O_i being the scattering phase function, the parameter ξ is either the scattering angle θ or the wavelength λ. The primary purpose is to find a solution vector $f(r)\, a(r)$ that minimizes differences between the measured and theoretically predicted values of O_i. In mathematical terminology, the Eq. (1) involves a mapping in the Hilbert space from $L^2[\xi_1,\ \xi_2]$ to $L^2[r_1,\ r_2]$ constructed over a set of quadratically integrable and continuous functions $f(r)\, a(r)$. The quadratic minimization principle is combined with a quadratic constraint because of the ill-posedness of the inverse problem (Twomey, 2002).

A simple solution concept can be demonstrated on two types of data-functions $O_i(\xi)$ and $O_j(\xi)$ measured concurrently, assuming the mean refractive index of aerosol particles is *a-priori* known. There is no doubt that aerosol size distributions, shapes, and refractive indices play together when determining the light scattered by the particles. However, the main intention of this short study is to present an iterative method for solving the inverse problem for aspect ratio; and, to make our methodology more clear we have assumed the refractive index to be known in the following analysis. Experimental noise also needs to be treated by a special technique that is, however, commonly known and thus beyond the scope of this manuscript. For instance Tikhonov's regularization is one of traditional techniques (Tikhonov et al., 1990).

The used iterative technique starts with zero approximation for $a(r) = a_0(r) \equiv 1$ that allows to obtain $f_0(r)$ based on the measured values of $O_i(\xi)$. The product of $f_0(r)$ and $K_j(r,\xi,a_0)$ forms the kernel for determining the theoretical values of $O_j(\xi)$. The first approximation to $a(r)$, i.e. $a_1(r)$, is obtained by solving the integral equation

$$O_j(\xi) = \int_0^\infty K_j(r,\xi,a_1)\, f_0(r)\, a_1(r)\, dr \tag{2}$$

Since the algebraization of the above equation results in a set of non-linear equations, we applied iterative technique to reconstruct $a_1(r)$. The solution starts with $a_1^0(r) = a_0(r)$ and determines the first approximation to the aspect ratio a_1^1 from Eq. (2), where $K_j(r,\xi,a_1^0)\, f_0(r)$ is the known kernel and a_1^1 is the sought function. This procedure is repeated until $\|a_1^n - a_1^{n-1}\| < \varepsilon$ for

$$O_j(\xi) = \int_0^\infty K_j\left(r,\xi,a_1^{n-1}\right) f_0(r)\, a_1^n(r)\, dr. \tag{3}$$

If convergent, the first approximation gives the aspect ratio $a_1(r) = a_1^n(r)$. Substituting $a_1(r)$ into Eq. (1), the unknown (underlying) function $f_1(r)$ is to be obtained and in turn used as a base for searching $a_2(r)$. The iterations are terminated when both conditions $\|a_n - a_{n-1}\| < \varepsilon_a$ and $\|f_n - f_{n-1}\| < \varepsilon_f$ are satisfied concurrently. Here ε_a and ε_f are predefined error margins. It has to be noticed that there is no guaranty for successful convergence of $a(r)$ and $f(r)$, especially when the measurement errors are large enough or when a chosen particle model doesn't fit conditions in the real experiment.

This kind of methodological approach is straightforward, but could show a few difficulties. Especially, the minimization runs over 2D “slices”, i.e. the best fit is found by minimizing the errors discussed above. If a minimization error defined in 3D has the form of a paraboloid, then the concept of slicing will be convergent, but generally it might fail if multiple local minima would exist with error function. The solution will definitely depend on the grid density as well as the choice of data functions. A more general approach is to minimize $\left|\vec{Q}_i - \overline{\overline{K_i}}\,\vec{g}\right|^2$ along with $\left|\vec{Q}_j - \overline{\overline{K_j}}\,\vec{g}\right|^2$, where solution vector $\vec{g}$ is a product of $\vec{f}$ and $\vec{a}$. It is expected that this method could benefit from faster convergence.

3. Numerical experiment on synthetically generated data

The numerical implementation based on the above methodological approach was used to retrieve $a(r)$ from synthetically generated scattering phase functions and volume extinction coefficients. The numerical computations of the kernel K are time