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A rapid approximate inversion of extinction data for partially absorbing particles

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ABSTRACT

Retrieval of particle size distribution from extinction data is solved analytically using a product kernel concept and its extension to partially absorbing materials. While a conventional analytical method overestimates the portion of large particles, the correction to the integral formula makes the solution acceptable also for larger absorbing particles. A set of numerical experiments on synthetically generated aerosol optical depths has proven improved accuracy of new formula that is simple and keeps requirements on computational time or memory consumption low enough. The numerical tests shown that original and reproduced size distributions are quite consistent, including the peak position and full width at half maximum. The main advantage of the analytical approach presented here is its easy applicability in fast sampling of rapidly evolving particulate systems, where quick estimation of modal radius or PM1.0, PM2.5 and other fractions of the aerosol population is required.

1. Introduction

Optical sensing is a traditional approach to retrieve the information on polydisperse discrete random systems containing particles with sizes comparable to the wavelength of observation [1,2]. It includes an atmospheric environment polluted by natural or man-made pollutants, clouds, biological aerosols such as bacteria and pollens, and especially dust, cosmic dust dispersed in interplanetary and interstellar space, but also small contamination embedded into semitransparent media, or other particles being a direct or a side product of industrial processes. The particles developed for medical or engineering applications represent a special class of tiny objects (powders) that are subject for lab diagnostics to control their microphysical properties, specifically the purity and/or monodispersity [3,4].

The electromagnetic interaction with a particulate system depends on microphysical properties such as chemistry, sizes, shapes, internal topology and total concentration of scattering domains. Small particles dispersed in a monitored environment redistribute the electromagnetic radiation to all directions, change the polarization state and/or remove part of the energy flux from the forward beam, and also modify the spectrum of an incident electromagnetic signal. Since the absorption coefficient is a material property showing a non-trivial dependency on the operational wavelength [5] the transmission as well as the attenuation spectra can differ markedly from that of emitting light source. However, the extinction curve is a product of both the absorption and scattering processes, where the latter manifests essential wavelength- and size-dependences [6]. Therefore an alteration of a particulate system can be identified by measuring its optical response at wavelengths comparable to the dominant particle sizes.

A conventional objective of optical sensing methods is to determine the size distribution of polydisperse system of particles, which is particularly important for the physics of nucleation, coagulation, aggregation, or crystal growth either in lab or in nature. As the full-featured multiangle scattering or polarization measurements are still possible for only a few media, the most commonly retrieved optical property of particulate system is the extinction. This property is routinely recorded in remote sensing of atmospheric environment or distant objects of astrophysical interest [7]. There exists a vast database of extinction data, which has great information content in respect to particle size distribution. A rapid increase in number of experimental data and also constantly growing interest in dynamically evolving particulate systems pose large demands on fast and reliable computational methods for estimating the temporal behavior of particle size distribution.

Motivated by a large variety of problems in which the instantaneous characterization of particle system plays a crucial role we

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have found a simplified solution to the first kind Fredholm integral equation with a modified product kernel. The solution concept uses Mellin transform technique and anomalous diffraction approximation to the Mie extinction kernel combined with an analytical extension to partially absorbing particles. Despite of tenuous theoretical foundation of this semi-analytical approach, the solution formula works quite well.

2. Retrieval of particle size distribution from extinction data

Reconstruction of particle size distribution is an ill-posed problem that is difficult to solve because small perturbations on data function typically translate to large discrepancies in solution vector [8]. To overcome such instability with a numerical solution the quadratic minimization principle is customarily combined with a quadratic constraint assuming one of the quadratic forms is non-degenerated and both are positive. Then the system reduces to the well-posed problem [9].

The mapping between particle optical depth $\tau(\lambda)$ and the projected-surface-area distribution function $s(r)$ is possible through the efficiency factor for extinction $Q_{\text{ext}}(r, m, \lambda)$

$$\tau(\lambda) = \int_0^\infty Q_{\text{ext}}(r, m, \lambda)\, s(r)\, \mathrm{d}r + \varepsilon, \tag{1}$$

where r is the particle radius, m is the particle refractive index measured relative to the surrounding environment, λ is the wavelength of an incident radiation, and ε is measurement noise. Numerical solution to Eq. (1) requires its algebraization in form

$$\vec{\tau} = \mathbf{Q}_{ext}\vec{s} + \vec{\varepsilon}, \tag{2}$$

where the solution vector $\vec{s}_\alpha$ satisfies the following condition (Zuev and Naac, 1982)

$$\left\|\vec{\tau} - \mathbf{Q}_{\text{ext}}\vec{s}_\alpha\right\|^2 \le \delta\left(\varepsilon^2\right) = c\varepsilon^2, \tag{3}$$

with $\delta\left(\varepsilon^2\right)$ being an error margin and c a real value. Traditional solution to Eq. (2) uses the regularization principles [10], where α is so-called parameter of regularization minimizing the functional Φ_α

$$\Phi_\alpha = \sum_i \left[\sum_j Q_{ij}s_j - \tau_i\right]^2 + \alpha\left\{\vec{s}^T \cdot \mathbf{H} \cdot \vec{s}\right\}. \tag{4}$$

here $\mathbf{H}$ is a symmetric matrix which has relation to concept of smoothing the vector $\vec{s}$. For instance Kabanov et al. [11] implemented the Legendre polynomials to smooth $\vec{s}$ resulting in a simple form of $\mathbf{H}$ with only few non-zero diagonal and near-diagonal elements. The second term in Eq. (4), i.e. the stabilizing functional, is weighted by the parameter α that can be found applying a discrepancy principle [12].

In practically important cases the extinction is measured at discrete wavelengths ranging from λ_1 to λ_2, so Eq. (1) is typically written in form

$$\tau(\lambda) = \int_{r1}^{r2} Q_{\text{ext}}(r, m, \lambda)\, s(r)\, \mathrm{d}r + \varepsilon, \tag{5}$$

which is a mapping in Hilbert space from $L^2[\lambda_1, \lambda_2]$ to $L^2[r_1, r_2]$ [13]. Since $\tau(\lambda)$ is measured only at bounded spectral interval, the information content of extinction data is limited to particles with radii varying from $\approx r_1$ to $\approx r_2$. Shifrin [14] has performed a few numerical experiments on monomodal size distributions and found approximate relation between particle modal radius and spectral interval needed for reconstruction the original size distribution function $s(r)$.

Although the regularization algorithm is frequently used in optical diagnostics of turbid media, it requires a reasonable estimation of radii interval $r_1 \to r_2$. If chosen inappropriately, the sought solution might become partly or fully inaccurate. In addition the error margin has to be known, otherwise the convergence of $s(r)$ can suffer from too weak or too strong constraints. If e.g. the error margin is unrealistically low, the solution becomes unstable with a few oscillations including unphysical (negative) values. Also, it has to be emphasized that too dense spectral grid does not lead to higher information content. In contrast, the ill conditioned level would increase since not all data $\tau(\lambda)$ contains new information, and consequently the number of independent equations does not exceed the number of unknowns. If however the spectral spacing is too sparse, the solution function might appear inaccurate. In principle, the optimum choice for spectral interval depends on the kernel of the integral equation. The regularization concept is usually implemented as iteration scheme, where the minimization is ruled by α. At present the high performance computers are well suitable for solving system of many equations, but CPU requirements are still large enough if online analysis of rapidly evolving media is intended or if vast database of archived optical data is to be processed routinely. In such a case, the approximate tools are more convenient even if they provide less accurate results.

3. Analytical solution for a product kernel

Applying analytic eigenfunction theory to anomalous diffraction approximation to Mie theory extinction kernel (Box and Box, 1983), the solution function can be found as an integral product of q_{ext} and data function. Here q_{ext} is the Mellin transform of Q_{ext}. This concept is well applicable to product-type kernels [15], where $Q_{\text{ext}}(r, \lambda)$ can be replaced by $Q_{\text{ext}}\left(r\lambda^{-1}\right)$. The Mellin transform of kernel Q_{ext} is based on two-sided Laplace's transform

$$\left\{MQ\right\}(s) = q\left(\beta\right) = \int_0^\infty X^{\beta-1} Q(X)\, \mathrm{d}X, \tag{6}$$

where the inverse transform is

$$\left\{M^{-1}q\right\}(X) = Q(X) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} X^{-\beta} q\left(\beta\right) \mathrm{d}\beta. \tag{7}$$

Following Shifrin's and Perel'man's approach [22], we can define the data function in the form of $T\left(\beta\right) = \beta\left[\tau\left(\beta\right) - \tau(\infty)\right]$, where $\beta = 2\pi(m-1)/\lambda$ has dimension m^{-1}. In contrast, Perel'man and Punina [21] rather introduced a dimensionless parameter $\beta^* \propto \beta r_0$ with r_0 being a particle radius. Since the projected-surface-area distribution function $s(r)$ is defined as $\pi r^2 f(r)$, the theoretical (errorless) optical thickness can be expressed in the form

$$\tau(\lambda) = \pi \int_0^\infty Q_{\text{ext}}(r, m, \lambda)\, r^2 f(r)\, \mathrm{d}r, \tag{8}$$

with $f(r)$ being the number size distribution function. Using the above convention, we have

$$T\left(\beta\right) = 2\pi \int_0^\infty p\left(2r\beta\right) rf(r)\, \mathrm{d}r, \tag{9}$$

with

$$p(z) = \frac{1-\cos(z)}{z} - \sin(z). \tag{10}$$