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Improved technique of retrieving particle size distribution from angular scattering measurements

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ABSTRACT

The paper presents results of the simulation study on improved technique of retrieving the particle size distribution (PSD) of dispersed systems from angular scattering measurements. Determining the PSD based on angular scattering measurements is the inverse problem which can be expressed in terms of the Fredholm integral equation of the first kind when single and incoherent scattering modeled by Mie theory is assumed. However, the problem is highly ill-conditioned when discretized. When one incorporates two constraints to be satisfied by the sought PSD: nonnegativity of PSD values and normalization of PSD to unity when integrated over the whole range of particle size into the Regularized Least Squares (RLS) method, one obtains the proposed Constrained Regularized Least Squares (CRLS) method which consists in solving the quadratic programming problem. Accuracy (bias) and uncertainty of the CRLS solutions were assessed in the study. The study refers to the one similar for spectral extinction measurements performed before. Results of the simulation sensitivity study prove that the CRLS method is capable of retrieving PSD functions with accuracy and uncertainty satisfactory for many industrial applications. However, the method does not manage to reconstruct bimodal PSD functions so that both modes are distinguishable. Retrieved PSD satisfies physical constraints inherently.

1. Introduction

The aim of the paper is to study the quality of reconstruction of the particle size distribution (PSD) function of a dispersed system by inversing angular scattering measurement data with the use of the proposed Constrained Regularized Least Squares (CRLS) method, which was described in [1,2].

Retrieving the PSD from angular scattering measurement data, which is the volume scattering function versus scattering angle, consists in solving the inverse problem which, under conditions assumed in the study that particles are spherical, single incoherent scattering occurs and hence Mie theory is applicable, is formulated in terms of the Fredholm integral equation of the first kind which relates angularly dependent volume scattering function with the sought PSD function. Discretization of the Fredholm equation yields the system of linear algebraic equations. The PSD function is retrieved directly by solving the system of equations without assuming any form (formula) of the distribution beforehand and retrieving only the parameters of the form. The inverse problem is strongly ill-conditioned which means that any small perturbation of the measurement data causes huge perturbation of the solution, and hence in order to find the accurate and realistic solution one needs to use some form of *a priori* information on the solution sought. A variety of inversion methods which use diverse forms of *a priori* information were proposed. In our previous study described in [1,2] we proposed the CRLS method for solving the discretized inverse problem in retrieving PSD from spectral extinction measurements. Since the inverse problem both in spectral extinction and in angular scattering is formulated in terms

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of the discretized Fredholm integral equation of the first kind under conditions assumed in the present study, the CRLS method can be applied for retrieving PSD from angular measurements.

In our previous study, published in [1,2], we showed that the CRLS method performs significantly better than a wide range of other inversion techniques in reconstructing the PSD by inversing spectral extinction measurements in terms of a smaller uncertainty of the yielded PSD and a smaller departure of the yielded PSD from the real (test) PSD. In the present paper a similar study for angular scattering measurements is presented. We intend to investigate whether the method is also suitable for inversing angular scattering measurement data.

The results of the study are important because the CRLS method, besides its superiority over other inversion techniques in terms of quality of PSD reconstruction claimed for spectral extinction measurement, has essential inherent advantage over other techniques: it takes into account the physical constraints imposed on the sought PSD which are nonnegativity of PSD and normalization of PSD to unity when integrated over the particle size. PSDs retrieved by means of the CRLS method satisfy these constraints inherently.

The CRLS method is derived from the ordinary Regularized Least Squares (RLS) method, also known as Tikhonov regularization or Twomey–Phillips method. The RLS method seeks for the solution which simultaneously fits the measurement data best and has minimal deviation from a property such as smoothness assumed *a priori*. The drawback of the RLS method as well as other inversion techniques is that they neglect the physical constraints imposed on the sought PSD: nonnegativity of the PSD and normalization of the PSD to unity when integrated over the particle size. The CRLS procedure takes into account both constraints on top of the RLS method. This leads to the quadratic programming problem which can be solved with the use of existing quadratic programming algorithms.

Wide variety of inversion techniques have been proposed for retrieving PSDs from light scattering measurement data. In [3] the improved forward model of extinction efficiency by spheroid particles which is derived from the anomalous diffraction approximation as well as the extended anomalous diffraction approximation has been suggested. Then principle component analysis (PCA) has been applied for selecting a set of measurement data which is optimal for direct PSD retrieval. PSD has been reconstructed by solving the discretized Fredholm integral equation of the first kind with the use of proposed iterative Tikhonov method with improved algorithm for selecting regularization parameter. In [4] the inversion of extinction efficiency measurement data with the use of genetic algorithm has been investigated for retrieval of the PSD of spherical and spheroidal particles. Improved anomalous diffraction approximation has been employed as the forward model of extinction efficiency. The paper [5] presents the method for retrieval of the PSD of atmospheric aerosol together with spectrally dependent complex refractive index and single-scattering albedo of the aerosol by inversion of the ground-based measurements of the Sun and sky radiance which are spectrally dependent optical thickness and spectrally and angularly dependent intensity of the scattered light. In order to ensure nonnegativity of retrieved PSD, refractive index and single-scattering albedo values, both the quantities retrieved and measurement values are transformed to their logarithms. The retrieval consists in solving nonlinear minimization problem. Applying Lagrange multipliers allows for incorporating smoothness constraints *a priori* (in terms of minimization of the norms of the differences of various orders of the sought vectors) as well as considering various levels of uncertainties of particular components of the measurement data. Levenberg–Marquardt type and steepest descent type iterations as well as matrix inversion using singular value decomposition have been studied for solving the nonlinear minimization problem.

The CRLS method unlike the above mentioned techniques is capable of providing regularized PSDs which inherently satisfy not only the nonnegativity constraint but also the constraint of normalization to unity when integrated over particle size. To be more accurate any linear equation-type and inequality-type constraint can be incorporated by the CRLS method.

2. Mathematical model of angular scattering measurements

In angular scattering measurements, also known as nephelometric measurements, one measures the volume scattering function of the dispersed system versus the scattering angle $\beta(\theta)$ which describes directional distribution of the scattered light intensity in the examined system [6,7]. Since the measured function $\beta(\theta)$ is related to the particle size distribution (PSD) function $f(a)$, where a is the volume radius of a particle of the dispersed phase of the dispersed system by a mathematical model of measurements:

$$\beta(\theta) = K[f(a)], \tag{1}$$

where K is a certain operator, one can retrieve the PSD based on the measurement data $\beta(\theta)$ by solving the inverse problem (1) [6,7]. In the study presented in the paper the following assumptions have been made:

- the dispersed phase of the particulate system is composed of homogenous, isotropic spherical particles of absolute complex refractive index $m_1 = n_1 + ik_1$;
- the dispersion medium of the particulate system is homogenous and isotropic and has absolute complex refractive index $m_2 = n_2 + ik_2$;
- incident light is a quasi-monochromatic, unpolarized, plane wave; and
- elastic, single and incoherent scattering occurs in the particulate system;

under which model (1) assumes the form of the Fredholm integral equation of the first kind [6,7]:

$$\beta(\theta) = \int_0^{\infty} K(\theta, a) f(a) \mathrm{d}a \tag{2}$$

The kernel function $K(\theta, a)$ is given by the formula [6–8]:

$$K(\theta, a) = N_v \frac{1}{k^2} S_{11}(a, \theta, m), \tag{3}$$