Multifrequency lidar inverse problem in atmospheric aerosol studies for simple marigenic aerosol models

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Abstract

The multifrequency lidar inverse problem in aerosol research is discussed. Two models of marigenic aerosol are presented for water and water/salt aerosol ensembles. The Tikhonov functional solution method of the inverse problem for both models is described. The algorithms for calculating the values of the scattering medium optical parameters and the size-distribution functions of the aerosol are discussed. The numerical simulations performed to test the inverse problem solution algorithms are described and a brief discussion of the results is given.

1. Introduction

The multifrequency lidar inverse problem is a term describing the problem of calculating the optical characteristics of the atmosphere (especially the extinction and backscattering coefficients) using the backscattered signal received by a multifrequency lidar in experiments consisting of remote sensing of the atmosphere with lasers. However, the lidar inverse problem in aerosol studies applies to calculating the aerosol microstructure parameters (of which the important is the size distribution) from the backscattered lidar signal data. This task is one of the most interesting problems that can be solved with the lidar sensing techniques.

The optical methods of aerosol microstructure analysis are a very effective tool which provides information about aerosols in their undisturbed state (which is virtually impossible with the methods of direct aerosol studies). They are especially well suited for marine aerosol research because of almost spherical shape of water aerosol droplets (Killinger, Mooradian, eds, 1983) allowing application of the Mie theory, the only accurate scattering theory (Naats, 1978).

The precise characterization of the lidar inverse problem discussed in this paper is the determination of optical parameters of the atmosphere and the size distribution of aerosol particles from data gathered with multifrequency laser sounding of the atmosphere. The aerosol particles will be assumed to be almost spherical as they are in the case of marine aerosol.

2. Lidar equation

A multifrequency lidar can be defined, for the purposes of this paper, as a device capable of of sounding the atmosphere with monochromatic light at n discrete wavelengths $\lambda_1, \lambda_2, \ldots, \lambda_n$ from a spectrum interval lying within the visible and IR regions. The signal is backscattered to the lidar (monostatic setting) and measured at the same wavelengths $\lambda_1, \ldots, \lambda_n$. That constrains the wavelengths used to parts of spectrum lying off absorption lines. Lidars used for aerosol studies should have the spatial resolution (the length of the laser beam path from which the signal is received in a single measurement) as fine as possible.

The backscattered signal $P(z, \lambda_l)$, received by a lidar from a given distance z at the wavelength λ_l, depends both on the backscattering coefficient $\beta_\pi(z, \lambda_l)$ and on the aerosol extinction coefficient $\beta_{ext}(z, \lambda_l)$ of the sounded volume of atmosphere. We assume that the lidar sounds the atmosphere in the boundary layer and we can neglect the influence of the Rayleigh scattering.

The basis of the multifrequency lidar theory is the lidar equation (Hinkley, ed, 1976; Zuev, Naats, 1983):

$$P(z, \lambda_l) = P_0(\lambda_l) B(\lambda_l) z^{-2} \beta_\pi(z, \lambda_l) \cdot \exp\left[-2 \int_0^z \beta_{ext}(z', \lambda_l) dz'\right], \tag{1}$$

where:

$P(z, \lambda_l)$ – the backscattered signal received by the lidar from the distance z at the wavelength λ_l,

$P_0(\lambda_l)$ – the lidar output at λ_l,

$B(\lambda_l)$ – the lidar efficiency factor at the wavelength λ_l.

It is assumed here that the spatial resolution of the lidar dz is negligible in comparison to the sounding distance $z (dz \ll z)$.

3. The inverse problems

It is obvious that to make a successful direct inversion of the lidar equation one needs to know at least one of the two optical characteristics. Unfortunately, the measured variable is none of them but the signal P. It would be very helpful to know a mathematical relationship between these two characteristics, which would reduce two unknown functions β_π and β_{ext} to one. There is a simple function relating the two optical characteristics for the molecular (Rayleigh) component of the scattering but, unfortunately, in the Mie theory it is impossible to find a relation between them in an explicit analytical form (Hulst, 1957; Zuev, Naats, 1983).

An analytical calculation of the optical characteristics $\beta_\pi(\lambda)$ and $\beta_{ext}(\lambda)$ is possible only for particles of a few simple geometric shapes. One of them is a spherical particle made of optically homogeneous material with a known complex index of refraction $\bar{m}$. The scattering of light by such particles is described by the classic Mie theory (Hulst, 1957). Another class of particles for which there is an analytical scattering theory consists of particles that may be best modelled as a spherical shell of homogeneous material with a complex index of refraction

$\bar{m}_1$ and an outer radius r_2, surrounding a homogeneous spherical kernel with a different refractive index $\bar{m}_2$ and radius r_1. There is an analytical theory of light scattering on such "double spheres" (Aden, Kerker, 1951; Güttler, 1952) (such a particle is also called "two concentric spheres"). The theory is analogous to the Mie scattering theory for single spheres; in fact, the difference is in formulae for the calculation of the Mie functions a_n and b_n. In marine aerosol studies, a typical representative of the first class of particles is a droplet of sea water, and of the second one – a particle consisting of a water droplet with a salt kernel. The marigenic aerosol is very convenient for optical aerosol studies and the lidar inverse problem is particularly well defined in its studies because of the well-known optical properties of sea water and salt.

The usual way of characterizing aerosol, consisting of spherical particles, is by a particle density distribution function with respect to the particles radii $n(r)$. It can be defined only for smooth size distributions (Zuev, Naats, 1983). It is defined by a formula for the total number of particles from size interval $R = [R_1, R_2]$ in unit volume:

$$\int_{R_1}^{R_2} n(r)\mathrm{d}r.$$

One can define an analogous size distribution function for the case of "double spheres" $n(r_1, r_2)$ defined by a similar formula (Piskozub, 1988) for the number of particles with outer radii from within the interval R *per* unit volume:

$$\int_{R_1}^{R_2}\int_{R_1}^{r_2} n(r_1, r_2)\mathrm{d}r_1\mathrm{d}r_2.$$

The equations linking the optical characteristics $\beta_\pi(\lambda)$ and $\beta_{\mathrm{ext}}(\lambda)$ to the distribution s in the classical Mie theory (uniform homogeneous spheres) have the form:

$$\beta_\pi(\lambda) = \int_{R_1}^{R_2} K_\pi(r, \lambda)s(r)\mathrm{d}r \tag{2}$$

and

$$\beta_{\mathrm{ext}}(\lambda) = \int_{R_1}^{R_2} K_{\mathrm{ext}}(r, \lambda)s(r)\mathrm{d}r, \tag{3}$$

where $s(r) = \Pi r^2 n(r)$ (by definition) and the integral kernels $K_\pi(r, \lambda)$, $K_{\mathrm{ext}}(r, \lambda)$ are dimensionless functions analytically calculable from the Mie scattering theory. They are equal to the differentiable cross-sections for respectively back-scattering and extinction divided by the optical cross-section of the particle Πr^2. Within the Mie theory the kernels $K_\pi(r, \lambda)$ and $K_{\mathrm{ext}}(r, \lambda)$ are expressed in terms of functions $a_n(x)$ and $b_n(x)$, the so-called Mie functions (Hulst, 1957), *viz*:

$$K_\pi(r, \lambda) = \frac{1}{x^2}\left|\sum_{n=1}^{\infty}(-1)^n(2n+1)(a_n(x)-b_n(x))\right|^2 \tag{4}$$

and

$$K_{\mathrm{ext}}(r, \lambda) = \frac{2}{x^2}\left|\sum_{n=1}^{\infty}(2n+1)\left[|a_n(x)|^2+|b_n(x)|^2\right]\right|, \tag{5}$$

where $x = 2\Pi r/\lambda$ is a dimensionless parameter (in the case of "double spheres" $x = 2\Pi r_2/\lambda$).

In the case of "double spheres" one needs not one parameter r but two (r_1 and r_2) to describe a particle. The distribution s and the scattering efficiency factors K_π and K_{ext} depend, in this aerosol model, on both the radii r_1 and r_2 and have the form:

$$s(r_1, r_2) = \Pi r_2^2 n(r_1, r_2); \quad K_\pi(r_1, r_2, \lambda); \quad K_{ext}(r_1, r_2, \lambda).$$

Thus, equations (2) and (3) become:

$$\beta_\pi(\lambda) = \int_{R_1}^{R_2} \int_{R_1}^{r_2} K_\pi(r_1, r_2, \lambda) s(r_1, r_2) \mathrm{d}r_1 \mathrm{d}r_2, \tag{6}$$

$$\beta_{ext}(\lambda) = \int_{R_1}^{R_2} \int_{R_1}^{r_2} K_{ext}(r_1, r_2, \lambda) s(r_1, r_2,) \mathrm{d}r_1 \mathrm{d}r_2. \tag{7}$$

The Mie functions $a_n(x_1, x_2)$ and $b_n(x_1, x_2)$ have different, much more complicated, formulae in the case of "double spheres" (Güttler, 1952; Hulst, 1957) than in the classical Mie scattering theory. However, formulae (4) and (5) are valid with this aerosol model (obviously both the kernels K_π, K_{ext} and the Mie functions a_n, b_n depend on two variables in the case of Mie functions $x_1 = 2\Pi r_1/\lambda$, $x_2 = 2\Pi r_2/\lambda$).

In fact it is not difficult to notice that every integral of a function $f(r)$ takes in the case "double spheres" the form of

$$\int_{R_1}^{R_2} \int_{R_1}^{r_2} f(r_1, r_2) \mathrm{d}r_1 \mathrm{d}r_2$$

instead of

$$\int_{R_1}^{R_2} f(r) \mathrm{d}r$$

as is in the case of homogeneous spheres. One can denote both the integrals with a common symbol $\int f(r)\mathrm{d}r$ where r stands for r in the case of simple spheres and the pair (r_1, r_2) in the case of "double spheres"; $\mathrm{d}r$ denotes $\mathrm{d}r$ and $\mathrm{d}r_1$, $\mathrm{d}r_2$, respectively, and R is the interval $[R_1, R_2]$ in the first case and a triangular area delineated by the integral

$$\int_{R_1}^{R_2} \int_{R_1}^{r_2} \mathrm{d}r_1 \mathrm{d}r_2.$$

4. The calculation schemes

It is convenient to write the integral equations

$$\beta_\pi(\lambda) = \int_R K_\pi(r, \lambda) s(r) \mathrm{d}r; \; \beta_{ext}(\lambda) = \int_R K_{ext}(r, \lambda) s(r) \mathrm{d}r$$

in the operator form

$$\beta_\pi(\lambda) = K_\pi(\lambda); \quad \beta_{ext}(\lambda) = K_{ext}(\lambda). \tag{8}$$

If one could assume that the optical characteristics β_π and β_{ext} are known, it would seem possible to calculate the values of the size distribution function $s(r)$ by a simple numerical inversion of equations (8) (in fact one of the equations would be sufficient). Unfortunately, it can be proved (Dahlquist, Björck, 1974) that the solutions produced in this way are very unstable with respect to measurement errors of the optical characteristics. That means that even in the simple case of homogeneous spherical particles two distributions can be selected, such that the relation $\|Ks_1 - Ks_2\| \leqslant \delta$ (where δ is comparable to the experimental error) does not imply that the value of $\|s_1 - s_2\|$ is small. That implies that the problem of solving the equation $Ks = \beta$ is "ill-posed" (Ivanov *et al*, 1978; Tikhonov, Arsenin, 1979) when there is an error in the measured value β. It can be proved that the problem is "correct" when the set of possible solutions is compact (Ivanov *et al*, 1978). The set can be restricted to a compact set by so-called regularization methods. One of the best verified ones is the method of smoothing functional (Tikhonov, Arsenin 1979). For the equation $Ks = \beta$, this functional is written in the case of homogeneous spheres

$$T_\alpha(s) = \|Ks - \beta\|_{c_1} + \alpha\Omega^2(s), \tag{9}$$

where

$$\Omega^2(s) = p_0\|s\|^2_{L_2} + p_1\|s'\|^2_{L_2}$$

is the stabilizing functional. The symbol s' denotes, as usual, the first derivative of s. In the case of double spheres this functional has an analogous form. The difference lies in the norms used:

$$T_\alpha(s) = \frac{1}{n}\sum_{l=1}^{n}\left[\int_R K(r, \lambda_l)s(r)\mathrm{d}r - \beta(\lambda_l)\right]^2 + \Omega^2(s), \tag{10}$$

where

$$\Omega^2(s) = \alpha\left\{p_0\|s\|^2_x + p_1\left[\left\|\frac{\partial s}{\partial r_1}\right\|^2_x + \left\|\frac{\partial s}{\partial r_2}\right\|^2_x\right]\right\}$$

and the norm $\|\ \|_x$ has the explicit form:

$$\|f\|^2_x = \int_R f^2(r)\mathrm{d}r.$$

The α, p_0 and p_1 used in both functional formulae are constants (α, p_0, $p_1 \geqslant 0$). The value $\Omega(s)$ is a measure of smoothness of the function s. It can be shown that all functions that obey the condition $\Omega(s) \leqslant c = \text{const}$ form a compact set of distributions (Ivanov *et al*, 1978).

According to the variational principle, the function s, which minimizes the functional $T_\alpha(s)$ in a compact set of distributions is taken as a regularized solution of the initial equation $Ks = \beta$.

The used value of parameter α is arbitrary. However, it is obvious that the value $T_\alpha(s)$ is a smooth function of α and as such it has a minimum in the interval

$[0, \infty)(T_\alpha(s) \to \infty, \alpha \to \infty)$. Changing α makes the solution more or less smooth. A decrease of α makes the solution more rugged and prone to be influenced by random errors. On the other hand, an increase of α makes the solution smoother with the implied loss of some information. Thus, the value of α should be chosen with care. One obvious condition helping in the choice of α is for the solution s_α to be equal to zero for $r = 0$. A real, physical solution of the distribution s must meet this condition by definition ($s(r) = \Pi r^2 n(r)$, where r is the outer radius of a particle). To be a minimum of the functional $T_\alpha(s)$, the size distribution s_α must satisfy the condition that the value of the functional first variance be zero. From that condition, in the simpler case of homogeneous particles, on performing the necessary calculations the Euler equation is obtained:

$$(K^*K + \alpha D_2)s = K^*\beta, \tag{11}$$

where K^* is the operator conjugate to K (the integral operator $\int K(r, \lambda)\, \beta(\lambda)\mathrm{d}\lambda$); the expression K^*K should be read in an explicit form as $K^*K(r, l) = \int K(r, \lambda)K(l, \lambda)\mathrm{d}\lambda$, and D_2 is the Sturm-Liouville operator:

$$D_2 = p_0 + p_1 \frac{\partial^2}{\partial r^2}.$$

The calculation scheme corresponding to this equation is:

$$s_\alpha = (K^*K + \alpha D_2)^{-1} K^*\beta, \tag{12}$$

where $K_\alpha^{-1} = (K^*K + \alpha D_2)^{-1} K^*$ is the regularized operator inverse to that in equation $Ks = \beta$. This scheme bases on the greater stability of the Euler equation (11) with respect to measurement errors, as compared to equations (2) and (3).

The Euler equation approach is unfortunately impossible with a more general case of two concentric spheres because of the more complicated character of the resultant equation. However, in both cases one can obtain the "smoothed" solution s_α through a direct numerical minimization of the Tikhonov functional $T_\alpha(s)$. One has to notice that this apporach is not so time-effective as an inversion of the Euler equation.

The above discussion on the inversion of the equation $Ks = \beta$ does not answer the question of reconstructing the aerosol size distribution from the received lidar signal. One needs to know the values of one of the two optical characteristics β_π and β_{ext} to calculate the values of the other from the lidar equation (1). However, if one could assume that one of the functions, for example the extinction β_{ext} is known, then it would be possible to calculate the size distribution s by the minimization of the Tikhonov functional for the assumed model of aerosol particles. Naturally, in the case of an ensemble of simple spherical particles it is possible to calculate s by an inversion of the Euler equation.

The idea of a full inversion of the lidar signal is to assume arbitrary initial values of the extinction $\beta_{ext}^{[0]}$ along the laser path. This makes possible the calculation of backscattering coefficients (Piskozub 1985; Zuev, Naats, 1983) $\beta_\pi^{[1]}$ and then the distribution $s_\alpha^{[1]}$ using one of the above described ways. This distribution is the basis for calculating a new set of extinction values $\beta_{ext}^{[1]}$ using the original equation $K_{ext}s = \beta_{ext}$. Repeating these steps one obtains increasingly

more accurate representations of optical characteristics and the real aerosol size distributions. Iterations stop when changes in the optical characteristics between two consecutive steps fall below a given level.

A formal algebraic representation of this iterative scheme which enables a complete solution of the inverse multifrequency lidar problem, in the case of known refractive coefficient is:

$$\begin{aligned} \beta_{\pi}^{[i]} &= F(P, \beta_{\mathrm{ext}}^{[i-1]}), \\ s_{\alpha}^{[i]} &= \hat{K}_{\pi_{\alpha}}^{-1} \beta_{\pi}^{[i]}, \\ \beta_{\mathrm{ext}}^{[i]} &= \hat{K}_{\mathrm{ext}} s_{\alpha}^{[i]}. \end{aligned} \tag{13}$$

The function F, associating the backscattering coefficients for every wavelength and sounding distance, may have various forms. It may employ a direct numerical integration scheme (Zuev, ed, 1976; Piskozub, 1985; Zuev, Naats, 1983) or an iterative process (Klett, 1982; Potter, 1987).

5. Simulation of the lidar inverse problem

The author has done some computer simulations of the above schemes for the lidar inverse problem solution. The simulations involved assuming an aerosol size distribution s and calculating with appropriate equations (2) and (3) or (6) and (7) the optical characteristics β_{π} and β_{ext}. Their values were the basis for calculating simulated values of the signal returning to the lidar P which, in turn, were the input data for the program solving the inverse problem.

The numerical simulations of the multifrequency lidar inversion for aerosol described by a simple sphere model were performed by the author according to the iteration scheme (13). The results of simulations were a very good prognosis for the utilization of the scheme in actual lidar investigations of the atmospheric aerosol. The convergence of the algorithm was very quick, the difference between the values of the distributions s obtained in the second and third iteration step was smaller than 1%. The scheme was very stable in terms of random errors of the lidar signal. The inversions obtained were good for the number of frequencies $n > 5$. The optical characteristics were inverted satisfactorily with $n = 3$ (Piskozub, 1985).

The author has solved the inverse problem for "double spheres" with direct minimization of the functional (10). The nonlinear optimization method of Davidson-Fletcher and Powell with the length of an optimization step sought with the Powell algorithm (Dahlquist, Björck, 1974) was used. A number of values of the distribution functions in the triangular area defined by the integrals were the arguments of optimization.

The simulated distributions $s(r_1, r_2)$ were inverted with errors under 50% of original values of the postulated distribution. This result is quite satisfactory when compared to the results obtained with use of impactors in marine salt/water aerosol studies.

6. Discussion

The methods described above of solving the multifrequency lidar inverse problem for aerosol complexes consisting of simple spherical and "double spherical" particles allow good reconstructions of the size distribution of the aerosol and the optical characteristics of the atmosphere with lidar devices working at at least 5 wavelengths in the first case and 7 in the second. The results acquired in the numerical simulations seem comparable in their accuracy to the results of conventional impactor measurements of aerosol microstructure parameters. The calculation programmes were quite fast, especially in the method involving an inversion of the Euler equation. The algorithms used in the solution of the lidar inverse problem require the knowledge of indices of refraction of the aerosol particles. However, the marigenic aerosol, the study of which was the main purpose of this research, is composed of substances of well-known refractive indices and its particles closely resemble spheres making applicable the algorithms utilizing the Mie scattering theory.

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