Multiwavelength lidar sensing of tropospheric aerosol using Raman scattering signals

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Received August 25, 2004

The problem of determination of optical and microphysical parameters of aerosol in turbid atmosphere from data of multiwavelength lidar sensing including Raman signals from atmospheric nitrogen is discussed. Solution of lidar equations along with retrieval of aerosol parameters is performed by iteration scheme using integral relations obtained by means of Fernald transform. Algorithm of retrieval of optical parameters in the iteration scheme is based on *a priori* information on the aerosol particle size-distribution function.

Introduction

The study of vertical distribution of the aerosol number density and microphysical parameters is important for solving the problems of radiative budget of the atmosphere, aerosol physics and chemistry, as well as of the pollution transfer in the atmosphere. Multiwavelength lidar sounding can provide for valuable data, in addition to contact measurements, because it enables one to carry out long-term continuous measurements with good spatial and temporal resolution. The problem of determination of the aerosol parameters from the backscattering coefficients at several wavelengths obtained from lidar signals is related to the class of the so-called ill-posed problems and is solved using one or another regularization algorithms.[1,2]

It is known that including the extinction coefficients obtained, for example, from the signals of Raman scattering (RS) by one of the main atmospheric gases, into the set of initial data together with the backscattering coefficients, essentially stabilizes the obtained solutions.[1] Recent development of the instruments for lidar sounding has led to appearance of combined lidars realizing traditional multiwavelength sounding with receiving the RS signals.[3–6] Among them, the most interesting are comparably simple systems based on Nd:YAG-lasers with frequency conversion to the second and third harmonics (wavelengths of non-shifted channels are 1064, 532, and 355 nm, and RS channels at 387 and 607 nm).[5,6] In its turn, this stimulated appearance of new approaches to interpretation of the data of sounding, in which one or another ways for regularization of solutions were proposed taking into account specific features of the considered problem.[7–9] But none of the developed approaches used *a priori* data on aerosol particle size-distribution function in explicit form, except of general ideas about it smoothness. The majority of numerical experiments[7–9] were carried out using single-mode initial distribution functions, and quite satisfactory quality of reconstruction of aerosol parameters were obtained for them. Nevertheless, it is known that real aerosol size distributions have at least two optically active fractions: submicron, which mainly contains particles of photochemical origin, and more coarse fraction, which is formed by particles of soil origin, large salt particles, etc. Results of numerical experiments for bimodal distributions are not so convincing, in particular, only initial backscattering signals without noise were used in Ref. 8 for reconstruction of the bimodal size spectrum. In contrast to Refs. 7–9, in this paper the algorithm explicitly taking into account *a priori* data on the size-distribution function of atmospheric aerosol is considered for obtaining solutions approaching actual distributions.

In interpreting data of multiwavelength sounding taking into account the extinction of radiation along the sounding path, it is necessary to solve the system of lidar equations together with reconstruction of the aerosol microphysical parameters. Different from iterative algorithms for solving the system of multiwavelength lidar equations developed earlier,[1,10] in which numerical solution of lidar equations were performed, the nonlinear integral relationships obtained by integration of the lidar equations with the known Fernald transform[11] taking into account both Rayleigh and aerosol scattering, are used in this paper. This enables one to choose the spatial resolution in different ways, and reconstruct the parameters of aerosol layers of the optical thickness close to 1. In this case, multiple scattering can be a restriction, the effect of which can be estimated beforehand.

Main equations

Lidar equation relating the measured signals at the wavelength λ_k $(k = 1, ..., N_\lambda)$ and the optical parameters of the atmosphere has the following form:

$$F(\lambda_k, z) = \left[\beta_a(\lambda_k, z) + \beta_m(\lambda_k, z)\right] \times$$
$$\times \exp\left[-2\int_0^z \left[\sigma_a(\lambda_k, z') + \sigma_m(\lambda_k, z')\right] dz'\right], \quad (1)$$

0235-6880/06/05 375-05 $02.00

where $F(\lambda_k, z) = A(\lambda_k)P_L(\lambda_k, z)z^2$, $A(\lambda_k)$ is the instrumentation constant, z is the distance along the sounding path, $P_L(\lambda_k, z)$ is the lidar return signal, $\beta_a(\lambda_k, z)$, $\beta_m(\lambda_k, z)$ are the aerosol and molecular backscattering coefficients, respectively, $\sigma_a(\lambda_k, z)$, $\sigma_m(\lambda_k, z)$ are the coefficients of light extinction due to aerosol and molecular scattering, respectively.

The coefficients $\beta_a(\lambda_k, z)$ and $\sigma_a(\lambda_k, z)$ are related with the aerosol size-distribution function by known integral relationships, the kernels of which were calculated, for spherical particles, using Mie formulas.[1]

One can write the solution of Eq. (1) for a certain wavelength λ_k obtained using the Fernald transform as follows (let us omit the variable λ_k for brevity of Eqs. (2) and (3)):

$$\beta_a(z) = -\sigma_m(z)\,\theta_m + \frac{\hat{F}(z,t)}{1 + 2\int\limits_z^t \frac{\hat{F}(z',t)}{\theta_a(z')}\,dz'}; \qquad (2)$$

$$\hat{F}(z,t) = \frac{F(z)\beta(z_g)}{F(z_g)} T^2(t, z_g) \times$$

$$\times \exp\left[-2\int\limits_z^t \sigma_m(z')\left[1 - \theta_a(z')^{-1}\theta_m\right] dz'\right];$$

$$T^2(t, z_g) = T_m^2(t, z_g) T_a^2(t, z_g),$$

$$T_{a,m}^2(t, z_g) = \exp\left[-2\int\limits_t^{z_g} \sigma_{a,m}(z')\,dz'\right],$$

where t is an arbitrary point on the sounding path, $\theta_a(z) = \beta_a(z)/\sigma_a(z)$ and $\theta_m = \beta_m(z)/\sigma_m(z)$ are the parameters called the lidar ratios for aerosol and molecular scattering, respectively (in literature often the inverse ratios are called lidar ratios). The point z_g is the reference point, at which $\beta_a(\lambda_k, z)$ is assumed known. The value $T(t, z)$ at $t \le z$ is interpreted as atmospheric transmission of the path (t, z). The value $T^2(t, z)$ is determined from the lidar return with the following formula

$$T_a^2(t, z_g) = \frac{\exp\left[2\int\limits_t^{z_g} \theta_a(z')^{-1}\theta_m \sigma_m(z')\,dz'\right]}{1 + 2\int\limits_t^{z_g} \hat{F}(z', z_g)\theta_a(z')^{-1}\,dz'}. \qquad (3)$$

Let us write the equation for Raman return signals $F(\lambda_{Rj}, z)$ at the Raman wavelengths λ_{Rj} ($j = 1, \ldots, N_R$, where N_R is the number of the used Raman channels) corresponding to the exciting wavelength λ_j in the form:

$$\ln\frac{F(\lambda_{Rj}, z_2)}{F(\lambda_{Rj}, z_1)} = \ln\frac{\rho(z_2)}{\rho(z_1)} -$$

$$-\int\limits_{z_1}^{z_2} [\sigma_a(\lambda_j, z') + \sigma_a(\lambda_{Rj}, z') + \sigma_m(\lambda_j, z') + \sigma_m(\lambda_{Rj}, z')]\,dz', \qquad (4)$$

where $\rho(z)$ is the density of the atmosphere.

The sum of the extinction coefficients at the wavelengths λ_j and λ_{Rj}, averaged over the range interval t $[z_1, z_2]$ is determined from Eq. (4):

$$\langle\sigma_a(\lambda_{Rj}, z) + \sigma_a(\lambda_j, z)\rangle = \Delta z^{-1}\left(-\ln\frac{F(\lambda_{Rj}, z_2)}{F(\lambda_{Rj}, z_1)} + \ln\frac{\rho(z_2)}{\rho(z_1)} -\right.$$

$$\left. - \int\limits_{z_1}^{z_2} [\sigma_m(\lambda_j, z') + \sigma_m(\lambda_{Rj}, z')]\,dz'\right). \qquad (5)$$

Iteration procedure for solving lidar equations

In processing the backscattering signals, it is usually assumed that the density of the atmosphere, and, hence, the value $\sigma_m(\lambda_k, z)$ are known. If it has been possible to set the aerosol lidar ratio $\theta_a(\lambda_k, z)$ at the wavelength λ_k from *a priori* ideas, the solution (2) taken from $t = z_g$ gives the profile $\beta_a(\lambda_k, z)$, and, hence, the profiles $\sigma_a(\lambda_k, z)$ and $T(z, z_g)$ along the sounding path.

In multiwavelength sounding Equations (2) are solved together for all wavelengths by iterative method. The lidar ratios involved into these equations are determined while solving, the extinction coefficients to be used are found from Eq. (5). Let us consider one of the possible variants of constructing such a solution.

Let us divide the sounding path into a number of intervals starting from the reference point: z_g, z_1, z_2, …, z_k. As a rule, the point z_g is selected so that the aerosol scattering $\beta_a(\lambda_k, z)$ is small compared to molecular scattering in the vicinity of z_g. So solution of the inverse problem in this vicinity is not expedient because of the large errors in determining the extinction and backscattering coefficients. Let (z_1, z_2) be the range interval nearest to z_g, for which the ratio of the mean aerosol to molecular backscatter $R_{am} = \langle\beta_a(\lambda_k, z_g)\rangle/\langle\beta_m(\lambda_k, z_g)\rangle$ increases up to some critical level R_{cr} (~ 0.1–0.2). As $\beta_a(\lambda_k, z_g)$ is small on (z_g, z_1), one can calculate $T_a(z_1, z_g)$ with *a priori* setting of $\theta_a(\lambda_k, z)$, without introducing noticeable error due to inaccurate account of the transmission of the path part (z_g, z_1). One can use, for example, the optical-location model[12] as *a priori* model of atmospheric aerosol. Then the problem is solved for the sequence of the range intervals (z_1, z_2), (z_2, z_3), etc., starting from the part (z_1, z_2), the following iteration procedure is performed for each of the range intervals, for which $R_{am} \ge R_{cr}$.

In Eq. (2) at pth iteration for the qth interval (z_q, z_{q+1}) it is assumed that $t = z_q$; $\theta_a(\lambda_k, z) = \theta_a(\lambda_k, z)^{(p)}$, and $\beta_a(\lambda_k, z)^{(p)}$ is calculated for all points $z_q \le z \le z_{q+1}$. Then the interval-mean values $\langle\beta_a(\lambda_k, z)\rangle^{(p)}$ are determined (calculation at the first iteration at small optical thickness of the path part is performed at $\theta_a(\lambda_k, z)^{(1)} \to \infty$, i.e., neglecting the aerosol extinction of the path part (z_1, z_2), and at large optical thickness one should set some initial value $\theta_a(\lambda_k, z)^{(1)}$, for example, obtained in the preceding part of the path). Then aerosol microphysical parameters are reconstructed using the set of the interval-mean optical coefficients $\{<\beta_a(\lambda_k, z)>^{(p)}, <\sigma_a(\lambda_{Rj}, z) + \sigma_a(\lambda_j, z)>\}$, after that, new values of the lidar ratios $\theta_a(\lambda_k, z)^{(p+1)}$ are calculated and one passes to next iteration. After stopping the iterations using some criterion of convergence, $T^2(\lambda_k, z_g, z_{q+1})$ is calculated, and one passes to the next part of the path $[T^2(\lambda_k, z_g, z_1)$ for the first part is determined based on the *a priori* data on $\theta_a(\lambda_k, z)]$. Numerical testing shows that at $z_{q+1} < z_q$ iterations converge at any optical thickness of the layer, at $z_{q+1} > z_q$ convergence is observed at the maximum optical thickness of the layer among all wavelengths, which does not exceed 1.

Replacing in Eqs. (2) and (3) the lidar ratio depending on the distance by its effective value, it is possible that errors are introduced into the determined value $\beta_a(\lambda_k, z)$. The following upper estimate is correct for the relative error ε_β in the value $<\beta_a(\lambda_k, z)>$ averaged over the range interval (z_q, z_{q+1}): $\varepsilon_\beta \le \varepsilon_\theta C|1 - D|/(1 + \beta_a\beta_m^{-1})_{\text{eff}}$, where ε_θ is the value of the relative variation of the lidar ratio about its mean value, C is the factor equal to 0.5 at small optical thickness and increasing up to 1 at increasing the optical thickness of the part of the path up to 1 and higher, the value $1 + \beta_a(z)\beta_m^{-1}(z)$ averaged over the interval with some weight is in the denominator, the value D is determined by the formula

$$D(z) = \exp\left[-2\int_{z}^{z_{q+1}} \sigma_a(z')\mathrm{d}z'\right]\times$$
$$\times\exp\left[-2\int_{z}^{z_{q+1}} \sigma_m(z')\theta_a^{-1}(z')\theta_m\mathrm{d}z'\right]. \quad (6)$$

$D(z) \to 1$ at the decreasing optical thickness, and $D(z) \to 0$ at its increase in the case of $z < z_{q+1}$. The condition $z < z_{q+1}$ is fulfilled in the case of the reference point z_g chosen at the end of the sounding path.

It follows from the presented estimate that the error in determining $\beta_a(\lambda_k, z)$ is small for sure in two cases, i.e., when variations of the lidar ratio ε_θ on the interval (z_q, z_{q+1}) are small, in this case there are no restrictions on the optical thickness of the path part (z_q, z_{q+1}); second, at small optical thickness of the path part (z_q, z_{q+1}) due to both aerosol and molecular extinction.

Algorithm for reconstruction of microphysical parameters

The algorithm for reconstruction of the microphysical parameters of aerosol particles is the part of the iterative procedure described above. The most suitable microphysical model of aerosol is selected from a set of possible models together with reconstruction of the size spectrum. On the whole, the algorithm for reconstruction is based on general ideas.[1] Due to the little bulk of the initial experimental data (3 to 5 optical coefficients), the aerosol particle size spectrum is presented in the form of a histogram f_i of $N_r = 8$ columns uniformly on the logarithmic size scale from $r_{\min} = 0.05$ to $r_{\max} = 3.0$ μm. Applying additional assumptions about smoothness of the particle size-distribution function, the integral relationships for the optical coefficients are reduced to the linear matrix relationships:

$$\beta_{ak} = \sum_{i=1}^{n} B_{ki} f_i, \quad \sigma_{ak} = \sum_{i=1}^{n} C_{ki} f_i.$$

The matrix elements B_{ki} and C_{ki} (of $N_\lambda \times N_r$, dimensionality, where N_λ is the number of sounding wavelengths) were calculated beforehand for each model. The assumption of internal mixture of the aerosol components is used when setting the models, when particles of same size are characterized by same complex refractive index that depends on wavelength. In the general case, particles can be non-spherical. According to the data of recent measurements[13] on the global network (AERONET), the model of spherical particles is applicable to aerosols of industrial regions as well as for aerosols generated from biomass burning. At the same time, it is evidently useless for aerosol of arid origin (Saharan aerosol, Asian dust), as it mainly consists of particles of irregular shapes and thus it makes the degree of depolarization of the lidar signal up to 40% as high. However, one can select out such cases by measuring the degree of lidar return depolarization.

In order to introduce *a priori* data, a two-stage procedure is used for reconstruction of the distribution function,[14] solution for the distribution function f_i is found in the form

$$f_i = g(r_i, \gamma) + s_i,$$

where $g(r_i, \gamma)$ is the *a priori* size-distribution function with free parameters γ; s_i is the correction. At the first stage, the parameters γ_0 satisfying the condition of the minimum of the discrepancy are found by the methods of nonlinear optimization. The procedure of minimization is repeated for all microphysical models set, and the most suitable model is selected according to the minimum of the discrepancy. If the discrepancy δ obtained at the first stage has exceeded some critical level δ_0, the second stage of solution is initiated, the correction s_i for the obtained model solution is found using Tikhonov regularization method.[15] Direct minimization of the regularization functional is carried out using the method of penalty functions[16] for avoiding negative solutions. The regularization parameter is determined by use of the discrepancy.[15]

It is an essential problem to set the type of *a priori* distribution function. The one-parameter Junge distribution[14] does not take into account real bimodal shape of spectra. Bimodal distributions, which are the sum of two lognormal distributions and have five free parameters, are used in some models, for example, in Ref. 17. To set the *a priori* data in such a model, it is necessary to place certain restrictions on the range of variations of the parameters and to set their correlation. This is difficult to do, because the number of parameters is too large, and some parameters are included into the formula for $\log(r)$ nonlinearly. So, it is proposed, for the purposes of this study, to take a simpler 3-parameter model and to use combination of parametric and statistical methods for introducing *a priori* data. To do this, *a priori* distribution function $g(r_i, \gamma)$ is represented in the form (Fig. 1):

$$\log\left[g(r_i,\gamma)\right] = \gamma_3 + (c_i + b_i\gamma_m)(\log r_i - \log r_5), \quad (7)$$

where $m = 1$ for the left (submicron) branch of the spectrum ($i = 1–4$) and $m = 2$ for the right branch ($i = 6–8$). The fifth column of the histogram is the boundary between two parts of the spectrum. The coefficients $c_3 = c_5 = c_7 = b_5 = 0$, $b_3 = b_7 = 1$, the residual coefficients c_i, b_i are determined in the way described below. It follows from Eq. (7), that $\gamma_3 = \ln g(r_5, \gamma)$, $\gamma_1 = \nu_{35}$, $\gamma_2 = \nu_{75}$, where $\nu_{i5} \equiv [\ln g(r_i, \gamma) - \ln g(r_5, \gamma)]/(\ln r_i - \ln r_5)$ for $i \neq 5$. The parameter γ_3 determines the value of the distribution function for $r = r_5$ and affects only the total aerosol content. The parameters γ_1 and γ_2 determine the slope of the left and right branches of the spectrum relative to r_5.

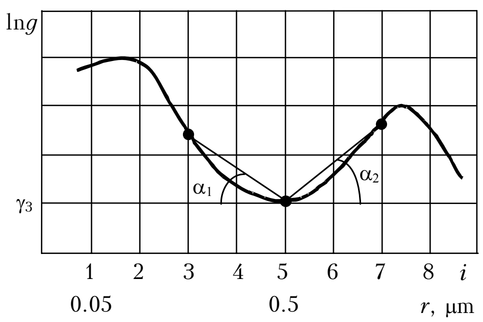

Fig. 1. Determination of the *a priori* distribution function. Number of points of the histogram i and the particle radii r are shown on the abscissa axis. Free parameters are $\gamma_1 = \tan\alpha_1$, $\gamma_2 = \tan\alpha_2$, and $\gamma_3 = \ln g(r_5)$.

Then we obtain linear relation from Eq. (7)

$$\nu_{i5} = c_i + b_i\nu_{L5}, \quad (8)$$

where $L = 3$ for the left branch ($i = 1, 2, 4$) and $L = 7$ for the right branch ($i = 6, 8$). Hence, the coefficients c_i, b_i for $i = 1, 2, 4, 6, 8$ can be defined as coefficients of linear regression, if applying Eq. (8) of the linear regression analysis to an ensemble of empirical distribution functions obtained in experiments *in situ* or from other measurements that may be considered as *a priori* in relation to lidar sounding.

Ensemble of 24 spectra formed using measurement data available from literature on the aerosol spectra in the boundary layer and in the free atmosphere for different aerosol types (urban, rural, etc.) was considered as the initial approximation.[18–26] The example of obtained correlations described by Eq. (8) is shown in Fig. 2.

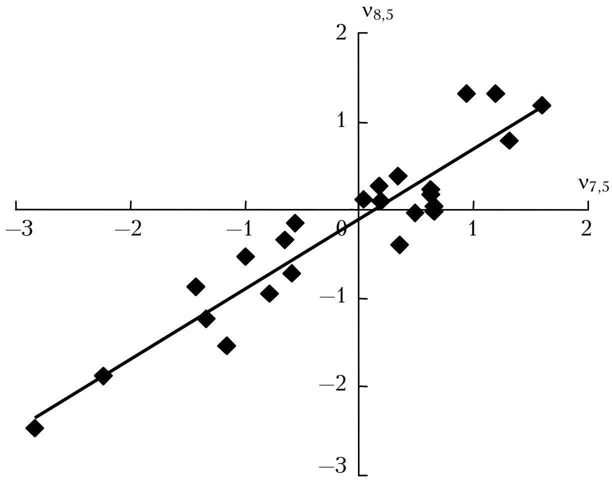

Fig. 2. Statistical relation between the parameters ν_{75} and ν_{85} for the set of empirical distribution functions.

It was obtained, based on the analysis of spectra in S-representation (particle surface distribution), that the mean values of the coefficients are $<\nu_{35}> = -1.245$, $<\nu_{75}> = -0.1444$, and their rms deviations are $\delta\nu_{35} = 0.7386$, $\delta\nu_{75} = 1.103$. Mean values of the coefficients b_i, c_i, and their rms deviations are presented in the Table.

In the future we plan to carry out similar analysis of individual types of aerosols of different origin.

Table. Mean values $<b_i>$, $<c_i>$ of the coefficients b_i, c_i and their variations δb_i, δc_i ($i = 1–8$)

$<b_i>$	0.6106	0.7571	1.0	1.121	0.0	1.174	1.0	0.7903
δb_i	0.13	0.089	0.0	0.21	0.0	0.10	0.0	0.063
$<c_i>$	0.416	0.08813	0.0	0.1507	0.0	–0.1974	0.0	–0.09992
δc_i	0.092	0.095	0.0	0.36	0.0	0.14	0.0	0.061

Conclusions

Iterative procedure is considered for solving the system of lidar equations of multiwavelength and Raman scattering with simultaneous determination of aerosol microphysical parameters. In constructing it, the integral relationships were used, which were obtained using known Fernald transforms.[11] The two-stage algorithm is applied for inverting the optical characteristics. *A priori* statistical data on the shape of the aerosol size-distribution function are used at the first stage.

Point-by-point iterative procedures were considered earlier for solving the multiwavelength sounding equations.[1,10] In this case, in passing from one point of the path to another one, numerical solution of lidar equations and solution of the inverse problem are realized simultaneously. To decrease the quadrature errors, the points of the path should be selected quite close to each other. Certain difficulties appear in the case of processing real signals, because, as a rule, signals are noised, and parameters of the aerosol medium fluctuate from point to point. So, preliminary averaging or smoothing of the signals along the sounding path are necessary. In our opinion, the most natural way for smoothing is the use of integral relationships of the type of Eq. (2), because they are exact integrals of the lidar equations taking into account both Rayleigh and aerosol scattering. So, one can consider the supposed iterative procedure as a generalization of the point-by-point procedure proposed earlier for solving the system of equations of multiwavelength sounding.

Iterative procedures can be constructed in two variants depending on what optical characteristics were selected for solving the inverse problem – backscattering or extinction coefficients.[1,10] In our opinion, in sounding atmospheric aerosol using calibration to molecular scattering signals at large heights, it is natural to use the variant of iteration of the backscattering coefficients. Indeed, in extending the solution from the reference point, the backscattering coefficients are determined, and a good first approximation of these coefficients is available in the beginning of iterations. Correction for extinction is introduced during iterations, which is small in this case. If iterate the extinction coefficients, it is necessary to introduce *a priori* the lidar ratios even at the first iteration, and the solution can strongly depend on the choice of *a priori* data.

The procedure described in the paper was applied to processing data of real experiments on multiwavelength sounding using additional Raman-lidar channel at the wavelength of 387 nm.[6] In order to study the possibilities of combined multiwavelength sounding, the vast cycle of numerical experiments on sounding atmospheric aerosol with Nd:YAG lasers was carried out. It is supposed to present the results in a separate paper.

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