ESTIMATE OF AEROSOL MICROSTRUCTURE BASED ON INTEGRAL METHOD OF MULTIPOSITION SOUNDING OF THE ATMOSPHERE

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We discuss here integral methods of k-position lidar determination of atmospheric optical parameters. Reliability of the results obtained using these methods is estimated. Thus obtained optical information is used in the reconstruction of microphysical parameters of aerosol with the account for radially varying refraction index of aerosol particles. The schemes of the multiposition lidar sounding of the atmosphere along the paths, making up a closed polygon in the intersection, are shown to be applicable to determination of parameters of realistic inhomogeneous aerosol formations. The optical model of aerosol proposed allowed us to explain the experimental results and difficulties in the interpretation of the findings on the particles spectral composition.

Aerosol essentially affects the radiation transfer process in the atmosphere, and it is one of the main climate forming factor. Information on the aerosol size distribution is useful when treating ecological problems. Optical methods and instruments are widely used for estimation of the atmospheric aerosol parameters. For example, the lidars are used for remote determination of the extinction and backscattering coefficients, and the aerosol microstructure characteristics are measured by the optical counters of particles or (remotely) by means of lidars. Interpretation of the results of such measurements should be based on the analysis of the optical-microstructural relations for particles. One should take into account the aerosols of different origin, related to the industrial and transport emissions. Such particles are different in size, chemical composition. Note that optical properties of aerosol are mainly formed by its fine particle fraction. In its turn, the coarse fraction characterizes its mass concentration.

The important problem of the lidar sounding is estimation of the reliability of determination of the optical and microphysical aerosol parameters.

Optical parameters are estimated in this paper by the integral techniques of the k-position lidar sounding of the atmosphere.[1,2] The technique is illustrated for the case of $k = 3$. The optical data obtained are used for reconstruction of the aerosol microphysical parameters taking into account the refractive index variation along the particle radius.[3]

Interpretation of the measurement results of the atmospheric aerosol parameters by means of laser sounding is based on solving the optical-radar equation (ORE). The difficulty of such an interpretation is related to the uncertainty of ORE, which contains two unknown values, the backscattering coefficient $\beta = \beta(\mathbf{r})$ and the atmospheric transmittance $T = T(\mathbf{R}_i, \mathbf{r})$. Here $\mathbf{r}$ is the radius-vector of the point of observation inside the scattering volume, $\mathbf{R}_i (i = 1, \ldots, k)$ are the radius-vectors of the points of transmitting the light pulses, with which the points of receiving the backscattering signals (RBS points) coincide.

$$T(\mathbf{R}_i,\mathbf{r}) = \exp\left(- \int\limits_{r(i)} \sigma(\mathbf{r}')\, \mathrm{d}\mathbf{r}'\right), \quad (1)$$

where $\sigma(\mathbf{r}')$ is the extinction coefficient, and integration is performed over the segment $\mathbf{r}(i)$ connecting the observation point $\mathbf{r}$ with the RBS point $\mathbf{R}_i$. Let us note that both different lidars and the same movable lidar can be placed at the RBS points. The ORE relates the backscattering signal power $P = P(\mathbf{R}_i, \mathbf{r})$ with the optical parameters of the atmosphere by the formula

$$P(\mathbf{R}_i,\mathbf{r}) = f(\Delta r)\, S(\mathbf{R}_i,\mathbf{r})\ ; \quad S(\mathbf{R}_i,\mathbf{r}) = A\beta(\mathbf{r})\, r^2(\mathbf{R}_i,\mathbf{r})\ , \quad (2)$$

where A is the lidar constant, $\Delta r = |\mathbf{r} - \mathbf{R}_i|$; $f(\Delta r)$ is the geometrical factor of the lidar, and $S(\mathbf{R}_i; \mathbf{r})$ is the power corrected for the geometrical factor of the lidar.

Different techniques for solving the multiposition ORE were developed in Refs. 1, 2, 4, and 5. In particular, in Ref. 2 ORE was solved by means of replacing it by a system of integral equations (SIE) corresponding to different paths of unpositioned sounding of the inhomogeneous atmosphere. The problem of uncertainty of this ORE is solved by the SIE technique on the basis of an *a posteriori* estimates of the relation between the unknown coefficients $\beta(\mathbf{r})$ and $\sigma(\mathbf{r})$ under the specific measurement conditions. This could be performed by means of the proposed procedure of processing the backscattering signals from

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the paths, the choice of which is determined by the conditions of their crossing inside the scattering volume under investigation. It is convenient to write the SIE in terms of the function[6]

$$\eta(\mathbf{r}) = \beta(\mathbf{r})\, \sigma^{-\xi}(\mathbf{r}) , \qquad (3)$$

to be determined. Here ξ is some constant value which should be found from the condition of minimum of the relative error in solving SIE.

Let us write the SIE for ORE. To do this let us introduce the values ($i = 1, ..., k$)

$$a_{ij} = [S(\mathbf{R}_i,\mathbf{r}_j)]^{1/\xi}; \qquad (4)$$

$$z_j = [\beta(\mathbf{r}_j)]^{-1/\xi}; \qquad (5)$$

$$b_1 = \pm \frac{2}{\xi} \int\limits_{d(i)} [S(\mathbf{R}_i,\mathbf{r})\, \eta^{-1}(\mathbf{r})]^{1/\xi}\, \mathrm{d}r , \qquad (6)$$

where $d(i)$ ($i = 1, ..., k-1$) means the side of the path polygon (PP) connecting the apices $\mathbf{r}_i$ and $\mathbf{r}_{i+1}$, and $d(k)$ is the line connecting the apices $\mathbf{r}_k$ and $\mathbf{r}_1$ of the PP. It was shown in Ref. 2 that in the case of coincidence of the number of RBS points and the PP sides the SIE has the form

$$\begin{cases} a_{ii}\, z_i - a_{i,i+1}\, z_{i+1} - b_i = 0 \quad (i = 1, \dots, k-1) , \\ a_{kk}\, z_k - a_{k1}\, z_1 - b_k = 0. \end{cases} \qquad (7)$$

Each equation of the system (7) is derived from the solution of the Bernoulli differential equations, the values $z_1, ..., z_k$ are the parameters determined from the subsequent solution of the equations from the system (7). Only the single path was considered in Ref. 6, and the unknown parameter was selected by means of the artificially constructed condition. The presence of PP introduced in Ref. 2 makes it possible to analytically determine the values $z_1, ..., z_k$.

The values (4) in the system (7) are known from measurements, and the values $z_1, ..., z_k$ can be expressed in terms of $b_1, ..., b_k$. Let us number the apex under consideration by 1. Then due to (7), any of values (5) may be represented in the form

$$\begin{cases} z_1 = B\, A^{-1} , \\ A = a_{kk} - \dfrac{a_{12}\, a_{23} \dots a_{k-1\ k}}{a_{11}\, a_{22} \dots a_{k-1\ k-1}}\, a_{k1} , \\ B = \dfrac{a_{kk}}{a_{11}} \left(b_1 + \sum\limits_{i=2}^{k} \dfrac{a_{12}\, a_{23} \dots a_{i-1\ i}}{a_{22}\, a_{33} \dots a_{ii}}\, b_i \right) . \end{cases} \qquad (8)$$

Let us consider two PPs with the common apex 1. Then one can exclude z_1 and obtain the equation relative to the values $b_1, ..., b_k$ depending on one unknown value $\eta(\mathbf{r})$. The general technique for calculating $\eta(\mathbf{r})$ by means of expanding it into a series near a given point of sounding. For simplicity let us consider only the first term of the series $\eta(\mathbf{r})$ = const, then according to (3) and (7) we obtain

$$\sigma_1 = A\, B^{-1} , \qquad (9)$$

where one can assume $\eta(\mathbf{r}) = 1$ without any loss of generality.

Let us estimate the random error of the extinction coefficient σ_1 at $k = 3$. According to the error transfer theory[7] we obtain

$$\delta_1 = \left[\sum_{i=1}^{k} \left(\frac{\partial \sigma_1}{\partial P_{ii}}\, \delta P_{ii} \right)^2 + \sum_{i=1}^{k-1} \left(\frac{\partial \sigma_1}{\partial P_{i,i+1}}\, \delta P_{i,i+1} \right)^2 + \right.$$
$$\left. + \left(\frac{\partial \sigma_1}{\partial P_{k1}}\, \delta P_{k1} \right)^2 \right]^{1/2} , \qquad (10)$$

where $P_{ij} = P(R_i, r_j)$.

Based on (8)–(10) at $k = 3$ we obtain

$$\delta_1\, \sigma_1^{-1} = \frac{1}{g(1-D)\sqrt{\sigma_1}} \left\{ \left[\Delta_4^2 + L_1^{-1} (\Delta_1 + \Delta_4)^2 \right] M_1 + \right.$$
$$+ \left[(\Delta_1 + \Delta_4)^2 + L_1^{-1} (\Delta_1 + \Delta_5)^2 \right] \times L_1^{-2}\, M_2 +$$
$$\left. + L_2^{-1} (\Delta_2 + \Delta_3)^2\, M_3\, D^2 \right\}^{1/2} . \qquad (11)$$

Here

$$\begin{cases} L_1 = e^{\sigma_1 \Delta_1} , \quad L_2 = e^{\sigma_1 \Delta_3} , \\ M_1 = e^{\sigma_0 \Delta_4} , \quad M_2 = e^{\sigma_0 (\Delta_1 + \Delta_4)} , \quad M_3 = e^{\sigma_0 \Delta_2} , \\ D = L_2\, L_1^{-2} , \end{cases} \qquad (12)$$

where σ_0 is the extinction coefficient of the medium outside of the PP; g is the experimentally determined parameter characterizing the error δ_p of the return signal power P. For the lidars where the laser meters of the cloud altitude (LMCA) are used[8] we have $\delta_p \sim \sqrt{P}$. Taking into account this fact, let us find the value g from the known range of operation of LMCA taking the signal-to-noise ratio equal to 2 and depending on the transmittance of the atmosphere. The value g for the distances in km found from the results of measurements in Sankt-Petersburg near the cross of Nepokorennyh ave. and Piskarevskii ave.[1] at η = const is presented in Table I.

TABLE I. The error in the return signal of LMCA.

Date	Time	g
23.01.91	11.00	5
24.01.91	10.30	5
30.01.91	12.40	5
20.02.91	16.00	10
22.02.91	15.00	10

Thus, the mean g value is approximately equal to 7. The results of calculations of the relative error (11) under the condition that the signal-to-noise ratio exceeds 2 are presented in Tables II and III.

TABLE II. Relative error in determining the extinction coefficient of the inhomogeneous atmosphere.

Δ_1, km	Δ_2, km	Δ_3, km	σ_1, km^{-1}	δ_1/σ_1, %
0.20	0.15	0.30	3.0	10
0.20	0.20	0.30	3.0	20
0.20	0.25	0.30	3.0	30
0.35	0.15	0.50	2.0	10
0.30	0.20	0.45	2.0	20
0.30	0.25	0.45	2.0	30
0.50	0.15	0.70	1.0	30
0.50	0.20	0.60	1.0	30

TABLE III. Relative error in determining the extinction coefficient of the homogeneous atmosphere.

Δ_1, km	Δ_2, km	Δ_3, km	σ_0, km^{-1}	σ, km^{-1}	δ/σ, %
0.20	0.25	0.30	1.0	3.0	10
0.20	0.40	0.30	1.0	3.0	20
0.20	0.50	0.30	1.0	3.0	30
0.35	0.20	0.50	1.0	2.0	10
0.35	0.35	0.50	1.0	2.0	20
0.35	0.45	0.50	1.0	2.0	30
0.50	0.20	0.60	0.5	1.0	30
0.50	0.30	0.60	0.5	1.0	30

The data given in the Tables show that the method considered allows one to determine the extinction coefficient of both homogeneous and inhomogeneous atmosphere with an acceptable error.

One can determine the number density of atmospheric aerosol particles (one can find a more detailed consideration of the problem in Ref. 1) and other aerosol characteristics from the measured optical parameters of the atmosphere. In this connection the modeling of the microscopic structure of aerosol particles is urgent. Modeling of the process of scattering of electromagnetic waves is based on solving the system of Maxwell equations and occurred effective for solving the problem of diffraction in the case of particles composed of a homogeneous core and inhomogeneous cover placed in a homogeneous nonabsorbing medium.[9] The method was proposed in Ref. 3 and allows one to obtain reliable approximations of the solution of Maxwell system for an arbitrary distribution of the complex refractive index over radius (both continuous and broken). The use of the new method has shown that the Pointing vector of the scattered wave change most significantly (by an order of magnitude or more) when the cover thickness increases, in the case of absorbing core, that should be taken into account when interpreting the data of laser measurements of the particle size-spectra. The results of calculations of a relative characteristic of the directional (at the angle of 90°) scattered radiation $R_k = I_k(\Delta)\ I^{-1}(0)$ for soot particles ($m = 1.80 - 0.64i$, wave radius $\rho_0 = 30$) are presented in Table IV as an example. Here $\Delta = |\rho_1 - \rho_0|$ is the wave thickness of the cover, the case $k = 1$ corresponds to the cover filled with dirty water, and the case $k = 2$ corresponds to the waterless cover. The results from Table IV agree with the estimate of the microstructure of aerosol particles obtained using aspiration filter and photoelectric methods.

TABLE IV. Relative error in determining the scattering coefficient of atmospheric aerosol.

Δ	0	0.6	1	1.6
R_1	1	0.8	0.7	0.2
R_2	1	0.5	0.3	0.1

Thus, it is shown that the schemes of lidar unpositioned sounding of the atmosphere along the paths forming closed polygons are applicable[3] for determining the parameters of actual aerosol formations. The use of these schemes makes it possible to adequately estimate the errors in the measured parameters, that is especially important for remote monitoring of pollution of the atmosphere over big industrial and transport centers.

The model of atmospheric aerosol created taking into account the microscopic inhomogeneities of the scatterers[3] and their significant variability allowed us to satisfactorily explain the results of experiments that did not fit the classic model, as well as the difficulties of interpretating the results of measurements of the particle size-spectra.

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