INVERSE PROBLEMS OF LIGHT SCATTERING BY AEROSOL SYSTEMS INTERACTING WITH PHYSICAL FIELDS

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The theory of inverse problems of the optics of aerosol interacting with different physical fields under the conditions of the real atmosphere is presented. It is shown that based on the numerical solution of such problems it is possible to develop methods for remote study of physical processes in the atmosphere and the spatial and temporal variability of the parametric fields. The starting information consists of the aerosol characteristics of light scattering, measured by optical sounding of the atmosphere. The basic integral equations for the inverse problems under study are presented and methods for solving the numerically are given.

The physical processes in which the aerosol systems in the atmosphere are involved can be studied by the methods of optical sounding by inverting the measured characteristics of the aerosol light scattering. To do so it is important that the spatial and temporal variability of these characteristics depends directly on the state of the physical fields in the atmosphere. Since optical methods do not "disturb" the medium under study the most reliable information about the parameters of the fields can be obtained. The main difficulties in this approach are largely related with the inversion of the optical measurements and unequivocal extraction of physical information from them. This paper is devoted to the development of numerical methods for interpreting data obtained by optical sounding of polydispersed systems of particles interacting with the physical fields. In order to present the material as clearly as possible the aerosol system in the moisture field is studied as the main example. We note that the interaction of aerosols with the moisture field plays an important role in the solution of forecasting problems, associated with the residence time of dispersed pollutants in the atmosphere.[1]

In developing a theory of optical sounding of a polydispersed systems of particles interacting with the moisture field the theory is significantly simplified if it is assumed that over the time of the experiment the total number of particles in the scattering volume remains constant. In other words, it is assumed below that sedimentation of particles, which accompanies condensation growth of particles in the moisture field, can be neglected to a first approximation.

It is convenient to start the formulation of inverse problems connected with the study of such aerosol systems with the integral representation of their optical characteristics, which is usually written in the form of the following integral:

$$\beta(\lambda) = \int_{R_1}^{R_2} K(\bar{m}, r, \lambda)\pi r^2 dN(r), \tag{1}$$

in which $K(\bar{m}, r, \lambda)$ is the efficiency factor for scattering of light with wavelength λ by a particle of size r $(R_1 \le r \le R)$ and $N(r)$ is the integral distribution of the number of particles in the local scattering volume. The attenuation efficiency factor $K_{ex}(\bar{m}, r, \lambda)$ can play the role of the function $K(\bar{m}, r, \lambda)$, if in the experiment the spectral behavior of the aerosol attenuation coefficient $\beta_{ex}(\lambda)$ or the backscattering factor $K_\pi(\bar{m}, r, \lambda)$ is measured using lidars for sounding the aerosols, etc. In all cases indicated above the quantity $\bar{m}$ characterizes the complex index of refraction of the particle material. It is well known that a particle in a moisture field grows as the humidity increases. This process is described with the help of the approximate relation $r(f) = r_d\,\varphi(f)$, in which f denotes the relative humidity and the function is called the growth factor. It is assumed that for some value $f = f_0$ (usually $f_0 \le 40$–50%) $\varphi(f_0) = \varphi_0 = 1$. It is obvious that $\varphi(f) \ge 1$ and $r(f_0) = r_d$. The quantity r_d is usually related with the so-called dry fraction of the atmospheric aerosols.[2]

As regards the change in the index of refraction of the particles owing to absorption of moisture from the air the following analytical model can be adopted: $\bar{m}(f) = \bar{m}_w + (\bar{m}_d - \bar{m}_w)\varphi^{-3}(f)$, where $\bar{m}_w$ is the index of refraction of water and $\bar{m}_d$ is the index of refraction of the aerosol material in its starting (dry) state. The dependence of the particle size and the index of refraction of its matter on the growth factor φ permits formulating the inverse optical problem, in which the spectral behavior $\beta(\lambda)$ ($\lambda_{min} \le \lambda \le \lambda_{max}$) is the measured function and the distribution $\varphi(f)$ if $f_0 \le f \le 1$) is the function sought. The function $\varphi(f)$ describes the interaction of the moisture field with the system of particles under study with the starting distribution $N_d(r_d)$ ($R_{d1} \le r_d \le R_{d2}$) and index of refraction $\bar{m}_d$. Information about the distribution $N_d(r_d)$ can be obtained from optical measurements of $\beta(\lambda, t_0)$, referred to the initial time t_0, to which the value of the humidity $f_0 = f(t_0)$ should correspond in the experiment. For this it is necessary to invert numerically the integral equation

$$\int_{R_{d1}}^{R_{d2}} K(\bar{m}_d, r, \lambda)\pi r^2 dN_d(r) = \beta(\lambda, f_0). \tag{2}$$

The index "d" in the variable of integration is dropped, since it is present in the limits of integration R_{d1} and R_{d2}. Methods for solving Eq. (2) numerically were presented in a previous work of this author.[3] We shall assume below that in the experiment the relative humidity f increases systematically in time. By virtue of the fact that the function $\varphi(f)$ increases monotonically as f increases and therefore the transformations $f \Rightarrow \varphi$ and $\varphi \Rightarrow f$ are one-to-one, the problem formulated above can be reduced to a sequence of equations of the form

$$\int_{R_j} K(\bar{m}(\varphi_j), r, \lambda)\pi r^2 dN_j(r) = \beta(\lambda, f_j), \quad j=1, \dots, m, \tag{3}$$

from which it is required to find the values of φ_j from $\beta_j = \beta(\lambda, f_j)$. In accordance with the assumption that the number of particles in any unit volume of the medium under study is conserved we can write the following equality:

$$\int_{R_j} dN_j(r) = N, \tag{4}$$

which is valid for all times t_j at which the values f_j are recorded and measurements of β_j are performed. Starting from the analytical properties of the integral distributions it can be shown that a stronger relation holds between the functions $N_j(r)$ and correspondingly the integrals R_j, namely,

$$dN_{j'}(r') = dN_{j''}(r''), \tag{5}$$

where $r' \in R_{j'}$, $r'' \in R_{j''}$ and $r'\varphi_{j'}(r') = r''\varphi_{j''}(r'')$. Assuming that $N_d(r)$ corresponds to $j = 0$ (the start of the increase in the humidity in the experiment, i.e., $\varphi_{j=0} \equiv 1$), the system (3) can be rewritten in the form

$$\int_{R_d} K(\bar{m}(\varphi_j), r\varphi_j, \lambda)\pi r^2 \varphi_j^2 dN_d(r) = \beta_j(\lambda), \; j=0, 1, \dots, m. \tag{6}$$

Since the distribution $N_d(r)$ and the interval R_d are assumed to be known, which was already mentioned above, equations (6) are determined for all j. Solving these equations numerically we find the collection $\{\varphi_j\}$ vector $\beta_f = \{\beta(f_j, \lambda)\}$, where the wavelength λ is fixed. Thus the transformation $\vec{\beta}_f \Rightarrow \vec{\varphi}$ is algorithmically fully determined and therefore the function $\varphi(f)$ is also determined by virtue of the unique correspondence between the values of β_j and f_j.

In the scheme presented above for interpreting the optical measurements, represented by the vector $\vec{\beta}_f$, the main point, of course, is the numerical solution of a nonlinear equation of the form

$$\varphi = \{\beta F^{-1}(\varphi)\}^{1/2}, \tag{7a}$$

where the notations $F(\varphi) = \int_{R_d} K(\bar{m}(\varphi), r\varphi) dS_d(r)$ and $S_d(r) = \pi r^2 dN_d(r)$ was employed. The analytical structure of this equation naturally leads to the interaction scheme

$$\varphi^{(p)} = \{\beta F^{-1}(\varphi^{(p-1)})\}^{1/2}, \tag{7b}$$

where p is the number of the iteration. In developing a theory for interpreting the experimental data based on iterative processes it is important not only to optimize their logical scheme bug also to indicate the condition under which they converge. The latter circumstance is directly related to the problem of planning an experiment, optimizing the volume of required data, and achieving the highest reliability of the results of interpretation in the face of one or another a priori uncertainty in the inverse problem. Recall that the quantities β and F depend on λ and therefore the sounding wavelength can be specifically chosen so as to make the iteration scheme (7b) converge rapidly. It should also be kept in mind that convergence of the scheme (7b), aside from everything else, indicates the existence of nontrivial solutions of Eq. (6). As regards the nonlinearity of these equations, the corresponding studies are best performed by the methods of numerical modeling taking into account the specific characteristics of concrete experiments.

The iteration scheme (7b) corresponds to the so-called simple iteration, the condition for convergence of which is well known and can be written down, for the case under study, in the form of the inequality

$$| -\varphi F'_{\varphi}/2F | < 1. \tag{8}$$

An examination of this condition suggests some simplifications that are completely acceptable in the optics of atmospheric aerosol. In particular, we shall assume that the factor $K(\bar{m}(\varphi), r\varphi, \lambda)$ is a function of one variable, for example, ρ, like, for example, in the case of the attenuation factor of soft particles, where

$\rho = 2(\bar{m} - 1)\ 2\pi r\lambda^{-1}$. We make this assumption solely to simplify the calculations of the derivative of the factor $K(\cdot)$ with respect to the variable φ, and it is in itself not related with any other fundamental restrictions. Accordingly, we can study the factor $K(\rho(\varphi))$, where $\rho(\varphi) = q(\varphi)r$ and $q(\varphi) = 2(\bar{m}(\varphi) - 1)\ 2\pi\varphi\lambda^{-1}$. Since $K'_\varphi = K'_\rho \cdot \rho'_\varphi$ and $K'_r = K'_\rho \cdot \rho'_r$ using the relation

$$K'_\varphi = K'_r\, \rho'_\varphi / \rho'_r = K'_r\, q'_\varphi / q, \qquad (9)$$

we find

$$F'_\varphi = (q'_\varphi / q) \int_{R_1}^{R_2} K'_r\, rs(r)\, \mathrm{d}r \qquad (10)$$

Subsequent integration by parts makes it unnecessary to calculate the derivatives of the factor, which is very nontrivial, if one starts from the working formulas of Mie's theory in which the indicated factors are expressed in terms of poorly converging series.[4] As a result it remains to evaluate two quantities, namely,

$$q'_\varphi/q \quad \text{and} \quad \left\{-\int_{R_2}^{R_2} K(r,\lambda)(rs)'dr \Big/ \int_{R_1}^{R_2} K(r,\lambda)s(r)dr\right\}.$$

The first quantity does not exceed the value $(-\varphi/2)$. As regards the ratio of the integrals, in accordance with the theory of differentiation of the spectral optical characteristics of light scattering by polydispersed systems of particles, developed in Refs. 3, 5, and 6, it is a function of $\omega(\lambda) = \lambda\beta'\lambda/\beta$, which plays an important role in the analysis of the spectral variability of the optical aerosol characteristics.[3] Finally, we find that the inequality (8) is equivalent to the condition

$$|w(\lambda)| < 2. \qquad (11)$$

As computational-analytical studies show[3,5,6] for typical atmospheric aerosol formations, described, in particular, by the optical models systematized in the monograph Ref. 7, the quantity $|w(\lambda)|$ is close to unity for visible wavelengths. For this reason the restriction (11) is not stringent. Representing the function $w(\lambda)$ analytically in terms of β and the derivative β^1_λ is convenient in that this makes it possible to evaluate it directly from the spectral behavior of $\beta(\lambda)$. Concluding this analysis of the computational scheme (7), we recall that the dimension of the vector being inverted $\beta_f = \{\beta(f_j, \lambda)$ at $j = 1, \ldots m\}$ is equal to that of the vector sought $\varphi = \{\varphi_j = \varphi(f_j)\}$ and the variability of the optical characteristic β with respect to the parameter f is the carrier of information about $\varphi(f)$. At the same time it is easy to see that if the growth function $\varphi(f)$ is known *a priori* the equations (6) can also serve as a source of information about the starting distribution $N_d(r)$ ($r \in R_d$). Of course, there arises the question of the information content of the vector $\vec{\beta}_f$ and comparing it with the information content of vector $\beta_\lambda = \{\beta(\lambda_1)$, for $i = 1,\ldots,n\}$ which, as it was previously proposed, should be used for preliminary estimation of $N_d(r)$. We shall make this comparative analysis on the basis of an entirely qualitative approach, using for $\varphi(f)$ the known empirical dependence $\varphi(f) = (1 - f)^{-\nu}$, where $0.2 \le \nu \le 0.3$ (Ref. 2). It is obvious that if all values of φ_j corresponding to $f_j = f(t_j)$ are known, then, introducing some a priori vector $N_d = \{N_d(r_1)$ for $l = 1,\ldots,m\}$ and writing Eqs. (6) in algebraic form, we arrive at a linear system of equations for the components $N_{dl}(l = 1,\ldots,m)$. Whether or not the system obtained, which can be regarded as the analog of the concept "information content," is well-conditioned depends on the variability of the scattering factor as a function of the parameter φ. This variability can be judged based on the total amplitude of the function $q(\varphi)$, i.e., from the value of the ratio $a_\varphi = \max\ q(\varphi)/\min\ q(\varphi)$. As f varies over range (0.7; 0.9) the value of α_φ reaches approximately 2.5. The quantity $\alpha_\lambda = (\max\ q(\lambda) / \min\ q(\lambda)$also reaches almost the same values as λ varies from 0.4 μm to 1 μm. This indicates that the variability of the scattering factor $K(\bar{m}(\varphi), \varphi r, \lambda)$ as a function of the parameters of the problem φ and λ is approximately the same. If, however, the presence of the factors φ_j^2 in Eqs. (6) is taken into account, the total amplitude of the function as a function of f should be much greater than its total amplitude as a function of the parameter λ, and this is confirmed by experimental studies.[8] At the same time, it should be kept in mind that if in a full-scale experiment it is possible to obtain complete optical information, i.e., the collection of data $\{\beta(\lambda_j, t_j)$, for $i, j = 1,\ldots\}$, then this should be done. The subsequent inversion gives the family of distributions $\{N(r, t_j) = N_j(r)\}$ and the corresponding intervals $\{R_j = [R_{ij}, R_{2j}]\}$ without any particular assumptions, for example, the equalities (4). Such microphysical information makes it possible to inverse problems for the equations of aerosol kinetics and to make a concomitant analysis of all physical processes determining the temporal variability of the spectrum over the of the experiment.[3]

In conclusion we point out the fact that the computational-analytical approach developed above to inverse problems of the real (interacting with air) aerosol can also be used in those cases when the aerosol system interacts with physical fields other than the moisture field. In particular, we can cite a polydispersed system of particles in a gravitational field. For this system the time variability of the size spectrum is determined by sedimentation. It is pertinent to note that the condensation particle growth, which was studied above, is necessarily accompanied by precipitation of large particles and, therefore, our constructions were of a somewhat idealized character. Let us assume that initially at the time $t = t_0$ the size spectrum is described by the function $s_0(r) = \pi r^2 n_0(r)$, where $R_{1,0} \le r \le R_{2,0}$, if it is assumed that sedimentation of particles started at this time, then the optical characteristic of the polydispersed system under study can be represented, to a first approximation, by the parametric integral

$$\beta(\lambda, t) = \int_{R_1}^{R(t_2)} K(r,\lambda) s_0(r) dr. \tag{12}$$

The application of this integral to the interpretation of optical data on the scattering of light by a liquid-drop aerosol, which were obtained in an artificial fog chamber, is given in Ref. 9. The unknown function $R_2(t)$ carries physical information about the state of the air. In particular, an empirical approximation of the form $\alpha t^{-1/2}$, where the coefficient α is determined by the kinematic viscosity of the air, can be used for $R_2(t)$.[2,9] As previously, we are interested in the inverse problem for the vector $\beta_t = \{\beta_j = \beta(t_j, \lambda)$, for $j = 1,\ldots, m\}$, which is solved in order to determine the function $R_2(t)$, i.e., to obtain physical information about the medium in which the aerosol system resides. To employ the standard algorithmic schemes in inverting the integral (12), for example, the method of linear systems, the method employed must be put into an appropriate analytical form. This can be done by introducing an auxiliary function $r(t, q) = R_1 + (R_2(t) - R_1)q$, where $0 \le q \le 1$. Denoting $(R_2(t) - R_1)$ by $\psi(t)$ we rewrite the integral (12) as follows:

$$\Psi(t)\int_0^1 K(\Psi(t),q,\lambda) s_0(\Psi(t),q) dq = \beta(\lambda, t). \tag{13}$$

Since $\psi(t)$ decreases monotonically as t increases the transformations $t \Rightarrow \psi$ and $\psi \Rightarrow t$ are one-to-one and therefore by fixing in the experiment the times j-th we obtain from Eq. (13) a sequence of equations for the numbers $\psi_j = \psi(t_j)$, namely,

$$\Psi_j \int_0^1 K(\Psi_j,q,\lambda) s_0(\Psi_j,q) dq = \beta_j \quad j=1,\ldots,m. \tag{14}$$

Equations (14) are analogous to equations of the type Eqs. (6) and can be studied and solved; numerically using the same methods as those described in detail above. By analogy to the preceding problem, the distribution $n_0(r) = n(r, t = 0)$ is assumed to be known. Another example of the possible use of the technique developed above for analyzing the inverse problems of aerosol optics and their numerical solution is the sounding of polydispersed liquid-drop aerosol interacting with a high-power radiation field.[10] Now the radius of the drop r is a complicated function of the time t and the initial radius $r_0 = r(t = 0)$. Here the function $r(t, r_0, E)$,where E is the energy characteristic of the field, can no longer be represented in the form $r = r_0\varphi(E, t)$. True, this is of no fundamental importance for the formulation and solution of inverse problems. Since it is possible to construct an inversion scheme based on iteration processes of the type (7b) a nonlinear integral equation of a general form, namely, an equation of the type Hammerstein equation of the first kind, must be solved numerically. The analysis of such inverse problems of aerosol optics for aerosol interacting with physical fields falls outside the scope of this paper.

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