

# Simultaneous determination of aerosol microstructure and refractive index from sun photometry data

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The inverse problem on simultaneous determination of microstructure characteristics and the refractive index of atmospheric aerosol from simultaneous measurements of the spectral transmission and solar aureole brightness is considered. To describe microstructure of the medium, integral functions of the particle size distribution were used. The similitude relationships have been derived, which the microstructure characteristics of scattering media having equivalent spectral extinction do obey. The algorithm of solving the inverse problem, based on minimization of the discrepancy between the measured scattering phase function and that calculated from the microstructure, retrieved from the inversion of the light extinction coefficient, is verified and tested in numerical experiments. The estimates of the error of determination of the real and imaginary parts of the refractive index are calculated, which do not exceed 0.05 and 0.03, respectively.

## Introduction

Determination of the aerosol disperse composition by optical methods requires, as a rule, that the refractive index of aerosol matter is known. Since the refractive index of the aerosol matter affects its scattering and absorption properties, it is natural to try to construct methods for its determination based on data of optical measurements. The data on the aerosol refractive index have independent interest, because these provide for information about its nature and physicochemical properties.

The purpose of this paper was to develop a technique for joint determination of the aerosol refractive index and disperse composition from measurement data on the spectral transmission and brightness of solar aureole using few *a priori* data on the solution sought. This paper continues the investigations,<sup>1,2</sup> where it was proposed to use the integral aerosol particle size distributions for microphysical interpretation of the optical measurements carried out by means of multi-wavelength sun photometers. In solving the above-stated problem, the approach was applied that earlier has been developed for inverting the angular measurements of the elements of the scattering phase matrix<sup>3</sup> based on the method of regularization for differential aerosol distributions. The advantage of using the integral aerosol distributions is the fact that in this case no restrictions are to be imposed on the smoothness of the distributions and no determination of the regularization parameter is needed. It is especially important in solving the inverse problems of aerosol light scattering using an approximately set operator, in which the approximate setting of the operator is caused by the errors in the refractive index of the particulate matter chosen. Another one advantage of the method proposed is the possibility

of using it in automation of the experimental data interpretation.

## 1. Peculiarities of interrelations between optical and microphysical properties of aerosol in the inverse problems of sun photometry

Let us consider the inverse problem on determination of the aerosol disperse composition from measurement data on the spectral dependence of the aerosol extinction coefficient

$$\varepsilon(\lambda) = \int_0^R K(\lambda, r) s(r) dr, \quad (1)$$

where  $K(\lambda, r)$  is the extinction efficiency factor depending on the complex refractive index  $m = n - i\kappa$ , which can be unknown,  $s(r) = \pi r^2 n(r)$ ,  $n(r)$  is the size distribution function of the particle number density. To understand specific features of such a problem, it is convenient to use the Hulst approximation<sup>4</sup> for description of the efficiency factor  $K(\lambda, r)$ , which was obtained for "soft" particles with the refractive index close to unity,  $|m - 1| \ll 1$  at  $kr \gg 1$ ,  $k = 2\pi/\lambda$  (anomalous diffraction). In reality, this approximation has wider boundaries of applicability and is quite suitable for describing the optical properties of aerosol particles in the atmosphere. According to Ref. 4, the factor  $K(\lambda, r, m) = K(\rho, \beta)$  is the function of two generalized parameters:  $\rho = 2kr(n - 1)$  and  $\beta = \kappa/(n - 1)$ . The parameter  $\rho$  characterizes the phase shift of the light wave passed through a spherical particle along its diameter.

For simplicity, let us consider first the case of non-absorbing particles ( $\kappa = 0$ ) with real part of the

refractive index  $n$ , which does not depend on the wavelength of radiation. Taking into account this assumption, let us write Eq. (1) in the following form

$$\int_0^R K[2kr(n-1)]s(r) dr = \varepsilon(\lambda). \quad (2)$$

Dependence of the factor  $K(\rho)$  on the particle size  $r$  and the refractive index  $n$  presented by the unified generalized parameter  $\rho = 2kr(n-1)$  makes it possible to determine the conditions, under which the spectral behavior of the extinction coefficient  $\varepsilon(\lambda)$  of two ensembles of particles with different refractive index and different disperse composition, is the same.

For example, let an aerosol medium, the optical-microphysical properties of which are characterized by the set  $\{m^*, s^*(r), \varepsilon^*(\lambda)\}$  satisfy Eq. (2). Then it follows from Eq. (2) that the equality  $\varepsilon(\lambda) = \varepsilon^*(\lambda)$  is valid for another medium with microphysical characteristics  $\{m, s(r)\}$ , if the distributions  $s(r)$  and  $s^*(r)$  have been related by the formula

$$s(r) = \eta s^*(\eta r), \quad (3)$$

where

$$\eta = (n-1)/(n^*-1). \quad (4)$$

Thus, two different ensembles of particles with the characteristics  $\{m, s(r)\}$  and  $\{m^*, s^*(r)\}$  related by the formulas (3) and (4) have the same spectral dependence of the extinction coefficient, and can be considered from this standpoint as optically equivalent. Obviously, one can find infinite number of ensembles of particles optically equivalent in this sense.

In making generalization to the case of absorbing particles, for keeping optical equivalence, the change of the imaginary part of the refractive index should correlate with the change of the real part by the rule

$$\kappa = \eta \kappa^*. \quad (5)$$

From the standpoint of solving the inverse problem, the consequence of above said is the impossibility, in principle, of unambiguously determining the distribution  $s(r)$  and the refractive index  $m$  only from the measured parameter  $\varepsilon(\lambda)$ . If a solution of such an inverse problem exists, it is not unique.

The transformation formula for the integral aerosol distribution

$$S(r) = \int_0^r s(r') dr'$$

has the form

$$S(r) = S^*(\eta r). \quad (6)$$

Formulas (3) and (6) describe transformations of the “stretching-compression” type and give simple explanation of the peculiarities of the transformation of the retrieved aerosol distributions at variations in the *a priori* selected refractive index, which were observed in numerical experiments.<sup>1,2</sup>

In particular, it follows from Eq. (6) that the value  $S = S(R) = S^*(\eta R)$  does not depend on the refractive index. So, in inverting the measured  $\varepsilon(\lambda)$  values, selection of the refractive index does not affect the value of the retrieved total geometric cross section of particles. Based on Eq. (6) and the property of monotonicity of the function  $S(r)$  at  $0 \leq r \leq R$ , one can also write the inequality

$$S(r) \leq S^*(r), \quad n < n^*, \quad (7)$$

which changes sign at  $n > n^*$ .

If one uses the size distribution of particle number density  $n(r)$ , instead of  $s(r)$ , for description of the disperse composition of optically equivalent media, then, analogously to Eq. (3), one can obtain the following relationship:

$$n(r) = \eta^3 n^*(\eta r), \quad (8)$$

from which, in particular, we obtain for the total number density

$$N = \eta^2 N^*. \quad (9)$$

It is seen from Eq. (9) that, on the contrary to the geometric cross section of particles  $S$ , the number concentration  $N$  of optically equivalent media changes depending on the ratio between real parts of the refractive index  $n$  and  $n^*$  proportionally to the square of the  $\eta$  value.

Finally, let us analyze the particle volume size distribution function  $v(r) = (4/3)rs(r)$ . Similarly to the previous cases, we have the following transformation formula for this function:

$$v(r) = v^*(\eta r), \quad (10)$$

and for the volume concentration

$$V = V^*/\eta. \quad (11)$$

It follows from Eq. (10) that “stretching” of the particle volume size distribution function  $v(r)$  along the positive direction of the abscissa axis occurs with decreasing the refractive index ( $n < n^*$ ). It leads to increase of the area under the curve  $v(r)$  proportionally to  $(1/\eta)$  value that agrees with Eq. (11), which determines the change of the total volume of particles. In contrast to the number density of particles  $N$  [Eq. (9)], which decreases with the decreasing refractive index, the total volume concentration  $V$  increases.

Taking into account the relation between the volume concentration  $V$  with the total geometric cross section  $S$ :

$$V = (4/3)\bar{r}_s S, \quad (12)$$

where  $\bar{r}_s$  is the mean particle radius over the  $s(r)$  distribution, and, taking into account the revealed constancy of the value  $S$  for the optically equivalent media, one can write the following relationship:

$$\bar{r}_s = \bar{r}_s^* / \eta. \quad (13)$$

It follows from Eq. (13) that, as in the case with the volume concentration, the decrease of the refractive

index of the particulate matter ( $\eta < 1$ ) causes redistribution of particles over size so that the mean particle radius  $\bar{r}_s$  increases.

Thus, we have revealed that spectral measurements of the extinction coefficient  $\epsilon(\lambda)$  contain insufficient information for simultaneous determination of the aerosol disperse composition and the refractive index, that leads to ambiguity of the solution. We have derived the similitude relationships, to which the microstructure characteristics of scattering media that have same spectral extinction at variations of the aerosol refractive index should satisfy.

Let us now address the question on what angular measurements of the solar aureole brightness can add under such conditions. According to Eq. (13), the mean particle radius  $\bar{r}_s$  in the reconstructed distributions depends on the refractive index selected. However, it is known that forward peakedness of the scattering phase function increases with the increase of the mean particle size. This gives grounds for expecting that the discrepancy between the measured scattering phase function and that calculated from the microstructure reconstructed by inverting the function  $\epsilon(\lambda)$  increases at deviation of the *a priori* set refractive index  $n$  from its true value  $n^*$ . In this case one can obtain the estimate of the refractive index  $n^*$  by minimizing the aforementioned discrepancy.

For example, let us consider reconstruction of the aureole scattering phase function in the Fraunhofer diffraction approximation

$$\mu(\theta) = \int_0^R K_\mu(\theta, r) s(r) dr, \quad (14)$$

where

$$K_\mu(\theta, r) = x^{(D)}(\theta, r) = \frac{1}{\pi} \left[ \frac{J_1(kr \sin \theta)}{\sin \theta} \right]^2 \quad (15)$$

is the normalized scattering phase function of an individual particle of the radius  $r$ ;  $J_1(\cdot)$  is the Bessel function of the first kind.

In this approximation, the scattering phase function  $\mu(\theta)$  does not depend on the refractive index of the particulate matter. This is true, if the particle size has not been related with their refractive index by a functional relation. However, if the function  $s(r)$  obtained by inverting the spectral dependence  $\epsilon(\lambda)$  at some value  $n$  has been taken as the aerosol distribution in the integrand of expression (14), one can show, based on Eq. (3), that

$$\mu(\theta) = \text{Ошибка! } \mu^*(\theta/\eta), \quad (16)$$

where the scattering phase functions  $\mu(\theta)$  and  $\mu^*(\theta)$  correspond to the distributions  $s(r)$  and  $s^*(r)$ . Equation (16) describes the changes in the scattering phase function reconstructed depending on the deviation of the refractive index  $n$  selected from its true value  $n^*$ . It follows from Eq. (16) that asymmetry of the scattering phase function  $\mu(\theta)$  monotonically increases with the decreasing refractive index  $n$

together with the increase of the discrepancy between the functions  $\mu(\theta)$  and  $\mu^*(\theta)$ . In particular, the ratio

$$\frac{\mu(0)}{\mu^*(0)} = \frac{1}{\eta^2} \quad (17)$$

at the point  $\theta = 0$  increases with decreasing  $\eta$  and, hence, with decreasing  $n$ . Due to continuity of the function  $\mu(\theta)$ , the increase of its forward peakedness should occur in some angular range  $0 \leq \theta < \tilde{\theta}$ . One can consider Eqs. (16) and (17) as equations for determining the true value of the refractive index  $n^*$  from measured scattering phase function.

The problem of determination of  $n^*$  is solved especially simply in the case of quite small scattering angles  $\theta$ , if one can use the following quadric approximation for the scattering phase function  $\mu(\theta)$  [Eq. (14)] with the kernel  $K_\mu(\theta, r)$  [Eq. (15)]

$$\mu(\theta) = \mu(0) - \alpha\theta^2. \quad (18)$$

In this case the value  $x = \eta^2$  can be determined from solution of the quadric equation for a specified scattering angle  $\theta$ :

$$(\alpha\theta^2)x^2 - \mu(0)x + \mu^*(\theta) = 0. \quad (19)$$

One can obtain another interesting relationship in the frameworks of the quadric approximation, by finding the point where  $\theta = \tilde{\theta}$ , i.e., the cross point of the curves  $\mu(\theta)$  and  $\mu^*(\theta)$ . It is easy to show that its position depends on the value  $\eta^2$  according to the formula

$$\tilde{\theta}^2 = \frac{\mu(0)}{\alpha} \frac{1}{1 + \eta^2}. \quad (20)$$

The value  $\eta^2$  can be determined from the value of the function  $\mu(\theta)$  at the point  $\tilde{\theta}$ :

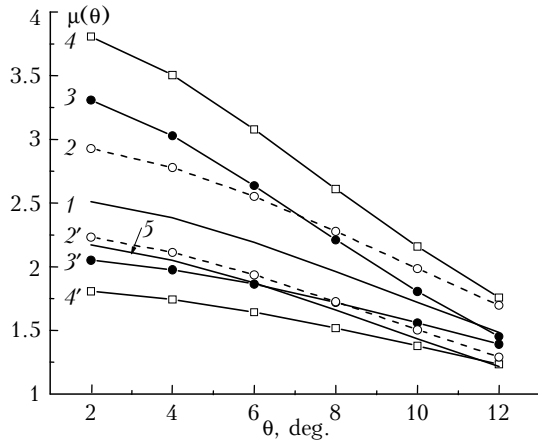
$$\mu(\tilde{\theta}) = \mu(0) \frac{\eta^2}{1 + \eta^2}. \quad (21)$$

This case only illustrates the extreme situation when the scattering phase function  $x^{(D)}(\theta, r)$  is independent of the refractive index  $n$ .

Another one characteristic case follows from the geometric optics approximation. In this case, as known,<sup>5</sup> the shape of the scattering phase function, on the contrary, does not depend on the particle size (if the absorption of light inside a particle is neglected) and is determined only by the refractive index. As is seen from the general formulas of geometric optics,<sup>5</sup> decrease of the refractive index in this case also leads to an increase in the asymmetry of the scattering phase function. An analogy is seen here with the effect of the refractive index of a lens on the structure of the light beam passing through it.

In the general case, when the kernel  $K_\mu(\theta, r)$  in Eq. (14) has been determined by formulas of the Mie theory,<sup>4-7</sup> the dependence of the scattering phase function on the refractive index of the particulate

matter can be calculated. For example, the behavior of the scattering phase functions at the wavelength  $\lambda = 0.31 \mu\text{m}$  for the models of submicron haze<sup>2</sup> with the parameter  $p = 0$  is shown in Fig. 1. The meaning of this parameter will be explained below.



**Fig. 1.** The scattering phase function for the model of microstructure with the parameter  $p = 0$ ; calculated data for the values of the refractive index  $n = 1.45, 1.35,$  and  $1.55$  at the same disperse composition (1, 2, 2'); calculated data for the extinction-equivalent distributions according to Eq. (3) at the exact value  $n_\mu = 1.45$  (3, 3'); total change of  $\mu(\theta)$  at  $n_\mu = n = 1.35$  and  $1.55$  (4, 4'); data calculated at  $n_\mu = 1.45$ ;  $\kappa_\mu = 0.05$  (5).

The scattering phase function  $\mu^*(\theta)$  (curve 1) corresponds to the aerosol model  $s^*(r)$  of the type "haze  $H$ "<sup>6</sup> with the refractive index  $n_\mu = 1.45$ . The index  $\mu$  at the refractive index means that the  $n$  value was used in calculation of the kernel  $K_\mu(\theta, r)$ . The imaginary part of the refractive index was assumed equal to zero. Curves 2 and 2' were calculated for the same model, but with other values of the refractive index, namely  $n_\mu = 1.35$  and  $1.55$ . It is seen from the data presented that the decrease of the refractive index causes monotonic increase of the scattering phase function in the aureole, i.e., the change has the same sign as in calculations using the geometric optics approximation.

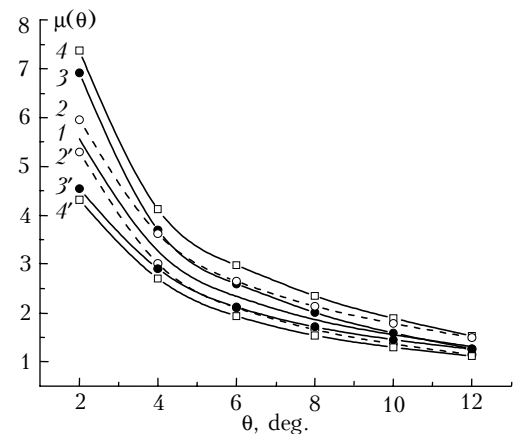
Transformation of the particle size distribution  $s(r)$  by Eqs. (3) and (4) coordinated with the change of the refractive index in inversion of the spectral dependence  $\epsilon(\lambda)$  causes additional transformation of the scattering phase function  $\mu(\theta)$ .<sup>14</sup> Such a transformation at the exact value  $n_\mu = n^* = 1.45$  yields  $\mu(\theta)$  functions shown in Fig. 1 by curves 3 ( $n = 1.35$ ) and 3' ( $n = 1.55$ ). Comparison of the curves 1, 3, and 3' shows an increase of the asymmetry of the scattering phase function occurring at decrease of the refractive index that agrees with the simultaneous increase of the mean particle radius  $\bar{r}_s$  according to Eq. (13).

It is important to emphasize that simultaneous effect of the refractive index in Eq. (14) on the kernel  $K_\mu(\theta, r)$  and the particle size distribution  $s(r)$  according to Eqs. (3) and (4) causes the change of

the scattering phase function  $\mu(\theta)$  of the same sign, which are then summed. Curves 4 and 4' in Fig. 1 give the idea of the total change of the scattering phase function under the effect of the aforementioned factors in the case of decreasing ( $n_\mu = n = 1.35$ ) and increasing ( $n_\mu = n = 1.55$ ) refractive index, respectively.

The effect of the imaginary part of the refractive index on the behavior of the aureole scattering phase function is illustrated by the curve 5 in Fig. 1 obtained at the exact values  $s^*(r)$ ,  $n_\mu = n^*$ , and  $\kappa_\mu = 0.05$ . It is seen that absorption of light leads to a systematic shift of the  $\mu(\theta)$  function. The same shift is observed at increase of  $n$  (compare with the curve 2'). This can be the reason of ambiguity in interpretation of the experimental data. So, for final estimation of the efficiency of determination of microstructure and refractive index from the measured parameters  $\epsilon(\lambda)$  and  $\mu(\theta)$  it is necessary to carry out numerical experiments on solving the inverse problem.

The regularities and tendencies observed in Fig. 1 are quite general and are reproduced for a wide set of aerosol models. To confirm this fact, the results obtained in the case of the model medium formed by two particle fractions, fine ( $f$ ) and coarse ( $c$ ), are shown in Fig. 2.



**Fig. 2.** The scattering phase function for the model of microstructure with the parameter  $p = 0.2$ ; calculated data for the values of the refractive index  $n = 1.45, 1.35,$  and  $1.55$  at the same disperse composition (1, 2, 2'); data calculated for the extinction-equivalent distributions according to Eq. (3) at the exact value  $n_\mu = 1.45$  (3, 3'); total change of  $\mu(\theta)$  at  $n_\mu = n = 1.35$  and  $1.55$  (4, 4').

The ratio between them can be regulated by means of the parameter  $p$ ,  $0 \leq p \leq 1$ , which determines the relative contribution of the coarse aerosol fraction to the total extinction coefficient  $\epsilon(\lambda)$  at the wavelength  $\lambda = 0.55 \mu\text{m}$ .<sup>2</sup>

## 2. Algorithm of solving the inverse problem

Based on the results obtained in the previous section, one can propose the following algorithm for simultaneous determination of the aerosol microstructure

and the refractive index from simultaneous measurements of the aerosol optical thickness and solar aureole brightness.

To describe the disperse composition of aerosol, let us use, as in Refs. 1 and 2, the integral distribution  $S(r)$ . Then, for the set of aerosol optical characteristics measured one can write the following system of operator equations:

$$Q_\alpha S = \alpha, \quad \alpha = \{\varepsilon(\lambda); \mu(\theta)\}. \quad (22)$$

The operators  $Q_\alpha$  depend continuously on the complex refractive index  $n - i\kappa$ , their form is determined by the kernels  $K_\alpha(r, n, \kappa)$  in Eqs. (1) and (14). Let us assume that the parameters  $n$  and  $\kappa$  are unknown and belong to some limited *a priori* defined set  $M$ , the elements  $m_0 = (n_0, \kappa_0)$  of which also belong to the set  $M$ . Let us also assume the existence of the exact distribution  $S_0(r)$ , which, as noted in Ref. 11, belongs to the set of monotonic bounded functions  $\Omega$ . It is known<sup>8</sup> that the set  $\Omega$  is compact in the space  $L_p[0, R]$ ,  $p > 1$ . Let us introduce the direct product of the sets  $U = \Omega \times M$ , the elements of which are pairs  $u = (S, m)$ ,  $S \in \Omega$ ,  $m \in M$ . The set  $U$  is compact because it is the product of compact sets  $\Omega$  and  $M$ .

If only approximate values  $\alpha_\delta$  are known in system (22) instead of the exact values of the right-hand parts  $\alpha_0$ , and these are such that  $\|\alpha_0 - \alpha_\delta\| \leq \delta_\alpha$ , one can select any element  $u_\delta \in U$  satisfying the condition

$$\|Q_\alpha(n, \kappa)S - \alpha_\delta\| \leq \delta_\alpha.$$

Let us denote the set of such elements  $U_\delta \subset U$ . This set is not empty, because it contains the exact solution of the problem  $u_0 = (S_0, m_0)$ . The following inequality is fulfilled for an arbitrary element  $u_\delta \in U_\delta$ :

$$\begin{aligned} \|Q_\alpha(n_0, \kappa_0)S_0 - Q_\alpha(n_\delta, \kappa_\delta)S_\delta\| &\leq \|Q_\alpha(n_0, \kappa_0)S_0 - \alpha_\delta\| + \\ &+ \|Q_\alpha(n_\delta, \kappa_\delta)S_\delta - \alpha_\delta\| \leq 2\delta_\alpha. \end{aligned} \quad (23)$$

As the elements  $u_\delta$  belong to a compact set, convergence of  $u_\delta$  to  $u_0$  at  $\delta_\alpha \rightarrow 0$  follows from inequality (23) and from the continuity of the inverse operator  $Q^{-1}_\alpha$  on the compact  $U$ . Thus, the problem on determination of the integral distribution  $S(r)$  and the complex refractive index of scattering particles  $n - i\kappa$  from the system of equations (22) is reduced to minimization of the functionals of discrepancy for these equations.

The considered approach to determination of the aerosol microstructure and refractive index is quite versatile in solving the inverse problems of aerosol light scattering and can be used for interpretation of joint measurements of different aerosol optical characteristics. Taking into account the specific features of the relations between optical and microphysical parameters in the problems of sun photometry mentioned in section 1, minimization of

the discrepancy functionals of the optical characteristics  $\alpha, \beta \in \{\varepsilon(\lambda); \mu(\theta)\}$  can be carried out separately for the variables  $S$  and  $m$  according to the following scheme.

Let us consider the solution  $\tilde{S}_\varepsilon$  reconstructed at minimization of the discrepancy functional of the extinction coefficient  $\varepsilon$  on the set  $\Omega$  at some point  $m \in M$ :

$$F_{\varepsilon\varepsilon}^2(S, m) = \|Q_\varepsilon(m)S - \varepsilon\|^2. \quad (24)$$

Obviously, the solution  $\tilde{S}_\varepsilon = \tilde{S}_\varepsilon(m)$  also is the function of the point  $m = (n, \kappa)$ . Let us determine the parameter  $\tilde{\mu} = Q_\mu \tilde{S}_\varepsilon$ . If the function  $\tilde{S}_\varepsilon$  approximates the exact solution  $S_0$  quite satisfactorily, one can say about transition from the parameter  $\varepsilon$  to the parameter  $\mu$ , which is performed by means of the operator  $W_{\mu\varepsilon} = Q_\mu Q_\varepsilon^{-1}$ .

The operator  $W_{\mu\varepsilon} = W_{\mu\varepsilon}(m)$  is defined on the set of measured functions  $\{\varepsilon(\lambda); \mu(\theta)\}$  and depends on the parameters  $n$  and  $\kappa$ . Using the operator  $W_{\mu\varepsilon}(m)$ , one can construct the discrepancy functional of the form

$$F_{\mu\varepsilon}^2(m) = \|W_{\mu\varepsilon}(m)\varepsilon_\delta - \mu_\delta\|^2 = \|\tilde{\mu} - \mu_\delta\|^2, \quad (25)$$

defined on the set  $M$  of the parameters  $(n, \kappa)$ . The functional  $F_{\mu\varepsilon}^2(m)$  [Eq. (25)] defines the difference between the experimentally measured scattering phase function  $\mu_\delta$  and  $\tilde{\mu}$  that is the same characteristics, but calculated using the integral distribution  $\tilde{S}_\varepsilon$  reconstructed by inverting the measured extinction coefficient  $\varepsilon_\delta$  values. In this case, one can consider the problem of estimation of the parameters  $n$  and  $\kappa$  sought as the problem on minimization of the functional  $F_{\mu\varepsilon}^2(m)$  [Eq. (25)] in the  $M$  domain.

Let us denote by  $\tilde{m} = (\tilde{n}, \tilde{\kappa})$  the point, at which the functional  $F_{\mu\varepsilon}^2(m)$  [Eq. (25)] reaches minimum on the set  $M$ . Then let us consider the set of parameters  $(\tilde{n}, \tilde{\kappa})$  and the function  $\tilde{S}_\varepsilon$ , bringing to minimum the discrepancy functional  $F_{\varepsilon\varepsilon}^2(S, m)$  [Eq. (24)] at  $m = \tilde{m}$  as a solution to the inverse problem on the aerosol optical characteristics  $\{\varepsilon(\lambda); \mu(\theta)\}$ . The efficiency of determination of the parameters  $(n, \kappa)$  and solution of the inverse problem, on the whole, depend on sensitivity of the discrepancy functional  $F_{\mu\varepsilon}^2(m)$  to their change in the vicinity of the exact solution. The results obtained by numerical simulation of the behavior of the functional  $F_{\mu\varepsilon}^2(m)$  under conditions corresponding to real observations by means of sun photometers for different aerosol models are presented in the next section.

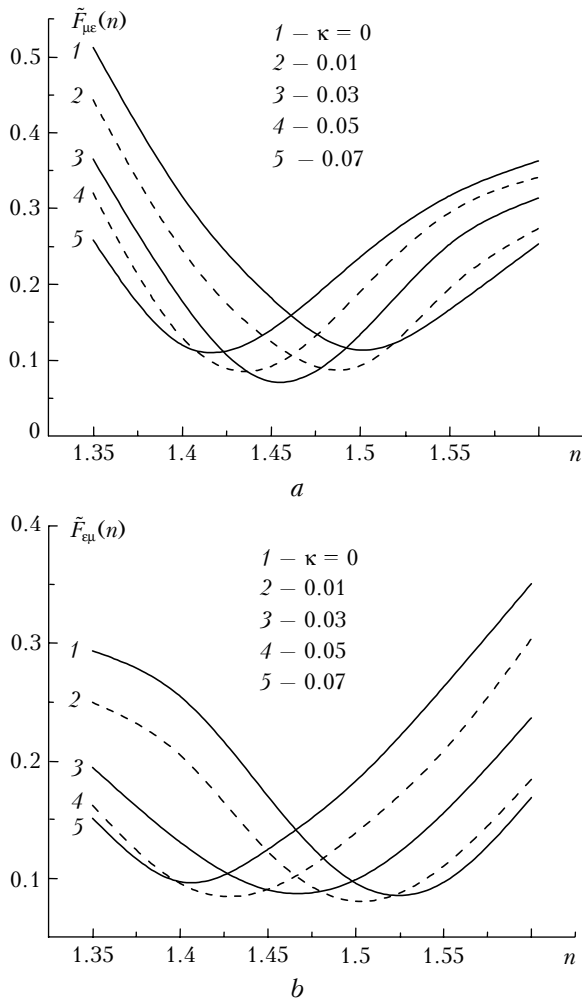
### 3. Results of numerical simulation

The optical and microphysical model of the medium and conditions of observations used in the numerical experiment presented below were exactly the same as in Refs. 1 and 2. Brief characterization of

the microstructure of the model medium has been given in section 1.

The values of the extinction coefficient  $\epsilon(\lambda)$  in the wavelength range  $[0.31, 4.0] \mu\text{m}$  and the scattering phase function  $\mu(\theta)$  at three wavelengths  $\lambda = 0.31, 0.4, \text{ and } 0.55 \mu\text{m}$  in the angular range  $2\text{--}12^\circ$  were calculated for the selected model of microstructure and the refractive index  $m_0 = 1.5 - i \cdot 0$ . Then these data, after adding a random error, served the input data for solving the inverse problem.

The dependences of the functional  $\tilde{F}_{\mu\epsilon}(n) = F_{\mu\epsilon}(n)/\|\mu\|$  for the model of microstructure at  $p = 0$  are shown in Fig. 3a.

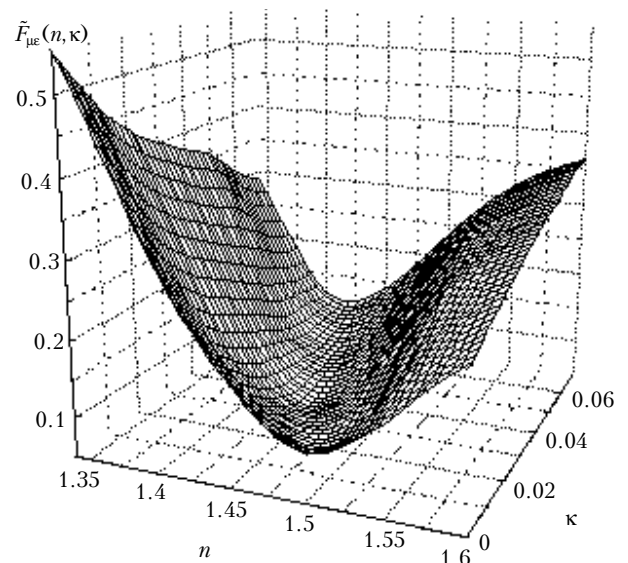


**Fig. 3.** Comparison of the discrepancy functionals  $\tilde{F}_{\mu\epsilon}(n)$  (a) and  $\tilde{F}_{\text{eq}}(n)$  (b) at  $p = 0$  and different values of the imaginary part  $\kappa$ .

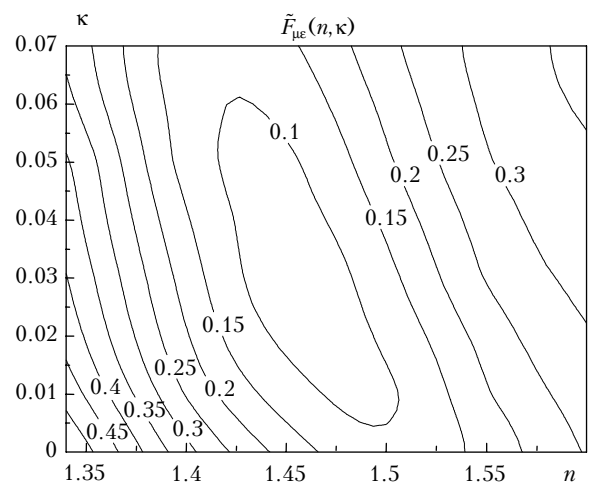
The error in the input data for the presented results was 10%. It is seen from Fig. 3a that the minimum of the functional  $\tilde{F}_{\mu\epsilon}(n)$  at a precisely set imaginary part of the refractive index ( $\kappa = 0$ , curve 1) is reached at the point  $\tilde{n} = n_0 = 1.5$ . The error in setting the value  $\kappa$  leads to the shift of the point of

the minimum of the functional  $\tilde{F}_{\mu\epsilon}(n)$  to the left. For example, at  $\kappa = 0.03$  we obtain  $\tilde{n} = 1.45$ , the point  $(\tilde{n} = 1.45; \tilde{\kappa} = 0.03)$  determines the global minimum of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  on both variables. Increasing the accuracy of determination of the refractive index is reached at introduction of *a priori* restrictions on the admissible range of variation of one of the parameters. For example, *a priori* set of the imaginary part of the refractive index with the error  $\Delta\kappa < 0.01$  causes restriction on the error in the reconstructed real part to be within the limits  $\Delta n < 0.015$ .

For clarity, the 3D surface of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  is shown in Fig. 4, and the map of isolines in the space of parameters  $n$  and  $\kappa$  is shown in Fig. 5.



**Fig. 4.** Surface of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  in the space of parameters  $(n, \kappa)$  for the model of the medium at  $p = 0$ .



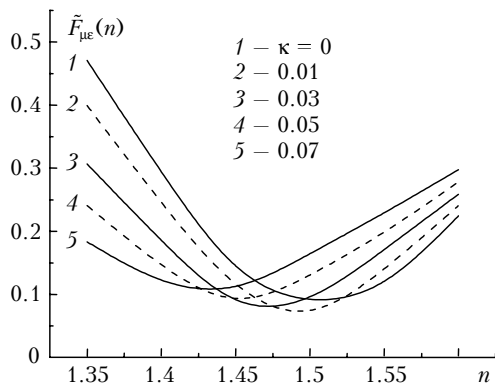
**Fig. 5.** Map of isolines of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  on the plane  $(n, \kappa)$  at  $p = 0$ .

The surface of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  has the shape of a ravine surface oriented mainly along the

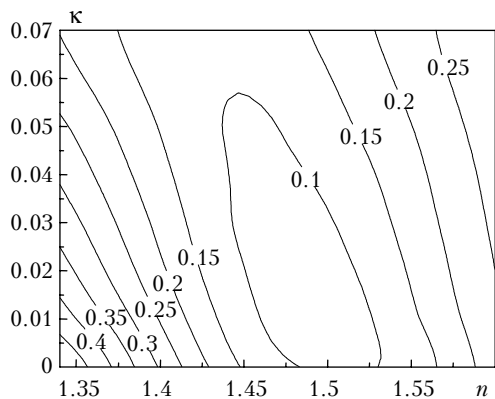
imaginary axis. Isolines in Fig. 5 determine the size of the uncertainty range of the estimate of the parameters  $n$  and  $\kappa$  at minimization of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  up to the selected level. The calculations show that the observed image will change inessentially if the scattering phase function is considered at a single wavelength.

If the sequence order of the characteristics  $\epsilon$  and  $\mu$  has been changed in the functional  $F_{\mu\epsilon}^2(m)$  [Eq. (25)], we obtain another functional  $F_{\epsilon\mu}^2(m)$  the behavior of which obviously changes. For a comparison, Fig. 3 shows the behavior of the functional  $\tilde{F}_{\epsilon\mu}(n) = F_{\epsilon\mu}(n)/\|\epsilon\|$ , dual relative to the functional  $\tilde{F}_{\mu\epsilon}(n)$ . As is seen from Fig. 3b, the functional  $\tilde{F}_{\epsilon\mu}(n, \kappa)$  keeps the extreme properties in the vicinity of the exact value of the refractive index, however, its surface has less steep slopes and narrower range of variations. Therefore the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  is less sensitive to variations of the parameters retrieved.

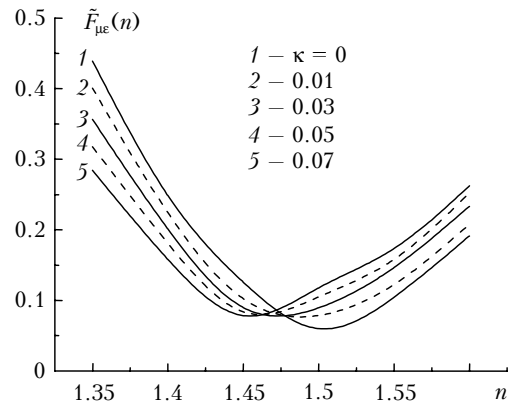
It is shown in Figs. 6–9 how the behavior of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  changes under the effect of variations of the model distributions caused by the change of the ratio between fine and coarse aerosol fractions. As the coarse fraction increases, the range of the values of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  changes weakly.



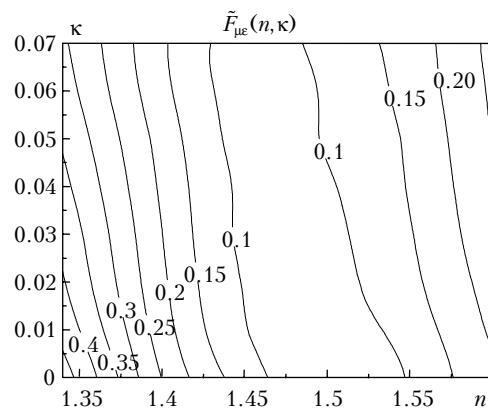
**Fig. 6.** Discrepancy functional  $\tilde{F}_{\mu\epsilon}(n)$  as a function of the real part of the refractive index  $n$  at  $p = 0.2$  and different values of the imaginary part  $\kappa$ .



**Fig. 7.** Map of isolines of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  on the plane  $(n, \kappa)$  at  $p = 0.2$ .



**Fig. 8.** Discrepancy functional  $\tilde{F}_{\mu\epsilon}(n)$  as a function of the real part of the refractive index  $n$  at  $p = 0.5$  and different values of the imaginary part  $\kappa$ .



**Fig. 9.** Map of isolines of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  on the plane  $(n, \kappa)$  at  $p = 0.5$ .

At the same time, the size of the range of uncertainty in estimation of the imaginary part of the refractive index  $\kappa$  increases and can exceed the size of the range of *a priori* admissible values. In this case, the experiment on determination of the value  $\kappa$  has low information capacity. This tendency is well observed in Figs. 8 and 9 for  $p = 0.5$ . In this example the level of the functional  $\tilde{F}_{\mu\epsilon}(n, \kappa)$  is lower than the error in setting the scattering phase function  $\delta = 10\%$  within the entire considered range of variations  $0 \leq \kappa \leq 0.07$  at the exact value  $n = 1.5$ .

### Conclusions

Analysis of the inverse problem of simultaneous determination of the size distribution function and the refractive index shows that, in the frameworks of the Van de Hulst approximation, the spectral dependences of the aerosol extinction coefficient do not provide for an unambiguous solution even if the measurement error is close to zero. The requirements are stated in this paper, and the similitude relationships are written, to which the ensembles of particles with different refractive indices and the disperse composition, optically equivalent in the

spectral extinction, should satisfy. In particular, it has been revealed that the mean radius of optically equivalent particles should increase with a decrease in the real part of the refractive index. The total geometric cross section of particles is kept constant, and the volume concentration increases. Hence, the noted factors lead to such a change of the shape of the scattering phase function in small angle range, at which the degree of asymmetry monotonically increases with the decrease of the refractive index of particles.

The obtained relations between optical and microphysical properties of the spectral and angular characteristics of light scattering of optically equivalent ensembles of particles enable one to propose and substantiate a technique for simultaneous reconstruction of the aerosol disperse composition and refractive index by minimizing the discrepancy between the measured scattering phase function and that calculated from microstructure reconstructed by inversion of the light extinction coefficient. The efficiency of the technique was numerically tested for a wide set of aerosol models. It was shown that simultaneous determination of the real and imaginary parts of the complex refractive index is possible with the errors  $\Delta n < 0.05$  and  $\Delta \kappa < 0.03$ , respectively.

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## References

1. V.V. Veretennikov, Atmos. Oceanic Opt. **19**, No. 4, 259–265 (2006).
2. V.V. Veretennikov, Atmos. Oceanic Opt. **19**, No. 4, 266–272 (2006).
3. V.V. Veretennikov, I.E. Naats, M.V. Panchenko, and V.Ya. Fadeev, Izv. Akad. Nauk USSR. Ser. Fiz. Atmos. Okeana **14**, No. 12, 1313–1317 (1978).
4. H.C. van de Hulst, *Light Scattering by Small Particles* (Wiley, New York, 1957).
5. K.S. Shifrin, *Light Scattering in a Turbid Medium* (ONTI, Moscow, 1951), 288 pp.
6. D. Deirmenjian, *Electromagnetic Waves Scattering on Spherical Polydispersions* (American Elsevier, New York, 1969).
7. C.F. Bohren and D.R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1983).
8. A.N. Tikhonov, A.V. Goncharkii, V.V. Stepanov, and A.G. Yagola, *Regularizing Algorithms and a priori Data* (Nauka, Moscow, 1983), 200 pp.