ON A SINGLE–PARAMETER MODEL FOR ATMOSPHERIC VARIABILITY OF ANGULAR FUNCTIONS OF AEROSOL LIGHT SCATTERING

R.F. Rakhimov and M.V. Panchenko

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
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We consider in this paper a single–parameter model for atmospheric variability of a scattering phase function of a near-ground haze. The domain of applicability of this model is considered as well. A role of random and regular geophysical factors in formation of the single–parameter model is analyzed based on model estimates. The analyzed model of correlation between aerosol light scattering characteristics is shown to correspond to the really observed wide variety of dispersion composition of the near-ground atmospheric haze, provided that optical constants of aerosol particles vary widely and the aerosol particle size spectrum changes characteristically with changing relative humidity of the atmosphere. Application of the single–parameter model is more justified for atmospheric situations, in which significant dynamics of the relative humidity is observed.

1. INTRODUCTION

The aerosol component in the atmosphere almost always is under influence of a complex set of geophysical processes. However, these processes occur randomly, and their duration is a random parameter too. Therefore, to estimate *a priori* light scattering parameters based on the methods of dynamic modeling of aerosol processes, the data of routine observations should be necessarily invoked. In this connection, a solution of the problem progressively shifts toward a study of statistical significance of different geophysical factors in variations of optical properties of the atmospheric aerosol. Dynamics of geophysical processes is so complicated that hardly can be modeled now. However, some stable regularities are observed in the correlation between the parameters to be predicted and some geophysical factors. A study of these regularities allows consideration of geophysical processes beyond the scope of purely theoretical analysis. The geophysical factors can be used as predictors of some change in optical properties of a dispersion component of the atmosphere and the efficiency of forecasts can be thus improved.

The problem of physical prerequisites for a single–parameter presentation of the atmospheric variability of angular functions of aerosol light scattering has been repeatedly discussed in the literature. In particular, this problem is considered in Ref. 1 with invoking model estimates. However, the number of model estimates is restricted by capabilities of computers, which are intensively developed in the last decades. In this paper, an effort is made to analyze the interaction between regular and random factors of variability of optical properties of the atmospheric haze.

2. STATISTICAL DESCRIPTION OF COEFFICIENTS OF DIRECTIONAL SCATTERING

According to the general scheme of empirical models construction,[2] any kth realization of a measured phase scattering function $g(\theta_k)$ or $g_\perp(\theta_k)$, $g_\parallel(\theta_k)$ can be presented as a random n-dimensional vector ($k = 1, \ldots, n$). In this case, a set of realizations of N such random vectors forms a statistical ensemble. Properties of this ensemble are described by the average values $\overline{g(\theta_k)}$, variance $D_g(\theta_k)$, rms deviation $\delta_g(\theta_k) = \sqrt{D_g(\theta_k)}$, and autocorrelation matrix $B_{gg}(\theta_k, \theta_l)$. Any random vector $g_i(\theta_k)$ from this ensemble can be approximated by some set of empirical orthogonal functions.

The first implementation of the similar scheme for studying statistical properties of angular dependence of scattering matrix components was the model described in Ref. 3 based on the results of optical measurements under continental conditions. In particular, it was shown that the lognormal presentation of the scattering phase function can be well approximated by the expansion in terms of the first eigenvector or by the regression dependence of the form

$$\ln[g(\theta, \lambda)] = k(\theta, \lambda) \ln\beta_\sigma(\lambda) + \ln[c(\theta, \lambda)], \quad (1)$$

where β_σ is the coefficient of aerosol light scattering; $k(\theta, \lambda)$ and $c(\theta, \lambda)$ are the parameters of the model.

A comparison between the results of measurements conducted at the Black Sea coast[4] and the continental data[3] has revealed (Table I) the similar, on average, transformation dynamics of angular functions of light scattering, even though the atmospheric haze formation is obviously specific in these regions.

External geophysical factors change the mesoscale structure of aerosol fields. These factors are nonregular,

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but steadily repeating. At the same time, the internal evolution of the dispersion system is governed by microprocesses. The lack of correlation between the external geophysical factors and the microprocesses evidently causes a statistical scatter of experimental points in regularities of correlation between integral and angular characteristics of light scattering. Thus, the identical regularities obtained for the two dissimilar regions with different mechanisms of the atmospheric haze formation clearly indicate a similarity in external factors, for example, stable diurnal variability of the relative air humidity.

TABLE I. Values of the empirical coefficients $k(\theta)$ for different sites.

θ°	Crimea, 1974–1977	Tomsk, 1975	Zvenigorod, 1971	Zvenigorod, 1972
5	0.22	−0.08	–	(3°)0.18
15	0.33	−0.21	0.34	0.25
20	0.31	0.18	0.33	0.18
30	0.16	0.12	0.22	0.06
50	−0.18	−0.05	−0.07	−0.12
70	−0.35	−0.18	−0.28	−0.26
90	−0.44	−0.26	−0.42	−0.36
110	−0.48	−0.35	−0.47	−0.44
120	−0.51	−0.38	−0.50	−0.47
140	−0.51	–	−0.48	−0.45
150	−0.52	−0.39	−0.46	−0.41
160	−0.52	−0.42	−0.46	−0.40
165	−0.52	−0.39	−0.47	−0.44

The numerical analysis of the information carried by the angular characteristics[5,6] shows that the magnitude of the directional scattering coefficient at $\lambda = 0.55$ μm and at the angle θ varying from 10 to 170° depends mostly on the contribution from the particles, whose radii are less than or comparable with the wavelength of the incident radiation. Therefore, the single-parameter model under discussion reflects predominantly the characteristic changes of submicron particles, which cause the atmospheric turbidity in the visible spectral range. This size region is just the region of interaction between fractions of the aerosol of primary and secondary formation. Consequently, some caution is required in extrapolation of the model to other spectral ranges because the size region of particles governing the angular light scattering diagram changes with the wavelength.

As to the lidar ratio, its values depend on variations of the relative humidity and local peculiarities of dispersion and chemical composition of coarse aerosols. Thus, there is a reason to expect ambiguous region-dependent dynamics of the lidar ratio.

3. PARAMETRIC DESCRIPTION OF MICROSTRUCTURAL CHANGES IN THE NEAR-GROUND ATMOSPHERIC HAZE

Available empirical data on the dispersion composition of the aerosol haze obtained mostly for the surface atmospheric layer are rather contradictory and can hardly be interpolated to other altitudes and climatic zones. Therefore, it is a problem to form an adequate ensemble of random realizations of the particle size spectrum in numerical simulation of statistical variety of the atmospheric haze states. On the one hand, the model must be as close to the real variety as possible; on the other hand, it may not be beyond the qualitative definition of the dispersion as the atmospheric haze.

Figure 1 shows a sample of some distribution spectra measured in a sufficiently wide size region. In particular, Fig. 1 demonstrates the aerosol size spectra observed in Ref. 7 (▼, ▽) and Ref. 8 (●, ❍), the model spectra (solid curves) recommended by the expert group[9] as the models for some typical aerosol formations, as well as the model estimates obtained in this work.

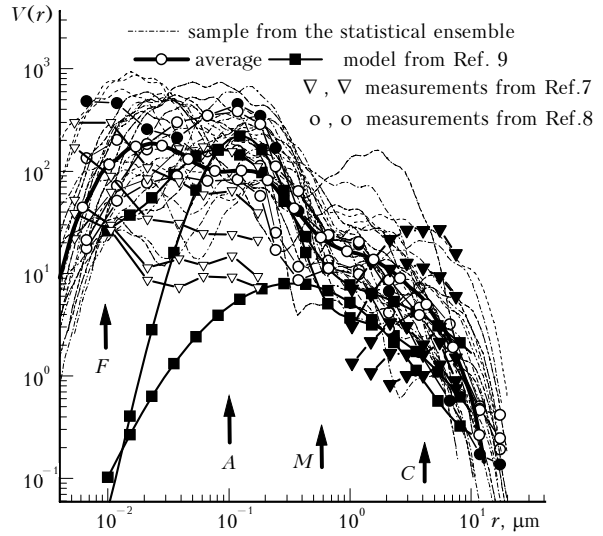

FIG. 1. Random sample of atmospheric haze particles size distribution from the universe (the parameters vary within the regions given in Table II) in comparison with the measured (Refs. 7 and 8) and model (Ref. 9) data for some typical formations.

As seen from Fig. 1, the variations in size distribution of the aerosol haze particles are significant and not correlated between different fractions. For example, both high and low number density of the coarse particles is observed for size spectra with high content of particles of accumulative fraction. This fact is indicative of relative independence of the processes governing the mechanisms of formation of the haze dispersion composition in different size regions. Therefore, in further model estimates we used the fraction-to-fraction method for parametrization of micro-structural changes. In particular, the variety of possible states of the atmospheric haze dispersion composition was modeled based on random and dynamic changes in the parameters of polymodal distribution.

At the first stage, the particle size spectrum was assumed consisting of four fractions (shown by arrows with letters in Fig. 1; the letters are abbreviations of the fractions: fine, accumulative, medium, and coarse) (Refs. 5 and 6):

$$f(r) = \frac{dN}{dr} = \frac{3v(r)}{4\pi r^3} = A^{(\nu)}\, r^{-\nu} \sum_{i=1}^{k} M_i^{(\nu)} \times$$

$$\times \exp\{- b_i\, [\ln(r/r_i^{(\nu)})]^2\}, \qquad (2)$$

where dN is the number density of aerosol particle with the radius varying from r to $r + dr$ in unit volume; $f(r)$ is the density function of the particle size distribution; $A_i^{(\nu)}$, $M_i^{(\nu)}$, b_i, and $r_i^{(\nu)}$ are the parameters of distribution modes.

Within the scopes of the qualitative definition of the dispersion as the atmospheric haze, an ensemble of random realizations of the size spectra was formed by randomly sampling the values of the model parameters (2) at $\nu = 3$ from the variation ranges presented in Table II. According to the qualitative definition of fraction elements, average, minimum, and maximum values for the regions of statistical variability of the following parameters are given here: $r_i^{(\nu)}$, b_i, and $F_i^{(\nu)} = A_i^{(\nu)}\, M_i^{(\nu)}$.

At the second stage, to provide qualitative variety of possible realizations of the spectra and to take the spectra with fine and irregular structure into the statistical ensemble, every random realization of the fractional spectrum (2) was replaced by a certain algorithm with optically equivalent (within 5–10% difference) superposition of lognormal modes. Every superposition consisted of 7 to 20 modes depending on relative positions of pre-separated fractions.

The initial static grid of lognormal modes is distributed uniformly over the logarithmic scale with the following parameters $b_k = 16$, $F_k = 1$, $r_{mk} = r^{(b)} (r^{(e)}/r^{(b)})^{k/28}$, where $k = 0, \ldots, 28$, at $\nu = 3$ hereinafter; $r^{(b)}$ and $r^{(e)}$ are the modal radii of the beginning and end elements of the grid, respectively.

Diurnal variations of relative humidity are considered as a regular (steadily and many times repeating) factor of microstructural transformation of the atmospheric haze.

Admittedly, the problem of parametrization of microstructural changes in the atmospheric haze in response to increasing relative humidity was widely discussed in the literature. In particular, systematic study of microstructural changes[10] led to the conclusion that the atmospheric haze responded to varying relative humidity, on the average, as the particles dispersion system containing both water soluble and insoluble ingredients.

Theoretical estimates of an equilibrium size of an aerosol particle in the humid atmosphere logically stem from the Wincler–Junge–Hanel model conception of mixed nuclei. Most models based on this conception use the well-known expression for partial pressure of water vapor on a partially soluble spherical particle[11]:

$$e_r = e_0(T) \exp\left[\frac{2\sigma_p}{\rho R T r}\right]\left(1 - \frac{A m_s}{r^3 - r_T^3}\right), \qquad (3)$$

where e_r and $e_0(T)$ are the partial pressure of water vapor on a spherical particle with radius r and the pressure on a plane surface at the temperature T, respectively; σ_p and ρ are the surface tension coefficient and the solution density; m_s is the mass of the soluble part; r_T is the effective radius of an insoluble particle. The parameter A characterizes the pressure lowering due to dissociation, hydration, and other solution effects. In the case of the phases thermodynamic equilibrium, the ratio $e_r/e_0 = q$ corresponds to relative humidity of the medium.

TABLE II. Values of the parameters of the particle size distribution function (2).

Parameters of a distribution mode (2)	Value			Distribution (type)
	Average	Minimum	Maximum	
	Fine fraction (FF)			
r_1, μm	0.015	0.003	0.026	uniform
b_1	1.64	1.29	2.01	"
F_1	62.0	17.3	227.3	normal
	Accumulative fraction (AF)			
r_2, μm	0.090	0.027	0.182	uniform
b_2	1.24	1.08	1.430	"
F_2	50.0	12.07	223.3	normal
	Medium fraction (MF)			
r_3, μm	0.490	0.266	1.2	uniform
b_3	1.43	1.15	1.77	"
F_3	2.6	0.5	14.0	normal
	Coarse fraction (CF)			
r_4, μm	3.9	1.2	6.10	uniform
b_4	1.84	1.21	1.99	"
F_4	0.54	0.04	5.43	normal

However, this approach cannot be directly used for prediction of microstructural responses of the atmospheric haze to variations of relative humidity because of an ambiguous *a priori* choice of values of the parameters A, m_s, and r_T for different fractions, which determine the physical and chemical individuality of some polydispersion mixture. Just this problem has initiated the development of a number of principally related theoretical models,[10–16] which can be generalized by the relation

$$r = U r_0^{\delta}, \qquad (4)$$

where r and r_0 are the radii of humid and dry particles; U and δ are the functions of the relative humidity q with particular values depending on the composition of the medium.

Table III gives some examples of U and δ functional dependence used in the model estimates.

TABLE III.

Author	Expressions for U and δ	Eq.
Hanel, Ref. 10	$U = \left\{1 + \frac{\rho_d}{\rho_w}\,\mu(q)\,\frac{q}{1-q}\right\}^{1/3}$, $\delta = 1$	(5)
Prishivalko, Ref. 12	$U = \left\{1 - G + \frac{\beta}{1-q}\right\}^{1/3}$, $\delta = 1$	(6)
Casten, Ref. 13	$U = (1-q)^{-\gamma}$, $\delta = 1$	(7)
Wells, Ref. 14	$U = \{1 - C_1 \ln(1-q)\}$, $\delta = 1$	(8)
Tuomi, Ref. 15	$\ln\delta = \frac{\alpha q}{(\zeta - q)}$; $\ln U = \frac{\xi q}{(\phi - q)}$	(9)

Note. ρ_d and ρ_w are the densities of a dry residue and the water; $\mu(q)$, G, q_1, α, β, γ, ζ, ξ, and ϕ are the particular parameters depending on the relative portion of a soluble substance and the water activity of the compounds entering the composition of aerosol particles.

The relative humidity is a factor regulating not only heterogeneous condensation processes, but also the entire complex of secondary processes determining the stage of aerosol phase synthesis within the atmosphere, in particular, heteromolecular processes.[4,16] The mechanism of coagulation joining aerosol particles, in its turn, activates the process of atmospheric humidity condensation due to formation of micropores at sticking together of two or more coagulants. A fraction-by-fraction simulation of the process becomes logical and necessary in view of an ambiguous response of aerosol formations to changes in the relative atmospheric humidity and wide variety of their possible initial states.

Since the process of an aerosol component adaptation to variations of the relative humidity is fast and accompanying other processes, deformations of the size spectrum were simulated taking into account the generalized data of direct microphysical measurements.[17] Besides, the correction for changes in values of the integral characteristics of the dispersion structure $S_i(q)$ and $V_i(q)$ was introduced separately for every fraction of the atmospheric haze.

Another important factor regulating the statistical variety of optical properties of the atmospheric haze is the complex refractive index. Changes of the refractive index of aerosol matter from one realization of chemical composition of the aerosol haze to another were also simulated using the fraction-by-fraction method in numerical estimates. Characteristic intervals of variations were stipulated both by regional peculiarities in formation of chemical composition of the haze fractions and by possible variations of the relative humidity.[10]

First attempts to take into account the variations of $m = n - i\varkappa$ in response to the relative humidity were undertaken by Volz in Ref. 18. Considerable study has been given to this problem by Hanel (Ref. 13). Applying the Dail—Glodstone rule, which determines the refractive index for slightly absorbing materials, and the functional dependence (4) for estimation of the water percentage, Hanelin has proposed the following relation in his model estimates[10]:

$$m = m_w + (m_d - m_w)\left\{1 + \frac{\rho_0}{\rho_w}\,\mu(q)\,\frac{1}{1-q}\right\}, \qquad (10)$$

where m_d and m_w are the values of m for the dry residual and the water, respectively.

However, in our opinion, the method based on *a priori* analysis of spectra of the molar absorption coefficients $\varkappa(\lambda)$ for different components of an aerosol matter seems more justified for model estimation of the spectral dependence of m. This method allows the synthetic dependence $\varkappa(\lambda)$ to be computationally constructed based on a particular chemical composition with regard to probable variation of the water content in the dispersed phase. The dependence obtained in such a way is more strict (especially, near absorption bands of the aerosol matter). With $\varkappa(\lambda)$ known in a wide spectral range, the spectral dependence of the real part $n(\lambda)$ is usually calculated using the classic Kramers—Croning dispersion relations.[19]

Different chemical composition of primary and secondary aerosols causes an ambiguity of not only hygroscopic properties, but optical constants as well. In our model estimates, the optical constants of particles of the accumulative and coarse fractions were determined separately following the first and the second versions of the synthetic model of the complex refractive index.[20] The versions were developed with regard for chemical composition of the continental haze. Besides, they used the data from Ref. 21 to determine optical constants for particles with sulfuric acid dominant having regard to their variations due to different water content.

4. DISCUSSION OF OBTAINED MODEL ESTIMATES

As a first step, we have considered the data of the numerical experiment. The ensemble of states for the experiment resulted from uniform filling the variability regions of microstructural parameters given in Table II. As expected, in this case the calculated results for most scattering angles have shown only a blurred cloud of points between the normalized scattering phase function and the volume scattering coefficient on the correlation diagram (Fig. 2).

This statistical set of realizations was formed based on the hypothesis of sufficiently wide variations of parameters in the distribution (1). The estimates have shown that under such neutral assumptions on possible variations in dispersion composition of the atmospheric haze, a somewhat stable regulation in mutual dynamics of the scattering coefficient and the normalized scattering phase function can be considered only conditionally, for example, at angle ranges from 5 to 10 and from 160 to 180°. At other scattering angles, an unambiguous tendency to intercorrelation can hardly be found, because $k(\theta)$ for these angles is close to zero.

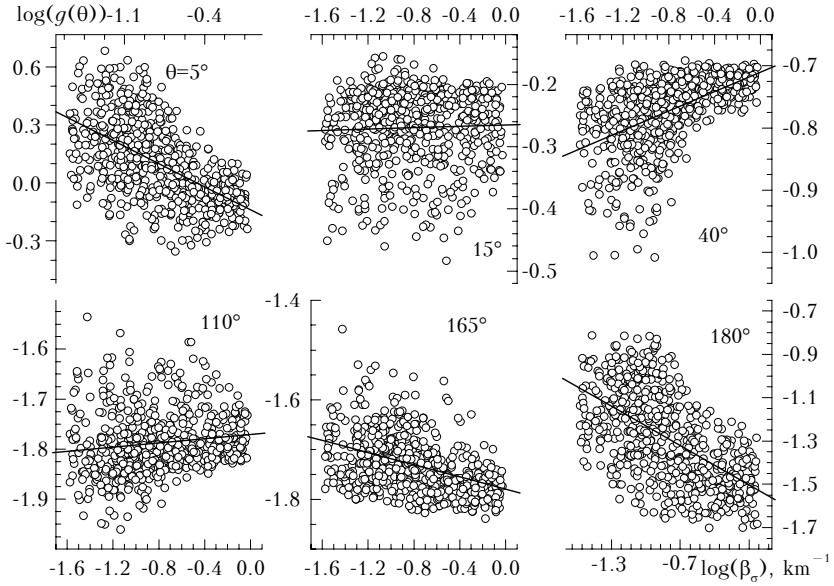

FIG. 2. Diagrams of correlation between the aerosol scattering coefficient and the normalized scattering phase function at angles of 5, 15, 40, 110, 165, *and* 180° *at random changes of the size spectrum parameters (2).*

Nevertheless, the attempt to approximate the slightly correlated set of points by the dependence (2) has revealed the analogy of angular dependence for the coefficient $k(\theta)$ (curves *1* and *6* in Fig. 3), which is rather close to the empirical model.

Curve *2* in Fig. 3 was obtained on the assumption that the optical constants of aerosol particles vary simultaneously with the dispersion composition of the atmospheric haze. These variations are assumed random and independent in every fraction. In particular, the complex refractive index varies from $m_u = 1.65 - i\ 0.009$ to $m_l = 1.34–0.001$. Curve *3* in Fig. 3 is obtained under the same assumption as the curve *2*, but variations of parameters F_i in this case are not neutrally uniform over the region but centered, although with sufficiently wide distribution about the mean value.

To simulate the normal distribution, we used sufficiently popular method of transformation of a pair of independent values ξ_1, ξ_2 uniformly distributed over the interval (0,1) into a pair of normally distributed independent values:

$$\eta_1 = \sqrt{-\ln\xi_1 \cdot \ln\sigma}\ \cos 2\pi\xi_2, \qquad (11)$$

$$\eta_2 = \sqrt{-\ln\xi_2 \cdot \ln\sigma}\ \sin 2\pi\xi_1. \qquad (12)$$

In this method, we can regulate a scatter of η_1 and η_2 values about the center of the variability range by changing $\ln\sigma$. In particular, $\ln\sigma = 2$ for curve *2* in Fig. 3. Curves *3–5* just illustrate the possible transformation of the angular dependence $k(\theta)$ at narrowing the statistical scatter of F_i values about the mean value at $\ln\sigma = 0.7$, 0.5, and 0.35, respectively.

Besides, it should be noted that just the assumption of statistical variations presence in the refractive index of aerosol particles gives grounds for bridging the gaps between the theoretically calculated curves and empirical data in Fig. 3. In particular, the model estimates show that in the absence of variations in the complex refractive index, a narrowing of the statistical variations of the F_i parameters alone does not imply the corresponding narrowing between the theoretical and empirical data. The angular dependence $k(\theta)$ in this case differs insignificantly from the curves *1* and *2*. At the same time, the calculations show that further narrowing of the variability region of the F_i parameters is undesirable, because the ensemble of $n(r)$ realizations losses its statistical variety and becomes incapable to cover the really observed variety of size spectra.[7–9]

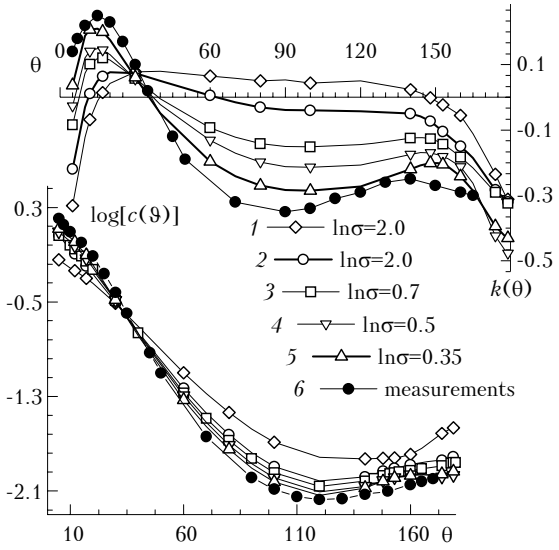

FIG. 3. Angular dependence of the coefficients $k(\theta)$ and $c(\theta)$ of the regression model (1) at narrowing the variability region of the F_2 model parameter in comparison with the data of the empirical model from Ref. 3.

Then we have conducted a numerical experiment with the results shown in Fig. 4. The variety of the size spectrum formed in the experiment was similar to that shown by curve *3* in Fig. 3.

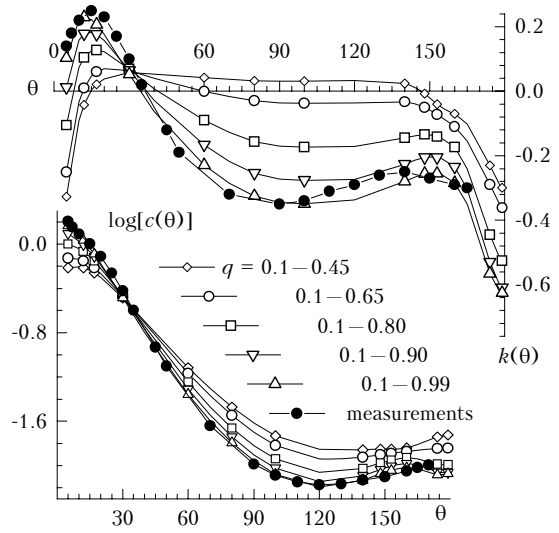

FIG. 4. Angular dependence of the coefficients $k(\theta)$ and $c(\theta)$ of the regression model (1) at widening the variability region of the relative humidity.

In addition, a random value of the relative humidity was modeled for every realization of $n(r)$. According to the technique described above, both the size spectrum and the refractive index for every fraction were corrected for the state of humidity. The variability range of the relative humidity was gradually widened (see the dynamics of curves *1–5* in Fig. 4).

Due to correction of a random realization of the size spectrum for a particular value of the relative humidity, the variability range of the aerosol particle size distribution function has been widened about four to five times. This has allowed the statistical variety of the ensemble of the $n(r)$ spectrum realizations to be adequate to field measurements. Curve *1* in Fig. 4 is calculated for narrow variability range of the relative humidity q from 0.1 to 0.45. This assumption was made in order to estimate the specificity of variations in optical properties of the atmospheric haze at relatively stable dry atmosphere as a close analogy of the arid climatic zone.

Curves *2–5* illustrate possible dynamics of the shape of the $k(\theta)$ dependence in response to increasing range of q variability up to 0.99. As seen from the obtained estimates, just the agreed changes in the optical constants and the particle size spectrum of the atmospheric haze due to stable diurnal variations of the relative humidity are most likely the main reason of formation of the angular dependence $k(\theta)$ found experimentally. In contrast to $k(\theta)$, different versions of the angular dependence $c(\theta)$ indicate that this model coefficient depends on the values of the microstructural parameters m_i, r_i, b_i, and F_i, averaged over the ensemble of realizations, rather than their variability ranges.

Figure 5*a* shows a random sample from the universe of realizations. This figure illustrates a change in the shape of the angular dependence of the normalized scattering phase function at only random changes of the particle size spectrum of the atmospheric haze. This means that curves in Fig. 5*a* were calculated based on the microstructural data similar to those used for calculation of curve *2* in Fig. 3. The data shown in Fig. 5*b* are similar to those shown in Fig. 5*a*, but they were obtained based on the additional assumption of varying relative humidity. The microstructural parameters of size spectrum and the refractive index of haze particles were simulated varying in response to variations of the relative humidity. Thus, the microstructural data used as initial for Fig. 5*b* are similar to those used for calculation of curve *5* in Fig. 3.

As seen from the above data, the amplitude of variations of the normalized scattering phase function increases with increasing relative humidity. Besides, of interest is the fact that the tendency to the scattering phase function variation is different for different angles. Thus, for angles from 20 to 145° the tendency to changing the scattering phase function is sufficiently stable and mutually agreed. At the same time, a variation of the relative humidity may result in irregular behavior of $g(\theta)$ with increasing turbidity near the halo, as well as at angles close to lidar ones.

To study possible reasons for formation of a particular shape of the angular dependence $k(\theta)$, model estimates have been performed. Empirical data to be compared with these estimates might be obtained from corresponding experiments in other regions. A particular attention in the model calculation was paid to variations of the microphysical parameters of the atmospheric haze and, first of all, its accumulative fraction.

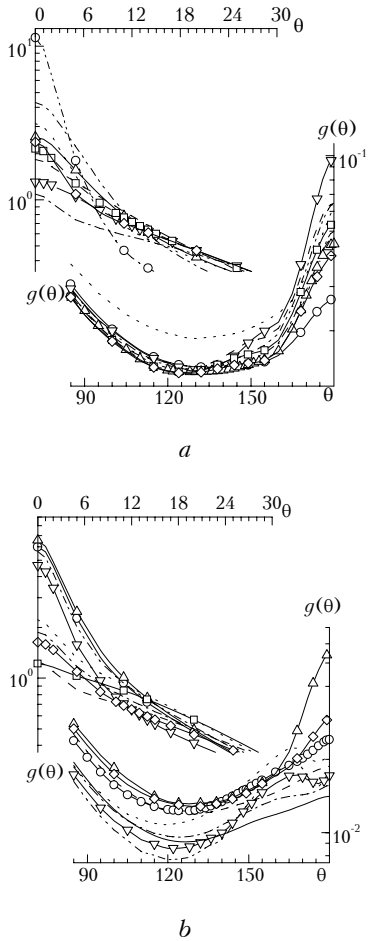

FIG. 5. Change in the shape of the angular dependence of the normalized scattering phase function: at random variations of the size spectrum neglecting possible variations of the relative humidity (a); at random variations of the relative humidity superimposed onto the statistics of random variations of the size spectrum (b).

In particular, a change in the shape of the $k(\theta)$ function was considered in relation to a shift of the distribution mode of the accumulative fraction along the size scale. The results of calculations are shown in Fig. 6 as dependence of $k(\theta)$ on the modal radius. The similar results are shown in Figs. 7 and 8, but as the dependence of $k(\theta)$ on the F_2 and b_2 parameters of the model (2), respectively. (The F_2 parameter describes the total content of particles of the accumulative fraction, while the b_2 parameter describes the statistically average width of the distribution mode.)

As follows from the model estimates shown in Figs. 6–9, the most significant factor responsible for the shape of the angular dependence $k(\theta)$ at the wavelength $\lambda = 0.55$ μm is the statistically average size r_2 of particles of the accumulative fraction. Although variations of the b_2 and F_2 parameters may lead to minor changes in $k(\theta)$ at the angles from 20 to 40 and from 90 to 165°, they do not change the shape of the angular dependence as a whole.

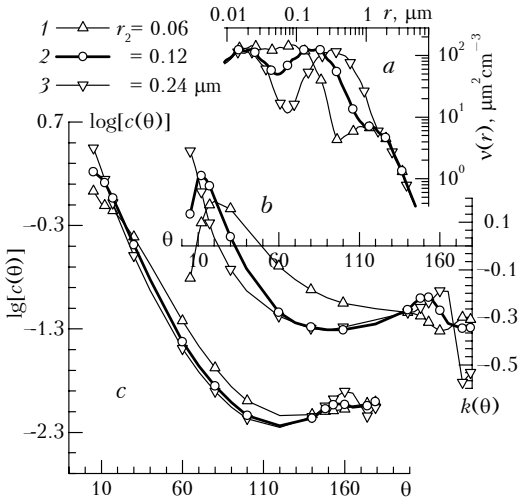

FIG. 6. Change in the shape of the angular dependence $k(\theta)$ (b) and $c(\theta)$ (c) at different values of the modal radius r_2 in Eq. (1) at $\nu = 3$ in the statistically average particle size spectrum of the accumulative fraction (a).

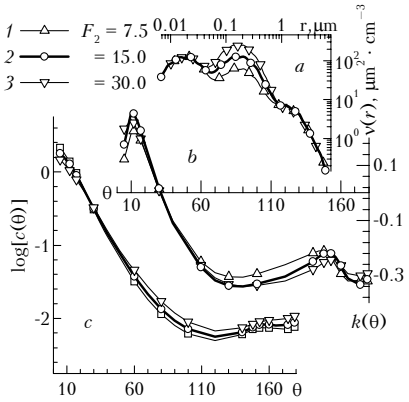

FIG. 7. Change in the shape of the angular dependence $k(\theta)$ (b) and $c(\theta)$ (c) at fourfold change of the statistically average value of the F_2 parameter of the particle size spectrum of the accumulative fraction (a).

The corresponding numerical experiments, performed for other fractions at the same wavelength, have shown that the influence of variations of size spectrum parameters on deformation of the $k(\theta)$ shape is of the second order of significance as compared to the accumulative fraction. For example, more than tenfold change in the mean content of the coarse fraction leads to only minor changes in the expansion coefficients in the halo region at angles close to the lidar ones (see Fig. 9).

The model estimates shown in Figs. 6–9 give grounds to expect that the dynamics of the aerosol scattering phase function, as well as that of the lidar ratio, is most complicated and region–dependent near the lidar angles.

Thus, it is seen from the numerical analysis of the model (1), that two factors can be separated out in the statistics of long-term atmospheric variability of aerosol light scattering parameters.

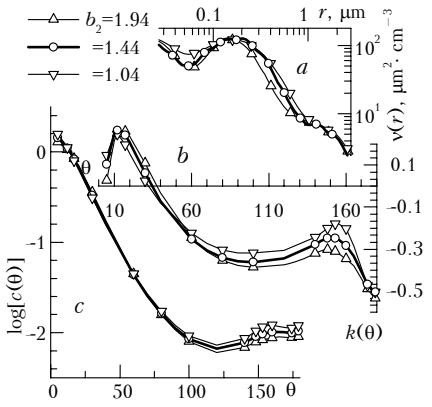

FIG. 8. Change in the shape of the angular dependence k(θ) (b) and c(θ) (c) at different values of the b_2 parameter, which describes the distribution of particles of the accumulative fraction in the statistically average size spectrum (a).

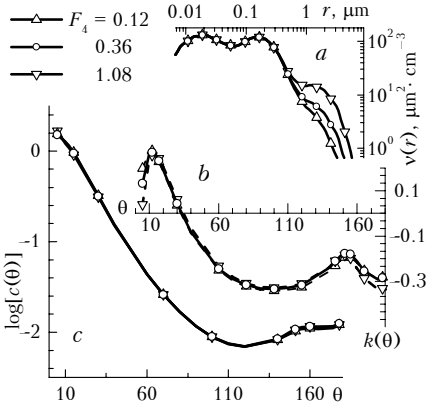

FIG. 9. Change in the shape of the angular dependence k(θ) (b) and c(θ) (c) at different values of the F_4 parameter of the coarse fraction in the statistically average size spectrum (a).

The first factor is sufficiently fast and local response of a dispersion system to changes in the relative air humidity. The air humidity, in its turn, depends on a particular hygroscopic activity of fractions and quantitative proportion between them, that is, on dispersive-chemical composition of the medium.

The second factor to be noted is geophysical processes, which govern the local statistics of variations of the relative humidity itself. The statistics of variations in the microstructural aerosol parameters is connected with geophysical factors by a weak cause-and-effect relation. Since the length of an observation series usually far exceeds the mean lifetime of the aerosol in a layer, the observed data characterize a wide variety of different synoptic situations. Besides, a wide spatial variety of distribution of aerosol fractions is observed in air masses of different types. Considering these facts, it is easy to understand why all attempts to find an universal empirical dependence between the integral light scattering parameters and the relative humidity failed. At the same time, such dependence can be sometimes found in individual measurement series. Some authors even noted a significantly irregular change of the aerosol light scattering coefficient and the relative humidity.[23,24]

As seen from the obtained model estimates, the view of relation between optical parameters in the model (1) may sometimes reflect a particular statistics of microstructural changes in aerosol due to effect of some geophysical factors, rather than a hygroscopic response of a particular dispersion composition of aerosol fractions to changes in the humidity.

Thus, our analysis of atmospheric variability in angular functions of aerosol light scattering has revealed a statistical set of partially related values of the microstructural parameters m_i, r_i, b_i, and F_i of the near-ground atmospheric haze. This statistical set was used as a basis of the numerical experiment aimed at prediction of spectral variability of the model. A possibility of the model extrapolation to other wavelengths was also analyzed. An analysis of correlation between other parameters of aerosol light scattering, including those, which can hardly be measured directly, was based on this statistical set as well. The results of model estimates are shown in Figs. 10–15.

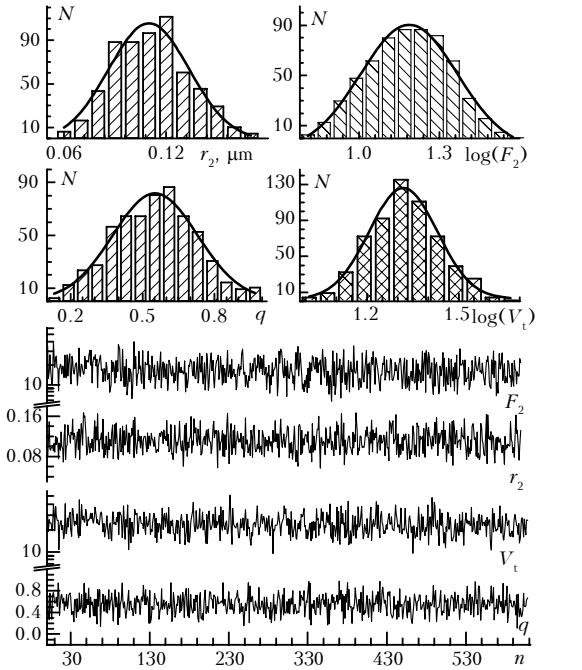

FIG. 10. Diagrams of the repetition rates of values of the microstructural aerosol parameters and the relative humidity and their change from one realization to another within a given statistical ensemble. The results were obtained at the same assumptions as curve 5 in Fig. 3.

Besides, it should be noted that the statistics of realizations of the microstructural parameters (except the F_i parameter) and the relative humidity were initially set as lognormal distribution at the linear scale (see Fig. 10). Nevertheless, the statistical ensemble of the values to be

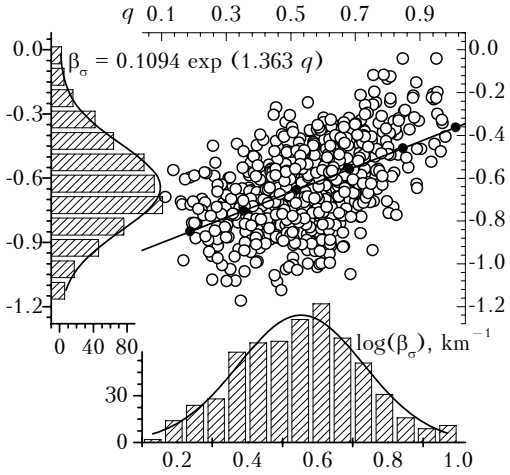

FIG. 11. Diagram of correlation between the aerosol scattering coefficient β_σ *and the relative humidity q, as well as their distribution over the variability ranges. The statistics of variations of the microstructural parameters was taken the same as for curve 5 in Fig. 3.*

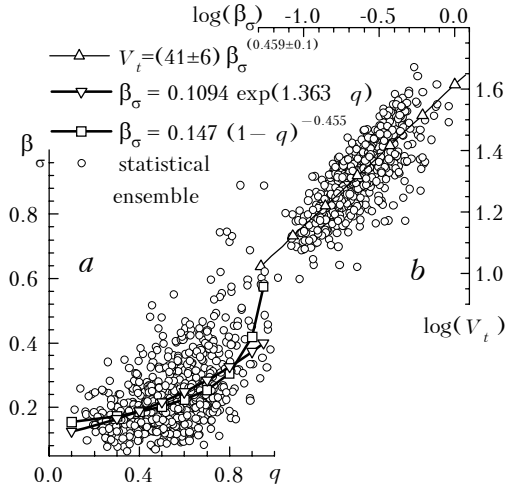

FIG. 12. Diagram of correlation between the aerosol light scattering coefficient β_σ *and the relative humidity q on the linear scale (a) and between the aerosol light scattering coefficient* β_σ *and the total volume* V_t *of particles of all aerosol fractions (b). The initial data were taken the same as for Fig. 11.*

predicted (see Figs. 11–13) proves to be closer to the lognormal law, which was used for determination of the statistics of F_i values. A particular view of statistical distribution of the meteorological and microstructural parameters may control not only the statistics of distribution of the light scattering parameters, but the shape of the statistically average regularity of their atmospheric variability as well.

In some papers, the statistically average relation between the aerosol light scattering coefficient and the relative humidity is analyzed on the linear scale of β_σ variations. However, on the logarithmic scale the statistical ensemble of points is distributed more uniformly along the approximating curve (see Fig. 12), although having almost triangular shape on the linear scale. Therefore, it makes sense to consider this relation on the logarithmic scale.

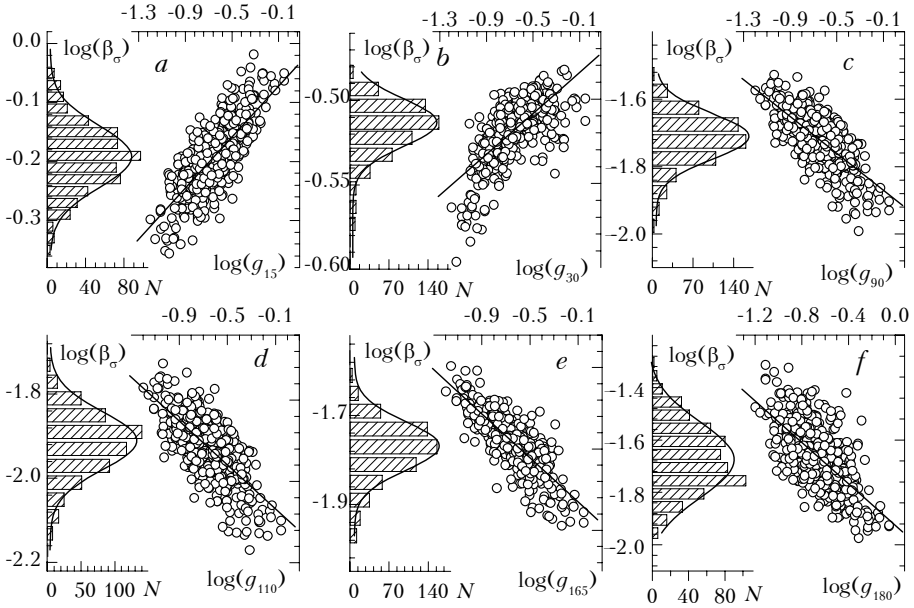

FIG. 13. Diagrams of correlation between the aerosol light scattering coefficient and the normalized scattering phase function at an angle θ = 15; 30; 90; 110; 165; 180°, *as well as repetition histograms for values of these parameters.*

In Figs. 12 and 13 one can notice a scatter in values of the parameters analyzed. However, the model estimates do allow some statistical regularities to be found in correlation between the aerosol light scattering coefficient β_σ and the total volume V_t of aerosol particles of all fractions at variations of the relative humidity q. In particular, a blurred spot of points with a correlation coefficient equal to 0.531 is characteristic for the scattering coefficient and the relative humidity. The relation between them can be approximated by the average exponential dependence

$$\beta_\sigma = 0.1094 \exp (1.363\, q). \quad (13)$$

At the same time, the scattering coefficient and the total volume of particles are characterized by closer correlation (0.777) and the power dependence

$$V_t = (41.0 \pm 6.0)\, \beta_\sigma^{(0.459 \pm 0.15)}. \quad (14)$$

If the statistical data shown in Fig. 12 are interpreted in such coordinate system as $\ln \beta_\sigma$ and $\ln (1 - q)$, the functional dependence (13) can easily be transformed into the traditional Kasten–Hanel form[22] with the following coefficients:

$$\beta_\sigma = 0.147\, (1 - q)^{-0.458}. \quad (15)$$

The dependence (13) is shown in Fig. 13*a* in comparison with the dependence (15) against the background of the initial statistical ensemble in the linear system of coordinates (β_σ and q). It is seen from the figure that the dependence (13) more closely corresponds to the statistics of realizations at $q < 0.8$, but is unsuitable at $q > 0.85$, where the dependence (15) better fits to the experiment.

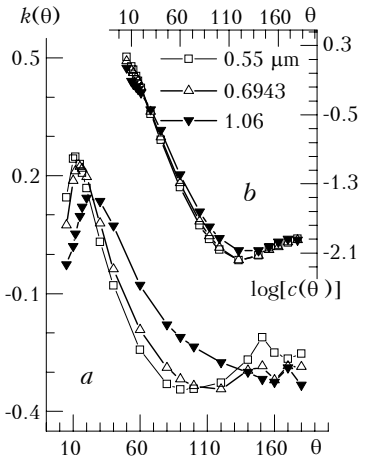

FIG. 14. Model extrapolation of the angular dependence of the coefficients $k(\theta)$ and $c(\theta)$ of the regression model to other lidar wavelengths: $\lambda = 0.53$, 0.6943, *and* 1.06 μm.

The purposeful field measurements[22] of intercorrelation between the above parameters have revealed that so-called parameter of condensation activity γ varies from 0.3 to 0.8 (in our case $\gamma = 0.458$ in Eq. (15)). Different measurements give different values for the second parameters, in particular, with regard parameter in Eq. (15). This uncertainty can be explained by peculiarities in formation of the haze microstructure and the statistics of atmospheric situations.

The obtained theoretical estimates were then used to consider the possibility of extrapolation of a single–parameter model empirically found at $\lambda = 0.55$ μm to other lidar wavelengths: 0.53, 0.6943, and 1.06 μm (see Fig. 14), including the case of sensing at the lidar angles. The latter case apparently requires an individual approach to *a priori* choice of the model for regional peculiarity of renewing the composition of the coarse particles.

5. CONCLUSIONS AND NOTES

The model estimates presented in this paper show that the single-parameter model of variation of the aerosol light scattering characteristics of the near-ground haze is suitable for description of really observed variety of the atmospheric haze dispersion composition providing both variability of the optical constants and the dynamic changes of the aerosol particle size spectrum with changing relative humidity of the atmosphere are characteristic for these variety. Application of a single-parameter model is more justified for atmospheric situations, in which significant dynamics of the relative humidity is observed.

In spite of linearly centered statistics of realizations of the initial microphysical aerosol parameters and the relative humidity, the lognormal statistics is more characteristic for normalized values of the angular functions of the aerosol light scattering.

Besides, the most significant factor governing the shape of the angular dependence $k(\theta)$ at the wavelength $\lambda = 0.55$ μm is the statistically average size r_2 of particles of the accumulative fraction. Variations of the b_2 and F_2 parameters may introduce minor changes in $k(\theta)$ at angles from 20 to 40 and from 90 to 165°, but they do not change the general shape of the angular dependence.

A particular shape of the angular dependence $k(\theta)$ is stipulated by the width of the variability range of the microstructural parameters (m_i, r_i, b_i, F_i) and their mutual dynamics. At the same time, the shape of the $c(\theta)$ dependence is dictated by the values of the microstructural parameters averaged over the ensemble of realizations. Thus, the single-parameter model as a statistically average regularity is a generalization of mean states of the atmospheric haze, which occur most often due to joint effect of geophysical factors.

This set of states acquires in fact a special status, relative to which other realizations, being in essence only deviations from the statistically average (atmospheric) variability of the haze, take the meaning of abnormal or, at large deviations, disturbed states. Stable quasi-cyclic variations of meteorological elements of the surface atmosphere favor joining of

dissimilar components (primary and secondary ones) into some unity. This forms a new quality, which is likely most promising for solution of the problem of determination of background properties of the aerosol component as a balanced or (following the Friedlander terminology) self-preserving aerosol background of atmospheric processes. A solution to the problem should be sought for by moving gradually along most probable states given by the model (1) towards the states with low atmospheric turbidity.

Diurnal changes of the meteorological parameters of the surface atmospheric layer smooth out peaks of sporadic microstructural changes in the near-ground haze. They also favor involving the secondary aerosols and water vapor into the microstructure of the optically most active fraction, thus providing really observed modulation of random variations of the aerosol light scattering parameters. In this sense, the surface atmospheric layer can be considered as a sort of assembly department, where different components of the atmospheric haze microstructure are processed and fit to each other before being carried away from the friction layer into free atmosphere already as a dispersion unity.

Thus, the calculated estimates show that the heuristic significance of the model (1) manifests itself against the background of complex external geophysical conditionality of local (internal) processes in the aerosol. This significance allows a revealing of most informative characteristics as predictors of light scattering properties of the atmospheric haze. Besides, it allows the information about most characteristic states of the atmosphere dispersion component (the surface atmospheric layer in our case) to be expressed in the accumulated form through particular values of the $k(\theta)$ and $c(\theta)$ parameters. By G.V. Rozenberg's graphic phrase, such few-variable models "store" the information about atmospheric processes, which are most significant for aerosols.

Let us finally note one more important circumstance, which follows from our results. Despite the study of the aerosol component of the atmosphere in its complex responses to various external actions, including natural effects of geophysical factors, adds noticeable complexity to optical experiments and the analysis of the obtained data, this approach opens principally new possibilities for deeper analysis of origins of some or another properties of the object under investigation. This conclusion follows not only from the results presented, but also from the analysis of the specificity of microstructural changes in pyrolysis smokes during relaxation in a closed volume.[25] In those experiments we have activated the microprocesses responsible for transformation of the smoke dispersion structure, in particular, dry sedimentation of particles onto chamber's walls and their coagulation.

The study of optical properties of an aerosol in dynamics or using a static set of states allows one to estimate not only most characteristic parameters of an object, but also the degree of its sensitivity to external factors and the processes of internal transformation. For example, a multidimensional vector $k(\theta)$ in Eq. (1) determines just the degree of mobility of the state of the near-ground atmospheric haze, i.e. its lability. When solving an inverse problem for some individual realizations of the scattering phase function $g(\theta)$, some fine specific properties of the object may remain unnoticed in static against the background of microstructural elements giving the major contribution into formation of a particular shape of $g(\theta)$. These fine properties can be studied from the analysis of microphysical conditionality of a specific shape of $k(\theta)$. At the same time, some microstructural elements of the atmospheric haze, little significant in static but strongly sensitive to external factors, manifest themselves as factors of formation of statistical variety of the aerosol light scattering angular functions.

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