Correlation between optical characteristics and microstructure of the near-ground aerosol

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The inverse problem was solved for the data of spectral nephelometer measurements of the directed scattering coefficients D at a scattering angle of 45°. Measurements were conducted in Moscow and Odessa regions both for ambient aerosol and its dry matter. The principle component method was applied to statistical analysis of logarithms of the retrieved size distributions of the particle cross section $S(r)$. A correlation between statistical characteristics of $\ln D$ and $\ln S$ is considered. A relation between first eigenvectors of their covariance matrices is suggested.

Introduction

The results of statistical analysis of the near-ground aerosol light scattering characteristics[1,2] testify that the variability of these characteristics can be described using a small number of statistical parameters, namely, the coefficients of expansion in terms of eigenvectors of the corresponding covariance matrices. The first coefficient, which describes about 90% the variance, is closely related to the scattering coefficient. This fact provided the basis for the one-parameter model of the aerosol optical characteristics.[2,3] Regularities of transformation of the aerosol optical properties represent variations of its microstructure. The one-parameter models[2,4] of aerosol microstructure were constructed through solving the inverse problem for the model optical characteristics corresponding to several values of the scattering coefficient (or meteorological range). These models enable one to follow the features of aerosol transformation at a change of the atmospheric turbidity. As it increases, particles become larger due to moistening and accumulation of dry substance. The refractive index of the aerosol matter decreases approaching to the refractive index of water in fog hazes.

There may be another approach to optical modeling of the aerosol microstructure, namely, simultaneous measurements of optical characteristics and particle size distribution, their separate analysis by the principal component method, and finding correlations between the obtained statistical parameters. Optical properties of aerosol in the visible wavelength range are largely determined by its submicron fraction. Unfortunately, the available methods and devices (counters of mobility and photoelectric counters) individually are not able to reliably determine the microstructure in the entire submicron size range. This significantly hinders the conducting of such measurements.

In this paper, an attempt is made to compare statistical characteristics of spectral dependences of the directed light scattering $D(\lambda)$ at the angle of 45° and the aerosol microstructure obtained from inverting these dependences. In spite of the fact that such an approach is to some extent artificial, this analysis, in our opinion, is of interest both for estimating the possibility of prompt determination of the aerosol parameters without solving the inverse problem and for constructing the optical models of the aerosol microstructure accounting for not only the main direction of its variability.

Experimental data arrays

Measurements of the spectral dependences of the directed scattering coefficient at the angle of 45° in the wavelength range 254–578 nm were carried out in winter 1986 at the Zvenigorod Scientific Station (ZSS) of the Institute of Atmospheric Physics and in fall 1987 in the vicinity of Odessa with the spectral nephelometer based on the MDR-2 diffraction monochromator. The DRS-250-2 mercurial lamp was a source of radiation. The nephelometer was equipped with a heater of the air flow pumped through the working volume, that enabled us to measure the characteristics both of the ambient aerosol and its dry matter. The length of the measurement cycle, including “dry” aerosol characteristics, was about 10 minutes. The scattering volume was approximately equal to 100 cm^3. For the subsequent processing, 118 measurement series at ZSS and 133 for the region of Odessa were selected.

Solving the inverse problem

The modified Twitty algorithm[5,6] was used for solving the inverse problem. Numerical experiments with model distribution showed that the particle size spectrum could be reconstructed in the range from 0.07 μm to ~ 0.8–1 μm. The rms deviation of the initial $D(\lambda)$ data from the reconstructed ones did not exceed a few percent.

One of the main problems in inverting the data of spectral measurements of the aerosol extinction is impossibility of determination of the refractive index. Analogous ambiguity exists also for light scattering at the angle of 45°. The real part of the complex refractive index n of the dry matter was set constant and equal to 1.5. The value of n for the ambient aerosol was estimated in two ways: from the one-parameter model[4] and from the ratio of the directed scattering coefficients of the dry and ambient aerosol. The difference between two estimates usually did not exceed 0.03. The mean of two n values was used in solving the inverse problem. The imaginary part of the refractive index was assumed equal to zero.

Analysis of results of inverting the same data with different n values shows that incorrect setting of the refractive index leads to a shift of the cross section size distribution $S(r)$ (in logarithmic scale) along the coordinate axis. The magnitude of the shift is equal to $0.5\ln(n-1)$. This result is shown in Figs. 1 and 2.

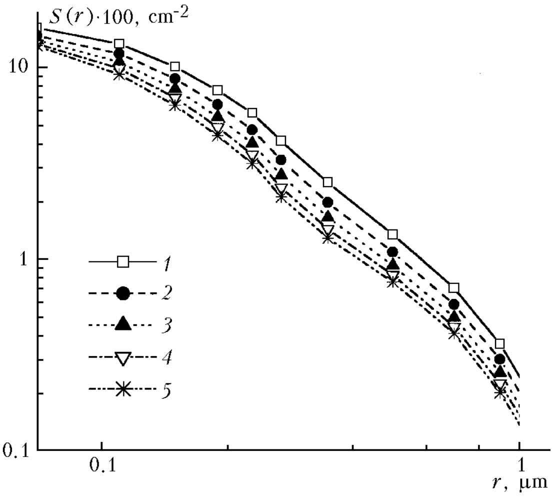

Fig. 1. Cross section size distributions obtained from solution of the inverse problem at different refractive indices. ZSS, January 29, 1986; $n = 1.35$ (*1*), 1.4 (*2*), 1.45 (*3*), 1.5 (*4*), and 1.55 (*5*).

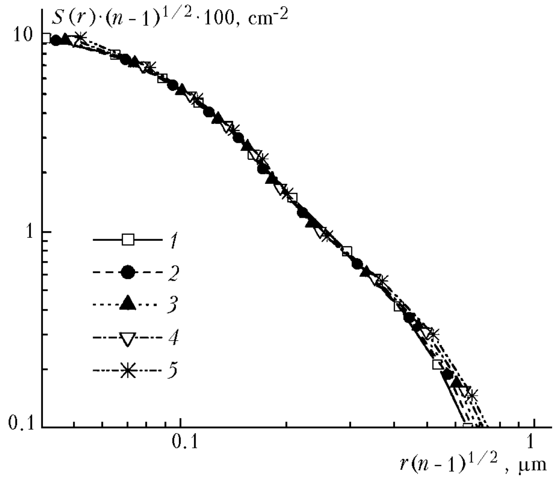

Fig. 2. Cross section size distributions as functions of $r(n-1)^{1/2}$. Designations the same as in Fig. 1.

The curves $S(r)$ obtained at different n in the range 1.35–1.55 are demonstrated in Fig. 1. Figure 2 shows $S(r)(n-1)^{1/2}$ as a function of $r(n-1)^{1/2}$. Note that the analogous result holds for interpretation of spectral measurements of transparency, but with the coefficient $n-1$, other than $(n-1)^{1/2}$. Hence, the influence of inaccuracy in setting n on inverting the directed spectral light-scattering coefficients at the angle of 45° is weaker than on inverting the aerosol extinction coefficients.

Statistical analysis of the size spectra

Statistical analysis of the obtained data arrays $\ln S(r)$ was carried out by the principal component method.[7] The choice of logarithmic representation was due to the fact that usually $S(r)$ quickly decreases as particle size increases at $r > 0.1$ μm, and small particles make a principal contribution into the expansion coefficient in terms of eigenvectors of the covariance matrix $S(r)$. Besides, covariance matrices and, hence, the systems of eigenvectors for $\ln S(r)$ are the same as for number and volume distributions.

In the first approximation, all four arrays $\ln S(r)$ can be described as one-parameter ones, for about 90% the variance fall on the first principal components (about 99% on the three first ones). Hence, $S(r)$ can be presented in the form

$$\ln S(r) = \ln S_0(r) + c_1\pi_1(r) + c_2\pi_2(r) + c_3\pi_3(r) + \ldots, \quad (1)$$

where $S_0(r)$ is the mean geometric distribution; $\pi_i(r)$ are the eigenvectors of the covariance matrix of $\ln S_0(r)$.

Eigenvalues of the covariance matrix normalized to its trace are presented in Table 1. Two first eigenvectors of the covariance matrix are shown in Fig. 3.

Table 1. Normalized eigenvalues of the covariance matrices

Value	ZSS, ambient	ZSS, dry	Odessa, ambient	Odessa, dry
L1	0.91	0.91	0.87	0.89
L2	0.074	0.062	0.078	0.063
L3	0.015	0.025	0.039	0.036

It follows from Fig. 3 that the shape of the particle size distribution function most varies in the range $r < 0.3$ μm. The eigenvector systems of the ambient aerosol and its dry matter are close to each other.

The initial spectral dependences $D(\lambda)$ can also be treated as one-parameter (about 97% the variance fall on the first component).[8] Within the limits of the measurement error, $\ln D$ can be presented in the form

$$\ln D(\lambda) = \ln D_0(\lambda) + f_1 p_1(\lambda) + f_2 p_2(\lambda) + \ldots, \quad (2)$$

where $D_0(\lambda)$ is the mean geometric spectral dependence of D; f_i are the expansion coefficients, $p_i(\lambda)$ are the eigenvectors.

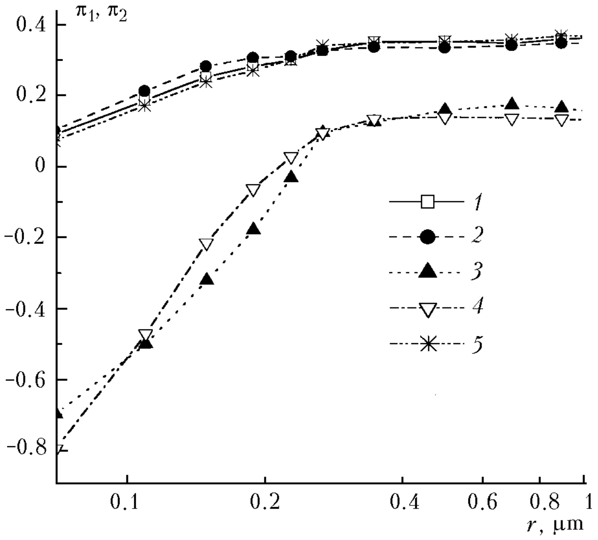

Fig. 3. Eigenvectors of the covariance matrix of ln$S(r)$: *1, 2, 5* — π_1; *3, 4* — π_2. *1, 3, 5* — ambient aerosol, *2, 4* — dry matter, *1–4* — ZSS, *5* — Odessa.

As logarithms of the directed scattering coefficients and differential number density of particles can be retrieved in the first approximation using the only parameter — the first expansion coefficient in terms of eigenvectors of the corresponding covariance matrix, it may be expected that these coefficients correlate. It turned out that not only the first but also the second and third coefficients correlate. The correlation coefficients exceed 0.9. Obviously, there is no cross-correlation between the expansion coefficients with different numbers.

Correlation diagrams for two first coefficients are shown in Figs. 4 and 5. Thus, it is possible to find direct relationships between optical characteristics and microstructural parameters obtained from solution of the inverse problem. It is supposed to test this result further using other data arrays including the measurements of polarized components of the directed scattering coefficient.

The considered ensembles of the particle size distribution were obtained from solution of the inverse problem. By virtue of the fact that the problem is ill-posed, one cannot be absolutely sure that they are adequate to realistic atmospheric situations. So, to confirm the reliability of the obtained results, it is necessary also to measure simultaneously the optical characteristics and aerosol microstructure in the submicron size range. As it was mentioned above, this is a rather difficult problem.

Another approach to interpretation of the revealed correlation is also possible. Assume that we have arrays of realistic particle size distributions, the variability of which is described by a small number of statistical parameters by formula (1). We calculate $D(\lambda)$ by the Mie theory and analyze them statistically. In this case we deal with the direct problem, and the results obtained testify that the statistical characteristics of $D(\lambda)$ directly relate to the statistical characteristics of $S(r)$. This is true both for the expansion coefficients and eigenvectors. Optical and microphysical characteristics are connected through the following formula:

$$D(\lambda) = \int S(r)K(r,n,\lambda)\mathrm{d}r\,. \qquad (3)$$

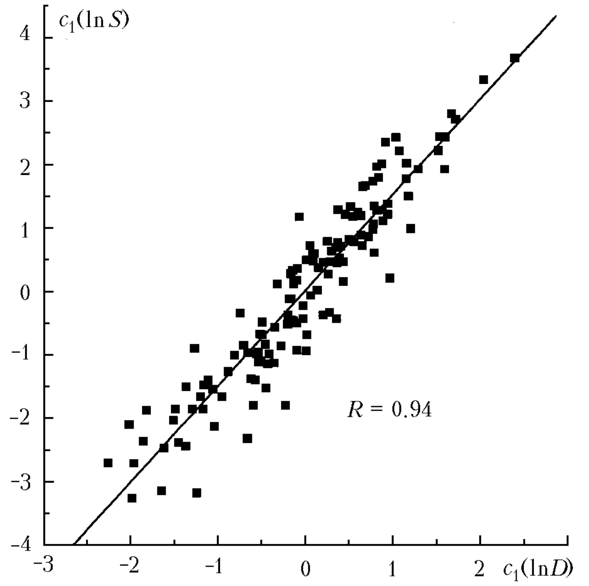

Fig. 4. Correlation diagram for the first coefficients of the expansions of ln$D$ and ln$S$, Odessa, 1987.

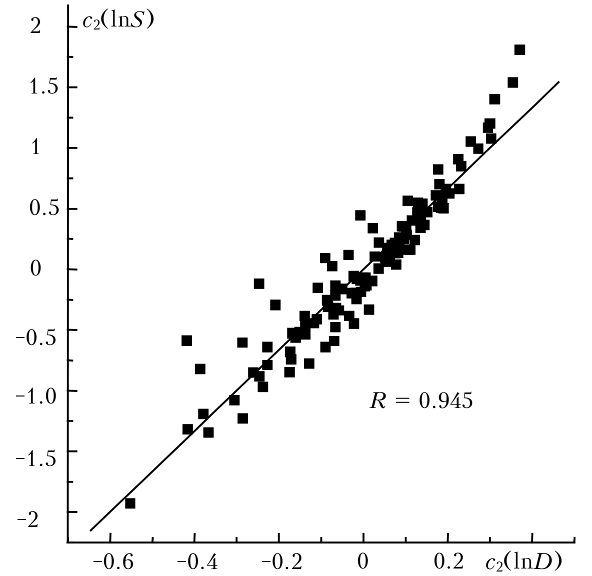

Fig. 5. Correlation diagram for the second coefficients of the expansions of ln$D$ and ln$S$, ZSS, 1986.

Assuming that ln$D(\lambda)$ and ln$S(r)$ are the differentiable functions of the expansion coefficients, and there is a linear relation between c_1 and f_1, as well as considering only first terms in (1) and (2), it is possible to derive the following relationship between first eigenvectors of the covariance matrices:

$$p_1(\lambda) \propto \left[\int \pi_1(r) S_0(r) K(r,\lambda) \mathrm{d}r\right][D_0(\lambda)]^{-1}, \qquad (4)$$

where $K(r, \lambda)$ is the mean kernel function $K(r, n, \lambda)$. The result of retrieving the first eigenvector by Eq. (4) is shown in Fig. 6.

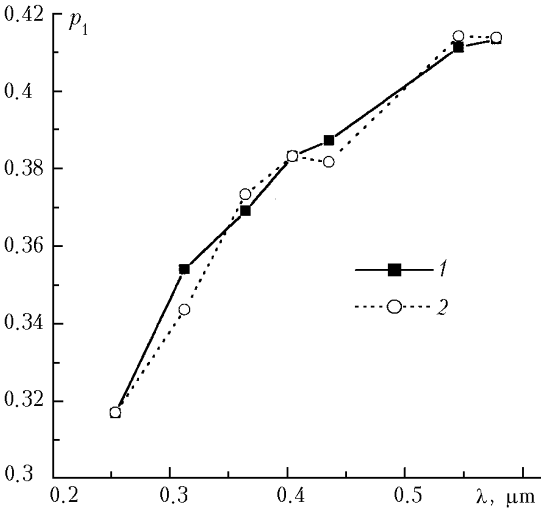

Fig. 6. The first eigenvector of the covariance matrix of ln*D* (curve *1*) and its calculation by formula (4), ZSS, 1986.

Acknowledgments

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