

OPTIMIZED LINEAR ESTIMATE OF INTEGRAL PARAMETERS OF MULTICOMPONENT AEROSOL

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The method for reconstructing the disperse medium characteristics by means of linear functions of measured parameters in application to studies of the atmospheric aerosol is proposed. The optimization problem with restrictions in the form of the system of linear equations is formulated to find the coefficients of linear regression. The optimization criterion and additional a priori information are chosen with respect to the peculiarities of formation and existence of real atmospheric aerosol related to its multifractional structure. Application of the method to data processing of multifrequency laser sounding of the atmosphere is considered. Some results of numerical calculations are presented.

Optical methods of studying the atmospheric aerosol are now widely used in practice. They are more efficient and informative when complicated data on aerosol properties at various altitudes are needed and long-term monitoring is desired. The reconstruction of aerosol parameter from optical measurement data falls into a class of an ill-posed problems whose solutions are unstable with respect to measurement and calculation errors.¹

The applicability of linear-estimation technique to determination of integral parameters of aerosol fine structure from optical characteristics was considered in Ref. 2. Simple linear relations between the parameters of scattering particles obtained by this approach allow the data processing to be done directly during measurements.

Efficiency of the linear-estimates method strongly depends on the choice of particle size-distributions ensemble for which the regression parameters are to be determined. If real objects do not agree with this *a priori* information, the reconstructed parameter values can be physically senseless. In this paper, to develop the method of Ref. 2, we assume the multicomponent structure of a disperse medium and introduce additional conditions concerning the physical models of formation and transformation of typical kinds of aerosol (continental, maritime, etc.). This allowed us to obtain the estimates of aerosol parameters accurate enough and quite stable to significant scatter of measured data. The common approach to making such estimates is given, and concrete formulas and numerical results for the calculation of extinction coefficient from backscattering spectral data are presented. Such a statement of the problem was inspired by the fact that determination of relations between extinction and backscattering coefficients is a key point in the methods of multifrequency laser sounding of atmospheric aerosol.³

1. CONSTRUCTION OF LINEAR ESTIMATE OF THE PARAMETERS OF MULTICOMPONENT AEROSOL

Let us analyze the aerosol parameters that can be presented as functionals of size-distribution density $\varphi(r)$

$$M[l \varphi] = \int l(r) \varphi(r) dr, \quad (1)$$

where $l(r)$ is the weighting function specifying the sense of $M[l, \varphi]$ (backscattering, extinction, mass concentration, etc.). The aerosol is considered as a polydisperse ensemble of uniform spherical particles.

The task is to present a sought parameter η as a linear combination of measured values of the parameter ξ

$$\eta = \mathbf{x}^T \boldsymbol{\xi} + \varepsilon, \quad (2)$$

where ε is the random deviation, and \mathbf{x}^T is transposed vector of expansion coefficients.

One can find these latter coefficients by minimizing the mean square difference over the ensemble of size-distributions

$$g = \overline{(\mathbf{x}^T \boldsymbol{\xi} - \eta)^2} = \min. \quad (3)$$

The bar means averaging over ensemble, and r is the particle radius.

To construct a linear estimate of η we use the representation of aerosol as an ensemble of statistically independent fractions with different formation mechanisms (maritime, dust, water-soluble, etc.).^{4,5} Then the parameters ξ and η are

$$\boldsymbol{\xi} = \sum_{j=1}^p c_j \boldsymbol{\xi}^j, \quad \eta = \sum_{j=1}^p c_j \eta^j, \quad (4)$$

where ξ^j and η^j refer to different aerosol fractions with corresponding distributions $\varphi_j(r)$ and optical properties of particles, p is total number of fractions, and c_j is the number density of the j th fraction. Distribution of every fraction is a stochastic function, but the common one can be presented as a linear combination of the mean distributions $\overline{\varphi_j(r)}$ with good accuracy in various typical situations, as it follows from experimental results on aerosol microphysical characteristics. In other words, the change of aerosol particle distribution is rather a change of densities

of individual conservative fractions than variation of the shape of their size distributions. Therefore, in order to reduce the η estimation error we impose the following additional restrictions on linear regression coefficients:

$$\mathbf{x}^T \overline{\xi^j} = \overline{\eta^j}, \quad j = 1, \dots, p. \quad (5)$$

Equation (5) requires accurate description, by means of a linear estimate, of the relation between averages $\overline{\xi^j}$ and $\overline{\eta^j}$.

By transforming Eq. (3) and combining it with Eq. (5), we derive the following conditions for regression coefficients²:

$$\begin{cases} \tilde{g} = \mathbf{x}^T \Phi \mathbf{x} - 2 \mathbf{L}^T \mathbf{x} = \min, \\ \mathbf{x}^T \xi^j = \eta^j, \quad j = 1, \dots, p, \end{cases} \quad (6)$$

where $\tilde{g} = g - \alpha$; $\Phi = \overline{\xi \xi^T}$ is the $n \times n$ matrix; $\mathbf{L} = \overline{\xi \eta}$ is the n -component vector; and, $\alpha = \overline{\eta^2}$ is a scalar. By definition the matrix Φ is symmetric and non-negative.

As a result, we obtain the linear regression problem with linear restrictions giving *a priori* information on the parameters, which can be solved with one of the known methods (see, e.g., Ref. 6).

2. LINEAR ESTIMATE OF EXTINCTION COEFFICIENT FROM THE BACKSCATTERING ONE

We have applied the above described procedure to the problem of reconstructing the aerosol extinction coefficient from a given backscattering spectrum. One has to determine the coefficients in the expression

$$\hat{\sigma}_l = \mathbf{x}^T \boldsymbol{\beta}, \quad (7)$$

where $\hat{\sigma}_l$ is the estimate of aerosol extinction at the wavelength λ_l , and $\boldsymbol{\beta}$ is the n -component vector of the backscattering coefficient at wavelengths λ_i , $i = 1, \dots, n$. The solution of this problem is a basis for methodology for processing data of multifrequency laser sounding, because it allows the system of lidar equations for different wavelengths to be closed.³ Besides, Eq. (7) can be used directly, i.e., for determination of aerosol extinction coefficient from the measured backscattering one.

Values of extinction (σ) and backscattering (β_i) coefficients at wavelengths λ_i in a multicomponent disperse medium depend on size-distribution densities for various fractions as follows:

$$\beta_i = \int \pi r^2 \sum_{j=1}^p c_j K_{\pi_{ij}}(r) \varphi_j(r) dr; \quad (8)$$

$$\sigma_i = \int \pi r^2 \sum_{j=1}^p c_j K_l(r) \varphi_j(r) dr, \quad (9)$$

where $i = 1, \dots, n$, n is the number of wavelengths; $K_{\pi_{ij}}(r)$ and $K_l(r)$ are the efficiency factors of backscattering and extinction for j th fraction, respectively.

We assume no correlation between distribution variations of different fractions. That does not affect the generality of the model, because two interacting fractions

can always be considered as a single one. Hence, the correlation function is of the form

$$\overline{\varphi_j(r) \varphi_k(r')} = \overline{\varphi_j(r)} \overline{\varphi_k(r')} + \text{cov}(\varphi_j(r) \varphi_k(r')) \delta_{jk}, \quad (10)$$

where δ_{jk} is the Kronecker symbol.

We also do not lose generality if suppose that $\varphi_j(r) = 0$ at $r > R$, for all j ignoring the behavior of the function out of $[0, R]$ interval. Therefore, the conditions (6) for the linear estimate (7) become

$$\begin{cases} \tilde{g} = \mathbf{x}^T \Phi \mathbf{x} - 2 \mathbf{L}^T \mathbf{x} = \min, \\ \mathbf{B}^T \mathbf{x} = \mathbf{L}^*. \end{cases} \quad (11)$$

Once Eqs. (8)–(10) are written, the vectors and matrices introduced earlier take the form

$$\begin{aligned} \Phi &= \|\Phi_{ik}\|, \quad i, k = 1, \dots, n, \\ \Phi &= (B \mathbf{c}) (B \mathbf{c})^T + \tilde{\Phi}, \quad \tilde{\Phi} = \sum_{j=1}^p c_j^2 \tilde{\Phi}^j; \\ B &= \|B_{ij}\|, \quad i = 1, \dots, n, \quad j = 1, \dots, p, \\ B_{ij} &= \int_0^R \pi r^2 K_{\pi_{ij}}(r) \overline{\varphi_j(r)} dr; \\ \tilde{\Phi}_{ik}^j &= \int_0^R \int_0^R \pi^2 r^4 K_{\pi_{ij}}(r) \text{cov}(\varphi_j(r) \varphi_j(r')) K_{\pi_{kj}}(r') dr dr'; \end{aligned} \quad (12)$$

$$\mathbf{L} = (B \mathbf{c}) (\mathbf{c}^T \mathbf{L}^*) + \tilde{\mathbf{L}}, \quad \tilde{\mathbf{L}} = \sum_{j=1}^p c_j^2 \tilde{\mathbf{L}}^j,$$

$$L_j^* = \int_0^R \pi r^2 K_l(r) \overline{\varphi_j(r)} dr, \quad j = 1, \dots, p,$$

$$\tilde{L}_i^j = \int_0^R \int_0^R \pi^2 r^4 K_{\pi_{ij}}(r) \text{cov}(\varphi_j(r) \varphi_j(r')) K_l(r') dr dr';$$

$$\alpha = (\mathbf{c}^T \mathbf{L}^*) (\mathbf{c}^T \mathbf{L}^*) + \tilde{\alpha}, \quad \tilde{\alpha} = \sum_{j=1}^p c_j^2 \tilde{\alpha}^j,$$

$$\tilde{\alpha}^j = \int_0^R \int_0^R \pi^2 r^4 K_l(r) \text{cov}(\varphi_j(r) \varphi_j(r')) K_l(r') dr dr'.$$

As it follows from Eqs. (12), when solving the system (11) absolute values of number densities do not affect the resulting linear regression coefficients; only fractions percentage is important. This allows one to choose numerical values of c_j for the convenience of making calculations.

The linearity of Eq. (7) provides simple evaluation of the error in the backscattering coefficient on resulting estimate uncertainty.

The construction of estimates for other integral parameters of the form (1) will obviously change only weighting functions K_{π} and K_l in all the above expressions

3. RESULTS OF NUMERICAL EXPERIMENTS

To perform concrete computations according to the above-mentioned scheme we should preliminarily define some properties of statistical ensemble considered. The *a priori* aerosol information was set as follows. We determine

the number p of independent fractions and optical constants $m = n - i \kappa$ for each of them, to calculate weighting functions of optical parameters. Then we find the mean values of number density and size distribution for each fraction as well as covariance functions $\text{cov}(\varphi_j(r) \varphi_j(r'))$. The choice of numerical values of the above quantities and analytical views of the functions has been done based on data on the properties of atmospheric aerosol available from literature.

The described algorithm was realized as a software package for an IBM PC/AT computer. The computations were carried out for continental and maritime types of aerosol according to the SRA-84 models proposed by International Radiation Commission.^{7,8} The results presented below refer to the maritime aerosol model.

The maritime model implies existence of two fractions: water-soluble and oceanic, each of them being described by the lognormal distribution with parameters presented in Table I.

Table I. Parameters of maritime aerosol model.

Parameters	Water-soluble	Oceanic
Refractivity, n (real part)	1.53	1.38
Absorption coefficient, κ (imaginary part)	0.007	5.e - 08
Dimensional parameter of lognormal distribution (μm), r_m	0.005	0.3
Width parameter of lognormal distribution, σ^*	1.094	0.921
Number density, c_j	9.9957413e - 01	4.2587e - 04

The covariance functions were defined as follows:

$$\text{cov}(\varphi_j(r) \varphi_j(r')) = \overline{\varphi_j(r)} \overline{\varphi_j(r')} \omega_j(r, r') \times ((\exp(\sigma^2(r)) - 1)(\exp(\sigma^2(r')) - 1))^{1/2}, \quad j = 1, \dots, p, \quad (13)$$

where $\sigma^2(r)$ is the variance of $\ln\varphi(r)$ at the point r , and $\omega_j(r, r')$ is the normalized correlation function of the form

$$\omega_j(r, r') = \exp(-(r - r')^2 v / D_j^2), \quad j = 1, \dots, p. \quad (14)$$

We have taken $\sigma^2(r) = \ln 2 = \text{const}$, $v = \ln 10$, and $D_j = 0.1 \mu\text{m}$, according to the results of measurements of the statistical parameters of aerosol size-distribution functions (in particular, from Ref. 9). The weighting functions $K_{\pi_{ij}}(r)$ and K_{I_j} of backscattering and extinction coefficients were calculated by Mie formulas.

The size-distribution function was considered to be normalized, i.e.,

$$\int_0^\infty \sum_{j=1}^p c_j \varphi_j(r) dr = 1. \quad (15)$$

In Table II we present in matrix form the coefficients for obtaining the extinction coefficients from given numerical values of the backscattering ones for the set of seven wavelengths of the multifrequency Gloria-M lidar.¹⁰

Table II. Matrix of coefficients for reconstructing extinction coefficients from the values of backscattering ones.

λ , nm	1	2	3	4	5	6	7
380	38.835	9.3629	1.8783	-12.415	14.044	-13.249	-35.788
430	32.436	7.9725	1.9715	-9.5500	11.614	-10.652	-27.829
510	23.598	6.5491	2.4238	-5.3609	8.2954	-6.9719	-18.331
585	18.405	4.1058	1.8990	-1.3087	5.8328	-4.3359	-11.362
694	14.341	0.53227	0.16042	1.6695	4.4344	-1.6419	-2.9240
860	7.7886	0.71204	-2.0632	3.8508	0.91496	2.1870	7.1123
965	2.7826	1.9188	-0.40919	4.4691	-1.3821	2.8027	12.218

Figure 1 illustrates the results of numerical experiment on the evaluation of measurement error influence on the accuracy of resulting linear estimates. The microphysical parameters of aerosol fractions were taken from Table I.

Three different situations concerning pure oceanic ($c_1 = 0$), pure water-soluble ($c_2 = 0$) fractions, and the mixture of both these fractions with the number density ratio as indicated in Table I, were considered. The "true" values of extinction (σ_i) and backscattering (β_i) coefficients were calculated for seven wavelengths (curves 1-3 in Fig. 1 are shown in relative units). Then we introduced a random normally distributed error in β_i with relative rms fluctuation of 10%. From spectral dependences of $\tilde{\beta}_i$, perverted by "measurement" errors we reconstructed the spectra of extinction coefficients $\tilde{\sigma}_i$ with the help of transfer matrix (Table II). To obtain statistically proved results this procedure was repeated 8 000 times. The rms fluctuations of $\tilde{\sigma}_i$ are shown by bars at corresponding curves.

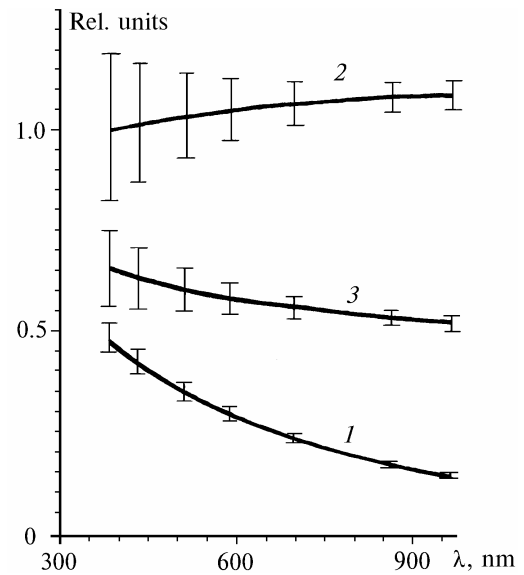


FIG. 1. Measurement error influence on a quality of reconstruction of extinction coefficient spectral behavior for maritime aerosol model: water-soluble (1), oceanic (2), and maritime (3).

It is seen from Fig. 1 that, first, the use of a given transfer matrix does not lead to systematic error even at strong variations of number densities of different fractions,

and, second, the strengthening factor of measurement errors in most cases is rather close to unity.

As it was already mentioned, significant source of errors in linear estimates calculations is the discrepancy between microphysical parameters of real atmospheric aerosol fractions and *a priori* model data. Below we present the results of two numerical experiments on investigating effects of this discrepancy.

In the first one, we analyzed errors caused by variation of parameters of lognormal distribution. A grid of values of r_m and σ^* was taken (see Table III) which produced 15 realizations for each fraction.

Table III. Variations of parameters of lognormal distribution for maritime aerosol fractions.

Water-soluble		Oceanic	
r_m	σ^*	r_m	σ^*
0.005	0.7	0.20	0.80
0.01	1.0	0.25	0.90
0.05	1.3	0.30	0.95
0.1	—	0.35	—
0.2	—	0.40	—

The size-distribution of aerosol particles was defined in such a way that microphysical parameters of one fraction were taken constant (from Table I) while those of second fraction varied according to Table III so that for every value of r_m the value σ^* varied thrice. The number density coefficients of varied fraction were taken in order that backscattering coefficient for λ_4 coincides with $\beta(\lambda_4)$ for corresponding maritime fraction. For these distributions, the "true" values of β_i and σ_i were calculated, and estimates of the extinction spectral dependence were derived from β_i with the transfer matrix. Then we obtained err_0 (maximum, of all λ_i , value of error of σ_i reconstruction) and err_2 (rms relative error). The results of calculations are presented in Table IV.

Table IV. Relation between errors of a priori information about size distribution of aerosol particles and those of reconstruction of the extinction coefficient from the backscattering one.

N	Variations of water-soluble fraction		Variations of oceanic fraction	
	err_0	err_2	err_0	err_2
1	4.0714e-001	3.1921e-001	3.3962e-001	2.1196e-001
2	3.5580e-002	2.0846e-002	1.8539e-001	1.1402e-001
3	7.8484e-003	5.1657e-003	1.0352e-001	6.4389e-002
4	1.9823e-001	1.5393e-001	2.6003e-001	1.5827e-001
5	5.0544e-003	3.8163e-003	9.7336e-002	5.9186e-002
6	4.2203e-002	2.7019e-002	1.7826e-002	1.1739e-002
7	2.9309e-002	2.2286e-002	1.8500e-001	1.1364e-001
8	9.8069e-003	7.4724e-003	2.1383e-002	1.4290e-002
9	2.5211e-001	1.4846e-001	5.5873e-002	3.2763e-002
10	1.1789e-001	5.8171e-002	1.1603e-001	7.4170e-002
11	1.2255e-001	6.9072e-002	4.4443e-002	2.6978e-002
12	3.7872e-001	2.3135e-001	1.1718e-001	6.9194e-002
13	1.7566e-002	1.0265e-002	5.9680e-002	3.9308e-002
14	2.8968e-001	1.6496e-001	1.0131e-001	6.0501e-002
15	5.3918e-001	3.5733e-001	1.6827e-001	9.9320e-002

In the same manner we performed the experiment on detection of linear estimates errors due to variations of refractivity (Table V). The results are presented in Table VI.

It is seen from Tables IV and VI that deviations of microphysical parameters of fractions from model values may cause significant systematic errors. Nevertheless, the obtained transfer matrix gives good accuracy in a wide range of variations of particle optical constants and size-distributions.

Table V. Refractivity variations for maritime aerosol fractions.

Water-soluble		Oceanic	
n	κ	n	κ
1.45	0.001	1.33	5.e-9
1.50	0.005	1.36	5.e-7
1.52	0.010	1.40	5.e-6
1.54	—	1.43	—
1.57	—	1.45	—

Table VI. Relation between errors of a priori information about refractivity and those of reconstruction of the extinction coefficient from the backscattering one.

N	Variations of water-soluble fraction		Variations of oceanic fraction	
	err_0	err_2	err_0	err_2
1	1.2080e-001	7.9176e-002	2.1576e-001	2.0018e-001
2	1.7240e-001	1.1616e-001	2.1602e-001	2.0054e-001
3	2.2524e-001	1.5523e-001	2.1811e-001	2.0362e-001
4	8.4971e-003	7.4493e-003	1.5579e-001	1.3000e-001
5	4.8782e-002	3.2000e-002	1.5686e-001	1.3070e-001
6	1.0931e-001	7.4697e-002	1.6111e-001	1.3381e-001
7	5.9966e-002	4.1099e-002	2.4560e-001	1.7003e-001
8	2.2482e-003	1.9913e-003	2.4429e-001	1.6917e-001
9	6.0911e-002	4.1803e-002	2.3337e-001	1.6195e-001
10	1.0903e-001	7.2973e-002	3.5256e-001	2.6636e-001
11	5.0693e-002	3.3948e-002	3.5132e-001	2.6550e-001
12	1.4165e-002	1.0457e-002	3.4291e-001	2.5944e-001
13	1.7555e-001	1.1562e-001	2.1950e-001	1.3067e-001
14	1.1555e-001	7.6192e-002	2.1919e-001	1.3010e-001
15	4.8812e-002	3.1409e-002	2.1661e-001	1.2608e-001

The obtained coefficients matrices of linear estimates of extinction coefficients from the backscattering ones for maritime and continental types of aerosol were applied to processing data of multifrequency laser sounding of the atmosphere performed with the use of Gloria-M lidar (Ref. 10) in the Atlantic territorial waters and in agricultural and industrial areas of Belarus Republic. The transfer matrix similar to the one presented in Table II, was calculated for each type of aerosol. Use of algorithms of aerosol optical characteristics calculations based on these relations improved the quality of reconstruction of extinction ($\sigma_a(\lambda, h)$) and backscattering ($\beta_a(\lambda, h)$) spectral dependences as compared with the scheme from Ref. 2 or with those obtained at a preset lidar ratio. For instance, we avoided solutions without physical sense. Thus calculated values of $\beta_a(\lambda, h)$ and $\sigma_a(\lambda, h)$ better describe spatial variations of particle microphysical properties during transition from aerosol atmosphere to optically thin cloud layers.

CONCLUSION

In this paper we propose a linear estimates method to obtain integral parameters of fine structure of disperse media and its application to the investigation of atmospheric aerosol. The basic method from Ref. 2 is modified by introducing additional restrictions in the form of system of linear equations based on physical properties of atmospheric aerosol. The results of numerical

experiments show that the method provides good accuracy of reconstruction of aerosol parameters and good stability of obtained linear estimates to variations of the initial data.

The presented scheme is convenient for calculation of parameter estimates in various synthetic aerosol models, because it allows one to use preliminary obtained calculations referring to basic aerosol fractions.

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REFERENCES

1. V.E. Zuev and I.E. Naatz, *Inverse Problems of Laser Sounding of the Atmosphere* (Nauka, Novosibirsk, 1982), 240 pp.
2. A.P. Chaikovskii and V.N. Shcherbakov, *Zh. Prikl. Spectrosk.* **42**, No. 5, 820–824 (1985).
3. A.P. Chaikovskii, *Zh. Prikl. Spectrosk.* **44**, No. 2, 183–197 (1986).
4. K.Ya. Kondratiev, ed., *Aerosol and Climate* (Gidrometeoizdat, Leningrad, 1991), 541 pp.
5. J. Lenoble and C. Brogniez, *Beitr. Phys. Atmos.* **57**, No. 1, 1–19 (1984).
6. R. Gabasov, F.M. Kirillova, O.I. Kostyukova, and V.M. Raketskii, *Constructive Methods for Optimization. P. IV. Convex Problems* (University Publishing House, Minsk, 1987), 223 pp.
7. *A Preliminary Cloudless Standard Atmosphere for Radiation Computation*, International Association for Meteorology and Atmospheric Physics, Radiation Commission, Boulder, Colorado, USA (1984), 53 pp.
8. M.Ya. Marov, V.P. Shari, and L.D. Lomakina, *Optical Characteristics of Model Aerosols of the Earth's Atmosphere (Thematical Issue)*, M.V. Keldysh Applied Mathematics Institute, USSR Academy of Sciences, Moscow (1989), 229 pp.
9. V.P. Ivanov, P.A. Maslennikov, V.I. Sidorenko, et al., in: *First Global PIGAP Experiment. Vol. 1. Aerosol and Climate* (Gidrometeoizdat, Leningrad, 1981), pp. 90–98.
10. A.N. Borodavka, E.M. Gitlin, E.M. Gubsky, et al., in: *Catalog of Instruments* (Nauka i Technika, Minsk, 1988), pp. 28–29.