APPROXIMATION OF SPECTRAL DEPENDENCE OF THE EXTINCTION COEFFICIENT FOR AEROSOL POLYDISPERSIONS COMPRISING LARGE STRONGLY ABSORBING PARTICLES

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The spectral dependence of the extinction coefficient is considered for aerosol polydispersions observed near sources of pollution in the atmospheric surface layer. It is shown that for polydispersions comprising large strongly absorbing particles this spectral dependence is well approximated by the quadratic one. Range of application of this approximation is specified based on model calculations. The effect of the measurement error on the approximation one is analyzed.

When solving the problems of reconstruction of the aerosol particle size spectrum by the method of inversion of optical measurement data (in particular, of the measured spectral dependence of the aerosol extinction coefficient for the electromagnetic radiation), in many cases one faces the problems of incomplete determinacy of these problems caused by a limited number of sounding wavelengths. To overcome these difficulties, various methods are used including a supplement to a definition of the problem by way of approximation of the spectral optical characteristics from measurement data[1] and calculation of these characteristics in intermediate points. In so doing, the approximation accuracy should be satisfactory for efficient solution of the inverse problem.[2]

In the present paper, we consider the spectral dependence of the extinction coefficient for aerosol polydispersions observed in the surface layer of the atmosphere. The salient features of the examined aerosol polydispersions are predominance of strongly absorbing particles, whose optical characteristics exhibit more regular spectral behavior,[3] and a great number of large particles, whose size varies in the range from 1 to 10 μm and even greater.[3,4] As a result, the range of particle size far exceeds the spectral range of radiation used for laser sensing of the atmosphere and the spectral dependence of the optical characteristics may essentially differ from the commonly observed one (see, for example, Refs. 5 and 6).

Model calculations were done for spherical polydispersions that approximate well an ensemble of convex particles of arbitrary shape in the atmospheric surface layer.[6] Based on a large volume of calculations of the spectral dependence of the extinction coefficient $\beta_{ex}(\lambda)$ by the Mie formulas from codes given in Ref. 2, the spectral dependence of the extinction coefficient of the form

$$\tilde{\beta}(\lambda) = a\lambda^2 + b\lambda + d \qquad (1)$$

was chosen.

To check this formula, model calculations were done for the spherical polydispersions with the lognormal particle size distribution.[7] The parameters of the distribution were as follows:

r_m = 0.5 μm, σ = 0.5 μm (S = 0.200);

r_m = 0.5 μm, σ = 4.0 μm (S = 0.680);

r_m = 6.0 μm, σ = 0.5 μm (S = 0.083);

r_m = 6.0 μm, σ = 4.0 μm (S = 0.541);

r_m = 2.0 μm, σ = 1.5 μm (S = 0.570).

Here, r_m is the mean geometric radius of particles, σ is its standard deviation, and S is the standard deviation of $\ln(r)$. These values of the parameters of the lognormal distribution are typical of a wide class of aerosol pollutants observed in the atmospheric surface layer near sources of pollution (for example, enterprises producing household chemical goods, nonferrous metallurgical plants, electric power plants, glassworks, enterprises of coal industry, and cement factories[4]). The monodispersed aerosols with σ = 0.1 μm (by definition of Ref. 8) and r_m = 0.5, 1, 2, 3, 4, 5, and 6 μm were also considered. The complex refractive index of aerosol particles was $m = 1.59 - 0.66i$ (carbonaceous and sootlike particles[9]). Its dependence on λ (for the same particles) was calculated from the Kramers–Kronig[2] formulas for the initial values $m(\lambda = 0.436\ \mu m) = 1.90 - 0.68i$ and $m(\lambda = 0.546\ \mu m) = 1.96 - 0.66i$ (see Ref. 9). Calculations were also done for $m = 1.33 - 0.0i$ (water droplets).

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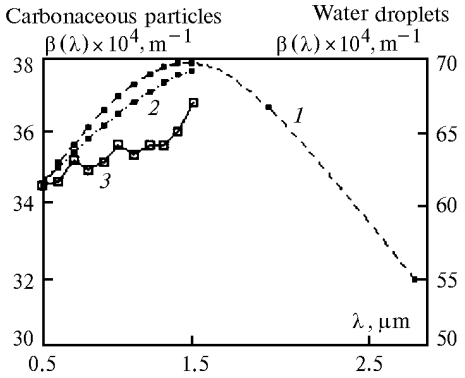

FIG. 1. Spectral dependence $\beta_{ex}(\lambda)$ *for ensembles of carbonaceous particles and water droplets having the same disperse composition* (r_m = 1 μm *and* σ = 1.5 μm*): 1)* $m = 1.59-0.66i$*, 2)* $m = m(\lambda)$ *(carbonaceous particles), 3)* $m = 1.33-0.0i$ *(water droplets).*

The calculations showed that the spectral dependence of the extinction coefficient for polydispersions comprising carbonaceous strongly absorbing particles actually resembled the ascending branch of an overturned parabola in the form of Eq. (1) in the wavelength range λ = 0.5–1.5 μm (see Fig. 1). At first glance the spectral dependence of this type seems to be inconsistent with numerous calculated and experimental results (for example, presented in Refs. 5 and 6). However, this contradiction will be eliminated if we consider that our calculations were done for large particles with $r_m \geq 0.5$ μm ($x_m = 2\pi r_m/\lambda \geq 2.09$), whereas the spectral characteristics given in Refs. 5 and 6 were determined for the finely dispersed fraction of atmospheric aerosols with $r_m \approx 0.2$ μm ($x_m \leq 2.51$). Calculations for λ > 2 μm and r_m = 1 μm ($x_m < 3.14$) showed that the value of the extinction coefficient decreased with the increase of the wavelength (curve *1* in Fig. 1), in complete agreement with the data of Refs. 5 and 6 considering the values of x_m. In addition, indirect evidence for the increase of $\beta_{ex}(\lambda)$ with λ in the wavelength range 0.5–1.5 μm for strongly absorbing particles can be found in Ref. 10.

The spectral dependence $\beta_{ex}(\lambda)$ for water droplets cannot be approximated by Eq. (1) with satisfactory accuracy because of its irregular behavior.

To construct the approximate dependence, the values of $\beta_{ex}(\lambda)$ calculated at λ = 0.5, 0.8, 1.2, and 1.5 μm were used. The relative approximation errors were calculated by the formula

$$\delta(\lambda_j) = [\tilde{\beta}(\lambda) - \beta_{ex}(\lambda_j)]/\beta_{ex}(\lambda_j) \qquad (2)$$

for λ = 0.5, 0.6,..., and 1.5 μm.

TABLE I.

λ, μm	$\delta\times10^3$, %											
	r_m, μm	0.5	0.5	6.0	6.0	2.0	0.5	1.0	2.0	3.0	4.0	5.0	6.0
	σ, μm	0.5	4.0	0.5	4.0	1.5	0.1	0.1	0.1	0.1	0.1	0.1	0.1
	S:	0.68	1.93	0.083	0.541	0.57	0.2	0.1	0.05	0.03	0.026	0.021	0.017
	$m = 1.59 - 0.66i$												
0.5		17	8	7	6	12	−17	24	15	12	10	9	8
0.6		−43	−18	−17	−13	−26	35	−52	−34	−27	−23	−19	−17
0.7		−55	−24	−23	−18	−36	56	−72	−46	−37	−31	−26	−23
0.8		−41	−19	−18	−14	−28	41	−59	−36	−29	−24	−21	−18
0.9		−16	−9	−8	−6	−13	8	−27	−15	−13	−11	−9	−8
1.0		9	3	3	2	5	−25	10	6	5	4	3	3
1.1		29	13	12	9	19	−43	41	25	20	17	14	13
1.2		39	19	18	14	27	−40	56	34	29	24	20	18
1.3		36	18	17	14	27	−22	51	32	28	23	20	18
1.4		18	10	10	7	14	1	22	17	15	13	11	10
1.5		−15	−8	−7	−6	−11	16	−22	−14	−11	−9	−8	−7
	$m = m(\lambda)$												
0.5		44	2	0	−1	9	−47	–	–	10	4	1	−2
0.6		−58	4	7	5	−2	66	–	–	−1	10	11	9
0.7		−106	0	5	5	−15	121	–	–	−15	−2	1	5
0.8		−103	−4	1	3	−21	117	–	–	−24	−19	−3	0
0.9		−62	−6	−3	−1	−18	71	–	–	−19	−12	−7	−4
1.0		−3	−5	−5	−3	−7	9	–	–	−7	−6	−5	−7
1.1		57	−1	−4	−4	6	−44	–	–	7	−1	−2	−6
1.2		98	4	−1	−2	20	−83	–	–	21	9	3	−2
1.3		105	8	3	0	27	−108	–	–	30	17	10	1
1.4		62	7	4	1	19	−70	–	–	20	11	7	3
1.5		−38	−2	0	1	−8	22	–	–	−4	−2	−1	1

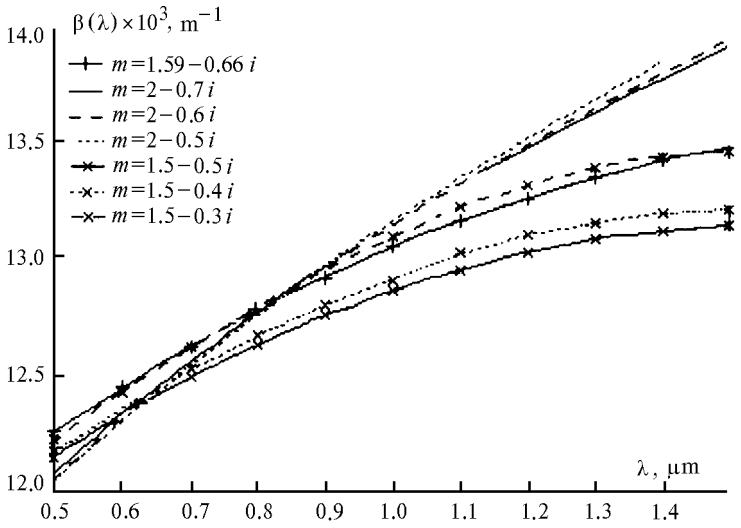

FIG. 2. Spectral dependence $\beta_{ex}(\lambda)$ *for an ensemble of aerosol particles with* $r_m = 0.5$ μm *and* $\sigma = 0.5$ μm *for different values of the complex refractive indices.*

Calculated results demonstrated that the approximation errors in the reference points λ_j never exceeded 0.15%, that is, were 5–10 times less than the standard errors in measuring $\beta_{ex}(\lambda)$ (see Table I). It was also established that the approximation errors decreased when the number of large particles in polydispersion increased.

All this confirms the validity of model (1) for aerosol polydispersions comprising large strongly absorbing particles.

To check additionally model (1) and to specify its application limits, $\beta_{ex}(\lambda)$ was calculated for different values of the refractive index m of strongly absorbing particles. It was found that the spectral dependence $\beta_{ex}(\lambda)$ was in good agreement with model (1) within the entire spectral range of sounding radiation (Fig. 2).

The errors given by Eq. (2) were also analyzed for the extinction coefficient $\beta_{ex}(\lambda)$ that deviated from its true value by ±1% at the wavelengths 0.5, 0.8, 1.2, and 1.5 μm. Calculations were done for carbonaceous particles with the parameters of the particle size distribution $r_m = 0.5$ μm and $\sigma = 0.5$ μm. The sign of the deviation was prescribed arbitrarily at each wavelength. The calculated results showed that the approximation error was no more than 1.5% for arbitrary combination of signs of deviations. This is quite acceptable for the inversion of these data into the particle size spectrum.[11]

To summarize the foregoing, we can state that the spectral dependence of the aerosol extinction coefficient for polydispersions of the atmospheric surface layer (and working rooms, workings, etc.) comprising a great number of large (with size greater than 1 μm) strongly absorbing particles can be approximated by the quadratic dependence with sufficient accuracy in the wavelength range 0.5–1.5 μm. This approximation can be successfully used to complete and to solve the problem of reconstruction of the aerosol particle size spectrum.

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