REDISTRIBUTION OF THE COARSELY DISPERSED AEROSOLS OVER THE SIZE SPECTRUM DUE TO ADVECTION BY EDDY FLUX

R.F. Rakhimov

Institute of Atmospheric Optics,
Siberian Branch of the Academy of Sciences of the USSR, Tomsk
Received January 15, 1991

A technique is described to simulate numerically the redistribution of the coarsely dispersed aerosols over the size spectrum due to advection by eddy flux. Some model estimates of the spatial transformation of the aerosol particle size spectrum affected by gravitational sedimentation are presented.

Entrainment of the air masses from the various water area of the global ocean and continenal climatic zones forms in the atmosphere the regional variety of the optical weather which is changed from pure arid to the oceanic ones. To construct a global (background) optical model of the atmospheric haze, the data on light scattering properties of the oceanic dispersed component provide the basis for the model accounting for the scale of presentation. Meanwhile, it is necessary to have well–founded techniques of an *a priori* estimate of the proportions in which aerosols of different origin, especially coarsely dispersed fraction, are mixed in order to elaborate the possible regional deviations from the global model. The kinetic model of secondary aerosols developed by Rozenberg[5] meets with significant difficulties in predicting the state of the coarsely dispersed aerosol fraction whose particles enter atmosphere virtually already formed.

Obviously, the gravitational sedimentation is important for modifying the size spectrum in the processes of global aerosol transport. This paper being the continuation of the previous studies[1,2], analyzes the problem of the deformations in the size spectrum of the coarsely dispersed particles due to advection by eddy flux. The technique for the analytic approximation of the experimental histograms of the atmospheric haze particle size spectrum developed in several stages starting from integral, linear (in the log–log scale, the so–called Pareto distribution) through the quadratic (lognormal distribution) to the structured fractional (the superposition of many fractions)

$$f(R,\ z,\ t) = \frac{d\Phi}{dR} = AR^{-\nu} \sum_{i=1}^{k} M_i \exp\{b_i[\ln(R/R_i)]^2\}, \qquad (1)$$

where A, M_i, b_i , R_i , and m are the variable parameters.

Depending on the particular problem, different functions are employed to describe the dispersed composition of the haze, in particular, not only the spectral distribution of the particle number density $n(R)$, but also the distribution of particles over sizes of geometric cross sections $s(R)$ and over their volumes $v(R)$. They differ in their modal radii R_i (maxima in the distributions over the indicated parameters) which are widely spaced along the R axis and can be estimated from the integral parameters of the dispersed composition of separate fractions based on the appropriate *a priori* choice of the approximate function (1) or the specific value of the parameter m of the distribution according to the formula

$$R_i^{(\nu)} = (3V_i)^{2/3}/[(4\pi N_i)^{1/6}\cdot]\cdot[(4\pi N_i)^{1/3}(3V_i)^{2/3}/S_i]^{3-\nu}. (2)$$

The dispersity parameter b_i and the normalization factor F_i can be found similarly from the relations

$$b_i = 1/\left\{\ln[(3V_i)^{4/3}(4\pi N_i)^{2/3}/S_i^2\right\}\ ; \qquad (3)$$

$$F_i = A\cdot M_i = S_i/\sqrt{16\pi^3/b_i}\ . \qquad (4)$$

When the aerosol particles are transported and redistributed in the atmosphere, the "old" local substructure distracts and the new one simultaneously forms. It seems that this interrelated unity of processes of destruction of the previous local structure and synthesis of the new one is caused to a considerable degree by the peculiarities of the eddy mixing, namely, by the impingement of the turbulent eddies of various size carrying the individual structural elements of the dispersed composition of the atmospheric haze, namely, its fractions, as a whole.

Model estimates[1,2,3] show that retaining the adequate description of the predicted optical object we can employ the data on integral parameters of the individual fractions of the dispersed composition instead of the measured histograms of the size spectrum assuming *a priori* that the particles on each fraction are distributed lognormally.

Postulating the qualitative stability of the shape of the particle size distribution in the individual fraction, we can reduce the problem of spatial deformations of the dispersed composition of primordial aerosols appearing as a result of mixing in the surface layer near the particle source to the solution of a system of differential equations describing the changes in the integral parameters of the size spectrum in the space of two variables

$$U(z)\frac{\partial N_i(x,\ z)}{\partial x} = \frac{\partial}{\partial z}D(z)\frac{\partial N_i(x,\ z)}{\partial z} - W_{in}(R_i)\frac{\partial N_i(x,\ z)}{\partial z}\ , \quad (5)$$

where $N_i(x,\ z)$ is the aerosol particle number density at the point with horizontal coordinate x and vertical coordinate y; W_{in} is the average velocity of the ordered vertical motion of particles including the Stokes gravitational sedimentation which is estimated for the weighted mean particle radius in of the ith fraction (averaged over the particle number density spectrum). The X axis is collinear to the vector of the mean wind velocity.

The vertical profiles of the horizontal velocity of the air flow $U(z)$ and the coefficient of the eddy exchange $D(z)$

0235-6880/91/05 389-05 $02.00

in this approach were assumed to be linearly dependent of an altitude

$$U(z) = U_0(z); \quad D(z) = D_0 z. \qquad (6)$$

Equation (1) is solved with the following boundary conditions:

a) vertical profile of the particle number density in the inflowing eddy flux

$$N_i(x, z)\big|_{x=0} = N_{i0} \exp(-\beta z^2), \qquad (7)$$

b) horizontal distribution of the particle number density at the altitude $z = 0$

$$N_i(x, z)\Big|_{\substack{z=0\\ x>0}} = N_{i1}(x), \qquad (8)$$

c) the horizontal distribution of the particle number density as $z \to \infty$

$$N_i(x, z)\big|_{z\to\infty} \neq \infty \qquad (9)$$

has the form[6]

$$N_i(x, z) = N_{io}\zeta^{1-p} \exp(-\beta\zeta z^2)\{1 - \Gamma(p,\zeta^2 z^2)\} +$$

$$+ \frac{\alpha^p}{\Gamma(p)}\left(\frac{z}{2}\right)^{2p} \int_0^x \frac{N_{i1}(\xi)}{(x-\xi)^{1+p}} \exp\left[-\frac{\alpha z^2}{4(x-\xi)}\right] d\xi, \qquad (10)$$

where $p = \dfrac{W_{in}}{2D_0}$; $\alpha = \dfrac{U_0}{D_0}$; $\Gamma(p, \kappa)$ is the incomplete gamma function; $\xi = \dfrac{\alpha}{(\alpha + 4\beta\chi)}$.

The rate of particle sedimentation in the Earth's gravitational field depends on the radius R_i, the density of aerosol substance ρ, and the air viscosity η (thereby it depends implicitly on an altitude z at which the particle is located), namely,

$$W_{in} = 2\rho g R_{in}^2 C_{ki}/9\eta. \qquad (11)$$

Since the Knudsen number $\mathrm{Kn} = l/R$ for aerosol particles (l is the mean free path of the molecules) varies from 0.1 to 1000 for the aerosol particles in the surface layer, the Cunningham adjustment factor must be taken into account in Eq. (11)

$$C_{ki} = 1 + \mathrm{Kn}\,(1.257 + 0.4 \exp[-1.1/\mathrm{Kn}]). \qquad (12)$$

Equation (5) is, in fact, the condition of the balance of fluxes of the particle number density. Obviously, the same equations can also be written for $S_i(x, z)$, $V_i(x, z)$ (with the appropriate boundary conditions). Since the modal radii of the integral parameter distributions are related by the inequality $R_{iv} > R_{is} > R_{in}$, the weighted mean Stokes sedimentation rate (averaged over the given parameter) also satisfies the inequality $W_{iv} > W_{is} > W_{in}$. These values determine parametrically the solution of Eqs. (1) – (6) for $V_i(x, z)$, $S_i(x, z)$, and $N_i(x, z)$, respectively.

In analogy with Ref. 3 we consider the process of the advection of "new" particles with enhanced number density taking into account their mixing with the background of "old" particles, whose integral characteristics are horizontally uniform while their profile decreases exponentially with altitude. In our modeling we used the data previously tested in Ref. 1 as the starting background optical model. The advected and the background aerosol particles are identical in their spectral composition, i.e., the conditional vectors (N_{ia}, S_{ia}, V_{ia}) and (N_{ibg}, S_{ibg}, V_{ibg}) are collinear and differ only in length.

In order to estimate the effect of gravitational sedimentation on altering the dispersed composition of different fractions of aerosol, we shall consider as the first approximation of the proposed model the advection of aerosol by the eddy flux near the surface free of aerosol sources, i.e., $N_{i1}(x) = S_{i1}(x) = V_{i1}(x) = 0$. We ignore the rotation of the geostrophic wind in the surface layer.

Data systematized in Table I describe the deformation of the aerosol particle size spectrum in the transversal section of the eddy flux along the Z axis (altitude) at $x = 200$ m. Table II presents such data on the longitudinal distribution of aerosol (along the flux) over the X axis at the altitude $z = 60$ m. The running coordinates are indicated in the first columns of both tables (in tens of meter for z and in kilometers for x). The values of the modal radii R_i are given in units of μm.

TABLE I. Parameters of the three–fraction model (1) vs. altitude

z	R_1	R_2	R_3	b_1	b_2	b_3	F_1	F_2	F_3
0.10	0.033	0.239	2.33+0	1.50+0	1.50+0	1.56+0	1.24+3	1.24+3	1.23+3*
0.35	0.033	0.239	2.39+0	1.50+0	1.50+0	1.50+0	7.48+2	7.49+2	7.50+2
0.60	0.033	0.239	2.41+0	1.50+0	1.50+0	1.48+0	2.56+2	2.56+2	2.57+2
0.85	0.033	0.239	2.41+0	1.50+0	1.50+0	1.48+0	5.14+1	5.14+1	5.16+1
1.10	0.033	0.239	2.40+0	1.50+0	1.50+0	1.49+0	7.87+0	7.87+0	7.89+0

*Here and below $1.23 + 3 = 1.23\cdot10^3$

TABLE II. Horizontal changes in the parameters of the three–fraction model (1).

x	R_1	R_2	R_3	b_1	b_2	b_3	F_1	F_2	F_3
0.05	0.033	0.239	2.40+0	1.50+0	1.50+0	1.49+0	1.91+2	1.91+2	1.92+2
2.55	0.033	0.239	2.34+0	1.50+0	1.50+0	1.55+0	8.35+1	8.35+1	8.26+1
5.05	0.033	0.239	2.33+0	1.50+0	1.50+0	1.56+0	4.65+1	4.65+1	4.58+1
7.55	0.033	0.239	2.32+0	1.50+0	1.50+0	1.57+0	3.27+1	3.27+1	3.21+1
10.05	0.033	0.239	2.31+0	1.50+0	1.50+0	1.58+0	2.55+1	2.55+1	2.50+1

Data in Tables I and II were calculated on the assumption that the density of the aerosol substance remains identical in all three fractions and is equal to the average density[7] $\rho = 2.7$ g/cm^3. According to the above boundary conditions, the integral parameters of the advected particles at $x = 0$ and at $z = 10$ m exceed the background values by the factor of 1000.

The estimates show that only the size spectrum of the coarse fraction ($R > 1.0$ μm) is noticeably modified under the effect of gravitational sedimentation. While the particles of fine ($R < 0.1$ μm) and accumulative ($0.1 < R < 1.0$ μm) fractions are virtually transported by the eddy flux without any qualitative changes in their dispersedcomposition, only the integral aerosol content changes, that is natural for the diffusion of the impurity with enhanced number density. In addition, as for the coarse fraction not only its number density is greater than the background value but also the pronounced qualitative deformations of its size spectrum are found to occur even at the distance $x = 10$ km from the source.

Since the physicochemical composition of the mineral dust particles is quite heterogeneous, we perform the model estimates of the process of the horizontal transport of the coarse fraction with different density of aerosol substance (see Table II).

In this study we assume that the inflowing eddy flux contains three fractions with different densities which were conditionally considered typical of water $\rho = 1.0$ g/cm^3, $i = 1$, for sea salt particles $\rho = 2.18$ g/cm^3, $i = 2$, and also for the mineral dust particles of iron–ore rocks $\rho = 6.9$ g/cm^3, $i = 3$ with the identical parameters of the lognormal particle size distribution at $x = 0$. Data are presented at the altitudes of 10, 35, 85, and 110 m.

If the deformation of the water droplet spectrum is insignificant (columns 2, 5, and 8 in the tables), then the noticeable narrowing of the spectrum (see the increase in the parameter b_2, column 6, $z = 10$ m) is found for the sea salt particles (columns 3, 6, and 9) at altitudes near the surface. These deformations are significant for the iron–ore minerals at all considered altitudes.

TABLE III Horizontal changes in the parameters of model (1).

x	R_1	R_2	R_3	b_1	b_2	b_3	F_1	F_2	F_3
$z = 10$ m									
0.05	4.675	4.535	3.81+0	1.56+0	1.64+0	2.43+0	1.17+3	1.16+3	1.15+3
2.55	4.389	3.963	2.56+0	1.73+0	2.11+0	1.21+1	4.65+1	4.47+1	5.53+1
5.05	4.366	3.933	2.74+0	1.75+0	2.13+0	4.51+0	2.49+1	2.37+1	1.77+1
7.55	4.365	3.943	2.94+0	1.75+0	2.10+0	2.95+0	1.76+1	1.66+1	1.04+1
10.05	4.371	3.966	3.13+0	1.74+0	2.07+0	2.35+0	1.39+1	1.31+1	7.64+0
$z = 35$ m									
0.05	4.833	4.833	5.07+0	1.48+0	1.46+0	1.40+0	4.32+2	4.36+2	4.56+2
2.55	4.576	4.330	3.27+0	1.61+0	1.78+0	3.68+0	4.59+1	4.48+1	4.48+1
5.05	4.538	4.258	3.18+0	1.64+0	1.83+0	3.73+0	2.49+1	2.42+1	2.28+1
7.55	4.524	4.235	3.22+0	1.64+0	1.85+0	3.31+0	1.77+1	1.71+1	1.49+1
10.05	4.518	4.230	3.29+0	1.65+0	1.85+0	2.92+0	1.40+1	1.35+1	1.11+1
$z = 85$ m									
0.05	4.804	4.825	4.95+0	1.49+0	1.48+0	1.42+0	4.45+0	4.46+0	4.48+0
2.55	4.717	4.617	4.07+0	1.54+0	1.59+0	2.08+0	3.88+1	3.85+1	3.83+1
5.05	4.666	4.517	3.79+0	1.56+0	1.65+0	2.42+0	2.31+1	2.28+1	2.22+1
7.55	4.642	4.470	3.68+0	1.58+0	1.68+0	2.54+0	1.68+1	1.65+1	1.58+1
10.05	4.629	4.445	3.65+0	1.58+0	1.70+0	2.55+0	1.35+1	1.32+1	1.24+1
$z = 110$ m									
0.05	4.761	4.729	4.57+0	1.51+0	1.53+0	1.61+0	2.56+0	2.55+0	2.50+0
2.55	4.754	4.704	4.35+0	1.52+0	1.55+0	1.83+0	3.36+1	3.35+1	3.39+1
5.05	4.703	4.596	4.01+0	1.54+0	1.61+0	2.13+0	2.15+1	2.13+1	2.11+1
7.55	4.678	4.543	3.87+0	1.56+0	1.64+0	2.29+0	1.61+1	1.59+1	1.54+1
10.05	4.662	4.512	3.80+0	1.57+0	1.66+0	2.36+0	1.30+1	1.28+1	1.23+1

Table IV presents the result of calculations classificated by the value of the parameter β from Eq. (7) which stipulates the initial vertical distribution of the particle number density in the inflowing eddy flux. The value of the parameter β are indicated in the headlines of Table IV.

Signific narrowing of the spectrum (the parameter b_3 increases from 1.30 to 2.9) and the shift of the maximum toward the small radius (R_3 decreases from 5.29 to 3.47 μm) takes place for small values of the parameter $\beta = 0.002$ for the heavy fraction of the coarsely dispersed particles with $\rho = 6.9$ g/cm. The increase of the values of R_3 at the end of the path is caused by the dominating role of the background particles in the mixture.

When the altitude gradient of the exponential decay of the particle number density increases (β equals to 0.01 and 0.10), the model estimates show that formation of the stable zones of the enhanced number density of the aerosol particles (see change in the parameters F_i) is possible at different distances from the initial vertical of the particle generation (at $z = 60$ the x axis varies from 100 to 500 m).

The model estimates given in Tables I–IV and plotted in Figs. 1–3 in the form of contours show that the size spectrum of the coarsely dispersed fraction acquires a complete spatial structure of both the particle number density (Fig. 1, *b*–3, *b*) and the characteristic radii of the particles (Fig. 1, *a*–3, *a*).

TABLE IV. Horizontal changes in the parameters of the model (1) at the altitude z = 10 m.

x	R_1	R_2	R_3	b_1	b_2	b_3	F_1	F_2	F_3
β = 0.002									
0.05	4.851	4.925	5.29+0	1.47+0	1.44+0	1.30+0	5.24+1	5.29+1	5.56+1
2.55	4.659	4502	3.73+0	1.57+0	1.66+0	2.54+0	4.30+1	4.24+1	4.18+1
5.05	4.615	4.412	3.52+0	1.59+0	1.72+0	2.88+0	2.42+1	2.38+1	2.29+1
7.55	4.595	4.375	3.47+0	1.60+0	1.74+0	2.89+0	1.74+1	1.69+1	1.58+1
10.05	4.585	4.357	3.48+0	1.61+0	1.75+0	2.77+0	1.38+1	1.34+1	1.34+1
β = 0.010									
0.05	4.850	4.927	5.37+0	1.47+0	1.44+0	1.26+0	5.11+0	5.15+0	5.37+0
2.55	4.682	4.554	3.95+0	1.56+0	1.63+0	2.15+0	1.08+1	1.07+1	1.02+1
5.05	4.664	4.552	3.95+0	1.56+0	1.64+0	2.06+0	7.03+0	6.91+0	6.33+0
7.55	4.664	4.527	4.03+0	1.56+0	1.64+0	1.94+0	5.63+0	5.53+0	4.97+0
10.05	4.668	4.540	4.11+0	1.56+0	1.63+0	1.85+0	4.91+0	4.82+0	4.30+0
β = 0.100									
0.05	4.783	4.778	4.78+0	1.50+0	1.51+0	1.49+0	2.81+0	2.81+0	2.78+0
2.55	4.745	4.694	4.47+0	1.52+0	1.55+0	1.67+0	3.50+0	3.48+0	3.36+0
5.05	4.749	4.704	4.53+0	1.52+0	1.54+0	1.62+0	3.11+0	3.09+0	2.99+0
7.55	4.753	4.713	4.56+0	1.52+0	1.54+0	1.60+0	2.97+0	2.96+0	2.86+0
10.05	4.756	4.720	4.58+0	1.52+0	1.53+0	1.59+0	2.90+0	2.89+0	2.80+0

In the surface layer the aerosol particles apart from the Stokes sedimentation can be entrained into the ordered upward motion of the convective heat fluxes and, vice versa, depending on the type of the underlying surface, they can also acquire a hypothetical component of the downward motion as a result of the dry sedimentation.

We consider three variants of calculated results shown in the figures: the convective component W_c dominates (Fig. 1), the dry sedimentation W_d dominates (Fig. 2), and both components compensate each other (Fig. 3).

As can be seen from the countors of the particle number density (F_i in the given case) are more uniform along the eddy flux when the convective component dominates (W_d = 0.06 m/s), though near the underlying surface the particle number density decreases with increase of x whereas we observe the increase in the particle number density when $z > 60$ m. Two last tendencies are manifested more clearly when the particle sedimentation rate increases, i.e., in the second and third variants. The "islands" of the implicit enlargement of the coarsely dispersed aerosol particles coinciding with the regions of the increase in the dispersity (the parameter b_i) located within the region dz = 60–90 m, dx = 600–1200 m are observed for all three variants.

The given results make it possible to consider in more detail only some typical features of the formation of the particle size spectrum of the coarsely dispersed fraction of the haze and to obtain a number of estimates which elucidate the mechanism of the formation of the anomalies in the spatial distribution of mineral particles which are useful for understanding the morphology of the so–called noncondensed clouds in which the particles of ground origin with R = 1.0–4.0 mm predominate. We simulate only the simplest variants when the transported aerosol component has the size spectrum identical to the background. Preliminary model estimates show that spatial deformations of the spectrum exhibit complex structure when heterogeneous aerosols are mixed and when the modal radius and the distribution width differ substantially from the background ones because in this case the integral parameters of the spectrum at different points are summed in various proportions and this fact leads to the ambiguous dynamics of the changes in R_i, b_i, and F_i (see relations (2)–(4)) even in the case of the accumulative fraction.

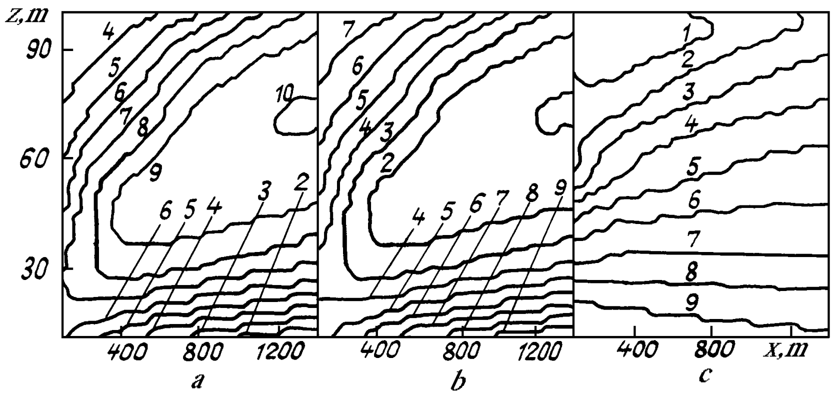

FIG. 1. Contours of the parameters of the spectrum of the coarsely dispersed fraction for W_c = –0.06 m/s *and* W_d = 0; *a) modal radius* R_i, *b) parameter* b_i *and c) parameter* F_i.

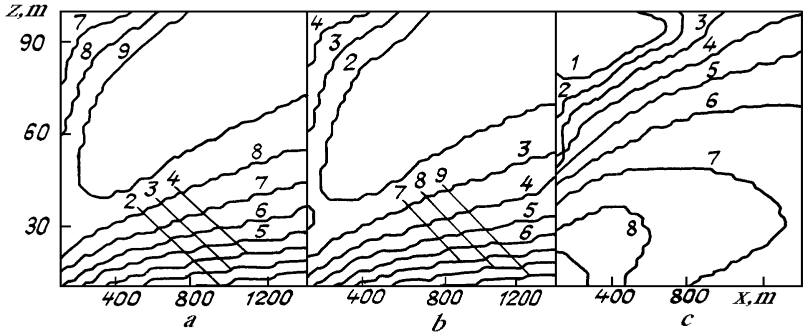

FIG. 2. The same as in Fig. 1 but for W_c = 0.05 *m/s and* $W_d = 0$.

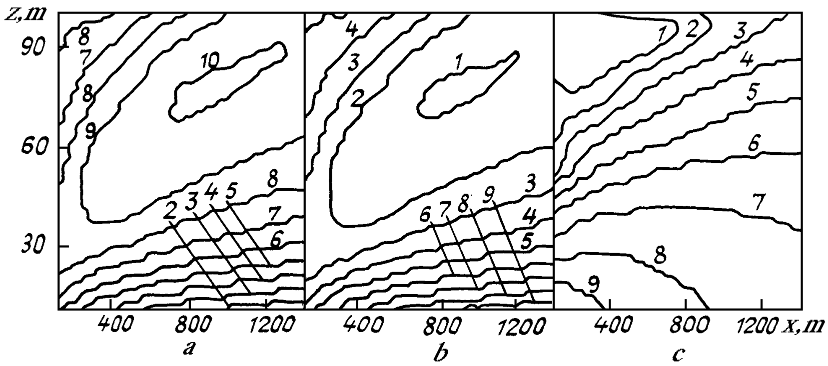

FIG. 3. The same as in Fig. 1 but for $W_c + W_d = 0$.

To study the mechanism of heterogeneous aerosol mixing, it is necessary to perform specific numerical estimates which take into account the differences in the strength and the spectrum of generation of the source as well as specific features of the physicochemical composition of particles determining the integral density of aerosol substance. In the conventional approaches based on the rigorous description of the processes in multiphase media one would face with the severe mathematical difficulties, and one would hardly succeed in overcoming these difficulties without conceptual simplifications. It seems that the developing tendency of the reduction of the model structures is justified and appears to be the natural improvement of the methodology of the prediction of the optical characteristics of the atmospheric aerosols.

The method of the fractional simulation of the atmospheric haze characteristics is one of such appropriate compromises. It makes possible to combine the quite simple description of the particle size spectrum governing the wide variety of local states retaining the qualitative common features (i.e., integrity) of these spectra.

REFERENCES

1. G.M. Krekov and R.F. Rakhimov, *Optical Models of the Atmospheric Aerosol* (Tomsk Affiliate of the Siberian Branch of the Akademy of Sciences of the USSR, Tomsk, 1986).

2. R.F. Rakhimov, Atm. Opt. **3**, No. 4, 305–314, (1990).

3. R.F. Rakhimov, Atm. Opt. **2**, No. 3, 206–212, (1989).

4. G.M. Krekov and R.F. Rakhimov, *Optical–Radar Model of Continental Aerosol* (Nauka, Novosibirsk, 1982).

5. G.V. Rosenberg, Izv. Akad. Nauk SSSR, Ser. FAO **19**, No. 3, 241–254 (1983).

6. S.N. Ermilov, Trudy LGMI, No. 49, 106–118 (1974).

7. L.S. Ivlev and S.D. Andreev, *Optical Properties of the Atmospheric Aerosols* (LSU, Leningrad, 1986).

8. V.E. Zuev, B.D. Belan, V.V. Veretennikov, et al. , Atm. Opt. **2**, No. 7, 605–609 (1989).

9. G.V. Rosenberg, Izv. Akad. Nauk SSSR, Ser. FAO **19**, No. 1, 21–35 (1983).