MODEL ESTIMATIONS OF THE POSTVOLCANIC RELAXATION OF THE OPTICAL PROPERTIES OF THE STRATOSPHERIC LAYER

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A technique for numerical simulation of the variability of optical–radar properties of stratospheric aerosols during the postvolcanic relaxation of the aerosol layer is discussed. The model estimation of the simultaneous effect of gravitational sedimentation and macroturbulent spreading of the eruptive cloud on the optical-radar properties of aerosols are presented.

The empirical[1-4] data on spatial and temporal trends in the integral parameters of dispersed composition of the stratospheric aerosol layer (SAL) were tested in Ref. 5 as the starting point for the numerical optical model. The main attention was devoted to the analysis of the effect of the vertical gravitational stratification of profiles of integral parameters. Considering the empirical data which demonstrated the difference in the rate of degradation of number and volume densities of the aerosol particles in the process of postvolcanic relaxation of the aerosol layer from the perturbed state to the equilibrium, the internal (kinetic) transformations of the particle size spectrum were indirectly taken into account in the calculations.

The optical model of the SAL proposed in Ref. 5, possesses not only some advantages over the others (it is, in fact, the dynamic version) but also some drawbacks. In addition to the previous results, in this paper the variability of the optical radar characteristics of the SAL is analyzed on the basis of the numerical model estimates taking into account the macroturbulent vertical spreading of the eruptive clouds.

The technique of fractional simulation of the dispersed composition of the atmospheric haze, which is used here, was described in ample detail in Refs. 5–7. As previously, the spatial deformations of the size spectrum of aerosol particles are evaluated from the transformations of the integral parameters: number density of the particles in the ith fraction $N_i(z, t)$ and their cumulative cross section $S_i(z, t)$ and volume $V_i(z, t)$.

The initial mathematical base of the model is the equation of macroturbulent diffusion of aerosol particles in the stratospheric layer[8]

$$\frac{\partial N_i(z, t)}{\partial t} + \frac{\partial}{\partial z}\left[W_{in}(z, t) \cdot N_i(z, t) \right] = \frac{\partial}{\partial z} D(z, t) \frac{\partial N_i(z, t)}{\partial z}, \quad (1)$$

where $W_{in}(z, t)$ are the velocities of the vertical motion of particles in the ith fraction, evaluated from the value of the modal radius R_i of the lognormal function of the number density distribution $n_i(R)$ and $D(z, t)$ is the eddy diffusion coefficient, which, according to Ref. 8, depends weakly on the altitude in the lower and middle regions of the stratosphere. The values of D vary from 0.1 to 0.6 m^2/s accounting for the seasonal variations.[9]

In general, the vertical velocity component of the ordered motion of aerosols represents the sum of the rate of gravitational sedimentation and the mean wind flows. The latter is negligible in the stratosphere, and can be practically set equal to zero at long times. The rate of gravitational sedimentation due to the fact that the Knudsen number varies within fairly wide limits is given by the Stokes–Cunningham formula

$$W_v = \frac{2\rho g R^2}{9\eta}\left(1 + C_{ki}\, Kn\right), \quad (2)$$

where R is the particle radius, g is the acceleration due to gravity, ρ is the density of the aerosol substance, η is the dynamic viscosity of air, C_{ki} is the value of the Cunningham adjustment factor averaged over the size spectrum of the particles in the ith fraction

$$C_{ki} = 1.257 + 0.40 \exp(-1.10\ /Kn), \quad (2a)$$

$Kn = l/R$ is the Knudsen number and l is the mean free path of the air "molecules".

Assuming that the mean free path for the air "molecules" changes following the barometric law with the constant temperature T independent of the altitude, one can use the following generalization for the vertical rate W_v:

$$W_v = -\frac{2\rho g R^2}{9\eta} - \mathrm{f}(2\rho g R, 9\eta)\, C_{ki}\, l_0 \exp\left(\frac{mgz}{k_B T}\right) = -A - B \exp(\gamma z), \quad (3)$$

where m is the mass of air "molecules", K_B is Boltzmann's constant and l_0 is the mean free path of the air "molecules" in the standard atmosphere.

The solution of Eq. (1) with the boundary conditions

$$N_i(z, t)\big|_{t=0} = N_{i0}\delta(z - z_0), \quad (4)$$

$$N_i(z, t)\big|_{t\to\infty} \to 0, \quad (5)$$

can be represented in the form[8]

$$N_i(z', t) = \frac{I(t)}{\sqrt{4\pi D \int_0^t I^2(\tau)\mathrm{d}\tau}} \exp\left\{ -\frac{z'^2 I^2(t)}{\sqrt{4D \int_0^t I^2(\tau)\mathrm{d}\tau}} \right\}, \quad (6)$$

where

$$z' = z - z_0(t); \quad (7)$$

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$$z_0(t) = z_0 - At - \gamma^{-1}\ln\left[1 + A^{-1}B\exp(\gamma z_0)\right]\left[1 - \exp(-\gamma At)\right]; \quad (8)$$

z_0 is the initial height of the aerosol emission;

$$I(t) = \exp\left\{\int_0^t B\gamma \exp\left[\gamma z_0(\tau)\right]d\tau\right\}. \quad (9)$$

Table I presents the values of $W_v(R)$ calculated from Eqs. (2) and (2a), according to which the values of W_v for the coarse fraction (columns 5 and 6) exceed, by more than a factor of 100, similar values from the accumulative fraction (columns 3 and 4) and by more than a factor of 1000 those from photochemical fraction (columns 1 and 2).

TABLE I.

R, μm	W_v, m/s	R, μm	W_v, m/s	R, μm	W_v, m/s
0.027	2.0E–6*	0.340	8.2E–5	1.887	2.1E–3
0.067	6.2E–6	0.742	3.5E–4	3.425	6.8E–3
0.106	1.2E–5	1.097	7.3E–4	4.615	1.2E–2

*Here and below 2.1E–6 denotes $2.1\cdot10^{-6}$.

We considered not only the first order gravitational sedimentation deformations of $n(R, z, t)$ related to fraction by fraction stratification of the profiles of integral parameters, but also the second order effects produced by stratification of the spectrum moments within each separate fraction, i.e., $N_i(z, t)$, $S_i(z, t)$, and $V_i(z, t)$.

It is evident that equations analogous to Eq. (1) with corresponding boundary conditions can be written also for the next spectrum moments $S_i(z, t)$ and $V_i(z, t)$. However, since $R_{iv} > R_{is} > R_{in}$, the mean values of the Stokes rate of sedimentation of the particles, which determine the solution of Eqs. (1)–(9) and, finally, the spatial redistribution of the spectrum moments $N_i(z, t)$, $S_i(z, t)$, and $V_i(z, t)$, differ also, namely, $W_{iv} > W_{is} > W_{in}$.

Since the chemical composition of the eruptive cloud has complex nature, the profiles of the integral parameters were predicted on the basis of the model (1)–(9) simultaneously for six subfractions of aerosol (the size intervals of accumulative and coarse fractions included three subfractions, which were subsequently additively mixed to form two main fractions). In this case the starting values of the integral parameters were selected so that the values of the modal radii of the subfractions were uniformly distributed over the interval 0.05–5.0 μm. Thus, varying the weight content of each subfraction, one can simulate the process of spreading the eruptive cloud with complex size distribution of aerosol particles.

Subsequently, during the macroturbulent spreading of the cloud from the localized region, the "anomalous" particles are mixed with the background ones. This effect was also taken into account in the model by addition of the values of integral parameters calculated from Eqs. (1)–(9) and the appropriate values of the background model. When determining the background terms, the generalized empirical data tested in Ref. 5 were used as initial profiles of the particle size spectrum of the SAL in constructing the background optical model.

Finally, within the framework of the hypothesis of additive mixing in the numerical model, several levels of initial localization of the eruptive cloud were considered with possible nonsimultaneous start of the processes at different altitudes.

The 12–29 km altitude range was considered in the calculations as most typical of volcanic aerosol emissions. Taking into account the reference data, the values of main parameters of the problem were the following: $T = 240$ K, $\eta = 1.5\cdot10^{-5}$ N s/m^2, $l_0 = 5.3\cdot10^{-8}$ m, and r = $2.3\cdot10^3$ kg/m^3.

The results shown in Figs. 1 and 2 illustrate the deformations of the size spectrum of the particles of accumulative and coarse fractions of the stratospheric aerosol during 210 days in the process of evolution of two peaks of enhanced concentration localized initially at the altitudes of 21 and 26 km.

The calculations showed that the factor of gravitational sedimentation is not so important for deformations of the size spectrum of accumulative fraction (Fig. 1), as for the coarse fraction (Fig. 2). All of the most important salient features of the deformation of the size spectrum of accumulative fraction are governed, to a considerable extent, by mixing of the background and "anomalous" particles. It should be noted that in the calculations we considered the case in which the aerosols with different initial localization were assumed to be similar in the dispersed composition, i.e., the initial spectrum of accumulative (and coarse) fraction of the emission at the altitudes of 21 and 26 km agree qualitatively.

Within the 20–22 km and 25–27 km altitude ranges with enhanced number density of "anomalous" particles (Fig. 1*a*) the modal size of particles of the accumulative fraction at the start of the process was close to the initial one, and in the process of vertical spreading of the local anomalies produced by macroturbulent diffusion, gradually approached the background value. The results of calculations also showed that the sedimentation of the layer with enhanced concentration as a whole is more typical of the particles of accumulative fraction. This fact was especially pronounced when analyzing the change in the parameters of the size spectrum within the 14–18 km altitude range (Fig. 1*a* and *c*). Simultaneously, in the course of evolution of the process at altitudes $z > 24$ km noticeable narrowing of size spectrum was observed (Fig. 1*b*).

For weak turbulent mixing ($D = 0.125$ m^2/s), the anomaly with enhanced concentration of the particles of accumulative fraction could exist during two and even more years (Figs. 1*c* and 3*c*), while for the coarse particles (Fig. 2*c*) the layer disappeared during half a year

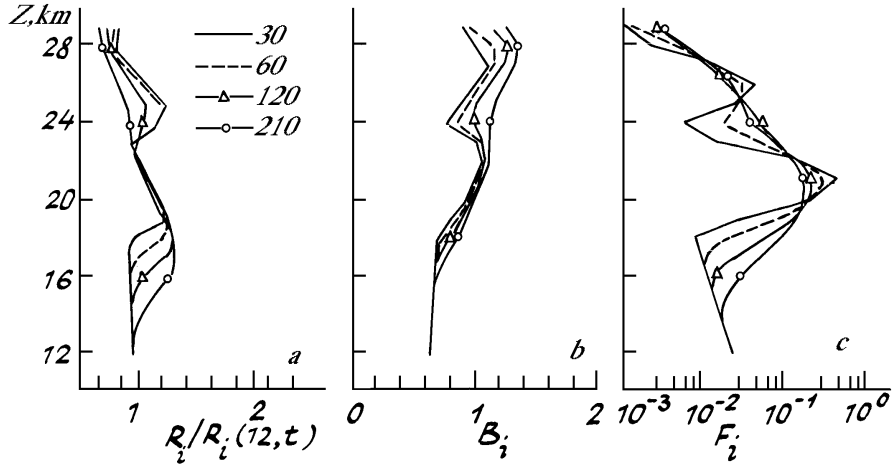

FIG. 1. Transformation of the parameters of the particle size spectrum of accumulative fraction of the SAL: a) normalized size of the particles $R_i/R_i(12, t)$. The current time of the process t = 30, 60, 120, and 210 days is indicated by the figures, b) dispersity parameter b_i, and c) parameter F_i (according to Ref. 7).

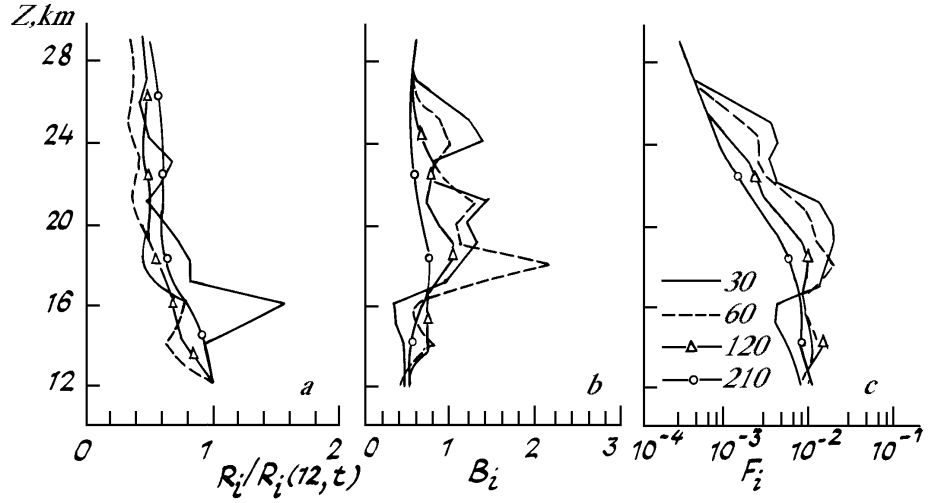

FIG. 2. Transformations of the parameters of the size spectrum of the coarse fraction of the SAL. The analyzed characteristics are the same as in Fig. 1.

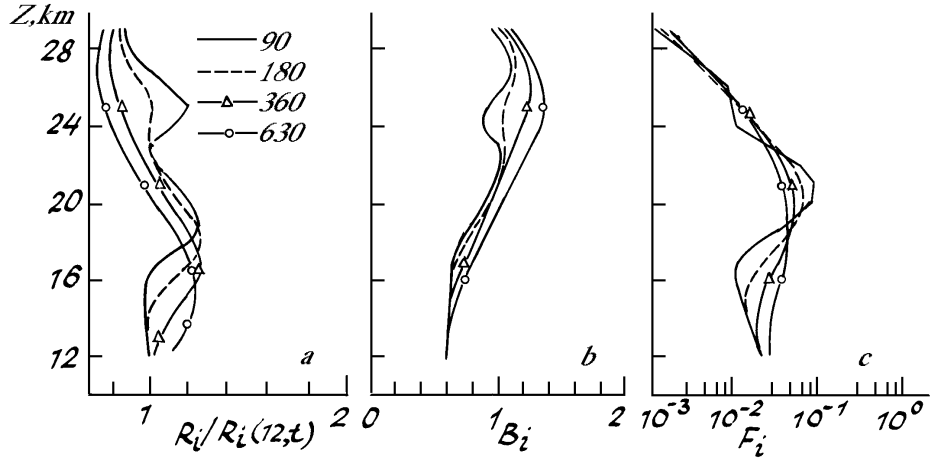

FIG. 3. The change in the parameters of the size spectrum of accumulative fraction of the SAL during 630 days.

In the case of evolution of the layer of coarse particles, the calculations showed the possible formation of aerosol layers (Fig. 2*b*, $z = 18$ km, calculated data for $t = 60$ days) with quite narrow size spectrum (the effect of monodispersity) which could gradually disappeared at the next stages of the process.

Certainly, such complicated and ambiguous transformations of the size spectrum of particles of the SAL must be especially pronounced in the changes of the optical radar properties of the stratification.

On the basis of the obtained data on the deformations of the size spectrum of the SAL (Figs. 1 and 2) the profiles of aerosol extinction coefficient β_t, in km^{-1}, lidar ratio $L_r = \beta_\pi/\beta_t$ (where β_π is the volume backscattering coefficient[5]), and Angstrom's coefficient ν in the approximation of spectral dependence of β_t by the power law function

$$\beta_t(\lambda) = \beta_{t0}(\lambda/\lambda_0)^{-\nu} \qquad (10)$$

were calculated.

The estimates for the wavelengths of laser sensing $\lambda = 0.53$, 0.6943, 1.06, and 10.6 μm are shown in Figs. 4 – 7, respectively. The comparison of calculated data (Figs. 1*c* and 2*c* and Figs. 4*a* and 5*a*) showed that the altitude and temporal changes of the extinction coefficient in the visible range were determined primary by the evolution of the composition of the accumulative fraction. Note the significantly irregular altitude behavior of the lidar ratio. At the same time, the profiles of the optical radar characteristics in the IR ($\lambda = 10.6$ μm), shown in Fig. 7*a*, are similar to the profiles obtained for coarse fraction (Fig. 2*c*).

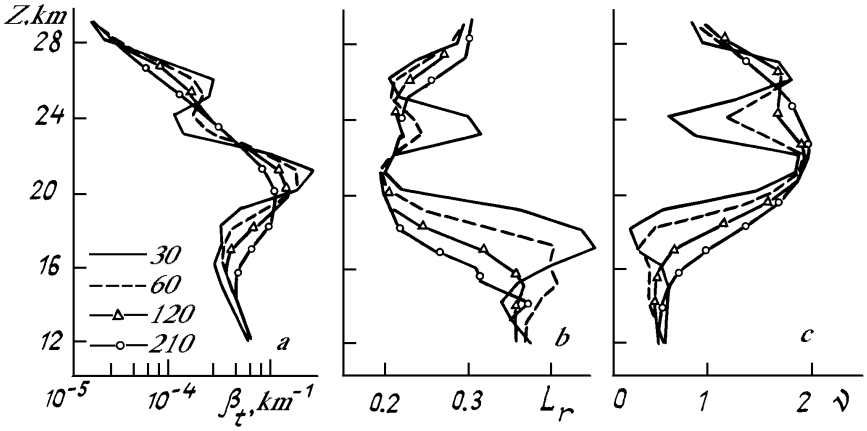

FIG. 4. Transformations of the vertical profiles of light scattering parameters for atmospheric aerosol at the wavelength $\lambda = 0.53$ μm*: a) extinction coefficient* β_t *(km); b) lidar ratio; and c) Angstrom's coefficient* ν*. Time interval is the same as in Fig. 1.*

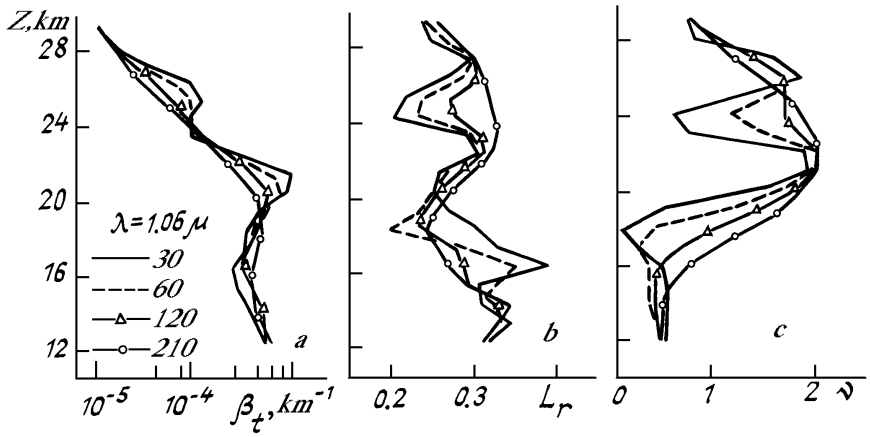

FIG. 5. Transformations of the vertical profiles of light scattering parameters for atmospheric aerosol at the wavelength $\lambda = 0.6943$ μm*. Notations are the same as in Fig. 4.*

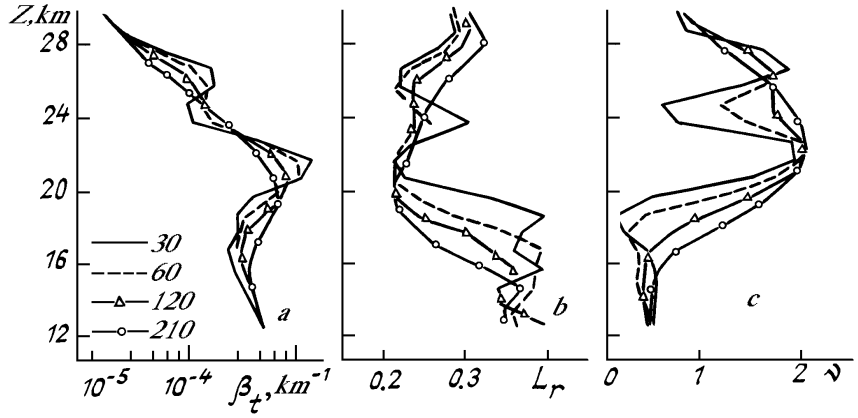

FIG. 6. Transformations of the vertical profiles of light scattering parameters for atmospheric aerosol at the wavelength $\lambda = 1.06$ μm. *Notations are the same as in Fig. 4.*

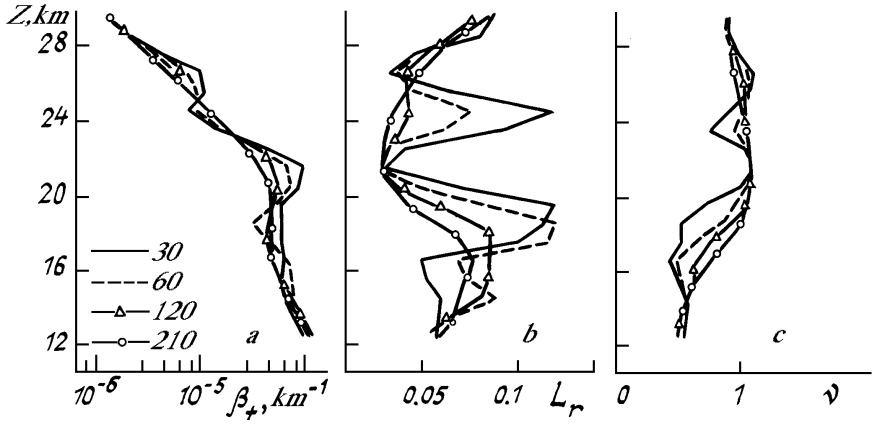

FIG. 7. Transformations of the vertical profiles of light scattering parameters for atmospheric aerosol at the wavelength $\lambda = 10.6$ μm. *Notations are the same as in Fig. 4.*

The results of calculations showed that when the strength of emission of the coarse fraction increases twice, the optical properties of stratospheric aerosols at the wavelength $\lambda = 1.06$ μm are formed by parity contributions of the properties of both fractions.

It is evident that the results of simulation depend strongly not only on the strength of emission and physical and chemical properties of element composition (density of substances and weight percent of fractions) but also on the intensity of eddy mixing. The analysis of the effect of the above–indicated factors presupposes quite a comprehensive study which remains outside the scope of initial description of the technique.

Taking into account the great variety of the variants of simulated process, it is expedient to fill the optical model of the SAL with the matter when the practical needs appear.

The proposed approach to the simulation of aerosol processes in the atmosphere is, in fact, an attempt to find new (intermediate) quantitative level of description of the properties of the atmospheric haze, namely, fractional, in which we consciously neglect the fine subfractional structure of the size spectrum. It is evident that in this case we have a rough approximation of the properties of the object, schematization of real microphysical processes. However, as the model estimates showed, the compromise can be justified, since the error of predicted optical image does not exceed possible instrumentation errors.

And, finally, the analysis of the process of postvolcanic relaxation of the SAL in the frameworks of two dimensional version convinces us of the urgency of further development of the model and of the investigation of gravitational sedimentation factor taking into account the horizontal spreading of local aerosol emission resulting from the eddy mixing, geostrophic drift of particles and local air jets.

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