# MODELLING OF THE AEROSOL PROCESSES FOR FORECASTING THE VERTICAL VARIABILITY OF LIGHT SCATTERING PARAMETERS

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*Received December 25, 1994*

*The problems are discussed on the dynamical modelling of vertical variations in the aerosol optical and microphysical properties as well as the methodology of constructing the information and modelling systems dealing with the aerosol light scattering properties. The question of the influence of gravitational sedimentation on the structure of aerosol anomalies under turbulent diffusion at different heights is considered in detail. A reduced approach to modelling of the coagulation transformation of particle size spectra is justified within the context of forecasting possible variations in the optical properties of hazes.*

## 1. INTRODUCTION

The aerosol component, albeit making a negligibly small portion of the air mass [$\mu g/m^3$], is one of the main factors of the instability of optical situation in the atmosphere, thus being the main factor of the light signal transformations. Atmospheric aerosol, as an open subsystem with a very developed surface, is the main factor forming the climate in the process of continuous evolution and interaction with different geophysical fields.

The cycles, with various durations, of renewal of the disperse component of the atmosphere due to income of new portions of aerosol–producing substances into the atmosphere and their spatial redistribution occur against the background of already existing ones. As a result, it is impossible to describe the process as whole[1–5] without taking into account possible interactions between them. It is practically impossible to encompass all the processes determining the state of disperse component of the atmosphere at a time. Evidently, for successful solving this problem it is necessary, first, to consider the question on the background processes of that type.

Light scattering parameters of the background aerosol component have been calculated in Ref. 1 based on a systematization and generalization of data on aerosol microstructure in the height range up to 90 km collected on a long–term scale.

Analysis of the background model showed physically good reliability of the estimates obtained and their adequacy to actual properties of atmospheric hazes. This, in turn, has allowed the diagnostics of the optical properties of a particular haze to be made in the context of estimation of their possible deviations from the background component.

In particular, when constructing the model, some preliminary estimates of possible deviations of the optical weather from the background one have been made on the basis of empirical data for industrial centers, different regions of the Ocean taking into account the wind velocity effect, and for a continent with the gradation of data with respect to relative humidity. Dynamics of the stratospheric aerosol optical properties is presented for the post–volcanic relaxation. However, the bulk of numerical data has reached its critical value to be tabulated even for that small number of typical situations.

Evidently, to develop dynamic versions of optical aerosol models, approach to formation of the informational systems different than that used in the models from Refs. 16–18 available is needed, when the data on aerosol light scattering should not be set in the final form, but could be modeled by a user on the basis of a compact database invoking current information.

This paper generalizes some methodical findings of the problem on modelling the aerosol processes that determine the spatiotemporal variations of the optical properties of disperse component of the atmosphere.

## 2. PARAMETRIC DETERMINATION OF ATMOSPHERIC AEROSOL MICROSTRUCTURE

In the beginning of seventieths the basic conceptions on the aerosol nature were essentially changed due to the admission of the important role of secondary processes in forming the fine–dispersed fraction in the atmosphere. It became clear that the conception of aerosol as a conservative admixture obviously does not stand up.[19–20]

Covering in its variety the size range from molecular complex–clusters of tens of angstroms to dust particles of hundreds of microns, aerosol occupies the transition zone between the micro– and macroscales of structure. The state of aerosol phase in the atmosphere is regulated by a combination of exchange processes both inside a gas–aerosol mixture cell at the micromolecular level and between stratifications and typical formations at the mesoscale level, including preliminary photochemical formation of aerosol producing compounds in the gas phase.[11,19,20]

The molecular complex–clusters[21] are formed due to homogeneous and heterogeneous nucleation from molecules. Then the optically significant aerosol particles are formed from them due to coagulation growth[22,23] and condensation swelling. In addition, the aerosol particle size spectrum is transformed due to income of new particles from primary sources as well as due to gravitational sedimentation, regular convective and advective transfer, turbulent diffusion, i.e., due to the mixing and dispersal.

This process of transformations can mathematically be represented in the form of an integral–differential equation describing the resultant effect of the most important of the aforementioned factors[8]:

$$\frac{\partial n(v, \mathbf{R}, t)}{\partial t} + \Delta \mathbf{V}\, n(v, \mathbf{R}, t) + \frac{\partial \left\{ I(v, t)\, n(v, \mathbf{R}, t) \right\}}{\partial v} =$$

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$$= \nabla D \nabla n(v, \mathbf{R}, t) + \frac{1}{2} \int_0^v K(\tilde{v}, v - \tilde{v})\, n(v, \mathbf{R}, t)\, n(v - \tilde{v}, \mathbf{R}, t)\, \mathrm{d}v -$$

$$- \int_0^\infty K(\tilde{v}, v)\, n(v, \mathbf{R}, t)\, n(\tilde{v}, \mathbf{R}, t)\, \mathrm{d}\tilde{v} - \nabla \mathbf{V}_g\, n(v, \mathbf{R}, t) . \quad (1)$$

The first two terms in the left–hand side describe the transformation of the size spectrum due to regular (advective or convective) transfer of particles; in the case of imcompressible flow, the second term is transformed to $\mathbf{V}\nabla n(v, \mathbf{R}, t)$, where $\mathbf{V}$ is the average velocity of the flow, $v$ is the particle volume, $\mathbf{R}$ is its coordinates, and $t$ is the time. The influence of the third term is determined by the intensity of the process of condensation growth of particles $I(v, t) = \mathrm{d}v/\mathrm{d}t$. The first term in the right–hand side is caused by the turbulent mixing process, where $D$ is the turbulent diffusion coefficient. Two next integral terms describe the process of coagulation transformation of the aerosol disperse structure, where $K(\tilde{v}, v)$ determines the intensity of particles coagulation. The last term describes the effect of different force fields, for example, gravitational.

It is difficult, using Eq. (1), to solve the problem of forecasting the dynamic variations in aerosol particle size spectrum without conceptual simplification of mathematical statement of the problem and isolation conditions, under which effects from different factors dominate, even for local volumes. So the mathematically justified reduction of the model constructions and preliminary analysis of the priority significance of different factors are important stages in approaching to solution of this problem.

Attempts of constructing the versatile model of $n(r)$ for the atmospheric haze based on Friedlander hypothesis[11,25–28] of keeping the shape of $n(r)$ unchanged undertaken in Refs. 29–31 did not give any convincing and unambiguous results. It has been shown that in order to reach the automodel properties, i.e., the quasiequilibrium shape of the spectrum

$$\frac{\mathrm{d}N}{\mathrm{d}r} = n(r, t) = \psi(t)\, \eta(r) , \quad (2)$$

in solving the kinetic equation, one needs for a time interval of some days, and so it is necessary to take into account not only inner aerosol–to–gas exchange processes, but also the external synoptical meteorological or mesoscale ones.

Junge,[32] who preferred the stochastic nature of the mechanism of formation of the atmospheric haze structure, suggested a hypothesis, according to which the regularity of the size spectrum shape is most likely to be the result of mixing the aerosols of different origin, and its structure can be represented by the statistical sum of the spectra that directly characterize individual peculiarities of the primary sources of aerosol.

The crisis in theoretical and numerical models of the disperse component (circumstantially formed classification of its typical forms) that has taken shape recently is caused by the developed, as the real data appeared, contradiction between the narrow frameworks of the model conception of atmospheric aerosol that interprets its local structure as a homogeneous uniform mixture of polydispersions and the wide limits of variations in its actual physico–chemical properties.

Assessing the prospects of the development of the models of atmospheric haze, one should accept the fact that neither the concept by Friedlander, who tried to assert the dominating role of microprocesses inside a disperse system in forming the atmospheric aerosols, nor the Junge concept, which considers the specific peculiarities of the haze microstructure to be the result of interaction between regional sources and external geophysical factors, can not provide the physically meaningful solution to the problem. At the same time, a mechanical combination of both approaches leads to a complex problem of allowing for the processes of different characteristic time.

The necessity of simultaneously taking into account all the exchange processes occurring in a gas–disperse mixture and between the cells of aerosol field under the perturbing action of regional geophysical factors gives rise to the problem on finding generalized parameters convenient for a parametrization of local variations and modelling of the mesoscale ones, as well for using them as predictors of the optical properties of the atmospheric aerosol.

Use of the number density of aerosol particles, $N(\mathrm{cm}^{-3})$, as a single criterion for estimating the energy losses of radiation when propagating through the aerosol atmosphere leads to noticeable errors, because this value is not a comprehensive characteristics of medium turbidity, and the accuracy of its measurement depends on the resolution ability of the instrumentation used.

As the model estimates[1] show, the data on quantitative content of the aerosol phase represented by three values, namely, the number density $N(\mathrm{cm}^{-3})$, surface density $S(\mu\mathrm{m}^2 \cdot \mathrm{cm}^{-3})$, and volume density $V(\mu\mathrm{m}^3 \cdot \mathrm{cm}^{-3})$, contain much more information necessary for adequate forecasting of the aerosol light scattering parameters.

In the approach proposed here we refuse from a relatively narrow framework of monoelement determination of the disperse structure and use instead the synthesis of $f(r)$ fraction–by–fraction based on to the integral parameters $\hat{Q}_i = \{N_i, S_i, V_i\}$ using the parametric model of the form

$$f(r) = \frac{\mathrm{d}\hat{Q}}{\mathrm{d}r} = A\, R^{-\nu} \sum_k^k M_i \exp\left\{ - b_i [\ln(r/r_i)]^2 \right\} \quad (3)$$

as an approximate. The prototype for the elements of this model is the well known lognormal distribution.

As the estimates[1] show, use of the model analog (3) constructed based on the integral parameters, instead of actual size spectrum measured in the form of a histogram, introduces the error into the estimate of its optical image not greater than 10–15%.

Depending on the problem to be solved, different variables are used for describing the disperse structure of a haze. In particular, it is not only size spectrum of the number density $n(r)$, but also of the surface $s(r)$ and volume $v(r)$ of particles. Maxima of the distribution over these parameters (modal radii $r_i^{(\mathrm{m})}$) are essentially separated on the size scale and can be estimated from the integral parameters of a fraction by a proper selection of the values of the parameters $v$ according to formula

$$r_i^{(\nu)} = (3\, V_i)^{(2\nu - 4)/3}\, (4\pi\, N_i)^{(2\nu - 7)/6}\, (S_i)^{(5 - 2\nu)/2} . \quad (4)$$

It is easy to estimate other parameters of the distribution (3) assuming the following form for them

$$1/b_i = \ln\left\{ (3\, V_i)^{2/3}\, (4\pi\, N_i)^{4/3}\, S_i^{-2} \right\} ; \quad (5)$$

$$F_i^{(\nu)} = AM_i = (3\, V_i)^{(3 - \nu)(1 - \nu)/3}\, (4\pi\, N_i)^{(\nu - 3)(\nu - 4)/6} \times$$

$$\times\, S_i^{(1 - \nu)(\nu - 4)/2} \,/\, \sqrt{16\pi^3 / b_i} . \quad (6)$$

When selecting the empirical data on the atmospheric aerosol microstructure, the method of fraction synthesis of the resulting spectrum based on the integral parameters was

used for validation of the background model,[1] what allowed us to make use of the data obtained by means of the instrumentation of different functional purposes and often limited in the information content.

Another one important factor regulating the variability of optical properties of the atmospheric haze are optical constants of the particulate matter.

In the context of the problem discussed, the method of model estimation of the spectral dependence of the complex refractive index (CRI) based on the *a priori* analysis of the spectra of molar absorption coefficients $\kappa_j(\lambda)$ of different components of aerosol substance has an important advantage because it makes it possible to construct numerically a synthetic dependence $\kappa(\lambda)$ more accurately (taking into account the behavior of $m(\lambda)$ near the absorption bands of aerosol substance), based on the knowledge of specific chemical composition of the aerosol substance. That means that it is possible to take into account possible variations in water content of the disperse phase. Once the function $\kappa(\lambda)$ is known in a wide spectral range, one can use the classical Kramers–Kronig disperse relationships to estimate numerically the spectral dependence of the CRI real part,[3] $\mu(\lambda)$.

When forecasting the aerosol light scattering parameters, the optical constants of particles were determined separately, in particular, for accumulative and coarse fractions according to the first and second versions of the synthetic model of CRI developed by Ivlev[3] for the chemical composition of the continental haze. When analyzing the regional peculiarities, the data on CRI of soot, mineral dust, silicate, sulfate, and organic compositions[3,33] were used in calculations.

## 3. COAGULATION TRANSFORMATION OF THE SIZE SPECTRUM

The kinetics of coagulation transformation of the size spectrum of atmospheric aerosol was modeled based on the Smolukhovskii integrodifferential equation supplemented with the terms describing the sink and source of particles

$$\frac{\partial n(x, t)}{\partial t} = \frac{1}{2}\int_0^x K(x, x - x')\, n(x - x', t)\, n(x', t)\, \mathrm{d}x' -$$

$$- n(x, t)\int_0^\infty K(x, x')\, n(x', t)\, \mathrm{d}x' + \gamma\, n_j(x) - \beta\, n(x, t)\,, \quad (7)$$

where $K(x, x')$ is the coagulation coefficient of particles of the size $x$ and $x'$; $n(x, t)$ is the particle size spectrum; $n_j(x)$ is the spectrum of particles generated by a source; $\beta$ is the effective rate of the particle sink; $\gamma$ is the rate of generation of new aerosols. For complex spectra the solution is only possible within the framework of iterative methods. By splitting the process into subprocesses for individual sub–fractions and integrating the Smolukhovskii equation, and using the effective value of the coagulation constant

$$K = \frac{4\,k\,T}{3\,\eta}\left[1 + \exp(1/2\, b_i) + l_0\, C_k\{\exp(1/4\, b_i) + \right.$$

$$\left. + \exp(5/4\, b_i)\}/r_i\right], \quad (8)$$

where $k$ is the Boltzman constant, $T$ is air temperature, $\eta$ is the viscosity of air, $l_0$ is the free path of molecules, and $C_k$ is the Cunningham correction, one can derive a system of differential equations for forecasting the dynamics of variations in the integral parameters of the size spectrum[3,6]

$$\frac{\partial N_i}{\partial t} = -\frac{K}{2} N_i^2 - \beta\, N_i + \gamma\, N_j\,; \quad (9)$$

$$\frac{\partial L_i}{\partial t} = -\beta\, L_i + \gamma\, L_j\,; \quad (10)$$

$$\frac{\partial W_i}{\partial t} = -\frac{K}{\pi} L_i^2 - \beta\, W_i + \gamma\, W_j\,, \quad (11)$$

where $N_i$, $L_i$, and $W_i$ are different moments of the particle size distribution function $n(x, t)$.

The common solution of Eqs. (9)–(11) is more convenient for an operative estimation of the consequences of the intensification of the process when anomalous aerosol accumulations appear. This approach was used in the analysis of the process of transformation of the smoke aerosol.[36] Comparison of the calculational estimates with the data of laboratory measurements (Fig. 1) in analysis dealt with the kinetics of the process of thermal sublimation of smoke aerosols made it possible to obtain a result that is worth noting. For achieving an agreement between theoretical estimates and empirical data it is necessary to assume that only every third collision of two particles results in their coagulation to one particle.

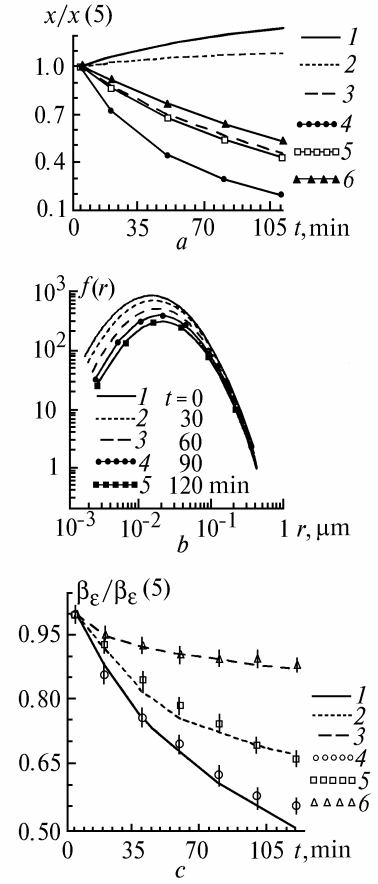


*FIG. 1. Dynamics of coagulation modification of the size spectrum parameters, extinction coefficient $\beta_\varepsilon$ of smoke aerosol in a closed volume at the wavelength $\lambda = 0.6943$ μm: a) parameters of the size spectrum: modal radius $r_d$ (1), spectrum width $b_d$ (2), parameter $f_d$ (3), integral attributes $N_d$, $S_d$, and $V_d$ (4–6); b) size spectrum shape; c) transformed values $\beta_\varepsilon$ ($t$ = 5 min) calculated for 0.007, 0.03, and 0.22 $m^{-1}$, respectively; curves 4–6 are for data of laboratory measurements.[36]*

The advantage of the method we propose is in the possibility, it provides, to analyze the process for several hypothesis on the coagulation mechanism, because this issue remains open, despite of the aforementioned results. The hypothesis on invariance of the total volume and integral linear size of the disperse system is the extreme variant among them.

Although the latter version can seem to be unlikely from the first sight, however, one cannot ignore it because aerosols in the atmosphere are always immersed into the water vapor whose mass is several orders of magnitude greater than that of aerosol phase. Formation of microcapillars at the mutual cohesion can stimulate the process of heterogeneous micropore condensation of moisture, providing an increase in the aerosol mass.

## 4. RELATIVE HUMIDITY

Analysis of calculational data, in the context of single–parameter model of the variability of the scattering phase function shape developed by Gorchakov and Sviridenkov[37] and Panchenko,[38] showed that the theoretically forecasted values of the coefficients in linear regression $\ln[g_a(\vartheta, \lambda)] = K(\vartheta, \lambda)\ln\beta_\varepsilon(\lambda) + \ln[C(\vartheta, \lambda)]$, where $g_a(\vartheta, \lambda)$ and $\beta_\varepsilon(\lambda)$ are the normalized scattering phase function and the volume extinction coefficient, respectively, are in a good agreement with empirical data (Fig. 2).

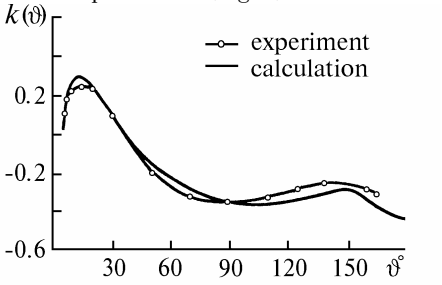


*FIG. 2. Angular dependence of the regression model coefficient k(ϑ) based on empirical data[38] and theoretical estimates obtained when modeling the variability of light scattering parameters of the atmospheric haze at relative humidity.*

The reason of closeness of the data is not only in uniformity of aerosol optical–microphysical properties of two geophysically different regions (Moscow region and Crimea), but in the uniform, or more exactly, dominating role of relative humidity in the mechanism of the near–ground haze variability.

Using theoretical estimates, we have managed to extrapolate the empirical single–parameter model (for the wavelength of 0.55 μm) to other wavelengths of optical sounding 0.6943 and 1.06 μm, and to other sounding angles (Fig. 3).

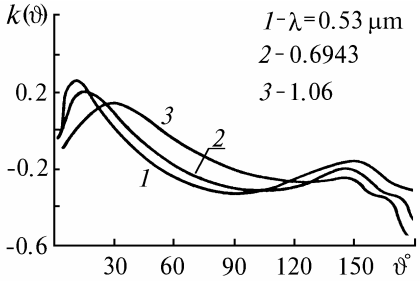


*FIG. 3. Model extrapolation of the angular dependence of the regression model coefficient k(ϑ) to other wavelengths of lidar sensing.*

## 5. VERTICAL VARIABILITY OF AEROSOL PARAMETERS

The following altitude gradation for typical states of the atmospheric aerosol[1] (Fig. 4) was used when constructing the model:

1) active turbulent mixing layer (up to 4 km);

2) the layer where the haze particles are involved into the cloud droplet structure (3 to 14 km);

3) the layer of long–term relaxation of anomalous accumulations of aerosol producing compounds of volcanic origin (11 to 30 km);

4) the layer of intrusion of interplanetary dust and remainders of meteor burning (25 to 90 km).

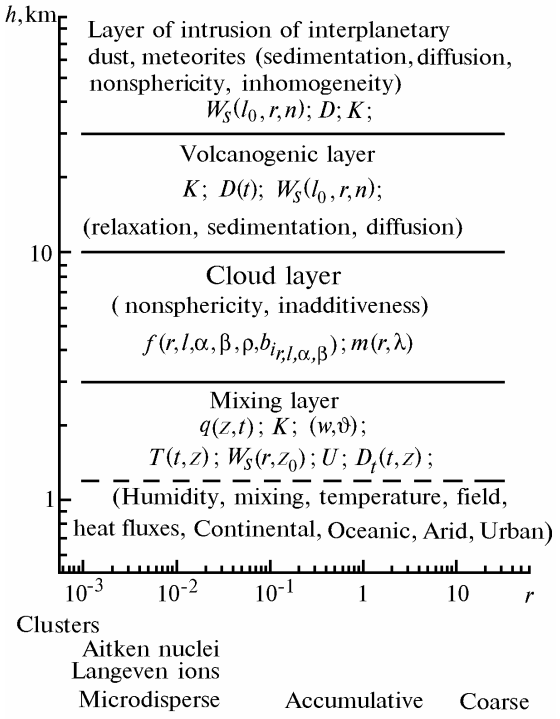


*FIG. 4. Principal types of formations determining the development of aerosol structure along the vertical direction.*

To investigate the mechanism of mixing the heterogeneous aerosols concrete numerical estimates are needed that take into account variations in power and specific nature of physicochemical composition of particles generated by different sources. Traditional approaches based on the exact description of the processes in multiphase media, run into great mathematical problems.

When transporting aerosol particles in the atmosphere, both the destruction of "old" local substructures and synthesis of the new ones occur simultaneously. Then, in spite of all variety of the effects of internal and external factors, the atmospheric haze particle size spectrum shows, on the average, the stability of shape. Synthesis of a new size spectrum, analogous to the initial one in its shape, is to a great extent caused by the specific nature of the mechanism of mixing various turbulent vortices of different scale containing their own structural elements of the disperse composition of a haze, or fractions.

The polydispersity of particles is of principle importance for the process of gravitational stratification of aerosol fraction structure along vertical direction. In contrast to a monodisperse system, the maxima of the number and volume density distribution of polydisperse aerosol particles are essentially well spaced on the size scale.

So an additional exchange process become more intense at the diffusion of particles between the cells of an aerosol layer. Larger and heavier particles leave behind the smaller and lighter ones when sedimentating, that in practice leads to the exchange by the structural elements of the size spectrum, subfractions, between the layers on large enough spatial scales of advection. Moreover, since the Cunningham correction for the Stokes sedimentation velocity varies by orders of magnitude at different heights, the size spectrum variation at different heights has certain peculiarities.

## 6. MIXING LAYER

Classification of aerosols in the near–ground atmospheric layer according to the their sources: oceanic, continental, arid, or industrial, without taking into account the intensity of turbulent mixing, is evidently insufficient. Diurnal variations of the temperature field caused by the relative humidity variations induce continuous spatial redistribution of the aerosol phase, in addition to the mobility of its local state.

To forecast the diurnal behavior of aerosol optical properties near the underlying surface, the Lille–Tennekes[34,35] micrometeorological model of the mixing layer was used (structural elements of the $C$ model are shown in Fig. 5). Diurnal variations of the entraping temperature inversion altitude and the average temperature of the layer were estimated based on this model:

$$h \frac{\partial \theta}{\partial t} = \langle w' \vartheta' \rangle_0 - \langle w' \vartheta' \rangle_h ; \quad (12)$$

$$\Delta \frac{\partial h}{\partial t} = - \langle w' \vartheta' \rangle_h ; \quad (13)$$

$$\frac{\partial \Delta}{\partial t} = - \frac{\partial \theta}{\partial t} + \gamma \frac{\partial h}{\partial t} ; \quad (14)$$

$$\langle w' \vartheta' \rangle_h = - c_h \langle w' \vartheta' \rangle_0 . \quad (15)$$

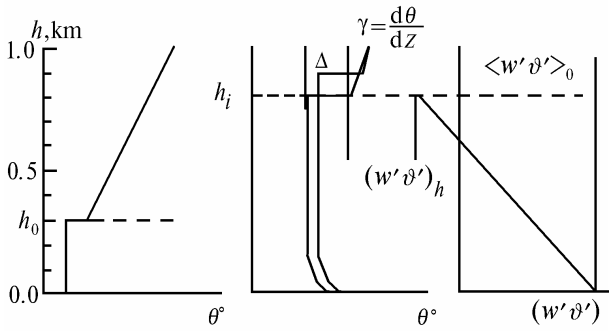


*FIG. 5. Diagram of the model of vertical distribution of potential temperature and turbulent heat flux inside and outside the mixing layer;* θ *is potential temperature in the layer.*

When solving the system of equations (6)–(9), the variations of the turbulent thermal flux were approximated by sinusoidal wave with a minimum at the sunrise and maximum at noon,

$$\langle w' \vartheta' \rangle_0 = B \sin[\omega ( t - t_s)] , \quad (16)$$

where $t_s$ is the sunrise time, $B$ is the amplitude of oscillations, and ω is the daily cycle frequency determined by the value

$$\omega = \pi / [2(12 - t_s)] .$$

Once, the specific boundary conditions are set, including ones related to the initial values $h_0$, $\theta_0$, and $\Delta_0$, the model makes it possible to forecast diurnal variations of the mixing layer height $h(t)$, which is an important parameter since the aerosol concentration inside the layer immediately adjacent to the surface strongly depends on that height as well as the potential temperature $\theta(t)$, whose value makes it possible to forecast the relative humidity variations as well as the inversion depth $\Delta(t)$.

When modeling the aerosol light scattering parameters in the mixing layer, we also took into account the effect of such factors as relative humidity variation, coagulation transformation inside anomalous accumulation of fine aerosols fraction, turbulent mixing, and gravitational sedimentation against the background of convective and advective transfer.

## 7. GRAVITATIONAL SEDIMENTATION AND CONVECTIVE TRANSFER OF AEROSOLS

The dynamics of transformation of the atmospheric haze microstructure inside an unstable mixing layer was modeled based on forecasting the joint variations of the profiles of the integral parameters of the particle size spectrum under the condition that mixing layer (its turbulent regime) develops during a day, and dynamic equilibrium is reached quite quickly between the upwelling and downwelling fluxes of the aerosol substance, if only for the average values of microphysical parameters of the spectrum.

The quasiequilibrium profile of spreading the aerosol particles inside the layer and corresponding variation rates of the average values of integral parameters of aerosol substance quantity over altitude is provided for the number, volume, and surface densities for each separate fraction. In other words, the condition of the balance of aerosol substance fluxes

$$D(z) \frac{\partial \hat{Q}_i}{\partial z} + W(z, r_i) \hat{Q}_i = 0 \quad (17)$$

holds simultaneously for $N_i$, $S_i$, and $V_i$. Here $\hat{Q}_i(z)$ is the average value of on integral parameter of the disperse structure $n(r, z)$; $D(z)$ is the profile of the turbulent diffusion coefficient ($m^2/s$); $W(z)$ is the mean, i.e., related to the characteristic size of a given fraction, resultant of the vertical component of their regular motion velocity (m/s).

The vertical component of the regular motion velocity of aerosol particles near the underlying surface in the general case is the sum of three principal components:

$$W_i(z) = W_s(r_i) + W_d(z) + W_t(z) , \quad (18)$$

where $W_s(r_i)$ is the Stokes sedimentation velocity of aerosols in the gravity field that is proportional to the square radius of a particle

$$W_s(r) = 2\rho g r^2 C_k / (9\eta) , \quad (19)$$

with η being the medium viscosity coefficient and ρ being the aerosol substance density, and it is a regular component of the value $W_i(z)$ and, practically, it does not vary with altitude inside the mixing layer.

In addition, in the framework of the proposed model, for the near–ground layer ($z < L$), we considered the so–called dry sedimentation velocity $W_d$ and took into account the process of aerosol capture from the turbulent flux by different obstacles (top of trees and plants, grass, other elements of the underlying surface roughness) that is an

important factor of sink of the disperse phase in the boundary layer.

For estimations we used linear dependence of the dry sedimentation velocity on altitude

$$W_d(z) = \begin{cases} \alpha_d (L - z) & \text{for } z < L, \\ 0 & \text{for } z \geq L. \end{cases} \quad (20)$$

Following the mixing layer model discussed above, for the approximation of the vertical profile of the aerosol regular convective motion velocity under the effect of the heat fluxes, upwelling from the surface and downwelling near the inversion base, we also used linear dependence of the following type:

$$W_t(z) = \begin{cases} \alpha_t \langle w' \vartheta' \rangle (z - h) & \text{for } z < h_0, \\ 0 & \text{for } z \geq h_0, \end{cases} \quad (21)$$

where $h_0 = 5h_i/6$ is the height of the zero flux of heat $\langle w'\vartheta' \rangle$ in the mixing layer (Fig. 5*c*) and $\alpha_t$ is the coefficient of proportionality between $W_t$ and $\langle w'\vartheta' \rangle$.

In addition, at the first stage of the numerical modeling of $n(r, z, t)$ we took into account only redistribution of aerosols over altitude under the condition of preservation of the quasiequilibrium level of their integral content in the layer as a whole

$$\int_0^{h_i} Q_i(z, t)\, dz = J_{Q_i} = \text{const}. \quad (22)$$

Just this circumstance often explains a significant decrease of the disperse phase optical density at the altitudes adjacent to the surface, not only because of heating of the layer and a decrease in the relative humidity of the carrier medium, but also because of a decrease in the aerosol concentration due to "thining" the layer turbidity as the mixing layer height increases.

Vertical profile of the turbulent diffusion coefficient was determined in the model with a correction for the state of layer stability, based on the known relationship[51]

$$D(z) = k u_* (1 - z / h_1) / \Phi(z / L), \quad (23)$$

where $k$ is the Karman constant; $u_*$ is the scale of friction velocity in the turbulent flux; $\Phi(z/L)$ is the semiempirical function of dimensionless parameter of stability

$$\xi = z / L = - k \langle w' \vartheta' \rangle g (z + z_0) / (\theta u_*^3), \quad (24))$$

where

$$L = (\theta u_*^3) / k \langle w' \theta' \rangle g \quad (25)$$

is the characteristic, Monin–Obukhov, length scale; $g$ is the acceleration due to gravity; and, $z_0$ is the underlying surface roughness parameter.

The value $u_*$ was estimated in the model calculations based on the logarithmic law for the wind speed profile $u(z)$ in the near–ground layer[52]:

$$u(z) = u_* / k \ln [(z + z_0) / z_0]. \quad (26)$$

According to Ref. 51, form of the stability function $\Phi(\xi)$ depends on the domain of the argument $\xi$ definition

$$\Phi(\xi) = \begin{cases} 0.74(1 - 9.0\xi) & \text{for } \xi \leq 0, \\ 0.74(1 - 6.4\xi) & \text{for } \xi > 0. \end{cases} \quad (27)$$

By varying the boundary conditions within the model framework, one can consider a great variety of atmospheric situations and study in detail the effect of the most important factors determining the altitude dynamics of variations of the optical and microphysical properties of the near ground haze.

The results obtained in Ref. 40 convincingly show that the information modeling system (IMS) block on the aerosol light scattering we have developed makes it possible to estimate the typological peculiarities of the possible deviations of the optical weather in the near ground layer depending on the underlying surface and aerosol sources, taking into account the peculiarities of the turbulent heat fluxes in different climatic zones.

## 8. ADVECTION OF AEROSOLS

In addition to the section devoted to analysis of mechanisms of forming the aerosol vertical structure in the mixing layer, we considered, in the model, the spatial deformations of their size spectrum at advection over the nonuniform underlying surface in a turbulent air flux. It is necessary for the analysis of such anomalous variations of the optical weather as dust storms, as well as for understanding methodical aspects of the problems of technical control of the air purity near pits.

The mathematical basis for corresponding IMS block is the turbulent diffusion equation[41]

$$U(z) \frac{\partial \hat{Q}_i(x, z)}{\partial x} = \frac{\partial}{\partial z} D(z) \frac{\partial \hat{Q}_i(x, z)}{\partial z} - W_i(r_i) \frac{\partial \hat{Q}_i(x, z)}{\partial z}, \quad (28)$$

where the vector–parameter $\hat{Q}_i = \{V_i, S_i, N_i\}$ is estimated at the point with coordinates $\{x, z\}$.

Theoretical estimates of the effect of mean flux velocity, mixing intensity, and degree of homogeneity of their distribution over altitude in the air flow show that the effect of gravitational sedimentation is essential for the coarse aerosol fraction, while the micro–disperse and accumulative fractions are transported practically without any variations in the spectrum, only their quantity decreases due to dispersal.

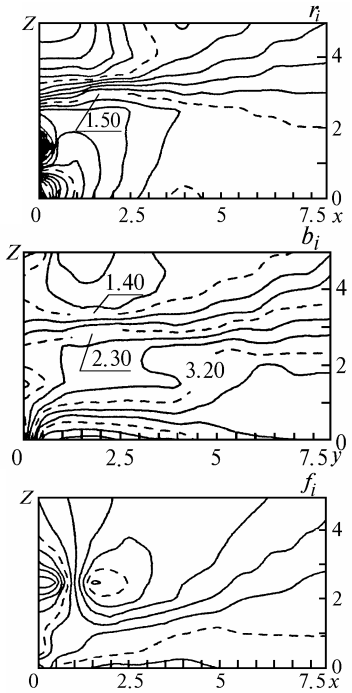


*FIG. 6. Isolines of spatial variations of the size spectrum parameters of the coarse fraction over uniform underlying surface for* $\beta = 0.1$ m, $U_0 = 0.18$ m/s, $D_0 = 0.005$ m$^2$/s, *and* $W_d = 0$ m/s. *Coordinates x and z are given in relative units, maximum values are* $x_{max} = 4$ km *and* $z_{max} = 50$ m, *respectively.*

It was also revealed that additional processes of exchange between the aerosol field cells by structural elements of the size spectrum, which other authors did not take into account earlier when working on this problem, appear at the advection due to aerosol polydispersity. The characteristic regions of spatial spectral accumulation of medium–sized aerosols (Fig. 6) can appear.

They create a background for forming the so–called noncondensation clouds at the convective instability in the near ground layer.

Finally, this block contains a possibility of analyzing of the effect of the specific density of the particle substance on the disperse composition of coarse aerosols and the mechanism of forming the spatial structure of aerosol fields.

## 9. LAYER OF THE INNERCLOUD WASH–OUT

The cells of a gas and particulate matter mixture penetrating into a cloud layer at 2 to 10 km in their motion are actively involved into the processes of phase transformation of the atmospheric water vapor, where the variations of atmospheric optical properties from the background ones are anomalously strong. So this atmospheric layer is presented in the model by a block providing the assessment of optical properties of different morphological structure, i.e., water droplets and crystals.

Active role of the atmospheric haze particles in the cloud formation process as the condensation centers causes breaks of the dielectric homogeneity of droplets and changes of their optical properties. This was investigated in artificial fogs within the framework of a laboratory experiment. Calculations showed that theoretical estimates made for droplets with foreign inclusions are in better agreement with the experimental data obtained at the early stage of fog formation. Data obtained 5–6 hours later show that these irregularities disappear and experimental values become close to the model estimates for homogeneous droplets, that can be explained by the processes of gradual washing out of big droplets and dissolving the hygroscopic nuclei.[42]

The difference in the fog and cloud optical properties at different stages of their formation demonstrated the need for data on the more complicated morphological structures to complete modern optical models of clouds.

Based on the model estimates, new explanation was proposed for the so–called "cloud grayness" effect revealed by Rosenberg based on the results of satellite measurements of the reflectivity of clouds.[43]

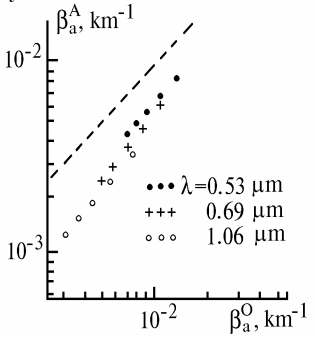


*FIG. 7. Diagram of a comparison between calculational estimates of the absorption coefficient of the atmospheric haze particles in the cloud droplets $\beta_a^O$ and independently in interdroplet space $\beta_a^A$ without the condensed moisture cover.*

The attempts to explaine the Wiscomba effect[44] by possibly inadequate account for the contribution of very big droplets failed. The correction was only 2–3% that is not enough for explaining this phenomenon even taking into account multiple scattering. We have shown that contribution of the haze particles into the absorption by clouds essentially increases (by 12–13%, Fig. 7) if they are considered as foreign inclusions into the droplets, and not as a filling of the interdroplet space. Estimates were obtained for the model of the droplets with three–layer dielectric structure, the scatter of values is due to the absorbing layer position inside the droplet.[45]

Specific character of formation of the phase composition of the upper layer clouds supposes that there are crystal aerosol structures inside them, and hence, their optical properties are anisotropic. In this connection, some optical light scattering effects appearing at the interaction with cirrus, cannot be interpreted in the framework of the models based on the Mie theory. Analysis of light scattering properties of the upper layer clouds taking into account the nonspherisity factor has been carried out in Ref. 46 based on the finite cylinder approximation obtained for far zone using the exact solution for the infinite cylinder in the near zone.

Analysis of the problem made out of the hypothesis of the spherical symmetry of the shape of aerosol particles leads to a significant extention of the list of initial parameters the problem needs for. The problem arises of summarizing the numerical estimates obtained in different coordinate systems to be presented in one system connected with the scattering plane.[46] Comparison of the estimates with analogous results obtained for elongated spheroids has confirmed the validity of the approach we have developed for the analysis of light scattering properties of crystal structures.[47]

Principal peculiarities of the formation of optical radar properties of cirrus clouds have been considered in Refs. 47 and 48. It has been shown that one cannot ignore even relatively small aerosol particles when forecasting the variations in the polarization characteristics of the scattered radiation. If small particles have symmetrical distribution of elongation axes over azimuth, and their CRI increases, they can noticeably change the polarization of scattered radiation. If there is preferred orientation, depolarization essentially decreases, but at the same time essential repolarization of the initial state occurs (for example, linear to elliptic).[47]

## 10. MODELING OF THE POSTVOLCANIC RELAXATION OF THE STRATOSPHERIC LAYER

Intense wash–out of aerosol–producing substances inside the cloud layer sereves as a powerful filter. Their invasion into the stratospheric layer of 12 to 30 km height occurs only during volcanic eruption episodes (approximately 50% of time it is in the state of relaxation from the disturbed state into the equilibrium background one), and plays an essential role in formation of the planetary albedo. Estimates of radiation effect based on the model of individual event are useful as a standard used in analysis of climate–forming process on the base of recurrence and strength of anomalous emissions of matter into the stratosphere.

Not ignoring the significance, at the initial stage of the volcanic invasion, of the processes of the inner–atmospheric synthesis of aerosols through the chain of heteromolecular phase transformations of sulfuric and nitric substances at the catalytic presence of water vapor breaking through the cloud layer and tropopause, in the approach we propose here the accent is done on the analysis of a more long stage of coagulation–sedimentation displacement of aerosol anomalies different in time and height of the initial location, because the more essential variations in the optical

state of the layer on the global scale occur just at the relaxation stage. Specific features of the process are predetermined by a significant decrease of density and viscosity of air. The Cunningham correction to the Stokes sedimentation velocity of aerosol particles increases by orders of magnitude. The diffusion spreading and sedimentation of the eruptive cloud are the main factors in evolution of the optical properties of the stratospheric layer.

The system of equations for the integral parameters

$$\frac{\partial \hat{Q}_i(z, t)}{\partial t} = \frac{\partial}{\partial z} D(z) \frac{\partial \hat{Q}_i(z, t)}{\partial z} - \frac{\partial}{\partial z} [W_{Q_i}(z, t) \hat{Q}_i(z, t)], \quad (29)$$

where $\hat{Q}_i$ is calculated for the height $z$ at the time moment $t$ has been used in Ref. 49 as the initial mathematical base of the model describing the variations in the eruptive cloud structure. Here $W_{Q_i}$ is the average Stokes sedimentation velocity of the $i$th fraction estimated for each equation from the modal radius of the distribution density function of the corresponding integral parameter of the size spectrum.

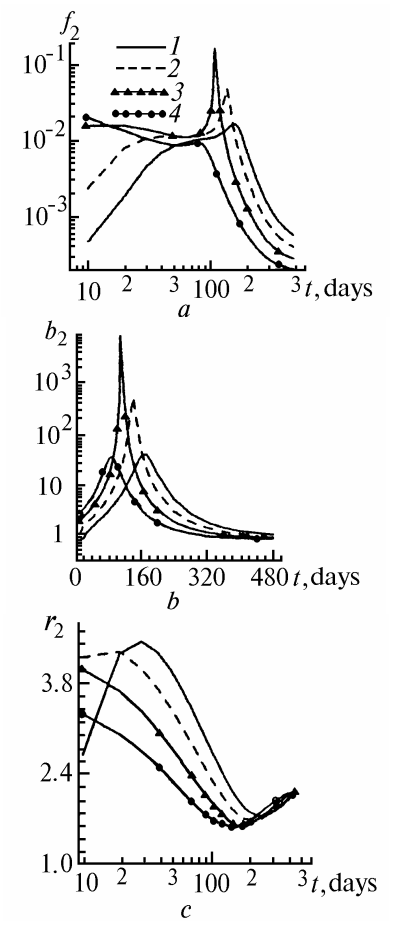


*FIG. 8. Variations of the size spectrum parameters of coarse fraction with time. Curves (1)–(4) correspond to z = 23, 24, 25, and 26 km.*

There is an interesting feature of the variations in size spectra of fractions revealed through the model estimates.[49] In particular, the unexpected tendency to narrowing the size spectrum (Fig. 8) was revealed in the evolution of the coarse aerosol fraction at weak turbulent mixing $D(z) \sim 0.1–0.3$ m$^2$/s (that is characteristic of summer). This layer is unstable and gradually spreads[50] at subsequent stages of the processes. Variations in the disperse composition of accumulative fraction are significantly due to the processes of mutual mixing of background and "anomalous" particles. Anomalous layer of the accumulative fraction can be retained during more than two years, but the coarse aerosol layer degrades during a period a little bit longer than a half of a year.

Analysis of the optical manifestations of this size spectrum narrowing effect showed[50] that the lidar ratio for this layer can vary by 110–150% of the mean value within the spectral interval of 40–50 nm. This effect and appearance of the fine structure in spectral variation of the backscattering coefficient $\beta_\pi(\lambda)$ (Fig. 9) can be used for lidar detection of the dielectric properties of the particulate matter, and, finally, of its chemical composition. The effect of turbulent mixing intensity, initial size spectrum, and different time of the process beginning at different heights has been also considered.

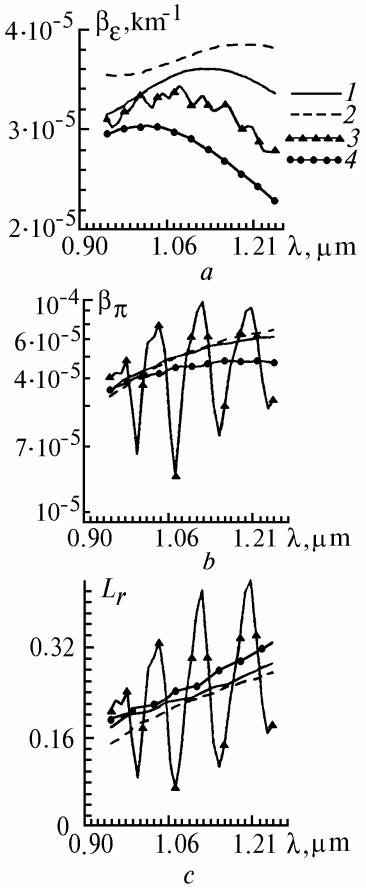


*FIG. 9. Spectral behavior of aerosol light scattering parameters: a) extinction coefficient; b) backscattering coefficient; c) lidar ratio at the height of* 25 km. *Curves (1)–(4) correspond to t =* 95, 105, 115, *and* 125 *days.*

## 11. GRAVITATIONAL SEDIMENTATION AND TURBULENT SPREADING OF AEROSOL ANOMALIES AT MESOSPHERIC HEIGHTS

In contrast to the stratospheric layer, the experimental data on mesospheric aerosol are very limited. In addition to the essential nonuniformity of the estimates of the interplanetary dust inflow intensity,[6] the problem of sinks and mechanisms of aerosol substance transformation in the layer is

still poorely investigated. The sedimentation instability of aerosol state in the layer is dramatized by an intense turbulent mixing $D \sim 3$–$50$ m$^2$/s. At the invasion of cosmic dust and micrometeors into the Earth's atmosphere, the evaporation of substance and fragmentation of structure occur due to the sharp retardation and conversion of kinetic energy into heat, that leads to accumulation of small particles with the size ~100 Å near the heights of retardation (80–100 km). Variations in the optical and microphysical properties of aerosol particles of cosmic origin have been investigated based on the model of diffusion–sedimentation spreading of randomly appearing anomalous accumulations (Fig. 10). Many scientists relate the noctilucent clouds appearing episodically near the mesopause to the process of covering of small hematite nuclei by dirty ice. So when modeling the optical properties the possible inhomogeneous dielectric structure of particles and their nonsphericity were taken into account in addition to the spatial variations of the size spectrum. In particular, depolarization properties of mesospheric aerosols were estimated in the approximation of elongated needle–shaped particles with chemical composition close to hematite fragments and dirty ice.

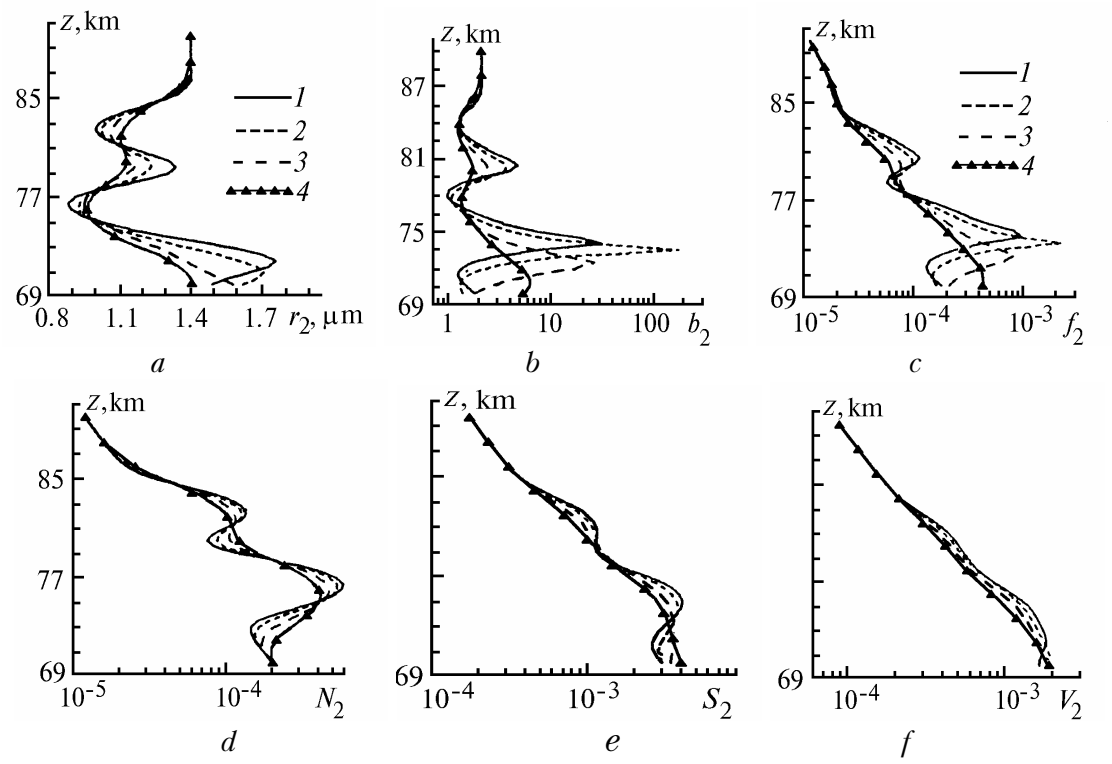


*FIG. 10. Deformation of vertical profiles of microstructural parameters of accumulative fraction of mesospheric aerosol at $D = 3$ m$^2$/s: model radius $r_a$ (a), parameter of dispersion $b_a$(b), $f_a$ parameter (c), particle number density $N_g$(d), surface parameter $S_g$(e), and volume parameter $V_g$(f). Curves 1–4 correspond to t = 2.9, 3.5, 4.7, and 6.6 days.*

## 12. CONCLUSION

It is necessary to note that the results presented in this paper show that the rate and character of deformations that the atmospheric aerosol structure experiences strongly depend on the initial parameters of the processes. Because of a great number of model parameters, it is necessary to investigate comprehensively various factors of the variability of aerosol state under different atmospheric conditions based on actual information and subsequent analysis of the effect of each of them.

Finally, it is necessary to note that the proposed approach to constructing the IMS structure well fits the capabilities of personal computers of a moderate speed and makes it possible to obtain data taking into account the fraction nonuniformity and aerosol nonspherisity both over height and time during 3–5 min computation. Also it gives the flexibility in planing field experiments and interpreting the experimental results.

## ACKNOWLEDGMENT

This work was supported by the Russian Foundation for Fundamental Research (Grant No. 94–05–16463–a).

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