Optics, microphysics, and kinetics of tropospheric aerosol

G.I. Gorchakov

A.M. Obukhov Institute of Atmospheric Physics, Russian Academy of Sciences, Moscow

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Systematic investigation of the scattering phase matrix of the atmospheric air allowed the investigation of time variability of the microphysical properties of atmospheric aerosol to be carried out. This in turn enabled the development of optical and microphysical models, as well as to find out the role of the basic transformation processes of the aerosol microstructure and particle composition, and to establish the laws of the atmospheric haze particles growth. The study of regimes of aerosol boundary layer formation has created grounds for better understanding evolution of the tropospheric aerosol. Analysis of multicomponent aerosol kinetics made it possible to assess the radiative effect of the transforming anthropogenic aerosol.

Introduction

The second half of the XXth century became a period of the atmospheric optics prosperity. Academician Vladimir Evseevich Zuev and Tomsk school of atmospheric optics headed by him made an important contribution to the development of atmospheric optics. Mainly owing to V.E. Zuev the Russian school of atmospheric optics as a whole is now on the worth place in the world science, that provides us the possibility of actively participating in the rapidly progressing investigations in the field of atmospheric physics, including physics of atmospheric aerosol.

In contrast to the approach to atmospheric aerosol investigation widely used abroad which is reduced to analysis of particular cases (case study), the tendency to the study geophysical peculiarities is characteristic of the investigations in Russia. In this paper we consider atmospheric optics as an inherent component and starting point of the development of physics of atmospheric aerosol: from aerosol optical features through microphysical features to features of kinetics and dynamics of the tropospheric aerosol.

Optics of aerosols

The problem of investigation of the atmospheric aerosol optical properties was stated in the most general form by Prof. G.V. Rosenberg.[1,2] Atmospheric aerosol optics got a significant development in the investigations by Academician V.E. Zuev and his school.[3,4] Let us note that the international conference "Atmospheric aerosol physics" devoted to 85th anniversary of G.V. Rosenberg's birthday was held in April 1999. It demonstrated the urgency of his ideas and approaches for the modern physics of atmospheric aerosol.

As known, extinction of light in the atmosphere is governed by the processes of scattering and absorption, so the total extinction coefficient ε is equal to the sum of the total scattering, σ, and absorption, α, coefficients:

$$\varepsilon(\lambda) = \alpha(\lambda) + \sigma(\lambda) , \quad (1)$$

where λ is the wavelength.

The scattering of light in the spectral regions out of the absorption bands of atmospheric gases is determined by the disperse (σ_a) and gas (σ_m) components of the air:

$$\sigma = \sigma_a + \sigma_m . \quad (2)$$

The angular distribution $V(\varphi)$ of electromagnetic radiation of the visible wavelength range scattered by a local volume of atmospheric air is characterized by the scattering phase matrix (φ is the scattering angle)

$$D_{ik}(\lambda, \varphi) = D_{ik}^{a} + D_{ik}^{m} , \quad (3)$$

that is the matrix of linear transformation of the Stokes parameters in the act of scattering (A is the instrumentation constant):

$$S_i'(\lambda, \varphi) = AV(\varphi)\sum D_{ik}(\lambda, \varphi)S_k(\lambda) \quad (i, k = 1, 2, 3, 4), \quad (4)$$

where S_k and S_i' are the Stokes parameters of the incident and scattered light beams, respectively. Obviously, the number of arguments of D_{ik} increases at the presence of anisotropic particles having a preferred orientation.

When analyzing the experimental data, it is convenient to deal with the components of the reduced scattering phase matrix

$$f_{ik} = D_{ik}/D_{11} . \quad (5)$$

In particular, the component f_{31} is numerically equal to the degree of linear polarization of the scattered light p if the scattering volume has been irradiated by an unpolarized light, and the component f_{43} is numerically equal to the degree of ellipticity of polarization of the scattered light q if the incident beam has been linearly polarized in the plane turned to 45° relative to the scattering plane.

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The main problem that arose at the first stage of experimental investigations of atmospheric aerosol optical properties was the accumulation of measurement data on some optical characteristics, including the scattering phase function or the coefficient of directional light scattering D_{11} (Refs. 5 and 6), degree of linear polarization p (Refs. 7 and 8) degree of ellipticity of polarization q (Ref. 9) and the spectral dependence of the extinction coefficient ε.[10–13] Soon it became clear that the small-angle scattering phase functions are worthy of special consideration.[14]

Great variety of observed angular and spectral dependences of the atmospheric aerosol optical characteristics[15] caused the necessity of solving the problem of their classification, that led to creation of the conception of the types of optical weather.[16–19] In our opinion, the term optical-meteorological weather is more exact here, because the case in point is such objects as drizzle haze, fog, ice haze, etc. Selection of the drizzle haze as a separate type of optical weather had the principal significance as well as the conception of thermodynamic equilibrium or equilibrium-solution haze.[16,20]

The need for the aerosol optical models led to the necessity of studying statistical features of the variability of optical properties of the atmospheric haze. Owing to reliable measurements of the coefficient of directional light scattering in the small-angle range of forward and backward directions of scattering[21,22] simultaneously with traditional measurements in the scattering angle range from 10 to 170°,[23] the near-ground aerosol scattering phase functions at the angles from 0.5 to 178° were obtained for the first time. This made it possible to construct a single-parameter statistical model of the angular dependence of D_{11} at the wavelength of $\lambda = 550$ nm, the input parameter of which is the scattering coefficient σ (or the meteorological visual range $L = 3.9/\sigma$). One can find the angular dependences of four components of the scattering phase matrix for different situations, including the dust haze, in Refs. 15, 24, and 25.

Long-term systematic investigations of the angular dependences of scattering phase matrix elements that have been being carried out in Moscow region made it possible to study the statistics of their variability and to construct single-parameter statistical models of the elements of reduced scattering phase matrix.[26,27]

As in the case with the coefficient of directional light scattering, the input parameter of the model is the scattering coefficient σ. Similar statistical features of the coefficient of directional light scattering and the degree of linear polarization were obtained later by researchers at the Institute of Atmospheric Optics.[28] Figure 1 shows the mean conditional angular dependences of three elements of the reduced scattering phase matrix and the coefficient of directed light scattering at the meteorological visual range of 20 km given by our model (solid lines) and analogous dependences constructed based on measurement data acquired in Abastumani[29,30] for the meteorological visual range of 50 km (dotted line). For convenience of a comparison the dependences $D_{11}(\varphi)$ for the cases shown in Fig. 1 are set equal at one point and for this reason these are presented in different relative units. From this it follows that, on the one hand, some similarity of the optical characteristics is observed for different seasons and different regions, and on the other hand, there is certain ambiguity in the relations between quantitative and qualitative aerosol optical characteristics.

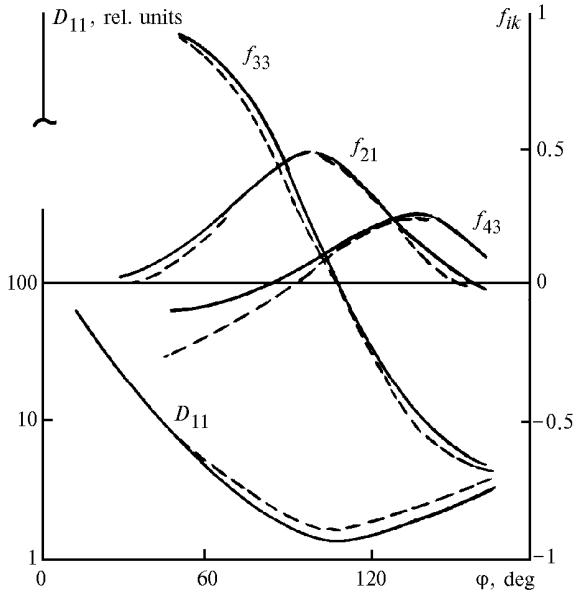

Fig. 1.

Investigations into the spectral behavior of aerosol extinction along horizontal paths have progressed in parallel, that made it possible to create statistical models of the spectral atmospheric transmission for different seasons and geographical regions.[11–13,28,31]

In connection with the problems of predicting optical weather, investigations of temporal variability of the aerosol optical characteristics, particularly their diurnal behavior were of great interest. Investigation of the diurnal behavior of the optical characteristics is necessary now for clarifying the features of the aerosol kinetics. One can consider round-the-clock measurements of the angular dependences of the scattering phase matrix elements carried out in summer 1979 in Abastumani[29,30] within the frameworks of the Soviet-American complex experiment on studying background aerosol as most informative optical experiment.

Important features of the temporal variability can be revealed by means of relatively simple nephelometric measurements. We carried out round-the-clock nephelometric measurements in 1984–1999 in Moscow region as well as in other sites. In particular, we revealed the long-period synoptic variability of the scattering coefficient,[32] studied its diurnal behavior, and inter-year variability.[33,34] The valuable advantage

of the measurements carried out using the instruments with a closed volume is the possibility of obtaining data on the scattering coefficient of dry matter of aerosol particles σ_d simultaneously with the scattering coefficient of natural aerosol σ. Let us note that the values σ are often recalculated to the submicron aerosol mass concentration.[34] The diurnal behavior of σ and σ_d together with the diurnal behavior of the meteorological parameters and the aerosol microphysical characteristics obtained from analysis of data on the scattering phase matrix measured in Abastumani in summer 1979 is shown in Fig. 2.

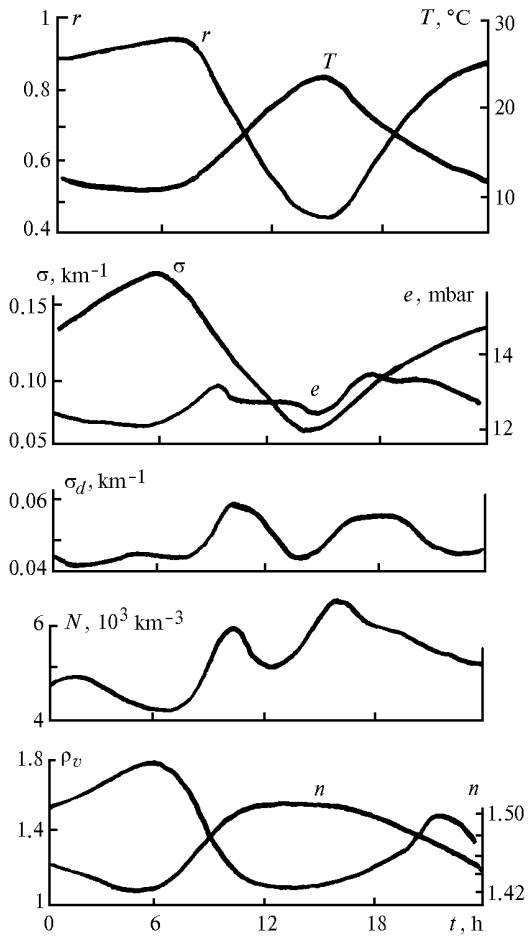

Fig. 2. Mean diurnal behavior of temperature T, relative humidity of air r, absolute humidity e, scattering coefficient of natural aerosol σ, of its dry matter σ_d, number density of particles N, dimensionless median radius of the volume size-distribution ρ_v, and the refractive index of natural aerosol matter n.

We calculated the mean diurnal behavior (Fig. 3) of the number density of aerosol particles with the radius of 0.0065 (*1*), 0.28 (*2*), and 1.41 μm (*3*) from the data of direct measurements of the near-ground aerosol microstructure in the same experiment by A. Wagoner, R. Wyes, and N. Alkvist (University of Washington, Seattle).

Optical methods are also a convenient tool for studying spatial distribution of aerosols in the near-ground layer of the atmosphere. An example is investigation of the spatial distribution of the mass concentration of submicron and soot aerosol along the Transsiberian railway by means of the laboratory installed in a carriage.[35]

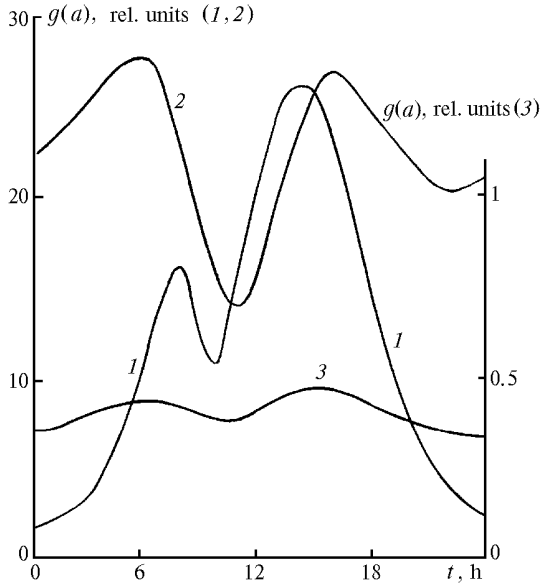

Fig. 3. Mean diurnal behavior of the number density of particles with the radius $a = 0.0065$ (*1*), 0.28 (*2*) and 1.41 μm (*3*) obtained from the data of measurements in Abastumani.

Microphysical characteristics of the near-ground aerosol

The observed variations of the aerosol optical characteristics are to a great degree determined by the variability of its microstructure, i.e., of the particle size-distribution function $g(a)$ and the refractive index $m = n - i\kappa$ of the particulate matter, where n and κ are its real and imaginary parts. In the case of moistened aerosol, one can calculate the angular and spectral dependences of the scattering phase matrix elements, for some preset particle size-distribution, by Mie theory[36] (C is the normalization constant):

$$D_{ik}(\lambda, \varphi) = C(\lambda) \int d_{ik}[\rho, \varphi; n(\lambda), \kappa(\lambda)]\, g(a)\, da, \quad (6)$$

where d_{ik} are the elements of the scattering phase matrix for a monodisperse aerosol with the particle radius a and $\rho = 2\pi a/\lambda$. It follows from the aforementioned formula that the inverse problem of light scattering is reduced to the system of integral equations more complicated that the Fredholm equation of the first kind, because the kernels of the integral equations depend in this case on the unknown parameters n and κ.

A few method have been developed for solving this inverse problem.[37–39] G.V. Rosenberg[37] proposed

the method for solving the inverse problem based on the parametric description of the aerosol microstructure. The method is based on expansion of the calculated optical characteristics Q_i into a Taylor series within quite a small vicinity of the point with known values of the parameters $s_k^{(0)}$:

$$Q_i(s_k) \cong Q_i(s_k^{(0)}) + \frac{\partial Q_i}{\partial s_k} \Delta s_k \ , \qquad (7)$$

where Δs_k are the unknown increments of the sought microphysical parameters s_k. The derivatives $\partial Q_i / \partial s_k$ were obtained[40] for the lognormal approximating size distributions. One should note that, to solve the inverse problem by this method, it is necessary, first, to analyze the information content of the experimental data.

We estimated the submicron aerosol microphysical parameters from data of systematic measurements of the angular dependences of elements of the scattering phase matrix in the approximation of a single-mode lognormal distribution (N is the total number density, a^* is the median radius of the distribution)

$$g(a) = \frac{N}{\sqrt{2\pi\gamma a}} \exp\left\{-\frac{\ln^2 (a/a^*)}{2\gamma^2}\right\} . \qquad (8)$$

Analysis showed that in the majority of cases it is possible to restrict oneself to calculations using the fixed γ value. We selected $\gamma = 0.7$. Besides, it occurred that it is expedient to restrict oneself to the approximation $\kappa = 0$, because the method does not allow one to reliably reveal the role of the aerosol absorption.

Continuous round-the-clock measurements of the scattering phase matrix elements and their dependences on relative humidity in Abastumani have made it possible to obtain the mean diurnal behavior of the microphysical characteristics of aerosol and its dry matter. Figure 2 shows the mean diurnal behavior of the refractive index n, dimensionless median radius ρ_v of the particle volume size-distribution, and the number density of submicron aerosol fraction N. Moistening and growth of the submicron particles are clearly seen to occur at nighttime, as well as more complex processes of the aerosol transformation during daytime.

As it follows from Fig. 3, the increase in the particle number density is observed in the morning for the microdisperse size range, then there occurs maximum in the afternoon that correlates with the maximum in the behavior of the scattering coefficient σ (see Fig. 2) and a less deep minimum in the evening. Diurnal modulation of the coarse particle number density time behavior (curve *3*) is pronounced in Abastumani more weakly. In general, direct measurements of aerosol microstructure and optical characteristics give a sufficiently complete pattern of the diurnal transformation of aerosol.

Regardless of a serious "roughening" of the problem, the use of a single-mode approximation makes it possible to reveal other essential features of the variability of the aerosol microphysical parameters. The example is the single-parameter microphysical model of the near-ground aerosol[41,42] that agrees well with the single-parameter statistical model of the angular dependence of the scattering phase matrix elements. As the statistical model, the microphysical one uses the scattering coefficient σ or the meteorological visual range L as the input parameter. The microphysical model allows one to reconstruct not only the filling factor V_w, number density N, median radius of the distribution a_w^*, and the refractive index n_w of the natural aerosol particle substance, but also the parameters of the aerosol dry matter V_d and a_d^*. Along with a_w^* and a_d^* let us also use the parameters a_w^v and a_d^v for the particle volume size-distribution $v(a) = 4\pi a^3 g(a)/3$ (Table 1). To calculate the principal microphysical parameters, one can use simple approximate formulas for V_w, a_w^v, and n_w:

$$V_w \cdot 10^{12} = 220\ \sigma - 22\ \sigma^2 \qquad (9)$$

$$a_w^v = 0.41 - 0.2 \log L \ , \qquad (10)$$

$$n_w = 1.36 + [\log (0.5\ L)]^{2.1}, \qquad (11)$$

as well as for the ratio

$$\frac{V_d}{V_w} = 0.71 - 0.46y - 0.05y^2, \qquad (12)$$

where $y = \log (50\ L^{-1})$.

The conditional mean values of the relative humidity r are also presented in Table 1.

Table 1

Values of the parameters	Parameters of the model				
σ, km^{-1}	0.078	0.195	0.39	0.78	1.95
L, km	50	20	10	5	2
r, %	45	67	78	88	95
a^*, μm	0.13	0.21	0.27	0.34	0.40
a_d^*, μm	0.117	0.17	0.20	0.225	0.225
γ	0.7	0.7	0.7	0.7	0.45
N, cm^{-3}	16500	10200	9100	8800	3200
n	1.45	1.42	1.4	1.38	1.36
$V \cdot 10^{12}$	18	44	40	165	350
$V_d \cdot 10^{12}$	13	23	33	48	62

It is also essential that the number density N of particles weakly changes in the range of variation of L approximately from 5 to 20 km ($N \cong 10^4$ cm^{-3}). In this case the increase of the atmospheric turbidity is caused by the processes of accumulation of the mass of the dry matter of aerosol particles and moistening of the particles. The noticeable deviation of N at $L = 50$ km is obviously explained by the fact that the single-parameter model is not sufficient for description of the aerosol microstructure at a long meteorological visual range.

The variability of the scattering phase matrices in Abastumani can be characterized by analogous

microphysical model (see Fig. 1). However, the mean number density of particles N in Abastumani in summer is 2.5 times lower ($\approx 4 \cdot 10^3$ cm^{-3}). Besides, we obtained the single-parameter microphysical models that agree with the statistical models of the angular dependence of the scattering phase matrix elements constructed based on measurement data in summer 1977 in Moscow and in winter 1976–77 in Moscow region.[43] Figure 4 shows the corresponding dependences of the parameter ρ^* on the meteorological visual range L. It is seen that in the general case the dependences $\rho^*(L)$ are nonlinear. At a long L there is the tendency toward a transition from the regime $N = \text{const}$ to $\rho^* = \text{const}$ when the turbidity of the atmosphere is determined mainly by variations of the number density of particles.

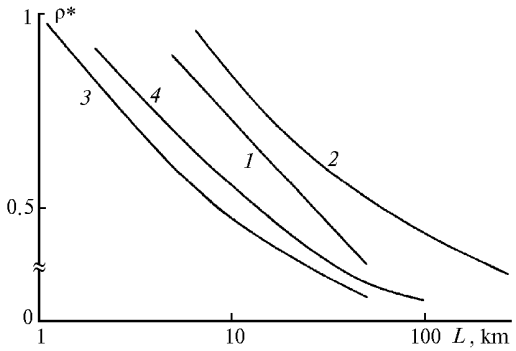

Fig. 4. Dimensionless radius of the particle size-distribution as a function of meteorological visual range in autumn in Moscow region (*1*), in summer in Abastumani (*2*), in winter in Moscow region (*3*), and in summer in Moscow (*4*).

Statistical analysis of the spectral dependences of the scattering coefficient[44] obtained in the ODAEX–87 experiment shows that the effective number density of the coastal haze particles is $N \cong 2.5 \cdot 10^4$ cm^{-3} that can be caused by the high rate of formation of aerosol particles (nucleation) in the coastal zone.

The dependences $\rho^*(L)$ together with the characteristic number density of particles N give an idea on the principal peculiarities in the optical and microphysical parameters of submicron aerosol.

The models we have developed can be used not only for the microphysical extrapolation of the optical characteristics but also in analyzing the features of the fine aerosol transformations.

Application of the method of inverse problem to analysis of angular dependences of the scattering phase function and the degree of linear polarization at light scattering by the coastal aerosol[45] lead to the results which satisfactorily agree with our results.

The single-parameter microphysical model does not pretend to the complete and exact reconstruction of the aerosol microstructure. For example, in some cases of a fog haze and at long meteorological visual range the effect of coarse aerosol fraction is well pronounced. The coarse or big-droplet aerosol in the near-ground layer of the atmosphere can be revealed by the method of spectral transparency.[46] The spectropolarimetry method[47] is also used now for this purpose.

Processes of aerosol transformation

Atmospheric aerosol is continuously transformed under the effect of various processes of different scales. The processes of intraatmospheric aerosol transformation including the gas-to-particle conversion processes and the processes of condensation of water vapor, coagulation, and nucleation are still of the primary interest. The complex process of intracloud aerosol transformation can also be referred to this group.

The problem of investigation of condensation of water vapor on aerosol particles (by G.V. Rosenberg,[48] assimilation and dissimilation) has a long history. The following empirical dependence[20,49] was obtained by a number of authors based on data of observations of the change of the coefficient ε (or meteorological visual range) and the relative air humidity r:

$$\varepsilon\ (r) = \varepsilon_0\ (1 - r)^{-\chi_\varepsilon}. \qquad (13)$$

However, not always simultaneous measurements of time dependences $\varepsilon(t)$ and $r(t)$ (t is time) yielded this dependence. Besides, the observed values of the parameter χ_ε not always occurred physically acceptable.

Owing to the creation of a pump through polarimeter[50] capable of introducing controlled effects on the air flow passing through the device, the techniques were developed for measuring the scattering phase matrix elements D_{ik} as functions of the relative air humidity r and measuring the components of the scattering phase matrix D_{ik}^d of the dry matter of aerosol particles, or of dried aerosol. In particular, the empirical dependence of the coefficient of directional light scattering was obtained from the measurement data acquired in Abastumani[29]:

$$D_{11}(\varphi; r) = D_{11}^d(\varphi)\ (1 - r)^{-\chi(\varphi)}, \qquad (14)$$

where $\chi(\varphi)$ is the empirical function.

Solving the inverse problem of light scattering for $D_{11}(\varphi; r)$, assuming single-mode approximation, shows that the specific samples $a^*(r)$ in a wide range of the relative air humidity are satisfactorily described by the formula of Kasten-Hanel type:

$$a^*(r) = a_0\ (1 - r)^{-\chi_a}. \qquad (15)$$

It is easy to obtain the dependence of the real part of the refractive index n on r in the approximation of additivity of the refractive index of a well mixed aerosol particles[20] (see Fig. 2).

Analysis of the diurnal behavior of the optical and microphysical characteristics of natural aerosol and its dry matter shows that the variability of the microphysical characteristics is governed not only by the processes of condensation of water vapor, but also by evolution of the dry matter of aerosol particles. The

estimate of the efficiency of the gas-to-particle conversion processes[43] (approximately 5 nm/hour) was obtained from the data on the rate of change in the effective size of dry particles. The role of the gas-to-particle conversion in the formation of submicron aerosol can be most distinctly revealed by means of a single-parameter microphysical model (see Table 1). Simultaneously, one can find an important peculiarity of the increase in the growth of the mass of dry matter of particles as the relative air humidity increases.

In some cases one can use the data of direct measurements of the aerosol microstructure for estimating the growth rate of particles due to the gas-to-particle conversions in the size range approximately from 3 to 30 nm, which are now called ultrafine particles. According to data from Ref. 45, the growth rate of particles of this size range can reach 10 nm/hour. Analysis of data of the round-the-clock measurements[52] of aerosol microstructure shows that the daytime rate of the particle growth in Western Siberia is approximately 2 nm/hour.

The main difficulty in estimating the growth rate of particles and the rate of their removal[25,30,43] is related to the absence of data on the aerosol lifetime in the near-ground atmospheric layer or on the rate of exchange of air between the near-ground and boundary layers of the atmosphere.

The process of aerosol emission from the underlying surface is among the poorly studied problems. It is expedient to consider this process as consisting of several stages, starting from the aerosol emission to a thin near-surface layer, from which the aerosol is then transported to the near-ground layer of the atmosphere, and from there to the boundary layer of the atmosphere. Thus, the process of aerosol emission from the land surface is closely related to the regime of turbulence in the near-ground layer of the atmosphere and to regime of the boundary layer of the atmosphere.

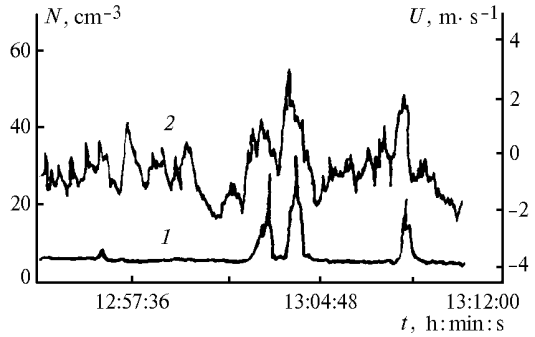

Fig. 5. The results of simultaneous measurements of the number density of aerosol particles N (1) and the pulsations of wind speed U (2) on September 29, 1998 in a near-Aral region.

New data were obtained when studying the aerosol emission from the desert areas in Kalmykia and near-Aral region. It was shown[53] that the aerosol emission often occurs under the effect of wind gusts[35] with the duration of the order of 100 seconds. Figure 5 shows the data of simultaneous measurements of the fluctuations of the number density of particles of approximately 0.3 μm in size (measurements by K.A. Shukurov) and the longitudinal component of the wind velocity (measurements by B.M. Koprov). The peaks of number density with a steep front are well seen in the figure, that is an evidence of the nonlinear nature of the effect. Further investigations have shown that some aerosol peaks over the desert area are related not to the wind gusts, but to the areas of the enhanced air temperature, or "thermics.B

Kinetics of the fine aerosol

The fine ("secondary") aerosol evolves and undergoes further transformations directly in the atmosphere. According to G.V. Rosenberg,[54,55] microdisperse and submicron fractions (nucleus and accumulative modes, according to K. Whitby[56]) should be related to the fine aerosol.

Nucleation, or formation of new aerosol particles of the size about 1 nm, is still being intensively studied. The mechanism of binary nucleation of water and sulfur acid vapor has been established quite reliably.[51] At the same time, it is known that other gas components of the atmosphere also play important role in this process. Now there are some reasons to suppose that the nucleation process intensifies[57] at the presence of a noticeable amount of ammonia in the atmosphere. However, a number of papers mention the important role of the fourth agent (or other agents) which can be organic compounds.[57] The hypotheses have been proposed that some organic compounds (for example, some bicarbon acids) are able to initiate the nucleation process at no any energy threshold. The rate of nucleation under some conditions[57] can reach hundred thousands $cm^{-3} \cdot s^{-1}$.

One should emphasize that inhomogeneity and nonstationarity of the nucleation process is of great importance for the evolution of fine aerosol. It is shown that the conditions favorable for nucleation are often realized in the free troposphere over ocean.[58] For some reasons, this process can not occur all time at one and the same rate. Obviously, the peaks of nucleation are more natural for the atmosphere.[59]

The particles appearing in the atmosphere are transported there and involved in the processes of mixing. Then they are removed from the atmosphere, as well as take part in the processes of intraatmospheric transformations. Evolution of the fine aerosol microstructure is described by the kinetic equation for the particle size-distribution function $g(a)$[60]:

$$\frac{\mathrm{d}g(a)}{\mathrm{d}t} = \left[\frac{\partial g(a)}{\partial t}\right]_{\mathrm{mix}} + \left[\frac{\partial g(a)}{\partial t}\right]_{\mathrm{cg}} + \left[\frac{\partial g(a)}{\partial t}\right]_{\mathrm{gpc}} + \left[\frac{\partial g(a)}{\partial t}\right]_{\mathrm{rem}} + B_{\mathrm{g}}(a)\,, \qquad (16)$$

which takes into account the processes of gas-to-particle conversion (gpc), coagulation (cg), mixing (mix), and removing of particles from the atmosphere (rem) in the explicit form. The term $B_g(a)$ characterizes the income of particles to the atmosphere both due to the nucleation and emission of particles from the underlying surface.

The practical impossibility of describing at present the totality of equations of chemical kinetics of the gas components of the atmosphere in detail, which is related to the formation of all principal aerosol producing compounds, is of principal importance. So now in the majority of cases one can restrict oneself to the semi-empirical theory of the kinetics of fine aerosol.

The variety of possible processes of gas-to-particle conversion[60] puts a question about the mechanism of particle growth, or the law of growth under specific conditions and in certain size ranges. It is also essential that the kinetics of water-soluble aerosol in a wet atmosphere can significantly differ from the kinetics of the waterless aerosol.[61]

According to Friedlander,[60] the laws of the growth of particles in the simplest case have the form:

$$\frac{dv}{dt} = A_\alpha\, a^\alpha, \qquad (17)$$

where a and v are the radius and volume of a particle, respectively. The rate of the growth of a particle under similar conditions in the wet atmosphere can be described by the relationships

$$\frac{dv_w}{dt} = A_\alpha\, \xi_v(r)\, a_w^\alpha, \qquad (18)$$

where a_w and v_w are the radius and volume of a liquid-droplet aerosol particle, respectively, and $\xi_v(r) = (1-r)^{-\chi_v} = (1-r)^{-3\chi_a}$ is the empirical function.

Evolution of the shape of the particle size-distribution of the submicron fraction is determined by the law of growth at a small number density of particles when one can ignore the process of coagulation. If the growth of particles has been determined by the process of condensation of the vapor of aerosol producing compounds ($\alpha = 1$ or 2 depending on the particle size) on the aerosol particles, the relative width of the distribution decreases. In the case when the growth of the liquid-droplet aerosol particles has been determined by the processes inside the particle (mechanism of the volume growth), the shape of the particle size-distribution remains unchanged. It is observed in the case of a single-parameter microphysical model that is obtained based on the data on the scattering phase matrix.[41,61]

Comparison of the few-parameter models of the kinetics of fine aerosol with the single-parameter microphysical model and the peculiarities of the diurnal variability of the aerosol microphysical parameters allows us to estimate the rate of some processes of aerosol transformation without the aforementioned estimates of the rates of the gas-to-particle conversion. In particular, one can explain the gradual decrease of the number density of particles N from 10200 to 8800 cm^{-3} at the decrease of the meteorological visual range from 20 to 5 km (single-parameter model) by the contribution of the coagulation process into the transformation of submicron aerosol. This hypothesis allowed us to obtain the rough estimate of the lifetime of submicron aerosol in the near-ground layer of the atmosphere[61] $\tau \approx 1$ hour.

The analysis of diurnal variability of the microphysical parameters of fine aerosol shows[43] that the rate of removing the submicron particles from the near-ground layer of the atmosphere during night is approximately 0.04 $hour^{-1}$ (underestimate). Coagulation essentially shortens the lifetime of the finest particles (clusters) and the particles of the microdisperse size range. The estimate is obtained of the rate of change of the number density of clusters due to their sticking to larger fine aerosol particles[43]: $\frac{1}{N_{cl}}\frac{dN_{cl}}{dt} \cong 1.1\ h^{-1}$. The mean rate of the cluster formation is approximately 5 $cm^{-3}\cdot s^{-1}$. One can also estimate the relative rate of condensation of vapor of the aerosol producing compounds[43]:

$$B_g \cong 0.1\ V_{sb}\ (h^{-1}),$$

where V_{sb} is the filling factor of submicron aerosol.

To exactly describe the aerosol kinetics, it is necessary to know the dependence of some aerosol characteristics on the particle size, in particular, the dependence of the parameter of condensation activity χ_a. The direct measurements we have carried out earlier revealed wide variety of the dependences $\chi_a(a)$ in the atmosphere.[62] The averaged over an ensemble mean dependences $\chi_a(a)$ were obtained[47] from data of spectropolarimetric measurements. It occurred so that the mean dependences $\chi_a(a)$ have a well pronounced maximum near $a = 0.3-0.4$ μm in a wide range of the meteorological visual range. The dependence $\chi_a(a)$ with the maximum in this range were revealed earlier from the measurement data on aerosol microstructure.[63] This result is an evidence of the complicated nature of the process of formation of aerosol microstructure in the submicron size range.

Let us assume that the growth of the dry matter of particles is determined by the processes of condensation of vapor of aerosol producing compounds. It is easy to show that the rate of growth of particles in a wide size range can be described by the relationship

$$\frac{da_w}{dt} = \frac{A^*\xi(r)}{a + a^*}, \qquad (19)$$

where a^* is the so-called free-molecular parameter.

If two types of the aerosol producing compounds were simultaneously present in the atmosphere, and the Kelvin effect[60] occurred in one of them, the rate of growth of particles in this case is equal to

$$\frac{\mathrm{d}a_w}{\mathrm{d}t} \cong \frac{A_1^*\xi(r)}{a_w + a_1^*} + \frac{A_2^*\xi(r)}{a_w + a_2^*}\,\frac{a - a_0}{a}\,, \qquad (20)$$

where a_1^* and a_2^* are the free-molecular parameters for the types of the aerosol producing compounds different in the free paths of molecules in air, and a_0 is the parameter determined by the Kelvin effect for the aerosol producing compound of the second type. The second term in Eq. (20) has maximum at some value of the particle radius exceeding a_0.

Organic compounds play an important role in the process of formation and growth of microdisperse aerosol.[51] As known, they are characterized by relatively small value of the parameter of condensation activity. The second aerosol producing compound (of the type of ammonium sulfate) begins to play a significant role in submicron size range. It provides for the inflow of substance with high condensation activity to the aerosol particle, and the submicron aerosol is formed with the parameter of condensation activity dependent on the particle size. The decrease of the parameter of condensation activity observed in the range $a \geq 0.5$ μm can be partially caused by the presence of particles of poorly soluble mineral aerosol particles in air.

The coagulation process significantly affects the transformation of the aerosol structure at small meteorological visual range.[64,65]

On the whole, one can assert that atmospheric haze is kinetically non-equilibrium, and one can consider it as kinetically equilibrium only in certain situations during only limited periods of time. But in both of these cases we have to refuse from the concept of a thermodynamically equilibrium haze, the concept that played important role in the development of the physics of atmospheric aerosol.

Let us note that interesting ideas appear at present in the kinetics of atmospheric aerosol, for example, the theory of condensation growth of particles with partially soluble salt core,[66] or the hypothesis of formation of liquid-droplet particles with a cover ("inverted micell").[67] The hypothesis of the possibility of realization of the "total absorption regime" in nature at condensation of organic compounds on the aerosol particles[68] deserves special attention too.

Aerosol in the boundary layer of the atmosphere

When interpreting temporal variability of the near-ground aerosol microphysical characteristics, serious difficulties appear because the kinetics of aerosol in the near-ground layer of the atmosphere is inseparably linked with the dynamics and kinetics of aerosol in the troposphere, and, first of all, in the atmospheric boundary layer, where the greatest part of tropospheric aerosol is usually contained and transformed. Noticeable success has been achieved in recent years in the study of the transformation of fine aerosol in the boundary layer of the atmosphere over ocean[69] and coastal regions.[70] However, many aspects of kinetics and dynamics of tropospheric aerosol over continent are studied insufficiently.

The problem on studying the features of the vertical distribution of aerosol in the troposphere, that became classical long ago, is far from being resolved. Among the results obtained earlier let us note the weak dependence of the spectral behavior of the scattering coefficient on the height in the boundary layer of the atmosphere,[43] as it follows from our analysis of airborne nephelometric measurements.[71] Systematic airborne sounding of the atmosphere carried out by the researchers of the Institute of Atmospheric Optics made it possible to reveal the peculiarities in the vertical distribution of the scattering coefficient[72] and to establish some features of formation of the vertical structure of submicron aerosol.

The more detailed data on the diurnal dynamics of aerosol in the boundary layer of the atmosphere were obtained in Tomsk by the method of lidar sounding from the ground surface.[73] Vertical structure of aerosol in the lower part of the boundary layer can be also studied by means of the instrumentation mounted on the tall masts. An example of the diurnal behavior of the number density of aerosol particles with the radius greater than 0.2 μm obtained from measurements on the Ostankino TV tower[74] is shown in Fig. 6.

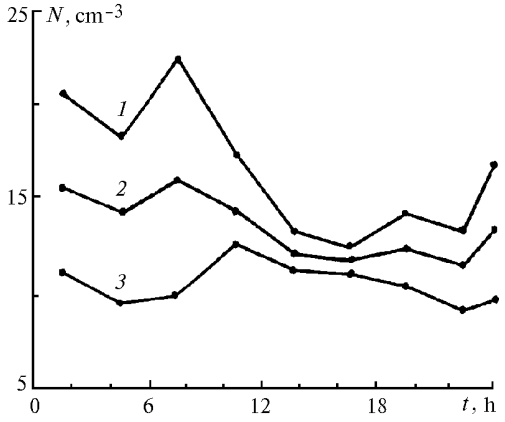

Fig. 6. Diurnal behavior of the number density of particles with radius greater than 0.2 μm from the data of measurements in Moscow (April 1987) at the height of 47 (*1*), 138 (*2*), and 350 m (*3*).

The examples appear now in literature of successful application of sounding of the aerosol vertical distribution by means of measuring the spectral phase functions of the sky brightness from onboard an aircraft.[75] This approach is very promising, because it allows to reconstruct the vertical profile of the particle size-distribution. The most effective method for the atmospheric column is the method of diagnostics of the aerosol microstructure using the data of simultaneous

measurements of the atmospheric transmission and the sky brightness phase function in the solar aureole in quite a wide wavelength range.[76] Let us note that the efficiency of the solar aureole method was successfully demonstrated when studying the dust haze by researchers of the Institute of Atmospheric Physics and Institute of Atmospheric Optics.[77] The successful selection of the method of microstructure parameterization is of a great importance for interpretation of data of optical sounding of the aerosol atmosphere. In our opinion, a good example is the three-fraction model proposed in Ref. 76.

To understand the aerosol evolution in the boundary layer of the atmosphere, it is necessary to study and examine the regimes of formation of the aerosol spatial distribution. When solving this problem, serious difficulties appear related to the insufficient study of "meteorological regimes" of the boundary layer of the atmosphere, especially under conditions of convection.[78,79]

We stated the problem of diagnostics of the regimes of formation of the aerosol boundary layer of the atmosphere in the early 90's. Different regimes of the aerosol boundary layer were revealed in 1991 by means of laser sounding from onboard a research vessel in Atlantics.[80] In particular, the high-amplitude waves were found at the boundary of inversion in the boundary layer of the atmosphere, that is an evidence of the unavoidable "uncertainty" in the diurnal behavior of the aerosol vertical distribution.

The fundamentally important results of investigation of the regimes of formation of the aerosol spatial distribution were obtained by the method of airborne lidar sounding. The characteristic structures of the aerosol distribution in the boundary layer of the atmosphere were observed during flights over Kalmykia in summer 1996 and 1997. That is an evidence of the existence of certain regimes of the convective boundary layer. In particular, the regime of nonpenetrating convection was revealed, which is characterized by the vertical position of the boundaries between ascending and descending convective cells.[82] It was revealed that the regime of nonpenetrating convection can occur as a regime of quasi-regular convection, when the ascending and descending cells form a quasi-periodic structure with the period of approximately 10 km, and as a regime of irregular convection, when the cell diameters randomly vary approximately from 1 to 5 km.[81] It is essential that the large-scale structure of the convective boundary layer is well pronounced in the temporal variability of the turbulent characteristics of the boundary layer of the atmosphere.[53] Apart from the nonpenetrating convection of the Benar scale, the situations were observed, for which the well known model of thermic convection is most appropriate.[83]

It is important to note that convection provides for a relatively quick mixing of admixtures in the boundary layer of the atmosphere. However, the process of mixing does not occur in the same way everywhere. So, when constructing the models of aerosol kinetics and dynamics in the boundary layer of the atmosphere, it is expedient to rely on the models with high spatial resolution, including the horizontal direction.[84]

Radiative effects of the transforming aerosol

It is necessary to take into account the kinetics of atmospheric aerosol in solving the problems of theory of climate and atmospheric ecology. It is expedient to illustrate the significance of the effects of aerosol transformation by analyzing, for example, the effect of the process of transformation of anthropogenic aerosol on the radiative regime in the atmosphere.

In studying the radiative processes, one cannot describe the microstructure of atmospheric aerosol by single particle size-distribution function $g(a)$. So the microphysical model of anthropogenic aerosol in the boundary layer of the atmosphere was proposed,[85] in which the use of preset particle size-distributions $g_i(a)$ (i = 1, 2, 3) of soot, dust, and water soluble aerosol was proposed. It is clear that the model is geophysically poorly stated, first of all, because it absolutely ignores the processes of aerosol transformation which can be described by the system of kinetic equations

$$\frac{\mathrm{d}g_i(a)}{\mathrm{d}t} = \sum \left[\frac{\partial g_i(a)}{\partial t}\right]_k, \qquad (21)$$

where the index i is the number of the distribution function of the ith aerosol component, and the index k is the number of the aerosol transformation process to be taken into account.

One can represent the rate of the coagulation transformation of $g_i(a)$ by a sum of two terms

$$\left[\frac{\partial g_i(a)}{\partial t}\right]_{\mathrm{cg}} = \left[\frac{\partial g_i(a)}{\partial t}\right]_{\mathrm{cg}}^{ii} + \left[\frac{\partial g_i(a)}{\partial t}\right]_{\mathrm{cg}}^{ij}, \qquad (22)$$

responsible for the intracomponent and inter-component coagulation.

Thus, the components of soot-water-soluble, soot-dust, and soot-dust-water-soluble aerosol are formed in the atmosphere instead of the external mixture of soot, dust, and water-soluble particles. From this it follows that it is necessary to examine the physical-chemical composition of mixed particles taking into account the processes of gas-to-particle conversion.

To illustrate the effect, it is convenient to restrict oneself to the particular case of soot-water-soluble aerosol.[86] One can use the single-reservoir approximation for the regime of quick mixing in the boundary layer of the atmosphere. Let us suppose that soot particles with the effective radius a_s = 0.005 μm are emitted into the atmosphere. Let us select the steady state number density N_s, or mass concentration M_s as the input parameter characterizing the soot aerosol component. The absorption coefficient values in

the visible wavelength range α_s corresponding to the selected values of a_s and N_s are presented in Table 2. Let us simulate the water-soluble aerosol by a sum of two fractions of submicron particles. The one of these two fractions is formed due to emission of water-soluble particles with the radius of the dry matter $a_0 = 0.05$ μm into the atmosphere, and the second one appears due to the growth of particles mainly caused by the processes of gas-to-particle conversion. In particular, the specific case was considered when the effective particle size of the former fraction increased by 40%, and the stationary number densities of particles of both of the fractions occurred to be equal to $9.15 \cdot 10^3$ and $0.85 \cdot 10^3$ cm^{-3}, respectively. The water-soluble particles were transformed to the soot-water-soluble ones due to the coagulation process. We have calculated the volume fractions of soot in the mixed particles (see Table 2). Then the extinction coefficient ε and the single scattering albedo Λ were calculated for this model in the approximation of homogeneous spherical particles of submicron aerosol. In particular, it occurred that the absorption coefficient of the soot-water-soluble fraction significantly exceeds the absorption coefficient of the soot fraction. One should note that the variations of the model parameters in acceptable limits do not contradict the main conclusion, that the account for the processes of aerosol transformation in the boundary layer of the atmosphere leads to a drastic change of its physical-chemical composition and radiative characteristics. The influxes of solar radiation to the aerosol layer ΔF_1 and the underlying surface ΔF_2 were calculated by means of the short-wave solar radiation transfer theory.[87] Let us note that the calculation was performed for the 30° zenith angle of the Sun and albedo of the underlying surface of 0.2.

Table 2. Radiative effects of the transforming aerosol

Parameter	Values		
M_s, μg/m^3	1.0	0.1	0.01
N_s, cm^{-3}	10^6	10^5	10^4
α_s, km^{-1}	$1.1 \cdot 10^{-2}$	$1.1 \cdot 10^{-3}$	$1.1 \cdot 10^{-4}$
δ_1	0.515	0.096	0.011
δ_2	0.150	0.017	0.002
ε, km	2.40	1.49	1.39
Λ	0.52	0.85	0.98
ΔF_1	0.75	0.30	0.05
ΔF_2	0.14	0.47	0.62

It is clear that complex aerosol experiments with certain instrumentation are necessary for providing better data for the radiative aerosol models. In addition to the scattering and absorption coefficients it is necessary to measure the mean cosine of the scattering phase function and the parameter of the aerosol condensation activity. To extrapolate the aerosol optical parameters to other wavelengths, it is necessary to examine the Angström parameter as well as the parameter that characterizes the spectral behavior of the absorption coefficient. One can estimate the contribution of the coarse fraction from the measurement data on aerosol microstructure. Among the available experimental data one can select the measured results on mass concentration of soot aerosol carried out by A.S. Emilenko and V.M. Kopeikin in Moscow, Beijing, and other sites.[88] According to these data, mean value of the single scattering albedo in Moscow is equal to 0.83, and in Beijing it is 0.78. The minimum measured value Λ in Beijing is 0.63. It follows from these data that the kinetic model we considered as an example makes it possible to reliably estimate the effects of the transforming aerosol in urbanized regions.

Conclusion

The principle results of investigations in the field of optics and physics of atmospheric aerosol carried out at the Institute of Atmospheric Physics are formulated in this paper. The genesis has been followed up of modern investigations into the kinetics of fine aerosol in relation to investigations of optical and microphysical parameters of the atmospheric aerosol.

The variations of angular dependences of the scattering phase matrix elements were studied, including the coefficient of directional scattering in the angular range from 0.5 to 178°. It is shown that the regular change of the scattering phase matrix of atmospheric haze as the turbidity of air increases is described by means of a single-parameter optical model. The peculiarities of the diurnal behavior of the aerosol optical characteristics in the near-ground layer of the atmosphere have been revealed.

It was shown that under conditions of moist haze aerosol optical characteristics in the visible wavelength range are mainly determined by the submicron fraction. The single-parameter model of the near-ground aerosol was developed, which agrees with the statistical model of the scattering phase matrix. The microphysical model well represents the main tendency of the variability of microphysical parameters of the submicron fraction of natural aerosol and its dry matter as the turbidity increases. When comparing the microphysical models for different seasons and regions, it was revealed that the principal difference between them is related to the variability of the mean number density of submicron aerosol. The features of the diurnal transformation of the microphysical parameters of the submicron aerosol fraction are revealed by the method of inverse problem of the light scattering from the measured data on aerosol optical characteristics.

The role of principal transformation processes in the submicron aerosol formation have also been analyzed. It was shown that the variations of the scattering coefficient, angular dependences of the coefficient of directional scattering, size and volume of particles, and the refractive index of the aerosol particle substance at the change of relative humidity can satisfactorily approximated by the Kasten-Hanel

formula. Optical characteristics (including the angular dependences of the elements of scattering phase matrix) and microphysical parameters of the aerosol dry matter were obtained. Analysis of the statistical features of the variability of the natural aerosol parameters, as well as its dry matter and features of the diurnal behavior of these parameters shows that, along with the process of condensation of water vapor, the processes of gas-to-particle conversion and the processes of condensation of the vapor of aerosol producing compounds play essential role in the formation of submicron aerosol. The rate of accumulation of the mass of the aerosol dry matter was estimated.

The rates of particle removal from the near-ground layer of the atmosphere, the generation rates and rates of cluster removal, as well as other kinetic parameters were obtained by means of few-parameter models of the kinetics of fine aerosol. It was revealed that the transformation of submicron aerosol under conditions of moist haze (autumn, night) is determined by the mechanism of growth characterized by the constancy of the particle size-distribution function. One can explain the recently found non-monotonic dependence of the parameter of aerosol condensation activity on particle size in the submicron range by the fact that, along with the processes of condensation of the aerosol producing compounds with relatively small solubility in a wide particle size range, condensation of the aerosol producing compounds with high solubility in water occurs on the particles with the size exceeding some threshold value determined by the Kelvin effect.

The process of emission of arid aerosol from the underlying surface is yet poorly studied. We have revealed the regimes of emission of the arid aerosol as peaks related to the wind gusts and zones of enhanced temperature of air in the near-ground layer of the atmosphere.

Kinetics and dynamics of aerosol in the near-ground and boundary layers of the atmosphere are mainly determined by the regime of the boundary layer of the atmosphere. From data of airborne lidar-nephelometric sounding we have revealed different regimes in the convective boundary layer of the atmosphere, including the regimes of quasi-regular and irregular nonpenetrating convection of the Benar scale.

The processes of aerosol transformation can lead to drastic changes in the microstructure and physical-chemical composition of the aerosol. In particular, the radiative characteristics of the multicomponent anthropogenic aerosol transforming in the boundary layer of the atmosphere significantly differ from the radiative characteristics of non-transforming aerosol. The results of direct measurements of the single scattering albedo in urban areas are an evidence of the reality of the models of transformation of the multicomponent aerosol. Significant changes of the radiation regime of the atmosphere and the underlying surface are observed at the enhanced aerosol filling of the boundary layer of the atmosphere over a city.

Thus, the prerequisites are created now for revealing the principal features of the microstructure and physical-chemical composition of the fine tropospheric aerosol particles over continent on the basis of the development of kinetics and dynamics of a multicomponent aerosol.

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