STUDY OF THE DYNAMICS OF EVOLUTION OF OPTICALLY DENSE WINTER HAZE BY THE TECHNIQUE OF INVERSION OF THE MEASURED SPECTRAL TRANSMITTANCE OF THE ATMOSPHERE

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Temporal transformation of the particle size spectrum of submicron and coarsely dispersed fractions of aerosols during formation and evolution of optically dense winter haze has been studied. Results have been obtained by the inverse–problem method from the data of experimental measurements of spectral dependence of the aerosol extinction coefficient within the wavelength range from 0.44 to 3.9 μm. The observed temporal behavior of absolute air humidity under conditions of constantly high (more than 95%) relative humidity was shown to correlate with the dynamics of the main integral microstructure characteristics of the aerosol accumulative fraction, namely, total geometric cross section, particle number density, and particle volume concentration. Variations in the modal radius and halfwidth of the accumulative particle size distribution function were synchronous.

Extensive experimental material on spectral transmittance of the ground atmosphere on a horizontal path in winter has been obtained in Tomsk as part of the long–term integrated program for atmospheric researches of the Institute of Atmospheric Optics. A statistical analysis of the experimental data on the aerosol extinction coefficients in the visible and IR ranges for different types of optical weather and of their interrelation with the synchronously measured meteorological parameters of the atmosphere was made in Ref. 1.

Further interpretation of the measured spectral dependence of the aerosol extinction coefficients by the inverse–problem method is made in this paper to investigate the poorly–known aerosol microstructure transformation in winter and its interrelation with the meteorological parameters of the atmosphere.

An instrumental complex for measuring the spectral transmittance of the atmosphere within the wavelength range from 0.44 to 12 μm and the experimental procedure have been described in Ref. 2. The algorithm used in this paper for solving the inverse problem was constructed by the Tikhonov regularization method and was described in detail in Ref. 3. Spectral dependence of the volume aerosol extinction coefficients in the atmospheric windows $\beta_{ex}(\lambda)$ measured in Tomsk on 1 km horizontal path at 10 different wavelengths from the range between 0.44 and 3.9 μm were the initial information in the inverse problem solution. As is well known, the volume aerosol extinction coefficients at the wavelength λ_i, on the assumption that scattering particles are spherical, are defined by the integrals

$$\beta_{ex}(\lambda_i) = \int_{r_1}^{r_2} K_{ex}(m, r, \lambda_i)\, s(r)\, \mathrm{d}\, r, \qquad i = 1, 2, \dots, n, \tag{1}$$

where $K_{ex}(m, r, \lambda)$ is the extinction efficiency factor calculated by the Mie theory, m is the complex refractive index of particulate matter, $s(r) = \pi r^2 n(r)$, and $n(r)$ is the particle size distribution function within the interval of particle radii $[r_1, r_2]$. The system of equations (1) is reduced to algebraic form

$$\sum_{l=1}^{\kappa} Q_{ex,\, il}\, s_l = \beta_{ex,\, i}\,, \qquad i = 1, 2, \dots, n\,. \tag{2}$$

The vector $\mathbf{s}$, which minimizes the quadratic form

$$T_\alpha(s) = \sum_{i=1}^{n} \left(\sum_{l=1}^{\kappa} Q_{ex,\, il}\, s_l - \beta_{ex,\, i} \right)^2 + \alpha\, \Omega(s) \tag{3}$$

on a set of positively defined vectors Ψ^+, is taken as a solution of this system, where α is the regularization parameter consistent with the level of the measurement error. A form of stabilizing functional $\Omega(s)$ depends on limitations imposed on the desired solution of the inverse problem.

Figure 1 depicts temporal transformation of the particle size spectrum in the successive run of measurements performed on December 14–16, 1992.

The values of the meteorological visibility range (MVR) are tabulated in Table I as a characteristic of optical situation in the atmosphere during the measurement period.

The numbers of the curves in Fig. 1 (also indicated in Table I) denote time counts in the measurements. Curves from *1* to *3* were recorded on December 14 from 17 to 21 h two hours apart, curves *4* and *5* were recorded on December 15 from 1 to 3 h, curves *6* to *13* were recorded on December 15 from 9 to 23 h two hours apart, and curves *14* and *15* were recorded on December 16, from 1 to 3 h, LT.

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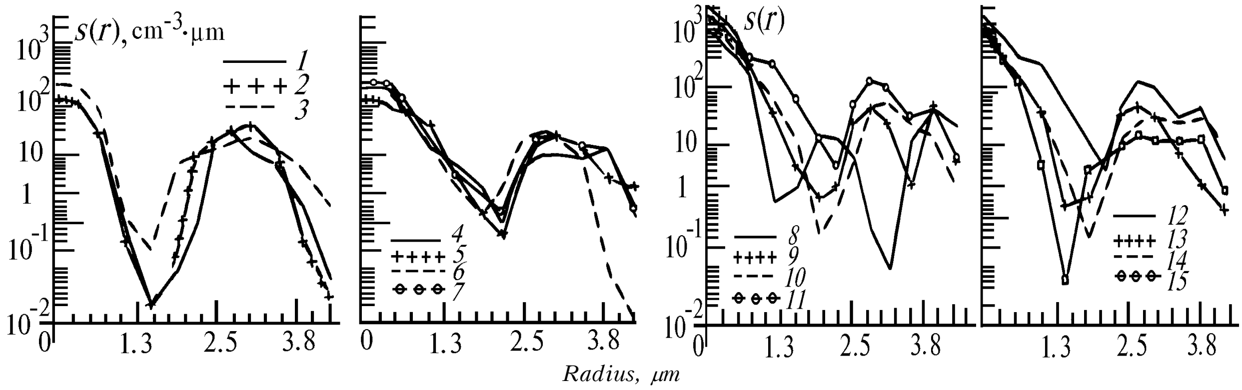

FIG. 1. Dynamics of the distribution function s(r) during the evolution of optically dense haze.

TABLE I. Characteristic of optical situation in the atmosphere in the run of measurements performed on December 14–16, 1992.

N	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
MVR, km	24	23	12	8.3	9.3	5.9	6.2	4.8	2.3	3.6	2.3	5	4	5	6
Wind direction, deg	180	193	142	171	152	133	143	343	120	74	136	–	–	–	–

From Fig. 1 it is apparent that there is an explicit boundary between accumulative and coarsely dispersed fractions of particles. In its vicinity in the range of particle radii between 1 and 2 μm, the particle size distribution is unstable, and the concentration of particles is minimum. Special attention must be given to the qualitative transformation of the particle size spectrum on December 15 at 13 h (curve *8* in Fig. 1), namely, to the occurence of intermediate fraction in the range of particle radii between 1.3 and 2.5 μm. It should be noted in this connection that the measurement station was located so that the main sources of industrial pollution of the atmosphere were in the western and north–western directions at a distance of longer than 3 km, while the south wind blew from rural regions.

As seen from the table, the occurence of the intermediate fraction in the particle size distribution coincides with sharp change of wind direction from south to north–west, what is indicative of possible anthropogenic origin of this fraction of particles.

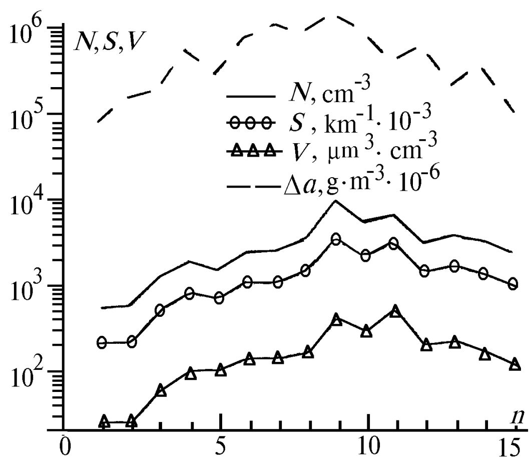

FIG. 2. Temporal behavior of the reconstructed parameters of aerosol microstructure during the formation of optically dense haze and dynamics of the absolute air humidity.

The joint analysis of the reconstructed integral parameters of aerosol microstructure and meteorological data (Fig. 2) shows that in this run of measurements the temporal behavior of the absolute air humidity correlated with the dynamics of the basic characteristics of submicron fraction of particles (total geometric cross section $S1$, particle number density $N1$, and volume concentration $V1$). The value Δa is represented in Fig. 2 in the form of "losses" of absolute humidity (g/m^3) with respect to its initial value. The numbers of counts along the horizontal axis in Fig. 2 (the same is valid for Fig. 3) denote the time intervals specified in description to the table. Simultaneously with the dependence shown in Fig. 2, during the evolution of dense haze we could observe the synchronous changes in the modal radius and halfwidth of the accumulative particle size distribution function $s(r)$ depicted in Fig. 3. The parameter $b = 1/2\ (\ln\sigma)^2$, where σ is the standard deviation from $\ln r$, defines the halfwidth of the accumulative particle size distribution function when it is described by lognormal distribution widely used in the analysis of aerosol microstructure.

It is significant that during this process the relative humidity of air remains practically constant with minor deviations from the mean value $f = 96$ %. In this period of observations in Tomsk there was an air mass of moderate latitudes with slight negative air temperature of about – 7.6° C.

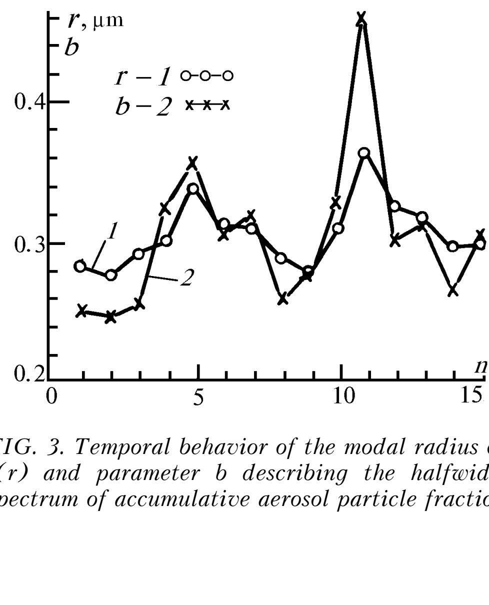

FIG. 3. Temporal behavior of the modal radius of function s(r) and parameter b describing the halfwidth of size spectrum of accumulative aerosol particle fraction.

The tendencies depicted in Figs. 2 and 3 were not observed for coarsely dispersed particle fraction which could be accounted for by different physical mechanisms of formation of these fractions.

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