Microphysical interpretation of the anomalous spectral behavior of aerosol extinction along a ground path

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The paper analyzes possible causes of the formation of situations rarely observed in the atmospheric boundary layer when anomalous spectral dependence of the aerosol extinction coefficient occurs with the invasion of the Arctic air mass. The solution of the inverse problem have shown that the invasion of Arctic air manifests itself in the aerosol disperse structure as a drastic fall in the content of the particles of accumulative fraction, narrowing of the medium-disperse fraction, and shift of the entire particle size spectrum to the region of small particles. At a more pronounced anomaly, the optical contribution of the accumulative aerosol fraction to the extinction of visible radiation decreased to a negligible level. The optical effect of coarse aerosols (at $r > 2$ μm) in the wavelength range 0.44 to 1.06 μm has a spectral behavior of the aerosol extinction coefficient close to a neutral one. Under these conditions the medium-disperse particles of the sizes from 0.3 to 1.8, μm with the main mode of particle geometric cross section distribution at about 0.6 μm and a broad second peak at about 1.4 μm have determined, in general, the anomalous spectral dependence of the measured aerosol extinction coefficients.

In September 2002 in the region of Tomsk we have observed a rare, for the near ground hazes, case of anomalous spectral dependence of the coefficients of the aerosol extinction $\beta_\varepsilon(\lambda)$ of radiation in the visible range.[1] At the same time, the formation of anomalous spectral dependence coincided in time with the process of change of air mass (AM), enriched with smoke aerosols, for arctic AM, whose coming was accompanied by a sharp decrease of temperature of the atmospheric boundary layer by 20 degrees. Aerosol extinction maximum located generally in the violet spectral range ($\lambda \le 0.44$ μm) was shifted to the range $\lambda \cong 0.8$–1.4 μm.

Possible causes for the occurrence of unusual spectral behavior of the aerosol optical thickness $\tau_a(\lambda)$ of the atmosphere, first determined by S.F. Rodionov,[2] were analyzed in detail in several papers.[3–5] The investigations showed that the specific spectral behavior of $\tau(\lambda)$ can be explained by the peculiarities of aerosol microphysical composition typical for relatively clear air. The detection of anomalous spectral behavior $\beta_\varepsilon(\lambda)$ by averaging of this value along a near ground horizontal paths at mid-latitudes is a more unique event. During warm seasons the formation of aerosol disperse composition close to the underlying surface usually proceeds on the background of emission of aerosol-forming compounds from different sources. Therefore in the cases of sharp decrease of the content of particles of the accumulative fraction, being a part of a near ground haze, their deficit is rapidly replenished from nearly local sources as well as the activity of the process of intraatmospheric kinetic development of optically low-active microdisperse fraction.

Figure 1 shows the mean, for September, spectral dependence of the aerosol extinction coefficients $\overline{\beta_\varepsilon(\lambda)}$ (curve *1*) with root-mean-square deviations and several single spectra obtained during the period of change of smoky air mass for the arctic mass (curves *2*–*5*). As seen from the figure, curve *2* measured on September 12 at 13^{00} (local time) throughout the entire spectral range exceeds essentially the upper limit of dispersion of the spread of measured values relative to the mean value. These high values of the coefficients $\beta_\varepsilon(\lambda)$ in the wavelength range analyzed are the result of the arrival to the measurement site of the AM enriched with aerosols of smoke origin. Analysis of AM motion trajectory and synoptic situation ensures that these smokes are transported from far regions on the European territory of Russia.

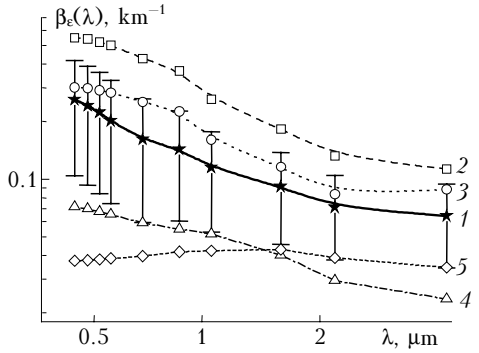

Fig. 1. Measured spectral dependences of the aerosol extinction coefficients: mean value in September (*1*) and individual realizations obtained on September 12 at 13^{00}, on September 13 at 7^{00}, on September 14 at 7^{00}, and on September 14 at 17^{00} (*2*, *3*, *4*, *5*).

Maximum of the atmospheric turbidity was reached approximately at noon, September 12, after

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which the optical contribution of accumulative fraction in the visible spectral range decreased from 85% to 73%. In the morning of September 13, the spectral dependence $\beta_\varepsilon(\lambda)$, even if it fell within the deviation corridor (curve *3*), nevertheless it exceeded considerably $\overline{\overline{\beta_\varepsilon(\lambda)}}$.

The invasion of the arctic air mass changed sharply the optical situation, and in the morning on September 14, the spectral dependence of $\beta_\varepsilon(\lambda)$ was found to be not only noticeably lower than the monthly mean curve but was beyond the corridor of the root-mean-square deviations over the entire wavelength range, thus belonging to the anomalous type only by this feature. At noon, on September 14 the values of the aerosol extinction coefficient in the visible range continued to decrease and, as a result, the spectral dependence was anomalous in the visible range (curve *5*).

The solution of inverse problem based on the spectral dependences of $\beta_\varepsilon(\lambda)$, presented in Fig. 1, shows (Fig. 2*a*) that the characteristic property of the smoke aerosol microstructure (curves *1*, *2*) is the shifted right boundary of the accumulative fraction $r_{2,a}$ up to the values ~0.7–0.75 μm. In this case the spectra of $s(r)$ shown in Fig. 2*b* (curves *1–3*) reconstructed according to data, measured on September 13 before the invasion of the arctic air mass, display a rare for accumulative fraction shape stability of the size spectrum and point to the conservation of smoke aerosols up to 21^{00} September 13.

Only in the morning of September 14 (curve *3*, Fig. 2*a*) when the arctic air mass fully replaced the preceding mass, enriched with smoke particles, the particle size spectrum of the accumulative fraction took the conventional shape with the right boundary of $r_{2,a}$ at about ~0.4–0.45 μm.

It should be noted that the right boundary of the monthly mean spectrum of the accumulative fraction, in spite of frequent disturbances of the atmosphere in September by smokes, is shifted to the left and has the same right boundary as in the curve *3* (see curve *5* in Figs. 2*a*,*d*).

Figure 2*c* shows the dynamics of daytime transformation of the disperse structure of the near ground haze on September 14, which shows that at the invasion of the arctic air mass the content of particles of the accumulative fraction decreased sharply and the intermediate disperse fraction played a leading part in the radiation extinction. The particle size spectrum of the intermediate disperse fraction, forming a part of the arctic air mass, was shifted to the region of smaller particles and was more narrow than that obtained from the average data for September (curve *5* in Fig. 2*a*).

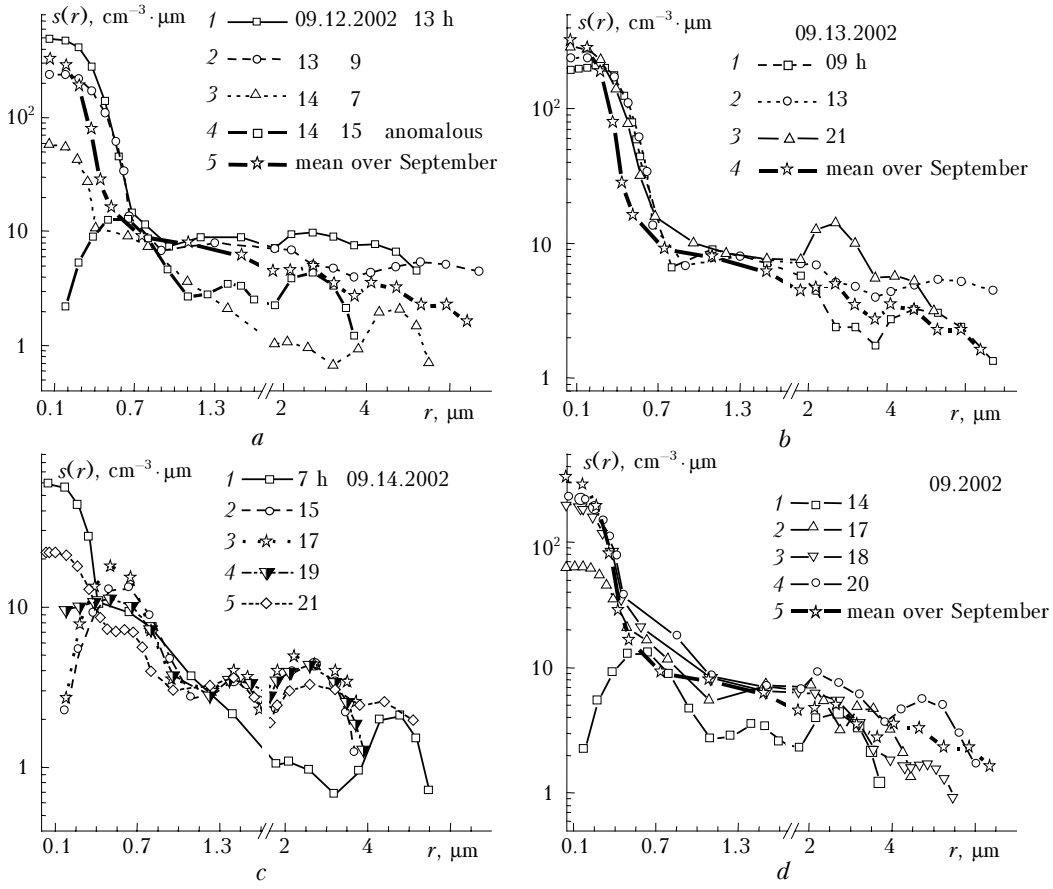

Fig. 2. Results of inversion of unit and average spectral dependences of $\beta_\varepsilon(\lambda)$ based on data for September 2002.

Optical contribution of the accumulative fraction to the radiation extinction in the visible range decreased up to negligible (against the background of the measurement errors) values. Stability of spectrum shape (curves *2–4*, Fig. 2*c*) points to a rather high homogeneity of the distribution of the intermediate disperse aerosol fraction entering into the composition of the arctic air mass.

The afternoon dynamics of the variation of the disperse composition of the near ground haze on September 14 shows (curves *3–5*, Fig. 2*c*) that the deficit of particles of the accumulative fraction occurring as a result of the invasion of the arctic air mass began to make up rapidly owing to the action of local sources. Stable increase during daytime of the optical effect of the largest particle fraction is observed. This fact also follows from the results of inversion of $\beta_e(\lambda)$ measured in warmer moderate air masses. In particular, in Fig. 2*b*, where the results of inversion of data measured on September 13 are shown, in spite of the relative stability of the size spectrum of the accumulative fraction, the content of coarse-disperse fraction in the daytime increases.

Dynamics of the development of the particle size spectrum of the near ground haze from anomalous situation to a conventional one is presented in Fig. 2*d*. In this case anomalous situation is presented by an individual spectrum (curve *1*), while other spectra (for September 17, 18, and 20) have been averaged per day. First of all, special attention must be given to the fast growth of the content of the accumulative fraction particles, which already by September 20 was fully reconstructed up to its usual concentration. Particle content of the intermediate disperse fraction during three days varied only slightly. It should be noted that the rise of air temperature in the ground layer results in an increase of the coarse particle content owing to strengthening of the convective aerosol lifting from the underlying surface. It is of interest to note that as the soil becomes warm, the right boundary of the coarse-disperse fraction distribution is gradually shifted (during 3 or 4 days) from 4.0 μm to 7–9 μm.

The quantitative characteristics of the variation in the volume content of particles of the coarse-disperse fraction V_{cr} over a period from 14 to 20 September are in a good agreement with the dynamics of the variation of daily mean temperature values of the ground layer t°C (Fig. 3*a*). The dynamics of the volume content variation of the accumulative fraction V_{ac} during those days agrees well with the variation of the absolute air humidity a (Fig. 3*b*).

However, one has to keep in mind that the agreed variation of V_{ac} and the absolute air humidity is only the result of the change of a relatively dry arctic air mass by a more humid air mass of midlatitude. Thus the dynamics of V_{ac} observed is due to the transition from one typical (for arctic air mass) level of content of aerosol particles of the accumulative fraction to the other one, in this case, characteristic of the mid-latitude air mass.

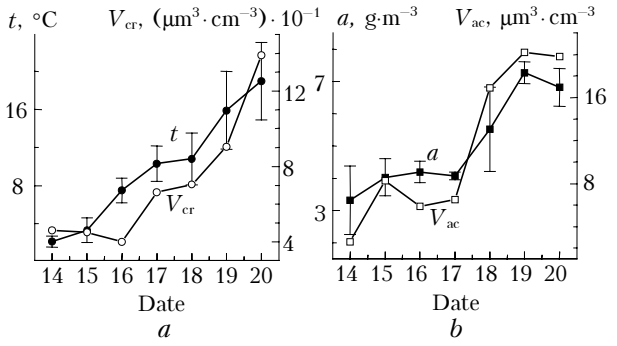

Fig. 3. Interdiurnal variability of the volume content of particles of the coarse-disperse fraction and daily mean temperature of the ground layer (*a*) and particle volume content of the accumulative fraction and absolute air humidity (*b*).

If we compare the particle size spectrum in the ground layer in anomalous situation $s(r)$ (Fig. 4*a*, curve *1*) with similar data on the atmospheric thickness $s_c(r)$ obtained using the inversion of $\tau(\lambda)$[3–5] (curves *2*, *3*), we must assure ourselves that the intermediate disperse fraction in the ground layer is the same in size as the average in thickness but wider in the distribution shape. In this case, Fig. 4*a* shows the results obtained by inversion of measurement data acquired in different time (dates are given in figure captions) but in both of these cases at the invasion of the arctic air mass to the region.

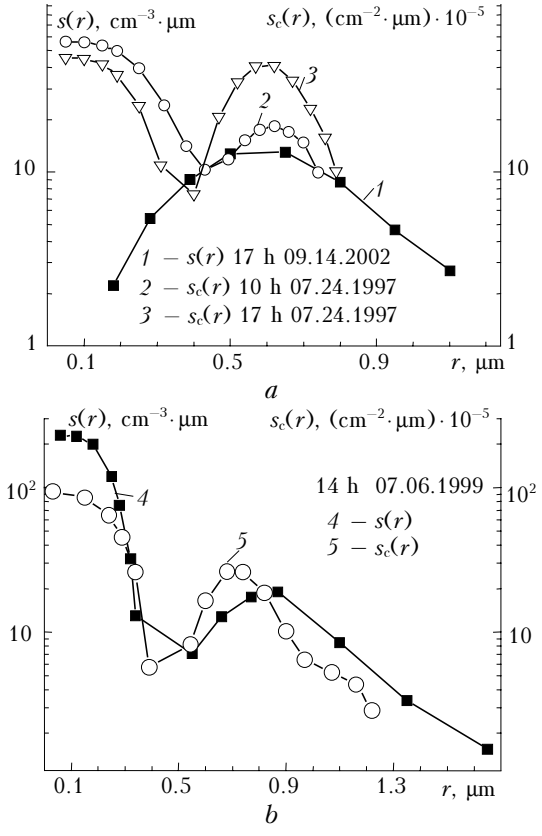

Fig. 4. Comparison of particle size distributions in anomalous (*a*) and ordinary (*b*) situations reconstructed from the inversion of $\beta_e(\lambda)$ and $\tau_a(\lambda)$.

Figure 4*b* shows the size spectra of haze obtained from data of simultaneous measurements of extinction along a horizontal (curve *5*) and slant (curve *4*) paths but already in the midlatitude continental air mass. In this case the intermediate disperse aerosol fraction in its whole atmosphere thickness and in the ground layer also is in one particle size range but shifted to the right as compared with the anomalous situation. Thus the results shown in Figs. 4*a* and *b* support once more a conclusion discussed before[4] about the fundamental significance of the intermediate disperse fraction of aerosol as an important component of the background state of the atmospheric haze.

When inverting the data of optical measurements, we obtained the estimates of the contribution of different fractions to the integral spectral dependence $\beta_\varepsilon(\lambda)$. In particular, Fig. 5 shows the contributions to the spectral dependence of the extinction coefficient coming from the accumulative, intermediate, and coarse-disperse fractions as well as the sum of these contributions. In our analysis we used the measurement data acquired on September 12 to 14, 2002. For September 13 the morning and midday dependences of $\beta_\varepsilon(\lambda)$ were used.

It is not difficult to understand from data shown in Figs. 5*a*–*c* that in the measurements of $\beta_\varepsilon(\lambda)$ on September 12–13 (in the smoky atmosphere) the increased contribution of accumulative fraction particles was observed (curves *2*). The closeness of spectral dependences of this fraction in the period of about 35–40 hours points to the stability of particle concentration and conservation of the spectrum shape. In particular, on a log-log scale we observe the convex shape of the spectral behavior of $\beta_\varepsilon(\lambda)$ different than the Angström law. The relative contribution of the accumulative fraction to the integral spectral dependence of $\beta_\varepsilon(\lambda)$ in the visible spectral range ($\lambda < 0.69$ μm) varies within this interval from 73 to 86%.

In the near infrared range at $\lambda \sim 1.5$ μm the optical effect of smoke aerosols decreases down to 30%. At the same time, in spite of high content of smoke aerosols on September 12–13, the total contribution of the intermediate and coarse fractions in the range $\lambda > 2.0$ μm is a defining factor not only in the afternoon but also in the morning (see Figs. 5*a*–*c*). Data presented in Fig. 5*d* show the variation of the relative contributions of fractions at the invasion of the arctic air mass, which took place on September 14. In this case the optical contribution of accumulative fraction turned out to be negligible and the contribution of intermediate disperse aerosols (curve *3*) that determined the anomalous spectral behavior of $\beta_\varepsilon(\lambda)$ (Fig. 5*d*) throughout the entire spectral range analyzed was more than 50% of the total aerosol contribution. The optical contribution of coarse-disperse aerosols (with $r > 2$ μm) in the wavelength range from 0.44 to 1.06 μm has the spectral behavior close to the neutral one (curve *4*). Besides, Fig. 5*d* shows for a comparison the total spectral dependence of $\beta_\varepsilon(\lambda)$ of the intermediate disperse and coarse-disperse fractions (curve *2*) calculated using the results of solution of inverse problem, which can be considered as the optical approximation of the measured values.

Summing up the obtained results as a whole it should be noted that the main factor of appearance of nonstandard spectral behavior $\beta_\varepsilon(\lambda)$ in the ground layer, as well as in the entire atmospheric thickness,[4]

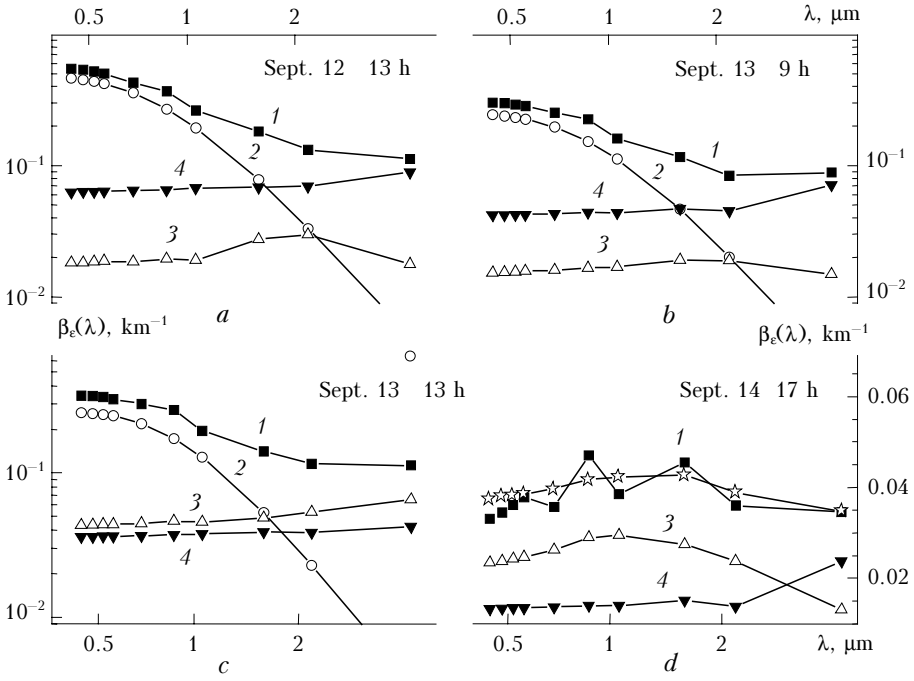

Fig. 5. The variation of the optical contribution of accumulative, intermediate, and coarse aerosol fractions (curves *2*, *3*, *4*, respectively) in smoky and arctic air masses and measured values of $\beta_\varepsilon(\lambda)$ (*1*).

is the invasion to the measurements site of a relatively dry and rather cold air mass. The invasion of arctic air mass predetermined the sharp decrease in the content of aerosol particles of the accumulative fraction in the ground layer and essential increase of the optical effect of the intermediate disperse fraction. With the reconstruction of typical for moderate air mass level of water content and temperature the proportions usually observed between the fractions in the disperse composition of the surface aerosol have been reconstructed.

Acknowledgments

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