Variations of aerosol microstructure under smoke effect assessed from inversion of spectral optical measurements

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The results of spectral aerosol optical depth (AOD) measurements obtained in 1999, 2003, and 2004 are analyzed. These periods were characterized by a significant increase of the atmospheric turbidity due to forest fire smokes near Tomsk, as well as the arrival of remotely transported air masses. The measurements were carried out with the multi-wavelength sun photometer (λ = 0.35–4.0 μm). The dynamics of daily averaged AOD values in conditions of the smoke presence in the atmosphere for several days, as well as the corresponding transformation of the aerosol microstructure are treated based on results of solution of the inverse problem.

Introduction

The study of mechanisms of generation and propagation of smoke aerosols, produced from forest and peatbog fires, is extremely important for analysis of the specificity of formation of the atmosphere optical state over Siberian regions. Especially urgent is the estimation of the priority of different factors determining variations of the atmosphere optical transparency in summer, when the intensity of such events dramatically increases. As the result of the forest fires, a significant amount of aerosol and heat is released, the processes of sublimation of volatile compounds become more intensive, and the emission of the aerosol-producing gases (APG) from wood essentially increases.

The peculiarity of the smokes emitted into the atmosphere by biomass fireplaces is a high concentration of the dispersion mixture of aerosol particles and APG. The content of the disperse component in smoke emissions is several orders of magnitude greater than its standard level. The time of relaxation of these inhomogeneities to the equilibrium state, as well as the dynamics of the aerosol optical-microphysical parameters vary depending on the type of air mass (AM).

Vertical shift of the wind velocity in the troposphere and turbulent mixing, as a rule, break smoke flows in AM into spatially inhomogeneous cells. The disperse composition of smokes in these cells undergoes asynchronous variations. Horizontal motions of smoke anomalies are recorded by instrumentation as significant oscillations of the aerosol light-scattering parameters (ALSP). So it is difficult to indicate some characteristic (typical) state of smokes in the atmosphere.

Parameters of the disperse mixture (including the refractive index of particles) vary within the wide range of values because of instability of the regimes of thermal generation of smokes, that predetermines especial attention to laboratory investigations of their variability under controllable conditions in aerosol chambers.[1–5] At the same time, the state of smokes depends not only on the conditions of their generation (during the process of combustion or pyrolysis), but also on the factors regulating the specific manner of their propagation in the atmosphere and the efficiency of sink, i.e., on specific meteorological conditions in the episodes of field observations. Some cycles of smoke researches were carried out at IAP RAS (Moscow),[6,7] IAO SB RAS (Tomsk)[8,9] and other organizations. However, some aspects of the problem are, as before, studied insufficiently. In particular, experimental data are necessary, which could make it possible to determine the characteristic, mutual (mean statistical) variability of optical and microphysical parameters of smoke aerosols.

The purpose of our work was to study the factors determining the specific manner of disturbance of the optical state of the troposphere haze at intensive emissions of smoke aerosols into the atmosphere from different large-scale fireplaces. Main attention was paid to the study of peculiarities of the development of the disperse composition of aerosol phase at different stages of the spread of smokes in the atmosphere, in particular, smokes of local formation and remote transport, which determine the change of the spectral transparency of the atmosphere.

Technique of investigations

Analysis of the optical microphysical properties of different types of smoke aerosols was carried out using the data of spectral measurements of the aerosol optical depth (AOD) in 1999, 2003 and 2004 in the periods of the essential increase of the

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atmospheric turbidity under the effect of forest fires near Tomsk and their remote transport with air masses. The spectral transparency of the atmosphere was measured with the multi-wavelength sun photometer in the wavelength range 0.35–4.0 μm. Observations were conducted in short ~5–30 min long series, when the Sun was not covered with clouds. Hourly mean values were calculated from the obtained data. The techniques for calibration of the photometer and determination of AOD of the atmosphere were considered in Refs. 10 and 11. The columnar water vapor of the atmosphere W was measured together with the aerosol component.

Variability of aerosol microstructure characteristics was analyzed using the results of the inverse problem solution by the regulating algorithm based on constructing the smoothing functional by A.N. Tikhonov method[12] and its direct minimization in the k-dimension vector space. Peculiarities of the technique for inversion were considered in detail in Ref. 13.

The complex refractive index of particles was selected, taking into account the results of investigation of smokes under controllable conditions in aerosol chamber[3] using the data of spectral nephelometric measurements of polarized components of the directed light scattering coefficient at five fixed angles $\theta = 15$, 45, 110, 135, and 165° in the wavelength range $\lambda = 0.44$–0.69 μm. The obtained estimates[3] of the real part of the refractive index at pyrolysis of coniferous wood samples lie in the range 1.45–1.54, the imaginary part is less than 0.005, and their variations are correlated with the level of the absolute humidity.

Results and discussion

Figure 1 shows variations of AOD in two wavelength ranges in July and August, 1999 in the region of Tomsk, when essential increase of the atmospheric turbidity caused by fires was twice observed.

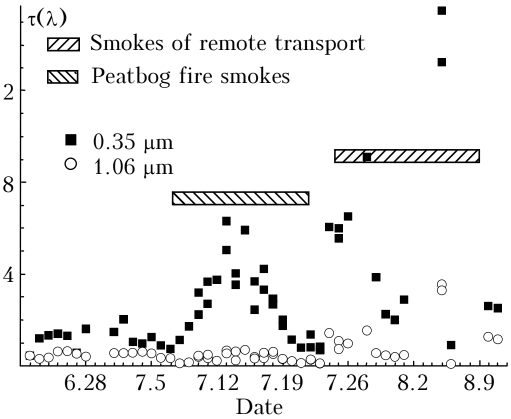

Fig. 1. Temporal dynamics of the atmosphere AOD in July–August, 1999 in two wavelength ranges.

Daily mean spectral dependences of AOD under different optical conditions are shown in Fig. 2.

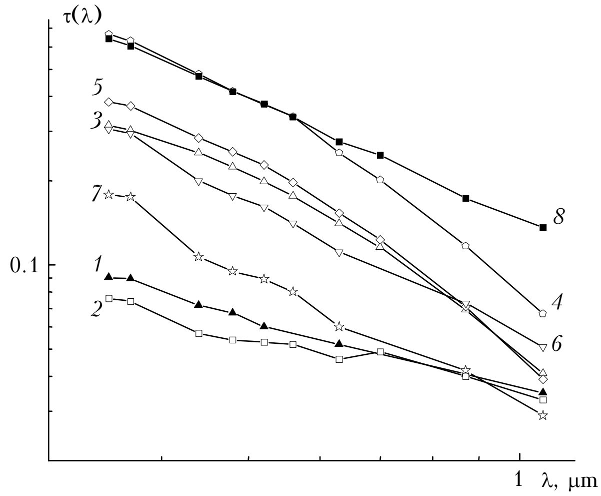

Fig. 2. Transformation of AOD spectra in the fire period: before smoke (on 6 and 7 of July, 1999) (*1*, *2*); in local smokes (peatbog burning in the vicinity of Tomsk), on 9 and 19 of July (*3*–*7*); in smokes of remote transport (forest fires in Krasnoyarsk Region, July 24) (*8*).

The decrease of atmosphere transparency in the first event (July, 9–19) was attributed to peatbog fires situated 10–20 km apart. Second increase of the atmospheric turbidity since July 24 till the first decade of August was related with remote transport of the smoke aerosol from forest fires in the Krasnoyarsk Region. It follows from the optical data (see Figs. 1 and 2) that essential increase (approximately by 4 times) of the aerosol turbidity in the short-wave range is common characteristic of both events (peatbog burning and forest fires). Main peculiarities of transformation of the aerosol microstructure under the effect of smokes are seen in the particle volume distributions (Fig. 3) retrieved from the data of Fig. 2.

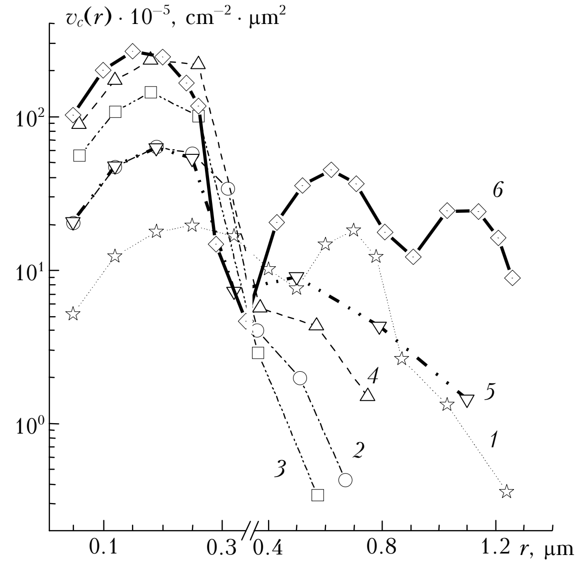

Fig. 3. Peculiarities of the columnar particle volume distribution: (*1*) before smoke (July 7, 1999); (*2*–*5*) in local smokes (peatbog burning in the vicinity of Tomsk, 9–19 of July); (*6*) in smokes of remote transport (forest fires in Krasnoyarsk Region, July 24).

The aerosol particle concentration of accumulative fraction ($r \sim 0.05$–0.35 μm) with the mode of the volume distribution $r_{\text{mod}} = 0.2$ μm increased under smoke conditions by more than an order of magnitude. At the same time, there are some peculiarities of smoke events with different prehistory.

When smoke from nearby sources has appeared in the atmosphere (peatbog smoldering), the content of large particles with the radius greater than 0.4 μm dramatically decreases together with increase of the concentration of small particles. At the same time, the results of inverting the spectral dependences of the aerosol extinction coefficient on horizontal path show the enhanced content of intermediately dispersed and coarse aerosols.[9] Under these conditions, the spectral dependence of AOD within the limits of the error in the measuring is determined by particles of accumulative fraction.

As the source of smoke aerosols weakened and the transparency increased, the content of particles $r > 0.4$ μm gradually increased and on July 18–19 it approached the level observed before the beginning of fire. Insufficiency of the data does not allow us to unambiguously relate the event of peatbog fires with the deficiency of large particles. Among the possible reasons of the effect, there may be weakening the processes of convection and turbulence (emission of particles) at decrease of the radiation heating of the underlying surface, change of air mass, and weak wind during the period of peatbog burning. Curves *1* and *2* in Fig. 2 show that the replace of continental mid-latitude air mass with subtropical one at night from 6 to 7 of July did not lead to essential change of daily mean spectral dependences of AOD. Peak values of the near-ground wind velocity during the period of observations did not exceed 5 m/s.

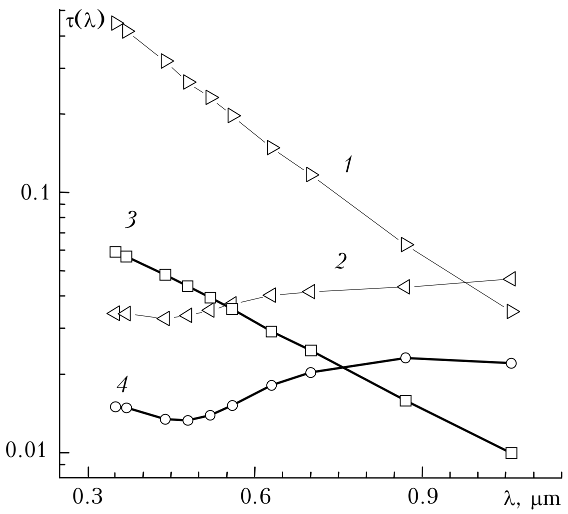

Fig. 4. Optical contributions of particles of accumulative and intermediately dispersion fractions into AOD in smokes of remote transport from forest fires in Krasnoyarsk Region (curves *1* and *2* are for July 25, 1999) and under pre-smoke conditions (*3* and *4* are for July 7, 1999).

Increase of the particle concentration in the size range 0.4–1.1 μm was observed in the event of remote transport of smoke from forest fires in Krasnoyarsk Region. The total volume of these particles was 2–3 times greater comparative to the clean atmosphere conditions.

The optical contribution of intermediately dispersed particles ($r > 0.4$ μm) was about 15% at $\lambda = 0.48$ μm and increases up to 50% in IR range (Fig. 4, curves *1* and *2*).

Another peculiarity was observed in the diurnal dynamics of the intermediately dispersed fraction in the atmospheric column. The particle size spectrum of this fraction varied during a day relative to some mean values and did not have a well-pronounced behavior with maximum at noon, which was observed in the atmosphere free of smoke (Fig. 5).

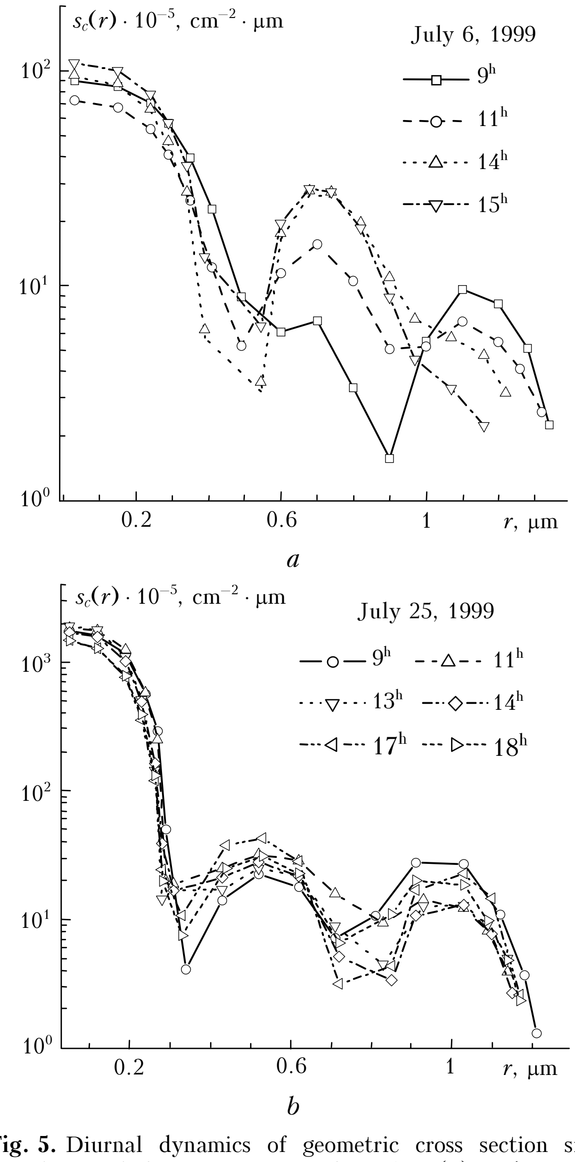

Fig. 5. Diurnal dynamics of geometric cross section size distribution before appearance of smokes (*a*) and at the presence of smoke due to the remote transport in the atmosphere (*b*).

It can be supposed that the intra-atmospheric transformation of the smoke aerosol accumulative fraction (gradual growth and partial transition to the size range $r > 0.4$ μm) is one of the factors of increasing concentration of intermediately dispersed aerosol.

The noted peculiarities of variations of the aerosol optical-microphysical characteristics under the effect of smoke were also observed in summers 2003–2004. The corresponding examples are shown in Figs. 6 and 7.

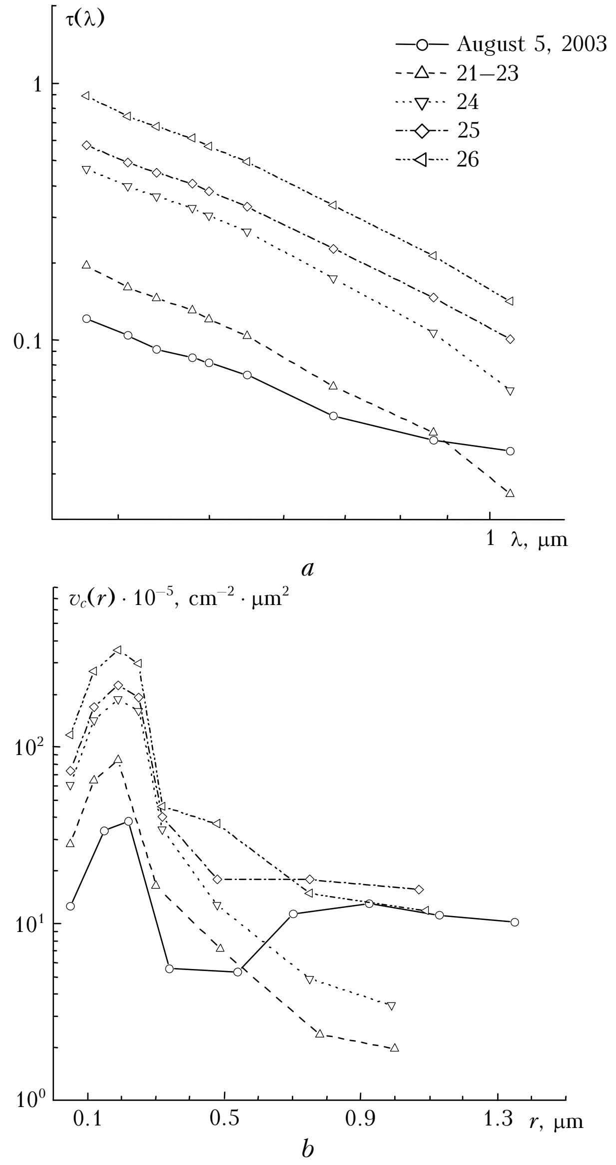

Fig. 6. Measured spectral dependences of AOD (*a*) and retrieved volume size distributions (*b*) under conditions of high atmospheric transparency (August 5, 2003) and in smoke event (August 21–26, 2003).

Temporal variations of τ(λ) and the retrieved distributions of $v_c(r)$ shown in Fig. 6 occur in the stable cloudless atmosphere under conditions of the Arctic air mass, being stayed for long time over the continent in mid-latitudes. When on August 21 (Fig. 6*a*), smokes have come, the aerosol microstructure changed analogously to the event shown in Fig. 3 during the period of peatbog burning near Tomsk. The total volume of accumulative fraction in the atmospheric column V_c in the range $r < 0.4$ μm increased from $8 \cdot 10^5$ to $16 \cdot 10^5$ cm^{-2} · μm^3. The total volume of larger particles in the size range $0.4 < r < 1.0$ μm dramatically decreased from $11 \cdot 10^5$ to $3 \cdot 10^5$ cm^{-2} · μm^3. Bimodal particle size distribution retrieved from the data on AOD under conditions of high atmospheric transparency before coming smoke was transformed into the monomodal size distribution (Fig. 6*b*), which could be considered as one fraction of primarily finely dispersion smoke. The mean particle size determined from the ratio $r_m = 0.75V_c/S_c$, where S_c is the total cross section of particles in the atmospheric column, is equal to 0.15 μm.

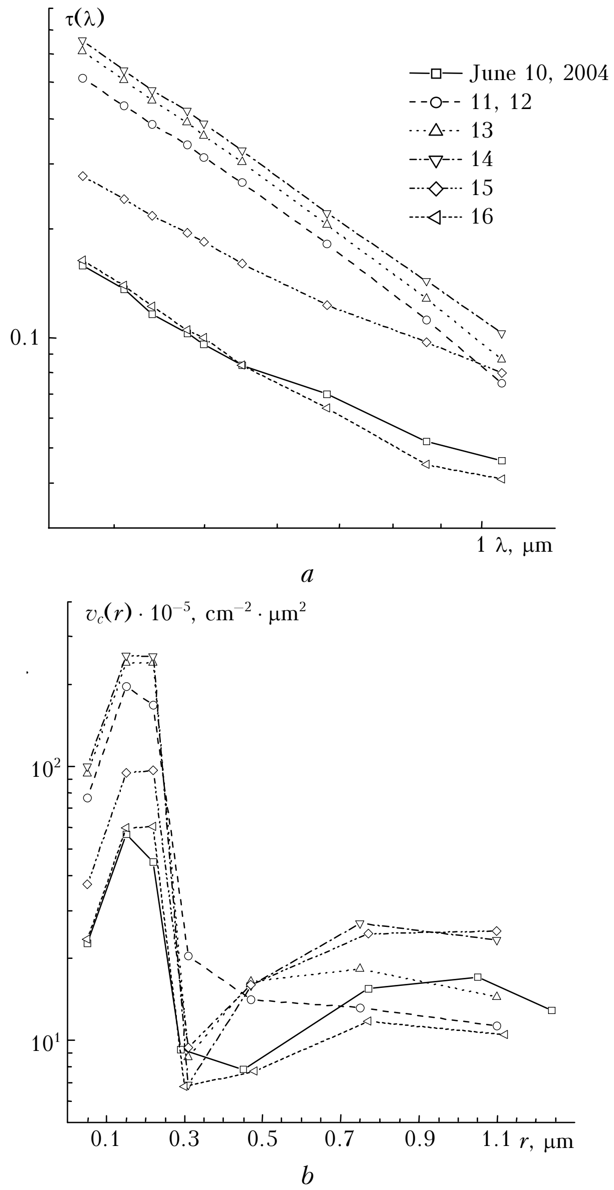

Fig. 7. Measured spectral dependencies of AOD (*a*) and retrieved particle volume distributions (*b*) under conditions of high atmospheric transparency (June 10, 2004), at the presence of smokes in the atmosphere (June 11–15) and after the change of air mass (June 16).

Spectral dependence $\tau(\lambda)$ in the initial period of smoke observation is convex in double logarithmic scale due to the decrease of relative contribution of large particles to AOD. According to daily average data, the measured values of AOD on August 21, 22, and 23, as well as the corresponding retrieved parameters of aerosol microstructure did not change essentially. The intensity of coming smoke significantly increased since August 24, the total volume of particles with $r < 0.4$ μm increased up to $40 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 on August 24 and reached $73 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 on August 26. Total volume of intermediately dispersed aerosol particles essentially increased in that period. The mean particle size increased to 0.17–0.18 μm on August 25 and 26. As the concentration of intermediately dispersed particles increased, the spectral dependence $\tau(\lambda)$ was transformed, which then approached to the linear function in the double logarithmic scale. The change of air mass on August 28 has led to significant increase of the atmospheric transparency. The total volume of particles with $r < 0.4$ μm decreased to $21 \cdot 10^5$ cm$^{-2} \cdot$ μm^3. However, the smoke event was quickly renewed, that indicated the closeness of the smoke source to the observation site. The measured values AOD in UV range increased on August 29–30 up to 0.5–0.6, the estimates of the total volume of particles with $r < 0.4$ μm were at the level of 35–40 $\cdot 10^5$ cm$^{-2} \cdot$ μm^3.

Another event is shown in Fig. 7. In this case smoke aerosol came to the atmosphere over Tomsk together with air mass changed at night from 10 to 11 of June. The measured values of AOD in the UV wavelength range increased with coming new air mass up to 0.55, the total volume of particles V_c ($r < 0.4$ μm) increased practically four-fold from $11 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 on June 10 to $40 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 on June 11. The inter-day variations of the concentration of finely dispersed aerosol in the period between 11 and 14 of June were not so essential at its high level. The maximum estimates of V_c ($r < 0.4$ μm) $5 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 were obtained on June 14.

The inter-day dynamics of intermediately dispersed aerosol is better pronounced (Fig. 7*b*), which is displayed as successive increase of V_c ($r < 0.4$ μm) from $10 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 on June 11 to $18 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 on June 14. Then, on June 15, the atmospheric transparency essentially increased (Fig. 7*a*). The V_c value ($r < 0.4$ μm) decreased from $50 \cdot 10^5$ cm$^{-2} \cdot$ μm^3 to $20 \cdot 10^5$ cm$^{-2} \cdot$ μm^3, however, the total volume of intermediately dispersed particles remained at the level reached on June 14. The change of air mass on June 16 practically reconstructed the situation observed before smokes came.

For conclusion, present some results of retrieval of the size spectrum of particles in the total data array obtained in summer periods 2003–2004, when the frequency of occurrence of smoke events was the highest (Fig. 8). The range of the measured AOD values (at λ = 0.48 μm) was divided into 5 levels: 0.03–0.059; 0.059–0.116; 0.116–0.228; 0.228–0.448;

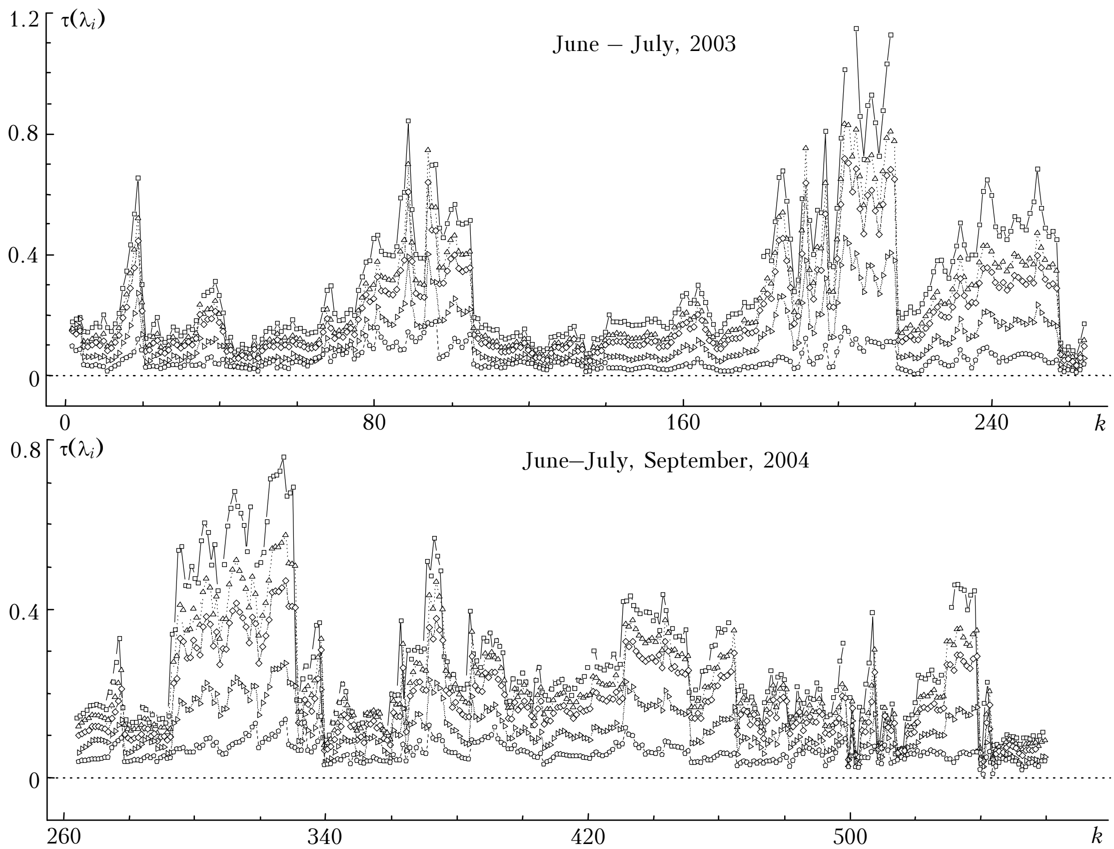

Fig. 8. Temporal variations of AOD measured in the wavelength range 0.37–4 μm in summer 2003 and 2004 (k is the number of realization).

0.448–0.88. The mean values of τ(λ) in the range 0.37–4 μm in each aforementioned interval were used as input data for solving the inverse problem (Fig. 9). The lower level was the background (minimum for the considered observation site) values AOD, the second and the third levels were sufficiently typical for summer (the third level contained a part of smoke events), the fourth and fifth levels reflected mainly smoke situations.

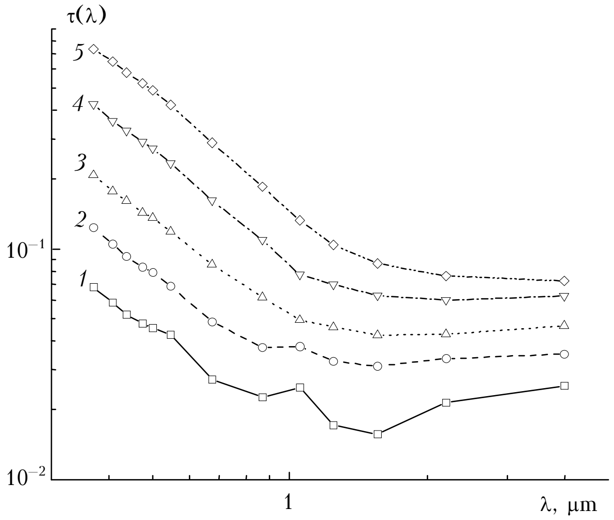

Fig. 9. Mean spectral dependences τ(λ) in the period of intensive effect of smokes in the atmosphere over Tomsk in the AOD intervals: 0.03–0.059; 0.059–0.116; 0.116–0.228; 0.228–0.448; 0.448–0.880 (curves *1*–*5*, respectively).

It is seen in Fig. 10 that as the atmospheric turbidity increases, aerosol filling of the intermediate size range increases.

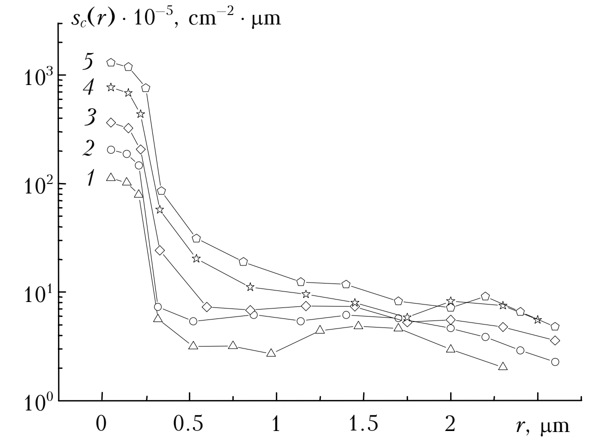

Fig. 10. Columnar particle geometric cross section size distributions retrieved from the optical data shown in Fig. 9.

The boundary of the first submicron mode is displaced to the right from 0.4 μm at the first and second levels of AOD up to 0.8 at the fourth level and exceeds 1 μm at the fifth level in the selected gradation of the atmospheric transparency.

Conclusions

1. The volume concentration of particles with $r > 0.5$ μm in the observed local smokes of forest and peatbog fires in the initial period was at the level of ignorable small values. The concentration of particles of this range in remotely transported smokes from forest fires in the majority of realizations exceeded the level of daily mean values in the atmosphere free of smoke.

2. The absence of diurnal behavior of large particles in the turbid atmosphere is, evidently, related with weakening of the processes of convection and turbulence (emission of particles) at a decrease of radiation heating of the underlying surface in the presence of smokes.

3. The inner atmospheric transformation of the finely dispersed smoke aerosol fraction (gradual growth and transition to the size range $r > 0.4$ μm) is one of the factors of replenishment of intermediately dispersed aerosol.

4. According to the averaged data for summer periods 2003–2004, the intensity of filling the intermediately dispersed particle size range increases with the increase of the concentration of finely dispersed smoke aerosol. The boundary of the first submicron mode is displaced to the right depending on the density of smoke and reaches 1 μm at a strong turbidity (τ(λ) > 0.5).

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References

1. V.S. Kozlov, M.V. Panchenko, and A.G. Tumakov, Atmos. Oceanic Opt. **6**, No. 10, 733–738 (1993).
2. V.S. Kozlov and M.V. Panchenko, Fizika Goreniya i Vzryva **32**, No. 5, 122–133 (1996).
3. R.F. Rakhimov, E.V. Makienko, M.V. Panchenko, V.S. Kozlov, and V.P. Shmargunov, Atmos. Oceanic Opt. **16**, No. 4, 308–317 (2003).
4. R.F. Rakhimov, V.S. Kozlov, E.V. Makienko, and V.P. Shmargunov, Atmos. Oceanic Opt. **15**, No. 4, 292–299 (2002).
5. A.A. Isakov, Atmos. Oceanic Opt. **12**, No. 1, 20–27 (1999).
6. A.A. Isakov, Atmos. Oceanic Opt. **16**, No. 10, 811–816 (2003).
7. G.I. Gorchakov, P.P. Anikin, A.A. Volokh, A.S. Emilenko, A.A. Isakov, V.M. Kopeikin, T.Ya. Ponomareva, E.G. Semutnikova, M.A. Sviridenkov, and K.A. Shukurov, Izv. Ros. Akad. Nauk. Ser. Fiz. Atmosf. i Okeana **40**, No. 3, 366–380 (2004).

8. V.S. Kozlov, M.V. Panchenko, V.V. Pol'kin, Yu.A. Pkhalagov, V.N. Uzhegov, N.N. Shchelkanov, and E.P. Yausheva, Atmos. Oceanic Opt. **12**, No. 5, 390–394 (1999).
9. E.V. Makienko, R.F. Rakhimov, S.M. Sakerin, D.M. Kabanov, and V.N. Uzhegov, in: *Proc. of the 5th Int. Conf. on Wildlife Fires: Initiation, Spread, Suppressing and Ecological Consequences* (Krasnoyarsk, 2003), 285–287 pp.
10. D.M. Kabanov and S.M. Sakerin, Atmos. Oceanic Opt. **10**, No. 8, 540–545 (1997).
11. S.M. Sakerin and D.M. Kabanov, J. Atmos. Sci. **59**, No. 3. Part 1, 484–500 (2002).
12. A.N. Tikhonov and V.Ya. Arsenin, *Methods for Solving Ill-Posed Inverse Problems* (Nauka, Moscow, 1974), 224 pp.
13. V.E. Zuev and I.E. Naats, *Inverse Problem of Lidar Sensing of the Atmosphere* (Springer–Verlag, 1983), 260 pp.