Some peculiarities in the dynamics of optical and microphysical characteristics of aerosol in smoke haze

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We analyze here measurement data on spectral dependence of the aerosol extinction coefficient within 0.44–12 μm spectral range, aerosol scattering coefficient at 0.52 μm wavelength, particle size distribution function within 0.4–10 μm, and mass concentration of soot in aerosol during a smoke haze observed in the West Siberia in October 1997. It is shown that the smoke haze results in an increase (by an order of magnitude or more) in the aerosol extinction and scattering coefficients in the visible range, as well as in the particle number density and soot content. Main changes of the parameters take place in the small-size aerosol fraction (sizes less than 1 μm) and, to a lesser degree, in the coarse aerosol fraction. It was found that the smoke haze smooths the peculiarities in the diurnal variation of the scattering coefficient and soot content characterizing the atmosphere free of smoke. Low relative content of soot in the smoke aerosol (1–4%) is indicative of a pyrolysis origin of the smoke haze particles formed during forest and peat fires. The smoke haze is assumed to contribute significantly to the planetary radiation budget.

1. Introduction

The study of peculiarities in the origin, development, and destruction of dense aerosol formations is an urgent problem in the field of climatology, physics, and optics of the atmosphere. In this aspect, such quite a unique and poorly studied aerosol formation as smoke haze is of a significant interest. Taking into account a great number of forest and peat fires on the Earth, there are certain reasons to suppose that smoke particles play a noticeable role in the formation of aerosol substance composition in the atmosphere and, therefore, in the radiation budget of the planet. To study the peculiarities of interaction of smoke haze with optical radiation, it seems to be important to carry out complex investigations by use of optical, microphysical, and meteorological measurements.

Such measurements were carried out in the region of Tomsk during the smoke haze event that occurred in October 1997 over a vast area of West Siberia. Measurements were performed of the spectral transmission of the atmosphere in the visible and infrared ranges on the near-ground path, the directional scattering coefficient at the angle of 45° for radiation at 0.52 μm wavelength, size distribution of the number density of aerosol particles, and the content of soot in the aerosol substance composition. The measurement campaign was supported by measurements of the meteorological parameters of the atmosphere as well.

2. Description of the experiment

Measurements of the directional scattering coefficient of dry matter of aerosol particles $\mu_0(45°)$ ($km^{-1}sr^{-1}$) and the mass concentration of soot M_s ($\mu g/m^3$) in the near-ground air layer were carried out round the clock (every hour) by means of an automated aerosol station[1] operated in the routine mode. The polarization nephelometer[2] equipped with the device for drying the air in the scattering volume was used for measuring $\mu_0(45°)$. Then the values of the scattering coefficients of dry matter of aerosol particles σ_0 (km^{-1}) were calculated from $\mu_0(45°)$ on the basis of known nephelometric method to determine the transparency that uses the fact that the relationship between the related parameters is close to a linear one. The mass concentration of soot was determined by means of a setup described in Ref. 3 from measurements of the diffuse attenuation of light by a layer of particles directly during the process of air pumping through and accumulation of particles on an aerosol filter, in a way similar to that discussed in Ref. 4. A single sampling cycle lasted 10 minutes. The values of the scattering coefficient were averaged over 600 readouts, and the value of the soot concentration was determined from two readouts: at the beginning and at the end of the aerosol sampling cycle. The measurement process is fully automated and controlled with a personal computer.

Measurements of the spectral transmission of the atmosphere were carried out on a 800-m long path with a fully automated dual-channel transmissometer instrumentation complex[5] in the 0.44 to 12 μm wavelength range. Measurements were performed round the clock every six hour during the initial period and then every two hours. To select the aerosol component from the total extinction of radiation in the IR wavelength range where the molecular absorption by atmospheric gases makes a significant contribution, we applied the statistical method for separating the components based on the linear multiple regression analysis.[6]

Measurements of the aerosol size distribution were carried out by means of an AZ-5 photoelectric counter of particles in 12 ranges of the particle diameters from 0.4 to

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10 μm. To improve the accuracy of measurements, the device was equipped with a CH3-63 frequency meter. Measurements were carried out selectively during daytime from 7 till 10 of October. The volume of air to be analyzed was one liter for fine particles (up to 1 μm in diameter) and three to five liters for the coarse particles.

The smoke haze considered in this paper was observed in the region of observations under the conditions of a wide and stable anticyclone formed over West Siberia. The weather in the Tomsk region in September and the first decade of October was stable, windless, warm, and dry that is quite rare for this season. Such weather conditions cause numerous forest fires and burning of peat (according to the satellite data, the most intense ones took place in Kazakhstan, Novosibirsk and Tomsk regions, and Altai).

The peak of the smoke haze development in Tomsk was observed in the first decade of October and was most pronounced on October 7 to 8, when the meteorological visual range in the city and its suburbs decreased down to 2 km. It is worth noting that the period of measurements was characterized by low relative humidity of air (mean value of 40%) and its weak dynamics.

3. Results of optical and microphysical measurements

Temporal behavior of the daily mean values of the aerosol scattering coefficient and the mass concentration of soot is shown in Fig. 1. The stages of the smoke haze appearance, condensation, and destruction with a significant amplitude of variations of σ_0 and soot content in the near-ground layer of air are well seen in these data. The stable trend of gradual accumulation of aerosol and soot was observed in the first decade of October until October 8. The scattering coefficient increased by eight times, on the average, and the soot content increased by four times during a comparatively short time interval. The change of air mass resulted in the fast destruction of the smoke haze in the region of observation after October 9.

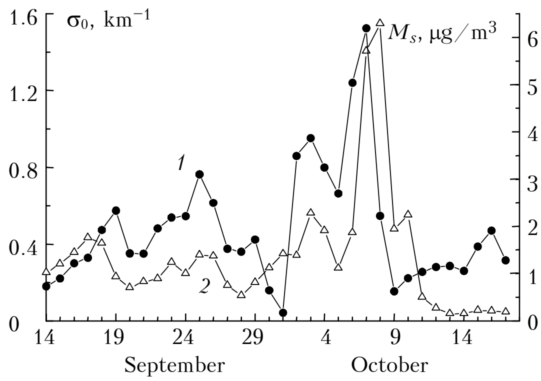

Fig. 1. Temporal behavior of the daily mean values of the mass concentration of soot (*1*) and aerosol scattering coefficient (*2*) during the period of formation and destruction of the smoke haze.

The mass concentration of the fine aerosol particles was estimated on the basis of the data on directional scattering coefficient (within the applicability limits of a one-parameter aerosol model[7]). The mass concentration (μg/m^3) was calculated for the particles with the density of 1.8 g/cm^3 (continental aerosol) by the following formula

$$M_a = 396 \cdot \sigma_0 = 3.56 \cdot 10^3 \cdot \mu_0(45°).$$

The quantitative estimates of M_a by this formula show that the aerosol mass concentration during the smoke haze peak on October 6–8 reached 385 μg/m^3, while its "background" value (in August) was 31.5 μg/m^3. The mean values of the soot content obtained from measurements were 3.9 and 0.9 μg/m^3, respectively. In turn, the maximum individual mass concentration values reached 840 μg/m^3 for aerosol (October 8, 12:00) and 9.7 μg/m^3 for soot (October 7, 14:00).

Thus, the results of measurements show that the mass concentration of aerosol and soot in smoke haze can exceed their background values by one order of magnitude and more. Let us note that the aerosol mass concentration obtained from the data of microstructure measurements by the particle counter was about 300 μg/m^3 that agrees with the aforementioned estimate of M_a from the data on the directional scattering coefficient.

The peculiarities in the dynamics of the aerosol extinction in different spectral regions during the event of smoke haze are illustrated by Figs. 2 and 3. Figure 2 shows the temporal behavior of the aerosol extinction coefficient $\alpha(\lambda)$ at several wavelengths since October 6 until October 10 (the break of curves is related to the malfunctioning of the instrumentation).

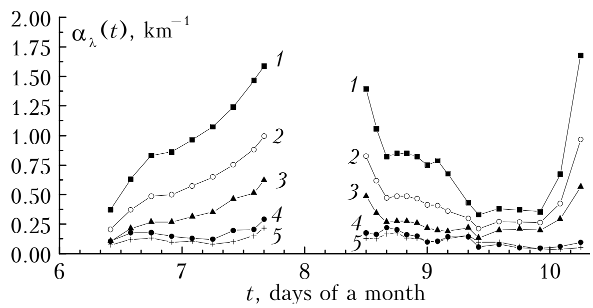

Fig. 2. Temporal behavior of the aerosol extinction coefficients at the wavelengths of 0.44 (curve *1*), 0.56 (*2*), 0.87 (*3*), 1.60 (*4*), and 10.6 μm (*5*).

It follows from Fig. 2 that the maximum variations of the coefficients $\alpha(\lambda)$ in smoke haze occurred in the visible wavelength range. The data shown in Fig. 3 are also evidence of this fact. It is seen that the coefficients $\alpha(\lambda)$ in the short-wave region rapidly increase after the onset of the smoke haze, while the extinction in the IR range practically does not change (moreover, even a small decrease of the extinction coefficients was observed). It is the evidence of the fact that the haze density at the initial stage increases due to the increase in the number density of submicron particles.

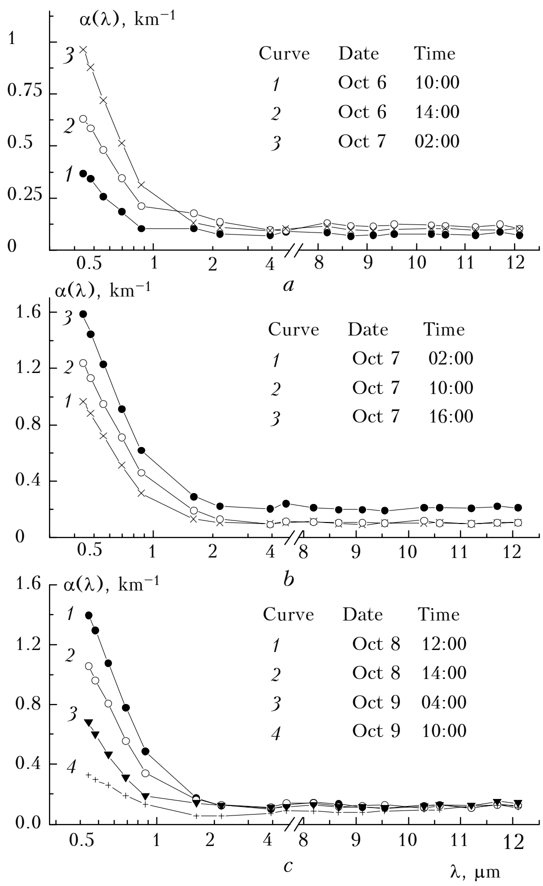

Fig. 3. Transformation of the spectral behavior of the aerosol extinction coefficients during the periods of appearance (*a*), development (*b*), and destruction (*c*) of the smoke haze.

However, a significant increase of the aerosol extinction has been observed as the haze developed (October 7, 16:00) not only in the visible, but in the IR range too (see Fig. 3*b*). The neutral spectral behavior of the aerosol extinction in the IR range allows us to suppose that it might be related to emissions from soil into the near-ground layer of the atmosphere during daytime. A significant decrease in $\alpha(\lambda)$ values, as the haze decays, occurs (see Fig. 3*c*) only in the short-wave range ($\lambda \le 1$ μm), while being practically absent in the long-wave range ($\lambda > 2$ μm). This means that the smoke haze formed is the quite fine dispersed system.

Figure 4 shows the particle size-distribution function during the period of maximum density of the smoke haze as estimated from the data of the photoelectric counter (curve *1*) in comparison with the atmospheric haze corresponding to the mean summer values of the particle number density in West Siberia in the lower 500-m layer (curve *2*). In general, the shape of the particle size-distribution of smoke corresponds to the mean distribution of the atmospheric aerosol and is characterized by the presence of two particle fractions: fine ($r < 1$ μm) and coarse ($r > 1$ μm). However, the decrease in the number density of particles of all sizes and, especially, of the fine particles is observed under the smoke haze conditions.

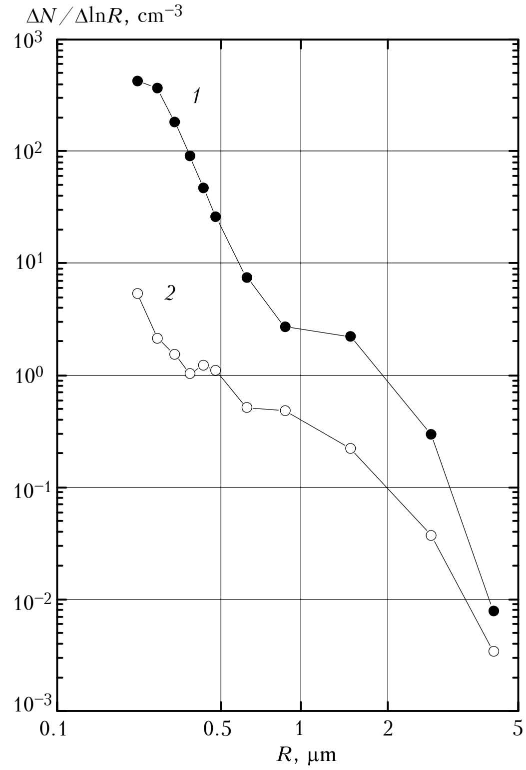

Fig. 4. Characteristic particle size distributions.

Dynamics of relative variations of the number density has some peculiarities in some particle radius ranges, at different stages of the smoke haze development, and in comparison with the background conditions. At the smoke haze destruction from the maximum smoke concentration to a low one, the change of the number density N_{max}/N_{min} was up to 50 times for fine particles (radius $r = 0.2$–0.4 μm) and up to 30 times for the coarse particles ($r = 1$–3 μm) (Fig. 5*a*). In turn, the maximum number density values during the haze can exceed the background characteristics (mean summer values in West Siberia in the lower 500-m layer) N_{max}/N_{back} by 175 times (0.2–0.4 μm) and by 10 times (1–3 μm) (Fig. 5*b*). Let us note that the comparison of the minimum haze number densities with the mean regional values N_{max}/N_{back} shows that the residue effect of the dissipated smoke haze leads to an enhanced number density, by up to 3.5 times, for fine fraction of aerosol particles (Fig. 5*c*). The decreased number density of coarse particles compared with the regional mean could be explained by almost windless weather and, hence, by a weak emission of the dust particles into the atmosphere.

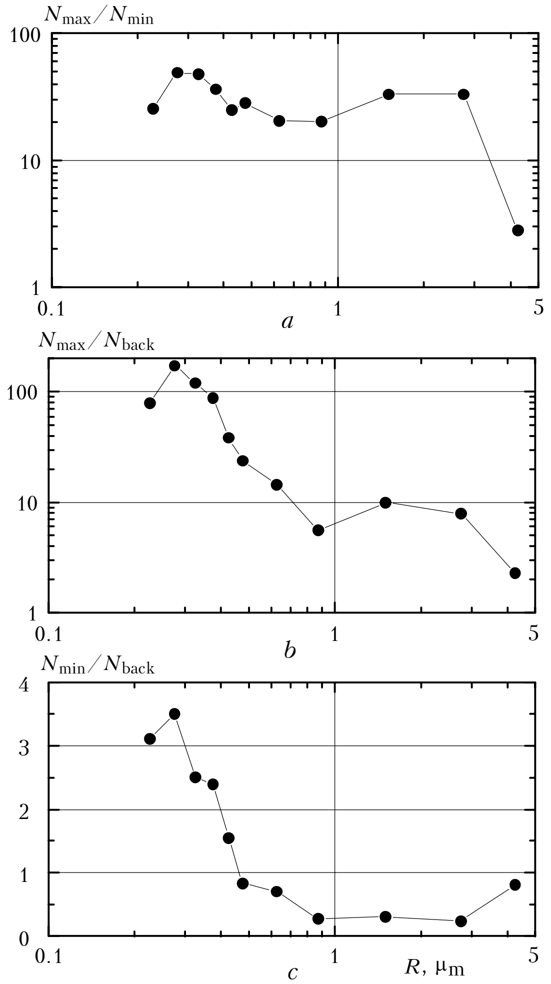

Fig. 5. Relative variations of the particle number density.

The data shown in Fig. 6 illustrate some peculiarities of the effect of smoke haze on the variations of the relative content of soot in the aerosol substance. Figure 6*a* shows the mean diurnal behavior of the relative soot content in aerosol P normalized to the daily mean value P_d (curve *1*) characteristic of the period since June till September 1997. It is characterized by quite a pronounced dynamics and by the presence of two maxima: one in the morning and the another in the evening–night hours. Situation changes dramatically under the smoke haze conditions (curve *2*). Diurnal behavior of the soot content becomes less contrast. Similar tendency is observed in the diurnal behavior of the aerosol scattering coefficient. The data obtained are evidence of the fact that formation of a dense smoke haze weakens the effect of the dynamical factors typical of the conditions of a smoke–free atmosphere, possibly, due to the anomalous turbidity of air.

Figure 6*b* presents the temporal behavior of daily mean values of the relative content of soot in aerosol (P, %) since September 11 till October 11. It is determined for every individual measurement as the ratio of the mass concentration of soot and aerosol. As analysis shows, the value of the relative soot content in particles is not high (from 0.5 to 5%) regardless a significant increase in the absolute content of aerosol and soot separately.

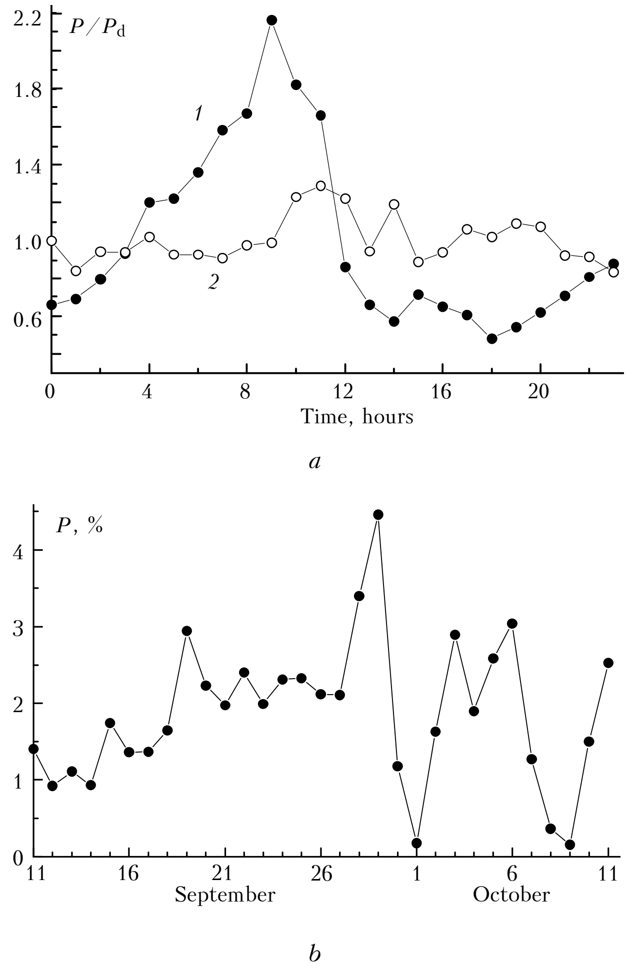

Fig. 6. Temporal behavior of the relative content of soot in aerosol: normalized diurnal behavior (*a*) (before the smoke haze (*1*), during the smoke haze (*2*)) and the dynamics of daily mean values (*b*).

In our opinion, a possible reason of low values of P, in %, in the smoke haze lies in the fact that, during the period of accumulation and the peak aerosol load, the principal portion of particles is characterized by high content of particles of the pyrolysis origin, i.e., from burning of the forest combustible matter. The results of our earlier investigations[8] of the influence of the conditions of burning on the soot content in the wood smokes under controlled conditions showed that the particles produced in pyrolysis (comparatively low temperature destruction of wood at 300–500°C without flame) contain little soot and, hence, are weakly absorbing. However, the soot content sharply increases as the as the pyrolysis changes for high temperature combustion with flame. One should suppose that the pyrolysis component of smoke gave the main contribution to the studied smoke haze appeared during the forest and peat fires.

4. Conclusion

Thus, the results of investigations show that intense forest fires under conditions of low activity of advective and convective processes in the near-ground air can lead to accumulation of the aerosol mass and thus cause the appearance of the anomalously high number density of aerosol with a significantly high content of fine particles.

The analysis made has allowed us to study some quantitative and qualitative peculiarities of the effect of smoke haze on the aerosol composition of the atmosphere and its optical properties:

– optical and microphysical parameters of aerosol in smoke haze can exceed the corresponding parameters of the smoke-free atmosphere by one order of magnitude and more;

– the smoke haze transformation is principally caused by the variability of the fine particle fraction (less than 1 μm in size) that has the highest amplitude of variations;

– dense smoke haze smooths the peculiarities of the diurnal variations of the main aerosol parameters characteristic of the smoke-free atmosphere;

– small relative content of soot in aerosol is most likely caused by the decisive contribution of the process of pyrolysis to the formation of aerosol in the smoke haze considered.

Acknowledgments

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