About the Rhythms of Variability of the Submicron Aerosol Characteristics in the Near-Ground Air Layer in West Siberia

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Introduction

The study of the role of atmospheric aerosol in climate change and radiative processes in the lower atmosphere requires knowing the peculiarities of the variability of the aerosol characteristics under the effect of geophysical factors of different temporal and spatial scale. Obtaining a detailed analysis of the long-term in situ measurements of the optical characteristics of near-ground aerosol is important in solving these problems.

In this paper, the year-to-year variability and the spectra of temporal variations of the submicron aerosol characteristics in the near-ground air layer are considered.

Instrumentation and Technique for Measurements and Calculations

Regular measurements of the directed scattering coefficient of the dry matter of aerosol particles at the wavelength of 0.52 μm (let us call it "aerosol"), the mass concentration of absorbing substance (let us call it "inorganic carbon" or "black carbon" [BC]) and the parameter of condensation activity of particles are carried out in the near-ground air layer at the aerosol monitoring station of IAO SB RAS located at the eastern outskirts of the city of Tomsk since 1998.

An aerosol sample was brought to the optical cells of the devices through the collectors installed at the height of 9 m. The available experimental setup is capable of measuring the characteristics of submicron aerosol particles. The air sample was preliminarily heated to 30°C to decrease relative humidity and enable the characteristics of practically dry particles to be measured. Measurements of the directed scattering coefficient at the 45° angle were carried out using the FAN nephelometer that has a sensitivity of 0.001 $km^{-1}ster^{-1}$. The device was calibrated using measurements of different pure gases of which the scattering coefficients are known. The data on the directed scattering coefficients can be used to estimate the total scattering coefficient and the mass concentration of submicron aerosol (Gorchakov and Sviridenkov 1981). The mass concentration of BC in the composition of aerosol particles was measured using an aethalometer analogous to that designed by Hansen et al. (1984). The aethalometer was capable of measuring the mass concentration of BC from 0.1 to 110 $\mu g/m^3$ with a sensitivity of about 0.1 $\mu g/m^3$ at the pumping of about 20 l of air. The aethalometer was calibrated by comparing the results of simultaneous optical and gravimetric measurements of BC particles of 50 to 200 nm generated during pyrolysis of buthanol vapor in the nitrogen atmosphere at temperature of about 1150°C (Baklanov et al. 1998). Measurements of the directed scattering coefficient and the mass concentration of BC at the aerosol monitoring station were carried out every hour using in automated regime (Kozlov et al. 1997). Measurements of the parameter of condensation activity characterizing the hygroscopic growth of the aerosol scattering coefficient were carried out once a day using artificial humidification of aerosol up to a relative humidity of 95% (Panchenko et al. 1999).

The results of the measurements were used for analysis of the peculiarities of the temporal dynamics of the aerosol characteristics and subsequent study of the principal rhythms of their variability. To do this, annual, seasonal, and total arrays of the data on the aerosol and BC characteristics were formed. The values of the parameters obtained under conditions of the effect of forest fires were removed from the data arrays. The data were statistically processed, and the histograms of the distribution of the aerosol characteristics were considered. The periodograms (Fourier spectra of the discrete data set) were calculated for all data arrays using spectral analysis of the series of observations (Bendat and Piersol 1980). The high-frequency (up to 1 month) and low-frequency (from 1 to 6 months) variations were considered. The significance of the amplitudes of oscillations of different periods was estimated upon the 5% level. The duration of the measurements is 6 years, taken once an hour; hence, the statistically provided frequency range is from 2 hours to 6 months. According to the classification of meteorological parameters (Monin 1969) such a range corresponds to the synoptic (from 10 hours to a few days) and the global (from weeks to months) scales of oscillations. To assess the peculiarities of year-to-year variability, we have also analyzed the dynamics of annual and seasonal mean values of all measured aerosol characteristics.

Results

Statistical processing of seasonal and annual data arrays has shown that, as a rule, the histograms of distributions of the aerosol characteristics are single-modal and are close to lognormal distributions. Figures 1–4 show the amplitude spectra of oscillations of the submicron aerosol and BC for seasonal (Figures 1 and 2), annual (Figure 3), and total (Figure 4) data arrays. Analysis of the periodograms makes it possible to determine the principal frequencies of oscillations in the short- and long-period ranges, to consider the peculiarities of seasonal variability of the spectra, and to estimate the differences in the spectra of temporal oscillations of aerosol and BC characteristics. The important peculiarity of the considered amplitude spectra is that, in spite of some frequency shifts and amplitude variations, the sets of the obtained mean values of the 5%-significant periods of oscillations of the characteristics of submicron aerosol and inorganic carbon are close to each other.

Seasonal data arrays provide estimation of only high-frequency periods of oscillations up to 10–15 days. It follows from the analysis of the spectra of aerosol and BC characteristics in different seasons that oscillations with the mean periods of 0.5, 1, 2, 3.5, 5.5, 7, 8.5, and 10-11 days are observed most often. Typical summer and winter spectra of the aerosol and BC characteristics are shown in Figures 1 and 2. The obtained frequencies are related with the principal daily and synoptic cycles of oscillations of the considered parameters. The periods of 0.5, 1, and 2 days reflect the peculiarities of the processes of formation and the sink of aerosol and BC in the near-ground air layer. The range greater than 3.5 days is, obviously, related with the processes of synoptic scale, such as the change of air masses.

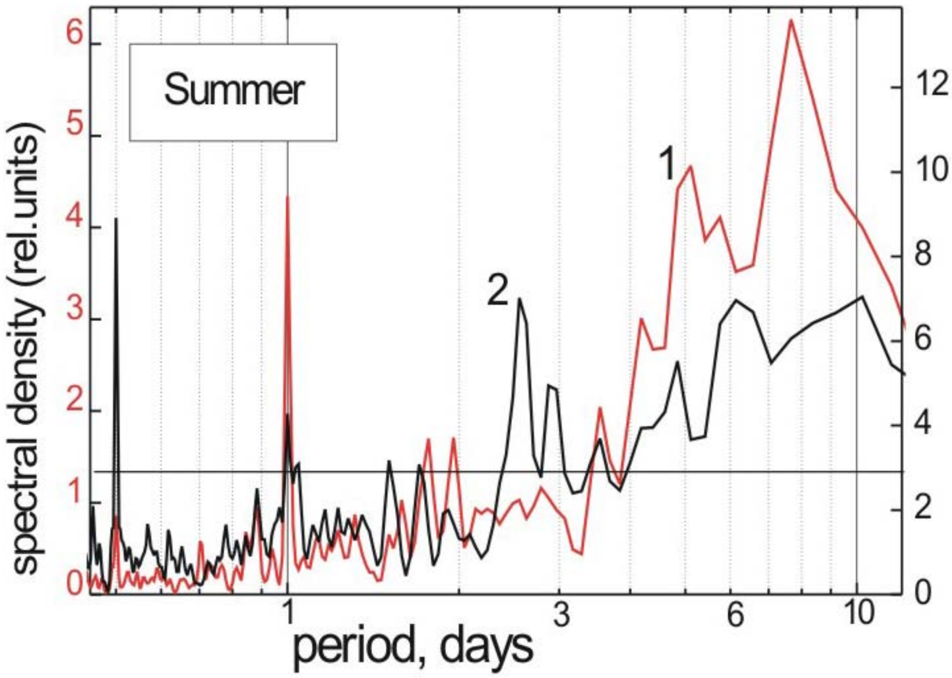

Figure 1. Periodograms of variations of the aerosol (1) and BC (2) mass concentrations in summer 2002 (5-point averaging using the Hamming window).

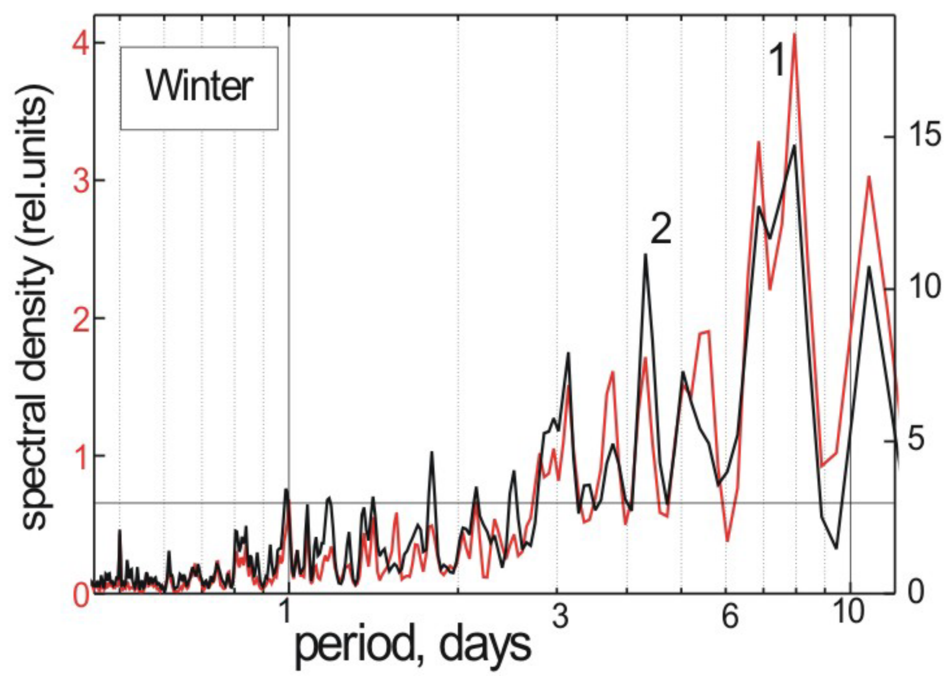

Figure 2. Periodograms of variations of the aerosol (1) and BC (2) mass concentrations in winter 2002 (5-point averaging using the Hamming window).

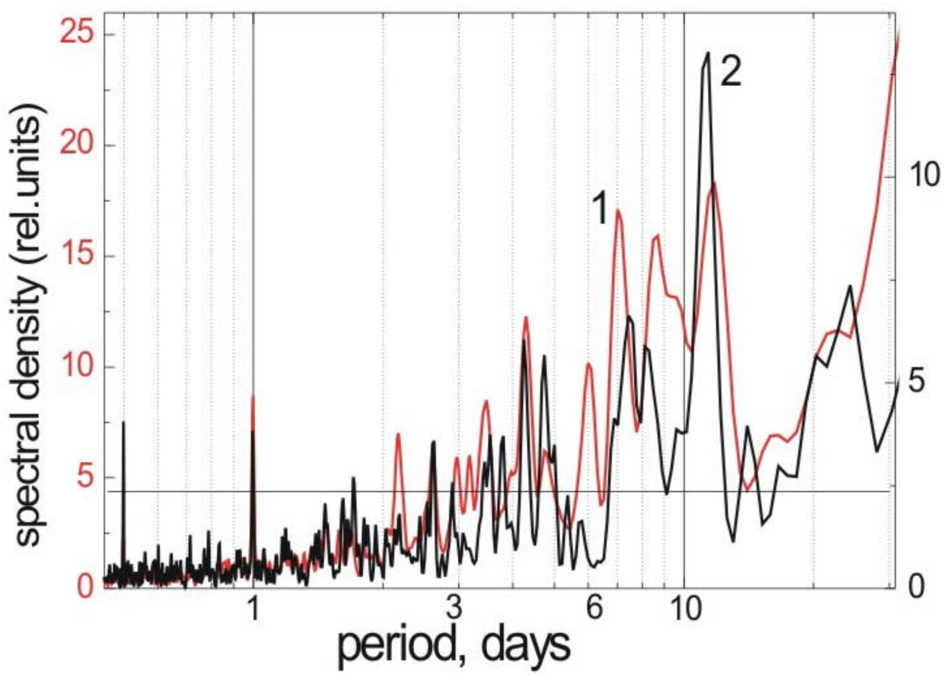

Figure 3. Amplitude spectra of temporal variations of the aerosol (1) and BC (2) mass concentrations obtained during 1 year (2000, 5-point averaging using the Hamming window).

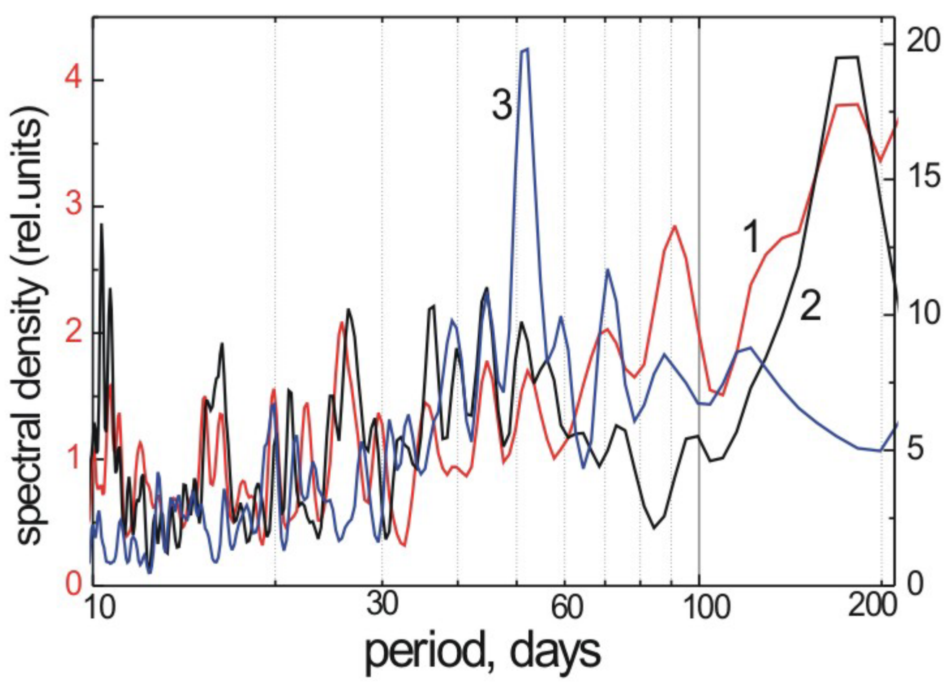

Figure 4. Amplitude spectra of temporal variations of the aerosol (1) and BC (2) mass concentrations and air temperature (3) assessed from the data obtained in 1998–2003 (5-point averaging using the Hamming window).

The periods of 3.5 and 5.5 days are close to that characteristic of the lifetime of the main synoptic formations, cyclones, and anticyclones. The greater periods are related with some alternation of the synoptic objects and, obviously, are formed as a superposition of the frequencies of individual oscillations (Belan et al. 1994).

Seasonal differences in the amplitude spectra of both aerosol characteristics are observed as the decrease of the amplitude of an individual oscillation up to its disappearance, as well as possible transformation of the spectrum structure due to the shift or splitting of frequencies near the main periods. This fact can be explained by taking into account that the intensity of synoptic circulation changes during a year. Summer is characterized by the most stable shape of the amplitude spectra and practically all periods of oscillations (Figure 1). The signs of the shift and splitting of the spectra are observed when passing to cold seasons, fall and winter (Figure 2). The greatest instability of frequencies of the amplitude spectrum is characteristic of spring and fall. The summer spectra are the most stable from year to year (the repetition rate of the principal frequencies in different years is from 65 to 100%).

Seasonal variations of daily oscillations (fewer than 3.5 days) lie in the decrease of the amplitude or the absence of oscillation. The 1, 2, and 3.5-day cycles of both aerosol characteristics are well pronounced in summer. However, when passing to the cold season, the amplitudes of, and 1- and 2-day oscillations significantly decrease, and the 3.5-day cycle becomes less pronounced. The amplitude of the 1-day period of the aerosol oscillation decreases and becomes comparable with the 5% level of significance. The 1-day period of the BC oscillation is weakly pronounced in spring. The period of 0.5 days is absent in the spectra of both characteristics in winter.

The principal difference in the amplitude spectra of aerosol and BC is that the 0.5-day period of the BC is observed in summer, while that of the aerosol is absent. Another difference in the dynamics of the aerosol and BC is that the aerosol spectra are more stable from year to year. The aforementioned peculiarities of the temporal variability are evidence of some “independence” of aerosol and absorbing substance (BC) life in the atmosphere. It confirms again that the hypothesis of additive mixture of absorbing substance and all submicron aerosol particles is only the first approximation.

Typical annual amplitude spectra of aerosol and inorganic carbon are shown in Figure 3. The frequencies up to 30 days are statistically provided in these spectra. As the figure shows, the annual spectra of aerosol and BC are also close to each other. Practically all periods revealed in seasonal ensembles are well pronounced in the annual data arrays. It is evidence that tendencies characteristic of the warm season are predominant at statistical mixing of the amplitude spectra. The set of frequencies of the annual data arrays is extended due to the following long-period oscillations: 15, 17, 20-21, 24-25, and 29-30 days. The revealed frequencies add the series of variations of the synoptic scale, and their amplitudes cogently exceed the level of significance. However, analogous to seasonal data arrays, amplitude spectra of aerosol are more stable from year to year than are those of BC.

Periodograms of the aerosol and BC contents in the total data array (1998–2003) are shown in Figure 4. The structure of the amplitude spectra in the high-frequency range (less than 1 month) is in good agreement with the data of separate years, shown in Figure 3. The low-frequency range of oscillations of aerosol and BC shows the following additional periods: 36, 45, 50-55, 70, 90, 130 and 180 days. The amplitude spectrum of air temperature oscillations in the near-ground layer obtained from the total 6-

year data array is also shown in Figure 4. Comparison of the periodograms of the aerosol characteristics and temperature shows their satisfactory agreement in a wide frequency range including low- and high-frequency oscillations. The appearance of the close periods of oscillations of the aerosol characteristics and air temperature is evidence of their correlation and coordinated transformation regulated by the processes of synoptic scale in the atmosphere.

The aerosol periodograms were also compared with the aerosol amplitude spectra obtained in the Moscow region in 1991–2002 (Sviridenkov et al. 2003). Comparison of the periodograms in the high-frequency (fewer than 30 days) and low-frequency (from 90 to 180 days) ranges shows their good agreement. The revealed similarity of the principal periods of aerosol oscillations shows that the processes determining formation of the rhythms of variability of the aerosol characteristics in two sites situated approximately 3000 km apart have the same origin and are characterized by the global spatial scale.

Year-to-Year Variability of the Measured Parameters

The 6-year-long series of measurements makes it possible to follow the year-to-year variability of the contents of aerosol and BC and the parameter of aerosol condensation activity.

The annual mean values of the parameter of condensation activity are shown in Figure 5. Its seasonal mean values in different years are also depicted here. As shown, the tendency of decrease is characteristic of not only the annual mean values but of each season separately. The estimates of the values of the directed scattering coefficient of the dry matter of aerosol particles and the mass concentration of BC (Figure 6) show there is no trend of these parameters. Therefore, one can conclude that the observed decrease of the parameter of aerosol condensation activity can be caused by the change of the qualitative composition of aerosol particles.

Conclusion

Analysis of the amplitude spectra of temporal variations of the contents of submicron aerosol and inorganic carbon in 1998–2003 is carried out. Principal periods of oscillations in high- and low-frequency ranges from 0.5 days to 6 months are obtained. It is shown that the amplitude spectra of 5%-significant frequencies of oscillations of aerosol and BC are close to each other and characterized by year-to-year stability. As a rule, the aerosol spectra are more stable than the BC spectra. When passing from the warm season to cold, the amplitudes of oscillations of the frequency of fewer than 3.5 days decrease, and year-to-year stability of the periodograms is destroyed. The main difference in the spectra of aerosol and BC lies in the fact that the 0.5-day period of BC oscillation is observed in summer, while that of aerosol is absent. It is evidence of the difference of the temporal dynamics of aerosol and BC, i.e., the hypothesis of the presence of absorbing substance as a passive admixture in the composition of particles is only an approximation. Good agreement of the aerosol spectra with that obtained by other researchers at a remote site in the same latitude zone shows the similar origin of the principal circulation processes in the atmosphere and their global spatial scale.

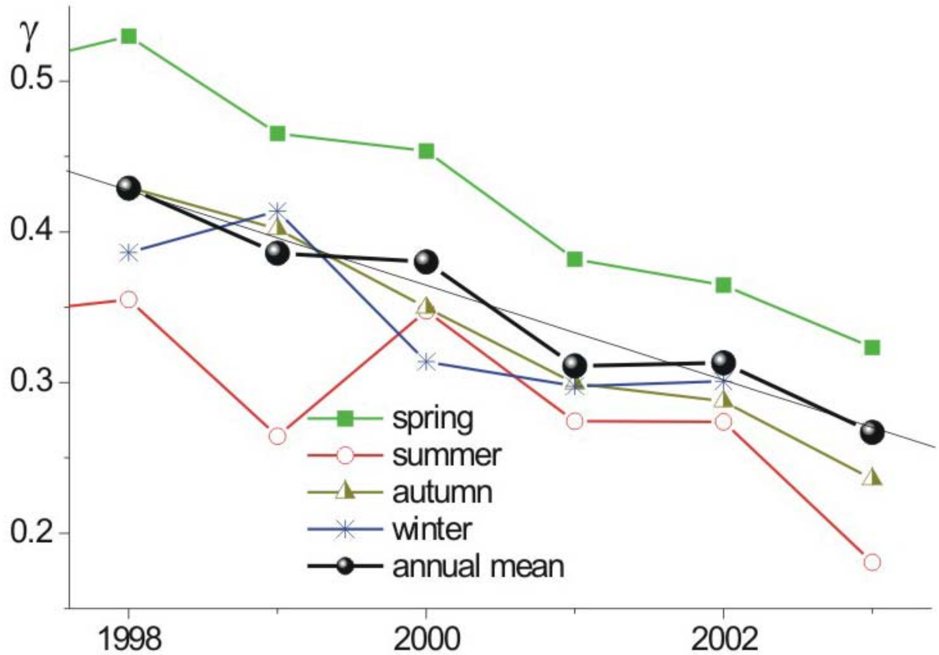

Figure 5. Year-to-year variability of the annual mean (1) and seasonal mean (2–5) values of the parameter of aerosol condensation activity in 1998–2003.

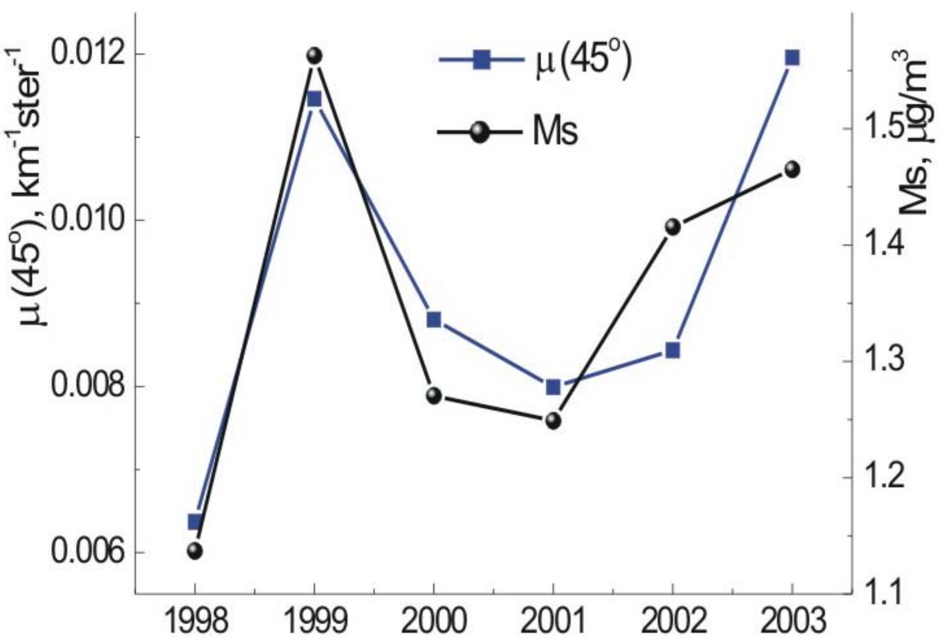

Figure 6. Year-to-year variability of the annual mean values of the aerosol scattering coefficient (1) and BC mass concentration (2) in 1998–2003.

Analysis of year-to-year variability of different aerosol parameters shows the well pronounced trend of the decrease of the parameter of aerosol condensation activity. Such a trend of other parameters is not observed, which allows us to conclude the change of the qualitative composition of submicron aerosol is gradual.

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