Time content variations of submicron aerosol and soot in the near-ground layer of the West Siberia atmosphere

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Based on the regular hourly measurements, we considered the specific features of interannual and seasonal variations of mass concentrations of the dry base substance of submicron aerosol and soot, as well as relative content of soot in aerosol particles in near-ground air layer for 1997–2006. We analyzed the arrays of annually, seasonally, and monthly mean values of aerosol parameters, as well as their root-mean-square deviations and coefficients of variation. For data subarrays, obtained under conditions of the atmosphere free of forest fire smoke, we studied the specific features of annual behavior, and investigated the tendencies in variations and trends of interannual variations of annually and seasonally mean values of aerosol characteristics.

Introduction

The important role of the atmospheric aerosol in atmospheric radiation processes determines the necessity of *in situ* study of time variation peculiarities of aerosol characteristics. Soot (Black Carbon (BC), crystalline carbon) is the main absorbing component in the atmospheric aerosol composition, making a considerable contribution to nonselective aerosol absorption in the shortwave spectral range. For understanding of the aerosol role in climate changes, of especial importance is the analysis of time series of multiyear measurements of atmospheric aerosol parameters in different geographic regions. The number of such experimental studies is small.

Among the works, performed recently and devoted to the study of long-term variations of aerosol content in near-ground layer, Refs. 1–3 can be underlined. For instance, based on the analysis of 12-year measurement series of submicron aerosol mass concentration in Moscow region, Gorchakov et al.[1] studied the interannual and seasonal aerosol variations and revealed a negative interannual trend in period between 1991 and 2002 with relative rate of about 2.6% per year.

The data of 4-year regular measurements of aerosol scattering and absorption coefficients in the visible spectral range at four ground-based stations in USA are discussed in Ref. 2; the authors, in particular, showed that the mean values of absorption and scattering coefficients experienced strong spatial variations, changing in transition from "background" aerosol station (Alaska) to the station with anthropogenic load (Bondwill, Illinois) by factors of 10 and 5, respectively. They also indicated that the mean annual behavior of the optical characteristics at the background station was characterized by maximum in winter and minimum in summer. Analogous features of annual behavior of the scattering and absorption coefficients were also reported by Man and Shin (Ref. 3), who considered the results of 1.5-year cycle of their measurements on Hong Kong island.

The goal of this paper is to analyze the interannual and seasonal variations of submicron near-ground aerosol in West Siberia, based on the regular measurements in 1997–2006.

Instrumentation

Since 1996, at the aerosol station of IAO SB RAS round-day measurements of the coefficient of directed scattering of dry base substance of submicron aerosol at the angle 45° at wavelength 0.52 μm are performed,[4] as well as BC mass concentration in the near-ground atmospheric layer. The current data are available in Internet (http://aerosol1.iao.ru). The measurements were conducted using the nephelometer of the type FAN with sensitivity up to 0.001 $km^{-1} \cdot sr^{-1}$. Data on the coefficient of directed scattering can be used for estimation of the total scattering coefficient and mass concentration of the submicron aerosol.[5] The BC mass concentration in the aerosol particle composition was measured by aethalometer using the method analogous to described in Ref. 6. The aethalometer allowed measurements of the BC mass concentration in the range 0.1–110 $\mu g/m^3$ with a sensitivity of about 0.1 $\mu g/m^3$ when pumping about 20 l of air through the device. The relative BC content P in the dry base substance was determined as the ratio of BC and aerosol mass concentrations with a mean error of about 20%. (From here on, P is given in fractions of unity or percent). The measurement results were used to compile the arrays of annually, seasonally, monthly, and daily mean values of aerosol parameters, as well as their root-mean-square deviations and coefficients of variation. Features of time dynamics of aerosol characteristics

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have been analyzed. In West Siberia, the forest fires, ignitions of peatbogs, and vegetation fires are frequently observed in warm period of the year. Therefore, to study the influence of fires on aerosol composition of the atmosphere, subarrays of data were additionally compiled with exclusion of fire episodes. Such an approach has made it possible to consider separately the conditions of the smoke-free (background) atmosphere.

Interannual variations of aerosol characteristics

Trends of interannual variations of annually and seasonally mean aerosol characteristics were calculated, based on the linear approximation of the time series for 10-year measurement period, by analogy with Ref. 7. In this regard, the obtained estimates of interannual variations of the aerosol and BC mass concentration, as well as variations of relative soot content in particles, reflect the features of a relatively short-period process on the considered timescale. We determined the relative rate of the trend (%), equal to the ratio of the rate of change of annually mean values to the mean value of aerosol characteristic for the entire measurement period. Analogous trend estimates were also obtained for seasonally mean values. The statistical significance of the trends was estimated using the Student test by analogy with Ref. 8.

The consideration of the whole data arrays for 1997–2006 has shown that the observed hourly values of the mass concentrations of aerosol M_a and crystalline carbon M_{BC}, as well as relative soot content P varied in the ranges 3–570 μg/m^3, 0.1–25 μg/m^3, and 1–20%. For monthly mean data, the corresponding variability ranges narrow to 12–95 μg/m^3, 0.7–3 μg/m^3, and 3–14%. The annually mean aerosol parameters mainly vary in the range 18–23 μg/m^3 for aerosol concentration, in the range 1.7–2 μg/m^3 for M_{BC}, and in the range 8–13% for relative BC content. The time scans of the annually and seasonally mean aerosol characteristics in 1997–2006 for arrays, free of the fire influence, are shown in Fig. 1, where in addition, the trends of annually mean values (statistically significant with 95% confidence probability) are illustrated by straight lines for the 10-year measurement period.

It is seen that the interannual M_a, M_{BC}, and P variation is characterized on the whole by nonlinear dependences, having local extremes, which illustrate a comparatively "fine" structure of interannual concentration variations. For instance, over the period 1997–2002 a tendency is observed of decrease of annually mean (and seasonally-mean winter) values of analyzed aerosol parameters, and from 2003, a tendency appears toward increase of aerosol and soot concentrations. Moreover, the estimates show that for the full 10-year period, the negative trends of variations of annually mean BC concentrations (–0.6% per year) and relative BC content in the particles (–2% per year) remain. However, the positive trend of aerosol concentration (0.4% per year) appears. One of the reasons for that is likely the gradual year-by-year decrease of anthropogenic load in the heating period of the year. Quite significant negative trend of decrease of annually mean P values indicates, in turn, the qualitative transformation of the composition of submicron aerosol for the 10-year period of measurements, which has led to decrease of absorptivity of aerosol particles, on average for P value from 10.5 to 8.5%.

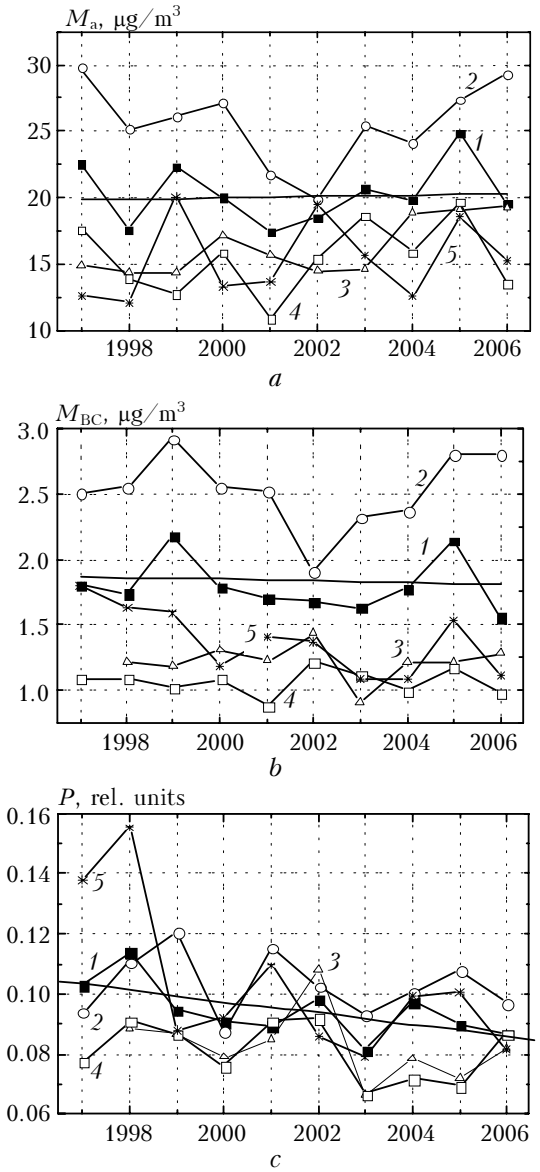

Fig. 1. Time behaviors of annually (curve *1*) and seasonally (winter- (curve *2*), spring- (curve *3*), summer- (curve *4*), and fall- (curve *5*)) mean mass concentrations of aerosol M_a (*a*), soot M_{BC} (*b*), and relative soot content P (*c*).

The time scans of seasonally mean aerosol characteristics exhibit a seasonal dependence of the trend magnitude. The largest trends correspond to spring and fall. For instance, the aerosol year mass

concentration increased by 3% in spring and by 2% in fall. The BC year mass concentration decreased by 0.2% in spring and by 3% in fall, while P decreased by 2% in spring and by 4% in fall.

The magnitudes of relative trends in winter and summer periods of the year for each aerosol characteristic are less than their values in the intermediate seasons; however, they are also statistically significant with the 95% confidence probability. On the whole, of note is insignificant winter negative trend of aerosol content, positive BC trend, and negative P trend (0.1% per year). This is a consequence of the tendencies of M_a and M_{BC} decrease, most pronounced in the winter period, up to 2002 and their subsequent increase from 2003. The revealed differences in tendencies of variations of seasonally mean M_a and M_{BC} values serve an indication of a certain independence of participation of aerosol and BC in the processes of particle formation and sink. This probably suggests that the consideration of the absorber as a passive admixture in the particle composition is only some approximation.

Figure 2 presents the time behaviors of annually mean variation coefficients V (V is the ratio of root-mean-square deviation to the mean value) of aerosol and soot concentrations and relative soot content, obtained by averaging their daily mean values. Each of the points characterizes the intraannual variability of aerosol parameters.

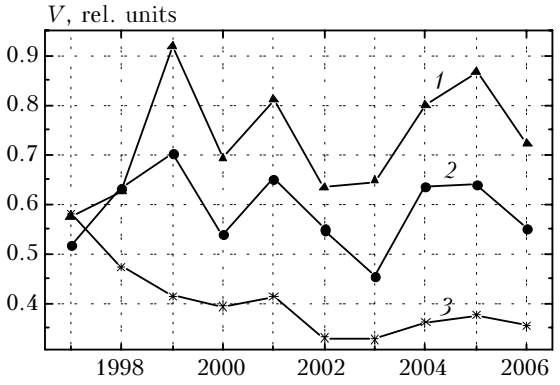

Fig. 2. Time behaviors of annually mean variation coefficients V_a (curve *1*), V_{BC} (curve *2*), and V_P (curve *3*).

As is seen, the variation coefficients of annually mean concentrations of aerosol (V_a) and BC (V_{BC}) change within quite wide limits, from 0.5 to 0.9, peaking in 1999 and 2005. Of note is the considerable decrease of annually mean variation coefficient of relative soot content V_P from 0.6 to 0.35 for the 10-year period (curve *3*).

Seasonal variations of aerosol and BC concentrations

Analysis of monthly and daily mean data arrays showed the presence of stable tendencies of seasonal variations of aerosol characteristics. This conclusion qualitatively agrees with results of the studies of aerosol time variations in different geographic regions for time series longer than a year.[1–3] Figure 3 presents 10-year average annual behaviors of the considered aerosol characteristics.

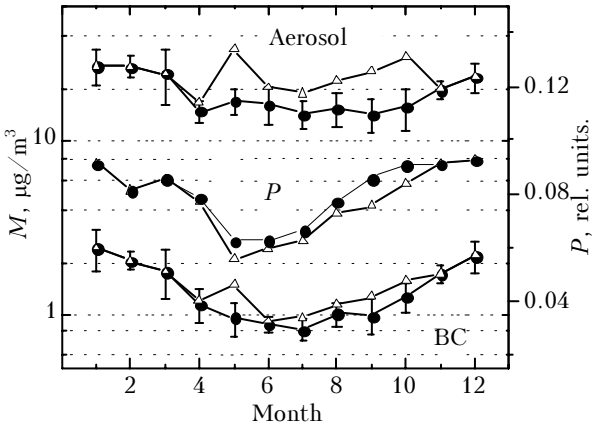

Fig. 3. Annual behavior of concentrations of M_a, M_{BC}, and P (1997–2006): taking into account fires (open triangles) and background aerosol (closed circles).

The results of the total data array averaging are denoted by triangles. The curves corresponding to data array free of the fire influence are denoted by solid circles (background aerosol). Error bars on the curves of aerosol and BC concentration indicate the root-mean-square deviations of monthly mean values over the 10 years of the measurements. In the smoke-free atmosphere the annual behaviors of aerosol parameters are characterized by winter maximum and summer minimum. The shape of the annual behaviors of aerosol characteristics possesses the interannual stability. The 10-year average monthly mean concentrations of aerosol and BC in the background atmosphere vary in ranges 14–28 μg/m^3 and 0.8–3 μg/m^3, respectively. The interseasonal concentrations vary, on the average, 2 times for aerosol and 3 times for BC. The 10-year-averaged P values for background aerosol vary from 0.06 to 0.1 (see Fig. 3).

The appearance of P annual behavior is caused by the fact that the seasonal dynamics of BC is more pronounced than the variations of aerosol concentration, which is determined by the seasonal differences of the intensity of the main processes responsible for emission of submicron aerosol and BC. Actually, when going to the warm period of the year, the processes of the photochemical generation of aerosol particles substantially intensifies. At the same time, the intensity of BC generation, upon completion of the heating season, substantially decays. These seasonal differences of directionality of the processes of aerosol emission and BC sink favor the development of the summer minimum in annual behavior of relative soot content.

Figure 4 illustrates the annual behavior of monthly mean V_a, V_{BC}, and V_p in 2002, obtained by averaging their daily mean values.

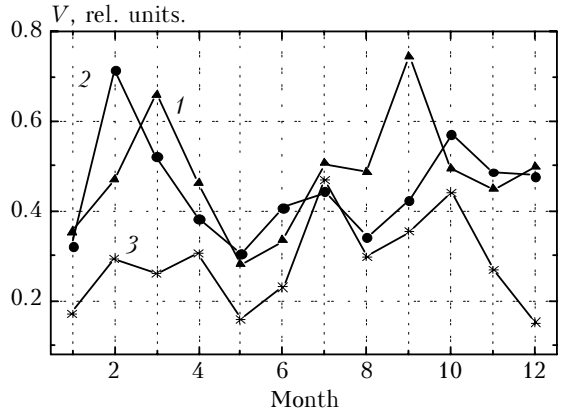

Fig. 4. Annual behavior of monthly mean variation coefficients V_a (curve *1*), V_{BC} (curve *2*), and V_P (curve *3*) in 2002.

For the monthly mean values, the variability is mainly determined by the synoptic-scale processes. The variation coefficients vary between quite wide limits, from 0.2 to 0.8. In the annual behavior, the largest variation coefficients of the aerosol and soot mass concentrations are observed in spring and fall-winter periods. The analogous tendency is also traced for monthly mean variation coefficients, averaged over 10 years of measurements.

Conclusion

We revealed the tendencies of decrease of annually mean and winter seasonally mean mass concentrations of aerosol, soot, and relative soot content in the particles in 1997–2002 and their increase from 2003 to 2006. Under these conditions, throughout the period, the positive aerosol trend and negative trends of variations of BC and relative BC content in the particles (0.4, –0.6, and –2% per year respectively) are observed. The trend magnitudes depend on the season and are maximal in the spring and fall periods. Therefore, the trends of annually mean mass concentrations of aerosol and BC over 10-year period were determined substantially by their dynamics in the spring and fall seasons.

The difference in tendencies of interannual variations of seasonally mean concentrations of aerosol and BC serves somewhat indication of a certain independence of participation of aerosol and soot in the processes of particle formation and sink.

The substantial decrease of annually mean values of P and its variation coefficient indicates that in period 1997–2006, the transformation of the qualitative composition of submicron aerosol and decrease in its absorptivity took place.

It is found that the annual behaviors of background aerosol characteristics are characterized by winter maximum and summer minimum. It is shown that this annual behavior of aerosol characteristics possesses the interannual stability.

Acknowledgements

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