Long-period variations of optical and microphysical parameters of the near-surface aerosol

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Using the data of measurements in 2001–2004 at the Zvenigorod Station of the Institute of Atmospheric Physics, variations of three basic characteristics of the near-surface aerosol are analyzed: the mass concentration M, the Angström exponent α, and the parameter of the condensation activity χ. Based on the data collected in the previous decade, it is shown that the decrease in the annual mean value of M, lasting for about a decade in Central Russia, has stopped in the beginning of this century, and its gradual increase for the past three years is observed. This is caused by a decrease observed in the frequency of occurrence of the cases with very low M values and an increase in the frequency of occurrence of the cases with very high, up to 250 $\mu g/m^3$, M values. Power spectra of aerosol parameters are analyzed. In addition to the well-known variation periods of ~ 5, 7, 10, and 45 days, the periods of about 14, 20, and 90 days have been revealed. The wavelet analysis performed has shown that variations group as time-limited series, and the periods and the amplitudes of the oscillations within the series do vary. It is shown that the long-period variations may be predominantly of the concentration origin when there is no relation between M and the Angström exponent during long time intervals, or may be due to changes in the mean particle size. Time intervals up to several months long have been revealed, which are characterized by high anti-correlation between M and χ (the absolute value of the correlation coefficient > 0.7).

Introduction

The variability of the near-surface aerosol is caused by various factors, depending on the geographic location and season. The alternation of air masses plays an important role in the variability of aerosol in Central Russia. The mass concentration of aerosol in the marine arctic air mass, located in the rear of the cold front of an arctic cyclone, can decrease to ~ 5 $\mu g/m^3$. In the turbid continental air mass of the warm sector of a cyclone or the rear part of an anticyclone, the aerosol mass concentration is much higher. The ratio of the periods, during which the observation point is in some or other air mass, determine, to a large extent, the annual average content of aerosol at the given place.

The time variations of the aerosol characteristics were analyzed by many authors. Historically, the first long series of atmospheric transmittance were obtained from measurements at the ozonometric network. Based on these data and using the spectral analysis, Zuev, Belan, and Zadde[1] have established the presence of atmospheric transmittance variations with the periods from 2 days to 2 months and undertook an attempt to relate some of the periodicities to the meteorological processes in the atmosphere.

The investigation of the optical characteristics of the surface aerosol began from obtaining relatively short series, only several months long. Thus, in Ref. 2 Sidorov et al. have revealed the diurnal behavior of the scattering coefficient and a "train" of several quasi-periodic oscillations, connected with the synoptic cycle. In Ref. 3, based on the longer observation series, the annual behavior of the mass concentration of the submicron aerosol was obtained. In 1991, Sidorov started regular measurements of the mass concentration of the surface aerosol at Zvenigorod Research Station (ZRS) of the Institute of Atmospheric Physics.[4] In mid-1990s the Institute of Atmospheric Optics SB RAS[5] and, somewhat later, Kazan State University[6] initiated combined observations of atmospheric constituents, in particular, the mass concentration of aerosol particles. The analysis of the obtained material revealed certain regularities in the variability of aerosol characteristics. In particular, Khutorova[6] has established the existence of variations with the period of about 50 days for atmospheric constituents and meteorological quantities. It is interesting to note that the variations with close periods were also found in the near-surface content of nitrogen dioxide (NO_2) at ZRS.[7] A rather good correspondence (in time) of the peak values of the NO_2 content in the surface layer and the mass concentration of the surface aerosol was found in Ref. 8 for the period of summer fires near Moscow in 2002.

The aim of this work was to analyze the time variability of the three main parameters of the submicron aerosol: mass concentration, parameter of condensation activity, and Angström exponent in order to study the variations of these characteristics on the time scales from several days to months.

Technique and instrumentation

The analyzed data were obtained in 2001–2004 at Zvenigorod Research Station (ZRS) of the

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Institute of Atmospheric Physics with a spectral polarimeter. The station is located in a rural area about 50 km west of Moscow. The spectral polarimeter measures the polarization components of the scattering phase function D at the scattering angles of 45, 90, and 135° in a spectral range from 0.4 to 0.75 μm with the wavelength step of 20 nm. The polarimeter is equipped with a low-temperature heater for controlled heating and drying of the aerosol studied.

The measurements were conducted almost every day, and with only few missing days in 2004 (because of technical problems, power supply failures), as an example. The time for obtaining one record was 13 min; usually, 5 records were recorded and the average values of the parameters were determined. Then they were used to calculate the diurnally average values of the parameters. In the spectral and wavelet analysis, the gaps due to missing days were filled by interpolation.

In this paper, we use three aerosol characteristics: the mass concentration M, the steepness of the spectral dependence of the scattering phase function α (Angström exponent), and the parameter of condensation activity χ. The mass concentration of aerosol M was estimated from the value of the scattering phase function D at an angle of 45° and a wavelength of 0.54 μm by the well-known equation $M = 3000D$, where M is measured mass concentration in $\mu g/m^3$, and D is in $km^{-1} \cdot sr^{-1}$. The mean accuracy of determination of M is about 20%. The parameter M is more common for use aerosol investigations than D. The condensation activity (or the Hanel parameter χ) was estimated by a short method from two values of the scattering phase function (D_1 and D_2) obtained at different values of temperature and, correspondingly, relative humidity of the air (Rh_1 and Rh_2) in the working chamber:

$$D(Rh_{1,2}) = D(Rh = 0)\,(1 - Rh_{1,2})^{-\chi}, \qquad (1)$$

whence it follows that

$$\chi = \ln(D_1/D_2)/(\ln(1 - Rh_2)/(1 - Rh_1)). \qquad (2)$$

Thus, χ is the aerosol optical response, calculated by Eq. (2), to the decrease of the relative humidity of the air at heating. The meaning of the parameter χ is close to that of the characteristics of condensation activity γ, used in IAO SB RAS (in IAO, the aerosol is, to the contrary, moistened[5]), and even coincides with it under conditions of applicability of Eq. (1) in the range Rh = 40–80%. The errors in determination of χ are caused, first of all, by fluctuations of the aerosol characteristics due to its spatiotemporal inhomogeneity and are about 0.1, on the average. In 2004, to provide for a large Rh difference, the measurements were conducted mostly in the morning or in the evening at a high initial value of Rh. This allowed obtaining an almost continuous series of daily χ values. In the past years, the measurements were mostly carried out in daytime, and the relative humidity in the spring–summer period was sometimes lower than 30%, which practically made it impossible the determination of χ from Eq. (2).

The Angström exponent is the exponent α in the approximation of the spectral behavior of the scattering phase function by the inverse power law:

$$D(\varphi = 45°,\lambda) \sim \lambda^{-\alpha}. \qquad (3)$$

The parameter α was determined for all spectral dependences of D by the method of linear regression. In more than 90% of cases, the relative error of its estimation was 5–7%.

Recall that the variations of the particle concentration are major contributors to the variations of M. The particle size distribution function in the range of radii providing for the largest contribution to M is described, in the first approximation, by the inverse power law, and the variations of the distribution reduce mostly to the variations of the exponent. This exponent is closely related to the parameter α. Thus, the value of α is almost independent of the particle concentration and is related to M, mostly, through the characteristics of the distribution. Our investigations show that the variations of the parameter α provide for about a half of the total variance of M variations. Thus, the consistency or inconsistency of M and α variations allow us to judge on the variations of which parameter, the concentration or the average size of particles, are major contributors to the variations of the aerosol mass concentration.

By the definition (2), the parameter χ is independent of the particle number density. The coefficient of correlation between χ and M, calculated based on a large data array (about 3 years), amounts to about –0.3; there is a tendency to the decrease in the value and the spread of the parameter χ with the increase of M.

Various methods have been used in analysis of measurement data. The sliding-average method allows the high-frequency components of the variability to be filtered out in order to make the pattern of low-frequency variations more illustrative. The spectral analysis with the method of maximum entropy, having the increased spectral resolution, reveals the periodicities of the time scale close to the length of the series and is useful for analysis of low-frequency variations. The wavelet analysis complements the spectral analysis and allows the separation of individual variations and the estimation of their time scale to be performed. In this work, we used the Morlet wavelet. In the wavelet analysis, the autoregression extrapolation of the series at the edges was applied.[9]

Results and discussion

The analysis of the annual mean values of the aerosol mass concentration at ZRS measured since 1991 through 2004 and that with the use of the

Sidorov's data for the period until 2001 reveals the minimum in the annual mean content $<M> = 23$ μg/m^3 in 2001 (hereinafter, the angular brackets denote averaging). The estimation of $<M>$ in 2002 over 10 months (the period of very strong smokes present in the atmosphere due to forest fires was excluded) gives a somewhat higher value than in 2001. In 2003 the value of $<M>$ was about 1.5 times higher than in year 2001, and in 2004 ($<M> \approx 40$ μg/m^3) it almost doubled as compared to the level of $<M>$ in 2001.

The increase of $<M>$ from one year to another proceeded as follows. The year 2001 was characterized by a large number of situations with low M and almost complete absence of dense hazes with M higher than 150 μg/m^3. The most probable value of M in this and the following years almost did not change and amounted to about 50 μg/m^3. As compared to 2001, in the year 2003 the relative number of days with low M decreased drastically and the number of hazes with M higher than 100 μg/m^3 increased somewhat. Finally, in 2004, at roughly the same number of cases of weak scattering as in 2003, the number of dense and very dense (with $M > 150$ μg/m^3) hazes increased sharply. From the meteorological point of view, this increase of $<M>$ could be attributed to the shift of cyclone trajectories to the north, which would result in a weaker effect of the cold sectors of arctic cyclones. However, the confirmation or denial of the role of this mechanism calls for a specialized investigation.

The above-said is illustrated in Fig. 1 by the histogram of frequency of occurrence of different M values in 2001, 2003, and 2004.

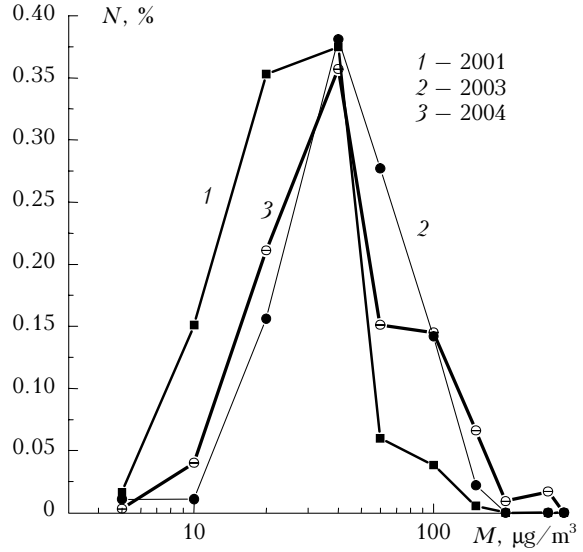

Fig. 1. Histogram of the frequency of occurrence of different values of the mass concentration M of the submicron near-surface aerosol at Zvenigorod Research Station measured in 2001, 2003, and 2004.

A somewhat uneven (in terms of the log M) division of histogram intervals is caused by the wish to emphasize the long right wing of the curve for 2004. It is just this behavior of the distribution wings at the almost unchanged most probable value of M that has determined the tendency toward the growth of the annual average value of M in these years. By the way, it should be added that in 2004 classical radiation fogs were observed near ZRS possibly for the first time in the last decade. (In meteorology, situations, classified by Rozenberg as dense hazes, when the relative humidity of the air is close to 100%, but the supersaturation needed for formation of the real fog is not achieved, are often mistakenly referred to as fogs).

Figure 2 shows the time profile of the diurnally mean values of M and α and the values of these parameters obtained in 2001 by applying the sliding average technique over 15 points. The sliding average allowed the almost synchronous large-amplitude oscillations with the mean period of about 45 days to be revealed in both of these characteristics more clearly. This period is close to the period detected in the variations of the content of atmospheric constituents and meteorological elements.[1,6,7] Note that neither earlier nor later that well pronounced variations of M with this period were observed at ZRS, though this period is usually present in spectra. The synchronism of M and α oscillations indicates that during almost the whole year the variations of M were mostly caused by the variations of the mean particle size. Despite a wide spread of these variations, the main features of the average seasonal behavior of M remained the same, namely, the April maximum and the summer minimum; the ratio of the M values at the maximum and the minimum was about two.

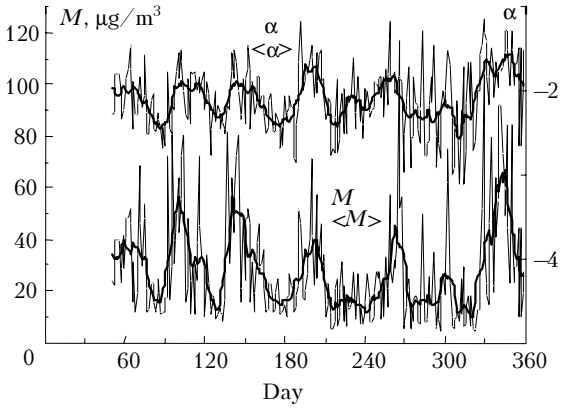

Fig. 2. Diurnally mean values (thin curves) and sliding 15-day average values (bold curves) of the mass concentration M and the Angström exponent α in 2001.

Quite different situation has been observed in year 2004. For 2004, Fig. 3 shows the behavior of the monthly mean values of the mass concentration M and of the parameter χ.

Curve *1* in Fig. 3 is the 8-year averaged behavior of the monthly mean values of M, borrowed from Ref. 4. The features of the annual behavior of

the parameter χ are in a good correspondence with the main regularities (established in Ref. 5) with the pronounced spring maximum and summer minimum. On the contrary, the seasonal behavior of the mass concentration M differs strongly from the many-year average behavior, and it cannot be called other than anomalous. The large-amplitude oscillations with the period of about 3 months are seen here even visually. As is shown below, the more accurate value of the period of oscillations is about 80 days.

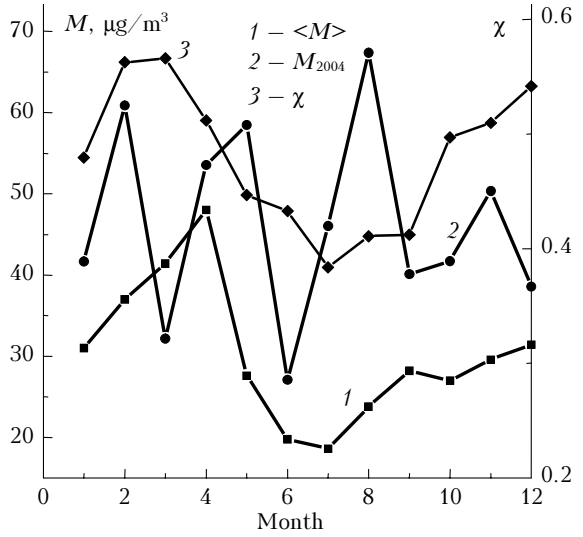

Fig. 3. Monthly mean values of the mass concentration M and the Hanel parameter χ for 2004 along with the many-year seasonal behavior of <M> based on Sidorov's data.[4]

Figure 4 depicts the time behavior of the diurnal and sliding 15-day average values of the mass concentration M and the parameter χ. The analysis of M and α has shown that in 2004 there were no correlation between them, which indicates the concentration origin of M variations. Therefore, for 2004 we restricted our consideration to only M and χ.

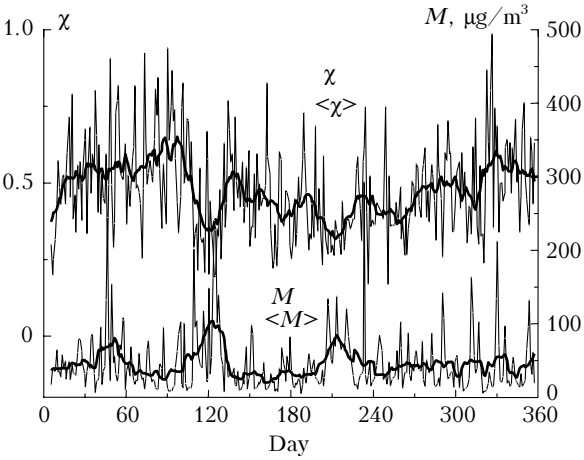

Fig. 4. Diurnally mean values (thin curves) and sliding 15-day averaged values (bold curves) of the mass concentration M and the Hanel parameter χ in 2004.

In the curve of the average M values, we can clearly see three complete oscillations with the increasing (see Fig. 6*b* below) period near 80 days. Their amplitude is so high that the ratio of M at the maximum and at the minimum achieves the value of five. The behavior of the curve for the average values of χ in Fig. 4 has its own features. It is possible to separate three parts of the curve with the gradual increase in January–March, decrease in April–August, and following increase of χ. Significant variations, which, most surprisingly, well correlate with the variations of M, are superimposed on the two last parts of the curve (spring – summer – fall).

The data for 2001 and 2004 were analyzed in a more detail using the spectral and wavelet analysis. The spectra of the studied parameters are shown in Fig. 5. In the low-frequency range of M and α spectra in 2001 and in the χ spectrum in 2004, we can clearly see the spectral maxima at the periods ~ 45 days.

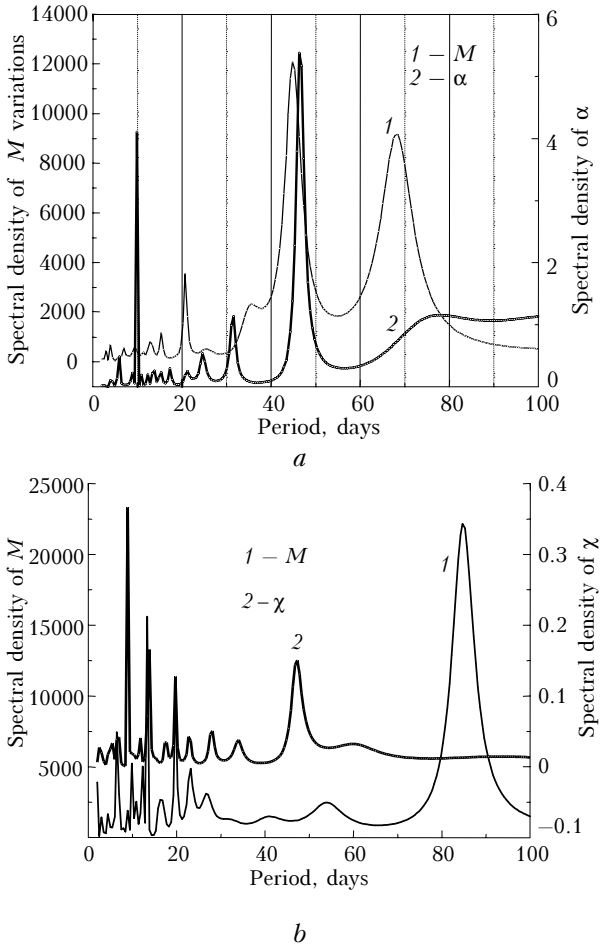

Fig. 5. Spectral densities of variations of the mass concentration M and the Angström exponent α in 2001 (*a*) and the mass concentration M and the Hanel parameter χ (*b*) in 2004.

The main feature of the low-frequency part of the M spectrum in 2004 is the spectral maximum with the period about 85 days. Perhaps, the only common

feature, pronounced in the M spectra for both of these years, is the presence of spectral peaks with the period ~20 days. In general, the spectral pattern of M variability in the high-frequency part (with periods shorter than 1 month) in 2004 is much richer than in 2001. It is worthy to note the presence of variations with the mean periods about 7, 10, and 14 days in 2004 (Fig. 5*b*). These periods are close to those noticed in Ref. 1. The spectral maxima of M with the periods of about 10, 14, and 20 days have analogs in the spectrum of χ as well. However, based only on the spectral analysis, it is difficult to judge on the correlation between M and χ at these frequencies.

The spectral analysis characterizes the mean power of fluctuations for the analyzed period. It is complemented by the wavelet analysis, which allows one to track the changes in the intensity and the period of variations. In particular, with the wavelet analysis used, we can try to reveal what processes are reflected in the 10-day peak in Fig. 5*b*. Figures 6*a* and *b* show the absolute values of the wavelet transform of the aerosol mass concentration in 2001 and 2004. The darker parts correspond to larger values. The parts with closed isolines correspond to individual, time-localized variations (both positive and negative with respect to the average level) on this time scale (see the vertical axis).

Figure 6*a* confirms the presence of variations with the mean periods about 40–50 and 20 days in the aerosol mass concentration for 2001. These variations occur in series during limited time intervals. The low-frequency oscillations consist of two series, and the period of variations inside these series increases gradually from ~30 to ~70 days. The period ~45 days in the corresponding spectrum shown in Fig. 5*a* reflects only variations with the maximum amplitude in Fig. 6*a*. It is worth noting the presence of a maximum at the period of 70 days in the spectrum. Obviously, it is the result of averaging of some low-frequency oscillations in late 2001 (see Fig. 6*a*). In Fig. 6*a*, the variations with the period of about 10 days are to be noted, which are weakly pronounced in the corresponding spectrum. The weakening of a signal in the spectral representation can be connected, in particular, with the phase mismatch between some oscillation series.

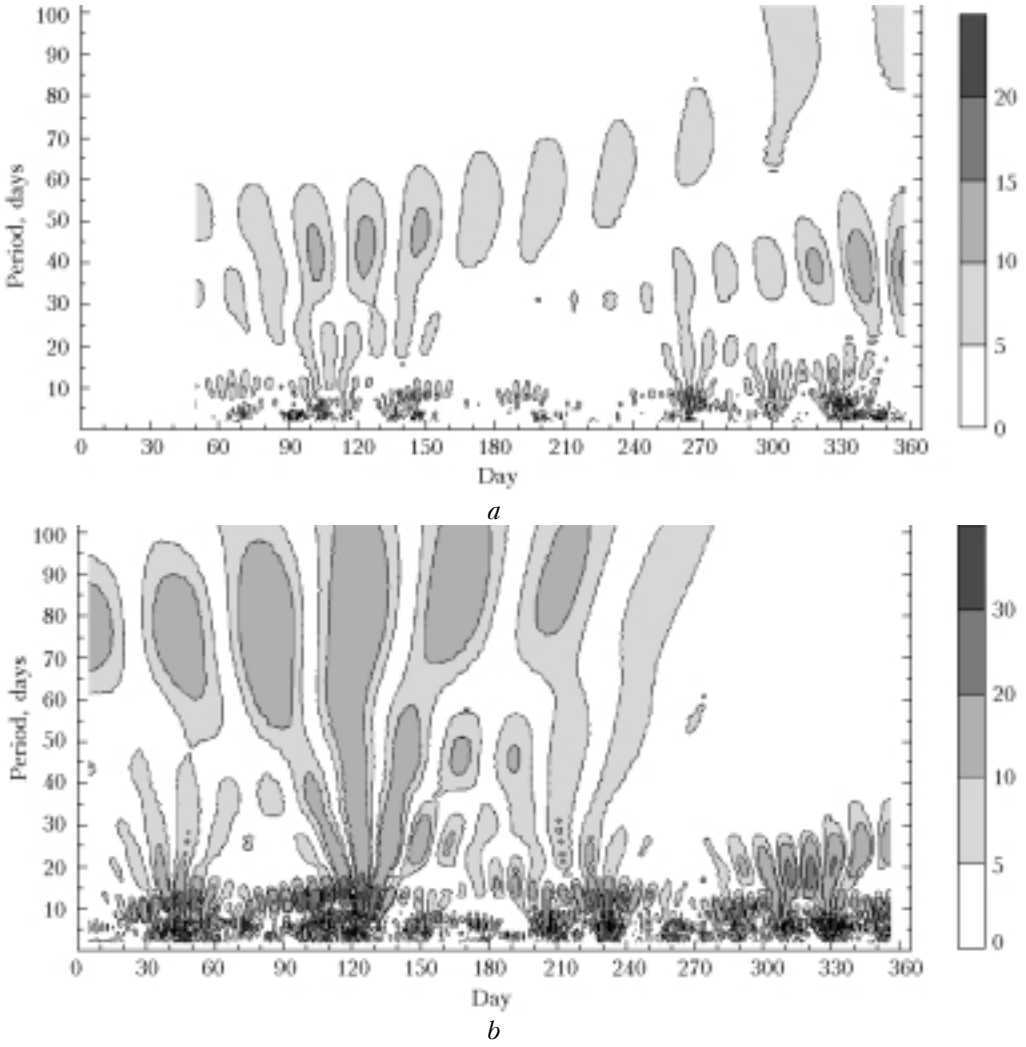

Fig. 6. Absolute value of the wavelet transform of the mass concentration M in 2001 (*a*) and 2004 (*b*).

The wavelet transform of M for 2004, as well as the spectral analysis, reveals the variations with the period of 80–90 days. In the early summer, it is also possible to see a weak variation with the period about 45–50 days, which almost does not manifest itself in the spectrum. A powerful 20-day peak in the spectrum shown in Fig. 5*b* is caused by a single series of variations in the end of the year (Figs. 6*b* and 4).

The variations with periods of about 7, 10, 14, and 20 days, reliably detected by the spectral analysis, manifested themselves in Fig. 6*b* as well. However, the variations with the mean periods of 10 and 14 days in the spring of 2004 merge into a single series, inside which it is possible to speak only about variations with the period varying in a wide range from 10 to 14 days. In general, in 2004 it is possible to separate out four series of variations with the periods near 10 days. Note that the return period of these series is close to the period of low-frequency variations (~ 90 days). Moreover, these series fall just on the intervals with the highest M values, which determine the 90-day component of the variations (see Fig. 6*b* and the nonsmoothed curve of M in Fig. 4). Thus, the correlation of these two components of aerosol variability can be considered as low-frequency 90-day modulation of high-frequency oscillations with the period of 10 days. The physical mechanism of the relation between variations at different scales (including, for example, the scales of 10–12 and 5–6 days; see Fig. 6*b*) calls for a specialized investigation.

In Ref. 1, Zuev, Belan, and Zadde proposed the mechanism of formation of the periods of 10–12 and 17–18 days long to be due to the passage of a series of 3–5 cyclones and an anticyclone through the observation point. This scheme can be useful for explanation of 20-day variations in the end of 2004. In the time scans of M, almost every cyclone in a series should "register" itself by its warm front and sector, that is, peaks of M. The series of three to five M maxima are just well seen in the end of the year in Fig. 4 (see the nonsmoothed curve of M) and Fig. 6*b* (where a series of variations with the period of 5 days is seen at the same time).

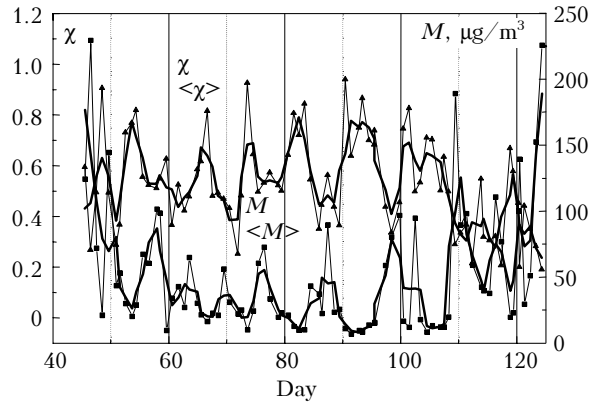

Fig. 7. Diurnally average (thin curves) and 2-daily average values (bold curves) of the mass concentration M and the Hanel parameter χ in February–April 2004.

In conclusion, Fig. 7 demonstrates the coordinated variations of M and χ with the characteristic scale of 6 days in the period from mid-February to late April of 2004. The bold curves in Fig. 7 correspond to two-day average values of M and χ. One can see clear and, what is most surprising, the anti-phase variations of M and χ even in nonsmoothed curves. At this time interval (about two and a half months!), the absolute value of the correlation coefficient of these parameters appeared to be higher than 0.7. This means that a half of the variance of M and χ variations in this period is connected with the same cause, having a periodic character.

Conclusions

The measurement data obtained in 2001–2004 have been used to analyze the variations of the three principal characteristics of the near-surface aerosol: the mass concentration M, the Angström exponent α, and the parameter of condensation activity χ. Using the data obtained in the previous decade,[4] it is shown that nearly decade-long decrease of the annual mean value of M in Central Russia stopped in the beginning of the current millennium, and during the past three years, its gradual increase has been observed. This is connected with the decrease in the frequency of occurrence of the cases with very low M values and the increase in the frequency of occurrence of the cases with very high, up to 250 μg/m^3, M values. It is shown that the long-period variations of M may have both the predominantly concentration origin, when the correlation between M and the Angström parameter is absent at large time intervals, and may be caused by the variation of the mean particle size. In the latter case, this manifests itself in the correlation between the variations of M and α. Time intervals with the duration up to several months and the high degree of negative correlation between M and χ (the absolute value of the correlation coefficient > 0.7) have been revealed. The spectra of aerosol parameters have been analyzed. In addition to the known periods of variations ~ 5, 7, 10, and 45 days, the periods of 14, 20, and 90 days have been revealed. The wavelet analysis has shown that the variations group in the form of time-limited series with the period and the amplitude of oscillations inside these series being variable.

Acknowledgments

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