Interannual variability of optical and microphysical parameters of the near-ground aerosol by measurements at the Zvenigorod Scientific Station

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During 2001–2006, spectral polarimeter and nephelometer measurements of mass concentration of near-surface submicron aerosol M and its condensation activity χ, were conducted at the Zvenigorod Scientific Station of IAP RAS. Variations in the above parameters at scales from several days to several months were analyzed. It is shown that there were more episodes of dense and super-dense hazes in Central Russia for these years. This has resulted in the change in the character of the aerosol mass concentration, M, in such a way that oscillations with increasing from-year-to-year amplitude appeared in the annual variation of M. Just due to more frequent episodes of dense and super-dense hazes in 2002–2006, a significant steady increase in the annual mean values of M is recorded. The annual mean value of M in 2006 increased more than twice as compared to 2001 (from 23 to 62 μg/m^3).

Introduction

The change of air masses plays the most important role in aerosol variability in the central Russia. The aerosol mass concentration in marine Arctic air mass, located at the cold front back of an arctic cyclone, can decrease to ~5 μg/m^3. The aerosol mass concentration in a turbid continental air mass of the cyclone warm sector or in back part of the anticyclone back is significantly higher (tens or hundreds of micrograms). Total durations of the observation points existence in one or another air mass mainly determines the recorded annual mean value of the aerosol content in the considered region.

The annual behavior of the mass concentration of submicron aerosol in the region of Caucasian Mineral Waters was reported in Ref. 1, based on the long series of observations. Seasonal behavior of the concentration of near-ground aerosol at the Zvenigorod Scientific Station (ZSS) of the Institute of Atmospheric Physics RAS was considered in Ref. 2 based on the 10-year series of observations. Some regularities in the aerosol parameter variability were revealed based on complex observations of the atmospheric components in Siberia and Tatarstan, including measurements of the aerosol mass concentration.[3,4]

Main attention was paid in Ref. 5 to the gradual increase of annual mean values of the submicron aerosol mass concentration M at ZSS since 2002.

Now, practically continuous 4-year long series of observation of M and χ (January, 2003–December, 2006) are in our disposal. This paper is devoted to analysis of variations of these two main integral parameters of near-ground aerosol in Moscow Region during 2003–2006. Measurements have been carried out by means of a spectral polarimeter and partially with the V.N. Sidorov nephelometer.[2] The station is situated at the rural zone approximately 50 km to the west from Moscow.

Instrumentation for determination of the aerosol parameters

The spectral polarimeter recorded the polarized components of the scattering phase function D at angles of 45, 90, and 135° within the wavelength range 0.4–0.75 μm with a wavelength step of 20 nm. The device was equipped with a low-temperature heater for controllable heating and drying of the aerosol under study.

Measurements were carried out almost every day, and the quantity of omissions (because of the device disrepair or the electricity switch-off), for example, in 2004, made only a few days. The duration of recording of one measurement series was 13 minutes, a recording block usually consisted of 5 series; the mean values of the parameters were stored. Then the daily mean values of the parameters were calculated based on these data.

The aerosol mass concentration M was estimated from the value of the scattering phase function D at an angle of 45° and a wavelength of 0.54 μm using the well-known formula $M = 3000D$, where M is in μg/m^3 and D is in km$^{-1}\cdot$ sr^{-1}. The mean accuracy of M determination was about 20%. The use of M (instead of D) is more habitual for aerosol researchers.

The condensation activity (or the Hanel parameter χ) was estimated using two values of the scattering phase function (D_1 and D_2) obtained at different values of temperature and relative humidity (Rh_1 and Rh_2) in the working chamber of the device.[6] Thus, χ is the optical response of aerosol to

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the relative air humidity decrease obtained by its heating. The parameter χ, in its meaning, is close to the parameter of condensation activity γ used at the Institute of Atmospheric Optics SB RAS (at IAO SB RAS, on the contrary, aerosol is humidified[3]). The parameters practically coincide in the range $Rh = 40–80\%$. The errors in determining χ are caused, first of all, by fluctuations of aerosol caused by its spatial-temporal inhomogeneity and are, in average, about 0.1. The parameter χ, by definition, does not depend on the number density of particles.[6]

As it was earlier noted,[6] the quality of the data on χ depends on the air relative humidity at the moment of measurements, so, starting from 2004, measurements were carried out in the morning or late in the evening, when the relative humidity is quite high, in order to obtain more reliable estimates of χ. Measurements in 2003 were often carried out at daytime, however, owing to cold rainy summer in Moscow Region, relative humidity of air was high even at daytime, that allowed us to obtain the data on χ. The nephelometer, designed by V.N. Sidorov, equipped with a low-temperature heater of air, operated most of time in 2003 together with the spectral polarimeter. This allowed filling the gaps in the data on M and χ with the nephelometer data in some cases.

Results and discussion

Temporal behavior of the daily mean values of the mass concentration M is shown in Fig. 1; the thick curve shows the 15-day moving average.

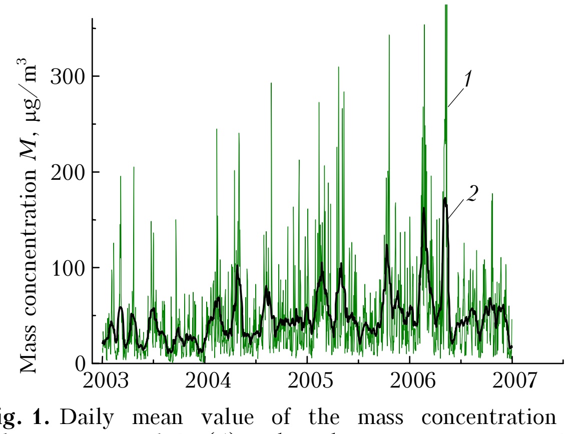

Fig. 1. Daily mean value of the mass concentration of submicron aerosol M (*1*) and 15-day moving average $\langle M \rangle$ (*2*) in 2003–2006.

Underline the following peculiarity of the daily mean M values: the well pronounced growth of the magnitude and frequency of occurrence of the M peak values is observed during the whole period 2003–2006.

The moments of the peaks appearance are quite ordered, namely, they are grouped into series separated by the intervals where the M values are close to the mean values. The fragment of the series corresponding to three first months of 2003 is shown in Fig. 2 for comparison. It is seen that the great levels of M in this period appear with an interval of about 4 days, and the length of such series is 2–3 weeks.

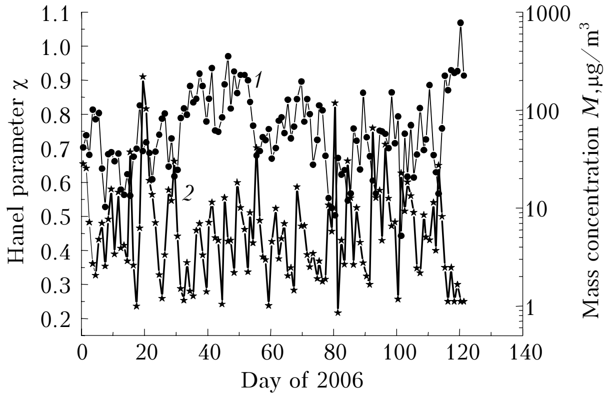

Fig. 2. Daily mean values of M (*1*) and χ (*2*) in January–April, 2006.

The characteristic oscillations with a time scale of about 3 months, related to appearance of dense and super-dense hazes, are observed in the M moving averages starting from winter 2004. There are 3 oscillations in 2003, two in 2005, and two in 2006, the beginnings of these series fall on the end of December – beginning of January. Most likely, it is related with the peculiarities of the winter regime of the atmospheric circulation.

The frequency of occurrence of such hazes monotonically increases from year to year. The histograms of M values obtained from the data of measurements in 2001 and 2003–2006 are shown in Fig. 3 (the data of 2002 were not used because of the severe smokes during forest fires in summer).

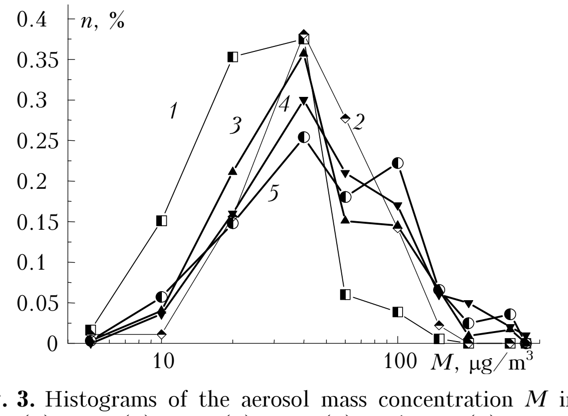

Fig. 3. Histograms of the aerosol mass concentration M in 2001 (*1*), 2003 (*2*), 2004 (*3*), 2005 (*4*), and 2006 (*5*).

As compared to 2001, relative quantity of days with M exceeded 100 μg/m^3 dramatically decreased in 2003. The quantity of dense and very dense hazes ($M > 150$ μg/m^3) increased in 2004 at approximately the same number of episodes of weak scattering in 2003. This increase continued in 2005–2006, and the super-dense hazes for the last decade were first recorded, when the M mass concentration of the aerosol dry matter exceeded 350 μg/m^3, and M = 770 μg/m^3 was observed on May 1, 2006.

The tendency of elevation of the right wing of the distribution from year to year is well seen in Fig. 3. This is caused by the increase of the frequency of occurrence of dense hazes. The histogram of M in 2006 is bimodal, the second M maximum appears in the range 100–150 μg/m^3 (i.e., dense hazes), however, in spite of the distribution deformation, position of the most probable M value (main maximum of the histogram) is invariable during these years and is equal to 40 μg/m^3.

Such manner of the M frequency distribution deformation strongly affects the seasonal M behavior. Curve *1* in Fig. 4 is the behavior of monthly mean M values in 1991–1998 [Ref. 2]. A close seasonal behavior of M in 2003 is not demonstrated, for to avoid overloading the figure. It is well seen how the peak values of winter–spring oscillation of the monthly mean M value increase from year to year.

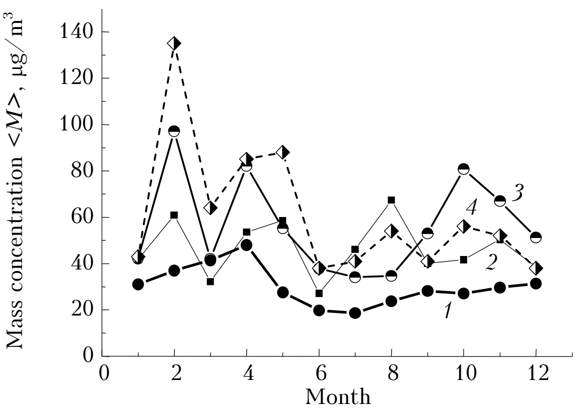

Fig. 4. Seasonal behavior of the monthly mean M values: average over 1991–1998 from the data by V.N. Sidorov (*1*), 2004 (*2*), 2005 (*3*), 2006 (*4*).

This process affected even the behavior of the annual mean M values (Fig. 5). The data for 1991–2000 were obtained by V.N. Sidorov. It follows from Fig. 5 that almost monotonic decrease of M in 1991–2001 is replaced in 2002 by quite quick monotonic increase of the annual mean M values (the annual mean M value in 2002 is obtained for 10 months, the data obtained during forest fires are excluded), that is evidence of the probable changes in the atmospheric circulation in 2002.

Dense and super-dense hazes appear in warm sectors of atmospheric cyclones, as well as in anticyclones under conditions of strong atmospheric inversions. Let us emphasize that they deliberately are not the consequence of some anthropogenic emissions in Central Russia. Neither “stove smog” nor forest fires are connected with formation of these hazes. This conclusion follows from the results of comparative analysis of particle size distributions in such hazes and smokes of forest fires.[6]

The narrow bell of the main distribution mode in the vicinity of the particle size $r \sim 0.2$ μm is characteristic of the smoke particle volume size distributions. This is expressed in almost neutral spectral behavior of $D(\varphi = 45°, \lambda)$ in smokes. Distributions obtained in dense hazes are wider, their mode refers to the r range 0.1–0.12 μm, and, as a rule, there present a part with inverse power law in such distributions in the radius range 0.2–0.6 μm characteristic of hazes. In other words, usual and dense hazes are different mainly in the concentration of particles and in the exponent of the inverse power law approximating the distributions.

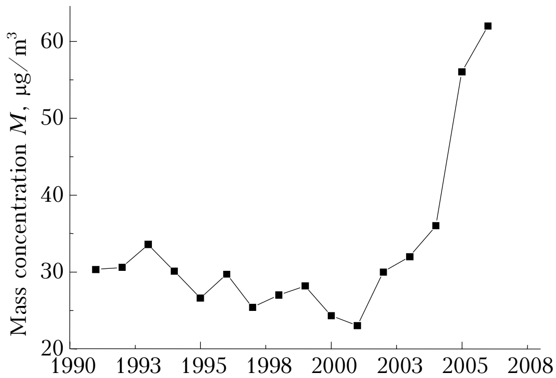

Fig. 5. Annual mean value of the mass concentration <M> in 1991–2006.

If from the standpoint of meteorology, the decrease with time of the number of episodes with weak scattering (i.e., increase of <M>) could be related to displacement of cyclone trajectories to the north, i.e., with weakening the effect of arctic air masses in cold sectors of arctic cyclones (confirmation or refutation of the role of this mechanism invites further investigations), then the much more often cases of super-dense hazes require a special explanation.

In some cases, the appearance of dense hazes is related with accumulation of aerosol in the ground layer under conditions of strong near-ground temperature inversions in anticyclones; however, the cases when high levels of the aerosol content have been observed in warm sectors of cyclones in the absence of any near-ground inversions are not rare as well. Exceeding of the M mean level in such cases reaches an order of magnitude. Therefore, the dense and super-dense hazes most strongly affect variations of the calculated monthly mean and even annual mean M values in 2003–2006.

Temporal behaviors of the Hanel parameter χ and the 15-day moving average <χ> are shown in Fig. 6, similarly even to Fig. 1; the behavior of monthly mean values of χ, which is well pronounced even in moving averages, is shown in Fig. 7.

Some peculiarities of the χ temporal behavior are similar to main regularities in γ variations.[3] In 2004–2005, the winter–spring maximum and summer minimum are observed against the background of variations of the monthly mean values. Such behavior is attributed in Ref. 7 to the snow cover disappearance and emission into the atmosphere of the substances accumulated in snow during winter. Pay our attention to the correlated opposite-phase variations of M and χ with the period of about 4 days in Fig. 2, where the amplitude of χ variations is much greater than its seasonal variations. Thus, the presence or the absence of the snow cover is not the main reason of χ variations.

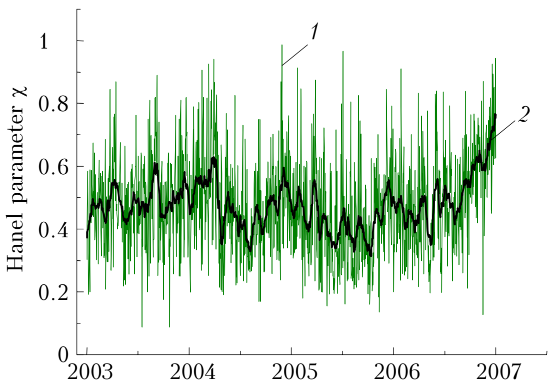

Fig. 6. Daily mean value of the Hanel parameter χ (*1*) and 15-day moving average (*2*) in 2003–2006.

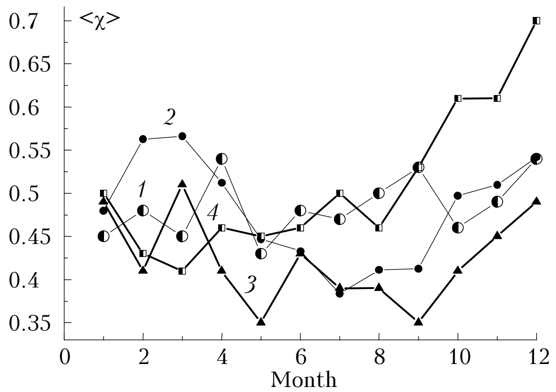

Fig. 7. Monthly mean values of the Hanel parameter in 2003 (*1*), 2004 (*2*), 2005 (*3*), and 2006 (*4*).

Oscillations with the period of about 7 weeks are observed in the χ temporal behavior, as though they modulate the 4-day χ variations even without the moving smoothing (see Fig. 6). They are just the periods, which were revealed in Ref. 7.

The frequency histograms of the Hanel parameter value are shown in Fig. 8 separately for each of 4 years of observation and for the total period. The combined histogram is well approximated by the normal distribution with the mean value $\chi = 0.5$ and the standard deviation $s = 0.21$.

At the same time, annual distributions very differ from each other – the distribution in 2003 is narrower, in 2005–2006 distributions are, on the contrary, noticeably wider, the first of them becomes wider owing to the left wing (smaller values χ), the second, on the contrary, widens owing to the right wing, so that the most probable χ value in 2006 is equal to 0.6. Note that the well-known weather anomalies observed at the end of 2006 become apparent in the last case – very warm and wet marine mid-latitude air masses from Atlantic were prevalent over Russia in November and December.

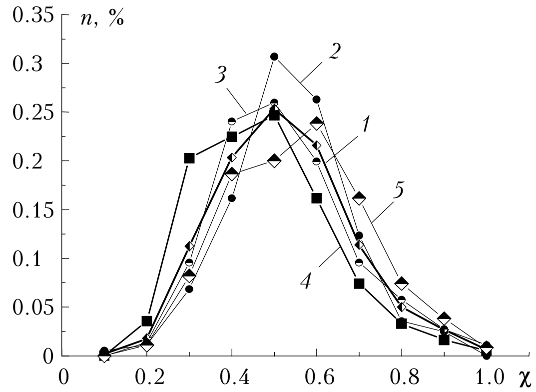

Fig. 8. Histograms of distribution of the Hanel parameter χ from the data of 2003–2006 (*1*), 2003 (*2*), 2004 (*3*), 2005 (*4*), and 2006 (*5*).

Conclusions

Variations of two main parameter of the near-ground aerosol, the mass concentration M and the parameter of condensation activity χ have been analyzed based on the data of measurements at the Zvenigorod Scientific Station of IAP RAS in 2001–2006. It is shown that the episodes of dense and super-dense hazes became more often in central Russia during these years, which has led to the change in the character of seasonal dependence of the aerosol mass concentration M: there appeared regular oscillations with increasing from year to year amplitude in the M seasonal behavior. Significant monotonic increase of the M annual mean values is observed owing to just these more frequent cases of dense and super-dense hazes in 2002–2006. The annual mean value of M in 2006 is approximately three times higher than that in 2001 (62 instead of 23 $\mu g/m^3$).

According to the data of 2003–2006, the spring maximum and summer minimum are present in seasonal behavior of the Hanel parameter. At the same time, the amplitude of χ variations with the periods of a few days is greater than the amplitude of its seasonal behavior. Therefore, it can be supposed that seasonal behavior of χ results from seasonal modulation of the processes, which are responsible for the short-period variations. The histogram of distribution of the Hanel parameter for the total period is well approximated by the normal distribution with the mean $\chi = 0.5$ and standard deviation $s = 0.21$, however, the distributions drawn separately for each year are different both in mean value and in the variance.

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