Annual behavior of the condensation activity of submicron aerosol in the atmospheric surface layer of Western Siberia

M.V. Panchenko, S.A. Terpugova, V.S. Kozlov, V.V. Pol'kin, and E.P. Yausheva

Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences, Tomsk

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We discuss the data of long-term measurements of the dependence of the submicron aerosol scattering coefficient on relative humidity in the near-ground layer of the atmosphere carried out at the Aerosol Monitoring Station of IAO SB RAS. To study the dynamics of the optical characteristics under the effect of relative humidity, the approach was applied based on separate investigation of the optical parameters of the dry matter of aerosol particles and their change when moistened. The characteristic peculiarities of the annual behavior of the condensation activity of submicron aerosol have been revealed. These peculiarities are stably observed from year to year. The effect of different air masses in every season is analyzed. It is shown that the processes that govern the formation of the principal optical characteristics of submicron aerosol are at least of a regional scale.

Relative humidity of the air is one of the most important factors determining the transformation of the optical and microphysical properties of the atmospheric aerosol. At the same time, the study of the effect of humidity in real atmosphere is quite difficult because it is practically impossible to discriminate the effect of relative humidity against the background of general variability of the optical state of the atmosphere determined by the whole complex of geophysical processes.

To isolate the pure effect of relative humidity on the dynamics of aerosol optical properties transformation at moistening, we have studied the optical properties of the dry aerosol matter, i.e., reduced to zero relative humidity and separately their change due to moistening of aerosol.

Optical properties of the dry matter are determined, first of all, by the total aerosol content and are governed by quite long processes of generation, accumulation, and sink of the aerosol from the atmosphere. The change of relative humidity is more quick process, which has diurnal behavior in the majority of events.

In due time, Kasten[1] and Hanel[2] proposed empirical formulas for describing the dependence of the parameters of aerosol particles (radius and scattering coefficient) on the relative humidity of air. In particular, for the directed scattering coefficient $\mu(45°)$ one can write:

$$\mu = \mu_0(1-RH)^{-\gamma}, \qquad (1)$$

where μ_0 is the coefficient of directed scattering by the dry aerosol particles, RH is the relative humidity of air, γ is the parameter of condensation activity of particles, which determines the dynamics of the optical characteristic as a function of humidity. This parameter depends on chemical composition and size distribution of dry aerosol particles and, first of all, on the ratio of soluble and insoluble substances in it.

At present, the majority of foreign research groups use Tandem Differential Mobility Analyzers (TDMA) for the study of condensation activity, in which the dependence of the particle size on relative humidity is measured.[3–5] This method enables one to describe in detail physics of the process of the influence of humidity on the particle size distribution. However, if it has been necessary to describe the change of any optical parameter at the change of relative humidity, serious difficulties appear in estimation of the optical constants of real aerosol.

As applied to optical problem, our approach based on direct measurements of the optical characteristics is free of these disadvantages.

To study the condensational transformation of aerosol, the nephelometric setup was used, the block-diagram of which is shown in Fig. 1. Measurements of the aerosol directed scattering coefficient at the angle of 45° at the wavelength of 0.51 μm were carried out with two nephelometers simultaneously. One of them was equipped with the device for artificial moistening of aerosol under study and was used for measuring the dependence of the scattering coefficient on relative humidity (a hygrogram). The second one was a part of the instrumentation set at the aerosol monitoring station[6] operated in a routine mode for measuring the directed scattering coefficient of the dry matter of aerosol particles, which characterizes the total content of aerosol in the air.[7]

Time behavior of the scattering coefficient recorded with this nephelometer enabled us to observe the variability of the aerosol content in the atmosphere. The nephelometers were calibrated in absolute units ($km^{-1} \cdot sr^{-1}$) by means of measuring

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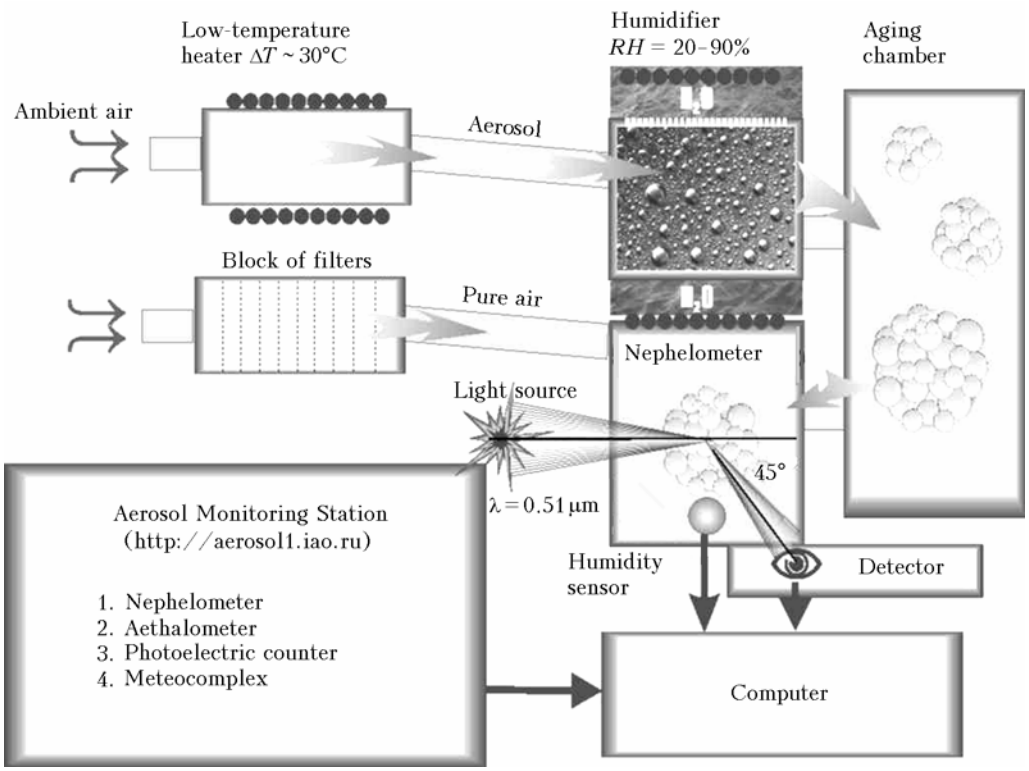

Fig. 1. Block-diagram of the experimental setup.

the signals of scattering by pure gases with the known values of the molecular scattering coefficients. Current calibration was performed before and after the measurements by means of the optical prism and using the molecular scattering of pure air.

Let us note that the air-sampling channels of the nephelometers provide for correct collection of particles with the diameters not exceeding few micrometers, while the larger particles settle in the intake channels. Hence, the main results and conclusions of this paper refer mainly to particles of submicron fraction.

As the directed scattering coefficient at the angle of 45° is closely related to the total scattering coefficient,[8,9] all conclusions drawn in this paper concerning the main peculiarities of the time behavior of the measured parameters are also applicable to the total aerosol scattering coefficient.

The low-temperature heater was additionally included into the air-sampling channel of the nephelometer for active artificial impact (Fig. 1) just after air intake. It provided heating of the air under investigation by $\Delta T \sim 30$°C higher than the initial temperature, what enabled us to decrease the value of relative humidity, from which the record of hygrogram started, to 20–40%. Then air passed through the humidifier designed as a cylindrical chamber with the diameter of ~ 10 cm and the length of ~ 30 cm. To humidify the air, distilled water was spilled on its walls; in addition, the walls were heated. Changing the water spilling rate and the intensity of heating, one could regulate the evaporation rate, hence, the rate of the change of relative humidity. To bring aerosol to equilibrium with water vapor during humidification, the aging chamber with the volume of ~ 4 l was included into the path between the humidifier and the nephelometer. The value of relative humidity was measured just near the scattering volume by the IH–3602–C sensor.

Before measurements, the air is blown through the filter system. The signal recorded at this time comprised the component proportional to the molecular scattering from the same air volume as at measurement of the aerosol characteristics, as well as the "noise" component. Subtraction of this value from the total signal recorded at the aerosol flow photometry enables one to obtain the pure signal proportional to the aerosol directed scattering coefficient.

The duration of recording a hygrogram was 10–12 minutes, during this time humidity gradually increased up to 90–95%. The higher values of humidity were not considered, because in dynamic regime it is difficult to control the effect of the possible contribution of the processes of condensation of water vapor when approaching the water vapor saturation point.

If variations of the scattering coefficient of the aerosol dry matter measured by the reference nephelometer in parallel to measurements of the hygrogram exceeded 10%, the corresponding

correction was applied to the signal recorded on the hygrogram at a relevant time moment. If variations of $\mu_0(45°)$ exceeded 50%, such measurements were rejected. Also, the situations were removed from processing, when the aerosol scattering coefficient had been less than the half of molecular scattering coefficient, because in this case the error in measuring the aerosol scattering significantly increased.

The error in measuring the aerosol directed scattering coefficient with our setup, at the mean level of turbidity observed in the atmosphere of Western Siberia, is not greater than 10%.[10] Taking into account the errors in data processing and calculating the parameter γ, the absolute error in determining the parameter of condensation activity is ~0.05.

Measurement of hygrograms have been carried out from March 1998 until now once a day. This enables us to study in detail temporal variability of the parameter of condensation activity. The obtained dependences of the directed scattering coefficient on relative humidity in the range 40–90% were approximated by Eq. (1). The values γ were determined by the least squares method in the coordinates $\ln\mu - \ln(1 - RH)$.

The change of the aerosol condensation activity in 1998–2004 is shown in Fig. 2. As is seen from the data obtained, the characteristic peculiarities of the annual behavior of γ is stable from year to year.

On the average, its values in winter are higher than in warm season. The aerosol condensation activity begins to dramatically increase in the beginning of March and reaches its maximum by the beginning or the middle of April. The presence of spring maximum is the most characteristic peculiarity of the annual behavior of this parameter. It is invariable and observed every year, although its value is different in different years and depends on weather conditions. However, we failed to reveal a relation of the position and the value of the maximum to any atmospheric parameter.

The decrease of the parameter of condensation activity continues from the middle of April until July, then the increase is observed since August until the middle of October, and the constant, on the average, value γ is recorded from November until March. Only local maxima and minima are observed in this period. The absolute values of them are significantly lower than the global spring maximum, and the position is not hard fixed.

Such structure of annual behavior is caused by two principal causes, either essential change of physicochemical properties of aerosol particles when passing from cold season to warm and back, or noticeable transformation of the particle size spectrum. Both hypotheses correspond to the existing ideas of the principal factors determining the reaction of particles on the change in relative humidity of air. Significant spatial contrast of the state and power of the sources of particles and aerosol producing vapor is observed in spring. In this season, the aerosol particles produced from various chemical compounds that have been accumulated in winter and emitted into the atmosphere because of snow melting in west regions, come into the atmosphere of Western Siberia. But the most significant increase of the values γ is observed, as a rule, after beginning of intense melting of the upper edge of snow cover immediately in the region of measurements.

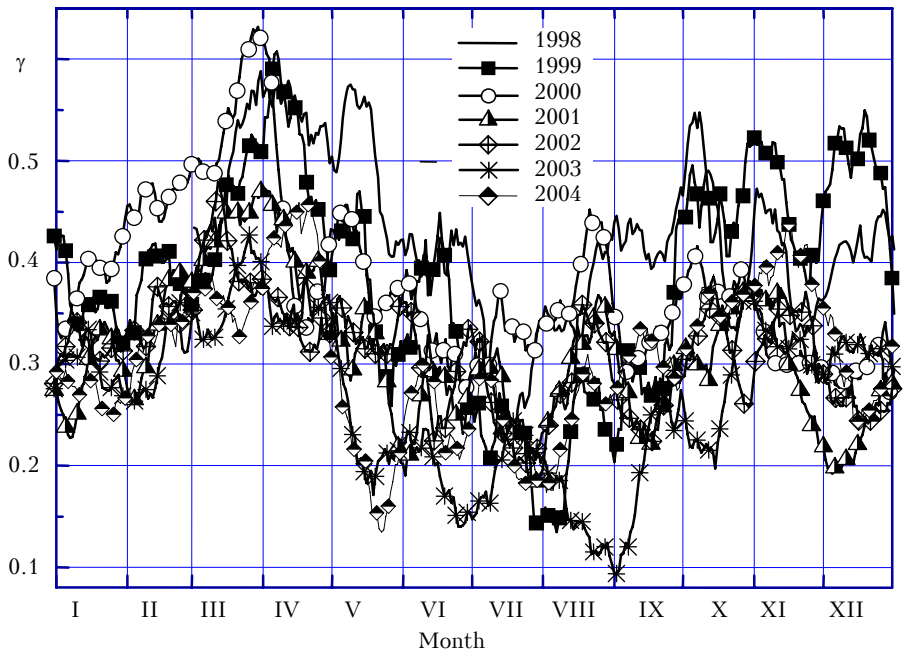

Fig. 2. Annual behavior of the parameter of condensation activity.

When analyzing practically any data obtained in the atmosphere at specific observation site, it is always necessary to answer two questions: is the revealed peculiarity characteristic of the whole region, and to what degree they are affected by the nearby industrial centers (Tomsk in our case). In order to reveal the effect of anthropogenic factor on the obtained results, a series of experiments was carried out, in which the aerosol and meteorological parameters were measured in parallel in Tomsk and in the background forest region near Kireevsk village, which is approximately 60 km far from the city. Comparison of the series of the total aerosol number density and scattering coefficients measured at these sites has shown that formation of the field of the submicron aerosol concentration in the near-ground layer of the atmosphere occurs on the regional scale, and the effect of anthropogenic factor on the obtained data is observed only in 25% of realizations.[11] The experiment carried out in spring of 2001, in addition to the parameters characterizing the aerosol concentration, included measurements of the condensation activity. The choice of spring season was dictated by the following reasons. First, the greatest variability of all atmospheric parameters is observed in this season. Second, the series of the values γ compiled by this date already had revealed the stable spring maximum repeated from year to year. Third, the question would be answered, is this maximum caused by the effect of the city on the measured parameters, or it is characteristic of the region as a whole. Temporal behavior of the parameter γ obtained during this experiment at two sites is shown in Fig. 3. It is seen that the difference between the measured values γ in Tomsk and Kireevsk in the majority of cases does not exceed the error in its determination, and, hence, confirms once again that the processes of formation of submicron aerosol have, at least, regional scale.

To study the effect of synoptic processes on the aerosol condensation activity, the data were analyzed separately in each season, for two main types of air masses determining the weather in West-Siberian region — Arctic and continental mid-latitude. The mean γ values are shown in the Table.

Season	Air mass		Reliability of the difference, %
	Arctic	mid-latitude	
Winter	0.37 ± 0.11	0.46 ± 0.12	99
Spring	0.55 ± 0.14	0.47 ± 0.15	99
Summer	0.32 ± 0.15	0.32 ± 0.14	< 10
Fall	0.42 ± 0.16	0.39 ± 0.17	< 40

The aerosol condensation activity in winter in mid-latitude air mass is greater than in Arctic (the reliability of the difference is 99%). In spring the difference between the masses is reverse. In summer and fall, the mean γ values in two air masses are practically the same. Obviously, it is related to different prehistory of air masses coming to Western Siberia in different seasons.

The seven-year long period of observations already enables us to perform some estimates of interannual variability of the parameter of condensation activity. The annual mean values of the parameter γ are shown in Fig. 4. Its seasonal mean values for different years are also presented here.

It is seen that the decrease of γ from year to year is observed in the period from 1998 to 2003. This peculiarity is characteristic of not only the year as a whole, but also for each season. At the same time, as is seen from Fig. 2, the decrease of the amplitude of annual behavior is observed in this period. The maximum values of γ observed in the end of March — beginning of April are ~ 0.6 in 1998–2000, and only ~0.45 in 2003. However, starting in 2004, the tendency of increasing of the annual and seasonal mean values of γ is observed. The mean value in spring 2005 already reaches the level of 1999–2000.

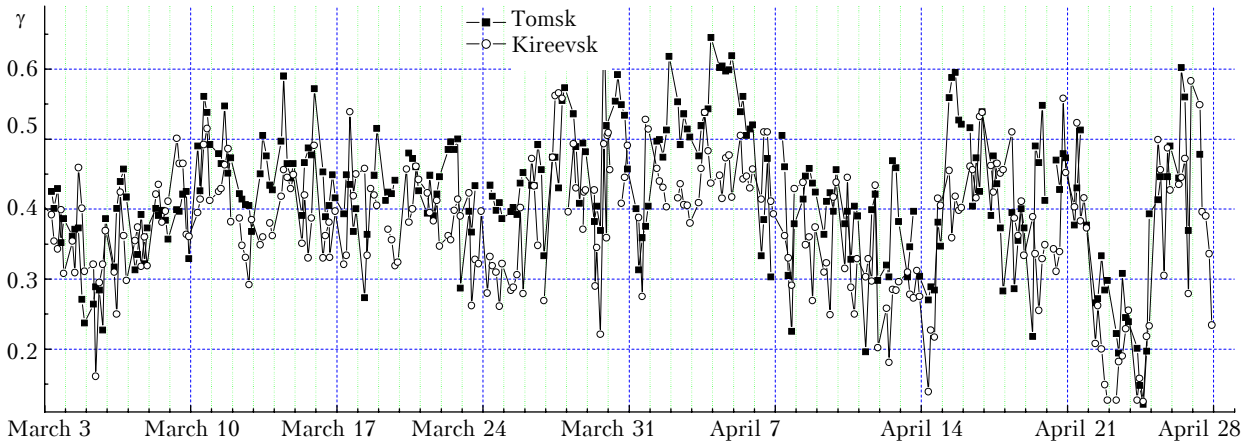

Fig. 3. Temporal behavior of the parameter of condensation activity in 2001 in Tomsk and in the background forest region near Kireevsk village.

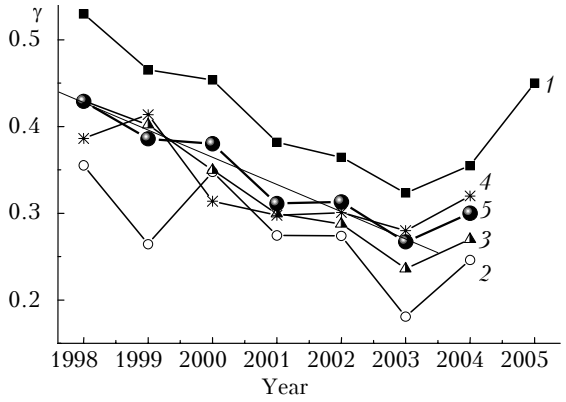

Fig. 4. Interannual variability of the parameter of condensation activity: spring (*1*), summer (*2*), fall (*3*), winter (*4*), annual mean value (*5*).

As was shown above, the main cause of the appearance of different γ values is the variation of the content of substances of different solubility in the composition of aerosol particles. Unfortunately, we have no available data of systematic measurements of the aerosol chemical composition in these years at the site of observations. One more possible cause can be different condensation activity of particles of different size. To check this hypothesis, the parameter X, which is the ratio of the number densities of submicron (the particle radius $r = 0.2–0.5$ μm) and coarse ($r = 0.5–2$ μm) fraction of particles was calculated from the data on the particle size distribution measured with a photoelectric counter at the aerosol monitoring station: $X = N_{sbm}/N_c$. Temporal behavior of daily and monthly mean values of this parameter during 1999–2000[12] is shown in Fig. 5.

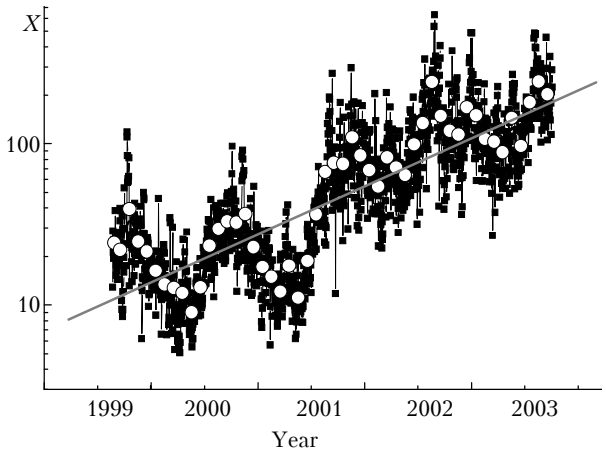

Fig. 5. Time behavior of the ratio of concentrations of submicron and coarse fractions: squares correspond to daily mean values, circles correspond to monthly mean values.

There is a direct evidence of the tendency toward an increase in its value, hence, the increase of the relative content of smaller particles in the aerosol size spectrum. It is early to interpret in this paper such interannual behavior of the relative content of submicron particles as the main cause of the decrease of the parameter of condensation activity. The study of condensation activity of particles of different size at our site of observation have started only recently,[13] however, this fact can confirm that noticeable interannual variations of the aerosol characteristics really occurred in this period in the region under study.

Summing up the results as a whole, let us emphasize that the revealed stable seasonal variations of the parameter of condensation activity of submicron aerosol repeating from year to year (it especially well pronounced when passing from cold period to the warm one) enables one to assume that this process is typical for the atmosphere (at least, for the considered geographical region).

Acknowledgments

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