DYNAMICS OF DIURNAL AND SEASONAL CYCLES OF AEROSOL FORMATION IN THE ATMOSPHERE AS IT FOLLOWS FROM MEASUREMENTS IN NOVOSIBIRSK REGION

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In this paper we present some results of the summer expedition in 1994 undertaken to study the dynamics of diurnal variations of the atmospheric aerosol concentration. The measurements are being conducted simultaneously at three points of Novosibirsk region. Comparison of the measurement data at different points of Novosibirsk region allows one to draw the conclusion on the dynamics of natural diurnal cycles and their anthropogenic distortions. Comparison of the summer measurements with those in other seasons reveals seasonal variations in the atmospheric aerosol concentration.

1. INTRODUCTION

The role the atmospheric aerosol (AA) plays in the Earth's climate changes is great both on global and local scales. This is because the AA particles actively affect the radiation transfer in the atmosphere by absorbing and scattering solar and thermal radiation as well as because they are condensation centers in formation of clouds and fogs.

The concentration and particle size spectrum are the most important characteristics of atmospheric aerosol. Recent investigations revealed the regularity in variations of AA concentration and its size spectrum. In addition to the diurnal cycles of aerosol concentration owing to anthropogenic activity (rush hours in traffic, etc.) there are natural diurnal and seasonal cycles of the atmospheric aerosol concentration of natural origin due to the processes of gas-particle transformation and mixing of the ground atmospheric layer.

However, the experimental data available are of fragmentary character and insufficient for creating a quantitative model describing the above cycles. The primary goal of our studies is acquisition of systematic and complete data sets from the observed diurnal and seasonal cycles of the aerosol concentration as well as the development of a model describing these cycles.

The investigations conducted in 1990-1993 near the Baikal and in Novosibirsk region within the framework of the Project "Siberian Aerosols" provided the basis of this work. During the following prolonged measurements near the ground, pronounced diurnal and seasonal cycles of variation of concentration of aerosol submicron fraction of natural and anthropogenic origin were observed. The dynamics of aerosol concentration can be explained in general by the known theories of aerosol formation.

2. CHARACTERISTIC OF OBSERVATION POINTS

Observations were performed at several points in Novosibirsk region.

1. The first point, where regular observations were conducted of dynamics of diurnal variation of aerosol concentration, is located at the eastern boundary of Novosibirsk Scientific Center, Akademgorodok SB RAS. The equipment was mounted in the building of the Institute of Chemical Kinetics and Combustion SB RAS. Sampling was made at 6 m height above the ground surface from the second floor of the Institute building. Since we mainly observed the aerosol particles of 0.1–1 μm size range, there was no need to keep track of isokinetics of sampling and minimizing of diffusion losses of particles in the intake hoses.

2. The second observation point was 12 km to the east of Akademgorodok close to the Klyuchi settlement. This sampling point can be characterized as "suburban".

3. The third observation point was located to the southwest of Novosibirsk region, not far from the Chany lake at a permanent station of the Biological Institute SB RAS about 500 km from the first two observation points. This means that the observations were performed on a regional scale.

3. EXPERIMENTAL TECHNIQUE

The aerosol particle concentration was measured in terms of total light scattering by aerosol particles using a nephelometer. The nephelometer is intended for measuring the integral light scattering from aerosol particles entering an illuminated volume. For measurements we used a device FAN-A produced in Zagorsk. A halogen lamp of 40 W power served as a

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light source. The image of a lamp filament was focused to the counting volume, light scattering was observed at an angle of 45° using a photomultiplier. Then the light passed through a yellow-green correcting light filter. The instrument signal value is given in relative units where as a scale the Rayleigh scattering from pure air was used, the latter being measured with the same instrument.

To arrive at correct conclusions based on the measurement data using a nephelometer we need to estimate the size of particles contributing considerably to the instrument readings. Using the data on the instrument spectral sensitivity we first calculated the relative light scattering by aerosol particles depending on their size. As a scale we took the Rayleigh scattering by pure air without aerosol particles. Calculations were made based on the Mie theory using the BHMIE program given in Ref. 2. The complex refractive index used in the calculations was taken from Ref. 3 and corresponded to the so-called "powderedB particles. The real part of the complex refractive index was equal to 1.53 and the imaginary part was equal to $8 \cdot 10^{-3}$. The results of calculations can be described as follows. In the size range $r < 0.1$ μm the light scattering depends on the particle size as r^6, that corresponds to the Rayleigh approach. For particles with $r > 1$ μm the light scattering asymptotically approaches the dependence r^2 corresponding to the geometric approximation. However, on the average, in the 0.2–1 μm size range the dependence of light scattering on the radius is of the form $r^{1.5}$.

For the convenience of calculations we used the following empirical dependence of light scattering on the radius:

$$f(r) = \frac{[1 + (r/R)^{1.5}]}{[1 + (R/r)^6]},$$

where $R = 0.16$ μm. This analytical expression approximates the Mie theory calculations for particle size interval 0.01 to 2 μm within 13% accuracy.

In order to identify the size interval of particles contributing into the nephelometer particular signal one has to multiply some typical size-distribution of aerosol by the above empirical function of the nephelometer sensitivity to particle size. The size-distribution function for a continental aerosol shown in Fig. 1 has been taken from Ref. 4 and describes that of continental aerosol with an admixture of industrial (urban) aerosol. The same distribution multiplied by the nephelometer size-sensitivity curve is shown in Fig. 1 by the dashed line. As seen from this figure, particles of 0.1 μm radius give the main contribution into the formation of this instrument response to light scattering. This conclusion will keep true even if we use, in our calculations, other size spectra of atmospheric aerosol.

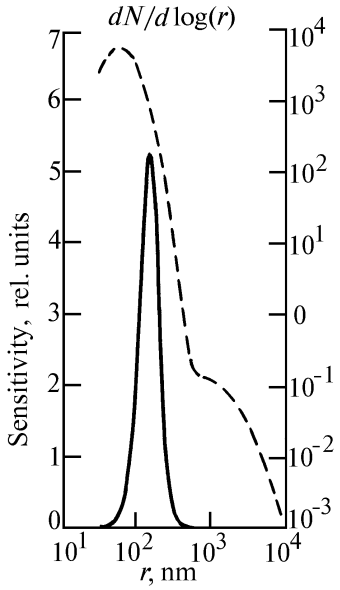

FIG. 1. The nephelometer signal amplitude as a function of particle size.

4. RESULTS AND DISCUSSION

Results of measurements of the diurnal mean variation of submicron particle concentration in terms of aerosol scattering for different seasons in Akademgorodok are given in Fig. 2. As can be seen from the figure, for the data of diurnal variations in summer the two maxima with a more pronounced evening maximum are typical. In winter the mean concentration value increases and the diurnal variation becomes smoother.

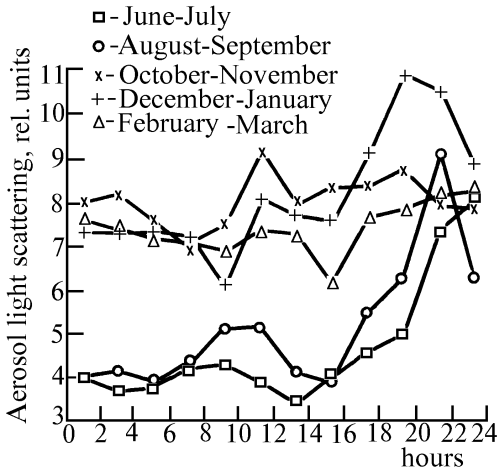

FIG. 2. Diurnal mean variation of light scattering by aerosol particles (σ–1) depending on the season in Akademgorodok.

Below we list the processes affecting the submicron particle concentration and the value of light scattering measured.

1. Formation of submicron particles in the transformation processes of the gas-to-particle type. These processes, as a rule, are of photoinitiated character being connected with the solar radiance value. Hence, it follows that the particle concentration increases at the daytime as compared with that at nighttime.

2. The air mass exchange is observed between the atmospheric boundary layer where the aerosol formation processes mainly occur and the higher atmospheric layers with lower aerosol concentration. These processes lead to the decrease of aerosol concentration in the middle of a summer day when the intensity of such an exchange is highest.

3. Water coating of aerosol particles is observed with the increase of air humidity resulting in the particle size growth and, hence, in the increase of light scattering efficiency by particles.

In winter, as compared with summer, the intensity of atmospheric vertical mixing decreases and the particles are accumulated in the atmospheric boundary layer. This results in the increase of the average value of aerosol scattering (Fig. 3). Besides, since in summer the above mixing has a pronounced diurnal dynamics, in winter its influence on this dynamics of aerosol concentration should be markedly weakened.

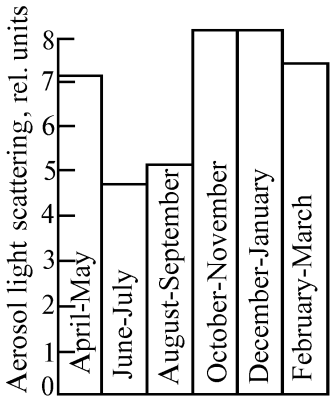

FIG. 3. Averaged values of light scattering depending on season.

In June 1994 the complex expedition was conducted for studying characteristics of atmospheric aerosol in Novosibirsk region and for investigating in detail diurnal dynamics of light scattering in summer. The measurements were made simultaneously in Akademgorodok, Klyuchi and in the vicinity of the lake Chany. These measurements should reveal the natural cycles of the number density variations and their change under the effect of moderate anthropogenic activity.

Figure 4 shows the averaged over one month diurnal cycles of aerosol scattering measured at the three measuring points. As one can see from the figure, the concentration and its dynamics, measured in Klyuchi and near the lake Chany, are similar (in their diagrams given in Fig. 5 in a magnified scale). At the same time, the light scattering diurnal variation measured in Akademgorodok is slightly different from those at two other sites, especially for evening hours. The similarity of diurnal variations for the stations Klyuchi and the lake Chany is striking (Fig. 5) if we take into account the fact that the measurements were carried out at a distance of 450 km. For the above stations we have to do with a "cleanB situation and natural diurnal dynamics of aerosol concentration.

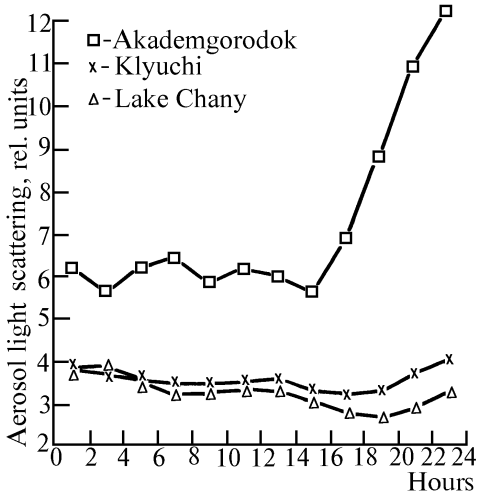

FIG. 4. Results of measurements in June 1994.

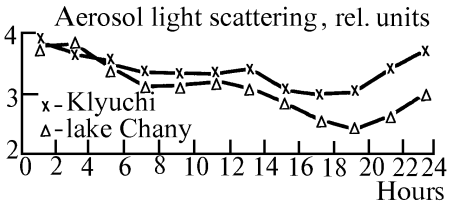

FIG. 5. The large-scale diagram of Fig. 4 for Klyuchi and the Lake Chany.

In Akademgorodok we observed the days when the diurnal mean value of aerosol scattering did not exceed the monthly mean values for the stations Klyuchi and the lake Chany. If the averaging is made based on these days, its daily dynamics is of the form as in Fig. 6. These days can be named as background when due to mass transfer from relatively ecologically clean regions the anthropogenic effect is minimal. As one can see from Fig. 6, the dynamics during the background days in Akademgorodok is similar to the daily mean dynamics at background stations with pronounced concentration dips at 7:00 and 12:00-16:00.

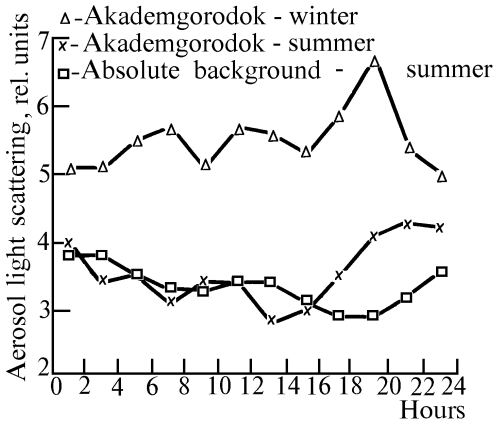

FIG. 6. Average selected values for summer and winter in Akademgorodok.

The observed diurnal variation can be explained qualitatively on the basis of known concepts of the main mechanism of formation and transformation of aerosol particles. The below explanation will then be used for constructing a numerical model, describing the diurnal variation of the aerosol concentration.

Before proceeding to an extended consideration of the form of diurnal variation of aerosol scattering, some comments should be made concerning the reliability of the data obtained. Figures 2-6 show that the relative amplitude of the averaged diurnal variations is small and, in addition, there is an uncertainty when determining the value of light scattering itself. This uncertainty comprises the instrument measurement errors and random fluctuations of light scattering in the atmosphere. Omitting the regular errors of the instrument resulting in constant overestimate or underestimate of the values measured, it can be shown that random errors are only several per cent of the value measured, and these errors cannot affect the reliability of conclusions on the diurnal variations. As to the aerosol concentration fluctuations, they are about 20 percent for the size range where the measurements are carried out, and they are compared with the amplitude of diurnal variations of light scattering. However, the complication of the problem occurs in calculating the value of the data standard deviation.

For example, in averaging the data, not all of those may be considered independent. An interesting example is given in Fig. 5. The amplitude of data variations for each of the measured daily mean variations is sufficiently large when calculating by the known formula for standard deviation. However, if each of the averaged curves is considered to be an independent measurement (and that is the case) the relative variation of data will be insignificant.

5. BASIC PRINCIPLES OF CONSTRUCTION OF A MODEL DESCRIBING DIURNAL DYNAMICS OF THE AEROSOL CONCENTRATION

First we divide arbitrarily all the aerosol particle size range with $r < 1$ μm into 6 subintervals.

Table I presents the radii of selected size intervals for estimates, the average particle lifetimes for the lower troposphere in these intervals depending on the radii,[3] and, based on the lifetime, possible values of diurnal variations of the number density, contribution to light scattering for the typical particle size spectrum of continental aerosol and possible amplitude of diurnal variations of light scattering by particles of one or another size.

TABLE I.

No.	Radii, nm	Lifetime, days	Relative amplitude of diurnal variations, %	Contribution to light scattering, %	Amplitude of diurnal variations, %
1	1–3	0.02–0.15	100	0	0
2	3–10	0.15–1.4	100–50	0	0
3	10–30	1.4–5.9	50–15	0	0
4	30–100	5.9–9.4	15–10	13	2–1
5	100–300	9.4–9.9	10	81	8
6	300–1000	9.9–9.4	10–15	5	1
	> 1 μm	< 9	> 15	1	0

The lifetime of particles was calculated by the semiempirical expression by Jaenicke[3]:

$$\frac{1}{T} = \frac{1}{C}\left\{\left(\frac{r}{R}\right)^2 + \left(\frac{R}{r}\right)^2\right\} + \frac{1}{T_{\text{wet}}}, \qquad (1)$$

where T is the lifetime of aerosol particles in the atmosphere; C is the constant equal to 4 years; r is the particle radius; R is the radius of particles with the largest lifetime (0.3 μm), T_{wet} is the largest lifetime determined by moist washing out of particles. The parameter T_{wet} is less determined and is equal, on the average, to 10 days for the lower troposphere.[3] The amplitude of possible diurnal variations was estimated using the expression for potential decrease of the number density:

$$N(t) = N(0)\exp(-t/T). \qquad (2)$$

The contribution coming from each fraction to the light scattering was determined based on the average size spectrum of Siberian aerosol determined in Ref. 5. The results, given in the third column of the table, do not vary essentially with variation of the particle size spectrum parameters within physically reasonable limits.

The last column of the table shows the contribution from each fraction to the relative amplitude of diurnal variation of aerosol scattering. The basic conclusion is that practically all the contribution to the light scattering variation is introduced by the aerosol fraction of 0.1–0.3 μm size. For $T_{wet} = 10$ days from Eq. (1) the amplitude of a given variation will be of the order of 10 percent. It should be noted that the value of T_{wet}, taken from Ref. 3, is an estimate, in this case an average, for the lower troposphere. This value may vary depending on weather conditions, the character of underlying surface, etc. The decrease of T_{wet} should also be expected in the surface layer since the particle removal and, hence, the particle lifetime become more effective. When decreasing T_{wet} down to 1.5 days the value of the diurnal variation amplitude increases up to 50 percent, in this case the contribution from the 0.1–0.3 μm fraction even increases.

First we try to understand the light scattering diurnal variation in cold season (November–March) when the vertical mixing is difficult and the variation of relative humidity during a day is small.[6] During this season for the conditions of cold winter of continental climate the diurnal variation of the turbulent diffusion coefficient is also small.[6]

The results of the 1994 summer complex expedition demonstrated that for Akademgorodok, where the basic measurements were carried out, the weather conditions are possible when both the atmospheric aerosol corresponds practically to the background one and the conditions with moderate anthropogenic influence. To exclude these conditions from the results of measurements the daily values were removed when the value of daily light scattering exceeded the average one, then the rest measurements (about 50 percent) were averaged again. The daily mean variation obtained is shown in Fig. 6.

The first main conclusion that can be drawn from Fig. 6 is the existence of the winter daily mean variation. In this case, as in summer, we observe an increase of light scattering during daytime, showing the possibility of photochemical gas-particle transformation to occur.

The second important fact is the fast (during several hours) decrease of light scattering after the evening peak down to the night level. We can explain this fact if we assume an additional particle sink to the surface or particle ascend to the upper atmospheric layers. First we estimate the contribution of turbulent diffusion to aerosol emission from the surface layer to higher layers. From the monograph by Laikhtman[6] the turbulent diffusion coefficient in the atmospheric boundary layer may be written as

$$K = k_1(h/h_0) \ . \tag{3}$$

Thus for winter $k_1 \approx 0.1$ m^2/s at $h_0 = 1$ m and it practically does not vary during a day. Assume that we conduct the measurements in the surface layer with the depth H. As H the depth of the internal mixing layer (IML) can be taken, equal approximately to 200–400 m. In this case

$$\frac{d\,n}{d\,t} \sim -\frac{d\,n}{d\,z}\,K\,\frac{d\,n}{d\,z}\,, \tag{4}$$

where n is the aerosol number density; z is the vertical coordinate. The vertical profile of aerosol number density can be written as[7]

$$n = n_0 \exp(-z/L), \tag{5}$$

where $L \sim 1000$ m (Ref. 7). Figure 6 shows that after the evening peak the number density decreases down to night quasistationary level during the time τ, which is several hours. Hence

$$\frac{1}{\tau} \sim \frac{k_1}{1\text{m}\,L} + \frac{H\,k_1}{1\text{m}\,L^2} \tag{6}$$

or

$$\tau \sim 10^3\ \text{m}^2/k_1 \sim 10^4\ \text{s} \sim 3\ \text{h}. \tag{7}$$

Thus, the decrease of number density from the evening peak to the night level can be explained by the particle emission from the surface layer to higher atmospheric layers due to the turbulent diffusion. It should be noted that the particle concentration will decrease until the height gradients of concentration equalize, if only inside the IML, i.e., the eddies from the higher atmospheric layers will bring air masses with the same aerosol concentration as from the lower layers. Thus, the variation of light scattering, owing to the turbulent mixing in the surface layer, can be written as:

$$\frac{\partial\sigma}{\partial\tau} = -\,(\sigma - \sigma_0)/\tau \ .$$

In Ref. 7 one can find a comparison of different literature data on particle sink to the surface in the surface layer. The sink velocity turned out to be no higher than 10^{-2} cm^{-3}/s for particles in the size spectrum under study. At $n \sim 10^2$ cm^{-3} the washing out time is also within 10^4 s. To answer the question, which of the mechanisms (turbulent diffusion in upper layers or precipitation to obstacles at the underlying surface) dominates, let us consider the diurnal mean

summer variation of aerosol concentration. Figure 6 shows that in summer the concentration decrease after reaching the evening maximum down to night level occurs approximately during 7 hours as compared to 4 hours in winter. The data given by Laikhtman[6] show that in summer the value of turbulent diffusion coefficient has a pronounced diurnal variation and is equal to 0.05 for the nighttime, that is, we should expect a twofold decrease of the velocity of turbulent ascend as compared with the winter night one (by day, however, this coefficient is larger in summer rather than in winter). This fact indicates that the upward turbulent diffusion is the principal mechanism for decreasing the number density at night when the evening maximum is achieved. If the dry precipitation on the roughness of surface is the principal mechanism resulting in the number density decrease, the faster fall of the above density at night in summer as compared with that in winter should be expected.

Reactions in gas phase resulting in aerosol formation can be schematically presented as

$$\text{GP} + \text{solar radiation} \Rightarrow \text{intermediate products} \Rightarrow \Rightarrow \text{aerosol particles (AP)}, \quad (8)$$

where GP denotes the gas precursors of aerosol particles. There may be many small chains of that kind leading to formation of aerosol particles of different chemical composition. If, however, the stage of the initiator photodissociation is a limiting stage of the process, the rate of aerosol formation will be approximately equal to $f(t)$, which is the time dependence of the solar radiation intensity. One can write for the mass of generated aerosol

$$\frac{\partial M}{\partial t} = M_0(t)\, f(t), \quad (9)$$

where $M_0(t)$ is the proportionality coefficient, depending on time as well. For the preliminary estimates, this coefficient can be taken to be constant in time, then it can be considered in more detail when refining the model.

First, the aerosol mass occurs as very fine particles coagulating with larger ones from the 0.1–0.5 μm fraction being of interest for us. The time of such a transfer, T, is determined by the initial concentration of particles observed and the constant of coagulation rate that is for normal conditions in the lower troposphere 2 or 3 hours. Thus the variation of aerosol mass in the size range being studied can be written as:

$$\frac{\partial M}{\partial t} = M_0\, f(t + T). \quad (10)$$

This mass variation can be formally presented as the particle size variation of the fraction being considered at a constant number density or as the number density variation at a constant radius. The exact solution of the problem calls for the solution of the set of the Smoluchowskii coagulation equations that is thus far impossible at a given stage of the model development. However, for estimates we take the version of the number density variation at constant radius. Then for light scattering we have

$$\frac{\partial \sigma}{\partial t} = C_0\, f(t + T) - (\sigma - \sigma_0)/\tau \quad (11)$$

for winter conditions. In these approximation the light scattering σ is proportional to n, and, if we know the diurnal variation of solar radiation intensity, the diurnal dynamics of the winter light scattering can be determined. If the sunrise and sunset times are known (for January 1st – 9:00 and 16.30, respectively) we can write $f(t)$ approximately as:

$$f(t) = f_0 \begin{cases} \sin((t - 9)\,\pi/\,7.5), & \text{for } 9 < t < 16.5, \\ 0, & \text{for } 0 < t < 9,\ 16.5 < t < 24. \end{cases} \quad (12)$$

Using $T = 3$ hours, $\tau = 3$ hours, and $\sigma_0 = 6$ for winter conditions, the diurnal variation is obtained which is given in Fig. 7. This figure shows also the winter diurnal variation presented in Fig. 6. As can be seen from the figure the formulas cited describe the occurrence of the evening peak. However, diurnal variation of light scattering turned out to be "blurred". This depends upon the following circumstances.

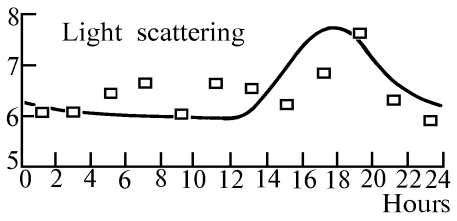

FIG. 7. Comparison of the daily mean variation for winter: measured in Akademgorodok and calculated numerically.

First, the number density of photoactive gases, resulting in aerosol formation at photodissociation, is not constant and varies during a day as well. For example, if the sink of such impurity is mainly determined by photodissociation and the emission rate does not vary significantly during a day, then the given impurity will be accumulated in the surface layer reaching its maximum before the sunrise, hence, high rate of aerosol formation is observed immediately after the sunrise thus accounting for the maximum between 10.00 and 14.00. This possible version of sequence of events is described in Ref. 5, where it is shown using a counter for measuring the condensation nuclei that the

morning nucleation peak is manifested every one or two hours after the sunrise. However, if the aerosol mass transfer from nucleation fraction to accumulation one takes place in a time interval of two hours, the morning nucleation peak should be expected in the nephelometric measurements at 13.00.

The above-mentioned studies describe the ways of the model development. Using this model we may obtain numerical results. A closer look at the reaction mechanisms is called for in the gas phase to determine a closer time dependence of the aerosol formation intensity.

When investigating the diurnal mean variation of aerosol scattering in Akademgorodok in winter and in summer, a strange peak of aerosol number density was observed at 5:00 in the morning. This peak, being of local nature, was not observed at the distant stations, and to explain this phenomenon further studies are necessary including element and chemical aerosol composition.

In spite of the above disadvantages, the described assumptions on diurnal variation of aerosol scattering enable one to describe correctly the seasonal transformation of diurnal variation from winter to summer. In summer, except for the above reasons of aerosol number density variations, in addition to variation in the character and intensity of sources, there are the following reasons:

1) increase of the daytime duration;

2) diurnal behavior of the turbulent diffusion coefficient;

3) diurnal behavior of humidity.

In the near future the diurnal behavior of humidity should be taken into account, however, nowadays the following facts can be noted. The evening humidity increase must result in the light scattering increase, which, in its turn, decreases only in the morning hours, after the sunrise and decrease of the relative humidity. In our measurements this dependence is not observed or it is masked by other processes. The function of particle size r dependence on humidity f is of monotonic character[7]:

$$r = r_0 (1 - f)^{-\varepsilon}.$$

The humidity itself varies also monotonically during 24 hours from 50% at daytime up to 80% at nighttime for a mean summer day. This means that the peak structure on the background of monotonic dependence will not vary. This is because the nephelometer (with counting volume) was inside the laboratory during measurements and its body was warmed up that could result in a decrease of the influence of relative humidity of the outer atmosphere on measurement results. By virtue of the above-mentioned reasons at this initial stage of development the humidity model was excluded from consideration.

The increase of the daytime duration in summer as compared with that in winter must result in the enhancement of the period of growth of aerosol number density (earlier morning growth, later evening peak). This fact is supported by curves in Fig. 6. As the summer minimum of the turbulent diffusion coefficient at nighttime is less than in winter (see the text given above), the evening peak of light scattering must be broader then in winter. This fact is confirmed by our observations. Besides, the large minimum of summer light scattering at 12:00–16:00 well correlates with the daily maximum of the turbulent diffusion coefficient.

Thus, in our opinion, the above scheme explains qualitatively and semiquantitatively the regularities of the submicron aerosol number density variation during 24 hours. It also shows the ways of further development and improvement of the model for obtaining a possibility for numerical simulations of the daily dynamics of the submicron aerosol number density.

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