

## Seasonal Changes in Diurnal Variation of Mass Concentrations of the Submicron Fraction of Continental Atmospheric Aerosol of Remote Areas

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### Abstract

A semi-empirical model is proposed for the description of seasonal changeability of the diurnal variations of the accumulation mode of continental atmospheric aerosol of remote territories. The data of synchronous measurements of diurnal variations of the mass concentration of accumulation mode of AA in winter and in summer are reported, obtained in two observation sites in the Novosibirsk Region, the distance between them being 450 km. Results of calculations according to the theoretical model are in good agreement with the experimental data.

### INTRODUCTION

The problem of photochemical smog formation has got the status of a fundamental research problem in the second half of the previous century. In order to solve this problem, large-scale investigations into the physics and chemistry of atmosphere were initiated in the 50-ies of the 20s century in California (USA) [1]. A large amount of experimental data on the size distribution and chemical composition of atmospheric aerosol (AA) particles of this region had been accumulated by the end of 70-ies; a generalized temporal dependence of the AA particle size distribution was proposed within a broad range from  $10^{-3}$  to  $10^2$   $\mu\text{m}$  [2, 3]. Investigation of the chemical composition of AA particles of different sizes allowed formulating the regularities concerning main sources, as well as mechanisms of formation and transformation of their disperse composition and concentration. The set of these results provided a distinct difference between the two fractions: submicron ( $d < 1$   $\mu\text{m}$ ) and coarse ( $d > 1$   $\mu\text{m}$ ). The major contribution into the submicron fraction mass is made by the aerosol mass formed in the photochemical gas-to-particle conversion.

Now it becomes clear that for solving the problem connected with the formation of photochemical smog of anthropogenic and natural origin, it is necessary to investigate the regularities of temporal variations of microphysical characteristics (disperse composition and concentration) of the AA submicron fraction and to study chemical composition of minor gaseous admixtures which are precursors of the disperse phase [5, 5a, 13–15]. However, the dynamics of transformation of the size distribution of AA submicron fraction includes many stages and is complicated, so the intensive investigation of these regularities goes on till present [5, 5a]. Other urgent problems of the physics of atmosphere are connected with the investigation of the role of the AA submicron fraction in the formation of clouds and fogs, and with the evaluation of its effect on optical weather and climate [16–32].

The data on the size distribution of submicron particles determining different types of optical weather were obtained by measuring various optical characteristics. An important result of these investigations was the model of kinetically determined parameters of the accumulation mode of AA under stationary conditions [17, 33, 34]. This theory is semi-

empirical; it is based on the measurements of optical characteristics of atmospheric aerosol. Because of this, correct data on the size distribution of AA can be obtained only for the accumulation mode of submicron fraction with  $d > 0.1 \mu\text{m}$ . For smaller particles ( $d < 0.1 \mu\text{m}$ ), this model allows obtaining qualitative estimations; this is why the data on the formation of the nucleation fraction (microdisperse one) are of qualitative character; as a rule, in order to explain spectral dependencies, the information on chemical composition of atmospheric aerosol is necessarily attracted [35–39], as well as the data on the changes of optical characteristics of aerosol under temperature action (changes of relative humidity). Using this procedure, temporal changes of the parameters of submicron aerosols were measured.

The present investigation is a continuation of earlier experimental and theoretical investigations aimed at building up a dynamic model of the formation and transformation of the particle size distribution for the submicron fraction of AA [4, 6–9]. First of all, this is a theoretical and experimental investigation of the diurnal variation of numerical concentration and dispersed composition of the submicron fraction of atmospheric aerosols of Siberia. Measurements of the dispersed composition of AA within the size range of  $10^{-3}$ – $10^2 \mu\text{m}$  in two sites of Siberia (Novosibirsk district, and Lake Baikal region) demonstrated that particle size distribution is almost identical in both sites [4]. Since the distance between the two observation sites is about 1300 km, this similarity means that the formation of AA particle size distribution in Siberia is regulated by global processes. Experimentally measured particle size distribution contains three modes, which corresponds to the atmospheric aerosol of remote continental territories, according to the classification of Whitby [3].

The submicron fraction of the AA of remote continental territories in summer is formed mainly by means of the photochemical gas-to-particle conversion of chemically active gas-phase precursors [5, 5a], which is confirmed by the results of experimental investigations in Siberia [4, 6]. A specific feature of the diurnal variation of numerical AA concentration

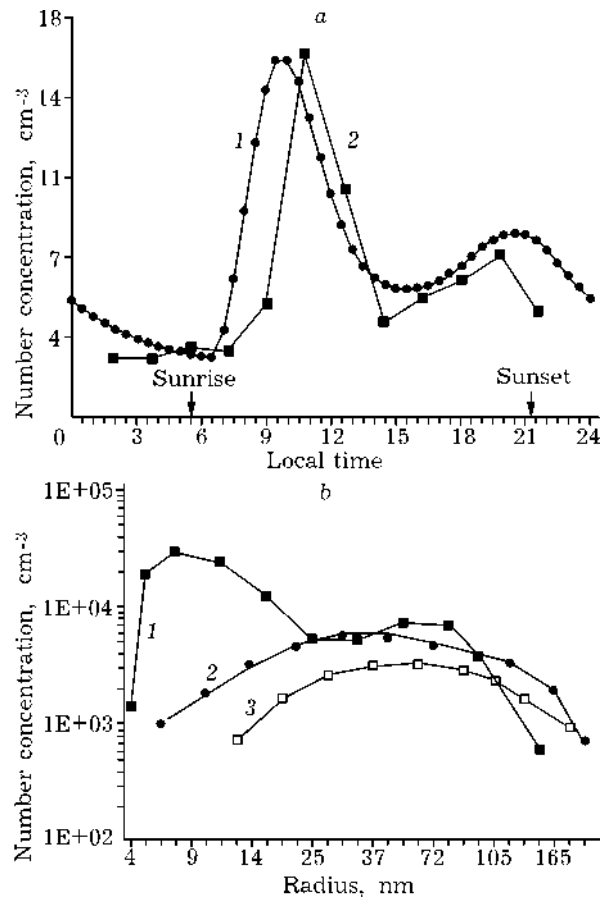


Fig. 1. Diurnal and seasonal dynamics of the dispersed composition and concentration of the submicron fraction of atmospheric aerosol: *a* – model (1), experiment (2); *b* – morning maximum (1), daytime (2), nighttime (3).

(Fig. 1, *a*, curve 2) is the existence of two peaks, in morning and in evening. Since gas-to-particle conversion rate is proportional to the intensity of solar radiation and to the concentration of gaseous precursors, this must be taken into account for the theoretical description of experimental data, along with taking account of the diurnal dynamics of the thickness of the boundary atmospheric layer. The latter process affects both the temporal change of the concentrations of chemically active gaseous admixtures and the concentration of newly formed aerosol particles.

The results of the calculation of the diurnal cycle of the submicron AA fraction according to the model developed in [4, 6, 8] are shown in Fig. 1, *a* (curve 1). The mechanism of photochemical formation of the submicron fraction of AA from gaseous precursors is also confirmed by the experimental data on the diurnal

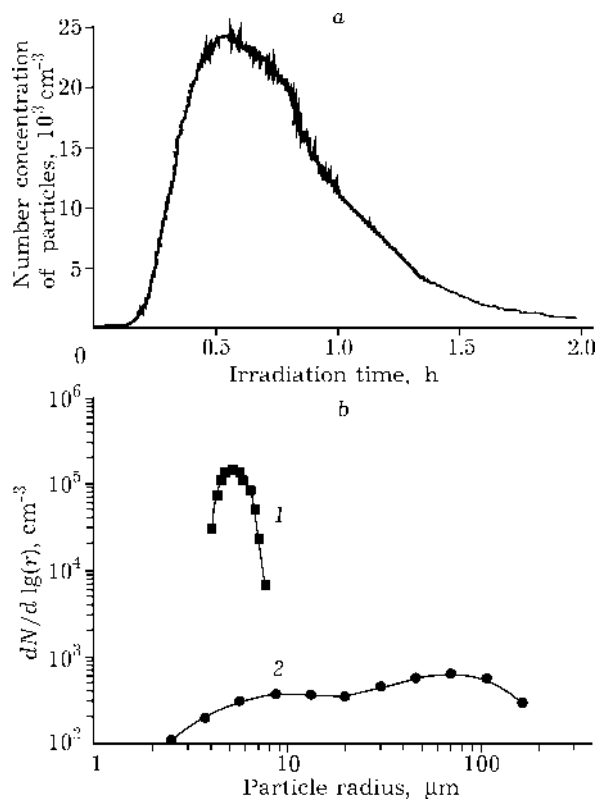


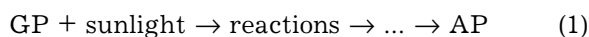
Fig. 2. Results obtained in smog chamber: 1 and 2 - after irradiation for 0.5 (1) and 2 h (2).

nal dynamics of the size of submicron particles (see Fig. 1, b) and the data obtained in a smog chamber [4, 6] (Fig. 2). In particular, during the morning maximum of the number concentration of AA (see Fig. 1, a, curve 2), the particle size distribution contains two modes. The first maximum is located at  $\sim 10$  nm radius, while the second is at  $\sim 100$  nm (see Fig. 1, b, curve 1). In the daytime, rather broad single-mode particle size distribution is observed (see Fig. 1, b, curve 2); a narrower single-mode distribution is observed at night (see Fig. 1, b, curve 3). These features of the diurnal variation of particle size distribution, as well as the results obtained in smog chamber (see Fig. 2), point to substantial effect of particle coagulation on the regularities of temporal evolution of AA particle size distribution within the diurnal cycle. In addition to clarifying the mechanism of temporal variability of the particle size distribution of submicron aerosol fraction formed during gas-to-particle conversion, an experiment in smog chamber allows us to estimate the characteristic scale of "pumping" the nucleation mode into accumulation one.

The accumulation mode ( $0.1-1 \mu\text{m}$ ) is the most long-lived size fraction of AA; its characteristics are most stable in time. Because of this, in order to study slow changes of mass concentration in seasonal cycles of atmospheric processes, it is preferable to use rather sensitive and specific measurement method. One of rather simple methods allowing one to investigate the dynamics of the mass concentration of accumulation mode is nephelometry. Earlier this technique was used to study diurnal dynamics in other regions [40-44]. These experiments also revealed the diversity of forms of the diurnal cycle of the numerical concentration of AA submicron fraction depending on weather type, season, as well as observation site (background territory, urban or suburban area). In order to explain the daytime minimum, the hypothesis was proposed concerning the role of convective mixing in the boundary atmospheric layer. However, only qualitative interpretation of the observed regularities was proposed; no theoretical models were presented to describe specific features of the diurnal dynamics (the value and time of extreme points on experimental curve, and the dependence of these features on external conditions).

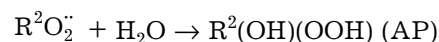
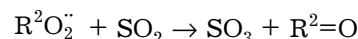
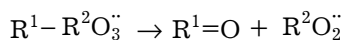
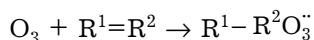
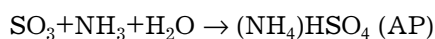
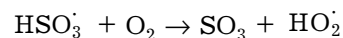
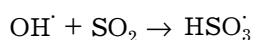
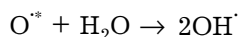
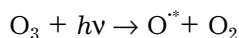
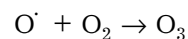
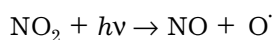
As it was mentioned above, the author previously proposed the dynamic model to describe diurnal variations of the number concentration and size distribution of the submicron AA fraction [4, 6, 8]. This became possible only due to the fact that the change of number concentration and size distribution of the submicron AA fraction in diurnal cycle is determined by the regularities of the behaviour of the nucleation mode ( $d < 0.1 \mu\text{m}$ ). The mass concentration of the particles of continental aerosol of remote territories, as it follows from the model [3], is determined by the characteristics of not only the accumulation mode by the coarse mode. It is demonstrated experimentally that in summer the mass fraction of the coarse fraction of AA in Siberia is up to 85% [45]. The submicron fraction accounts for about 15%. In winter, because of the decrease of the emission of soil erosion component, the mass fraction of coarse fraction decreases [45]. However, due to the global transfer of air, an insignificant fraction of the submicron parti-

cles of soil erosion nature can also participate in the formation of the accumulation mode of AA in continental territories. This contribution can be estimated using the data on the chemical composition of the accumulation fraction of AA (see [46]). If we assume that the gas precursors (GP) of the aerosol particles can be converted into aerosol particles (AP) by a sequence of photochemically initiated reactions, the reactions in the gas phase can be written down as the following scheme:



The mean reaction rate constant of the sequence depends on the quantum yield of the GP photodecomposition and on the intensity of sunlight. It is assumed that the photodecomposition of the initiating agent is the limiting stage of the process.

The following examples of the sequences of atmospheric reactions can be listed:



These reactions [49] lead to the formation of low-volatile components like sulphates ((NH<sub>4</sub>)HSO<sub>4</sub>) or hydroperoxides (R<sup>2</sup>(OH)(OOH)) which can be condensed in the atmosphere forming aerosol particles. In these reactions, R<sup>1</sup> and R<sup>2</sup> mean hydrocarbon residues of more complicated organic compounds.

In this reaction sequence, two components, playing key role according to modern notions, can be stressed. They are ozone (O<sub>3</sub>) and sulphur oxides. The special role of ozone is connected with the fact that its decomposition re-

sults in the formation of excited oxygen, which can react with water vapour leading to the formation of OH radicals; in turn, they are likely to be the major active basis for the atmospheric processes, since they oxidize almost all the atmospheric admixtures.

Sulphur oxidation is a very important process from the viewpoint of aerosol formation. The reason is that the compounds of sulphur (IV) are present in the atmosphere mainly in the gaseous state, while the compounds of sulphur (VI) are much less reactive; under the conditions of competition between numerous atmospheric reactions, this leads to preferable formation of sulphates with very low saturated vapour pressure. This means the formation of sulphate aerosol particles.

An alternative way of using ozone as oxidizer leading to the formation of aerosol particles is its participation in the oxidation of unsaturated hydrocarbons. This reaction mechanism may be probable over territories with large natural emission of unsaturated hydrocarbons. For example, plants emit rather large amount of terpenes, which are the products of isoprene dimerization or trimerization. These hydrocarbons are unsaturated (since they contain double bonds); their concentration can be substantial over territories densely populated with plants (for example, tropical or boreal forests).

The reaction of ozone with the double bond of unsaturated hydrocarbon leads to the formation of double radicals, the so-called Criegee radicals (R<sup>2</sup>O<sub>2</sub><sup>··</sup>). These radicals can in turn participate in the oxidation of sulphur, which leads to the formation of sulphate aerosol, as it has been already mentioned. At the same time, organic aerosol particles are formed, which are better known as the blue haze. Thus, intensive gas-phase emission from plant sources helps forming both organic and sulphate aerosol, which is considered mainly as anthropogenic one, since the major source of sulphur dioxide in atmosphere is believed to be the combustion of coal and oil. This example confirms the postulate stating impossibility to distinguish atmospheric aerosol by relating them to anthropogenic or natural origin.

Thus, even a very limited set of atmospheric chemical reactions leading to the formation of aerosol particles demonstrates the complexity

of the interpretation of experimental data for revealing the aerosol formation mechanism. In order to reduce the listed system of equations to the eq. (1), it is necessary that the initial stage of photodecomposition of the process initiator (for example, nitrogen dioxide) would be the limiting one.

In the model proposed in the present study, such detailed notions are not taken into account, because specific features of the nucleation and accumulation modes are modelled by their lifetimes. The lifetime of the nucleation mode is much less than a day, while that of the accumulation mode substantially exceeds 24 h [47]. This fact allows one to reveal the features of the behaviour of mass concentration of the accumulation mode within a day cycle as small deviations from the daily mean value. Physical processes leading to temporal change of the mass concentration are considered in such a manner that the numerical concentration of the accumulation mode is taken constant (this follows from the previous model developed by the author), while the mass concentration changes due to photochemical conversion of gaseous precursors which contribute in increasing numerical concentration of the nucleation mode. After this, small particles pass into the accumulation mode due to coagulation.

## EXPERIMENTAL

As it was demonstrated in [7], the major part of light scattering in Siberia is connected with the particles within size range 0.1–0.3  $\mu\text{m}$ . Within this size range, the sensitivity of a FAN-A type nephelometer changes as a function of  $r^3$  [7]. This dependence was obtained by calculating light scattering by particles of different sizes according to the Mi theory [10] taking into account experimentally measured disperse composition of AA, illumination characteristics in the counting volume of FAN-A nephelometer, and spectral sensitivity of a photomultiplier. This means that the overall light scattering by AA is proportional to the mass concentration of the accumulation mode or submicron particles.

For the light scattering by aerosol, measured with a FAN-A nephelometer, one can calculate the proportionality coefficient for the mass concentration of the submicron fraction ( $M_{\text{sub}}$ ,  $\mu\text{g}/\text{m}^3$ ) [10], at the density of aerosol particles 1  $\text{g}/\text{cm}^3$ :

$$M_{\text{sub}} = 2.6(S - 1) \quad (2)$$

where  $S$  is the ratio of total light scattering to molecular scattering. The eq. (2) is obtained on the basis of the results of the Siberian AA size distribution determinations. Earlier, a similar equation was obtained from the experimental data by measuring the indicatrix of scattering by AA in the bottom layer of the atmosphere [48].

The results of experimental determination of seasonal variations in the diurnal cycle of light scattering by the submicron fraction of AA in Novosibirsk district are shown in Fig. 3 (the observation site was in Akademgorodok). In winter (curve 4), when the intensity of solar radiation and the duration of illumination are minimal, the daily peaks of light scattering which are proportional to the mass concentration of the accumulation mode exhibit less contrast; the time between them is the shortest. In warm season (curve 2), in comparison with winter, light scattering by aerosol is lower. This can be explained by the fact that in winter the height of the boundary layer is less than in summer, while the intensity of indust-

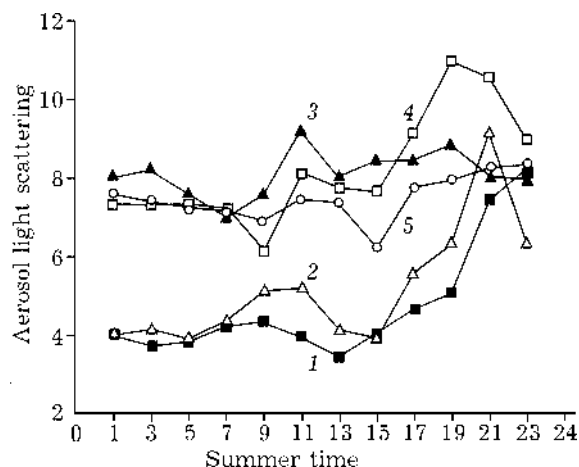


Fig. 3. Dynamics of diurnal and seasonal variation of the concentration of accumulation mode: 1 - June-July, 2 - August-September, 3 - October-December, 4 - December-January, 5 - February-March.

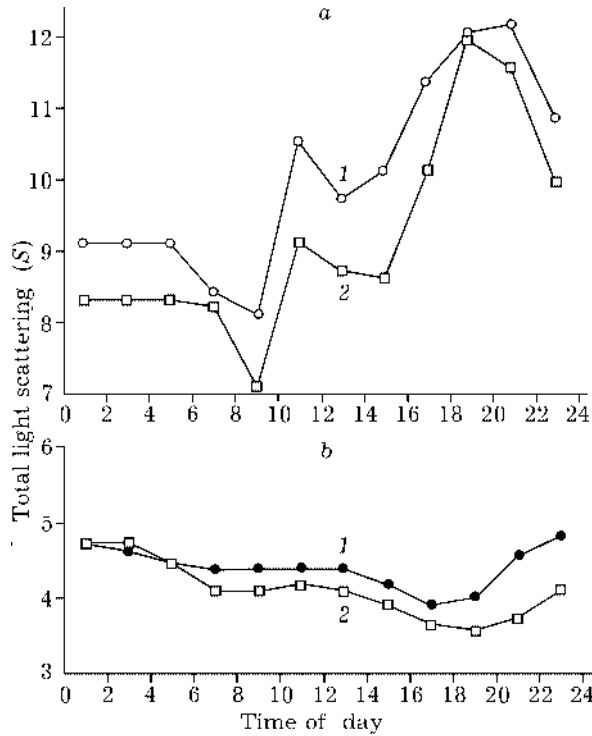


Fig. 4. Month-averaged diurnal variation of aerosol scattering measured in winter (a) and in summer (b) in the Novosibirsk district: 1 - Klyuchi, 2 - Chany.

rial emission of chemically active gases and aerosol particles is higher.

The results of simultaneous winter and summer measurements of the intensity of aerosol light scattering in different observation sites (Klyuchi settlement, and Chany), distance between them being 450 km, are shown in Fig. 4. One can see that at any season the diurnal variations in both sites are very much similar; however, the shapes of diurnal cycles of light scattering in warm season are substantially different from those in cold season.

#### THEORETICAL MODEL

Let us assume that the submicron fraction of the atmospheric aerosol is a mixture of two monodispersed fractions. In this case, the total number concentration ( $N$ ) will be a sum of the numerical concentrations of individual fractions:

$$N = N_1 + N_2 \quad (3)$$

Here  $N_1$  and  $N_2$  are number concentrations of the nucleation and accumulation modes, re-

spectively. The lifetime of the latter mode is about 10 days; these particles are uniformly spread over the boundary atmospheric layer. The lifetime of small particles of the nucleation mode depends on the rate of coagulation with larger particles and on the rate of transfer from the boundary atmospheric layer (BAL) [6].

$$\frac{dN_1}{dt} = k_{\text{coag}} N_1 N_2 = -\frac{1}{\tau_1} N_1 \quad (4)$$

According to [11], the coagulation constant ( $k_{\text{coag}}$ ) for the particles of the radius of 23 nm with the particles 0.16  $\mu\text{m}$  in size is equal to  $5 \cdot 10^{-8} \text{ cm}^3/\text{s}$ . According to the experimental data [4, 6], the numerical concentration of the accumulation mode is  $2600 \text{ cm}^{-3}$ . This makes  $\tau_1$  equal to approximately 3 h. The time within which aerosol particles are transferred from the BAL in summer is about 3 h. Since the mass concentration of the AA submicron fraction is proportional to the total light scattering ( $\sigma$ ), we can write down the following equation:

$$\sigma = \sigma_1 + (\sigma_0 + \sigma_2) \quad (5)$$

where  $\sigma_1$  is light scattering by the nucleation mode;  $\sigma_0$  is a constant constituent of light scattering by the accumulation fraction;  $\sigma_2$  is an additional contribution arising from the coagulation of small and large submicron particles. Temporal changes of light scattering of each mode can be represented as

$$\frac{d\sigma_1}{dt} = -\frac{1}{\tau_1} \sigma_1; \quad \frac{d\sigma_2}{dt} = +K_M \frac{1}{\tau_1} \sigma_1 \quad (6)$$

$$K_M = -\frac{d\sigma_2}{d\sigma_1} = +\left(\frac{d\sigma_2}{dM_2}\right) / \left(\frac{d\sigma_1}{dM_1}\right) \quad (7)$$

According to mass conservation law,  $dM_2/dM_1 = 1$ .

Theoretical consideration [7] demonstrates that the  $\sigma(r)$  function can be approximated by the following expression:

$$\sigma = \sigma_0 \frac{1 + (r/R)^{1.5}}{1 + (R/r)^6} \quad (8)$$

In eq. (8),  $R = 0.16 \mu\text{m}$ . Using (7) and (8) at  $r = 23 \text{ nm}$ , we obtain  $K_M = 84$ . This result shows that one should take into account an increase of light scattering by large particles when considering the coagulation of small particles with

larger ones. Because of this, let us represent  $ds/dt$  in the following form:

$$\frac{d\sigma}{dt} = C_0 f(t + T) - \frac{\sigma - \sigma_0}{\tau_2} Z(t) \tag{9}$$

where  $1/\tau_2$  is the amplitude of the rate of vertical transport in daily cycle ( $0.3 \text{ h}^{-1}$ );  $Z(t)$  is the rate of vertical daily mass exchange;  $C_0$  is a parameter connected with the amplitude of diurnal variations of the intensity of light scattering by aerosol. Using experimental data [12], one can determine all the parameters necessary for solving the eq. (9).

For winter, one can write down

$$T \sim 1 \text{ h}, f(t) = \begin{cases} 0, & \text{if } 0 \leq t \leq 9; 17 \leq t \leq 24 \\ \sin\left(\frac{\pi}{8}(t - 6)\right), & \text{if } 9 \leq t \leq 17 \end{cases} \tag{10}$$

$$Z(t) = 1 \tag{11}$$

because in winter the coefficient of turbulent diffusion remains almost constant during a day. The relative humidity of the atmosphere is also almost constant and close to 100 % in winter, so its effect on the variability of aerosol light scattering can be neglected.

In summer,

$$T \sim 2 \text{ h}, f(t) = \begin{cases} 0, & \text{if } 0 \leq t \leq 6; 22 \leq t \leq 24 \\ \sin\left(\frac{\pi}{16}(t - 6)\right), & \text{if } 6 \leq t \leq 22 \end{cases} \tag{12}$$

$$Z(t) = 1.2 + \sin\left(\frac{\pi}{2}(t - 1) + \frac{3}{2}\pi\right) \text{ (see Fig. 32 in [12])} \tag{13}$$

In summer one must take into account also the diurnal variation of relative humidity ( $f_{RH}$ ):

$$\sigma = \sigma_0 (1 - f_{PH}(t))^{-0.5} \tag{14}$$

$$f_{RH}(t) = \begin{cases} 0.75, & \text{if } 0 \leq t \leq 6 \\ 0.75 - 0.25 \sin\left(\frac{\pi}{18}(t - 6)\right), & \text{if } 6 \leq t \leq 24 \end{cases} \tag{15}$$

The results of theoretical calculations according to the proposed model are shown in Fig. 5; for comparison, experimental data are shown. One can see that the proposed model provides a good description of the features of diurnal dynamics of the mass concentration of AA submicron fraction in different seasons (the appearance of maxima and minima, their sea-

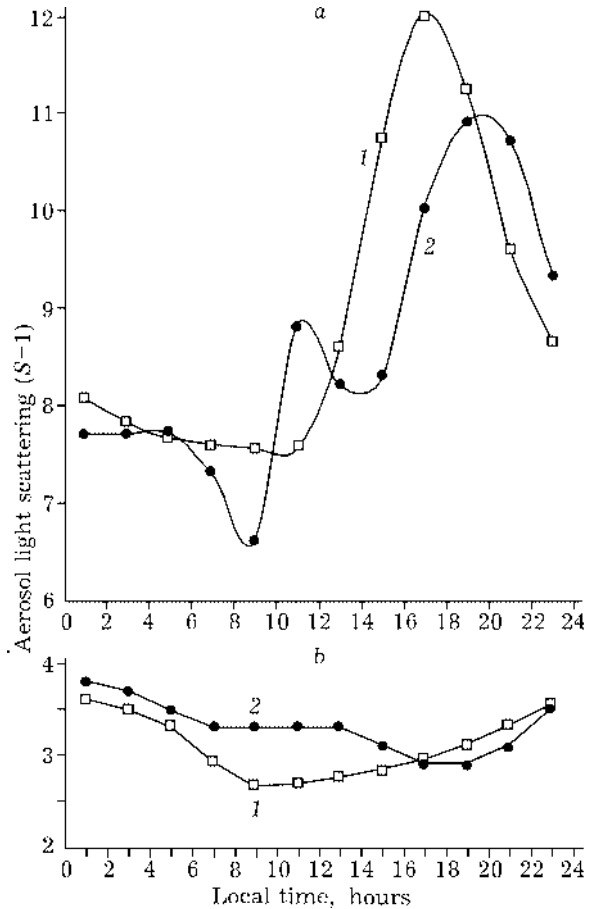


Fig. 5. Changes of the diurnal aerosol light scattering in winter (a) and in summer (b): 1 - model, 2 - experiment.

sonal variability, intensity of extreme points and the time of their appearance).

**CONCLUSIONS**

A model is proposed to describe seasonal variations of the mass concentration of the AA submicron fraction. The model takes into account photochemical formation of particles, coagulation of the formed particles, diurnal dynamics of the height of the boundary layer of the atmosphere, and relative humidity of the air. The comparison of calculation results obtained using this model with the experimental data shows that the model allows one to describe the time of the appearance of maxima and minima of the mass concentration of AA accumulation mode, their values, as well as seasonal variations of these major characteristics of the daily cycle. The proposed mo-

del is based on clearly established physicochemical processes. The parameters of the dynamics of these processes are defined functionally (they can be determined either experimentally or theoretically). This allows using the model to investigate the regularities of the diurnal dynamics of the characteristics of AA submicron fraction under other conditions, as well as to perfect and develop it.

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