Model of seasonal variations of the diurnal cycle of mass concentration of submicron fraction of the continental aerosol over remote territories

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A semiempirical model is proposed to describe the seasonal variations of diurnal cycle of mass concentration of continental accumulation-mode aerosol over remote territories. The experimental data of synchronous measurements of this cycle are presented; the measurements have been carried out in winter and summer seasons at two observation sites in Novosibirsk Region 450 km apart. The regularities revealed at the two observation sites are shown to closely agree in each season, but strongly change from season to season. The theoretical model calculations well fit the results of field measurements.

Introduction

The problem of studying photochemical smog formation had evolved into a fundamental scientific issue in the second half of the past century. To solve this problem, complex large-scale studies into the atmospheric chemistry and physics were initiated in the 1950s of the past century in California (USA).[1] In the late 1970s, abundant experimental data on size spectra and chemical composition of atmospheric aerosols (AA) had been accumulated for this region. After assimilation of the data on microphysical AA parameters, a generalized empirical dependence of AA size spectrum in a wide size range from 10^{-3} to 10^{2} μm was proposed.[2,3]

Study of the chemical composition of AA of different sizes has also made it possible to formulate ideas on the main sources and mechanisms of formation and transformation of microphysical AA characteristics responsible for AA variability. The set of these results showed quite clearly the fundamental difference of two size fractions: submicron ($d < 1$ μm) and the coarse ($d > 1$ μm) ones. The submicron fraction mainly consists of aerosol particles formed by photochemical conversion gas – dispersed phase. Now it is clear that, to solve the problem of photochemical formation of anthropogenic and natural smog, precisely the time variations of microphysical characteristics (disperse composition and concentration) of submicron AA fraction and chemical composition of minor gas admixtures, as precursors of dispersed fraction, must be studied.[5,5a,13–15] However, since the transformation of the spectrum of submicron AA fraction has multistage and complex dynamics, it is now actively studied.[5,5a] Other urgent problems of atmospheric physics are associated with the study of the role of submicron AA fraction in the cloud and fog formation and with estimation of its influence on the optical weather and climate.[16–32]

Data on disperse composition of submicron particles, determining different types of optical weather, were obtained during measurement of different optical characteristics. An important result of these studies has been the development of the model of kinetically caused parameters of accumulation-mode AA under stationary conditions.[17,33,34]

This is a semiempirical theory, based on experimental data on the optical characteristics of atmospheric aerosols. Therefore, correct qualitative data on disperse AA composition can be obtained only for accumulation-mode submicron fraction ($d > 0.1$ μm). For smaller particles ($d < 0.1$ μm), the theory provides qualitative estimates. Therefore, the information on kinetics of formation of nucleation-mode (microdisperse) fraction is qualitative in character and, to explain the spectral dependences, the information on chemical composition of atmospheric aerosols,[35–39] as well as data on changes of aerosol optical characteristics in response to temperature impact (effect of varying relative humidity) on atmospheric aerosol is generally used. This method was used to measure time variations of submicron aerosol parameters. This work extends upon earlier experimental and theoretical studies performed by the author and aimed at constructing a dynamical model of formation and transformation of size spectrum of submicron AA fraction.[4,6–8] Primarily, it presents the results of theoretical and experimental studies of dynamics of the diurnal variations of characteristics (number concentration and disperse composition – size spectrum of aerosol particles) of submicron fraction of atmospheric aerosols in Siberia.

The measurements of disperse AA composition in the size range from 10^{-3} to 100 μm, made at two sites of Siberian region (in Novosibirsk Region and near Lake Baikal) have shown that the particle size distributions at the two sites are very similar in shape (Fig. 1*a*).[4] The two observation sites are separated by about 1300 km, suggesting a global scale character of the processes responsible for formation of AA size spectrum in Siberia. The experimentally measured spectrum is a three-mode distribution, which, by the Whitby[3] classification, corresponds to atmospheric aerosols of remote continental territories.

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The submicron AA fraction of remote continental territories in the summer season is hypothesized to be formed by photochemical conversion gas – disperse phase of chemically active gaseous precursors.[5,5a] The experimental study[4,6] of dynamics of diurnal variations of size spectrum and concentration of submicron AA fraction in Siberia supports this hypothesis. The specific feature of diurnal variations of AA number concentration (curve *2* in Fig. 1*b*) is the presence of two peaks, in the morning and evening hours. The theoretical descriptions of experimental data must take into account the dependence of gas–particle conversion rate on the intensity of solar radiation and concentration of gaseous precursors, as well as the diurnal variation of planetary boundary layer (PBL) height. The latter process influences both the time variations of concentration of chemically active gas admixtures and the concentration of newly formed aerosol particles.

Figure 1*b* shows diurnal cycle of number concentration of submicron AA fraction, calculated by the model developed in Refs. 4, 6, and 8. The mechanism of photochemical formation of submicron AA fraction from gaseous precursors is also supported by experimental data on diurnal variations of size spectrum of submicron particles (Fig. 1*c*) and by data obtained in smog chambers.[4,6] In particular, in the period of morning maximum of AA number concentration (see Fig. 1*b*, curve *2*), the size spectrum of submicron particles is bimodal. The first maximum is in the radius range ~ 10 nm, and the second near ~ 100 nm (see Fig. 1*c*, curve *1*). The particle size distribution is quite wide and unimodal during a day (see Fig. 1*c*, curve *2*), and it is narrower and unimodal at night (see Fig. 1*c*, curve *3*).

These features of diurnal variations of size spectrum indicate that the particle coagulation influences substantially the time evolution of AA particle size distribution in diurnal cycle. The accumulation mode (0.1–1 μm) is the longest-lived size fraction of atmospheric aerosol, and its characteristics are most stable in time. Therefore, to study slow changes of mass concentration in seasonal cycles of atmospheric processes, it is preferable to use quite sensitive and specific measurement technique.

One of the fairly simple methods, allowing one to study time variations of mass concentration of accumulation-size mode, is the nephelometry, already used recently to study the diurnal variations in other geographical regions.[40–44] Those experiments also revealed the diversity of forms of diurnal cycle of mass concentration of submicron AA fraction for different weather types, seasons, as well as observation sites (background territory, urban and suburban locations). To explain the daytime minimum, it is hypothesized that the convective mixing plays an important role in PBL. However, aside from qualitative interpretation of observed dependences, no theoretical model was proposed to describe the specific features of diurnal variations (magnitude and timing of extrema on the experimental curve, as well as dependence of these characteristics of diurnal variations on the environmental conditions).

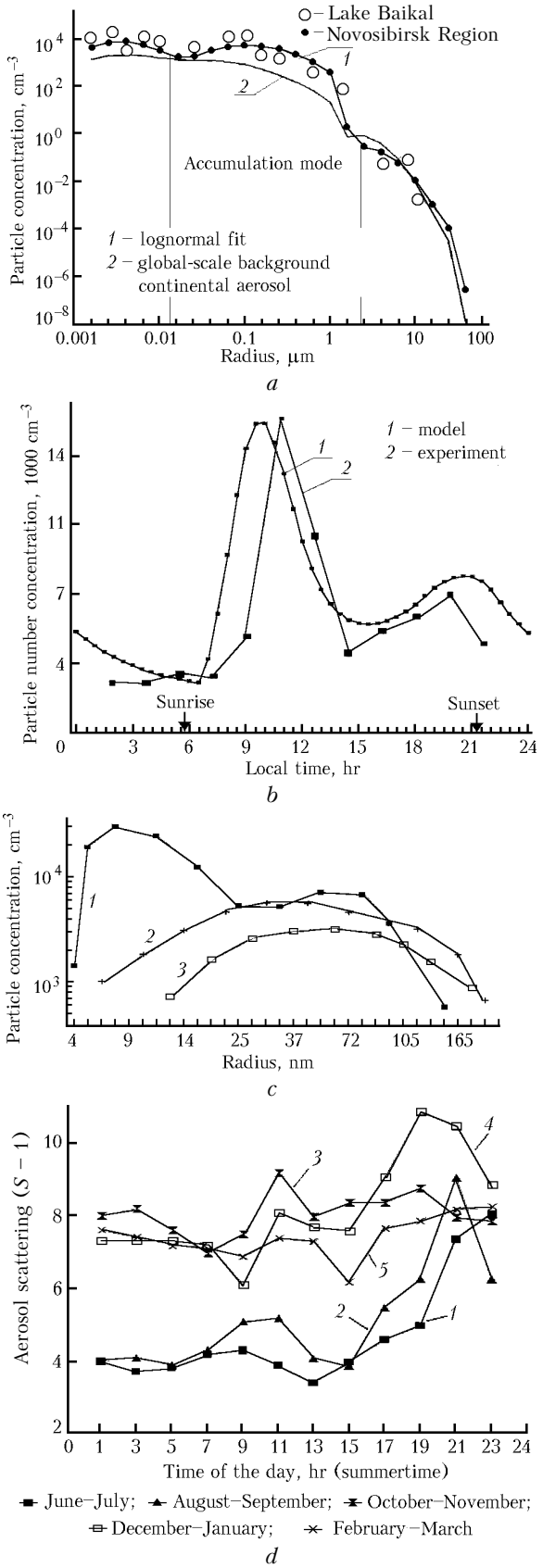

Fig. 1. Dynamics of the diurnal and seasonal variations of disperse composition and concentration of submicron fraction of atmospheric aerosol.

As was already mentioned above, the author has earlier proposed a dynamic model to describe the diurnal variations of number concentration and size spectrum of submicron AA fraction.[4,6,8] This was stimulated by the fact that the variations of number concentration and size spectrum of submicron AA fraction in diurnal cycle are determined by the behavior of the nucleation mode ($d < 0.1$ μm). Mass concentration of continental aerosol of remote regions is predicted by the Whitby model to depend not only on the characteristics of accumulation mode but also on those of coarse fraction. For AA in Siberia, the experimental studies showed that, in the summer season, the coarse fraction amounts for up to 85% of the total mass concentration of the aerosol.[45] The submicron fraction amounts for about 15%. In winter season, due to reduction of emission of soil-erosion component, the fraction of mass concentration of the coarse aerosol decreases.[45] However, due to global transport of air masses, an insignificant portion of soil-erosion derived submicron particles can also contribute to the formation of the accumulation-mode of AA over continental territories. This can be estimated only using data on chemical composition of the accumulation-mode of AA (see Ref. 46).

For the model proposed in this paper, such detailed information is not taken into account because the specific features of nucleation and accumulation modes are simulated according to their lifetimes. Whereas the nucleation mode particles have lifetime much shorter than a day,[47] the accumulation mode particles are much longer lived, allowing one to treat the specific features in the behavior of mass concentration of the accumulation mode in diurnal cycle as small deviations from a daily mean value. The physical processes leading to temporal changes of the mass concentration are characterized by the fact that the number concentration of accumulation mode is considered (such as in the previous model by the author) to be constant; whereas the mass concentration varies due to photochemical conversion of gas-phase precursors, acting to increase the number concentration of nucleation mode. Then the small particles "coalesce" into accumulation mode by coagulation.

Experimental results

As shown in Ref. 7, the AA scattering in Siberia is primarily associated with particles in the radius range 0.1–0.3 μm. In this size range, the sensitivity of nephelometer of the FAN-A type varies as a function of r^3 (Ref. 7). This dependence was obtained in Mie scattering calculations for particles of different sizes[10] taking into account the experimentally measured disperse AA composition, characteristics of illumination in counting volume of a FAN-A nephelometer, and spectral response characteristics of the photomultiplier used. This means that the total AA scattering is proportional to mass concentration of the accumulation-mode AA or mass concentration of submicron particles.

For aerosol scattering ($S - 1$) measured by a FAN-A nephelometer, it is possible to calculate the proportionality constant of mass concentration of submicron fraction[10]:

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

where M_{sub} is in μg/m^3 for aerosol particulate density of 1 g/cm^3; and S is the ratio of total to molecular scattering.

Relation (1) is inferred from the results on AA size spectrum determined in Siberia. Similar relation was obtained earlier from experimental data during measurement of AA scattering phase function in near-ground atmospheric layer.[48]

Figure 1*d* presents some experimental results on seasonal variations in diurnal cycle of scattering by submicron AA fraction in Novosibirsk Akademgorodok. In winter (Fig. 1*d*, curve *4*), when intensity of solar radiation and daylight portion of the day are minimum, the diurnal peaks of scattering, which are proportional to mass concentration of the accumulation mode, are less pronounced and are closer in time. In the warm part of the year (see Fig. 1*d*, curve *2*), the aerosol scattering is weaker than in winter, probably because in winter the PBL height is lower, while industrial emissions of chemically active gases and aerosol particles are more intense, than in summer.

Figure 2 presents simultaneously acquired data on the intensity of scattering at different observation sites (Klyuchi and Chany villages), separated by 450 km, for winter and summer periods. From this figure we can see two specific features: (1) in either season, the diurnal cycles at both sites are very similar; and (2) the diurnal variations of scattering have very different shapes in summer and winter.

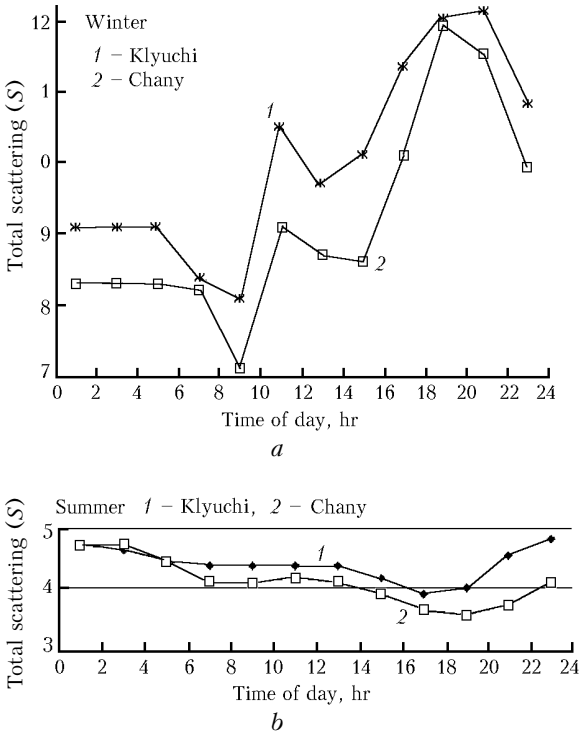

Fig. 2. Monthly mean diurnal behavior of scattering measured in winter (*a*) and in summer (*b*) in Novosibirsk Region.

Theoretical model

Suppose that the submicron fraction of atmospheric aerosol is a mixture of two monodisperse aerosol fractions. In this case, the total number concentration N will be a sum of number concentrations of individual fractions

$$N = N_1 + N_2, \tag{2}$$

where N_1 and N_2 are number concentrations of nucleation- and accumulation-mode particles, respectively.

The lifetime of the second mode is about 10 days, and these particles are uniformly distributed within the PBL. The lifetime of small, nucleation-mode, particles depends on the rate of coagulation with larger particles and transport from PBL[6]:

$$\frac{dN_1}{dt} = - k_{coag} N_1 N_2 = - \frac{1}{\tau_1} N_1. \tag{3}$$

In Ref. 11 it was shown that the coagulation constant k_{coag} between particles with radii 23 and 160 nm is $5 \cdot 10^{-8}$ cm^3/s. According to experimental data,[4,6] the number concentration of the accumulation mode is 2000 cm^{-3}. Finally, τ_1 is approximately 3 hr. In summer, the aerosol particles are transported from PBL during about 3 hr. Since the mass concentration of submicron AA fraction is proportional to the total scattering (σ), we can write the following equation:

$$\sigma = \sigma_1 + (\sigma_0 + \sigma_2), \tag{4}$$

where σ_1 is the scattering coefficient of the nucleation-mode particles; σ_0 is a constant component due to scattering by the accumulation-mode particles; and σ_2 is an additional component arising from particles formed due to coagulation of small and large submicron particles. The time variations of scattering by each of the two modes will be presented as

$$\frac{d\sigma_1}{dt} = - \frac{1}{\tau_1} \sigma_1; \quad \frac{d\sigma_2}{dt} = + K_M \frac{1}{\tau_1} \sigma_1; \tag{5}$$

where

$$K_M = - \frac{d\sigma_2}{d\sigma_1} = + \left(\frac{d\sigma_2}{dM_2}\right) / \left(\frac{d\sigma_1}{dM_1}\right). \tag{6}$$

From the laws of mass conservation it follows that $-dM_2/dM_1 = 1$, where M_1 is the nucleation mode mass, and M_2 is accumulation mode mass.

The theoretical calculations[7] showed that the function $\sigma(r)$ can be fitted by the formula:

$$\sigma = \sigma_0 \frac{1 + (r/R)^{1.5}}{1 + (R/r)^6}. \tag{7}$$

In equation (7), $R = 160$ nm. Using Eqs. (6) and (7), for $r = 23$ nm we obtain $K_M = 84$. This result suggests that, when small particles coagulate with larger ones, the increase of scattering by the large particles must be taken into account. Therefore, we present $d\sigma/dt$ as:

$$\frac{d\sigma}{dt} = C_0 f(t + T) - \frac{\sigma - \sigma_0}{\tau_2} Z(t), \tag{8}$$

where $1/\tau_2$ is the amplitude of diurnal variations of vertical transport rate, equal to 0.3 hr^{-1}; $Z(t)$ is the diurnal vertical mass exchange rate; C_0 is the adjustment parameter chosen according to the amplitude of diurnal variations of intensity of aerosol scattering; T is the time, during which the nucleation mode is converted into the accumulation mode. Using experimental data,[12] it is possible to determine all parameters required to solve equation (8).

Winter

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

$$Z(t) = 1, \tag{10}$$

since in winter season the coefficient of turbulent diffusion varies little during a day. In the winter period, the relative air humidity is practically constant during a day and it is close to 100%. Therefore, in this season the effect of relative humidity on variations of aerosol scattering can be neglected.

Summer

$$T \sim 2 \text{ hr}, \; f(t) = \begin{cases} 0, \text{ if } 0 \le t \le 6; \; 22 \le t \le 24, \\ \sin\left[\frac{\pi}{16}(t - 6)\right], \text{ if } 6 \le t \le 22; \end{cases} \tag{11}$$

$$Z(t) = 1.2 + \sin\left[\frac{\pi}{2}(t - 1) + \frac{3}{2}\pi\right] \tag{12}$$

(see Ref. 12, Fig. 32). In summer period, it is also necessary to take into account the diurnal variations of relative humidity (f_{RH}):

$$\sigma = \sigma_0 \, [1 - f_{RH}(t)]^{-0.5}; \tag{13}$$

$$f_{RH}(t) = \begin{cases} 0.75, \text{ if } 0 \le t \le 6, \\ 0.75 - 0.25 \sin\left[\frac{\pi}{18}(t - 6)\right], \text{ if } 6 \le t \le 24. \end{cases} \tag{14}$$

Figure 3 shows the results of theoretical calculations in the frameworks of suggested model. The curve *1* shows the calculated dependences, obtained using numerical solution of equation (8). For comparison purposes, experimental data (curves *2*) are also presented.

From a comparison of theoretical calculations with the measured data it is seen that the model proposed well describes the specific features of diurnal variations of mass concentration of submicron AA fraction in different seasons (development of maxima and minima, their seasonal variations, magnitude, and timing).

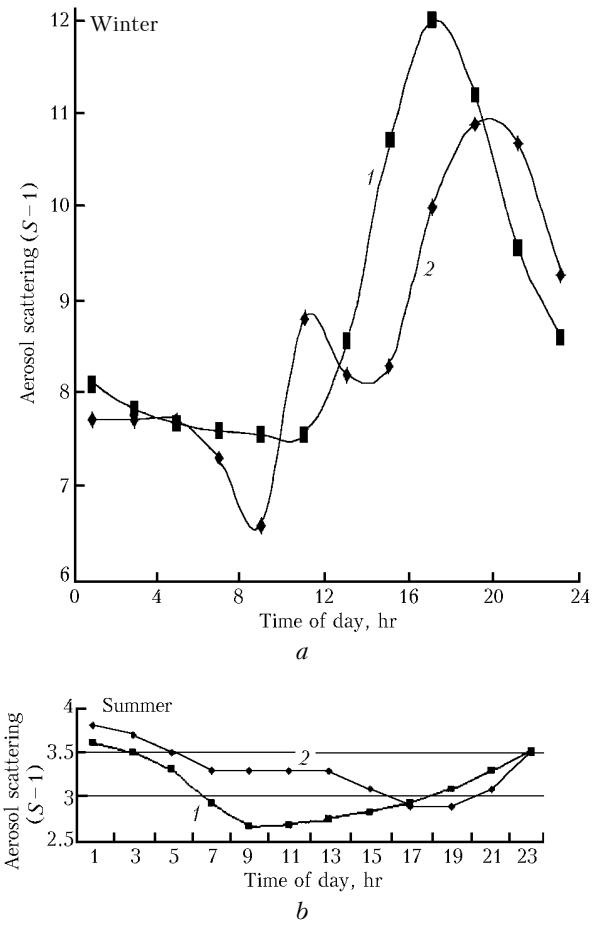

Fig. 3. Diurnal variations of scattering in winter and summer: calculations (curve *1*) and experiment (curve *2*).

Conclusions

To describe the seasonal variations of mass concentration of submicron AA fraction, a model is proposed which takes into account the photochemical process of particle formation, coagulation of formed particles, and diurnal variations of the planetary boundary layer and relative air humidity.

Comparison of the calculations, performed using the model suggested here, with the experimental data has shown that the model is successful in reproducing the diurnal maxima and minima of mass concentration of accumulation-mode AA concentration, their timing and magnitude, as well as seasonal variations of these main characteristics of diurnal cycle.

The model proposed here is based on clearly defined physical and chemical processes. The parameters describing the dynamics of these processes are specified functionally. They either can be measured experimentally or calculated theoretically, so the model can be used to study the diurnal variations of characteristics of submicron AA fraction under other conditions, and admits further refinement and development.

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