COMPUTER SIMULATION OF DIURNAL VARIATIONS OF ATMOSPHERIC AEROSOL SIZE SPECTRA

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A mathematical model describing the processes of coagulation, dilution, and precipitation of particles is proposed for a numerical study of aerosol number density dynamics. The paper presents the results of calculations of daily mean behavior of atmospheric aerosol parameters in summer period.

This paper continues the investigations of one of the authors[1,2] into the nature of diurnal variation of number density and size spectra of atmospheric aerosol in Siberia. References 1–3 give the results of measurement of dynamics of the number density and light scattering of continental aerosol in summer period and propose the mechanism of aerosol formation, which describes qualitatively properly the special features of the obtained dynamics of measurements. Based on the above mechanism, the mathematical model of aerosol formation is proposed including the description of processes of coagulation, mixing, dilution of small particles, and precipitation of large particles. A detailed description of the technique of experimental measurement of particle size spectra, number density, and light scattering is given in Refs. 1–3 and 6.

For numerical simulation of dynamics of atmospheric aerosol formation, we use the following set of coagulation equations:

$$\frac{dN_1}{dt} = K_1(t)G - K_3(t)N_1 - N_1 \sum_{i=1}^{\infty} \beta_{1i} N_i , \qquad (1)$$

$$\frac{dN_l}{dt} = -C_i(t)N_l + \frac{1}{2} \sum_{i+j=1} \beta_{ij} N_i N_j - N_l \sum_{i=1}^{\infty} \beta_{li} N_i , \qquad (2)$$

where $N_l(t)$ is the number density of aerosol particles consisting of l monomers; β_{ij} is the rate of coagulation adhesion of particles in the regime of freely molecular collisions; G, W, and $K_l(t)$ are the concentration, the rate of emission, and the photodissociation function of gas precursors of aerosol particles, respectively; $K_3(t)$ is the dilution function of monomers of aerosol particles; $C_i(t)$ is the function characterizing the rate of number density decrease of particles with volume i as a result of processes of dilution of small particles ($2 \le i \le 50$) and precipitation of large particles ($i \ge 10^5$). The quantity G of the gas precursors concentration is given by the solution of the following boundary-value problem:

$$\frac{dG}{dt} = W - G(K_1(t) + K_3(t)); \; G(0) = G(24). \qquad (3)$$

When calculating the diurnal dynamics of aerosol formation in summer period, the values of functions and constants of gas precursors photodissociation and monomer dilution are given by the following relations:

$$K_1(t) = K_{01} f(t), \qquad K_{01} = 2.1 \cdot 10^{-4} \text{ s}^{-1},$$

$$f(t) = \begin{cases} 0 & \text{at } 0 \le t \le 6, \;\; 22 \le t \le 24; \\ \sin\left(\dfrac{\pi}{16}(t-6)\right) & \text{at } 6 \le t \le 22, \end{cases}$$

$$K_3(t) = K_{03} z(t), \qquad K_{03} = 2.2 \cdot 10^{-4} \text{ s}^{-1},$$

$$z(t) = \begin{cases} 0 & \text{at } 0 \le t \le 7, \;\; 18 \le t \le 24; \\ \sin\left(\dfrac{\pi}{16}(t-6)\right) & \text{at } 7 \le t \le 18, \end{cases}$$

where t is the daytime, h. The values of the dilution function of l-mers $2 \le l \le 50$

$$C_l(t) = c_l K_3(t), \; c_l = \frac{\log(l) - \log(50)}{\log(1) - \log(50)}$$

are taken with regard to the fact that the source of aerosol particles is located in the surface layer and, consequently, small particles are diluted more actively due to atmospheric convection. The value of the constant $C_l(t) = c = 1.5/(8 \cdot 24)$ in the expression for the rate of precipitation of large particles of the volume ($l \ge 10^5$) is found from the suggestion that the large particle number density decreases by about a factor of three in the absence of the source over the period of eight days that is consistent with the experimentally determined mean life time of aerosol particles in the lower troposphere.

The numerical values of the constants of rates of coagulation adhesion of particles β_{ij} are taken from Ref. 5. The value $W = 0.68 \cdot 3000 \text{ cm}^{-3}\text{s}^{-1}$ of the emission rate of gas precursors is calculated from the condition of conservation of sum aerosol matter daily.

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The initial aerosol particle size spectrum is approximated by the lognormal function

$$\frac{dN(r)}{d\log r} = \frac{N_0}{\sqrt{2\pi}\,\log\sigma} \exp\left(-\frac{\log^2 r/R}{2\log^2 \sigma}\right)$$

with the parameters $\sigma = 1.5$; $R = 41$ nm; $N_0 = 2000$ cm^{-3}, that corresponds to the accumulation mode of bimodal distribution of average size spectrum of submicron function of atmospheric aerosols.[1,2] The mode of large particles is not included in the model since mode practically does not affect the dynamics of calculated below functionals of light scattering and sum aerosol number density.[3] Therefore, in the expression for the initial size distribution function of aerosol particles, we use only the single modal distribution, which parameters correspond to those of accumulation mode.

To solve the Cauchy problem, Eqs. (1) and (2), we use the section finite-element algorithm proposed by us in Ref. 4, in which 50 point and 80 linear finite elements were employed for the approximation of aerosol number density spectrum. The detailed description of the algorithm of transition from a continuous spectrum to a discrete one, as well as integration procedure and the study of its accuracy are also given in Ref. 4. Note that in the case of freely molecular collisions, the accuracy of numerical solution for typical test problems is not worse than 10^{-4}. The set of 130 ordinary differential equation derived when approximating Eqs.(1) and (2) was solved before determining the diurnal particle distribution spectrum from day to day. That corresponds to physical time about 20 days.

Figure 1 shows the daily mean behavior of the aerosol number density, which has the marked maximum during daytime from 10:00 to 14:00. In our calculations, the ratio of the sum number density maximum to the minimum was in the range of 2–2.5 that was in qualitative agreement with the measured data.[1–3]

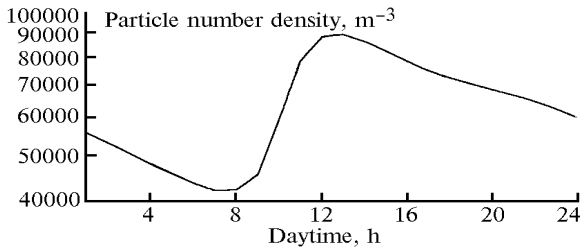

FIG. 1. Established mean daily behavior of particle number density.

The functional $\psi(t)$ of optical light scattering for the calculated particle size distribution spectrum was found from the following expression:

$$\psi(t) = \int \frac{dN(t)}{d\log r}\,\phi(r)\,d\log r,$$

where

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

$R = 0.16$ μm.

Transition from the time variation of number density to the time dependence of the light scattering coefficient is justified in Ref. 3.

The established daily dynamics of optical light scattering is given in Fig. 2.

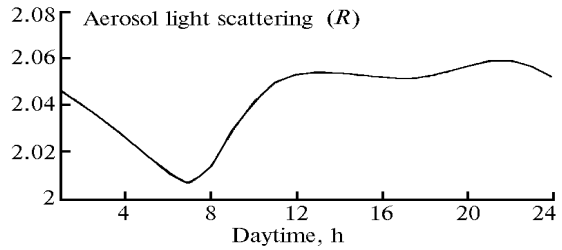

FIG. 2. Established mean daily behavior of optical scattering. R is the ratio of aerosol light scattering to the molecular one.

It is evident that, for the given daily variation, two peaks with more marked evening maximum between 20:00 and 24:00 are typical. The first peak correlates with the maximum of the number density connected with the growth of the number of new small particles, the second peak is due to the growth, as a result of coagulation process, of the number of large particles which more intensively scatter light. Although the absolute value of the oscillation amplitude and the light scattering intensity obtained in the numerical simulation is less than the experimentally observed one (about 10–15% in the experiment and 1–3% in calculations), the calculated value exceeds the accuracy of the numerical solution.

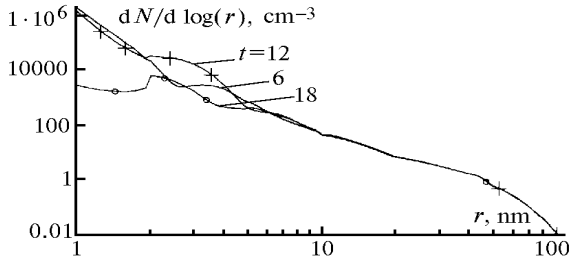

FIG. 3. Examples of established spectra at different daytime.

Therefore, the shape of theoretical dependence is beyond question. This shape agrees qualitatively with the experimentally observed regularities (positions of maxima and minima in the diurnal dynamics).

Figure 3 shows the character of particle size spectrum transformation at different instants of time. The particle size spectra are given for $t = 6$ a.m., 12 a.m., and 6 p.m.

As is seen from the figure, the number density of small particles varies by three orders of magnitude during a day. For the particles of $r > 10$ nm size, the number density obtained in our calculations varies only slightly. This agrees well with the characteristic features of behavior of accumulation fraction of atmospheric aerosols.

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