OPTICAL CHARACTERISTICS OF SMOKE PARTICLES

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Smoke particles can be regarded as aggregates of small soot particles fractal in structure. A model of a smoke particle has been proposed based on results of study of such aggregates, and calculations of the extinction, absorption, scattering, and backscattering coefficients have been done. Results of modeling furnish a qualitative explanation of optical properties of smoke of various type.

It is well known that atmospheric aerosol particles represent complex agglomerations of a lot of small homogeneous (in the first approximation) particles. Smoke particles are the most widespread example of such agglomerations.[1] It seems natural that practically all physico–chemical properties of such agglomerations differ essentially from corresponding properties of both homogeneous large particles and a system of independent small particles composing aggregate of the same size (or mass) as the agglomeration.

In the last few years concepts and methods of the theory of fractal systems[2,3] have been successfully employed for the study of various properties of such formations. Fractal aggregates are the specially formed structures in which each element is similar to the entire system.[4] As applied to atmospheric aerosol problems, one can consider the aggregates formed by many particles (N | 10^2–10^3) having the same physico–chemical properties as fractals. The size of such particles is essentially less than the size of the system and is approximately the same for individual particles. Their arrangement is described by general statistical laws. Even this definition, although not very strict and exhaustive, is evidence of the fact that atmospheric aerosols can be regarded as fractal systems only in rough approximation, because the primary particles composing aggregate are not identical under real atmospheric conditions. They are polydisperse and may differ in their chemical composition. Nevertheless, the study confirms that usage of such an approximation allows one to describe reasonably adequately both structure and properties of smoke particles. (It seems likely that multifractal approximation considers more adequately the properties of formations similar to aggregated aerosol particles. However, the theory of optical properties of multifractal aggregates is still far from completion.)

To describe a fractal aggregate, one usually uses such parameters as the size of structural element a (for aerosols it is the average radius of primary particles), the number of elements in the aggregate N, and the fractal dimension of the aggregate D. In Ref. 5 it has been shown that the smoke particles can be regarded as fractal structures with $a = 0.01$–0.05 μm, $N = 3\cdot10^2$–10^4, and $D = 1.78$. The specific values of the parameters of the fractal structure depend on the mechanism of smoke particle formation, and although this dependence has not yet been explained, it is determined on an empirical level.

A series of investigations into the optical characteristics of aggregates fractal in structure was performed at the Laboratory of Aerosol Physics.[6] In particular, the applicability of the theory of fractal optical properties was examined,[7] and it was shown that although this theory cannot be regarded as strict, it allows one to obtain the realistic estimates of the optical characteristics of aggregate that describe adequately results of experimental optical investigations taking into account the effect of multiple scattering on particles inside the aggregate.

Using the results of Ref. 6, we calculated the optical characteristics of smoke particles. The results of model calculations are presented below.

The construction of the model of the object under study and the choice of its characteristics are the first problem of any attempt at numerical modeling. As has been mentioned above, the average size of primary particles is $a \approx 0.01$–0.05 μm depending on a smoke source, and the fractal dimension of aggregates is $D \approx 1.78$. The aggregate size R varies from several tenth to ~10 μm. The smoke particles in the atmosphere are long–lived. Taking into account their friable structure, gravitational sedimentation of such particles is ineffective. They are removed from the atmosphere by washing out or are entrained by descending air flows.

The smoke particles of natural origin produced by combustion can be accumulated at great heights immediately in the process of their production. Even produced by a small fire, they reach several tens of meters; in the case of heavy forest fires, they can reach heights up to 1.5–2 km. Industrial smoke is deliberately emitted into the atmosphere at heights up to several hundreds of meters. As a result, the lifetime of smoke particles can be several hours or even days. For so long time the particles can be transported within several tens and hundreds of kilometers in the atmosphere, being subject to some structural transformations. In the field of variable humidity, the smoke particles became denser as the air humidity increases. Then, taking into consideration the fractal structure, the fractal dimension increases with simultaneous decrease of the aggregate size.

If one considers primarily the optical properties of such aggregates, as it is in this particular case, one cannot ignore the fact that when the humidity of air surrounding a particle varies, the effective particle size and the effective values of the complex refractive index vary due to interaction of both the aggregate and individual primary particles composing it with atmospheric moisture. However, the process of interaction of soot particles with atmospheric water vapor is as yet very little understood. At the same time, the probability exists that the small particle approximation becomes inapplicable to the primary particles because of their growth due to moistening. The theory of the optical properties of aggregates fractal in structure is based on precisely this approximation (the main assumption of this theory reduces to isotropy of scattering on primary particles that allows one to obtain analytical expressions connecting the optical characteristics of a system with the corresponding

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parameters of particles composing it; for strongly elongated scattering phase function of primary particles, one can consider only numerical solution of the problem under specific conditions with some restrictions).

Moreover, the term "soot" is hypothetical in this case. Smoke particles do not consist of the chemically pure substance like soot. In this case, we consider soot as a complex mixture of incomplete combustion products. Chemical composition of smoke and hence its physico–chemical properties differ essentially for smoke of different origin. In this connection, we consider in this paper only variations of smoke optical properties associated with different structure of particles without a single attempt at considering the possible variations of the characteristics of small particles composing aggregates under the effect of varying air humidity.

But even in this case significantly simplified in comparison with real situation some difficulties emerge connected with variability of the composition of smoke particles. In particular, when interpreting the measurement results reported in Ref. 6, the choice of the values of complex refractive index of the particulate matter involved difficulties, and finally this problem was solved by the adjustment technique, i.e., the values of the refractive index were chosen to provide the best agreement between experimental and calculated data. Evidently, now we can say only about the qualitative relationship between the values of real and imaginary parts of the complex refractive index of the smoke particles of different origin.

In calculations, we used the model value of the refractive index $m = 1.75 - i\,0.65$ for the visible spectral range (this value of the complex refractive index provided the best agreement between the experimental and theoretical data presented in Ref. 6) and proceed to the model values m in the spectral range $\lambda \geq 1 \mu m$ according to Ref. 8.

The curves of extinction (a), absorption (b), scattering (c) and backscattering (d) coefficients are shown in Fig. 1 for one model of smoke particles in comparison with corresponding characteristics of analogous (by mass) system of small non–interacting soot particles. Even straightforward comparison of the curves drawn in Fig. 1 shows that a consideration of the smoke particles as aggregates fractal in structure leads to essential overestimation of their optical characteristics.

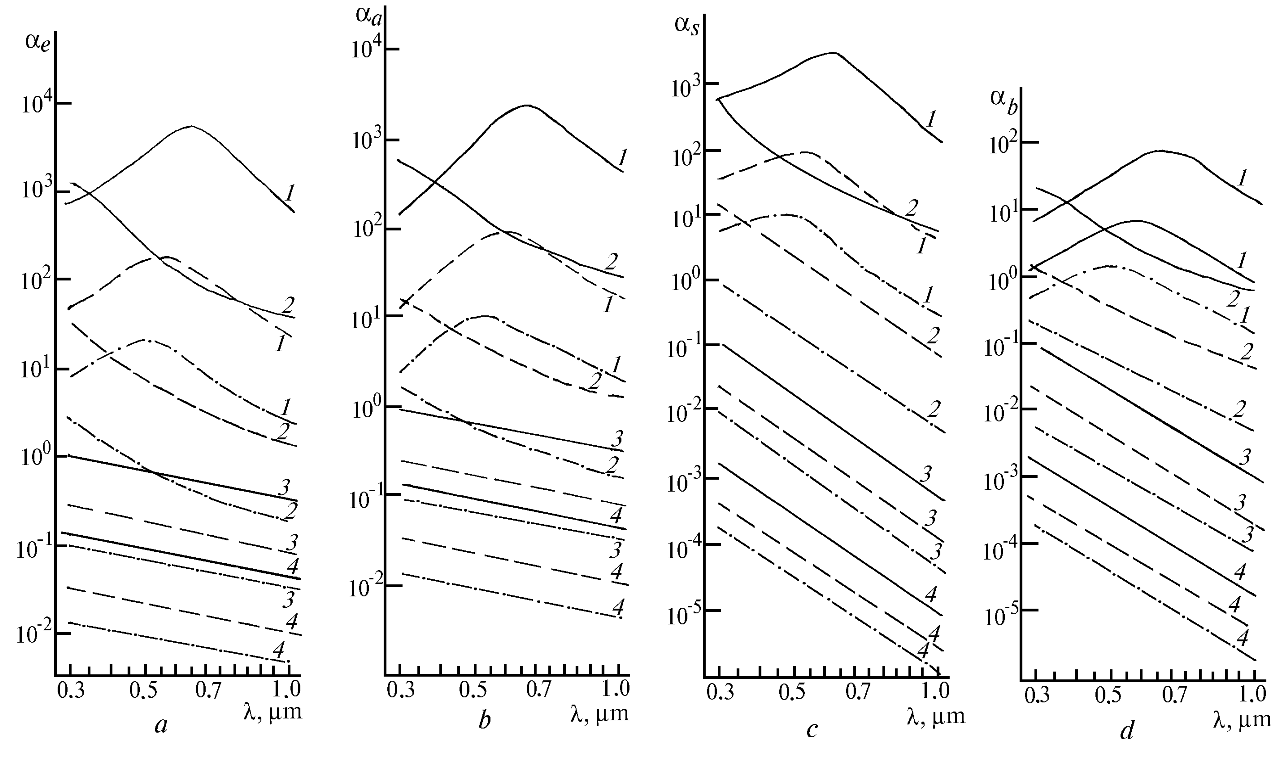

FIG. 1. Extinction α_e(a), absorption α_a(b), scattering α_s(c), and backscattering α_b(d) coefficients for model smoke particles: solid line is for N = 2000, dashed line is for N = 500, dash–dot line is for N = 200: 1) and 3) a = 0.02; 2) and 4) a = 0.01 μm; 1) and 2) aggregate fractal in structure with D = 1.78; 3) and 4) system of noninteracting particles.

The scattering phase functions of the considered aggregates are shown in Fig. 2, and the comparison of calculated estimates with the results of experimental measurements under laboratory and field conditions is shown in Fig. 3.

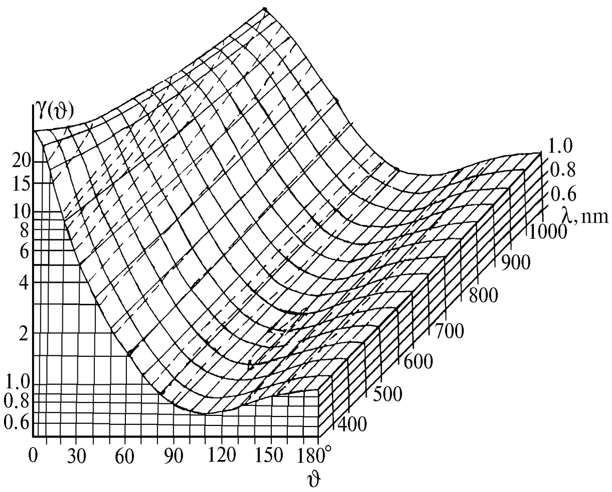

FIG. 2. Absolute brightness phase function of fractal aggregate in the wavelength range 0.3–1.0 μm.

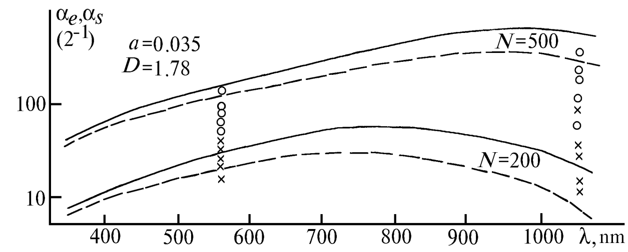

FIG. 3. Comparison of the results of experimental measurements (symbols) with numerical estimates (curves) of the extinction (solid lines and circles) and scattering (dashed lines and small crosses) coefficients of smoke particles.

It seems that the results obtained provide strong evidence of the fact that application of concepts of the theory of fractal aggregates, on the one hand, allow us to describe realistically the results of experimental measurements and, on the other hand, made us to revise the estimates of the role of smoke pollution of the atmosphere that are used in the problems associated with the study of climatic consequences of nuclear war, effect of anthropogenic activity on climate formation, and so on.[9,10]

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