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ON THE CALCULATION OF OPTICAL CHARACTERS OF ATMOSPHERIC AEROSOL AND SOOT

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Introduction

This study deals with optical properties of the medium which is polydisperse ensemble of nonhomogeneous spherically symmetrical balls.

Soot-dust atmospheric pollution of aerosol type is considered in the paper. Real particles of such a pollution are composite. Optical properties of such a medium are defined by optical properties of every particle, size and structure distribution of particles.

An algorithm of calculation of absorption and scattering coefficients and phase function of nonhomogeneous spherically symmetrical particle (perhaps, with several layers) was constructed on the basis of the Mie theory.

There are considered some models for size and structure distributions of soot-dust balls. A weak dependence of optical characters of such a medium on model parameters of composite was shown.

§ 1. Coagulation models

For researching the influence of composite on optical properties of aerosol pollution mentioned it is necessary to construct a model for size and structure distributions of soot-dust balls.

During interaction of homogeneous "soot" and "dust" particles composite "soot-dust" particles appear and it is supposed that soot particles are accumulated on "dust" particles. Such a supposition is based on the fact that average dust particles volume is equal to 100 average soot particles volume (soot particles are essentially smaller than dust particles).

Let V be some atmosphere volume; then $V_D V$ and $V_S V$ are dust and soot volumes in this atmosphere volume. We shall use next the designation: $\tilde{V}_D = 10^9 V_D$, $\tilde{V}_S = 10^9 V_S$. In [1], there are next values: $\tilde{V}_D = 0.8$, $\tilde{V}_S = 0.2$. As soot particles are smaller than dust particles, it is supposed that the soot is a continuous medium. Moving in such a medium, dust particles are covered by soot. During such accumulation process, spherical symmetry of "soot-dust" particles is conserved.

Let us consider a law of volume growth of composite particle. This law is like a law of volume change of a particle when water is accumulated on the particle [2]:

$$\frac{\partial X^3(t,x)}{\partial t} = A\big(t, \alpha_D v_D + \alpha_S v_S - V^*(t)\big) g(X,x), \tag{1.1}$$

where $X = R/r_M$, $x = r/r_M$, $r = 1$ μm; r_M is the average particle radius, R, r radii of composite and dust particles, α_D $(0 \leqslant \alpha_D \leqslant 1)$ a part of coagulated dust, $\tilde{\alpha}_S$ $(0 \leqslant \tilde{\alpha}_S \leqslant 1)$ a part of soot that may coagulate, $V^*(t)$ the hole composite particle volume at t time of moment per unit atmosphere volume:

$$V^*(t) = \alpha_D v_D \int_{x_m}^{X_M} N_D(x) X^3(t,x)\,dx, \tag{1.2}$$

where $x_m = 0.001$, $X_M = 3$; x_m, X_M are, respectively, undimensioned minimum and maximum particles radii, $N_D(x)$ radius distribution of homogeneous dust with norm condition

$$\int_{x_m}^{X_M} N_D(x) x^3\,dx = 1. \tag{1.3}$$

An initial condition for (1.1) is

$$X(0,x) = x, \quad x_m \leqslant x \leqslant X_M. \tag{1.4}$$

Function $A(t,p)$ has the following properties:

$$\begin{aligned} &A(t,p) \geqslant A_0 > 0, && t \geqslant 0,\ p > 0; \\ &A(t,p) = 0, && t \geqslant 0,\ p \leqslant 0. \end{aligned} \tag{1.5}$$

Let us consider (1.1)–(1.5). A limit $X(x) = \lim_{t\to\infty} X(t,x)$ does not depend on function type $A(t,p)$, but only depends on properties (1.5). So, when we study the $X(x)$ function, we can take without loss of generality $A(t,p) = 1$.

In the case of small particles, dust balls are considered "fluid". Interaction between the balls and soot is taken as continuous mixing. A cross-section of such interaction is proportional to dust particle volume. In the case of balls larger than some average, the process of soot accumulation on these balls is considered as a process of soot accumulation on large balls. The cross-section of such the interaction is proportional to ball surface.

On the basis of assumption above, let us consider several cases of interaction function $g(X,x)$. At first, let us take

$$g(X,x) = g_1(x) = \begin{cases} x^3, & x_m \leqslant x \leqslant x_* = (x_D x_S)^{1/2}, \\ x_* x^2, & x_* \leqslant x \leqslant X_M. \end{cases} \tag{1.6}$$

Here and later, $x_D = r_D/r_M = 0.25$, $x_S = r_S/r_M = 0.05$, r_D, r_S are "average" dust and soot particles radii.

From solution (1.1)–(1.6) it follows that there exists T time moment such that

$$X(t,x) < X(T,x), \quad 0 \leqslant t < T;$$
$$X(t,x) = X(T,x), \quad t \geqslant T.$$

And we have $V^*(T) = \alpha_D V_D + \alpha_S V_S$ where α_S $(0 \leqslant \alpha_S \leqslant \tilde{\alpha}_S)$ is a part of coagulated soot.

Then the limit solution $X(x)$ will be

$$X(x) = \left[x^3 + \frac{\alpha_S}{\alpha_D} \Delta \frac{g_1(x)}{\int\limits_{x_m}^{X_M} g_1(x) N_D(x)dx} \right]^{1/3} \qquad \Delta \equiv V_S/V_D.$$

In case of such interaction function g, volume of soot, accumulating on a dust particle, may be infinite, and as a result, composite particles may grow up to extremely large dimensions that is not real.

Let us consider another interaction function, more naturally. We have

$$g(X,x) = g_2(X,x) = \begin{cases} x^3, & X \leqslant x_*, \\ x_* X^2, & x_* \leqslant X \leqslant X_M, \\ 0, & X \geqslant X_M. \end{cases} \tag{1.7}$$

A solution of (1.1)–(1.5), (1.7) is

$$X(t,x) = \begin{cases} x(t+1)^{1/3}, & 0 \leqslant t \leqslant t_*(x), \\ \dfrac{x_*}{3}[t - t_*(x)] + x[t_*(x)+1]^{1/3}, & t_*(x) \leqslant t \leqslant t_M(x), \\ X_M, & t \geqslant t_M(x), \end{cases}$$

where $t_*(x)$ is a time of growth of dust particle from x initial radius to x_* dimension, $t_M(x)$ is time of growth of dust particle from x initial radius to X_M dimension. These functions are defined as follows:

$$t_*(x) = \int\limits_x^{x_*} \frac{ds^3}{g(s,x)} = \max\left\{0, \left(\frac{x_*}{x}\right)^3 - 1\right\},$$

$$t_M(x) = \int\limits_{x_m}^{X_M} \frac{ds^3}{g(s,x)} = \max\left\{0, t_*(x) + \frac{3}{x_*}[X_M - \max(x, x_*)]\right\}.$$

If we assume that all dust particles of x dimensions, where $x_m \leqslant x \leqslant X_M$, will grow up to X_M dimension, their volume v_M will be

$$v_M = X_M^3 \int\limits_{x_m}^{X_m} N_D(x)dx.$$

Time T is the moment of the end of coagulation. In the case

$$v_M > 1+\frac{\alpha_S}{\alpha_D}\Delta, \tag{1.8}$$

we have that T is less than the time of growth of smallest particles up to critical dimension: $T < t_M(x_m)$. Note that $t_M(x)$ is a monotonous increased function, i.e. $t_M(s') < t_M(s)$, if $x_m \leqslant s' < s \leqslant X_M$. In case (1.8), value T is defined from an equation

$$\Phi(T) = \int_{x_m}^{X_M} X^3(T,x)N_D(x)dx = 1+\frac{\alpha_S}{\alpha_D}\Delta.$$

Function $\Phi(t)$, $0 \leqslant t \leqslant t_M(x_m)$, is a continuous monotonous increased function. So, solution T exists

$$T = \Phi^{-1}\left(1+\frac{\alpha_S}{\alpha_D}\Delta\right).$$

In this case, time T characterizes time moment, when all dust and soot particles, which are allowed to coagulate, have coagulated.

In the case

$$v_M = 1+\frac{\alpha_S}{\alpha_D}\Delta.$$

we have that time T is equal to time of growth of smallest particle up to critical dimension: $T = t_M(x_m)$. That means that for a particle of an arbitrary dimension $x_m \leqslant x \leqslant X_M$ the following is true: $X(T,x) = X_M$. Here, time T characterizes a time moment, when all composite particles have fallen down, because of the fact that dust particles had accumulated so much soot that they have grown up to X_M dimension. And as a result, they have fallen down. Value α_S is defined as follows:

$$\alpha_S = \begin{cases} \tilde{\alpha}_S, & T < t_M(x_m), \\ [\alpha_D(v_M - 1)]/\Delta, & T = t_M(x_m). \end{cases}$$

If $\alpha_S \leqslant \tilde{\alpha}_S$, then dust part α_D and soot part α_S have fallen down.

So, in this coagulation model we have that dimension of composite particles X is a function of the dimension of dust nucleus only:

$$X(x) = X(T,x).$$

We also note that T time stabilization for such soot-dust particles is many times less than "cleaning" time of atmosphere (washing-out, sedimentation, etc.). For this reason we consider the situation for instantly stabilized distribution of composite particles.

In models discussed we assume that the size distribution function of composite particles is

$$N_{SD}(x) = \begin{cases} N_D(x), & X(x) < X_M, \\ 0, & X(x) = X_M. \end{cases}$$

These properties allow to describe the composite particles structure by means of a single independent variable dust nucleus coagulation dimension x. This allows us to simplify essentially a computation of optical characteristics of soot-dust pollution.

§ 2. Calculation of optical characteristics of polydisperse ensemble of composite particles

We shall describe a computation algorithm of optical properties of a medium consisting of composite and homogeneous particles. The algorithm is based on a calculation of optical properties of homogeneous soot and dust particles and composite "soot-dust" particles [3].

As noted above, we assume that only dust part α_D coagulates with soot. The rest dust part $(1-\alpha_D)$ has the following $N_D(x)$ size distribution [1]:

$$N_D(x) = \begin{cases} Bx^{-1}\exp\left[-\dfrac{\ln^2(x/x_D)}{2\ln^2\sigma}\right], & x_m \leqslant x \leqslant 1, \\ Bx^{-4}\exp\left[-\dfrac{\ln^2 x_D}{2\ln^2\sigma}\right], & 1 \leqslant x \leqslant X_M, \end{cases}$$

where $\sigma = 2$ and the condition (1.3) takes place.

Absorption coefficient $\varkappa_{a_D}$, scattering coefficient $\varkappa_{s_D}$, phase function γ_D of the rest dust part are, respectively (see [3]):

$$\varkappa_{a_D}(\lambda) = \tilde{\varkappa}_{a_D}(1-\alpha_D)\hat{V}_D = \hat{V}_D(1-\alpha_D)\int_{x_m}^{X_M} Q_{a_D}(x;\lambda)\,N_D(x)x^2\,dx, \tag{2.1}$$

$$\varkappa_{s_D}(\lambda) = \tilde{\varkappa}_{s_D}(1-\alpha_D)\hat{V}_D = \hat{V}_D(1-\alpha_D)\int_{x_m}^{X_M} Q_{s_D}(x;\lambda)\,N_D(x)x^2\,dx, \tag{2.2}$$

$$\varkappa_{s_D}\gamma_D(\chi;\lambda) = \hat{V}_D(1-\alpha_D)\int_{x_m}^{X_M} Q_{s_D}(x;\lambda)\gamma_D^*(\chi, x, \lambda)N_D(x)x^2\,dx, \tag{2.3}$$

where $\hat{V}_D = 0.75\,\tilde{V}_D$ and index D shows that these functions relate to dust. Note that $\tilde{\varkappa}_{a_D}$, $\tilde{\varkappa}_{s_D}$, γ_D do not depend on α_D. Here $\pi x^2 Q_{a_D}(x;\lambda)$, $\pi x^2 Q_{s_D}(x;\lambda)$ are absorption and scattering coefficients, and $\gamma_D^*(\chi, x;\lambda)$ phase function of some dust particle of radius x.

Absorption coefficient $\varkappa_{a_{SD}}$, scattering coefficient $\varkappa_{s_{SD}}$, phase function γ_{SD} of the medium of "soot-dust" balls are, respectively,

$$\varkappa_{a_{SD}}(\lambda) = \tilde{\varkappa}_{a_{SD}}(\lambda)\alpha_D\hat{V}_D = \hat{V}_D\alpha_D\int_{x_m}^{X_M} Q_{a_{SD}}(x;\lambda)N_{SD}(x)X^2(x)\,dx,$$

$$\varkappa_{s_{SD}}(\lambda) = \tilde{\varkappa}_{s_{SD}}(\lambda)\alpha_D\hat{V}_D = \hat{V}_D\alpha_D\int_{x_m}^{X_M} Q_{s_{SD}}(x;\lambda)N_{SD}(x)X^2(x)\,dx,$$

$$\varkappa_{s_{SD}}\gamma_{SD}(\chi;\lambda) = \hat{V}_D\alpha_D\int_{x_m}^{X_M} Q_{s_{SD}}(x;\lambda)\gamma_{SD}^*(\chi, x;\lambda)N_{SD}(x)X^2(x)\,dx,$$

where index SD shows that these functions relate to the medium of "soot-dust" balls. Here $\pi X^2(x) Q_{as_D}(x;\lambda)$, $\pi X^2(x) Q_{ss_D}(x;\lambda)$ are absorbtion and scattering coefficients of some composite particle with x radius of dust coagulation nucleus.

We assume that the rest soot part $(1-\alpha_S)$ has the following $N(x)$ size distribution [1]:

$$N_S(x) = C x^{-1} \exp\left[-\frac{\ln^2(x/x_S)}{2\ln^2\sigma}\right], \qquad x_m \leqslant x \leqslant X_M,$$

with a norm condition

$$\int_{x_m}^{X_M} N_S(x) x^3 \, dx = 1.$$

Absorption coefficient $\varkappa_{a_S}$, scattering coefficient $\varkappa_{s_S}$, phase function γ_S of the rest soot part are, respectively,

$$\varkappa_{a_S}(\lambda) = \tilde{\varkappa}_{a_S}(1-\alpha_S)\Delta \hat{V}_D = \hat{V}_D \Delta (1-\alpha_S) \int_{x_m}^{X_M} Q_{a_S}(x;\lambda) N_S(x) x^2 \, dx,$$

$$\varkappa_{s_S}(\lambda) = \tilde{\varkappa}_{s_S}(1-\alpha_S)\Delta \hat{V}_D = \hat{V}_D \Delta (1-\alpha_S) \int_{x_m}^{X_M} Q_{s_S}(x;\lambda) N_S(x) x^2 \, dx,$$

$$\varkappa_{s_S}(\lambda)\gamma_S(\chi;\lambda) = \hat{V}_D \Delta (1-\alpha_S) \int_{x_m}^{X_M} Q_{s_S}(x;\lambda)\gamma_S^*(\chi, x;\lambda) N_S(x) x^2 \, dx,$$

where index S shows that these functions relate to soot. Here $\pi x^2 Q_{a_S}(x;\lambda)$, $\pi x^2 Q_{s_S}(x;\lambda)$ are absorbtion and scattering coefficients, $\gamma_S^*(\chi, x;\lambda)$ phase function of some soot particle of x radius. Note that $\tilde{\varkappa}_{a_S}$, $\tilde{\varkappa}_{s_S}$ do not depend on α_S, Δ, $\hat{V}_D$ values.

Absorption coefficient $\varkappa_a$, scattering coefficient $\varkappa_s$ and phase function γ of soot-dust atmosphere pollution are

$$\begin{aligned}\varkappa_a(\alpha_D, \alpha_S, \Delta; \hat{V}_D, \lambda) = \hat{V}_D[&(1-\alpha_S)\Delta \tilde{\varkappa}_{a_S}(\lambda) \\ &+ (1-\alpha_D)\tilde{\varkappa}_{a_D}(\lambda) + \alpha_D \tilde{\varkappa}_{a_{SD}}(\alpha_S\alpha_D^{-1}, \Delta; \lambda)];\end{aligned}$$

$$\begin{aligned}\varkappa_s(\alpha_D, \alpha_S, \Delta; \hat{V}_D, \lambda) = \hat{V}_D[&(1-\alpha_S)\Delta \tilde{\varkappa}_{s_S}(\lambda) \\ &+ (1-\alpha_D)\tilde{\varkappa}_{s_D}(\lambda) + \alpha_D \tilde{\varkappa}_{s_{SD}}(\alpha_S\alpha_D^{-1}, \Delta; \lambda)];\end{aligned}$$

$$\begin{aligned}\gamma(\alpha_D, \alpha_S, \Delta; \chi, \lambda) = [&(1-\alpha_S)\Delta \gamma_S(\chi,\lambda)\tilde{\varkappa}_{s_S}(\lambda) \\ &+ (1-\alpha_D)\gamma_D(\chi,\lambda)\tilde{\varkappa}_{s_D}(\lambda) + \alpha_D \tilde{\varkappa}_{s_{SD}}(\alpha_S\alpha_D^{-1}, \Delta, \lambda) \\ &\times \gamma_{SD}(\alpha_S\alpha_D^{-1}, \Delta; \chi, \lambda)] \hat{V}_D \varkappa_s^{-1}(\lambda).\end{aligned}$$

Note that functions Q_a, Q_s, γ^* were calculated due to algorithm (see [3]).

§ 3. Calculation results

In numerical calculation, the following parameters were used: $\Delta = 0.25$, $\lambda = 1.55 r_M$, $\hat{V}_D = 0.6$. So, $\varkappa_a$ and $\varkappa_s$ coefficients are the following functions: $\varkappa_a = \varkappa_a(\alpha_D, \alpha_S), \varkappa_s = \varkappa_s(\alpha_D, \alpha_S)$. Calculations were performed for two cases discussed in § 1.

At first, let us consider the case of interaction function $g(X,x) = g_1(x)$.

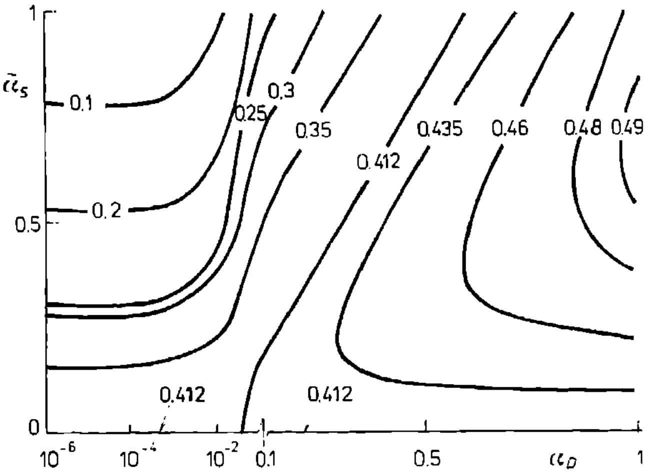

Fig. 1

Figure 1 shows isolines of function, depending on α_D and $\tilde{\alpha}_S$. In Figures 1 and 2, a dimension unit of function $\varkappa_a$ is km^{-1}. A case $\tilde{\alpha}_S = 0$, $0 \leqslant \alpha_D \leqslant 1$, corresponds to the medium of homogeneous soot and dust balls. Note that due to composite ($\tilde{\alpha}_S > 0$) in a region of parameters $0.1 \leqslant \{\tilde{\alpha}_S, \alpha_D\} \leqslant 1$ the absorption coefficient grows not greater than two times. The increasing of absorption coefficient is due to layers structure, but the decreasing is due to a process of falling down of composite particles. The case is characterized by the fact all soot, which is allowed to coagulate, involved in coagulation independently on dust quality, i.e. $\alpha_S = \tilde{\alpha}_S$. In the case where the coagulating dust part is small,

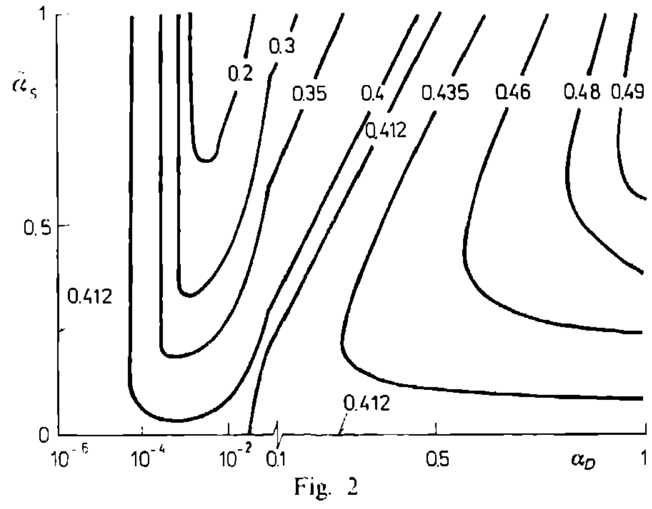

Fig. 2

the composite particles may reach extremely large dimensions. Correspondingly, when $\alpha_D \leqslant 10^{-4}$, all composite particles (and, $\tilde{\alpha}_S$ soot part) fall down.

The case connected with interaction function $g(X,x) = g_2(X,x)$ is more realistic. Figure 2 shows isolines of function $\varkappa_a$ depending on values α_D and $\tilde{\alpha}_S$. The case $\alpha_D = \tilde{\alpha}_S = 1$ corresponds to the medium of composite "soot-dust" balls; the case $\tilde{\alpha}_S = 0$, $0 \leqslant \alpha_D \leqslant 1$; $\alpha_D = 0$, $0 \leqslant \tilde{\alpha}_S \leqslant 1$ corresponds to the medium of homogeneous soot and dust balls. The behaviour of function $\varkappa_a$ in Figures 1 and 2 differs just only in a region $\alpha_D \lesssim 0.1$. This difference is based on the following. In a case $g(X,x) = g_2(X,x)$ and $\alpha_D \lesssim 10^{-3}$, there is a situation when all composite particles fall down ($\alpha_S < \tilde{\alpha}_S$) and, as a result, value $\varkappa_a$ mainly depends on $(1-\alpha_S)$ soot part. And we have $\alpha_S(\alpha_D, \tilde{\alpha}_S) \to 0$ as $\alpha_D \to 0$. In a region $0.1 \leqslant \alpha_D \leqslant 1$ $\varkappa_a$ functions in Figures 1 and 2 coincide, because in this case we have $g_2(X,x) \approx g_1(x)$.

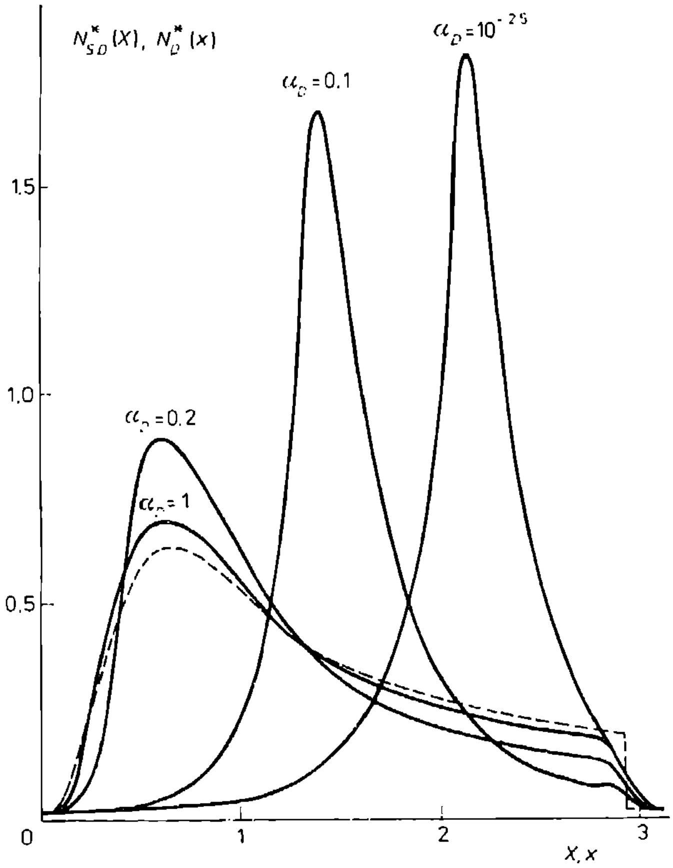

Fig. 3

Figure 3 shows size distributions of dust and composite particles; the distributions were calculated numerically. For obviousness graphs show $N^*_D(x) = N_D(x)x^3$ and $N^*_{SD}(X) = D\tilde{N}_{SD}(X)X^3$ values, where $\tilde{N}_{SD}(X)$ is a function of distribution of composite particles on outer radius X with the norm condition

$$\int_{x_m}^{X_M} D\tilde{N}_{SD}(X)X^3 dX = 1.$$

A dashed line shows the initial size distribution of dust particles $N_D^*(x)$. Graphs are made for several values α_D, when $\alpha_S = 1$. We see that in region $0.2 \leqslant \alpha_D \leqslant 1$ the number of composite particles with the middle dimension of coagulation nucleus increases. In this case, the distribution function becomes more stretched when α_D decreases. For small α_D values ($\alpha_D \leqslant 10^{-2}$), the maximum of function $N_{SD}^*(x)$ grows and displaces to large dimensions. Here the main role is played by small particles, as large and middle particles have already fallen down.

Functions $\alpha_D^*(\alpha_D, \alpha_S)$ and $\alpha_S^*(\alpha_D, \alpha_S)$, which define dust and soot parts, fallen down are of great interest. They are

$$\alpha_D^*(\alpha_D, \alpha_S) = \alpha_D\Big(1 - \int_{x_m}^{x_M} N_{SD}(x)x^3\,dx\Big),$$

$$\alpha_S^*(\alpha_D, \alpha_S) = \alpha_S \frac{\int_{x_m}^{x_M} (N_D(x) - N_{SD}(x))(X^3(x) - x^3)dx}{\int_{x_m}^{x_M} N_D(x)(X^3(x) - x^3)dx}.$$

In the Figure 4, solid line shows functions $\alpha_D^*(\alpha_D)/\alpha_D$ and dashed line — functions $\alpha_S^*(\alpha_D)/\alpha_S$. For these functions, value $\tilde{\alpha}_S$ is equal to 1. There exists value α_D, when $\alpha_D^*/\alpha_D = \alpha_S^*/\alpha_S = 1$. It characterizes the situation when all soot and dust, that was involved in a coagulation, have fallen down. Up to this point $\alpha_S^*(\alpha_D)/\alpha_S$ function increases, but $\alpha_D^*(\alpha_D)/\alpha_D = 1$. After this point the functions observed decrease, because of the fact, that the process of falling down relaxes, when the quality of coagulated particles grows.

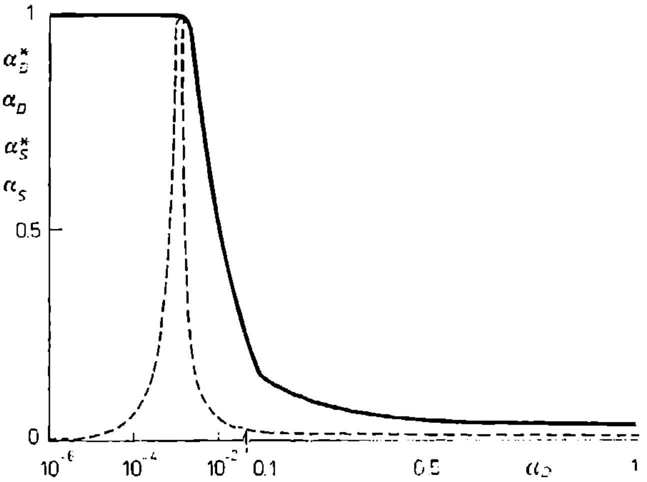

Fig. 4

Figure 5 shows phase function $\gamma(\alpha_D, \alpha_S, \Delta; \chi, \lambda)$ for considered polydisperse ensemble of particles, depending on cosines scattering angles χ. Dashed line corresponds to $\alpha_D = \alpha_S = 1$ case (composed particles medium), solid line to $\alpha_D = \alpha_S = 0$ case (homogeneous particles medium). Parameters Δ and λ were defined above. We see that phase functions is sharply stretched in forward direction (is sharply increased as $\chi \to 1-0$). This behaviour is typical for difraction picture which occurs when scattering of plane electromagnetic waves on particles is larger than wavelength.

Numerical calculations show that composity of particles practically does not change (changes are less than 10%) scattering coefficient, phase functions.

In calculation performed optical depth of soot column of 1 gr mass and 1 m^2 cross-section is equal to 2.0–2.1. This result is in a good agreement with date [1] where optical depth is equal to 2.2–2.4.

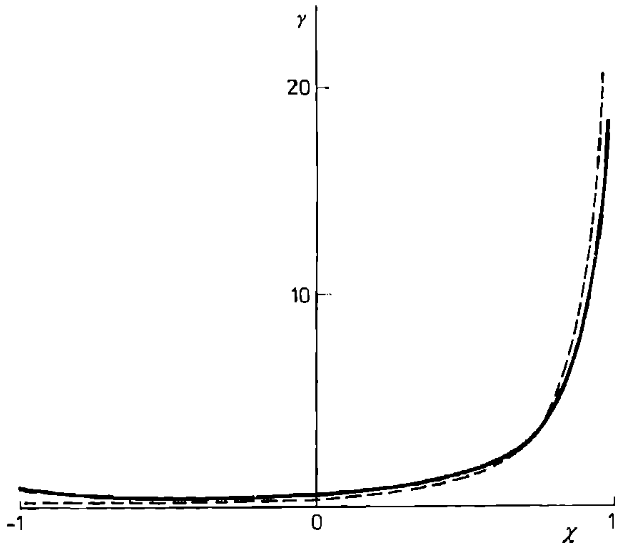

Fig. 5

Conclusions

From the numerical calculations performed we may draw a conclusion that, due to assumptions made on the optical characteristics of a medium which consists of homogeneous "soot" and "dust" particles and a medium which consists of composite "soot-dust" particles, are practically equivalent.

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