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INVESTIGATION OF RADIATION SCATTERING BY SULFURIC ACID DROPS*

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A study was made of the scattering of radiation by polydispersed drops of sulfuric acid, with gamma and log-normal distributions of the drops according to size. Scattering functions, attenuation coefficients, and backscattering coefficients were calculated.

Radiative transfer plays an important role in the development of thermal and other dynamic processes on atmospheric planets, ultimately determining their thermodynamic state. For earth, the thermodynamic state of the atmosphere and its dynamics affect global climate and, thus, environmental conditions for human life. The problem of the climate on the earth has become important in recent years since, given the present state of science, technology, and industry, man's activities may be affecting the climate on a global as well as local scale. The earth's atmosphere is being continually fouled by industrial wastes on an enormous scale, comparable to natural contamination of the atmosphere from volcanoes, dust storms, and hurricanes. Incomplete combustion of fossil fuels leads to pollution of the atmosphere with carbon dioxide, sulfur dioxide, etc. and to a qualitative change in the composition of the atmosphere. All this has a significant effect on processes of radiative transfer in the atmosphere and on global climate.

Many atmospheric processes are determined by solar radiation, along with thermal radiation from the heated earth.

Study of the radiative state of the atmosphere is being given much attention in an international program of investigations of global atmospheric processes (PIGAP); study of the atmospheres of other planets by means of spacecraft has also become important.

In connection with the rapid growth of industry and proliferation of hazardous wastes, the problem of protecting the atmosphere from pollution has become more acute in recent decades and can be solved only by cooperation on a global scale, involving the efforts of many nations. Calculations show that the following tonnages of natural and industrial pollutants are received by the atmosphere every year [1]: carbon dioxide $7 \cdot 10^{10}$ and $1.5 \cdot 10^{10}$; SO_2 $1.4 \cdot 10^8$ and $7.3 \cdot 10^7$; natural H_2S sulfates $1.3\text{-}2 \cdot 10^8$, industrial H_2S sulfates $1.3\text{-}2 \cdot 10^8$

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natural nitrogen N_2 $1.4 \cdot 10^9$ and $7.3 \cdot 10^7$, natural ozone O_3 $2 \cdot 10^9$. It is clear from the above data that, after natural carbon dioxide, gaseous and solid sulfates contribute the most in terms of quantity to atmospheric contamination. Combining with water vapor in the atmosphere, these sulfates form sulfuric acid H_2SO_4. Recent studies of the stratosphere have established that it contains a fairly large quantity of concentrated (up to 75%) sulfuric acid. The stratosphere extends around the earth from an altitude of 11-50 km and is characterized by a drop in temperature to -56.5°C at altitudes of 10-15 km, a constant temperature of -56.5°C at 12-25 km, and an increase in temperature from -56.5°C to 1°C at altitudes from 25 to 54 km. The increase in temperature in the upper layers of the atmosphere is due to intensive absorption of ultraviolet solar radiation by ozone O_3.

As studies of the atmosphere of Venus have shown, the clouds of this planet also contain large quantities of sulfuric acid and contaminants, so that it is natural to suggest that sulfuric acid aerosols play an important role in scattering solar radiation in planetary atmospheres [2-7].

Studies of the aerosol composition of the atmosphere [3-5] indicate that sulfuric acid H_2SO_4 is the most likely component of aerosols in the stratosphere. Calculations have established that stratospheric aerosols of H_2SO_4 may exist either in the form of drops of supercooled liquid or as a solid phase. Remsberg [7] proposes that the optical properties of solid-phase H_2SO_4 are not appreciably different from those of the liquid phase [8].

Scattering of radiation by single drops of H_2SO_4 occurs on the assumption that the drop is a sphere with radius r_0, in conformity with the rigorous theory of diffraction by a sphere developed in 1909 by Mie [9]. The main parameters for computer calculation are: 1) diffraction parameter $\rho = 2\pi r_0/\lambda$, 2) wavelength of light λ, 3) complex refractive index $m = n - i\varkappa$ where n is its real part and $\varkappa$ is its imaginary part.

Absorption is very small for H_2SO_4 drops [2] and may be ignored in calculations, so that the drops may be practically regarded as nonabsorbers of light energy. The dependence of the real part of the refractive index on the wavelength of light for sulfuric acid is given in Table 1.

The scattering coefficient is defined as the ratio of the energy flux scattered by a particle in all directions to the incident flux and in conformity with the Mie theory [9] is determined by the expressions

$$k_s = \frac{2}{\rho^2} \sum_{l=1}^{\infty} \frac{l^2(l+1)}{2l+1} (|c_l|^2 + |b_l|^2), \tag{1}$$

where $|c_l|$ and $|b_l|$ are coefficients in the Mie series defining the field scattered by the particle and expressed through Bessel and Hankel functions [9-15].

The scattering function is defined as the angular distribution of radiation intensity and is determined at large distances from the particle by the expression

$$I(\beta) = I_0 \frac{i_1 + i_2}{2k^2 R^2}. \tag{2}$$

The normalized scattering function is written in the form

$$\bar{I}(\beta) = \frac{I(\beta) R^2}{I_0 \pi r_0^2 k_s}, \tag{3}$$

where I_0 is the intensity of the incident radiation; $k = 2\pi/\lambda$ — the wave number; I is the intensity of the scattered light; i_1 and i_2 are orthogonal components of the scattered radiation polarized perpendicular and parallel to the scattering plane, respectively.

The values of i_1 and i_2 are represented in the form of a Mie series [15]:

$$i_1 = \left| \sum_{l=1}^{\infty} (c_l Q_l + b_l S_l) \right|^2, \quad i_2 = \left| \sum_{l=1}^{\infty} (c_l S_l + b_l Q_l) \right|^2. \tag{4}$$

TABLE 1. Backscattering Coefficient b, Scattering Coefficient k_s', and the First Two Coefficients a_0 and a_1 in a Legendre Polynomial Expansion of the Scattering Functions of H_2SO_4 Drops for Different Wavelengths λ and Refractive Indices n

n	λ	k_s'	$b/4\pi$	$a_0/4\pi$	$a_1/4\pi$
			For gamma distribution		
1.344	2.5	0.154394	0.0328389	0.0794947	0.0544983
1.384	2.0	0.398292	0.0296503	0.0794644	0.0793754
1.398	1.667	0.745580	0.0268430	0.0794362	0.101011
1.406	1.429	1.20413	0.0245580	0.0794116	0.118289
1.413	1.25	1.77946	0.0227525	0.0793908	0.131594
1.418	1.111	2.44030	0.0213344	0.0793737	0.141721
1.422	1.00	3.16660	0.0202054	0.0793597	0.149510
1.424	0.909	3.91649	0.0192806	0.0793483	0.155670
1.426	0.833	4.69423	0.0185202	0.0793395	0.160558
1.427	0.769	5.45592	0.0178858	0.0793329	0.164500
1.427	0.714	6.18649	0.0173442	0.0793281	0.167762
1.431	0.556	8.85054	0.0159912	0.0793247	0.175222
1.452	0.357	12.1373	0.0156754	0.0793674	0.174493
			For log-normal distribution		
1.344	2.5	0.175661	$2.80511\cdot10^{-2}$	$7.94463\cdot10^{-2}$	$9.13878\cdot10^{-2}$
1.384	2.0	0.420517	$2.54748\cdot10^{-2}$	$7.94173\cdot10^{-2}$	0.110544
1.398	1.667	0.737060	$2.36691\cdot10^{-2}$	$7.93968\cdot10^{-2}$	0.123569
1.406	1.429	1.12494	$2.23231\cdot10^{-2}$	$7.93826\cdot10^{-2}$	0.133066
1.418	1.111	2.08792	$2.05179\cdot19^{-2}$	$7.93665\cdot10^{-2}$	0.145514
1.422	1.00	2.62476	$1.98987\cdot10^{-2}$	$7.93619\cdot10^{-2}$	0.149705
1.424	0.909	3.16932	$1.93824\cdot10^{-2}$	$7.93584\cdot10^{-2}$	0.153160
1.426	0.833	3.73063	$1.89584\cdot10^{-2}$	$7.93556\cdot10^{-2}$	0.155963
1.427	0.714	4.82281	$1.82603\cdot10^{-2}$	$7.93513\cdot10^{-2}$	0.160480
1.431	0.556	6.95386	$1.73483\cdot10^{-2}$	$7.93502\cdot10^{-2}$	0.166076
1.452	0.357	11.3283	$1.66153\cdot10^{-2}$	$7.93656\cdot10^{-2}$	0.169632

In the absence of absorption, the attenuation coefficient is equal to the scattering coefficient:

$$k_{\text{att}} = k_s + k_{\text{absr}} = k_s. \qquad (5)$$

If $k_s(r, \lambda, n, \chi)$ is the coefficient for scattering of light by a certain particle, then in the presence of N particles in a unit volume of the medium the volume coefficient of scattering by particles of identical radius r is equal to

$$k_s = N\pi r^2 k_s(r, \lambda, n, \varkappa). \qquad (6)$$

The microstructure of a polydispersed medium is described by a distribution function defining its composition according to size. If the medium is dynamic, then the function generally depends on many arguments: time t, coordinate x_i, velocity v_i, particle size r, and other parameters. An integral-differential kinetic equation [16, 17] is used to determine the function

$$\frac{\partial f_i}{\partial t} + (\vec{v}\cdot\nabla f_i) + \frac{\vec{F}_i}{m_i}\nabla_v f_i + \frac{\partial f_t \dot{r}}{\partial r} = I_{st\,\text{els}} + I_{st\,\text{inels}}, \qquad (7)$$

where $\vec{F}_i/m_i$ is the force acting between drops (particles); $\dot{r}$, rate of change in drop (particle) dimensions; $I_{st\,\text{inels}}$, integral of the inelastic collisions, also accounting for processes such as fragmentation, etc.

In principle, solving this equation would determine distribution function f. In practice, investigators often examine model problems where the distribution function of the particles with respect to size alone is known from indirect measurements made by photographic, holographic, or other methods.

According to Remsberg's data [7], the distribution of aerosol drops in the stratosphere may be approximated by the following distribution function:

$$f = 0.142r^{-3.82},\ 0.3 \leqslant r \leqslant 1\ \mu\text{m};$$
$$f = 2.28 \cdot 10^3 r^{4.19},\ 0.1 \leqslant r \leqslant 0.3\ \mu\text{m};$$
$$f = 0.435 \cdot 10^4,\ 0.03 \leqslant r \leqslant 0.05\ \mu\text{m}, \tag{8}$$

where f is the number of particles per 1 cm^3, the dimensions of which fall within the interval r + dr.

Apart from Remsberg's distribution function (8), there are several other distribution functions for aerosols with respect to size in the literature on atmospheric physics [18-25]. For small (R < 10 μm) drops, Young's distribution function [26] is widely used

$$f(r) \cong ar^{\nu}, \tag{9}$$

where ν and a are certain parameters dependent on the height and type of particles.

In 1954, Levin [27, 28] proposed that the log-normal law of Kolmogorov [29] be used to approximate the distribution functions of cloud drops:

$$f(r) = B \exp\left\{-\frac{1}{2}\frac{1}{\ln^2 \sigma}\left(\ln^2 \frac{r}{r_0}\right)\right\}, \tag{10}$$

where σ is the standard deviation of the distribution; r_0 is the most probable radius; B is a parameter.

Shifrin and D'yachenko [30, 31] showed that many actual size-distribution functions for cloud and mist particles may be approximated by the function

$$f(r) = Ar^{\mu} e^{-\gamma r^b}, \tag{11}$$

where A, μ, γ, and b are certain parameters that can be chosen with a known empirical or assigned distribution function by methods of the theory of approximations or the least-squares method. A particular case of this function is the gamma distribution obtained at b = 1 and proposed by Levin in 1958 [28]:

$$f(r) = Ar^{\mu} e^{-\gamma r} = Ar^{\mu} e^{-\mu \frac{r}{r_0}}, \tag{12}$$

where μ is a parameter characterizing the relative half-width of the distribution function; r_0 is the most probable or modal radius of the principles; $\gamma = \mu/r_0$, at μ = 2 this is the Khrgian—Mazin distribution [32].

According to the data in [2], the distribution of H_2SO_4 drops may be approximated by a modified gamma distribution

$$f(r) = 4 \cdot 10^5 r^2 \exp(-20r), \tag{13}$$

in which the modal radius $r_0 = \mu/\gamma = 2/20 = 0.1$ μm, and the half-width of function (13) at which $f(r) = 0.5f(r_0)$ is equal to

$$\theta = 2.48r_0/\sqrt{\mu} = \frac{2.48 \cdot 0.1}{\sqrt{2}}\ \mu\text{m}.$$

If the particle-size distribution function f(r) is known, then the total number of particles in a unit volume

$$N = \int_{r_1}^{r_2} f(r)\,dr. \tag{14}$$

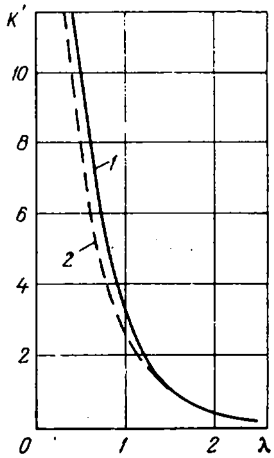

Fig. 1

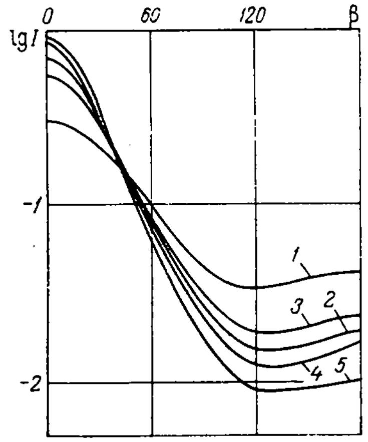

Fig. 2

Fig. 1. Dependence of scattering coefficient k_s' on wavelength λ, μm of radiation for different distribution functions: 1) gamma distribution, $f(r) = 4 \cdot 10^5\, r \exp[-20\, r]$; 2) log-normal distribution, $f(r) = 1.8 \cdot 10^3 \exp[-\ln^2 \frac{r}{r_m} / 2 \ln^2 \sigma]$, $\sigma = 2$, $r_m = 0.035\ \mu m$.

Fig. 2. Dependence of form of scattering function of sulfuric acid aerosols on refractive index n and wavelength λ for a log-normal distribution: 1) n = 1.344, λ = 2.5 μm; 2) 1.418 and 1.111, respectively; 3) 1.406 and 1.429; 4) 1.426 and 0.833; 5) for the gamma distribution, n = 1.406 and λ = 1.429 μm. β, deg.

In particular, for distribution (12) we have

$$N = A \int_0^\infty r^\mu e^{-\gamma r^b} dr = \frac{A}{b} \gamma^{-(\mu+1)} \Gamma\left(\frac{\mu+1}{b}\right), \tag{15}$$

while in the case of the modified gamma distribution (13) at b = 1

$$N = A\gamma^{-(\mu+1)} \Gamma(\mu+1). \tag{16}$$

At $\mu = 2$, $A = 4 \cdot 10^5$, and $\gamma = 20$ 1/μm

$$N = 4 \cdot 10^5 \Gamma(3)\, 20^{-3} = 4 \cdot 10^5 \cdot 1 \cdot 2 \frac{1}{8 \cdot 10^3} = 10^2 \frac{\text{particles}}{\text{cm}^3}.$$

Using Eq. (12), it is easy to also compute the second moment of the distribution function

$$\int_0^\infty r^2 f(r)\, dr = A \int_0^\infty r^{\mu+2} e^{-\gamma r^b} dr = \frac{A}{b} \gamma^{-(\mu+2+1)} \Gamma\left(\frac{\mu+2+1}{b}\right) \tag{17}$$

and at $b = 1$, $\mu = 2$, $\gamma = 20$, and $A = 4 \cdot 10^5$ we have

$$4 \cdot 10^5 \Gamma(5)\, \gamma^{-5} = 4 \cdot 1 \cdot 2 \cdot 3 \cdot 4 \frac{1}{2^5 \cdot 10^5} = 3\ \mu\text{m}.$$

Mean particle size is characterized by the ratio of the third moment of the distribution function to the second:

$$r_{32} = \int_{r_1}^{r_2} r^3 f(r)\, dr \Big/ \int_{r_1}^{r_2} r^2 f(r)\, dr \tag{18}$$

and in the case of the gamma distribution (12)

$$r_{32} = r_0\left(1+\frac{3}{\mu}\right) = 0.1\left(1+\frac{3}{2}\right) = 0.25\ \mu\text{m}, \tag{19}$$

while the mean diffraction parameter

$$\rho_{32} = \frac{2\pi r_{32}}{\lambda} = \frac{2\pi r_0}{\lambda}\left(1+\frac{3}{\mu}\right) = \frac{2\pi\cdot 0.1}{\lambda}\left(1+\frac{3}{2}\right). \tag{20}$$

At wavelengths 2.5; 2.0; 1.667; 1.429; 1.25; 1.111; 1.00; 0.909; 0.883; 0.714; 0.556; 0.357, the mean radius ρ_{32} of the drops for a gamma distribution is, respectively, equal to 0.63; 0.785; 0.945; 1.1; 1.255; 1.41; 1.57; 1.73; 1.885; 2.2; 2.82; 44.

The scattering coefficient of polydispersed media with a function is determined by the expression

$$k_s' = \int_{r_1}^{r_2} r^2 k_p(r,\ \lambda,\ n,\ \varkappa) f(r)\,dr \Big/ \int_{r_1}^{r_2} r^3 f(r)\,dr, \tag{21}$$

where $k_s(r, \lambda, n, \chi)$ is the scattering coefficient. Its value is shown in Fig. 1 and Table 1 in relation to wavelength λ and type of distribution function $f(r)$.

In the case where the distribution is in the form of a gamma function for Rayleigh particles, an expression was obtained for the scattering coefficient at $\varkappa = 0$ (11):

$$k_s = \frac{3}{4} c_v \rho_{32}^3 \frac{(\mu+6)(\mu+5)(\mu+4)}{(\mu+3)} \frac{8}{3} \frac{[(n^2+2)\,n^4]^2}{(n^2+2)^4}. \tag{22}$$

The scattering function for polydispersed media with a particle-size distribution function $f(r)$ is determined from the expression

$$I'(\beta) = \int_{r_1}^{r_2} [i_1(\beta) + i_2(\beta)]\, f(r)\,dr \Big/ \int_{r_1}^{r_2} r^3 f(r)\,dr, \tag{23}$$

The appearance of the scattering function is shown in Fig. 2 for a gamma distribution and log-normal distribution.

The coefficient of scattering-function asymmetry is the ratio of the flux scattered in the leading hemisphere to the flux scattered in the trailing hemisphere and for monodispersed particles is determined from the formula

$$\eta = \int_0^{\pi/2} I(\beta) \sin\beta\,d\beta \Big/ \int_{\pi/2}^{\pi} I(\beta)\sin\beta\,d\beta. \tag{24}$$

The normalized scattering function may be expanded into a series of Legendre polynomials P_n:

$$I'(\beta) = \sum_{n=0}^{\infty} a_n P_n(\cos\beta),\ a_0 = 1, \tag{25}$$

where a_n is the expansion coefficient. The first two coefficients in the expansion of $a^* = a_n/4\pi$ are shown in Table 1. The backscattering coefficient was also calculated

$$b = \frac{1}{2\pi}\int_0^{\pi} \beta I'(\beta)\sin\beta\,d\beta,$$

and these values are also shown in Table 1.

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REFLECTIVITY OF A LAYER OF POLYDISPERSED WATER DROPS

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UDC 536.3

Spectral and integral reflectivities of a semi-infinite layer of water droplets are calculated. Particle size distribution is assumed to be of Γ-form. The results of numerical calculations for integral reflectivity are presented by a simple formula.

The calculations are based on the assumption of the "softness" of the particles $|n-i\varkappa| \approx 1$. Hulst's formula [1, 2] is valid for determining the dimensionless attentuation coefficient of such particles

$$K = 2 - \frac{4}{\rho'} \sin \rho' + \frac{4}{\rho'^2}(1 - \cos \rho'), \tag{1}$$

where $\rho' = 2\rho(n-1)$, $\rho = 2\pi r/\lambda$. We will use a modification of this formula for the dimensionless absorption coefficient

$$K_a = K_{a\infty}\left\{1 - 2\left[\frac{1}{\omega^2} - e^{-\omega}\left(\frac{1}{\omega} + \frac{1}{\omega^2}\right)\right]\right\}, \tag{2}$$

where $K_{a\infty}$ is the dimensionless absorption coefficient of particles with an infinite radius:

$$\omega = 4a(n)\varkappa\rho/K_{a\infty}. \tag{3}$$

The appearance of function $a(n)$ is shown in [3], and the following approximate expression was derived in [4]

$$a(n) = n^2\left[1 - \left(\frac{n^2-1}{n^2}\right)^{3/2}\right]. \tag{4}$$

Table 1 shows values of $K_{a\infty}$ calculated in accordance with the electromagnetic theory at $\varkappa\rho \gg 1$, $\varkappa \ll n-1$. Table 2 shows the results of calculations with Eq. (2) compared to the results of a rigorous solution. Equation (1) for K_a leads to a substantial divergence from the correct values compared to Eqs. (2) and (3).

To solve the transfer equation, apart from K and K_a we need to know the scattering function. We will limit ourselves to an approximate solution with one functional parameter, for which we will use the mean cosine of the scattering angle for minimum scattering. The following design formula was derived from an analysis of data obtained on an electronic digital computer in accordance with the Mie theory

$$\bar{\mu} = \bar{\mu}_\infty[1 - e^{-1.2\rho}(1 + 1.2\rho)], \tag{5}$$

and is valid for $n \approx 1.3$ and $\varkappa \ll 1$. The following approximate expression was derived for the

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