Radiation absorption asymmetry factor and photophoresis of aerosols

S.A. Beresnev and L.B. Kochneva

Ural State University, Ekaterinburg

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We present theoretical analysis of the role of the radiation absorption asymmetry factor J_1 in photophoresis of aerosol particles. The efficiency of applying the mentioned microphysical optical characteristic in predicting direction and magnitude of the photophoretic force and velocity is demonstrated. Based on the Mie theory, within the framework of optically homogeneous spherical particle model, the J_1 values for weakly, moderately, and strongly absorbing model particles are calculated. The J_1 dependences on size parameter ρ and incident radiation wavelength λ are considered. The known asymptotic results for J_1 obtained for the Rayleigh limit and for the limit of ray optics are analyzed. Further investigations of the asymmetry factor properties for the model of two-layer particles taking into account the spectral distribution of incident radiation are considered.

Introduction

Absorption of electromagnetic waves by atmospheric aerosols is the primary cause of different dynamical processes including motion of particles in the field of radiation. When analyzing such phenomena, it is expedient to distinguish three related parts of the problem: electrodynamics problem (calculation of the characteristics of absorbed electromagnetic field in the particle volume), thermal problem (calculation of temperature field in the volume and on the surface of a particle) and gas-kinetic problem (description of heat and mass transfer in the gas phase, calculation of forces, force moments, particle velocity, etc.).[1,2] The above noted complexity of the problem is characteristic in full measure of aerosol photophoresis, i.e., the motion of nonuniformly heated suspended particles in the field of directed radiation. The history of the development of the ideas on the causes and nature of this phenomenon is quite extraordinary. The author of the term “photophoresis” F. Erenhaft explained the various observed forms of the motion of particles in the radiation field by an “electrodynamical effect of the second type” (different from the phenomenon of light pressure).[3] Such an interpretation was explained by the fact that dependence of the aerosol motion velocity on gas pressure was not observed in the experiments by the author, and the shape of the particles under investigation strongly differed from spherical. Shortly after this a more realistic interpretation of this phenomenon was suggested[4]: photophoresis was explained by the known radiometric effect for particles not uniformly heated by radiation. Let us note that the principle of splitting the problem into the electrodynamical and gas-kinetic parts was formulated for the first time in Ref. 4. A comprehensive review of the early theoretical and experimental papers is presented in Ref. 5.

Traditional understanding of the nature of radiometric photophoresis is the following. The particles in the field of directed radiation absorb electromagnetic waves, the energy of which is then converted into heat and causes nonuniform heating of its surface. In its turn, radiometric (in this case photophoretic) force affects the nonuniformly heated particle placed to a rarefied gas and drives it. If the particle shape has been different from a sphere, the force moment appears, which leads to complex and not always linear motion of aerosols in the field of directed radiation.[5,6] However, the primary cause of all these various forms of motion is the heterogeneous distribution of the absorbed electromagnetic energy over the particle volume.

First calculations of the distribution of the electromagnetic field density over the volume of spherical particles by Mie theory[7,8] confirmed the necessity of detailed analysis of electromagnetic part of the problem and allowed the qualitative explanation of the nature of positive and negative photophoresis (the motion of particles along and against the direction of propagation of radiation) to be given. However, calculations of the intensity of the internal field do not provide for unambiguous data on the direction and magnitude of the photophoretic force and the velocity of a particle motion. In this regard, the factor of asymmetry of the absorption of radiation J_1, related to the complex of microphysical optical characteristics (MOC) of the particulate matter responsible for the dynamics of aerosols in the radiation field, is a comprehensive characteristic.[1,2] First this parameter was determined as the asymmetry factor of the particle surface temperature.[9,10] This terminology is useful in analyzing the thermal problem, however the way for calculation of it is electrodynamics. In the general case, the factor J_1 is a function of intensity of the internal field in the particle volume and depends on the value of the

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diffraction parameter and the complex refractive index of the matter. Let us note that the same or similar, by the physical meaning, characteristics were given different names in other papers (for example, factor of asymmetry[8,11,12] and photophoretic factor of asymmetry[13] coinciding with the definition[9,10]; asymmetry of sources,[14] which has a little bit different meaning).

It is known[15] that electrodynamical part of the problem is separated from the thermal and gas-kinetic parts in the frameworks of the theory of photophoresis linear on the intensity of incident radiation. This makes it possible to write the formulas for the force and velocity of a particle motion in the form of the product of electrodynamical and gas-kinetic functions. The factor J_1 includes all necessary data on the optical properties of particles and incident radiation. At a fixed particle radius, radiation wavelengths, and the complex refractive index this numerical parameter characterizes not only the direction of particle motion but to a large extent determines the absolute values of the photophoretic force and velocity.

The ways for solving the problem without selecting the factor J_1 in an explicit form (for example, Refs. 16 and 17, where electromagnetic description is performed only for the intensity of the internal field) that are possible, though, not for all gas-kinetic regimes, make the problem mathematically more complicated and can lead to the errors in calculations and even to the errors in final results.[16]

Practically no systematic analysis of the properties of the factor J_1 as a function of the determining parameters of the problem was carried out. Only fragmentary data can be found on this topic in many cited papers, however, on the whole, the problems of the general dependences of the factor J_1 on the diffraction parameter, wavelength of incident radiation, complex refractive index of particulate matter of the principal types of atmospheric aerosol are not studied. This paper, continues investigations of the complex of MOC[1,2] responsible for the dynamics of particles in the radiation field, should compensate for this gap. Besides, analysis of the factor J_1 is directly related to the problem of photophoretic levitation of particles in the stratosphere and the development of new models of the vertical transfer of aerosols at large heights.[18]

1. Analysis of the problem of photophoresis and determination of the factor J_1

Let us consider a spherical particle of the radius R_0 in the field of a plane wave of a monochromatic radiation of the intensity **I**. Heating of the particle occurs resulting from absorption of electromagnetic energy. It is described by non-stationary heterogeneous thermal conductivity equation

$$c_p \rho_{\mathrm{p}} \frac{\partial T_{\mathrm{p}}}{\partial t} = \lambda_{\mathrm{p}} \Delta T_{\mathrm{p}} + q(\mathbf{r}), \qquad (1)$$

where c_p is the specific thermal capacity, ρ_{p} is the density, λ_{p} is the thermal conductivity coefficient, T_{p} is local temperature inside the particle.

$$q(\mathbf{r}) = 2\pi k k_0 \, I B(\mathbf{r})$$

is the volume density of the inner heat sources;

$$B(r,\theta,\varphi) = \frac{1}{2\pi}\int_0^{2\pi} \frac{|E(r,\theta,\varphi)|^2}{E_0^2}\,\mathrm{d}\varphi = B(r,\theta,\frac{\pi}{4})$$

is the dimensionless function of the sources of electromagnetic energy in the case of nonpolarized incident radiation. Here $E(r, \theta, \varphi)$ is the local intensity of electric field inside the particle; E_0 is the electric field strength of the incident wave; $k_0 = 2\pi/\lambda$ is the wave number, λ is the wavelength, $m = n + ik$ is the complex refractive index of the particle substance, t is the time, $\mathbf{r}$ is the radius-vector in the spherical coordinate system r, θ, φ with the origin at the particle center. General solution of Eq. (1) in the stationary case can be represented in the form[15]:

$$T_{\mathrm{p}}(x,\theta) = \sum_{l=0}^{\infty} P_l(\cos\theta)[A_l x^l + B_l x^{l+1}], \qquad (2)$$

where $P_l(\cos\theta)$ are the Legendre polynomials of the first kind, θ is the polar angle, $x = r/R_0$ is the dimensionless radial coordinate. The expansion coefficients A_l and B_l can be determined from the boundary conditions on the particle surface and take into account the characteristics of the incident radiation and the parameters of the gas medium. The coefficients B_l are determined only by the characteristics of incident radiation and thermal conductivity of the particle $B_l = -IR_0 J_1/\lambda_{\mathrm{p}}$, and the coefficients A_l depend on both thermal properties of the gas and particle substance, and on the gas-kinetic regime realized.[15] The momenta of the electromagnetic energy source function J_1 involved into the coefficients B_l can be represented in the form

$$J_l = (2l+1)kn\rho \int_0^{\pi} \sin\theta \;\; P_l(\cos\theta)\mathrm{d}\theta \times$$
$$\times \int_0^1 x^{l+2} B(x,\theta,\varphi=\pi/4)\mathrm{d}x, \qquad (3)$$

where $\rho = 2\pi R_0/\lambda$ is the particle diffraction parameter. The momentum of zero order at $l = 0$ is proportional to the dimensionless radiation absorption efficiency factor $J_0 = (1/4)\,Q_{\mathrm{abs}}$, and the momentum of the first order at $l = 0$ is the radiation absorption asymmetry factor J_1. The results for thermal and electrodynamic problems (1)–(3) are not the final result of solving the problem of photophoresis, because it is closed by the solution of the gas-kinetic problem. The molecular-kinetic theory of radiometric photophoresis, which covers the entire range of variations of the Knudsen number $\mathrm{Kn} = l_g / R_0$, where l_g is the mean free path of gas molecules) taking into account optical, thermal, and accommodation properties of the gas–particle system, is presented in Ref. 15. Its principal results are formulas for the photophoretic force and the velocity of the particle motion. In particular, the photophoretic force

$$\mathbf{F}_{\mathrm{ph}} = -\frac{2\pi}{3}\left(\frac{\pi M}{8RT_0}\right)^{1/2} R_0^2 \;\; \mathbf{I} \;\; J_1(\rho, m) F(\mathrm{Kn}, \Lambda), \quad (4)$$

where T_0 is the gas temperature far from the particle, $\Lambda = \lambda_p / \lambda_g$ is the thermophysical parameter equal to the ratio of thermal conductivity values of the gas and a particle, M is the molar mass of the gas, R is the universal gas constant. The value $F(\mathrm{Kn}, \Lambda)$ is a complex function of the number Kn, parameter Λ, accommodation coefficients of the pulse and energy of gas molecules on the particle surface are, in the general case, numerically calculated in solving the gas-kinetic Boltzmann equation. Linearity of Eq. (4) with respect to electrodynamical and gas-kinetic characteristics is caused by the statement of the problem linear with respect to the intensity of incident radiation $\mathbf{I}$.

The factor J_1 is the mean integral characteristic of the source function $B(\mathbf{r})$ and estimates the position of the "gravity center" of the sources of electromagnetic energy in the particle volume:

$$J_1 = 3nk\rho \int_0^{\pi} \sin\theta \;\; P_1(\cos\theta) \mathrm{d}\theta \int_0^1 x^3 B(x, \theta, \varphi = \pi/4) \mathrm{d}x. \quad (5)$$

As shown in Refs. 9 and 10, the factor J_1 does not depend on the degree of polarization of radiation incident on the particle. The same property is characteristic of the absorption efficiency factor[2] Q_{abs}, which is also necessary for solving the problem of photophoresis. Let us note that the relationship (5) determines the factor J_1 for the case of monochromatic incident radiation, i.e., is the spectral microphysical characteristic (the account for spectral composition of the incident radiation is discussed below). The value of the factor J_1 varies within the limits $-0.5 < J_1 < 0.5$. Negative values correspond to the prevalent heating of the front side of the particle and refer to the so-called "positive" photophoresis (motion of particles along the direction of radiation propagation), and positive values correspond to heating of the back side and "negative" photophoresis (motion against the radiation propagation direction). Analysis of the results obtained using the molecular-kinetic theory of the phenomenon[15] shows that in varying any gas-kinetic and thermophysical parameters, as well as the accommodation coefficients in the entire possible range of their values does not lead to any change of the sign of the photophoretic force and velocity. Thus, the direction of particle motion is completely determined by the sign of the factor J_1.

2. Algorithm for calculating the factor J_1 by Mie theory

To calculate the dimensionless source function $B(\mathbf{r})$, it is necessary to use the solution of Mie problem for internal field[13] in Eq. (5):

$$E_r = -\frac{\cos\varphi}{m^2\rho^2}\sum_{l=1}^{\infty} i^{(l+1)}(2l+1) \;\; d_l \psi_l(m\rho) P_l(\mu),$$

$$E_\theta = \frac{\cos\varphi}{m\rho}\sum_{l=1}^{\infty} i^l \frac{2l+1}{l(l+1)} \times$$
$$\times \left[c_l \psi_l(m\rho) \frac{P_l(\mu)}{\sin\theta} - i d_l \psi_l'(m\rho) \frac{\mathrm{d}}{\mathrm{d}\theta} P_l(\mu) \right], \quad (6)$$

$$E_\varphi = -\frac{\sin\varphi}{m\rho}\sum_{l=1}^{\infty} i^l \frac{2l+1}{l(l+1)} \times$$
$$\times \left[c_l \psi_l(m\rho) \frac{\partial}{\partial\theta} P_l(\mu) - i d_l \psi_l'(m\rho) \frac{P_l(\mu)}{\sin\theta} \right].$$

Here E_r, E_θ, E_φ are the components of the internal electric field strength vector; $\mu = \cos\theta$; ψ_l are the Rikatti-Bessel functions of the first kind; a_l, b_l, c_l, and d_l are the expansion coefficients of the solution into a series over vector spherical harmonics (Mie coefficients). Direct numerical integration in Eq. (5) according to this representation is, in principle, possible,[19] however, it requires significant time for calculations (especially at large ρ values), to achieve high accuracy of calculations of the Rikatti-Bessel functions in Eq. (6), and potentially entails the loss of accuracy of calculations at multiple numerical integration in Eq. (5). Another way for calculating the factor J_1 was proposed in Ref. 13 based on the transformation of the integral relationship (5) into an infinite series over the vector spherical harmonics:

$$J_1 = -\frac{6nk}{|m|^2\rho^3} \;\; \mathrm{Im}\sum_{l=1}^{\infty} \left\{ \frac{l(l+2)}{m}(c_{l+1}c_l^* R_l + d_{l+1}d_l^* R_{l+1}) - \right.$$
$$\left. - \left[\frac{l(l+2)}{l+1}(c_{l+1}c_l^* + d_{l+1}d_l^*) + \frac{2l+1}{l(l+1)} d_l c_l^* \right] S_l \right\}, \quad (7)$$

where the functions R_l and S_l have the form

$$R_l = \int_0^x |\psi_l(m\rho)|^2 \mathrm{d}\rho = \frac{\mathrm{Im}\left[m\psi_{l+1}(m\rho)\psi_l^*(m\rho) \right]}{\mathrm{Im}(m^2)};$$

$$S_l = \int_0^x \rho\psi_l^*(m\rho)\psi_l'(m\rho)\mathrm{d}\rho =$$

$$= -\frac{i}{2\mathrm{Im}(m^2)} \left\{ \rho \left[m\,|\,\psi_l(m\rho)\,|^2 + m^*\,|\,\psi_{l+1}(m\rho)\,|^2 \right] - \right.$$

$$\left. - \left[m + 2(l+1)\frac{\mathrm{Re}(m^2)}{m} \right] R_l + (2l+1) m^* R_{l+1} \right\}.$$

Representation of J_1 in terms of the Mie coefficients (7) allows one to effectively calculate this characteristic with required accuracy analogously to other MOC (efficiency factor Q_{abs} and the light pressure factor Q_{pr}).[1,2]

As was mentioned above, the formulas (5) and (7) correspond to the determination of the spectral asymmetry factor. In principle it is possible to determine the total (or integral) factor $\bar{J}_1$ taking into account spectral composition of the incident radiation. For example, the following way for such a determination was proposed in Ref. 13:

$$\bar{J}_1 = \frac{1}{\sigma T_r^4}\int_0^\infty J_1(\rho, m) I_\lambda(T_r)\,d\lambda, \qquad (8)$$

where $I_\lambda(T_r)$ is the Plank function for radiation of the absolutely black body; σ is the Stephan–Boltzmann constant; T_r is the effective temperature of radiation. One can represent $\bar{J}_1$ as a function of $\bar{\rho}$ and m by introducing the term of the diffraction parameter for thermal radiation $\bar{\rho} = 2\pi R_0 T_r/(\lambda T)_{max}$, where the value $(\lambda T)_{max}$ is determined by the Wien displacement law. The detailed data on the dependence of the refractive index m on the wavelength λ should be available for specific calculations of J_1, what is not always possible.

In calculating the factor J_1 numerically, its representation (7) was realized in the form of the algorithm in the software package Mathematica 3.0, as a modification of the known algorithm BHMIE[20] was used, which was initially intended for calculation of the absorption efficiency factor Q_{abs}. The absolute accuracy was set in calculations (six decimal digits for any values of the parameters ρ and m). It made it possible to calculate the factor J_1 on a PC with a Pentium II processor up to the values $\rho \le 50$ within an acceptable time of computations.

3. Discussion of the results

3.1. Factor J_1 as a function of the diffraction parameter ρ

Analysis of the behavior of the complex of MOC of the principal types of atmospheric aerosol makes it possible to select three main groups of particles depending on the values of the index m: weakly, moderately, and strongly absorbing particles.[1,2] According to this classification, one can form the groups of model particles with preset values of the real and imaginary parts of the complex refractive index (naturally, without violation of the Kramers–Kronig relationships[20]). Calculations of J_1 were performed for the model of the optically homogeneous spherical particles for monochromatic nonpolarized radiation in quite a wide range of variation of the parameter ρ. The calculated results for some selected values n and the set of the values k are shown in Fig. 1.

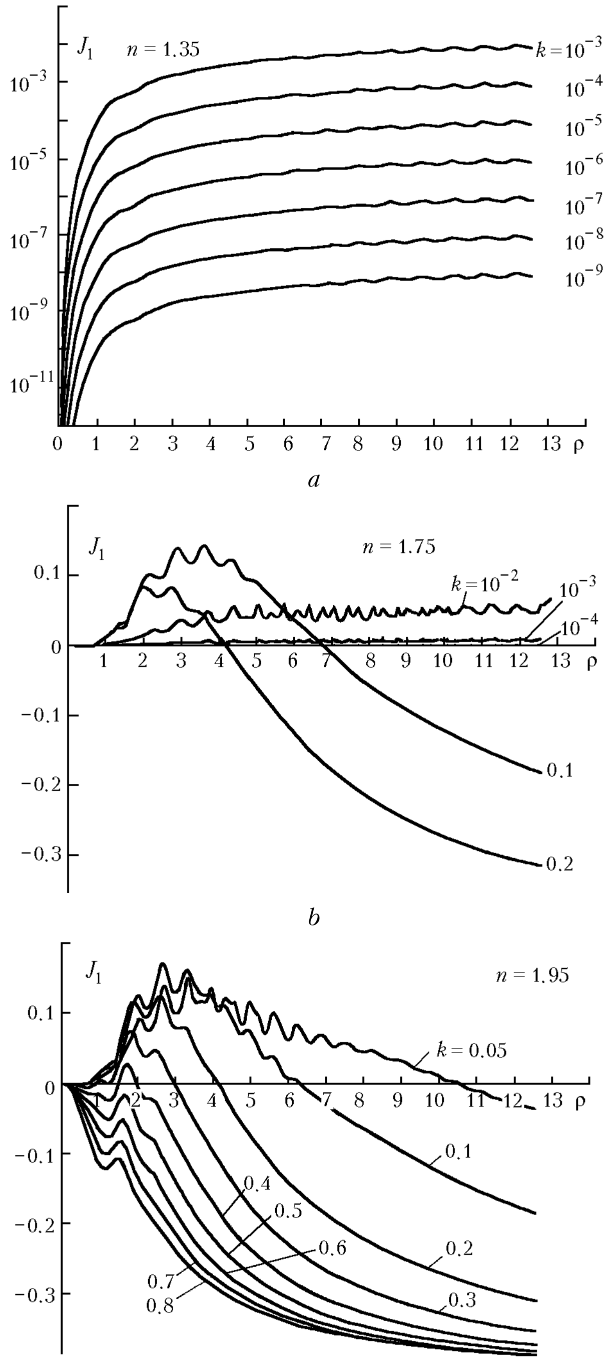

Fig. 1. Calculation of the radiation absorption efficiency factor J_1 for three groups of model particles: weakly absorbing (*a*), moderately absorbing (*b*), and strongly absorbing (*c*) particles.

Weakly absorbing particles (Fig. 1*a*) demonstrate similarity of the curves of J_1, increasing the absorption index k by one order of magnitude (at a fixed value of the refractive index n) increases the factor J_1 by approximately the same magnitude. The periodic resonance structure is observed at $\rho \ge 10$ (the so-called interference structure[20]). The values of the factor J_1 of these particles are only positive (that corresponds to negative photophoresis), however, its absolute values (and, hence, the values of photophoretic force and velocity) are negligible.

Strongly absorbing particles (Fig. 1*c*) demonstrate a different dependence of the factor J_1 on ρ. Calculations show that there is the initial range of positive values of J_1 for the particles of $n \approx 1.5–3.0$ and $k = 0.01–0.4$, the length of which is quite large (up to $\rho \approx 10$) and depends on the complex combination of the values n and k. Absolute values of the factor J_1 reach here the values ≈ 0.2. This range is then followed by the range of negative J_1 values (also with great absolute value) that corresponds to positive photophoresis of particles. It is the evidence of the fact that at a fixed wavelength λ the particles of the same substance, but different size, can move both against and along the direction of radiation propagation. For some characteristic types of atmospheric aerosol (for example, carbonaceous particles) this means that, in principle, the photophoretic levitation of particles of a certain size range is possible at different heights in the stratosphere in the field of solar radiation.[18]

Moderately absorbing particles (Fig. 1*b*) demonstrate an intermediate variant of the dependence on ρ, where at different k values one can observe the peculiarities characteristic of both weakly (small values k) and strongly absorbing particles (quite great k). Evidently, the only type of the dependence is observed: the initial range of positive values J_1, the length of which can reach tens of ρ units is certainly followed by the range of negative values.

Let us note that the used classification of particles according to their absorption properties is quite conventional. In particular, absorption of radiation by many of atmospheric aerosols is radically different in the visible and infrared regions. Some particles pass from the category of weakly absorbing to the category of strongly absorbing (for example, water droplets and ice particles at λ = 0.5 and 10.6 μm).

3.2. Asymptotic of J_1 and comparison with numerical calculations by the Mie theory

It is known[20] that, because of the peculiarities of the solution of the Mie problem (representation of the solutions for internal and scattered fields in the form of infinite series), it is impossible to derive analytical formulas for MOC for the whole range of the parameters ρ and m. However, it is possible in two limiting cases, for small (Rayleigh) particles ($\rho \ll 1$) and in the extreme case of geometric optics ($\rho \gg 1$). The necessity of such asymptotic formulas is obvious: the error in calculations by Mie theory at very small ρ is great because of very small absolute values of J_1, and significant time is necessary for calculation at very great ρ for summing the great number of terms of the series in Eq. (7). Besides, asymptotic provides for the possibility of general consideration of analytical results for the force and velocity of photophoresis in the extreme regimes.

Two extreme results for J_1 in the case of $\rho \ll 1$ are obtained in Ref. 9 by means of asymptotic expansion of the Mie solution. The first estimates the asymmetry factor for weakly absorbing particles ($\rho \ll 1$ and $k\rho \ll 1$)

$$J_1 = \frac{12n^2k^2\rho^2}{5|2+m^2|^2|3+2m^2|^2}(5-|3+2m^2|^2), \qquad (9)$$

and the second describes the behavior of strongly absorbing particles ($\rho \ll 1$, but $k\rho \gg 1$)

$$J_1 = -\,9n^2/(2|m|^4). \qquad (10)$$

Then authors of Ref. 11 also obtained the result (9) in a slightly different way. Comparison of the numerical calculation of the factor J_1 by full Mie theory and asymptotic results (9) for the same groups of model particles as in Fig. 1 is shown in Fig. 2. The comparison lead to unexpected conclusions. First, formula (9) does not describe in principle, the behavior of weakly absorbing particles, for which it was obtained (formula (9) provides for only negative values J_1 that does not agree with full calculations; quantitative differences are unacceptably great even at very small $\rho \le 0.001$). Second, it well describes the dependences of J_1 on ρ for moderately absorbing particles and satisfactorily for strongly absorbing ones that also was not supposed when deriving this formula. Third, the asymptotic formula (10) for strongly absorbing particles absolutely disagrees with the full calculation by Mie theory (not shown in Fig. 2). In our opinion, the reason is an obvious discrepancy of the conditions $\rho \ll 1$ and $k\rho \gg 1$ and, hence, mathematically artificial character of this extreme case. Let us note that doubts in the efficiency of formula (9) and the necessity of calculations by full Mie theory in this case were also discussed in Ref. 11.

The extreme case of geometric optics ($\rho \gg 1$) was studied in Ref. 10. The results obtained are based on the Fresnel formulas for amplitudes of reflected and refracted waves for plane surface neglecting the interference effects. It was obtained[10] for large weakly absorbing particles under conditions that $\rho \gg 1$, but $k\rho \ll 1$

$$J_1 = 2nk\rho\left[\frac{3(n-1)}{8n^2} - \frac{2}{5}nk\rho\right]. \qquad (11)$$

The following result was obtained[10] for large strongly absorbing particles:

$$J_1 = \begin{cases} -0.5; & |n-1| \ll 1,\ k \ll 1,\ k\rho \gg 1, \\ -\dfrac{9}{4k^2}; & k \gg n,\ k \gg 1,\ k\rho \gg 1. \end{cases} \qquad (12)$$

Correctness of the Fresnel formulas in this case was discussed in Refs. 12 and 16 and was subject to well founded criticism. Significant error characteristic of formulas (11) and (12) was revealed both in comparing with the results calculated by full Mie theory[16] and with the experiment.[12] The results of comparison of calculations of J_1 by full Mie theory and by formula (11) are presented in the Table for the characteristic groups of model particles at ρ = 12.

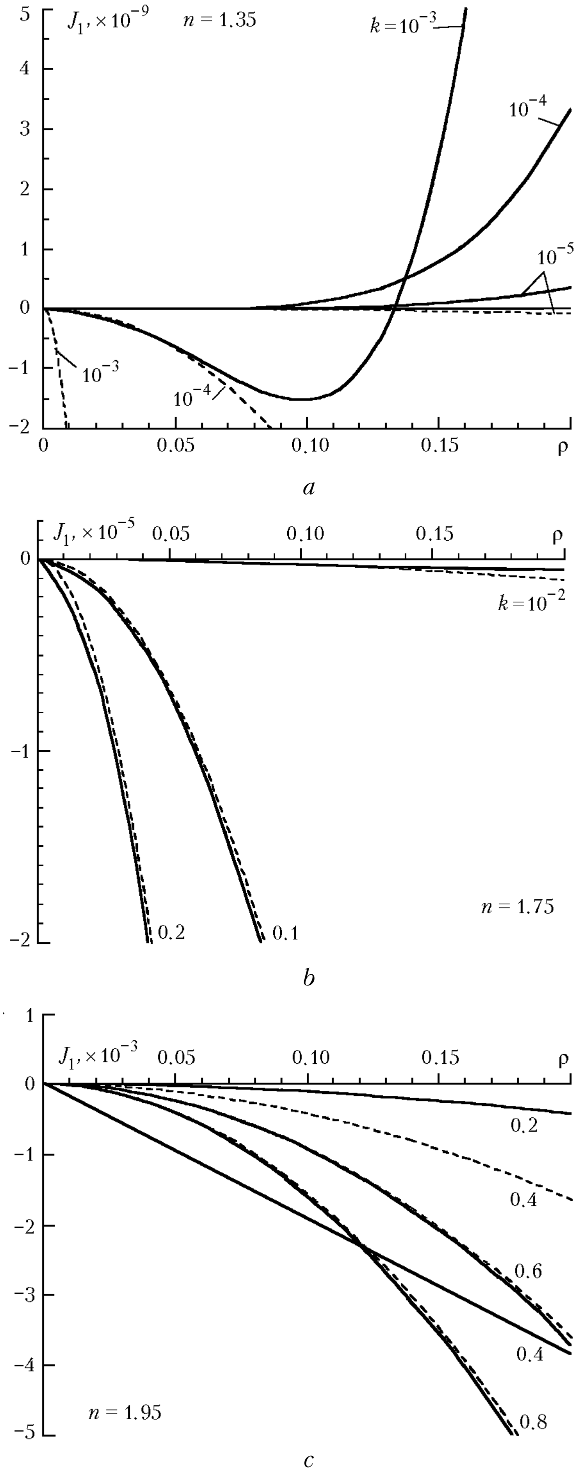

Fig. 2. Comparison of calculations of the factor J_1 by full Mie theory (7) (solid lines) and asymptotic (9) (dotted lines) for model particles.

Although such a comparison demonstrates qualitative agreement of the results, quantitative differences again are too great. As in the case with formula (10) for Rayleigh particles, formulas (11) and (12) describe not real, from the standpoint of the optical properties of particles, events, which are poorly suitable for use in practice. The factor J_1 for strongly absorbing particles at ρ ≥ 10 reaches the extreme negative values close to −0.5 (Fig. 1*c*) as it was predicted by formula (12). Thus, one can ascertain that to date the problem of asymptotic analytical results for the factor J_1 is open and requires additional consideration.

Table. Comparison of numerical calculation of the factor J_1 by full Mie theory (7) with the asymptotics (11) for characteristic groups of model particles at ρ = 12

$m = n + ik$	Mie theory (7)	Asymptotics (11)
$n = 1.35$		
$k = 10^{-9}$	8.69(−9)	2.33(−9)
10^{-8}	8.74(−8)	2.33(−8)
10^{-7}	8.78(−7)	2.33(−7)
10^{-6}	8.33(−6)	2.33(−6)
10^{-5}	9.01(−5)	2.33(−5)
10^{-4}	8.54(−4)	2.31(−4)
10^{-3}	8.10(−3)	2.12(−3)
$n = 1.75$		
$k = 10^{-3}$	7.43(−3)	3.50(−3)
10^{-2}	5.41(−2)	3.29(−3)
$n = 1.95$		
$k = 5{\cdot}10^{-2}$	−3.02(−2)	−8.76(−1)

3.3. Dependence of J_1 on the wavelength of incident radiation λ

In discussing the effect of the spectral composition of incident radiation on photophoresis of atmospheric aerosols, analysis of the dependences of the factor J_1 of particles of different size on the wavelength λ is of doubtless interest. In particular, such calculations were performed for pure water droplets and soot particles; the data on their optical characteristics at different λ were taken from Refs. 21 and 22.

The dependences of the factor J_1 on λ are shown in Fig. 3 for pure water droplets of the size 0.01, 0.1, and 1.0 μm in the wavelength range 0.2 to 1.0 μm. It is seen that the curves are similar. They are characterized by a sharp decrease of J_1 in the UV range, plateau in the visible and near IR range, and the tendency toward an increase at λ ≥ 0.9 μm. The values J_1 are positive that corresponds to negative photophoresis. Let us note that the absorption efficiency factor Q_{abs} of water aerosol of the droplet size 0.01 to 100 μm demonstrates similar dependence on λ (Ref. 1). It follows that the great contribution of UV and IR spectral ranges of solar radiation to the total asymmetry factor J_1 is possible for water droplets. Perhaps, this fact forces reconsideration of the estimate of the role of negative photophoresis of water droplets obtained from calculations of J_1 at λ = 0.55 μm (maximum of intensity in the solar radiation spectrum). The dependences of J_1 on λ for soot particles of the same size and the same wavelength range as for water droplets are shown in Fig. 4.

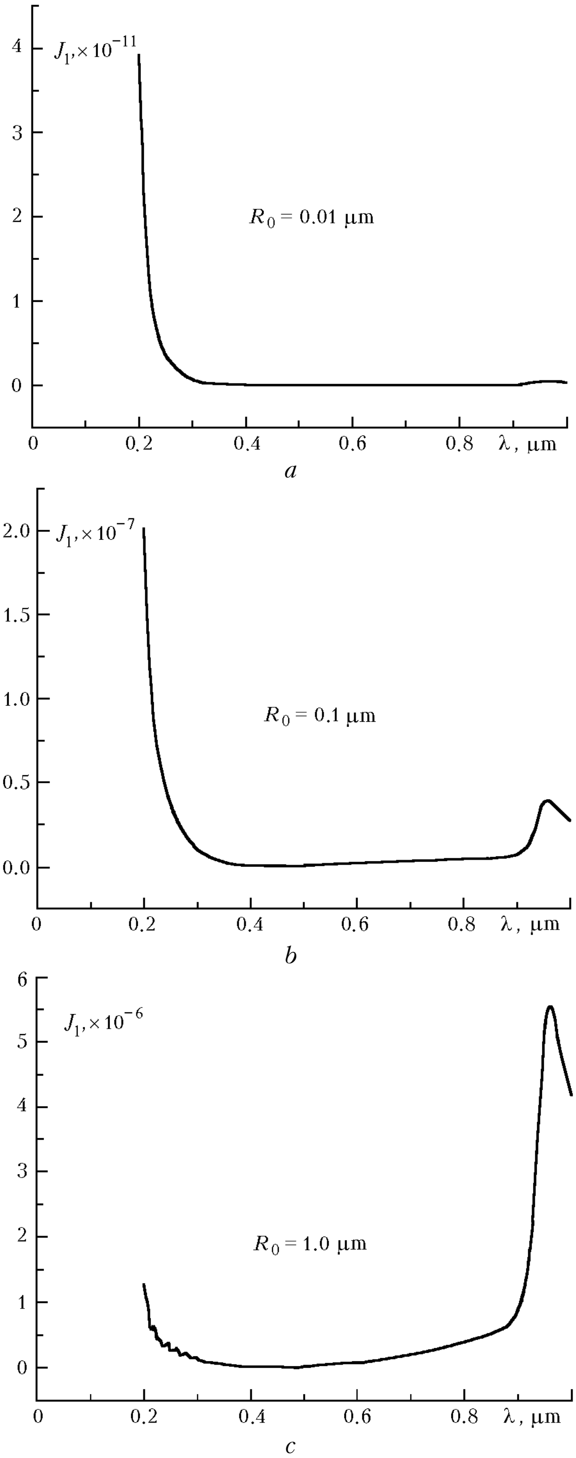

Fig. 3. The factor J_1 as a function of wavelength of the incident radiation for pure water droplets of different size.

It is seen that the dependences are distinctly different: practically monotonic increase of J_1 as λ increases, the values of J_1 are only negative. It is of principle importance that the absolute value of the factor J_1 of strongly absorbing carbonaceous aerosol decreases as the wavelength of incident radiation increases, hence, the force of positive photophoresis of such particles decreases.

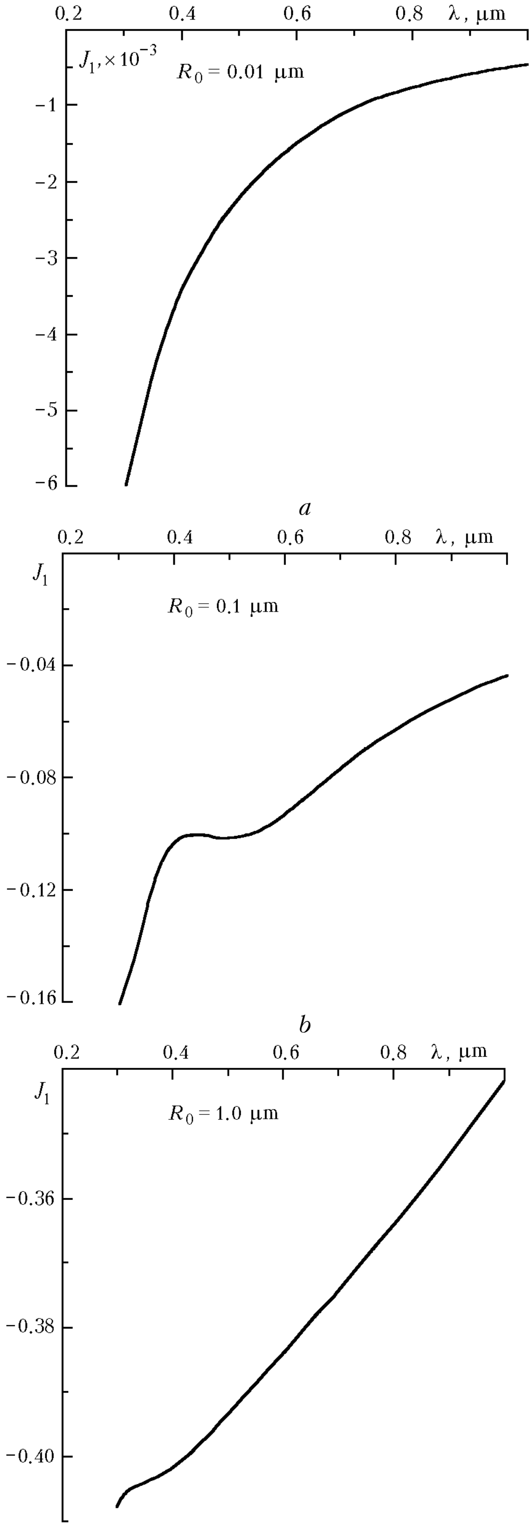

Fig. 4. The factor J_1 as a function of wavelength of the incident radiation for soot particles of different size.

Conclusion

This paper presents the results of systematic analysis of the spectral asymmetry factor J_1 for the model of optically homogeneous spherical particles. It is planned to continue these investigations using the model of two-layer particles, that would enable extending the results to description of the photophoretic characteristics of a more wide class of atmospheric aerosols. Advantage and efficiency of generalizing the asymmetry factor for non-spherical particles (at least, for cylindrical particles and ellipsoids of revolution) are obvious. Authors do not know such results. The necessity of taking into account the spectral composition of incident radiation is also obvious, because quantitative estimates of the role of photophoresis of particles of different types of atmospheric aerosol on the basis of only spectral asymmetry factors can lead to incorrect physical conclusions. It is supposed to use the data obtained for the development of a new model of the vertical transfer of stratospheric aerosol, where photophoresis of aerosol particles should be taken into account in addition to sedimentation–diffusion mechanism.

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References

1. L.B. Letfulova, A.V. Starinov, and S.A. Beresnev, Atmos. Oceanic Opt. **14**, No. 1, 62–68 (2001).
2. S.A. Beresnev, L.B. Kochneva, and P.E. Suetin, Atmos. Oceanic Opt. **15**, Nos. 5–6, 472–479 (2002).
3. F. Ehrenhaft, Physik. Zeitschr. **17**, 352–358 (1917).
4. A. Rubinowicz, Annalen der Physik. **62**, No. 16, 691–737 (1920).
5. O. Preining, in: *Aerosol Science*, ed. by C.N. Davis (Academic Press, New York, 1966), pp. 111–135.
6. H. Rohatschek, J. Aerosol Sci. **16**, No. 1, 29–42 (1985).
7. P.W. Dusel, M. Kerker, and D.D. Cooke, J. Opt. Soc. Am. **69**, No. 1, 55–59 (1979).
8. W.M. Greene, R.E. Spjut, E. Bar-Ziv, A.F. Sarofim, and J.P. Long-well, J. Opt. Soc. Am. B. **2**, No. 6, 998–1004 (1985).
9. Yu.I. Yalamov, V.B. Kutukov, and E.R. Shchukin, Inzh. Fiz. Zh. **30**, No. 6, 996–1002 (1976).
10. Yu.I. Yalamov, V.B. Kutukov, and E.R. Shchukin, J. Colloid and Interface Sci. **57**, No. 3, 564–571 (1976).
11. S. Arnold, A.B. Pluchino, and K.M. Leung, Phys. Rev. A. **29**, No. 2, 654–660 (1984).
12. S. Arnold and M. Lewittes, J. Appl. Phys. **53**, No. 7, 5314–5319 (1982).
13. D.W. Mackowski, Int. J. Heat and Mass Transfer **32**, No. 5, 843–854 (1989).
14. H. Rohatschek, J. Aerosol Sci. **26**, No. 5, 717–734 (1995).
15. V. Chernyak and S. Beresnev, J. Aerosol Sci. **24**, No. 7, 857–866 (1993).
16. M. Kerker and D.D. Cooke, J. Opt. Soc. Am. **72**, No. 9, 1267–1272 (1982).
17. S. Tehranian, F. Giovane, J. Blum, Y.-L. Xu, and B.A.S. Gustafson, Int. J. Heat and Mass Transfer **44**, 1649–1657 (2001).
18. S.A. Beresnev, F.D. Kovalev, L.B. Kochneva, V.A. Runkov, P.E. Suetin, and A.A. Cheremisin, Atmos. Oceanic Opt. **16**, No. 1, 44–48 (2003).
19. A.B. Pluchino, Appl. Opt. **22**, No. 1, 103–106 (1983).
20. G.F. Bohren and D.R. Huffman, *Absorption and Scattering of Light by Small Particles* (Willey, New York, 1983).
21. G.M. Hale and M.R. Querry, Appl. Opt. **12**, No. 3, 556–558 (973)
22. V.M. Zolotarev, V.N. Morozov, and E.V. Smirnova, *Optical Constants of Natural and Technical Media. Handbook* (Khimiya, Leningrad, 1984), 216 pp.