Photophoresis of atmospheric aerosols in the Earth's thermal radiation field

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A new type of photophoretic motion of atmospheric aerosols is considered, namely, the photophoresis of particles in the field of outgoing thermal radiation of the Earth (thermal photophoresis). It is shown that the soot particles (as well as the others light and slightly heat conducting particles) in the stratosphere demonstrate high spectral response to photophoretic effects. In particular, the levitation and ascend against the force of gravity are predicted for the soot particles with the size of 1 to 2 μm in the field of thermal radiation. At the same time, the thermal photophoresis, apparently, is not a significant mechanism in maintaining the stability and long-term existence of the known aerosol layers in the stratosphere and mesosphere.

Introduction

In many publications devoted to the investigations of photophoresis, a question on the significance of this phenomenon for atmospheric aerosols is discussed. Traditionally the following problem is analyzed. The plane wave of solar radiation with the intensity being equal to the solar constant is incident on particles located at high altitudes. The intensity maximum occurs within the wavelength interval $\lambda = 0.50–0.55$ μm, that, in the first approximation, makes it possible to consider the radiation monochromatic. Due to the absorption of radiation, the particles are heated inhomogeneously, and in the rarefied gas the force of radiometric nature, i.e., photophoretic force, acts on these particles. If this force is counterbalanced by the force of viscous resistance from gas, then the particle moves uniformly along a straight line at the so-called photophoretic speed. Later on we shall name this phenomenon the "solar" photophoresis. The fundamental problem on the effect of the Brownian rotation is solved by evaluating characteristic times of the temperature relaxation in the volume of particles.[1] They turn out to be much shorter than the characteristic time of turn to a marked angle for micron-sized particles.

In earlier published papers[2–5] the interest in the "solar" photophoresis of aerosols was determined by the investigations of delayed fallout of particles at the nuclear tests in the atmosphere[5] and at the first assessments of residence times of highly dispersed aerosols in the stratosphere.[2,3] On the whole the results of investigations did not show a considerable effect of photophoretic effects on the stratospheric-tropospheric transfer of aerosols. A specific role in support of this point of view can be found in recent studies,[6] where it was stated, based on the calculations by the Lorentz–Mie theory and the molecular-kinetic representations, that in the free-molecular regime the forces of negative and positive "solar" photophoresis cannot exceed several percent of the force of gravity affecting the particles. Recent analogous results[7] correct the errors and inaccuracies in the calculations[6] and increase this ratio for similar conditions up to 10–20%, however, the role of "solar" photophoresis in the vertical transport of particles in the stratosphere is estimated as insignificant.

Incidentally, the assessments of the feasibility of radiometric photophoresis in the aerosol transport at high altitudes were not completed. The complexity of the photophoresis phenomenon implies the need for analysis of both the electrodynamics and gas kinetic aspects of the problem taking into account morphology of the particles considered and variations of their thermophysical properties. In particular, in Ref. 8 new assessments are made of the possibilities of "solar" photophoresis in the vertical transport of stratospheric particles. They show that the negative "solar" photophoresis can cause levitation and even the ascend of certain types of soot particles in the stratosphere.

Numerous observations point to the presence in the stratosphere on a long-term scale of absorbing soot aerosols of different character and light particles of volcanic aerosol existing at much higher altitudes than the altitude of the aircraft flights and the heights of gaseous-dust plumes of volcanic origin (e.g., Ref. 9). The traditional question is on the conditions and reasons of the formation of sporadic or sufficiently stable aerosol layer at different altitudes in the stratosphere and mesosphere observed, in particular, at tangential sounding in the ultraviolet range from space.[10]

In recent years the role of polar stratospheric and mesospheric clouds is being studied intensely in the processes of ozone depletion.[11] All these problems explain the search for different power systems

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appearing under specific conditions of high altitudes and providing an efficient vertical transport of particles. Traditional models of the stratospheric transfer based on the idea of sedimentation-diffusion equilibrium (see, for example, Ref. 12), face problems in describing the observed aerosol altitude stratification. New models of vertical transfer are often based on the hypotheses, which are not confirmed, on the existence of forces of exotic physical nature.[9,13]

Thus the questions formulated early in the investigations of this phenomenon remain open on whether or not the radiometric photophoresis affects considerably the dynamics of stratospheric and mesospheric aerosol. In what quantities and characteristics can this effect be discovered? The discussion and analysis of these questions can also take into account the spectral dependence of photophoretic effects, which cannot be reduced only to their solar manifestations. In this paper a new type of photophoretic motion of atmospheric aerosols is proposed: photophoresis of particles in the field of outgoing thermal radiation of the Earth (further "thermal" photophoresis). The goals of this work were formulation of the problem, description of the methodology of calculations of basic characteristics, and analysis of the results obtained.

1. Statement of the problem on aerosol photophoresis in the field of thermal Earth's radiation

Now we consider a spherical particle of radius R_0 suspended at the height z in the field of electromagnetic radiation (Fig. 1). Such a particle is characterized by the density ρ_p, the coefficient of thermal conductivity λ_p and the complex refractive index $m = n + ik$, depending on the wavelength λ of incident radiation.

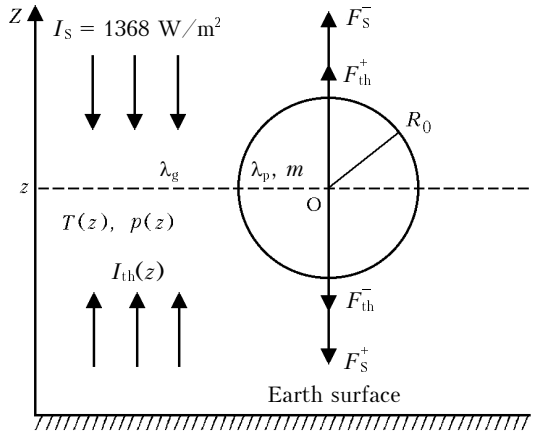

Fig. 1. Formulation of the problem on the photophoresis of atmospheric aerosols.

It is believed that at the height z the air pressure equals $p(z)$, its temperature is $T(z)$ and the corresponding gas thermal conductivity coefficient is $\lambda_g(T)$. In addition to solar radiation of the intensity $I_S = 1368$ W/m^2 with the effective wavelength $\lambda_S = 0.50–0.55$ μm, the particle is in the field of a plane wave of outgoing thermal Earth's radiation with the effective wavelength $\lambda_{th} = 10–11$ μm. In this case the thermal radiation intensity I_{th} is the function of height z due to the absorption of thermal radiation by gaseous components of the atmosphere and aerosol. Photophoretic forces of radiometric character induced by the incident solar radiation and the outgoing thermal radiation affect the particle:

$$F_{ph} = F_S^+ + F_S^- + F_{th}^+ + F_{th}^-, \quad (1)$$

where the sign "+" refers to the forces of positive photophoresis (motion of particles along the direction of radiation propagation) and the sign "–" refers to the forces of negative photophoresis (motion of particles opposite to the direction of radiation propagation). It is known[14] that the direction of motion of particles is determined only by their optical properties, and the size and is independent of gas-kinetic regime and accommodation characteristics of the surface.

It should be noted that the statement of the problem on the "thermal" photophoresis implies the existence of a plane wave of incident longwave radiation (then it is possible to use the results of Mie theory to calculate the asymmetry factor J_1 (Refs. 15 and 16)) and the knowledge of the dependence of the intensity I_{th} on the height of particle in the atmosphere z. Otherwise the statement of the problem is similar to the case of photophoresis of atmospheric aerosols in the field of solar radiation.[8] The change of the spectral range of incident radiation is not a fundamental factor, for example, we know the experiments[17] on photophoresis of microparticles in the field of longwave electromagnetic radiation (wavelengths 10.63 μm and 9.58 μm).

2. Method of solution and the quantities determined

The calculation of the force and velocity of aerosol photophoresis is based on the molecular-kinetic theory of this phenomenon.[14] This theory is based on the solution of gas kinetic model equation with the corresponding boundary conditions for the function of molecular distribution on the particle surface and covers the entire range of variation of the Knudsen number Kn ($Kn = l/R_0$) where l is the mean free path of gas molecules, R_0 is the particle radius) at an arbitrary ratio between the coefficients of thermal conductivity of the particulate matter and gas, $\Lambda = \lambda_p/\lambda_g$, taking into account the optical and accommodation properties of the particle–gas system.

The expression for the photophoretic force in the entire range of Kn numbers is of the form:

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

where I is the intensity of the radiation incident on the particle, M is the molar mass of the surrounding gas, T_0 is the typical temperature, R is the universal gas constant, $F(\mathrm{Kn}, \Lambda)$ is the function of the Kn number, thermal physical parameter Λ, coefficients of accommodation of momentum and energy of gas molecules on the particle surface. The expression for the photophoretic speed of a spherical particle is written in the form:

$$U_{\mathrm{ph}} = -\frac{\pi}{2(8+\pi)} \frac{IJ_1}{p_0} \Phi(\mathrm{Kn}, \Lambda), \qquad (3)$$

where p_0 is the pressure of the surrounding gas, $\Phi(\mathrm{Kn}, \Lambda)$ is the new gas kinetic function similar to $F(\mathrm{Kn}, \Lambda)$. In the general case the functions are calculated using numerical methods and presented in the tables. If we use an assumption about the total accommodation of the momentum and energy of gas molecules on the particle surface (that is acceptable for atmospheric aerosols) then one can use effective analytical approximations of numerical calculations of the functions $F(\mathrm{Kn}, \Lambda)$ and $\Phi(\mathrm{Kn}, \Lambda)$. In particular, in Ref. 14 it is shown that the structure of gas kinetic functions

$$F(\mathrm{Kn},\Lambda)=\Psi_1/(1+\Lambda\Psi_2),\ \Phi(\mathrm{Kn},\Lambda)=\Psi_1/[\Psi_3(1+\Lambda\Psi_2)] \qquad (4)$$

is precise, and the expressions for Ψ_1, Ψ_2, and Ψ_3 should be chosen as follows

$$\Psi_1 = \frac{\mathrm{Kn}}{\mathrm{Kn}+5\pi/18}\left(1+\frac{2\pi^{1/2}\mathrm{Kn}}{5\mathrm{Kn}^2+\pi^{1/2}\mathrm{Kn}+\pi/4}\right),$$

$$\Psi_2 = \left(\frac{1}{2}+\frac{15}{4}\mathrm{Kn}\right)\left(1-\frac{1.21\pi^{1/2}\mathrm{Kn}}{100\mathrm{Kn}^2+\pi/4}\right), \qquad (5)$$

$$\Psi_3 = \frac{\mathrm{Kn}}{\mathrm{Kn}+0.619}\left(1+\frac{0.310\mathrm{Kn}}{\mathrm{Kn}^2+1.152\mathrm{Kn}+0.785}\right).$$

Then it is possible to approximate the numerical calculations of the force and the photophoresis speed within the experimental error no worse than 3% for the entire range of Kn numbers (maximum error is at $\mathrm{Kn} \approx 1$). Note that the formulae (4)–(5) provide correct limiting transitions in case of large and small numbers Kn (in these cases, the known asymptotic expressions for the force and speed of photophoresis are precisely reproduced). In addition to the force and speed of photophoresis, other values characterizing the significance of this phenomenon for atmospheric aerosols are of interest. Among those, there is the ratio of photophoretic force to the force of gravity

$$\alpha = F_{\mathrm{ph}}(z)/F_{mg}, \qquad (6)$$

where $F_{mg} = 4/3\,\pi R_0^3 \rho_{\mathrm{p}} g$; g is the free fall acceleration; and the ratio between the photophoretic speed of the particle and its speed of sedimentation due to gravity

$$\beta = U_{\mathrm{ph}}(z)/U_{mg}(z), \qquad (7)$$

where U_{mg} is calculated by the formula

$$U_{mg} = \frac{\pi^{1/2} R_0 \rho_{\mathrm{p}} g}{(\pi+4) p_0 \left(\frac{M}{2RT_0}\right)^{1/2} \Psi_3}. \qquad (8)$$

When writing Eq. (8) the results are used for isothermal force of the particle resistance in the entire range of Kn numbers.[18] The resultant speed of vertical particle transport U_{sed} is determined as

$$U_{\mathrm{sed}}(z) = U_{mg}(z) + U_{\mathrm{ph}}(z). \qquad (9)$$

Using the expressions (3), (8) and (9) we can estimate the times of particle sedimentation from some height to the boundaries of tropopause with or without the account of the effect of photophoretic forces (the term "time of sedimentation" is not identical to the term "time of residence" of particles at some height).

2.1. Calculation of intensities of the outgoing thermal radiation of the Earth

Owing to the absorption of the outgoing longwave Earth's thermal radiation by clouds, water vapor, optically active gases and aerosol, the major fraction of thermal radiation emitted by the Earth's surface is absorbed in the troposphere. As a result, the vertical profiles of upwelling fluxes of longwave radiation are formed, which are characteristic of different latitudes and seasons. The experimental data on the radiation fluxes, obtained using actinometric radiosondes, indicate that the vertical profiles of mean values of the intensity of the outgoing thermal radiation at different observation points are similar to each other. In the troposphere the mean value of thermal radiation intensities decreases with height and in the stratosphere the mean intensity value increases slowly at altitudes higher than 20 km. A peculiarity of the thermal radiation flux is its low daily variations as opposed to the solar radiation flux with its maximum at noon and zero at night. Densities of integral fluxes of the outgoing thermal radiation I_{th} at different latitudes are daily hundreds of $\mathrm{W/m^2}$, at the same time, about 50% of the flux can be observed in the atmospheric transmission window at 8 to 12 μm. Setting the temperature of the Earth's surface, the concentration of greenhouse gases and the temperature profile in the atmosphere it is possible to calculate the spectral density of the brightness of thermal radiation in the atmosphere at different heights $W_\nu^{\mathrm{up}}(z)$ when solving the transport equation for infrared radiation[19]:

$$W_\nu^{\mathrm{up}}(z) = \varepsilon_\nu B_\nu(T_0) \exp\left(-\int_0^z K_\nu^{\mathrm{abs}} \mathrm{d}h\right) +$$

$$+ \int_0^z K_\nu^{\mathrm{abs}} B_\nu \exp\left(-\int_h^z K_\nu^{\mathrm{abs}} \mathrm{d}h'\right) \mathrm{d}h, \qquad (10)$$

where K_ν^{abs} is the coefficient of absorption by atmospheric components at the frequency ν; B_ν is the spectral density of the black body brightness; ε_ν is the emissivity of the Earth's surface; T_0 is the temperature of the Earth's surface; z is the height at which the upwelling flux is calculated. The absorption coefficient K_ν^{abs} incorporates the following components: (1) the absorption coefficient by gas constituents, which is calculated in the approximation of local thermodynamic equilibrium by the method of summation over spectral lines (line-by-line) with the use of parameters of known spectral data base HITRAN-96 (Ref. 20); (2) the extinction coefficient by the atmospheric aerosol components calculated using model representations (for example, Ref. 21); (3) the coefficient of continuum absorption by water vapor calculated using a parametrization.[22]

For cases of the radiation transfer at an angle to the vertical the substitution $dh \to \sec\theta\, dh$ is made in Eq. (10), where θ is the angle between the direction of the radiation propagation and the vertical. In order for the transfer equation to be written in this case, it is necessary to perform the integration in Eq. (10) over the angle θ from 0 to π/2 and over the azimuth angle from 0 to 2π. Then the transfer equation for the monochromatic longwave radiation retains the same form as Eq. (10) at θ = 0, however, secθ should be equal to 1.66 (Ref. 23). This coefficient in a modified equation (10) accounts for the effect integration over θ angle. The total intensity of the outgoing thermal radiation I_{th} is calculated by integrating the solution of the modified equation (10) over the entire spectral range:

$$I_{th}(z) = \int_0^\infty W_\nu^{up}(z)d\nu. \qquad (11)$$

The calculations of $I_{th}(z)$ were made using a previously developed FIRE-ARMS software package.[24]

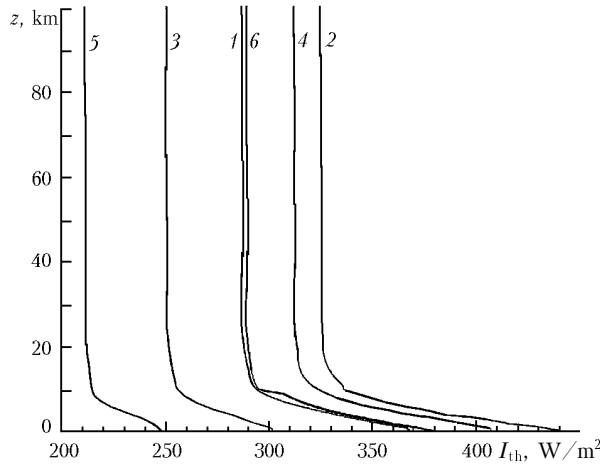

Fig. 2. Integrated over wavelength intensity of the outgoing Earth's thermal radiation as a function of height: standard atmosphere of USA (*1*); model of tropical atmosphere (*2*); mid-latitudes (winter) (*3*); mid-latitudes (summer) (*4*); subarctic (winter) (*5*); subarctic (summer) (*6*).

Fig. 2 shows the calculations of values of $I_{th}(z)$ in the altitude range from 0 to 100 km using latitude-seasonal models of the atmosphere.[25]

The magnitude of the emittance of the Earth's surface ε_ν is assumed to be equal to 0.95. Despite the fact that calculations were made traditionally for a cloudless atmosphere, the averaged effect of cloudiness is considered, although indirectly. The matter is that the calculations were made with the use of statistically mean model profiles of the temperature and humidity, which are, in turn, the average of numerous field measurements using sondes and meteorological rockets. Consequently, the intensities $I_{th}(z)$ in Fig. 2 show the influence of the characteristic cloudiness for the latitudes and seasons indicated.

2.2. Calculation of the asymmetry factor of the radiation absorption J_1

The spectral asymmetry factor of the radiation absorption J_1 is the basic result of solution of the electrodynamic problem of photophoresis.[8,15,16] This factor is determined by the internal field in a particle and depends on the magnitude of the diffraction parameter ρ and the complex refractive index of the particle matter m

$$J_1 = 3nk\rho \int_0^\pi \sin\theta P_1(\cos\theta)d\theta \int_0^1 x^3 B(x,\theta,\phi = \frac{\pi}{4})dx, \qquad (12)$$

where $B(x, \theta, \phi)$ is the dimensionless function of sources of electromagnetic energy inside the particle, $\rho = 2\pi R_0/\lambda$ is the diffraction parameter, $x = r/R_0$ is the dimensionless radial coordinate, $P_1(\cos\theta)$ denotes the Legendre polynomials of the first kind.

The quantity J_1 is normalized and varies within the limits $-0.5 < J_1 < 0.5$. Negative values of J_1 correspond to the heating of the front particle side and fit the positive photophoresis, positive values correspond to heating mostly of the rear particle side and the negative photophoresis. In Refs. 15 and 16 it was noted that in calculations it is preferable to use not the spectral but the integral (or the total) asymmetry factor $\bar{J}_1$, determined in Ref. 26 as

$$\bar{J}_1 = \frac{1}{\sigma T_R^4} \int_0^\infty J_1(\rho,\ m)B_\lambda(T_R)d\lambda, \qquad (13)$$

where $B_\lambda(T_R)$ is the Planck function for the radiation of ideal black body, σ is the Stefan–Boltzmann constant, T_R is the effective radiation temperature. Having determined the diffraction parameter as $\bar{\rho} = 2\pi R_0 T_R/(\lambda T)_{max}$, where the quantity $(\lambda T)_{max}$ is determined according to the Wien displacement law, we can express $\bar{J}_1$ in terms of the function of $\bar{\rho}$ and m. In Ref. 26 it is assumed that such an approach will result in the curve smoothing out for J_1 as well as in the removal of morphologically specified resonances, but this approach will not change the character of the dependence of this quantity on the diffraction

parameter ρ. However, for making numerical calculations of $\overline{J}_1$ according to Eq. (13) it is essential to have the detailed data on the dependence of m on the radiation wavelength λ that it is either impossible or a very cumbersome process. As an alternative, this paper describes the calculation of the effective spectral asymmetry factor J_1 for the wavelengths λ_{th} = 10–11 μm corresponding to the intensity maximum of outgoing Earth's thermal radiation.

Figure 3 shows the distributions of relative intensity of the internal field $B(x, \theta, \phi)$ for the soot particles in the visible and infrared regions. These calculations do not yield single-valued information on the direction and the magnitude of the photophoretic force and the velocity of particle motion.

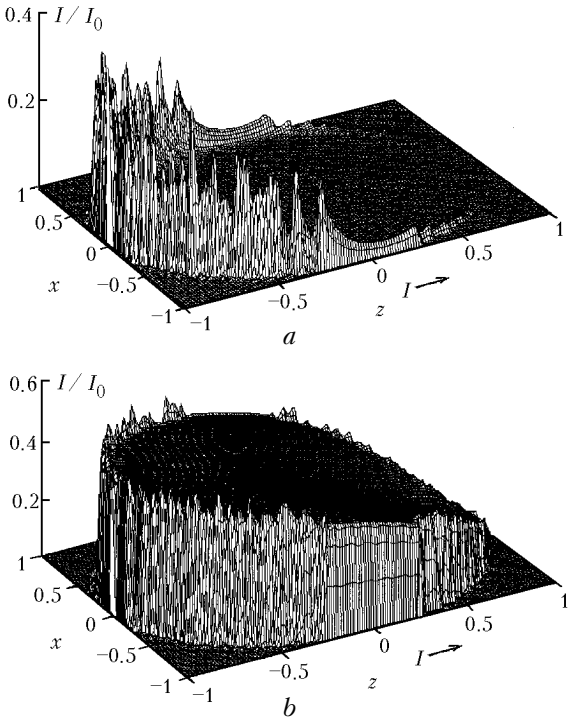

Fig. 3. Dimensionless intensity of internal field for soot particle R_0 = 1.6 μm; λ = 0.5 μm (ρ = 20) (*a*); λ = 10.6 μm (ρ = 0.95) (*b*).

The asymmetry factor is the averaged integral characteristics of inhomogeneous power distribution over the particle volume of the radiation absorption J_1 (Fig. 4). In particular, Fig. 4*b* shows the calculated results on J_1 obtained according to the values of complex refractive index for the soot particles in the range λ = 10.0–10.6 μm.[27,28] It is evident that in the field of thermal radiation such particles can undergo only positive photophoresis (factor J_1 is negative at any values of ρ).

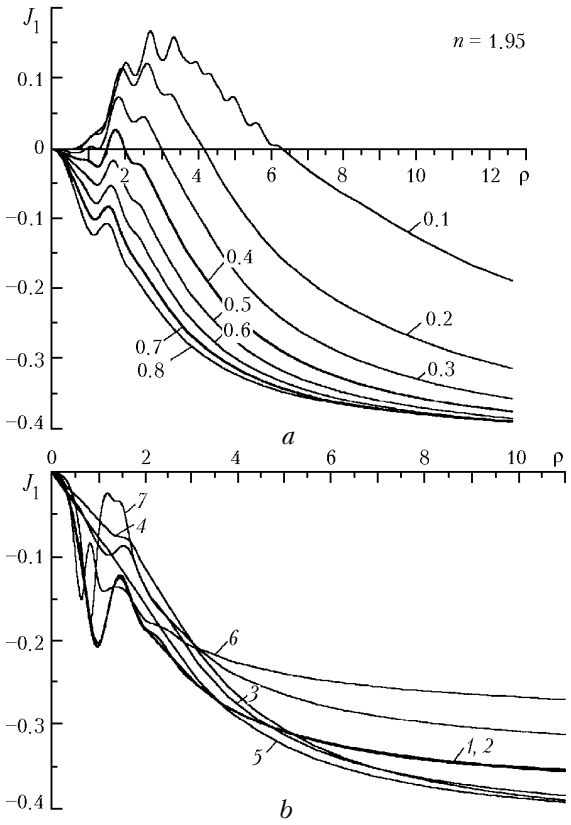

Fig. 4. The absorption asymmetry factor J_1 for soot particles: λ = 0.5 μm, n = 1.95; k = 0.1–0.8 (*a*); λ = = 10.0–10.6 μm (*b*): $m = 2.42 + 1.02i$ (*1*); $m = 2.40 + 1.00i$ (*2*); $m = 1.9 + 0.7i$ (*3*); $m = 1.75 + 0.62i$ (*4*); $m = 1.70 + 0.75i$ (*5*); $m = 4.40 + 1.24i$ (*6*); $m = 3.38 + 0.77i$ (*7*) (Table 1).

Table 1. Optical and thermal physical characteristics of soot particles

Characteristic	Value of parameter
$m = n + ik$	2.42 + 1.02*i* at λ = 10.6 μm; 2.40 + 1.00*i* at λ = 10.0 μm (Ref. 28); 1.9 + 0.7*i*; 1.75 + 0.62*i*; 1.70 + 0.75*i* at λ = 10.0 μm (Ref. 27); 4.40 + 1.24*i* (polycrystalline graphite); 3.38 + 0.77*i* (dispersed graphite) at λ = 10.6 μm (Ref. 27)
ρ_p, kg/m^3	280–570 (Ref. 29) (charcoal); 165 (Ref. 30); 80 (Ref. 4) is the minimum value for the compact units; 2250 (Ref. 29) is the maximum value for the mass samples; 450–850 (Spherocarb – synthetic porecarbon particles of the size more than 0.5 μm of the firm Analabs Inc., Foxboro, USA)
λ_p, W/(m · K)	0.167 (Ref. 29) (charcoal); 0.07–0.12 (Ref. 30); 75–100 (Ref. 29) (reactor graphite) is the maximum value for mass samples
$\Lambda = \lambda_p/\lambda_g$ at $T(z)$= 220 K	2.7–4.6 (based on (Ref. 30)); 5 (optimal value when comparing the photophoresis theory (Ref. 14) with the experiment (Ref. 4)); 150 (maximum value for mass samples)

3. Main results

Analysis of the results obtained was made using the following representation: first the above-mentioned characteristics were considered for most suitable in thermal-physical characteristics (i.e., light and slightly heat-conducting) types of atmospheric aerosol (first, for soot particles), and then the characteristics were considered for the known types of stratospheric and mesospheric aerosol layers. In analyzing the second group of aerosols the classification and conclusions of the survey were used.[11] Characteristics of the considered soot particles are classified in Table 1.

As in the case of "solar" photophoresis,[8] the soot particles demonstrate high sensitivity to possible photophoretic effects. Figure 5 shows the calculated results on the values α and β obtained using Eqs. (6) and (7) for the real but very low, from the viewpoint of characteristics of mass samples, values of density and thermal conductivity of particles.

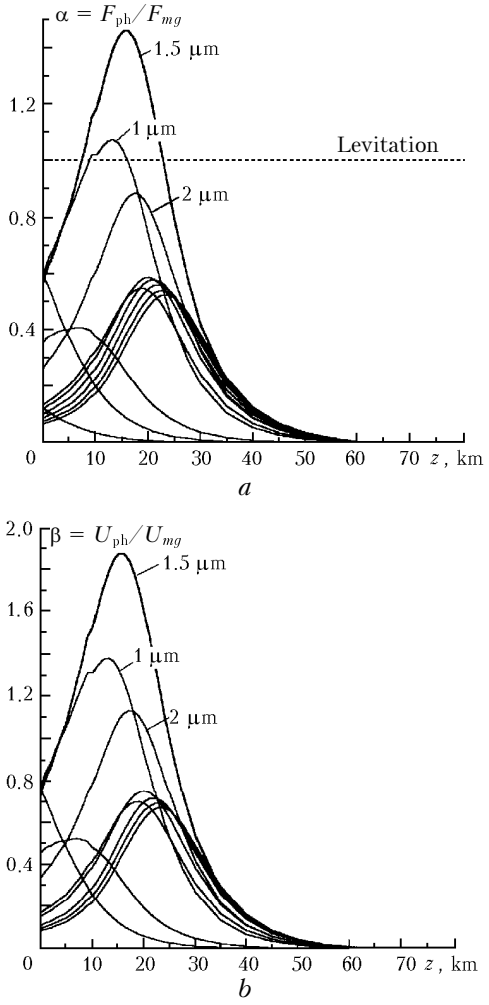

Fig. 5. Relationships between the photophoretic force and the force of gravity (*a*) and the photophoretic velocity and the velocity of gravitational sedimentation (*b*) for soot particles of different size (ρ_p = 165 kg/m^3, Λ = 5) depending on the height *z*.

In particular, for our analysis we have chosen the values ρ_p = 165 kg/m^3 and Λ = 5 because those give good agreement between the theory of photophoresis[14] and the laboratory experiment.[4] The calculations were made with the use of the U.S. standard atmosphere and the value $m = 2.42 + 1.02i$ (λ = 10.6 μm) for the particle dimensions R_0 = 0.05–5.0 μm. For the soot particles of smaller dimensions the photophoretic effects are maximum at altitudes lower than 10 km (i.e., in the troposphere) where the developed model is unlikely to be rather correct.

It is evident that the particles of dimensions $R_0 \approx$ ≈ 1.5–1.6 μm demonstrate the maximum photophoretic effect (this dimension corresponds to the gas kinetic maximum of photophoretic force and speed for the soot particles). Sufficiently large particles of the size R_0 = 0.95–1.9 μm can move vertically against the force of gravity.

Figure 6 illustrates the conditions of maximum height such particles ascend (if they move from bottom to top) or the minimum descend due to precipitation (if for some reasons they first were at a higher altitude). For a comparison some results are given on the geometrically optimum conditions of the soot particles levitation under the action of "solar" photophoresis[8] (it is assumed that the Sun is at zenith, therefore the radiation flux is maximum).

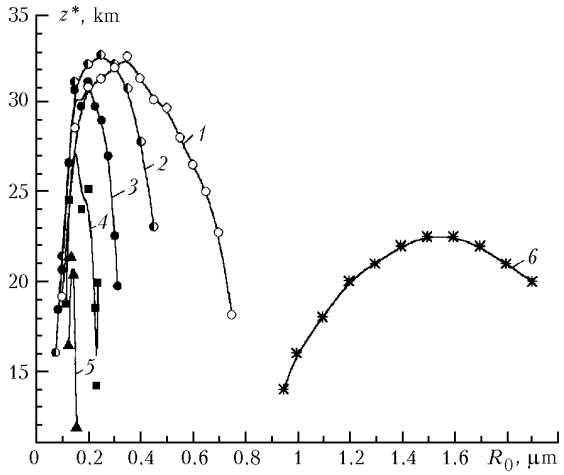

Fig. 6. Heights of photophoretic levitation *z** for soot particles of different size (ρ_p = 165 kg/m^3, Λ = 5, *T* = 220 K). "Solar" photophoresis *n* = 1.95, *k* = 0.05 (*1*); 0.1 (*2*); 0.2 (*3*); 0.3 (*4*); 0.4 (*5*). "Thermal" photophoresis: $m = 2.42 + 1.02i$ (*6*).

The particle levitation at altitudes 14–23 km for the thermal photophoresis and at 18.5–32 km for the "solar" photophoresis denotes a particular stratification of the soot aerosol in the stratosphere. The thickness of the predicted aerosol layer is rather significant (more than 15 km in the lower stratosphere). In this case the "solar" levitation at $m = 1.95 + 0.2i$ is possible for soot particles of smaller size (0.08–0.3 μm) but approximately in the same altitude range. These conclusions quantitatively agree with the data on the detection of soot aerosol

at altitudes above 20 km presented in Refs. 9 and 32, but call for a careful analysis and comparison. Of course, in this case the questions arise on the conditions of the formation, chemical and disperse composition, morphology, consequences of interaction of soot particles and water vapor (one of the effective mechanisms of compactness of the volume fractional soot clusters.[33] All this, unfortunately, is beyond the scope of the present paper and requires additional analysis and discussion.

The calculations of the above-mentioned characteristics were made under changes of the values of density and thermal conductivity of particles in the intervals $\rho_p = 165–2000$ kg/m^3 and $\Lambda = 5–150$. The variation of possible values of the complex refractive index (see Table 1) results in a slight change of the factor J_1 (Fig. 4*b*) that affects the values of the force and velocity of photophoresis of soot particles. In all cases we have obtained essentially the same qualitative behavior of the values α and β, similar to the variations shown in Fig. 5. The photophoresis effect is found to appear also at maximum considered densities and thermal conductivities of particles corresponding to the values for massive samples. The increase of thermal conductivity in this case has a stronger effect on the decrease of photophoretic effects as compared with the increase of particle density. It is to be recalled that the particle density determines the particle force of gravity while the thermal conductivity determines the photophoretic force: when increasing the values of Λ the temperature of the particle surface becomes more homogeneous thus decreasing the radiometric effect.

Figure 7 shows the estimations of times of the soot particle sedimentation with $\rho_p = 500$ kg/m^3 and $\Lambda = 5$ from altitudes down from 100, 30, and 20 to 10 km altitude (the tropopause boundary) with and without the account of the effect of photophoretic forces. Note that the term "time of sedimentation" is not identical to the mean time of particle residence at some altitude in the atmosphere because this term does not take into account different mechanisms of particle sink of a given chemical composition and dimensions. It is evident that the photophoresis significantly increases the time of sedimentation of particles of different size as compared with the times of gravitational sedimentation. Especially this is well-defined for soot particles of size 1–2 μm, for which the values of photophoretic forces are maximal.

Thus, it is assumed that the combined effect of "solar" and "thermal" radiometric photophoresis can be a significant and effective mechanism of vertical transport of soot aerosol in the lower stratosphere. Clearly, this fact is based on the results of calculations using the model of optically homogeneous spherical soot particles (in a more detail of the compact isometric particles), taking correct account of their thermal physical characteristics. Later on it will be necessary to analyze photophoretic characteristics of morphologically more complex particles (for example, two-layer particles) of different composition with a soot nucleus, shell of nucleus and volume inclusions, which are widely distributed in the stratosphere.[33,34]

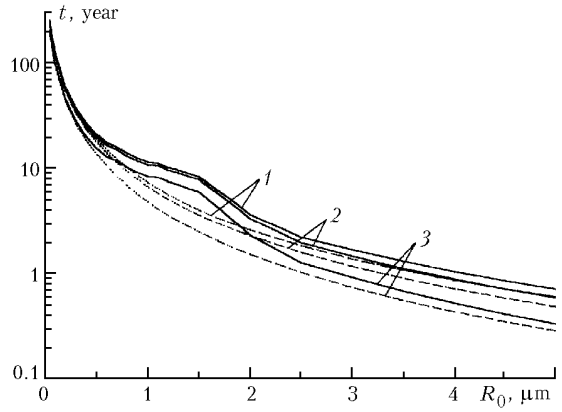

Fig. 7. Times of sedimentation of soot particles (ρ_p = = 500 kg/m^3, $\Lambda = 5$) from the stratosphere to the tropopause boundary: solid curves denote the gravitational sedimentation and photophoresis; dashed curves denote the gravitational sedimentation: 100–0 km (*1*); 30–0 km (*2*); 20–0 km (*3*).

Preliminary analysis of the influence of "thermal" photophoresis on the particle dynamics of the known aerosol layers has shown that the Junge layer (drops of 60–80% solution of sulfuric acid) is characterized by strong absorption in the infrared spectral range ($\lambda > 2.5$ μm).[31] This fact explains large absolute values of the asymmetry factor J_1 for such particles calculated by the authors (these values are not cited here). However, at the cost of large density and moderate thermal conductivity of such particles the parameter α is small as compared with the soot particles; this implies that the competition of the photophoresis forces with the forces of gravity is impossible. Similar conclusion can also be drawn for polar stratospheric clouds of the first kind,[11] the particles of these clouds are too heavy and heat-conducting, although they possess clearly defined absorbing properties in the longwave range. For polar stratospheric clouds of the second kind (crystal water ice) photophoretic forces are small due to low radiation absorption and very high thermal conductivity of particles. Finally, for particles of polar mesospheric clouds the photophoresis is small due to very high altitudes and low air pressures (in the free-molecular regime $F_{ph} \sim p(z)$). Thus the radiometric photophoresis of particles in the field of outgoing Earth's thermal radiation is not, evidently, a significant mechanism for the maintenance of stability and long times of existence of the known aerosol layers in the stratosphere and mesosphere. This problem requires further analysis and discussion.

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