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Vertical Aerosol Transport in the Middle Atmosphere

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This study continues the analysis of the determining forces responsible for vertical aerosol transport in the middle atmosphere (upper troposphere, stratosphere and mesosphere) on global time and spatial scales.
One of possible regular mechanisms of vertical transport can be the well-known radiometric photophoresis. The developed theory forecasts, that for some types of soot aerosols the negative “solar” and positive “thermal” photophoresis (motion of particles in the field of outgoing thermal radiation) can result in levitation and vertical lifting of particulates against gravity in the upper troposphere–lower stratosphere.
The results of theoretical and experimental investigations of so-called “accommodation” forces acting on particles with asymmetry of surface properties in rarefied gas in the directed electromagnetic radiation field (gravito-photophoresis) are presented. The estimations show that action of the investigated force is not capable to provide effective vertical transport of aerosol particles against gravity at altitudes of the lower and middle stratosphere.
Among regular and effective mechanisms of aerosol transport it is necessary to note the vertical stratospheric wind. The unique opportunity of this analysis is given with a database of the research satellite UARS (1991-2005) together with the UKMO global circulation model of the middle atmosphere. It is shown that the vertical wind caused by specific long-term circulation of air masses in stratosphere and mesosphere can play a determining role in vertical transport of particles of any thermal-physics and optical properties at high altitudes.

1. Introduction

In thermally and mechanically stable stratosphere the marked manifestations of independent transport characteristics of particles should be noticeable, which are not limited only to the convective aerosol transfer due to the tropospheric circulation. Vertical forces of different physical nature and magnitude can affect the particles of stratospheric aerosol of different origin, chemical composition and morphology in the radiation field. It is well known that the ponderomotive forces (forces of light pressure) are decisive for the aerosol dynamics in the intensive laser beams, but are negligible for aerosol particles in the field of solar radiation. At low gas pressures and intensities of incident radiation the vertical thermoconvective forces are not significant, produced by carrying away the particles by gas volumes surrounding a heated particle.
The forces of radiometric photophoresis are reasonably traditional in the analysis of vertical motions of stratospheric aerosol. In particular, in recent publications (Beresnev

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et al., 2003; Beresnev and Kochneva, 2003) the results are presented of calculations of characteristics of photophoresis particle motion of some types of atmospheric aerosol (mainly – soot particles) at altitudes of lower and middle stratosphere. The obtained data enable us in a new way to assess the potentialities of the radiometric photophoresis in the vertical transport of absorbing aerosol in the middle atmosphere.

Among regular and effective mechanisms of aerosol transport it is necessary to note the vertical stratospheric wind. The unique opportunity of this analysis is given with a database of the research satellite UARS (1991-2005) together with the UKMO unified model of middle atmosphere. It is shown that the vertical wind caused by specific long-term circulation of air masses in stratosphere and mesosphere can play a determining role in vertical transport of particles of any thermal-physics and optical properties at high altitudes.

2. Radiometric Photophoresis

Consider a spherical particle with radius R_p suspended at the altitude z in the field of a directional electromagnetic radiation (Fig. 1). The particle is characterized by density ρ_p , the heat conductivity ratio $\Lambda=\lambda_p/\lambda_g$, and the complex refractive index $m=n+ik$ depending on the wavelength of incident radiation λ. The particle is in the solar radiation field with intensity I_S=1368 W/m^2 (solar constant) with effective wavelength λ=0.50÷0.55 μm, and in the outgoing thermal radiation field with effective wavelength of the order λ=10÷13 μm. The intensity of thermal radiation I_{th} is a function of altitude z due to the radiation absorption by aerosol and gaseous components of atmosphere.

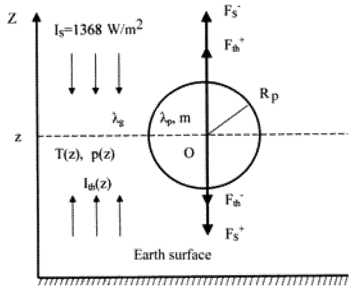

Fig. 1. Statement of the atmospheric photophoresis problem.

Photophoretic forces of radiometric nature 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) and the sign «-» refers to the forces of negative photophoresis (motion of particles opposite to the direction of radiation). It is established (Beresnev and Kochneva, 2003) that the direction of particle motion is determined only by their optical properties and size, and is independent of gas-kinetic regime and accommodation characteristics of the particle surface.

Owing to the absorption of outgoing long-wave 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 long-wave radiation are formed, which are characteristic of different latitudes and seasons. Fig. 2 shows the calculations of $I_{th}(z)$ in the altitude range from 0 to 100 km using standard latitude-seasonal models of atmosphere.

The spectral photophoretic asymmetry factor J_1 is the basic result of solution of the electrodynamic problem of photophoresis (Beresnev and Kochneva, 2003). This factor is determined by the internal field in particle and depends on the magnitude of size parameter ρ and complex refractive index m:

$$J_1 = 3nk\rho \int_0^{\pi} sin\,\theta \cdot P_1(cos\,\theta) d\theta \int_0^1 x^3 B(x,\theta,\phi = \frac{\pi}{4}) dx\,, \tag{2}$$

where $B(x,\theta,\phi)$ is the dimensionless source function, $\rho = 2\pi R_P/\lambda$ is size parameter, and $x = r/R_P$ is dimensionless radial coordinate. The quantity J_1 is normalized and varies within the limits $-0.5 < J_1 < 0.5$. Negative values of J_1 correspond to heating of front particle side and fit the positive photophoresis; positive values correspond to heating mostly of rear particle side and the negative photophoresis. Figs. 3,4a show the calculated results for J_1 obtained according to the values of m for soot particles in the range $\lambda = 0.5$ µm and $\lambda = 10.0 \div 10.6$ µm. 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 on Fig. 4a).

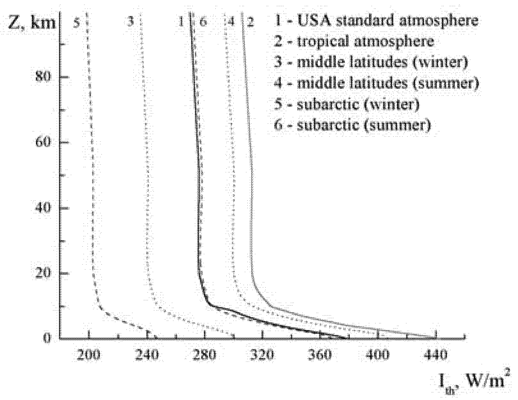

Fig. 2. The intensity of outgoing thermal radiation I_{th} as a function of altitude z (Beresnev et al., 2003).

The calculation of force and velocity of radiometric photophoresis is based on the molecular-kinetic theory of this phenomenon (Beresnev et al., 1993). This theory is based on the solution of gas-kinetic model equation with the corresponding boundary conditions for the velocity distribution function on the particle surface, and covers the entire range of the Knudsen number Kn ($Kn = l/R_P$, where l is the mean free path of gas molecules) at arbitrary ratio between the thermal

conductivities 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 expressions for the photophoretic force and velocity in the entire range of Kn number are of the forms (Beresnev et al., 1993):

$$F_{ph} = -\frac{2\pi}{3}\left(\frac{\pi M}{8RT_0}\right)^{1/2} R_P^2 I J_1(\rho, m) F(Kn, \Lambda), U_{ph} = -\frac{\pi}{2(8+\pi)}\frac{IJ_1}{p_0}\Phi(Kn, \Lambda), \qquad (3)$$

where $F(Kn,\Lambda)$ and $\Phi(Kn,\Lambda)$ are the functions of the Kn number, parameter Λ, coefficients of accommodation of momentum and energy of gas molecules on the particle surface. The full velocity of vertical particle transport U is determined as

$$U(z) = U_{mg}(z) + U_{ph}(z). \qquad (4)$$

Using the Eqs. (3-4) we can estimate the particles sedimentation times from some high altitudes to the tropopause boundary with or without of the photophoretic effect.

2.1 "Solar" radiometric photophoresis

Here calculations of photophoretic characteristics have been executed for typical representatives stratospheric aerosols at reliable values of thermal-physics and optical parameters. The basic interest are data for the well absorbing, light and low heat-conductivity carbonaceous particles. Some results are presented on Fig. 3.
Maxima of photophoretic force on 10÷12 km are caused it gas-kinetic extremum at $Kn \approx 0.30 \div 0.35$, corresponding to the given altitudes. Absolute values of forces are small (10^{-17}÷10^{-14} N), but quite sufficient for a competition to gravity. Velocities photophoretic motion make units and tens μm/s, and they are maximal for particles with $R_p = 0.2 \div 0.4$ μm. Increase in heat conductivity and density of particles predictedly reduces photophoretic velocities, and above 15 km U_{ph} values for the particles with different sizes do not depend practically on altitude z. Appreciably, that in stratosphere negative "solar" photophoresis generates the vertical velocities comparable on size and opposites on a sign to velocities of gravitational sedimentation. At the certain altitudes z^*, where the gravity is counterbalanced by opposite directed negative "solar" photophoretic force, the levitation of particles is possible. Soot particles with $R_p = 0.075 \div 0.75$ μm and $k = 0.05 \div 0.4$ can levitate at heights 12÷32 km in the lower and middle stratosphere.
Thus, the "solar" photophoresis in a stationary stratosphere can lead to the formation of wide layers of submicronic soot aerosol. For particles with $R_p = 0.2 \div 0.4$ μm the characteristic times of photophoretic lifting from Earth' surface up to heights of levitation make 25÷30 years.

2.2 "Thermal" radiometric photophoresis

As in the case of "solar" photophoresis, the soot particles demonstrate a high sensitivity to possible photophoretic effects in the thermal radiation field. Soot particles in the field of thermal radiation demonstrate only positive photophoresis, and its photophoretic velocities change from the tenth shares up to hundreds μm/s (Fig. 4).

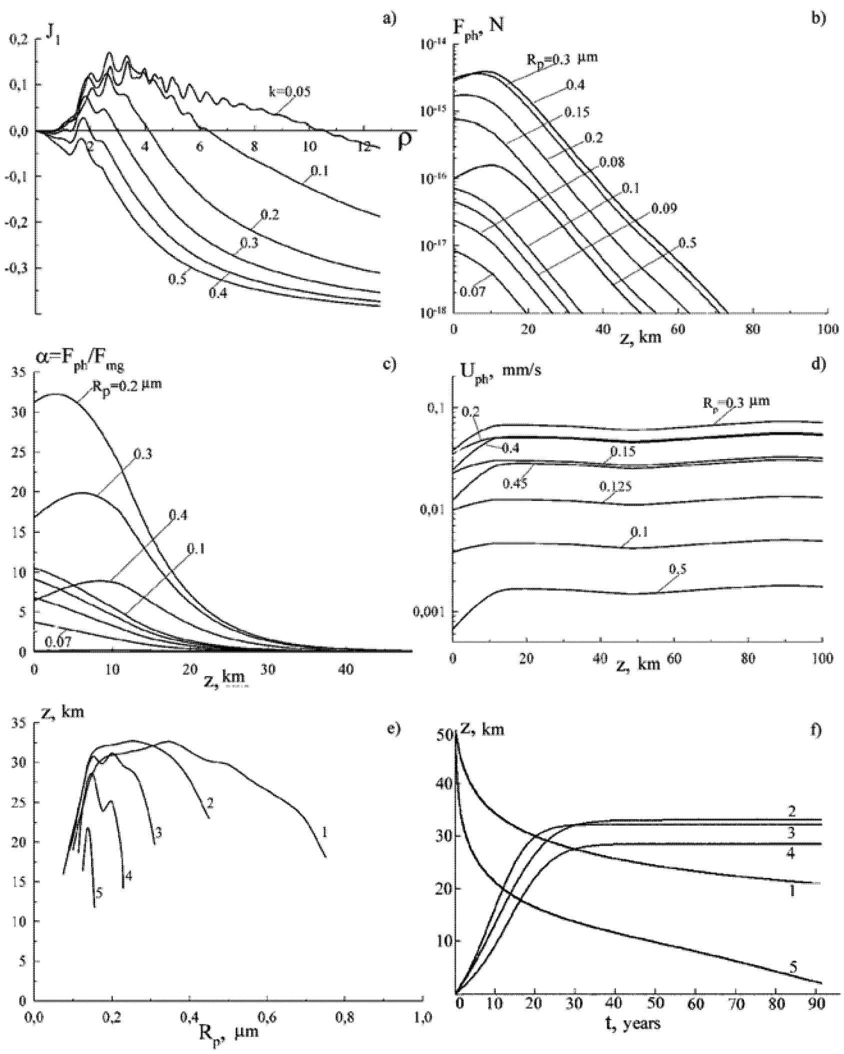

Fig. 3. Characteristics of "solar" photophoresis for compact carbonaceous particles with m=1.95+0.1i, Λ=5 and ρ_p=0.165 g/cm^3:

a) photophoretic asymmetry factor J_1 vs size parameter ρ at λ=0.5 μm for soot particles with n=1.95 and k=0.05÷0.5;

b) negative "solar" photophoretic force F_{ph} as function of altitude z;

c) ratio $\alpha=F_{ph}(z)/F_{mg}$ as function of altitude z;

d) velocities of soot particles under action of negative "solar" photophoretic forces U_{ph} as function of altitude z;

e) levitation altitudes z^* for particles with size R_p in stratosphere under action of negative "solar" photophoresis: 1 - k=0.05, 2 – 0.1, 3 – 0.2, 4 – 0.3, 5 – 0.4;

f) sedimentation times for particles with R_p=0.07 μm (1) and 0.5 μm (5) from altitude of 50 km and rising times up to levitation altitudes in the short-wave radiation field for particles with R_p=0.2, 0.3 and 0.4 μm (lines 2,3 and 4, respectively).

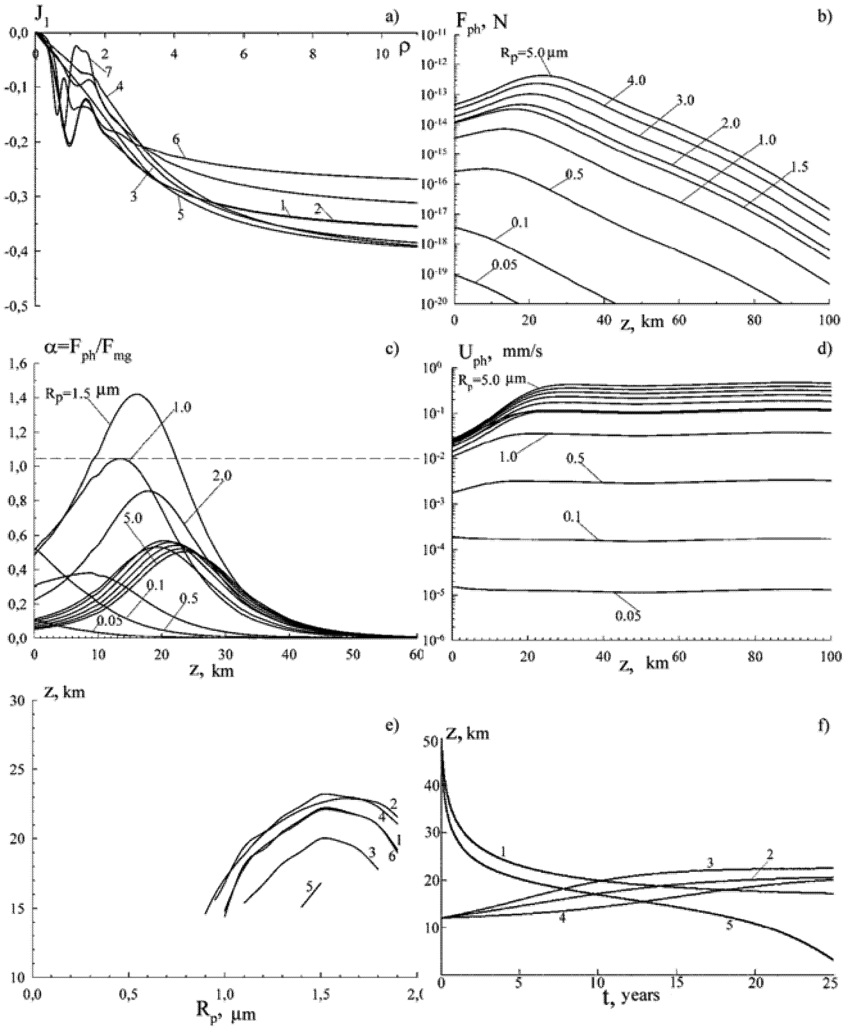

Fig. 4. Characteristics of "thermal" photophoresis for compact carbonaceous particles with m=2.42+1.02i, Λ=5 and ρ_p=0.165 g/cm^3:

a) photophoretic asymmetry factor J_1 vs size parameter ρ for soot particles at λ=10.0÷10.6 μm for the most reliable values of $m=n+ik$;

b) positive "thermal" photophoretic force F_{ph} as function of altitude z;

c) ratio $\alpha=F_{ph}(z)/F_{mg}$ as function of altitude z;

d) velocities of soot particles under action of positive "thermal" photophoretic forces U_{ph} as function of altitude z;

e) levitation altitudes z^* for particles with size R_p under action of positive "thermal" photophoresis for different standard seasonal-latitude models;

f) sedimentation times for particles with R_p=1.0 μm (1) and 2.0 μm (5) from altitude of 50 km and rising times up to levitation altitudes in the long-wave radiation field for particles with R_p=1.3, 1.5 and 1..8 μm (lines 2,3 and 4, respectively).

Particles with $R_p = 1.5$ μm show maximal photophoretic effect (this size correspond to gas-linetic maximum of photophoretic force). Particles with $R_p = 0.95 \div 1.9$ μm can test vertical rise against gravity, and then levitate due to forces of "thermal" photophoresis at heights 15÷23 km. Particles with $R_p = 1.3 \div 1.8$ μm achieve heights of levitation for times 20÷25 years. In spite of the fact, that intensity of thermal radiation make about a quarter of intensity of a short-wave solar radiation, the "thermal" photophoresis can be considered as the effective power mechanism of vertical transport for stratospheric aerosol. Possessing similar with "solar" photophoresis qualitative characteristics (vertical lifting of particles against gravity up to opportunity of a levitation), the "thermal" photophoresis operates effectively on larger particles, and the corresponding heights of particles levitation decrease. The expressed daily run for "thermal" photophoresis is absent, as the fluxes of outgoing thermal radiation possess small variability on time scales from day till a season. Total action of "solar" and "thermal" photophoresis can be shown in vertical transport of soot particles against a gravity in size interval $0.075 < R_p < 1.9$ μm with actual overlapping two ranges of the sizes.

3. "Accommodation" Forces (Gravito-Photophoresis Phenomenon)

In 1951 the Austrian scientists F. Erenhaft and E. Reeger have reported about the observations of a so-called "transverse" photophoresis: aerosol particles at illumination by a horizontal beam of solar radiation demonstrated the various motion trajectories including a vertical rise opposite the gravity. The discovered phenomenon (appearance of the force and the positive vertical velocity of the particle motion in the field of arbitrarily directed radiation) was called as gravito-photophoresis. The goal of this investigations was, firstly, the independent development of the successive gas-kinetic theory of the phenomenon in the free-molecular regime and, secondly, an attempt of the measurement of predicted forces using methods of model thermal physical experiment with macroparticles with the goal of a quantitative comparison of the results with the theoretical predictions.

As an object measuring the "accommodation" forces a steel particle was chosen (R_p = 0.52 cm; p_p = 7.8 g/cm^3; λ = 14.8 W/mK) in helium at sufficiently low pressures corresponding to the near-free-molecular regime of gas flow. Three different experimental situations were realized serially: a well-polished homogeneous particle; a particle with highly rough frontal and polished back hemispheres; a rough homogeneous particle in the field of directed radiation.

The developed gas-kinetic theory of "accommodation" forces in the free-molecular regime explains the mechanism of their occurrence by the difference in values of the accommodation coefficients of a normal momentum of gas molecules α_n (but not the energy accommodation coefficients α_E) on different sides of a model particle. The evaluations when using the known values of α_n show that in the stratosphere the "accommodation" forces can be compared with the forces of the radiometric photophoresis but do not exceed the gravity. These conclusions both qualitatively and quantitatively differ from the conclusions of the semiempirical theory of gravito-photophoresis developed for explaining the experiments (Pueschel et al, 2000).

The measurements made using methods of model thermal physical experiment with macroparticles confirm the existence of “accommodation” forces affecting a particle with artificial asymmetry of surface characteristics together with the forces of radiometric photophoresis. The experimental values for the system “steel particle-helium” are in good agreement with theoretical predictions; in this case the relation between the “accommodation” force and the photophoretic force does not exceed 3%.

In the authors opinion, the effect of action of the “accommodation” forces is insufficient for explaining large velocities of vertical particle motion against the gravity in the model of Pueschel et al. (2000), but the theory of gravito-photophoresis can explain these phenomena. A further analysis of the above experimental data is required with the goal of detection of action of the other forces, which were not considered in the analysis.

It is obvious that to explain the phenomenon of accumulation of soot particles from the air transport engines at altitudes of lower and middle stratosphere (Pueschel et al., 2000) the other reasons must be searched, except for the action of “accommodation” forces and even forces of radiometric photophoresis. One of constantly and profitably acting factors can be the influence of the positive vertical velocity of stratospheric wind.

4. Vertical Wind in the Middle Atmosphere

The vertical wind probably is the determining factor of particles motion up to altitudes 30÷40 km and can change essentially the characteristics of sedimentation and residence times of atmospheric aerosols. The latitudinal and seasonal dependences of vertical wind at different altitudes averaged for the various time intervals since 1992 to 2006 according to the model UKMO are analyzed. It is established, that monthly-averaged amplitudes of vertical wind make values ±5 mm/s, and the annual-averaged are ±1 mm/s. Ascending wind provides vertical lifting against gravity for the sufficiently large aerosol particles (up to 3÷5 μm) with density up to 1,0÷1,5 g/cm^3 at stratospheric and mesospheric altitudes. The structure of the averaged vertical wind fields supposes the opportunity of formation of dynamically stable aerosol layers in the middle stratosphere. Details of climatological investigation of vertical wind field in the middle atmosphere are presented in separate paper of authors (Gryazin and Beresnev, 2008).

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5. References

Beresnev, S., V. Chernyak and G. Fomyagin, 1993, Phys. Fluids A 5, 8, 2043.
Beresnev, S. and L. Kochneva, 2003, Atmos. Ocean. Optics 16, 2, 134.
Beresnev, S., F. Kovalev, L. Kochneva et al., 2003, Atmos. Ocean. Optics 16, 1, 52.
Beresnev, S., L. Kochneva, P. Suetin et al, 2003, Atmos. Ocean. Optics 16, 5–6, 470.
Gryazin, V. and S. Beresnev, 2008, Chem. Eng. Trans., this issue.
Pueschel, R.F., S. Verma, H. Rohatschek et al., 2000, J. Geophys. Res. 105D, 3, 3727.