Photophoretic effects for the black carbon aerosols at stratospheric altitudes

L.B. Kochneva, S.A. Beresnev, V.I. Gryazin, M.V. Vasiljeva, K.G. Gribanov and V.I. Zakharov

Ural Federal University, Ekaterinburg, 620083, Russia
Keywords: fundamental aerosol physics, stratospheric aerosols, photophoresis, soot aerosol
Presenting author email: Louisa.Letfulova@usu.ru

This report continues and summarizes analysis and estimations of the photophoretic effects for soot aerosols in stratosphere. One of possible mechanisms of aerosol transport in the thermally and mechanically stable stratosphere can be radiometric photophoresis as the regular factor of aerosol vertical motion on the synoptic and global temporal scales. It can lead, for example, to the uncontrollable accumulation of soot particles at the certain altitudes in stratosphere from aircraft engines and biomass burning.

At the first stage of studies the estimations of transport opportunities of negative "solar" (motion of particles in the field of short-wave solar radiation against gravity) and positive "thermal" (motion in the field of thermal outgoing radiation) photophoresis of soot particles have been given at the assumptions of the highest possible intensities of atmospheric radiation fluxes. In this report we present updated and expanded calculations of the photophoretic motion characteristics according to the advanced radiation block of the model. Firstly, the results for “solar” photophoretic motion are calculated in the framework of advanced model for short-wave solar radiation (Beresnev *et al*, 2012a). Secondly, the characteristics for “thermal” photophoresis are specified taking into account the downward flux of the long-wave thermal radiation (Beresnev *et al*, 2011).

The results of theoretical analysis of photophoretic motion of soot particles in the short-wave solar radiation field for stationary atmosphere are presented. The integrated fluxes of solar radiation were calculated by the Monte Carlo method with approximation of a plane-parallel, horizontal-homogeneous molecular-aerosol atmosphere. The analysis confirms again an opportunity essential photophoretic effects for soot aerosol particles in stratosphere: the "sun" photophoresis can be considered as the effective mechanism of vertical transport of well-absorbing sub-micrometer particles up to heights of the middle stratosphere. The forces of "sun" photophoresis can compete to gravities up to heights 30-35 km for particles of sub-micrometer and micrometer sizes. On other hand, the estimations have shown again the small efficiency of radiometric photophoresis for weakly-absorbing, dense and high-conducting particles of atmospheric aerosol. The results for photophoretic motion of soot particles in the field of the Earth’ thermal radiation in a stationary atmosphere are also presented. In calculations the up- and down-fluxes of thermal radiation are taken into account. It is shown that positive "thermal" photophoresis potentially can be the effective mechanism of vertical transport for micron-sized soot particles at stratospheric altitudes.

Besides, in this report the theoretical and experimental estimations for so-called “accommodation” (gravito-photophoretic) forces which can compete partly with radiometric photophoretic forces are presented. We discuss, first, the independent development of the consecutive and rigorous gas-kinetic theory of the phenomenon for the free-molecular regime with detailed consideration of gas-surface interaction features and, second, an attempt of the measurement of predicted forces using an experimental technique with model macroparticles for quantitative comparison with the theoretical predictions (Beresnev *et al*, 2012b).

The theoretical estimations show that in the upper troposphere and in the middle stratosphere the “accommodation” (gravito-photophoretic) forces can be comparable with the radiometric photophoretic forces, but do not exceed the gravity force. These conclusions both qualitatively and quantitatively differ from the conclusions of the semi- empirical theory of gravito-photophoresis by Rohatschek (1984).

The measurements with the appropriate model macroparticles in vacuum chamber have confirmed 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, and the ratio of the “accommodation” force to the photophoretic force does not exceed 3%.

Thus, the action of "accommodation" (gravito-photophoretic) forces, in our opinion, is not capable to provide effective vertical transport of aerosol particles against gravity at stratospheric altitudes.

This work was supported by the Ministry of Education and Science of the Russian Federation (research project implemented by leading scientist at Russian institution of higher education, agreement No.11.G34.31.0064).

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