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Photo-Thermophoresis as a New Tool for Aerosol Characterization

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Abstract. Aerosols are particles in the size range of nanometers to some micrometers, suspended in air or other gases. Their characterization and separation according to different physical and chemical properties is of great interest for environmental analysis and certain industrial applications. Photo-thermophoresis is an optothermal effect, which occurs when aerosol particles are illuminated by intense light. Locally inhomogeneous heating, resulting in locally increased impingement rates of the gas molecules, generates driving forces. These photophoretic forces can move fine particles either towards or away from the light source. We employ this effect for characterization and separation according to optical and thermal properties of aerosols. We hope to establish this new approach as a helpful supplement to the existing separation methods based on electrical, thermal or flow field forces.

1. Introduction

Atmospheric aerosols originate from a broad diversity of natural sources (e.g. volcanic eruption, air guided raise of mineral dust or sea salt) as well as anthropogenic sources (e.g. combustion of biomass or fossil fuels). The air-suspended particles can have significant influences on the earth's atmosphere. They scatter and absorb solar and terrestrial radiation and contribute to the formation of clouds and rainfall by serving as condensation respectively ice nuclei [1]. In addition, air-suspended particles play an important role in the distribution of microorganisms, reproductive material and pathogens and can cause several diseases themselves [2]. This broad variety of interferences concerning human health and the environment make aerosols an important subject of research. A wide range of techniques for aerosol measurements is therefore understandable [3]. Separation according to size or size related quantities is possible using diffusion batteries [4], cascade impactors [5], electrical low pressure impactors [6] or electrostatic classifiers such as differential mobility analyzers [7]. To our knowledge, there exists no method for the separation of particles according to optical or optothermal properties.

Photophoresis denotes the phenomenon that small particles illuminated by a beam of light of sufficient intensity move on paths of varying degrees of complexity. Besides the characteristics of the light beam as well as pressure and composition of the carrier gas, the motion depends on size, shape, thermal conductivity and optical properties of the particles [8]. A separation method based on photophoretic mobility can therefore fill the gap mentioned before.

2. Theory

Illumination of a particle is always attended by absorption of light and therefore heat production within it. A thermo-photophoretic force arises when unevenly heating of the particle leads to temperature differences in the surrounding gas. Molecules of the heated gas in close proximity of hot particle regions start to impinge on the surface with higher kinetic energy than molecules in close proximity of cold surface regions resulting in a net force on the particle contrary to the temperature gradient.

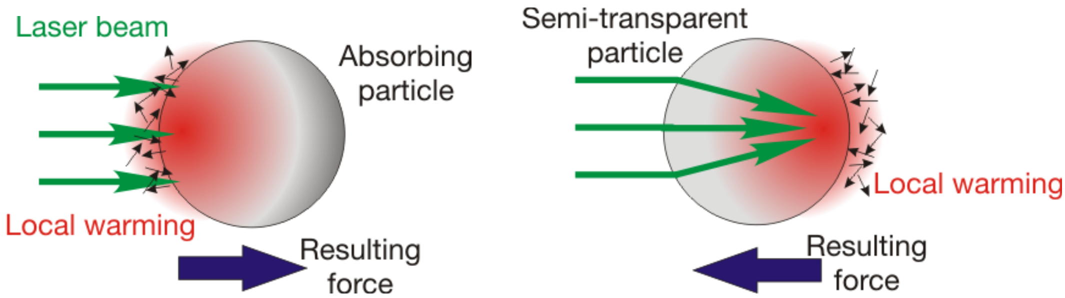

Figure 1 Positive (left) and negative (right) photo-thermophoresis.

The distribution of the heat-source function within an illuminated particle is directly determined by the internal electric field distribution. Calculations based on Mie theory demonstrate that the internal field distribution strongly depends on the size and complex refractive index of the sphere [9]. Only minor changes in size and/or refractive index can cause a change of direction of the thermo-photophoretic force (see figure 1).

There are numerous theoretical studies on photo-thermophoresis, whereas experimental studies are sparse. Several authors deduced expressions to quantify the thermo-photophoretic force. We only want to refer to the analytical appoach obtained by Reed [10]. Reed developed a formula for the thermo-photophoretic force on a spherical particle suspended in gas for the slip flow regime (small Knudsen number). The Knudsen number denotes the ratio of the mean free path λ of the gas molecules to the particle radius *r*. Descriptions in the slip flow region are only justified for Knudsen numbers up to about 0.25 [11]. Reed's expression reads as follows:

$$F_P = \frac{3}{2}\pi\eta^2 \cdot P_L \cdot f \frac{2r \cdot R}{p \cdot M \cdot K_s} \left\{ \frac{1}{1 + 3 \cdot C_m \frac{\lambda}{2r}} \right\} \left\{ \frac{1}{1 + 2 \cdot \frac{K_f}{K_s} + 2 \cdot C_t \frac{\lambda}{2r}} \right\}$$

Here, ρ is the density, T the temperature (far from the sphere) and k_g the thermal conductivity of the gas. In addition, the thermal conductivity of the particle is required as well as the total light intensity I. The fraction f of the incident light that is absorbed involves particle optical properties in the calculation. κ is a dimensionless constant called thermal creep coefficient and takes values between 0.75 and 1.50. C_m and C_t are dimensionless constants as well, and take values between 1.00 to 1.35 respectively between 1.875 to 2.48 for most technical applications.

The friction force acting on the moving particle can be determined by Stokes' law:

$$F_f = 6\,\pi\,\eta\,r\,v$$

were η is the viscosity of the medium. It counteracts the thermo-photophoretic force, resulting in an equilibrium velocity for a given set of particle properties and light intensities, which is denoted to photophoretic velocity in the latter.

Another force acts on a particle in a strong laser field, the well-known radiation pressure or direct photophoresis. It is due to the transfer of momentum from photons being scattered on the particle. For particles suspended in gases it is generally significantly lower than the thermo-photophoretic force, while for particles suspended in liquids becomes dominant [12].

3. Experimental Setup

3.1. Flow cell
Experiments were carried out in a one-piece aluminum flow cell, made in-house (see figure 2). It has a square cross section of 1 cm^{-1} and a total length of 5 cm. Aerosol in-and outlet are positioned at both ends of the measuring cell under an angle of 45° with respect to the z-axis. Where the laser beam enters respectively leaves the cell, fused silica windows are embedded flush with the inner cell wall. The recording of particle movements is possible using a CCD camera equipped with a macro objective located in front of one of the two glass viewing windows. The aerosol flow through the cell is chosen upwards, against gravitation. It can be controlled with a needle valve behind the flow cell controlling the suction of a diaphragm pump.

3.2. Optical System
In a first setup, a cw laser diode (λ = 806 nm, P = 0.5 W) served as light source. Using fiber optics, the laser beam was directed towards the flow cell and locked into position by means of a metal socket. The diverging beam of light was collimated using a plano-convex lens with a focal distance of 40 mm. Another lens focuses the laser beam to the middle of the cell. In addition to the laser diode, a frequency-doubled cw Nd:YAG laser (λ = 532 nm, P = 20 mW) was employed to illuminate the particles in a larger region for camera observation. In a second setup, a cw Nd:YAG laser λ = 532 nm, P = 1 W, Changchun New Industries Optoelectronics Tech. CO,. Ltd) was used as light source. The optical output power of the laser could be adjusted. The laser beam was directed towards the flow cell by means of two dichroic mirrors and focused to the middle of the cell using a plano-convex lens with a focal length of 50 mm. To straighten out the distorted laser profile a movable pinhole (∅ 2.5 mm) was placed in front of the focusing lens.

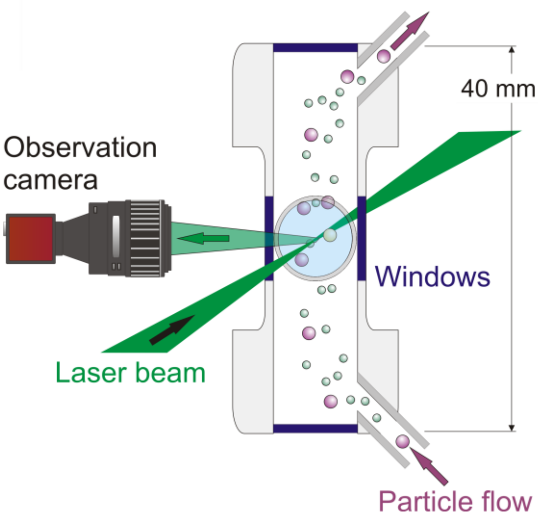

Figure 2 Schema of the experimental setup for the measurement of photo-thermophoresis on aerosols.

3.3. Evaluation system

The particle movements were recorded using a CCD camera (Guppy, GF 080B, Allied Vision Technology GmbH, Germany) adapted to a macro objective (AF Micro-Nikkor 50 mm 1:2.8DG, Sigma GmbH, Germany). The field of view is reduced to a rectangle with a width of 3.89 mm and a height of 0.97 mm, resolved on 800 × 200 pixels (1 pixel corresponds to 4.861 µm). Particles are tracked with a LabView tool, allowing the real-time evaluation with 15 frames per second. The collected data was further processed with Matlab (version 7.1, National Instruments). The employed particle tracking tool was also developed using LabView. The core distribution was supplemented by NI IMAQ Vision, an image acquisition and processing add-on package. It includes a number of image analysis virtual instruments (VIs) which are the basis for our tracking algorithm. To make real time tracking possible, the CCD camera was connected to the computer via an IEEE 1394 interface.

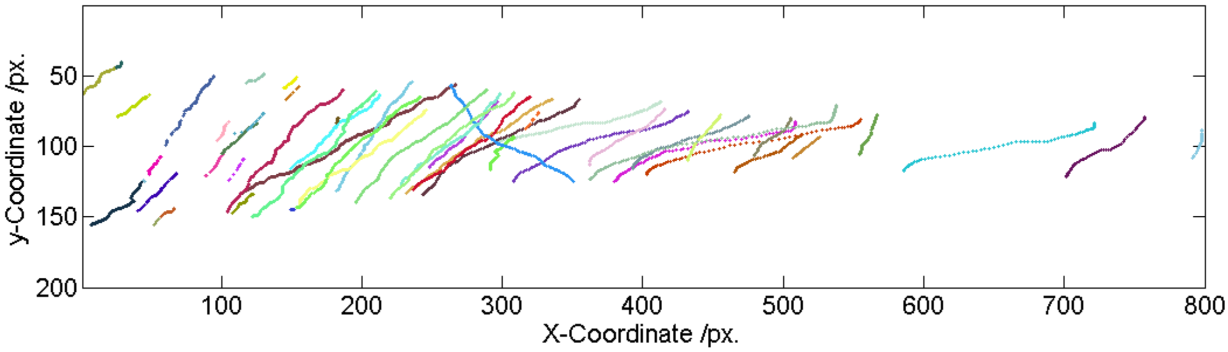

Figure 3 Typical tracking results of aerosol particles flowing upwards, while the laser beam comes from the left side. The measurement was carried out with the focused light of the laser diode.

3.4. Aerosol generation

Aerosols were generated using different techniques. Suspensions of white polystyrene latex particles (Ø 990 nm, 500 nm and 110 nm; distributor not specified) as well as sodium chloride solutions were sprayed using a cross-flow nebulizer. Previous to conducting the aerosol through the flow cell, it was run through a diffusion dryer. Suspensions of nanodiamonds were prepared by dispersion of a commercially available powder (Dynalene Inc., NB 98) in a 10:1 mixture of deionized water and isopropanol resulting in a mass concentration of 40 mgL^{-1}.

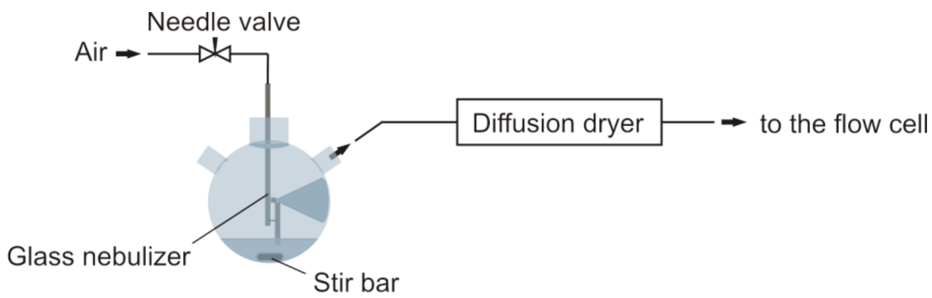

Figure 4 Scheme of the setup to generate dispersion aerosols.

4. Results

To revisit the topic of laser power dependency on the photophoretic velocity [12], nebulized polystyrene particle suspensions (Ø 500 nm) were measured by means of photophoretic velocimetry. The velocity distributions for 250 mW, 325 mW and 550 mW are shown in figure5. It can be noticed that the average photophoretic velocity increases with increasing laser power. Additionally, the velocity distribution seems to broaden towards higher velocities with increasing laser power. This effect is assumed to be connected to the overvaluation of slow particle movements which inherent to the evaluation system we are using currently.

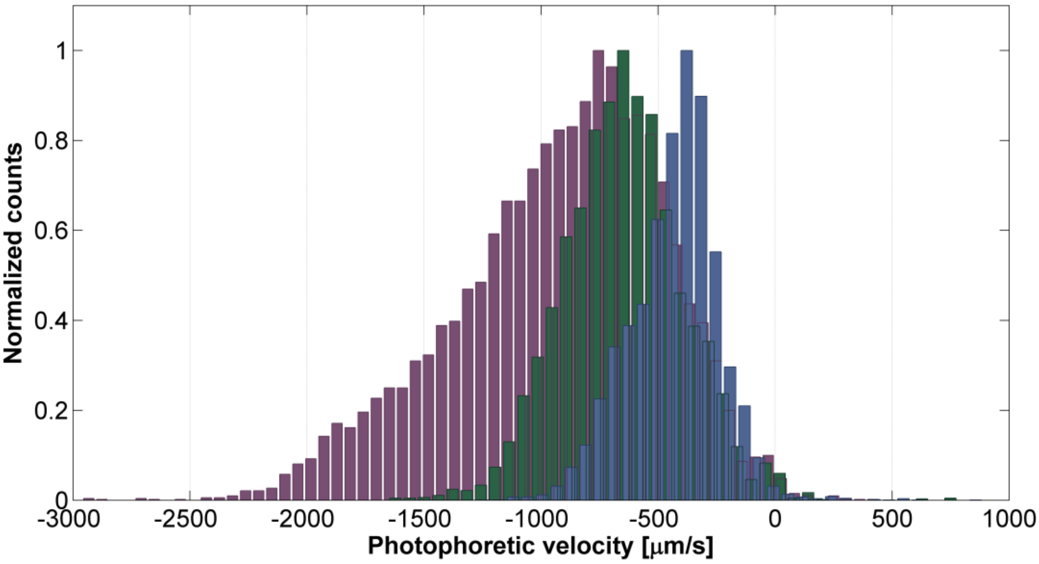

Figure 5 Change in the photophoretic velocity of polystyrene spheres (Ø 500 nm) due to different laser power. Velocity distributions at 250 mW (blue), 325 mW (green) and 550 mW (violet) are shown.

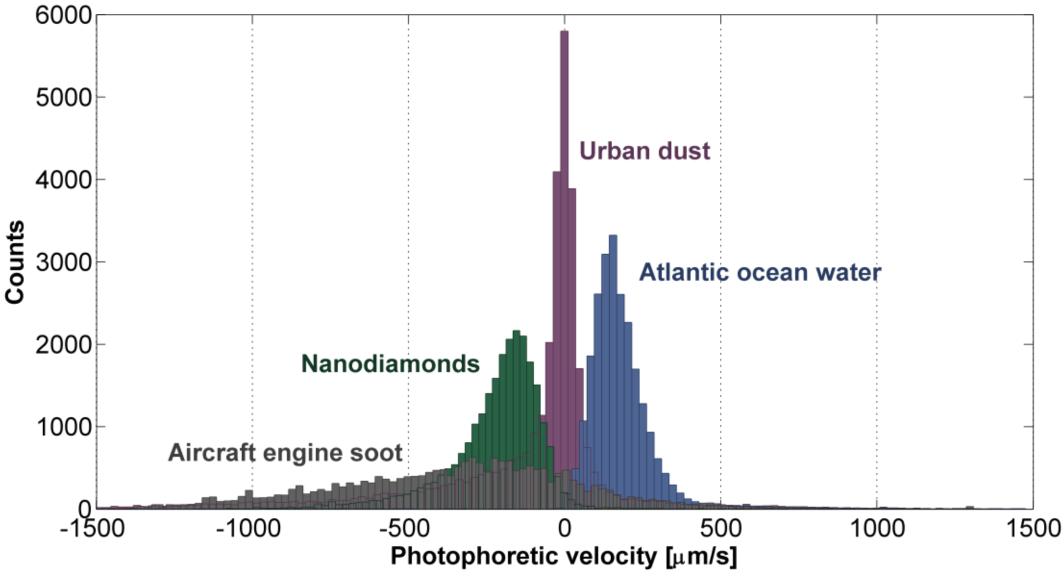

Figure 6 Thermo-photophoretic velocity distribution for different environmentally relevant aerosols.

The abilities of photophoretic velocimetry are demonstrated on the example presented in figure 6. Four significantly different, environmentally relevant particle systems were analyzed regarding their photophoretic properties. Characteristic response patterns of the different particle collectives when illuminated with an intense beam of light are the reason for that. For example, the particularly broad velocity distribution for aircraft engine soot is the result of irregular particle movements in various directions. The causative thermo-photophoretic force is by far not understood. In our investigation, an overall tendency of the movement of aircraft engine soot particles is found in direction towards the light source and therefore caused by a negative thermo-photophoretic force. Measurements with spark generated soot led to a similar result.

5. Summary and outlook

Considering the fact that only few experimental studies exist on the photophoretic motion of aerosol particles, characterization of different particle collectives is required. The attempt to use photophoretic velocimetry to derive a characteristic velocity for a certain particle species provides valuable information on the capacity of a possible separation method. Further studies should concern velocity measurements of strongly absorbing particles. In addition, photoacoustic measurements can provide information on the absorption properties of the investigated aerosol particles. Considerations should as well be made concerning the magnitude of the different photophoretic forces. Can the radiation pressure force really be neglected compared to the photothermophoretic force if we investigate weakly absorbing particles?

Our current activities are directed to the practical realization of a separation method based on photophoretic forces. Flow rates in the same range as the photophoretic velocities are difficult to realize with mechanical pump systems. New approaches therefore dispense with any mechanical flow system. The aerosol particles are charged in a first step; subsequently they can be manipulated in an adjustable electric field. Charged particles could then be guided through a focused laser beam by means of electrostatic forces.

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