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Opto-aerodynamic focusing of aerosol particles

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ABSTRACT
We describe a new method for focusing and concentrating a stream of moving micron-sized aerosol particles in air. The focusing and concentrating process is carried out by the combined drag force and optical force that is generated by a double-layer co-axial nozzle and a focused doughnut-shaped hollow laser beam, respectively. This method should supply a new tool for aerosol science and related research.

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1. Introduction

An aerosol is a suspension of particles in a gas or liquid. The particles may be solid, liquid, or a mixture of both. Ambient aerosol particles commonly have a wide size distribution, ranging from a few nanometers to a few hundred micrometers. They can be composed of a wide variety of materials and play an important role in climate and health. In aerosol science and its instrumentation, delivering aerosol particles into a small interrogation volume for particular measurements or characterization remains an essential research task. For instance, Raman scattering measurements often require gathering limited photons from single particles, and large numerical aperture (NA) collection optics are generally used for signal gathering. However, using large NA optics always results in a small effective range of focus, hence a small effective interrogation volume of particles. Therefore, particle focusing and concentrating is a desired approach to facilitate these aerosol studies. A specially-designed nozzle, based on aerodynamics, is often used to aerodynamically focus aerosol particles. Aerosol particles, because of their inertia, can take different trajectories depending upon their size, shape, density, and velocity, and the density and viscosity of the surrounding gases. Using a nozzle, particles in a particular size range can be concentrated in air (Geller et al. 2005; Romay et al. 2002; Wu et al. 1989), or be collected based on their inertial properties (Murphy et al. 2004; Riediker et al. 2000). Also, a nozzle may be used to increase the speed of particles so that they can be impacted upon a surface (Burwash et al. 2006; Riediker et al. 2000). Nozzle-based focusing technology is widely used to concentrate particles into a relatively small-diameter jet (Choi 2015; Dahneke et al. 1966; Deng et al. 2008; Frain et al. 2006; Fuerstenau et al. 1994; Hwang et al. 2015; Hutchins et al. 1991; Lee et al. 2003; Pan et al. 2003, 2009a, 2009b; Park et al. 2009; Schreiner et al. 1998; Tafreshi et al. 2002; Vidal-de-Miguel et al. 2011), increasing the sampling rate so the particles can be analyzed using laser induced breakdown spectroscopy (Murphy et al. 2004), laser-induced fluorescence (Pan et al. 2003, 2009a), particle deflection (Frain et al. 2006), mass spectrometry (Park et al. 2009), etc. The classical aerodynamic focusing methodologies are designed based on the particle inertia and the Stokes force of gas–particle interaction. The widely used nozzles are the aerodynamic lens (Fuerstenau et al. 1994; Schreiner et al. 1998; Liu et al., 1995a, b; Wang and McMurry, 2006; Vidal-de-Miguel et al. 2011), specially curved single nozzle, or sheath nozzle (Pan et al. 2009b). In some cases, electrostatic force is also used for focusing charged aerosol particles (Choi 2015; Hutchins et al. 1991).

Optical force is another means to manipulate aerosol particles. It has been used to trap or control the motion of nanometer- or micron-sized particles (Ashkin et al. 1986; Brzobohaty et al. 2013; Desyatnikov et al. 2009; Grier 2003; Pan et al. 2014; Redding and Pan 2015a; Rohatschek 1985; Wang et al. 2016). The applied optical forces are mainly radiative pressure force (scattering force and gradient force) and photophoretic force (limited to absorbing particles). A particular optical configuration can trap both transparent and absorbing airborne particles, enabling studies of single particles

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(Redding and Pan 2015a). In addition to particle trapping, the optical forces can also be used for manipulating the movement of particles in air.

Here, we demonstrate a new method that can focus and concentrate a stream of moving aerosol particles in air using the combined drag force and optical force.

2. Materials and methods

Figure 1 illustrates the experimental arrangement for observing the aerosol stream focused using the combination of drag force and optical force. The general operating principle is as follows. A double-layer co-axial nozzle operated in the counter flow mode in which particles are focused and concentrated into a small-diameter stream via the aerodynamic flow in the inner layer and then the particles are slowed down by a counter flow of air in the outer layer. As a result, the particles exiting the nozzle are formed into a narrow, slowly moving particle stream. In order to further focus and concentrate the particles into a localized jet which flows into an even smaller interrogation volume where the particles can be analyzed individually as they flow through one-by-one, a focused doughnut-shaped hollow laser beam illuminates the stream of particles along the axial orientation and optical forces further focus the particle stream into an even smaller-diameter aerosol stream.

The design details of the double-layer co-axial nozzle assembly for aerodynamic focusing have been reported previously (Pan et al. 2019b). The optical focusing is configured by a focused doughnut-shaped hollow laser beam (Civillaser, 532 nm CW laser, 3 W). The laser beam is formed into a conical region surrounded by a high-intensity-light at the surface of the cone. The hollow beam is produced by two axicon lenses (Del Mar Photonics, cone angle 175°), passes through a quartz window into an air-tight chamber, then is reflected by an elliptical mirror at 45°, propagates upward to a concave spherical mirror ($f = 19$ mm, diameter $= 25.4$ mm), and reflects downward from it to form a high-intensity-light cone. The high-intensity-light cone forms a focal spot right below the tip of the nozzle aligned to be co-axial with the double nozzles. The photograph in Figure 1 shows the cone shape formed by the doughnut-shaped hollow laser beam. Both the elliptical mirror and the concave spherical mirror inside the chamber have a center hole to let the aerosol or airflows pass through. The four sides of the chamber are covered by quartz windows to provide a large solid angle ($>0.3\pi$ for each side) for observation and signal collection.

As we previously showed, the double-layer co-axis nozzle can focus the super micron-size aerosol particles into a stream with a diameter less than 250 μm, where the airflow from both the inner and outer layer nozzles is

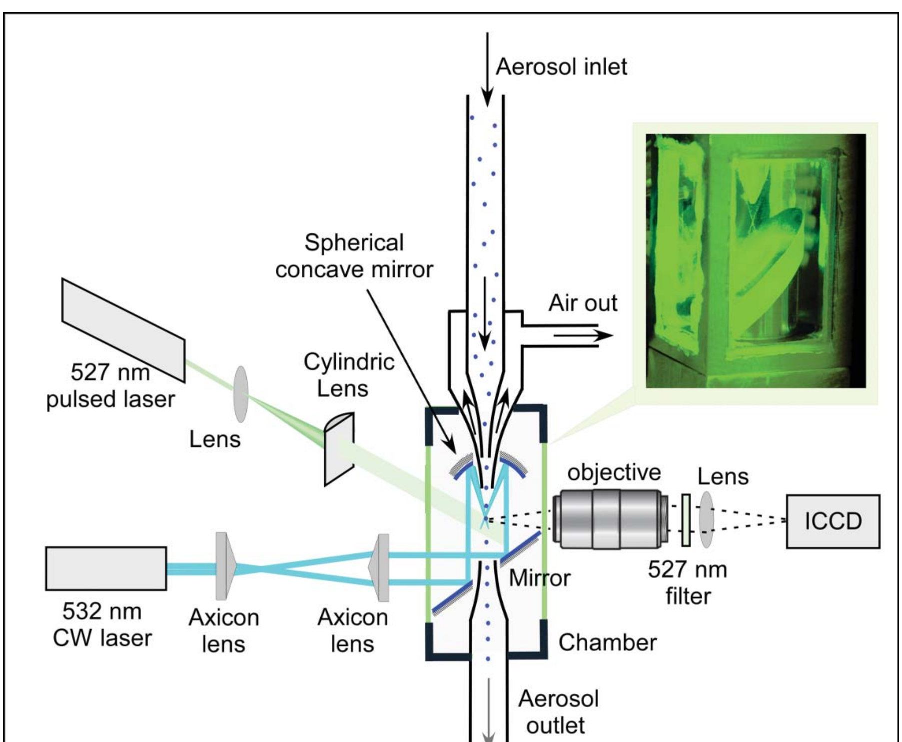

Figure 1. Schematic of the experimental setup for observing the aerosol stream focused using the combination of drag force and optical force.

moving in the same direction with the same speed (Pan et al. 2009b). Here, we run the airflow in the double-layer nozzle in a counter flow mode. We visualized the particle trajectories using a sheet of pulsed 527-nm laser (Photonics Industries, DC-150-263) beam operating at 1 KHz, 100 mW and recorded the light scattered from the particles using an Image-Intensified Charge-Coupled Device (ICCD, AndorIstar DK720-25-UV) camera. The scattering image from a 100-μm diameter fiber was used for size estimation. Tryptophan has no absorption at 532 nm while Bermuda grass smut spore has high absorption at 532 nm. We aerosolized the two materials to test the focusing effect of this configuration for either transparent or absorbing particles.

3. Results and discussions

The focusing ability of converging nozzles depends on the nozzle design, particle size, and the sample flow rate. Generally, particles with different aerodynamic diameters can only be focused into a shorter distance at different focal points. However, using the special designed gradually curved nozzle, we were able to focus super micron-size (2–10 μm) particles into a relatively small aerosol stream less than 300 μm diameter and keep the stream collimated for a distance longer than 5 mm, when the particles were drawn through the inner nozzle between 0.5–1.4 L/min. In order to optimize the focusing effect for the bioaerosol particles at 1–10 μm in diameter that we are interested in, various combinations of the sample and reverse flow rates were tested. In general, higher sample flow rate (close to 1.4 L/min) supplied better aerodynamic focusing by the inner nozzle, but resulted in large focusing disperse for different size particles, and easily generated turbulences as reversed flow applied, especially for small-size particles. Smaller particles (less than 2 μm) got bigger loss than bigger particles that were carried away by the reverse flow. The lower initial particles (smaller) are quicker in following the air flow than the larger particles, so both the speed acceleration (by the sample flow) and reduction (by the reverse flow) are all size dependent. We tried to compromise all the factors and tested out that it works well at a flow rate 0.6 L/min and 0.5 L/min for the inner and outer nozzle respectively. Therefore, the aerosol particles were drawn into the inner nozzle at0.6 L/min, where they were aerodynamically accelerated and focused by the drag force formed through the air flow in the inner layer of the nozzle, then rapidly decelerated by the gas flow moving in the reverse direction in the outer layer of the nozzle (0.5 L/min). The particles kept moving parallel to the axis by their inertia and continue moving forward by the remaining air flow of 0.1 L/min that exits from the bottom of the chamber. Consequently, the particles can be focused into a stream with a diameter less than 250 μm (2–10 μm particles). This center of the aerodynamically focused aerosol beam is aligned with the focusing laser cone for further optically concentrating.

For a spherical particle, the drag force F_d can be expressed as Davis et al. (2002)

$$F_d = -6\pi a \mu_a v \quad [1]$$

which is the well-known Stokes' Law, for a particle with radius a moving at a velocity v in air with a viscosity $\mu_a = 1.73 \times 10^{-5}$ Ns/m^2.

The radiation pressure force F_{rp}, attributable to the light illuminating of a plane wave, can be expressed as Shvedov et al. (2009):

$$F_{rp} = \frac{P}{c} = \pi a^2 I_0 \, / \, c, \quad [2]$$

where c is the speed of light and P is the laser power acting on the particle, it can then be expressed in terms of the particle radius a, the illumination laser intensity I_0.

The photophoretic force F_{pp}, can be expressed as Shvedov et al. (2009):

$$F_{pp} = -J_1 \frac{9\pi \mu_a^2 a I_0}{2\rho_a T\left(k_f + 2k_a\right)} \quad [3]$$

where μ_a is the air viscosity, $\rho_a = 1.29$ mg/cm^3 is the density of air, T is the temperature, k_f is the thermal conductivity of the particle, and $k_a = 0.0262$ W/mK is the thermal conductivity of air. In addition, the sign and magnitude of the photophoretic force depends on the coefficient J_1, which describes the asymmetric heating of the particle. For plane-wave illumination of an ideal black-body, $J_1 = -0.5$, which results in a positive photophoretic force, pushing the particle in the same direction as the light propagates. For a partially absorbing particle, the asymmetry factor can be expressed in terms of the optical field distribution within the particle, and it can be shown that for strongly absorbing spherical particles, the photophoretic force is always positive. However, spatially non-uniform particles are known to exhibit more complex photophoretic forces, including negative forces and forces which result in complex motions such as elliptical or helical orbits. These forces are explained by variations in the thermal accommodation coefficient across the particle surface. The thermal accommodation coefficient of a material describes the ability of the material to transfer heat to a surrounding air. If the accommodation coefficient varies across a particle surface, then even if it is uniformly heated, it will experience a force pushing the particle toward the side with the lower accommodation coefficient. In practice, it is very challenging to estimate the asymmetry coefficient J_1, and the accommodation coefficient for partially absorbing, non-spherical particles. As a result, the magnitude and even the direction of the photophoretic force can be difficult to predict.

Nonetheless, we used Equations (1)–(3) to roughly estimate the relative strength of the drag force, photophoretic force, and radiation pressure force acting on spherical particles. The estimation conditions are set as, illumination intensity at $I_0 = 3$ W/mm^2, the particle diameter at 5 μm, the absorption length at 0.1 μm, and the thermal conductivity at 0.159 W/mK, which is the value reported for leather in the range of thermal conductivities of biological materials ranging from wood to animal tissues. The asymmetry coefficient was set $J_1 = \max\{-0.5, -2\, a/3l_{\text{abs}}\}$. It turned out that the photophoretic force acting on particles with a diameter of 1–10 μm is about 2 orders of magnitude stronger than the radiation pressure force, or the gravitational force for weak and strong absorbing particles with thermal conductivity below ~1 W/mK. While the drag force is on the same order as the photophoretic force (Redding et al. 2015b). As the magnitude of the drag force is proportional to the particle moving velocity v, and we need comparable optical force to affect the particle moving trajectory, therefore, we reduce the particle moving speed to be around or below 0.1 m/s especially for the non-absorbing particles (or increase the laser power).

As the optical force is strongly dependent on the particle parameters including particle size, shape, refractive index (both the real and imaginary parts), density, thermal conductivity, laser power and beam profile, and the residence time of a particle within the laser beam, it is a big challenge to figure out what are the major factors in optical focusing. Hereby, we present the primary experimental result that showed optical force combined with drag force can effectively focus super micron aerosol particles.

Figure 2 shows scattering images of the aerosol stream focused by the drag force (the top row) or the combine of drag force and optical force (the middle and bottom rows). The left column in Figure 2 represents a cartoon drawing of the possible movements of a particle under the influence of the various forces; mainly, the drag force, gravitational force, optical radiation pressure force, and photophoretic force. The middle column in Figure 2 shows typical trajectories of a single particle movement. The images were recorded using the ICCD with a 300 ms accumulation time targeting an area about 1.2 mm in length and centered 0.6 mm above the focal point of the optical cone. During the 300 ms recording time interval, only one particle passed through this imaging area taking about 6 ms to cross the 1.2 mm (only 5–6 particle positions recorded by the illumination of 300 pulses). The right column in Figure 2 shows the trajectories from hundreds of particles accumulated during a 3 second time interval, these particles were imaged from the area centered at the focal point of the optical cone. From the top to the bottom row in Figure 2, they represent: (a) the particles were focused only with drag force; (b) the transparent particles were focused with the combination of drag force and optical force; and (c) the absorbing particles were focused with the combination of drag force and optical force.

Comparing the images between row (a) and (b) and (c) in Figure 2, it clearly indicates that the optical force does indeed further focus aerosol particle's stream into a smaller diameter. A closer look at the images finds that the particle streams in (b) and (c) are less than 100 μm in diameter, meaning the optical force further enhanced the particle focusing by a factor of 2.5. Correspondingly, the effect of particle density concentrating is enhanced by more than $(250/100)^2 \approx 6$-fold. This approach will greatly increase the sample rate and help deliver particles into even smaller interrogation volumes. Without the further focusing by the optical force, particles went straight as indicated on the upper row. For transparent particles, both the scattering force (pushing particles along the laser propagating direction) and gradient force (pulling particle toward the higher intensity area) bend the particle trajectories further toward the focal point of the laser beam along the high intensity wall of the cone to further focus the particle stream. For the absorbing particles, the photophoretic force confines the particles within the low laser intensity region and also pushes them toward the center axis of the particle stream. In order to make this idea work, a strong laser beam (3 W) was applied to supply a large enough focusing optical force. It must be noted that the particles should have a moving momentum to avoid particles being optically trapped, but sufficiently slow enough to allow the optical force to modify the particle's trajectory toward the beam center within the confinement of the optical cone. Unlike particle trapping in which a moving particle is deliberately slowed down close to a zero velocity to facilitate the particle capture and trapping in the desired trapping region (Desyatnikov et al. 2009; Redding and Pan 2015a), this particle focusing requires delicate control of the particle's downward motion in the focusing region so that optical force takes effect.

In conclusion, we demonstrate a new method that can focus and concentrate micron-sized aerosol particles into a small stream. The focusing process is carried out by the combination of a double co-axial nozzle and a focused

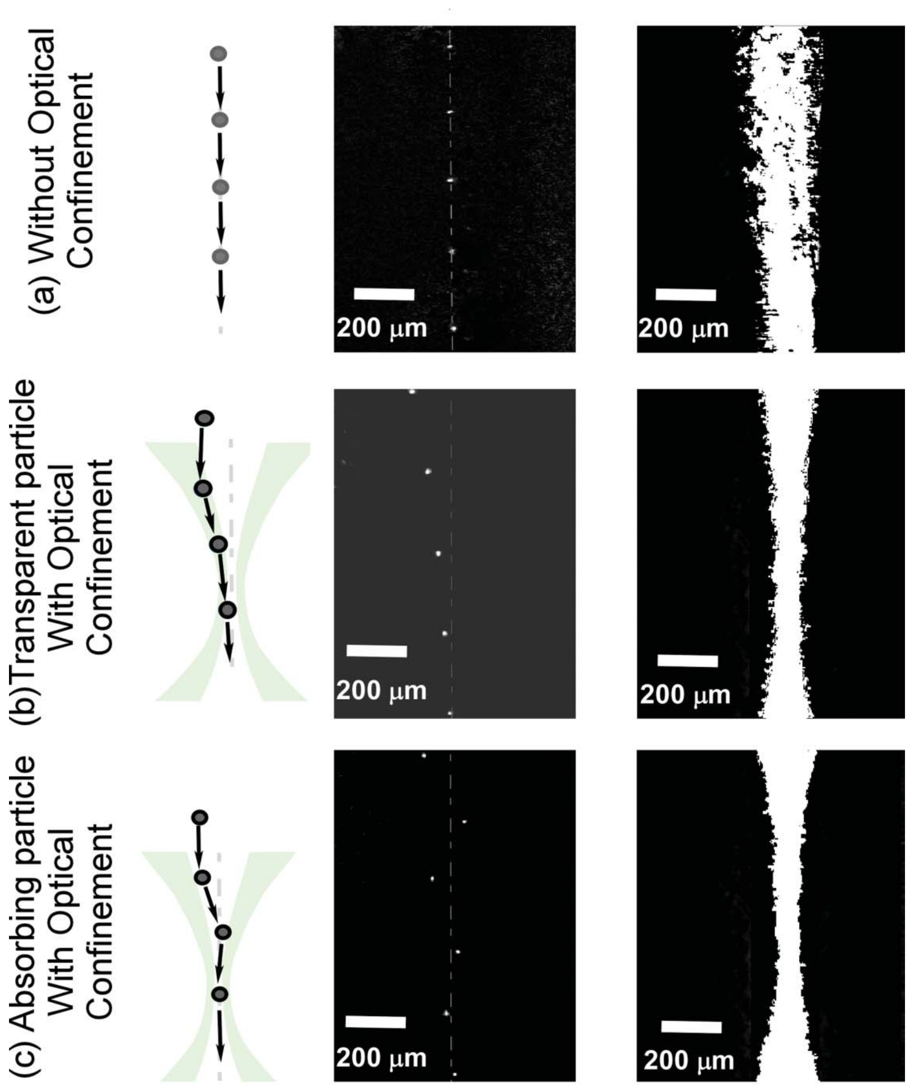

Figure 2. Illustration of particle focusing (left column) and images of the aerosol particles focused with the drag force only or the combine of drag force and optical force (middle and right column). (a) Particles are focused by drag force only; (b) transparent aerosol particles are focused with the combination of drag force and optical forces; and (c) absorbing aerosol particles are focused with the combination of drag force and optical forces (c).

annular hollow laser beam combining drag forces and optical forces. With the help of the optical forces, the particles can be concentrated six times more than using the aerodynamic nozzle alone. This method provides a new approach for focusing and concentrating aerosol particles that may be used in a variety of aerosol research and instrumentation applications.

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