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Multi-angle, dual wavelength scattering measurement chamber for the structural measurement of combustion generated particles

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ABSTRACT

Soot particles are formed through combustion processes and have fractal-like shapes. They influence our climate by absorption and scattering of the solar radiation directly, and indirectly by their influence on cloud (liquid or glaciated) formation and their deposition on snow and ice surfaces. An Instrument, which we would like to introduce in this paper was developed and built at the University of Applied Sciences Northwestern Switzerland to measure light scattering properties of combustion type aerosols. A closed chamber was designed where the scattered light can be detected at 7 fixed, predefined angles simultaneously. Two lasers at 405 nm and 852 nm serve as light sources, and with that almost simultaneous measurements at the two different wavelengths can be carried out. The instrument is calibrated with aerosol particles of known scattering properties and concentration. We used nebulized, size selected ammonium sulphate particles for this purpose.

The angular light scattering is influenced by the shape of the particles. In the case of fractals information on the fractal dimension and characteristic size can be derived from the angle dependence of the scattered light. In this paper we demonstrate and test the performance of this new light scattering instrument through the measurement of different soot or soot–containing aerosols. These examples include the measurement of artificial soot particles generated by two different soot generators (Cast soot and Palas soot generators) and different kind of combustion produced aerosols like: flaming ethanol, smouldering wood and mixed smouldering/flaming wood smokes.

1. Introduction

Carbonaceous particles are a major constituent of the atmospheric aerosols, these particles are either emitted as primary particles or formed from precursor gases in the atmosphere. Soot particles originate from different kind of combustion processes (automobile and aircraft emissions, fossil-fuel combustion and biomass burning) and often are of anthropogenic origin. Their amount can reach as much as 10–50% of the total particulate matter. The optical and other physical properties of black carbon (BC) and soot are of significant importance as these species have been recognized as the greatest anthropogenic aerosol contributors to global warming (Stocker et al., 2013). They cause radiative forcing by direct solar absorption, their influence on cloud (liquid or glaciated) formation and their deposition on snow and ice surfaces.

Soot particles produced in combustion processes have fractal-like shape. These fractal aggregates have a self-similar structure and are composed of spherical primary particles (monomers) of elemental carbon. Soot particles and/or their internal mixture with other inorganic or organic particles define the combustion aerosols. Due to the fractal-like shape the number of primary particles

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(N_{mon}, proportional to the aggregate's mass) scales as a power law with the characteristic size of the aggregate (Sorensen, 2001):

$$N_{\mathrm{mon}} = k_0 (R_g/a)^{D_f} \tag{1}$$

where k_0 is a scaling prefactor, a is the radius of the monomer, D_f is the fractal dimension and R_g is the characteristic size of the aggregate: the radius of gyration. A fractal dimension of 3 means a particle shape where the 3 dimensional space is completely filled, whereas a fractal dimension of 2 refers to a particle which fills the space as much as a plane does. The radius of gyration can be defined as follows:

$$R_{\mathrm{gyr}}^2 = \frac{1}{N_{\mathrm{mon}}} \sum_{i=1}^{N_{\mathrm{mon}}} |\vec{r_i} - \vec{r_c}|^2 \tag{2}$$

where $\vec{r_i}$ is the position of the ith primary particle and $\vec{r_c}$ is the position of the fractal's center of gravity.

One of the most popular methods for soot particle morphology determination is the use of elastic light scattering (e.g. Chang, Lin, & Biswas, 1995; Hull, Shepherd, & Hunt, 2004; Keller, Loepfe, Nebiker, Pleisch, & Burtscher, 2006; Köylü & Faeth, 1994; Oltmann, Reimann, & Will, 2010). Next to the size and chemical composition of the particles, the structure of the soot particles highly influences their scattering behaviour as well which can be used to gain information on the aggregates size, expressed as the radius of gyration, and on the fractal dimension. An approximate theory describing the scattering of fractal aggregates is presented in the next section. Oh and Sorensen (1997b) showed that combining elastic light scattering and extinction measurements enables the determination of all important soot morphological parameters including the number of primary particles per aggregate as well.

Most of the instruments measuring the angular elastic light scattering can be categorised into two types: The first category of the instruments using a scanning goniometer. This method can have a very good angle resolution, however the single detector needs substantial time for a full scan and, therefore, such an instrument is only applicable if the aerosol source stays stable during the scan. The other type uses multiple detectors and therefore can have very good time resolution, but the number of detectors and with this the angular resolution is limited. Chang et al. (1995) have built an open measurement system which can reach high time resolution with measuring the light scattering at a single wavelength and 5 different scattering angles simultaneously. They performed measurements on non-absorbing, spherical particles and invented an inversion algorithm in order to derive the particles number size distribution and refractive index.

Oltmann et al. (2010) have developed a special instrument which uses an ellipsoidal mirror to collect the scattered light over a wide angular range, which is then detected by a CCD camera. With this tool both high time and angular resolution can be achieved, however the sensitivity of the instrument is limited.

2. Rayleigh–Debye–Gans theory for fractal aggregates

The Maxwell equations cannot be solved analytically for fractal aggregates, however very precise numerical solutions exist like the T-matrix or the DDA (dipole–dipole approximation) methods (e.g. Skorupski et al., 2013; Smith & Grainger, 2014). These numerical calculations can provide us the differential cross section of soot particles but require the knowledge of the exact geometry (location of all primary particles in space) of the aggregate. The Rayleigh–Debye–Gans theory for fractal aggregates (RDG-FA) is an approximate method to describe the scattering of aggregates, able to deliver differential cross sections averaged over all random orientations, but still captures the fractal-like behaviour and has a reasonable agreement with the more precise methods (Ma, 2011).

This theory assumes that every primary particle see the same incident light wave and with this ignores multiple scattering. The primary particles are assumed to be Rayleigh scatterers and therefore the differential scattering cross section of a monomer for vertically polarized light can be written as:

$$\frac{\mathrm{d}\sigma_{\mathrm{mon}}(\Theta)}{\mathrm{d}\Omega} = k^4 a^6 F(m) \tag{3}$$

where Θ is the scattering angle, $k = 2\pi/\lambda$ is the magnitude of the wave vector, a is the diameter of the primary particle and:

$$F(m) = \left|\frac{m^2 - 1}{m^2 + 2}\right|^2 \tag{4}$$

with m being the complex refractive index of the monomer. The differential cross section of the aggregate according to the RDG-FA theory can be written as:

$$\frac{\mathrm{d}\sigma_{\mathrm{agg}}(\Theta)}{\mathrm{d}\Omega} = N^2 \frac{\mathrm{d}\sigma_{\mathrm{mon}}(\Theta)}{\mathrm{d}\Omega} S(q) \tag{5}$$

q is the amplitude of the scattering wave vector $q = 4\pi/\lambda \cdot \sin(\Theta/2)$ and $S(q)$ is the so-called structure factor of the soot aggregate which includes the structural information of the fractal. We used the following structure factor definition from Dobbins and Megaridis (1991):