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Diesel soot mass calculation in real-time with a differential mobility spectrometer

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Abstract

This paper presents a methodology to allow a real-time particle size spectrometer to produce a mass concentration output by calculation from its electrical mobility response. As part of this, a Bayesian statistical algorithm for parametrising spectral data from the Cambustion DMS500 in terms of a number of lognormal functions is outlined, allowing the nucleation and accumulation modes of a Diesel aerosol to be treated separately and also to reduce mass calculation noise and improve spectral resolution. Previous literature is combined with new experimental results to develop a size:mass power-law relationship for this instrument. The effective density as a function of size for this instrument is found to be closer to that for water droplets than equivalent relationships for DMA/SMPS measurements in the literature, therefore making DMS500 mass calculation less susceptible to error from liquid adsorbed on agglomerates. The technique is validated with two Diesel engines against the gravimetric methods of filter paper and Diesel particulate filter (DPF) weighings. Good agreement is achieved over a variety of engine conditions, with the mean and standard deviation of the percentage difference of the calculated mass concentration from DPF weighings being $-12.1 \pm 11.4\%$ and from filter paper weighings being $-15.1 \pm 18.0\%$. The calculated mass concentrations are systematically below those of the gravimetric methods.

Keywords: Diesel; DMS; Particulate mass; Lognormal; Fractal

1. Introduction

1.1. Background

Particles are generated from a variety of sources, both natural and anthropogenic; and the emissions from internal combustion engines are one of the major global sources of particulate matter (PM). PM is known to contribute to climate change, reduce visibility, and affect human health. To reduce the environmental impact of internal combustion engines, governments regulate the mass of PM emitted by vehicles. Current particulate air quality regulations and diesel PM regulations from the US Environmental Protection Agency (EPA) and the EU are typically based on a gravimetric

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filter method. However, it is well documented that gravimetric filter measurements can be affected by a variety of artefacts. These artefacts include adsorption of vapour onto the filter, volatilisation of semi-volatile compounds from filtered particles, and an array of chemical reactions between filtered particles, the gas, and filter substrate (Hinds, 1999; Patashnick, Rupprecht, Ambs, & Meyer, 2001; Zhang & McMurry, 1987). Another problem with gravimetric filter measurements is that long sampling times are required for adequate measurement resolution and as future emission standards tighten, gravimetric methods may not be feasible for such small amounts of accumulated mass. Also, due to the nature of filter measurements, transient mass concentration measurements cannot be made, making the task of reducing particulate emissions by engine calibration difficult.

Besides filter-based methods, various instruments and methods can be used to measure mass concentrations or mass distributions. Firstly, a tapered element oscillating microbalance (TEOM) can be used to measure real-time mass concentrations (Chan & He, 1999; Morawska, Johnson, Ristovski, & Agranovski, 1999; Saito & Shinozaki, 1990). However, like filter measurements, the TEOM is affected by sampling artefacts and cannot provide size-resolved data. Secondly, light scattering measurements can be used to estimate particle mass concentration (Sioutas, Kim, Chang, Terrell, & Gong, 2000; Thomas & Gebhart, 1994); however, the relationship between light scattering and particle mass is empirical and light scattering methods will not be accurate in transient measurements if the size-distribution or refractive index of the aerosol changes during a test. Thirdly, cascade impactors such the micro-orifice uniform deposit impactor (MOUDI; Marple, Rubow, & Behm, 1991) can be used to measure mass distributions by a gravimetric analysis of particle mass collected on each impactor substrate. However, this method is time consuming and the MOUDI suffers from the same sensitivity issues as filter measurements. Fourthly, acoustic resonance from laser heated soot can be used to measure mass (photoacoustic soot sensor, Petzold & Niessner, 1996). This method promises a direct mass measurement for most particle sizes but does not provide size information.

Particle sizing instruments can also be used to measure mass distributions if the particle density is known. Particle sizing instruments generally measure the aerodynamic equivalent diameter of particles, such as the electrical low pressure impactor (ELPI; Ahlvik, Ntziachristos, Keskinen, & Virtanen, 1998), the mobility equivalent diameter, such as the scanning mobility particle sizer (SMPS; Wang & Flagan, 1990), or the electrical mobility equivalent diameter (see Section 4.1 for a definition) such as the differential mobility spectrometer (DMS; Reavell & Hands, 2002), or engine exhaust particle sizer (EEPS; Johnson, Caldow, Pocher, Mirme, & Kittelson, 2004). A hybrid inertial and electrical size classification instrument, the Dekati mass monitor (DMM) is also produced (Lehmann, Niemelä, & Mohr, 2004).

In early studies with ELPI and SMPS mass measurements, a uniform particle density was assumed. However, it was found that for fractal-like structures such as diesel particulates, assumptions of uniform particle density resulted in large uncertainties in mass measurement (Andrews, Clark, Rojas, Sale, & Gregory, 2001; Peters, Chein, & Lundgren, 1993; Sioutas, Abt, Wolfson, & Koutrakis, 1999). The uncertainty in this method can be greatly reduced by measuring or assuming a size-dependent particle density. The density of particles can be measured by using a differential mobility analyser (DMA) and an ELPI and using the relationship between mobility and aerodynamic equivalent diameters (see Eq. (11)), as shown by Maricq and Xu (2004). The size-dependent density of particles can also be found using a DMA and a centrifugal particle mass analyser, such as the APM (aerosol particle mass, Ehara, Hagwood, & Coakley, 1996) or Couette centrifugal particle mass analyser (CPMA) (Olfert & Collings, 2005), which measures the mass of particles. From these studies size-dependent density functions for diesel particles have been obtained which can be used to calculate the mass distribution and mass concentration of particles measured with mobility sizing instruments.

These functions are usually represented in form $M \propto D^i$ for mass M, particle diameter D, and fitted index, $i \in \mathbb{R}^+$. When $i = 3$ this expression reduces to the trivial case for spherical particles, when $i \notin \mathbb{N}$ the expression represents a fractal structure (e.g. Mandelbrot, 1983). In this paper, a method is described which builds upon these principles to determine mass concentration in real-time from spectral data produced by the Cambustion DMS500 real-time particle sizing instrument. As part of this method, a new software algorithm to parametrise spectral data in terms of multiple lognormal modes is also presented. Validation data from Diesel engines is given to compare with gravimetric methods.

1.2. Overview of method

The method of mass calculation used in this paper has three distinct parts: (1) the particle spectrometer and sampling system, (2) the novel lognormal fitting software, and (3) the derivation of a specific mass weighting expression