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The effective density and fractal dimension of particles emitted from a light-duty diesel vehicle with a diesel oxidation catalyst

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Abstract

A differential mobility analyzer (DMA) and a Couette centrifugal particle mass analyzer (Couette CPMA) were used to measure the effective density and fractal dimension of particles emitted from a light-duty diesel vehicle fitted with a diesel oxidation catalyst (DOC). It was found that at high engine loads, the DOC increased in temperature, sulphate levels in the particulate matter increased, and a transient nucleation mode was observed. The increase in sulphate levels resulted in a drastic increase in the effective density and fractal dimension of the particles. At low engine loads (8–15%), sulphate levels were much lower, no nucleation mode was present and the fractal dimension varied from 2.22 to 2.48, which is in good agreement with previous studies. At 40% load, sulphate levels were much higher and the fractal dimension was 2.76.

Keywords: Diesel soot; Effective density; Fractal dimension; Centrifugal particle mass analyzer

1. Introduction

Particles in the environment are known to affect climate change and reduce visibility. There is also evidence that suggests that particles can have a negative effect on human health. Particles can be generated by a variety of sources, both natural and anthropogenic; however, particulate emissions from diesel vehicles are a major source of ultra-fine particles in the atmosphere. Particles and aerosols can be characterized by many different properties such number concentration, mass concentration, particle size, mass, density, volume, dynamic shape factor, fractal dimension, etc. By measuring the mobility size and mass of particles, other important particle properties can be determined such as effective density and fractal dimension.

The particle effective density is an important parameter because it determines particle transport properties, it can be used to convert size distributions to mass distributions, and it provides a relationship between mobility and aerodynamics sizes. The morphology of an agglomerate is characterized by its fractal dimension. The morphology affects the behaviour

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of particles in the atmosphere (Friedlander, 2000) and possibly impacts upon the effect particles have on human health (Bérubé et al., 1999).

There have been several studies that have measured the effective density and/or fractal dimension of diesel exhaust particles. These studies have generally used a differential mobility analyzer (DMA) and an electrical low-pressure impactor (ELPI) to determine the particle's mobility and aerodynamic diameters, respectively. In general, two different experimental methods have been used to find the effective density and fractal dimension of diesel particles using a DMA and ELPI. The first method (used by Ahlvik et al., 1998; Maricq et al., 2000 to measure effective density, and Maricq and Xu, 2004 to measure effective density and fractal dimension) involves using a DMA and an ELPI in series, where particles are first classified by mobility size with the DMA and then the aerodynamic size is determined with the ELPI. The effective densities of diesel particles measured by Ahlvik et al. (1998) and Maricq et al. (2000) were much higher than later studies found. It was later suggested by Maricq and Xu (2004) that the effective densities were overestimated due to soot accumulation on the impactor plates. A similar technique was used by Skillas et al. (1998) and Van Gulijk et al. (2004) to measure the fractal dimension of diesel particles. A second technique, used by Virtanen et al. (2002), and Virtanen et al. (2004), involves using a scanning mobility particle sizer (SMPS) in parallel with an ELPI, where the difference between the two corresponding size distributions is minimized by fitting the data with a size-dependent effective density function (also see Virtanen et al., 2004 and Ristimäki et al., 2002 for a detailed description of this method). The effective density of diesel particles could also be determined by using other systems such as: a DMA, optical particle counter, and aerodynamic particle sizer (Hand and Kreidenweis, 2002), an SMPS and aerosol mass spectrometer (Katrib et al., 2004), and a DMA and a single particle laser ablation time-of-flight mass spectrometer (Zelenyuk et al., 2005). However, experiments using these other systems have not been conducted with diesel particles.

Park, Cao, Kittelson, and McMurry (2003) have shown that the effective density and fractal dimension of diesel particles can also be measured using a DMA and an aerosol particle mass analyzer (APM; Ehara et al., 1996) to measure particle mobility diameter and particle mass, respectively. Recently, a new particle mass classifier has been developed called the Couette centrifugal particle mass analyzer (Couette CPMA), which is similar to the APM, but with an improved transfer function (see Olfert and Collings, 2005 and Olfert et al., 2006). In this study, a DMA and Couette CPMA will be used to measure the effective density and fractal dimension of diesel particles produced from a light-duty diesel vehicle with a diesel oxidation catalyst (DOC).

Previous studies by Maricq et al. (2002); Vogt et al. (2003), and Giechaskiel et al. (2005) have shown that the size distribution of diesel soot can change with fuel sulphur levels and DOC temperature. However, previous studies have not shown how increased levels of sulphuric acid in the exhaust can drastically change the effective density and fractal dimension of diesel soot. The purpose of this study is to show how the Couette CPMA can be used to measure the effective density and fractal dimension of non-spherical particles (i.e. diesel particles), and show how increased levels of sulphate in the exhaust, due to increased DOC temperatures, can greatly increase the effective density and fractal dimension of diesel particles.

2. Experimental set-up

2.1. Sampling diesel exhaust

A schematic of the experimental set-up is seen in Fig. 1. The light-duty diesel vehicle used in this study was a 2002 Peugeot 406 (2.2 L common rail diesel) certified to Euro III standards. The vehicle was fitted with a DOC. The diesel fuel used had < 50 ppm of sulphur, although the exact concentration of sulphur was not measured, the same tank of fuel was used for all the tests. The vehicle was operated on a chassis dynamometer at six different engine speed/load settings. For each of the tests the vehicle was warmed-up by running the vehicle at the required setting for approximately 10 min before the test was started. Each test ran for approximately 1 h. The exhaust from the vehicle was diluted immediately (with air at 25 °C and 45% relative humidity) as it entered the constant volume sampling (CVS) dilution tunnel. The particles in the dilution tunnel were measured with three different systems. Firstly, a sample of the diesel soot was taken with filter paper (Pallflex Emfab filters; TX40HI20WW) for sulphate analysis. For each of the tests two sequential filter measurements were taken with each filter collecting samples for approximately 20 min. The sulphate was extracted from the filter and analysed with ion chromatography using a similar method as described by SAE J1936 (1989). Secondly, the size distribution was continuously measured with a differential mobility spectrometer