A novel diesel soot particle generator for calibration purposes

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Introduction

Particulate emissions are a major hazard for human health and the environment. Especially nanometre sized particles are found detrimental to health as they may enter the alveolar region of the lungs. In urban areas the main source of nanoparticles is combustion generated particles from vehicles, especially diesel fuelled vehicles. In order to reliably measure these particles, the measuring instruments needs to be calibrated using particles that closely resemble diesel soot particles. The physical and chemical nature of the particles, however, depends on the vehicle engine design, fuel, lubricant, operating conditions to name a few. Real diesel engines are not suitable for generating particles in calibrations where a reproducible and well defined particle output is needed. Moreover, the generated particle size distribution and number concentration are only poorly or not at all controllable in real engines.

Due to the apparent shortcomings of using real diesel engines to generate calibration soot particles, alternative methods have been developed. Helsper *et al.* [1] introduced a graphite spark generator in which the aerosol is produced by spark discharge between two graphite electrodes. Jing [2] developed the combustion aerosol standard (CAST) which uses a propane diffusion flame for generating soot like particles. An apparent drawback of these methods is that the material from which the particles are generated differs from diesel fuel. Thus, the generated particles may only be considered an approximation of real diesel soot particles.

In this study a novel diesel fuelled soot generator was applied to generate diesel like soot particles for calibrating real-time measuring instruments. In a recent study [3] the generated soot particles were found to represent "real" exhaust particulates with respect to size, morphology and material. The application of the soot generator for calibration purposes is demonstrated by calibrating real-time measuring instruments Dekati Mass Monitor (DMM) and Scanning Mobility Particle Sizer (SMPS) using gravimetric filter sampling as a reference.

Soot generator

The soot generator is made from a commercial diesel fuelled vehicle heater (Webasto Air top 2000 ST). The heater is modified in such a way that the fuel pump and combustion air are controllable. This allows one to change the air to fuel ratio, which determines the generated soot particle size distribution. The flow which normally acts as the heating flow in the intended use of the heater is used to cool the combustion chamber. The performance characteristics are summarized in table 1. A more detailed description of the soot generator is given in [3].

The generated soot particle size distribution was found to obey a log-normal distribution with geometric mean diameter (GMD) and geometric standard deviation (GSD) values similar to real diesel exhaust [3]. Transmission electron microscopy images confirmed that the generated particles are fractal-like agglomerates consisting of primary spheres of sizes 20 - 30 nm [3]. The effective density of generated particles was derived from concurrent Electrical Low Pressure Impactor (ELPI) and SMPS measurements using the method of Ristimäki *et. al.* [4]. It was found that the effective density gets smaller with larger particle sizes (see table 1). This is also the case for fractal-like soot particles from real diesel exhaust. Therefore, the soot generator is considered well

suited for generating diesel like soot particles for calibrating instruments measuring diesel exhaust particles.

Particle size range (GMD)	27 – 164 nm (adjustable)
Width of distribution (GSD)	2.5 – 1.78 (for GMD 27 – 164 nm)
Effective density of particles	$1.16 - 0.51 \text{ g/cm}^3 \text{ (for GMD 36 - 155 nm)}$
Particle output concentration	up to 10^8 particles/cm ³
Stability (relative std.)	4.0% (during a 900 s test run)
Exhaust gas flow rate	35 – 50 L/min

Table 1. Performance characteristics of the soot generator

Experimental setup

A part of the generator output was taken as the calibration aerosol. The aerosol was diluted with dry compressed air so that there was no condensation of water at ambient laboratory temperature. Thereafter, the aerosol was allowed to stabilize in the mixing chamber (volume 6.6 l). A thermodenuder (Dekati® Thermodenuder), operating at 270°C, was used to remove semivolatiles, which would otherwise introduce inaccuracies in filter sampling due to the filter artefact [3]. The aerosol was slightly heated so that the aerosol temperature requirement ($47^{\circ}C \pm 5^{\circ}C$) for gravimetric filter sampling was fulfilled. Finally, an ejector diluter was used to ensure homogeneity of the generated calibration aerosol.

The calibration aerosol output was split between the real-time measuring instruments (SMPS and DMM) and the filter sample. The SMPS (TSI 3071, TSI CPC 3775) was operated at 0.6/6 (polydisperse/sheet) L/min flow rates and the DMM and filter sample flow rate was 8.6 L/min and 10 L/min, respectively. Teflon-bonded glass fibre filters (Pallflex Emfab) with a diameter of 47 mm were used, since they are specifically designed for diesel exhaust measurements. A filter bypass was used to ensure that soot was only sampled during the measurement period.

Measurements

The calibration was performed by comparing the soot mass sampled on the filters with the response of the real-time measuring instruments (DMM and SMPS). The soot mass sampled on the filters was determined by pre- and post weighing the filters using a microbalance (Sartorius CCE6) with 0.1 μ g resolution. The effect of background contamination on filter mass load was measured by bypassing the soot generator. The background contamination was typically 10 μ g for a 20 min measurement. The result of gravimetric sampling was corrected for this effect.

Measurements were performed at three different soot particle concentrations 128, 418 and 613 μ g/m³ (gravimetric measurement) using a sampling time of 20 min. The GMD of the calibration aerosol was 60 – 90 nm during the calibration. A particle density of 1 g/cm³ was used for calculating the SMPS results.

Results

Real-time measuring instruments SMPS and DMM had a linear response to increasing soot particle concentration (figure 1). The slope of the linear fit was 1.64 and 1.9 for the SMPS and DMM respectively. The results for the SMPS agree well with the results reported by Sioutas *et al.* [5], stating that SMPS overestimates mass concentration by a factor of 1.4 - 1.9. The slope of the DMM is somewhat higher than the result reported by Mamakos *et al.* [6] stating that DMM overestimates mass concentration by a factor of 1.42 ± 0.34 . The small discrepancy is probably due to the fact that the calibration aerosol is free of volatiles which would normally contribute to the filter mass through adsorption from gas phase (positive filter artefact).



Figure 1. Comparison of gravimetric sampling and real-time measuring instruments. The thin line represents the line of equality (y = x).

Conclusions

A novel soot generator was used to generate diesel soot particles for calibration purposes. The application of the soot generator was demonstrated by calibrating real-time measuring instruments SMPS and DMM using gravimetric filter sampling as a reference. The results of the real-time measuring instruments show good linear agreement with results of gravimetric filter sampling. The slope of the linear fit was found to be 1.64 and 1.90 for the SMPS and DMM respectively. The soot generator in conjunction with the aerosol conditioning of the experimental setup presented here, show high potential for providing a diesel like soot particle output which can be used for instrument calibration and testing purposes.

Acknowledgements

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INTRODUCTION

• Particulate emissions are a major hazard for human health and the environment. Especially nanometre sized particles are found detrimental to health

• In urban areas the main source of nanoparticles is combustion generated particles from vehicles, especially diesel fuelled vehicles.

• In order to reliably measure these particles, the measuring instruments needs to be calibrated using particles that closely resemble diesel soot particles.

• Real diesel engines are not suitable for generating particles in calibrations where a reproducible and well defined particle output is needed.

• In current commercial particle generators for calibration the material of the particles differ from diesel soot particles.

• In this study a novel diesel fuelled soot generator was applied to calibrate real-time measuring instruments.

SOOT GENERATOR

• The soot generator is made from a commercial diesel fuelled vehicle heater (Webasto Air top 2000 ST).

• The heater is modified in such a way that the air to fuel ratio is controllable allowing control over generated particle size (table 1).

• In a recent study [1] the generated soot particles were found to represent "real" exhaust particulates with respect to size, morphology and material.

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Particle size range (GMD)	27 – 164 nm (adjustable)
Width of distribution (GSD)	2.5 – 1.78 (for GMD 27 – 164 nm)
Effective density of particles	$1.16 - 0.51 \text{ g/cm}^3 \text{ (for GMD 36 - 155 nm)}$
Particle output concentration	up to 10 ⁸ particles/cm ³
Stability (relative std.)	4.0% (during a 900 s test run)
Exhaust gas flow rate	35 – 50 L/min

GMD = geometric mean diameter, GSD = geometric standard deviation

CALIBRATION

• The soot generator was applied for the first time to calibrate real-time measuring instruments Dekati Mass Monitor (DMM) and Scanning Mobility Particle Sizer (SMPS) using gravimetric sampling as a reference.

• The calibration aerosol was split between the realtime measuring instruments and the filter sample.

 \bullet Measurements were performed at three different soot particle concentrations 128, 418 and 613 $\mu g/m^3$ (gravimetric measurement) using a sampling time of 20 min.

• The GMD of the calibration aerosol was 60 - 90 nm during the calibration.

 \bullet A particle density of 1 g/cm 3 was used for calculating the SMPS results.

• A thermodenuder was used to remove semivolatiles, which would otherwise introduce inaccuracies in filter sampling due to the filter artefact [1].

RESULTS

Results of the real-time measuring instruments show good linear agreement with the gravimetric method (figure 1).





CONCLUSIONS

The soot generator was successfully applied for calibrating real-time measuring instruments DMM and SMPS using gravimetric filter sampling as a reference.
The presented soot generator was found a unique source of diesel soot particles, which can be used for instrument calibration and testing.

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