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Comparison of three nanoparticle sizing instruments: The influence of particle morphology

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HIGHLIGHTS

- We examine particle sizer equivalency with particles of contrasting characteristics.
- Some instruments at times significantly overestimate concentrations.
- Overestimation occurred in the presence of agglomerates.
- The overestimation affects near-road measurement of vehicle plumes.

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ABSTRACT

The TSI Fast Mobility Particle Sizer (FMPS), Engine Exhaust Particle Sizer (EEPS), and Scanning Mobility Particle Sizer (SMPS) provide size distributions for 6–560 nm particles. The aim of this study was to perform comprehensive equivalence testing of these three particle sizing instruments with particles of contrasting chemical and physical characteristics (urban ambient, diesel exhaust, and laboratory-generated particulate). It was observed that the EEPS and FMPS measurements agreed to within 15% thus concluding that data from these instruments may be considered equivalent. Parallel measurements with the SMPS showed that when measuring diesel exhaust particulate during ISO8178 Mode 9 operation there is significant overestimation of particle concentrations by both the EEPS and the FMPS in the 20–120 nm size range (25–38% overestimation). This overestimation also occurred for near-road measurement of heavy emitter vehicle plumes in ambient samples (up to 75% overestimation). Laboratory-generated soot agglomerate particles, whose shape was verified by transmission electron microscopy, were also tested. The agglomerate nature of diesel soot particulate was the dominant cause of the overestimation; parallel measurements with an FMPS and an Ultrafine Condensation Particle Counter of the laboratory-generated soot particulate showed overestimation by up to a factor of three.

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1. Introduction

Diesel fuels are a significant energy source in Canada, accounting for 97% of the fuel consumed by heavy-duty vehicles, and 6% of that consumed by the total vehicle fleet (Office of Energy Efficiency, 2009). Tailpipe emissions from these diesel vehicles release numerous pollutants into the airshed, of which the most well-known is diesel particulate matter (PM). Concentrations of PM can be classified into three particle size modes: the ultrafine mode (particle diameter, $D_p < 100$ nm); the accumulation mode (100 nm $< D_p <$ 2500 nm); and the coarse mode (2.5 μm $< D_p <$ 10 μm). The smallest size fraction of these categories, the ultrafine mode, is only attributable to 1–20% of the PM mass but comprises more than 90% of the PM number concentration (Kittelson, 1998). This is significant given ultrafine mode particles deposit with the greatest efficiency in the nasal cavity and alveolar region, where they are capable of inducing oxidative stress (Alföldy et al., 2009; Creutzenberg, 2012; ICRP, 1994; Mudway et al., 2004; Xia et al., 2004).

The current study was conducted as part of the EMITTED project (Exhaust Measurement and Inhalation Toxicology Testing for Emerging Diesel Fuels) where the aim is to evaluate emission reductions using emerging control technologies and diesel fuel types. Increasingly stringent regulatory standards on diesel emissions have required engine modifications and implementation of exhaust line after-treatment controls by manufacturers. Evaluating the

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Table 1
Summary of instrument operation settings and software.

Instrument	Model	Software	Aerosol air flow rate (LPM)	Sheath air flow rate (LPM)	Notes
SMPS	Classifier: 3080. DMA: 3081/3085. CPC: 3786	Aerosol Instrument Manager (ver. 9.0.0)	0.6	6.0	Gas diffusion correction; 1 μm impactor
FMPS	3091	FMPS Software (ver. 3.1.0)	10.0	40.0	1 μm impactor
EEPS	3090	EEPS Software (ver. 3.1.1)	10.0	40.0	1 μm impactor

effectiveness of diesel engine emission reduction technologies requires a measurement platform that is capable of high-time resolution particle characterization over a wide concentration range. This high-time resolution instrumentation may also be applied to characterize the impact of control devices on near-road concentrations of traffic-related air pollutants and real-world vehicle emission factors. The EMITTED study measurement platform includes two spectrometer-based instruments capable of measuring size distributed number concentrations between 6 and 560 nm with 32 size channels: TSI Model 3090 Engine Exhaust Particle Sizer (EEPS) and the TSI Model 3091 Fast Mobility Particle Sizer (FMPS). This work presents a comprehensive comparison study of these instruments for particles of varying characteristics. Agreement of both these sizing instruments was further assessed against another TSI sizing instrument, the Scanning Mobility Particle Sizer (SMPS, Model 3080). In general, acceptable agreement (<15% difference) was observed between the EEPS and the FMPS; however, by extending the comparison to include the SMPS it was determined that size-fractioned correction is required due to the agglomerate nature of diesel PM. Two types of laboratory-generated soot PM were produced with spherical and agglomerate geometries to confirm the effect of particle agglomeration on FMPS and EEPS overestimation of particle number concentration.

2. Methods

2.1. Particle sizing instruments

Particle size distributions and number concentrations were obtained with an EEPS and an FMPS. Both instruments function according to the same operating principles, measuring particle sizes from 6 to 560 nm using a series of electrometers; particles smaller than 8 nm and larger than 300 nm were excluded from analysis as concentrations were below detection for two of the four PM sources tested. In the EEPS and the FMPS, the sampled particles are passed through an impactor to remove particles with an aerodynamic diameter exceeding 1 μm. The remaining particles are subsequently charged in a unipolar diffusion charger and subjected to an electrical field in a high-voltage electrometer column. This causes the particles particles to impact specific electrometers based on their electrical mobility: smaller particles are impacted at the top of the electrometer column with particles of increasing size collected down the length of the column. A data inversion algorithm yields size distribution curves given in 32 equally spaced size bins on a logarithmic scale (TSI Incorporated, 2005; Wang et al., 2006). The FMPS generates particle size distributions at 1 Hz while the EEPS operates ten times faster at 10 Hz.

An SMPS was operated in parallel with the FMPS and EEPS for further equivalency testing with a scanning period of 120 s. The extended time scale required for a complete size distribution measurement limits the use of this instrument to steady-state testing. The SMPS used in this study consisted of a differential mobility analyzer (DMA) and an ultrafine water-based condensation particle counter (UWCPC, TSI model 3786). The size distribution of the particle sources tested determined the DMA used: the nano- (TSI 3085) or long- (TSI 3081) DMA. The SMPS used in this study has been previously described by Jeong and Evans (2009). A summary of the critical parameters for instrument operation is provided in Table 1. Further details on SMPS and FMPS operation are described in Jeong and Evans (2009); the EEPS was operated in an identical fashion to the FMPS. For all parallel measurements, carbon filled conductive tubing was used to minimize particle line losses and sample lines to the EEPS, FMPS and SMPS were identical in length (3 feet). Instrument flow rates were verified at the beginning of each experiment with a mass flow meter (TSI 4045) to ensure accuracy within 10% of the set point. Ongoing comparison with additional particle counting and sizing instrumentation (API651 CPC, additional FMPS 3091) co-located at the Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR) Field Measurement Facility was used as a reference to assure that the performance of the test instruments had not changed over the duration of this study.

2.2. Particle generation and sampling

Equivalency studies were conducted in four operating environments: diluted diesel engine exhaust, urban ambient air, electrospray-generated aerosols from a 0.3% $(NH_4)_2SO_4$ and a 0.5% sucrose solution, and laboratory-generated combustion soot (Fig. 1).

The urban ambient air study was conducted at the SOCAAR Field Measurement Facility located in downtown Toronto, Canada. Ambient air was sampled at the intersection of two high-trafficked roads. The EEPS and the FMPS were operated over four consecutive days in May 2012, yielding a total of 252,000 measurements. For all four days of measurement the SMPS was operated in parallel with the EEPS and the FMPS.

The second set of equivalency studies was conducted with diesel engine (1997 Cummins B3.9-C) exhaust diluted by a TSI Rotating Disk Thermodiluter (Model 379020A) during ISO8178 Mode 9 operation, representative of a low-load, urban driving condition. Five dilution ratios, ranging from 118 to 202, were tested for 10 min each, generating 3000 measurements. Experiments conducted with the diesel particles were performed in triplicate. During the first of the triplicate runs, the SMPS was operated in parallel with the EEPS and FMPS. Discrepancies were identified in the size distributions produced by the SMPS and the EEPS/FMPS. To determine whether this discrepancy was due in part to engine exhaust particle volatility subsequent equivalency experiments with a Dekati Thermodenuder set to an operating temperature of 250 °C were conducted.

As neither the engine nor the ambient environments produced high enough concentrations of sub-25 nm particles to properly compare the EEPS and the FMPS, a third set of equivalency studies was conducted using an aerosol generator (TSI Model 3480 Electrospray Aerosol Generator). Electrospray-generated aerosol measurements were collected from two different ionic solutions: 0.3% ammonium sulphate and 0.5% sucrose with particle diameter modes of 12 and 14 nm, respectively. Particles generated by the electrospray were supplemented with HEPA-filtered air to meet the air flow requirements of parallel FMPS and EEPS operation; a mixing tube was used to ensure a homogeneous stream of diluted electrospray particles. The electrospray flow was diluted with varying amounts of HEPA-filtered air to produce a gradient of

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