Atmospheric Environment 100 (2015) 178–184

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

A source-independent empirical correction procedure for the fast mobility and engine exhaust particle sizers

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HIGHLIGHTS

- Accuracy of high time resolution particle sizers is affected by particle morphology.
- These instruments also under-size particles larger than 80 nm.
- We provide a source-independent correction protocol to address this issue.
- The correction protocol ensures agreement within 20% of a reference instrument.

ARTICLE INFO

Article history:
Received 17 March 2014
Received in revised form
29 October 2014
Accepted 30 October 2014
Available online 31 October 2014

Keywords:
EEPS
FMPS
SMPS
Morphology
Vehicle emissions
Particle size distributions

ABSTRACT

The TSI Fast Mobility Particle Sizer (FMPS) and Engine Exhaust Particle Sizer (EEPS) provide size distributions for 6–560 nm particles with a time resolution suitable for characterizing transient particle sources; however, the accuracy of these instruments can be source dependent, due to influences of particle morphology. The aim of this study was to develop a source-independent correction protocol for the FMPS and EEPS. The correction protocol consists of: (1) broadening the >80 nm size range of the distribution to account for under-sizing by the FMPS and EEPS; (2) applying an existing correction protocol in the 8–93 nm size range; and (3) dividing each size bin by the ratio of total concentration measured by the FMPS or EEPS and a water-based Condensation Particle Counter (CPC) as a surrogate scaling factor to account for particle morphology. Efficacy of the correction protocol was assessed for three sources: urban ambient air, diluted gasoline direct injection engine exhaust, and diluted diesel engine exhaust. Linear regression against a reference instrument, the Scanning Mobility Particle Sizer (SMPS), before and after applying the correction protocol demonstrated that the correction ensured agreement within 20%.

1. Introduction

Shifting trends in particle-phase measurements of traffic-related air pollutants have produced a need for high time resolution (≤1 s) particle number and size characterization over a wide concentration range. For example, high time resolution instruments may be required to characterize the performance of emission control technologies, such as diesel particulate filters (Bergmann et al., 2009; Biswas et al., 2008; Kittelson et al., 2006; Zervas and Dorlhène, 2006); the effect of fuel type on particle number concentrations during transient drive cycles (Liu et al., 2007; Wang et al., 2006); or to characterize particulate matter (PM) in microenvironments near roadways (Baldauf et al., 2008; Choi et al., 2013; Kozawa et al., 2009; Massoli et al., 2012; Reponen et al., 2003; Zhu et al., 2002). Increasingly stringent emissions standards and the need for real-world emission factors necessitate that the employed particle sizing and counting instrumentation be accurate and precise; however, assessing accuracy is difficult due to the lack of traceable measurement standards. These instruments are generally validated against auxiliary, lower time resolution instrumentation, such as the TSI Scanning Mobility Particle Sizer (SMPS). Two commonly applied instruments capable of high time resolution particle number and size characterization are the TSI Fast Mobility Particle Sizer (FMPS) and the Engine Exhaust Particle Sizer (EEPS). When measuring traffic-related particle sources in parallel, agreement between the SMPS, the FMPS,

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http://dx.doi.org/10.1016/j.atmosenv.2014.10.054
1352-2310/

and the EEPS has been inconsistent (Asbach et al., 2009; Awasthi et al., 2013; Kaminski et al., 2013; Leskinen et al., 2012).

In Zimmerman et al. (2014) it was established that measuring concentrations and size distributions of different particle sources with an FMPS or an EEPS- including ambient PM, laboratory-generated soot, aerosolized ionic solutions, and diesel PM- all required unique correction procedures based on linear regression with a Scanning Mobility Particle Sizer (SMPS). Differences in the correction procedures were largely attributed to particle morphology; particles with a more agglomerate structure required greater correction than those with a more spherical structure. Reliance on an additional particle sizer to correct for source-specific discrepancies is time consuming, dependent on the user having access to an SMPS, and limits the correction to particle sources that are constant over the SMPS measurement interval. Thus, the objective of this study was to develop an empirical correction protocol independent of PM source that is suitable for high time resolution applications (i.e., independent of an SMPS). The source-independent correction procedure presented here involves an existing correction protocol developed in Jeong and Evans (2009), broadening the upper fraction of the size distribution, and a scaling factor from parallel measurement by an ultrafine water-based condensation particle counter (UWCPC), which has the same time resolution as the FMPS. Application of this correction protocol ensures accuracy between an FMPS or EEPS and an SMPS within 20%. If more accurate agreement is required, linear regression with an SMPS, as described in Zimmerman et al. (2014), is required.

2. Methods

2.1. Particle sizing instruments

Particle size distributions and number concentrations were obtained with an SMPS and either an FMPS or an EEPS. The EEPS (TSI model 3090) and FMPS (TSI model 3091) both 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 350 nm were excluded from analysis as concentrations were generally below detection. Operation of the EEPS and the FMPS used in this study has been previously described by Zimmerman et al. (2014). The SMPS used in this study consisted of an electrostatic classifier (TSI 3080), differential mobility analyzer (DMA) and ultrafine water-based condensation particle counter (UWCPC, TSI model 3786). Both the long DMA (TSI 3081) and nano DMA (TSI 3085) were included in the SMPS configuration. For the urban ambient measurements, an additional SMPS (TSI 3080 classifier) was used configured with the nano water-based ultrafine condensation particle counter (N-WCPC, TSI model 3788) to allow for parallel measurements with the nano DMA and long DMA; for the engine exhaust comparisons, the SMPS was equipped with the long DMA for two replicates and the nano DMA for the third replicate. The extended time scale (2 min, 100 s for scan up time + 20 s for scan retrace) required for a complete size distribution measurement limited the use of this instrument to steady-state testing. The SMPS used in this study has been previously described by Jeong and Evans (2009). For all parallel measurements, carbon filled conductive tubing was used to minimize particle line losses and sample lines to the instruments 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. As there is no particle counting measurement standard, auxiliary particle counting and sizing instrumentation (API 651 CPC, additional FMPS 3091) co-located at the Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR) Field Measurement Facility was used to verify agreement between instruments with the same working principle. The assumption underlying this approach was that it was highly improbable that a number of different instruments would all drift or fail in such a way as to continue to agree with each other. A summary of the operating parameters and settings for the SMPS, EEPS and FMPS are provided in Table 1.

2.2. Condensation particle counters

In addition to the TSI 3786 UWCPC employed as a component of the SMPS configuration, two additional condensation particle counters (CPCs), a TSI 3788 Nano Water-Based CPC (N-WCPC) and a Teledyne API 651 CPC, were used to measure total particle number concentration. These CPCs use water as their condensing fluid to grow particles for optical counting. A summary of the operating parameters and settings for the CPCs is provided in Table 1.

2.3. Particle generation and sampling

Experiments were conducted with three particle classes: (1) laboratory-generated aerosols, (2) urban ambient air, and (3) diluted heavy-duty diesel and light-duty gasoline direct injection (GDI) engine exhaust (Fig. 1).

Aqueous 1% v/v solutions of ammonium nitrate (NH_4NO_3) and sucrose were prepared using deionized water and standard stock chemicals (sucrose: BioShop Canada, Burlington, ON, Canada; NH_4NO_3: Anachemia, Mississauga, ON, Canada). Two polystyrene latex (PSL) aqueous solutions (147 and 240 nm) were prepared by mixing 3–5 drops of stock solution (NanoSphere™ Size Standards, Thermo Scientific, Fremont, CA, USA) in 100 mL of deionized water and mixed with a vortex mixer. The four aqueous solutions were aerosolized using a constant output atomizer (TSI 3076) connected to a 20 psi filtered air supply (TSI 3074B). The generated aerosol mixture was dried using a silica diffusion drier (TSI 3062) and

Table 1
Summary of instrument operation settings and software.

Instrument	TSI SMPS	TSI FMPS	TSI EEPS	TSI N-WCPC	API CPC
Model	Classifier: 3080 DMA: 3081/3085 CPC: 3786	3091	3090	3788	651
Software	Aerosol Instrument Manager (ver. 9.0.0)	FMPS software (ver. 3.1.0)	EEPS software (ver. 3.1.1)	Aerosol instrument manager (ver. 9.0.0)	Aerosol instrument manager (ver. 9.0.0)
Aerosol sample flow rate (LPM)	0.6 (CPC: 0.3)	10.0	10.0	0.3	0.3
Sheath air flow rate (LPM)	6.0 (CPC: 0.3)	40.0	40.0	0.3	0.3
Time resolution (s)	120	1	0.1	1	1
Size range	3085 DMA: 3–93 nm 3081 DMA: 12–340 nm	6–560 nm	6–560 nm	3 nm–3 μm	7 nm–3 μm
Notes	TSI diffusion correction; 1 μm impactor	1 μm impactor	1 μm impactor	–	–