



# Metrological Assessment of a Portable Analyzer for Monitoring the Particle Size Distribution of Ultrafine Particles

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## ABSTRACT

Adverse health effects caused by worker exposure to ultrafine particles have been detected in recent years. The scientific community focuses on the assessment of ultrafine aerosols in different micro-environments in order to determine the related worker exposure/dose levels. To this end, particle size distribution measurements have to be taken along with total particle number concentrations. The latter are obtainable through hand-held monitors. A portable particle size distribution analyzer (Nanoscan SMPS 3910, TSI Inc.) was recently commercialized, but so far no metrological assessment has been performed to characterize its performance with respect to well-established laboratory-based instruments such as the scanning mobility particle sizer (SMPS) spectrometer. The present paper compares the aerosol monitoring capability of the Nanoscan SMPS to the laboratory SMPS in order to evaluate whether the Nanoscan SMPS is suitable for field experiments designed to characterize particle exposure in different microenvironments. Tests were performed both in a Marple calm air chamber, where fresh diesel particulate matter and atomized dioctyl phthalate particles were monitored, and in microenvironments, where outdoor, urban, indoor aged, and indoor fresh aerosols were measured. Results show that the Nanoscan SMPS is able to properly measure the particle size distribution for each type of aerosol investigated, but it overestimates the total particle number concentration in the case of fresh aerosols. In particular, the test performed in the Marple chamber showed total concentrations up to twice those measured by the laboratory SMPS—likely because of the inability of the Nanoscan SMPS unipolar charger to properly charge aerosols made up of aggregated particles. Based on these findings, when field test exposure studies are conducted, the Nanoscan SMPS should be used in tandem with a condensation particle counter in order to verify and correct the particle size distribution data.

**KEYWORDS:** diesel particulate matter; dioctyl phthalate particles; laboratory SMPS; metrological assessment; Nanoscan SMPS; occupational aerosol; particle size distribution; portable analyzer; total particle number concentration; ultrafine particles

## INTRODUCTION

Airborne particles are considered to be one of the main air pollutants in terms of environmental and occupational health problems for people exposed in both indoor and outdoor microenvironments and in the workplace (World Health Organization, 2005, 2006). Epidemiological and toxicological studies show that possible negative effects on human health can be related to particle exposure, including breathing difficulties, cardiovascular diseases, and general increases in mortality and morbidity (Kreyling *et al.*, 2006; Pope and Dockery, 2006; Miller *et al.*, 2007; Schmid *et al.*, 2009; Strak *et al.*, 2010). The health effects strongly depend on the aerosol particle size (International Commission on Radiological Protection, 1994), chemical composition (Eiguren-Fernandez *et al.*, 2010; Janssen *et al.*, 2011), and assumption rate (i.e. short/long exposure, Manigrasso *et al.*, 2013). The size of the particles is used to regulate certain fractions of airborne aerosols,  $PM_{10}$ ,  $PM_{2.5}$ , and total dust (Council of the European Union, 1999; U.S. Environmental Protection Agency 2004, 2006; European Parliament and Council of the European Union, 2008, 2010). Meanwhile, scientific attention is moving toward sub-micrometric and ultrafine particles (UFPs, particles <100 nm) since these particles can easily cross the human respiratory system, following the airstream all the way down to the alveolar region and deposit in the deepest areas of the lung carrying toxic compounds. In fact, a number of recent studies have related particle effects on health to particle number (Peters *et al.*, 1997; Schulte *et al.*, 2010; Franck *et al.*, 2011a,b) and surface area concentrations (Oberdörster, 2000; Nel *et al.*, 2006; Waters *et al.*, 2009; Cauda *et al.*, 2012).

In order to characterize the exposure to particles in different environments, the scientific community involved in aerosol measurements developed several portable instruments to measure total particle number [condensation particle counters (CPCs) and diffusion chargers, Fierz *et al.*, 2009; Marra *et al.*, 2010; Asbach *et al.*, 2012], deposited surface area (unipolar diffusion chargers, Fissan *et al.*, 2007; Asbach *et al.*, 2009a), and mass concentrations (e.g. photometers, Wang *et al.*, 2009, and the portable tapered element oscillating microbalance, Volkwein *et al.*, 2004).

Particle size distribution represents an essential parameter for the evaluation of a particle exposure/

dose profile, and a complete dimensional characterization of sub-micrometer particles cannot be limited only to integrated data, such as total particle number and surface area concentrations. As mentioned, the inhalation and deposition of particles in the human lungs is strongly related to their size (International Commission on Radiological Protection, 1994). In addition, particle size distribution measurements are essential for the evaluation of a personal particle dose for each resided microenvironment (Buonanno *et al.*, 2012a,b,c; Manigrasso *et al.*, 2013).

Monitoring the real-time evolution of particle size distribution can be performed through a complex measurement process involving particle charging, classification, and counting (McMurry, 2000). To this purpose, differential mobility particle sizer spectrometers, such as the scanning mobility particle sizer (SMPS) (Wang and Flagan, 1990), are used. SMPS spectrometers generally include an electrostatic classifier, where particles are electrically charged/neutralized and then classified according to their electrical mobility [in a differential mobility analyzer (DMA), Knutson and Whitby 1975; Kinney *et al.*, 1991], and a CPC, where particles, previously classified, are counted (Agarwal and Sem, 1980). By varying the voltage in the electrostatic classifier, it is possible to obtain the particle size distribution of the aerosol in the sub-micrometric range.

To perform particle size distribution measurements in different microenvironments for the evaluation of occupational exposure/dose, the SMPS spectrometer cannot be considered since it is a laboratory-intended instrument—i.e. in most cases, it is not a very practical solution to transport it; moreover, it also requires a power supply. Therefore, a novel differential mobility particle sizer spectrometer, usable in the field, was recently developed by TSI Inc., Shoreview, MN, USA (Nanoscan SMPS 3910, Tritscher *et al.*, 2013). The Nanoscan is quite similar to the laboratory SMPS spectrometer in that it has sizing and counting features separated, but unlike the laboratory SMPS it uses (i) a non-radioactive particle charger, (ii) a smaller mobility analyzer, and (iii) a compact particle counter. Moreover, it is battery operated and it can operate and store data without the necessity of a separate computer. A comparison of the performances of the Nanoscan and laboratory SMPS was conducted by TSI using laboratory standard

aerosols, NaCl and Emery Oil particles (Tritscher *et al.*, 2013).

The aim of this study is to compare the metrological performance of the portable Nanoscan SMPS to the laboratory SMPS spectrometer for their ability to measure various aerosols in terms of particle size, concentration, and morphology. Both Marple chamber and environmental experiments were performed, and particle size distributions and total particle number concentrations were evaluated.

## METHODS

### Experimental apparatus

The following instruments were used for this study: two Nanoscan SMPS spectrometers 3910 (TSI Inc., Shoreview, MN, USA), two laboratory SMPS spectrometers 3936 (TSI Inc., Shoreview, MN, USA), and a butanol-based CPC 3775 (TSI Inc., Shoreview, MN, USA).

The Nanoscan is a portable differential mobility particle sizer spectrometer designed to measure particle size distributions and total concentrations in the range of 10–420 nm (13 size channels, when used in the ‘scan mode’) with a sampling time equal to 60 s. A polydisperse aerosol flow is drawn from the inlet section where a cyclone is used to remove large particles. Next, the aerosol is positively charged in the unipolar charger (corona jet type charger, Medved *et al.*, 2000), which uses the opposed flow technique. In particular, the unipolar charger works through the introduction of an opposed charger flow that is filtered with both an active carbon filter and a high-efficiency particulate air (HEPA) filter. After passing over the charger needle, a jet of positive ions flows into the field-free mixing chamber, allowing the interaction of ions with the polydisperse sample flow using the opposed flow technique, which improves mixing and charge repeatability (Tritscher *et al.*, 2013). Next, the charged sample flow is flowed through the classification region, which is made up of a radial DMA (Zhang *et al.*, 1995) whose top plate is at ground with the bottom plate at a high negative voltage. The electric field in the region between the two electrodes is able to modify the positively charged particle trajectories (particles are introduced radially through an inlet channel at the top) according to their electrical mobility. Particles having a certain mobility diameter

are classified through the monodisperse outlet and continue to the counting section; particles with an electrical mobility lower than the fixed one are flowed through the excess flow exits, then this flow is cleaned through a HEPA filter and reused as sheath flow rate in the radial DMA, whereas particles with a higher electrical mobility are captured on the negative electrode. The counting is performed by means of an isopropanol-based CPC (CPC 3007, TSI Inc.). During the 60 s of the ‘scan mode’ measurement, the radial DMA’s voltage is ramped up to scan the entire size range so that particles ranging from 10 to 420 nm are sent to the CPC in sequence and then the complete particle size distribution is obtained. The Nanoscan can be operated in standard or extended mode. In the standard mode, a wick cartridge need to be manually wet with isopropanol every 8 h, whereas, in the extended mode, a bottle need to be filled to supply isopropanol automatically to the wick, allowing for longer sampling periods. The experimental analysis here presented was carried out using the Nanoscan in extended mode.

The SMPS 3936 was used, made up of an electrostatic classifier 3080 (TSI Inc.) and a CPC. In the electrostatic classifier (EC) 3080, particles are electrically neutralized through a Kr<sup>85</sup> neutralizer, then classified in a long DMA 3081 (TSI Inc., Knutson and Whitby, 1975; Mulholland *et al.*, 1999). Particles classified are counted through the CPC. Two different CPCs were used during the experiments in the laboratory SMPS configuration: a CPC 3010 and a CPC 3775. They differ in terms of minimum detectable particle size (10 nm for the CPC 3010 and 4 nm for the CPC 3775) and particle concentration range (up to 10<sup>4</sup> particle cm<sup>-3</sup> for the CPC 3010 and up to 10<sup>7</sup> particle cm<sup>-3</sup> for the CPC 3775; in particular, the CPC 3775 measures up to 5 × 10<sup>4</sup> particle cm<sup>-3</sup> in single particle counting mode and in photometric mode for higher concentration levels). Nevertheless, they have similar metrological performances when used in an SMPS configuration within the size range we tested. A sampling time of 120 s (plus a retrace of 15 s) was used for the laboratory SMPS measurements; aerosol flow and sheath flow were set equal to 0.3 and 3.0 l min<sup>-1</sup>, respectively, so that the corresponding measurement range was 14.6–661.2 nm (64 channels per decade). When compared to the Nanoscan data, only the overlapping 14–420 nm size range of the distribution was considered.

### Procedures

Two different setups were used to assess the metrological characteristics of the Nanoscan: tests in a Marple chamber to measure dioctyl phthalate (DOP) particles and diesel particulate matter (DPM); micro-environment tests to measure different indoor and outdoor aerosols. Tests were performed in the period September 2012–March 2013 at the Office of Mine Safety and Health Research [CDC, National Institute for Occupational Safety and Health (NIOSH), Pittsburgh, PA, USA] and at the European Accredited (EA) Laboratory of Industrial Measurements (LAMI) of the University of Cassino and Southern Lazio (Italy). The Nanoscans being evaluated were calibrated by the manufacturer before the study. In fact, to properly perform experimental analyses on field, the manufacturer suggests that the Nanoscan should be serviced annually for maintenance of the charger, pumps, filters, and RDMA. Nonetheless, when used on field, a calibrated Nanoscan does not need any particular maintenance procedure apart from filling the bottle of isopropanol. The authors performed further comparisons of the Nanoscans with freshly calibrated CPCs measuring NaCl-generated particles were performed at both the measurement sites. The laboratory SMPS spectrometers used for this experiment were freshly calibrated by the manufacturer too.

### Marple chamber tests

Experimental analysis to measure DOP and DPM particles was performed in a calm air Marple chamber (Marple and Rubow, 1983) at the Office of Mine

Safety and Health Research (NIOSH, Pittsburgh, PA, USA). The chamber is hexagonal in cross section, 2.44-m high with an inside diameter of 1.19 m. Aerosol is introduced at the top of the chamber, is thoroughly mixed by an air jet, and flows through a 10-cm-thick honeycomb structure in which turbulence is reduced and low-velocity flow exits and passes through the test section. Past work has shown the sampling zone of the chamber to be very uniform with relative standard deviation in different area of the chamber <0.05 (Marple and Rubow, 1983).

DOP particles were generated through a Constant Output Atomizer 3076 (TSI Inc.) (Stabile *et al.*, 2013). Oil particles were obtained by dissolving DOP in deionized water. In particular, DOP particles were produced through the atomization of the solution and the evaporation of the solvent. A solution with 1.28% oil volume concentration was used. Before entering the Marple chamber, particles flowed through a Kr<sup>85</sup> Aerosol Neutralizer 3012 (TSI Inc.) in order to remove pre-existing charges on particle surfaces (Table 1).

The emissions of a Kubota diesel engine connected to a 10-kW electrical generator (constant speed 1800 r.p.m.) were used as a source of DPM aerosol. The exhaust of the diesel engine was not retrofitted with any control technology, with previous studies on the engine showing a DPM aerosol very rich in EC and with a very low concentration in volatile compounds (Noll *et al.*, 2005; Cauda *et al.*, 2012). The engine was run on ultralow sulfur, road-grade diesel fuel. Resistive load conditions were employed

**Table 1. Summary of the tests performed in the calm air Marple chamber at the NIOSH Office of Mine Safety and Health Research**

Label	Aerosol tested	Experimental apparatus	Notes
DOP1.28	DOP, 1.28% volume	SMPS 3936 (CPC 3010), Nanoscan SMPS 3910	Fresh aerosol; DOP particles neutralized before entering the Marple chamber
DPM5%	Diesel, engine load 5%		Fresh aerosol
DPM30%	Diesel, engine load 30%		
DPM50%	Diesel, engine load 50%		
DPM70%	Diesel, engine load 70%		
DPM80%	Diesel, engine load 80%		

(from 5 to 80%) to mimic typical working conditions for the engine. During tests, a portion of the exhaust emitted by the engine was mixed with particle-free fresh air just before the introduction into the Marple chamber.

During DOP and DPM measurements, samplings were performed using the SMPS 3936 (with a CPC 3010) and the Nanoscan. SMPS and Nanoscan were placed outside the chamber and 1-m-long conductive tubes were used to sample aerosol from the chamber. The sampling lines were placed approximately at the center of the chamber itself at a height of ~40 cm from the bottom. The two sampling points were at ~10 cm one from the other. The following procedure was used during the experiments: particles were generated and continuously flowed to the chamber; thus, the particle concentration suddenly increased. After initiating air flow through the generator, sufficient time was allowed to reach a steady chamber aerosol concentration, i.e. ~10 min. When constant concentrations were reached, 11-min sampling (5 SMPS samples, 11 Nanoscan samples) was performed. During post-processing, the mode of distribution was also checked in order to detect any possible shift toward larger diameters due to coagulation processes. The authors only considered tests to be valid if no coagulation phenomena inside the Marple chamber were detected.

### Tests in different microenvironments

As part of this study, the following aerosols/microenvironments were monitored:

- (1) Outdoor aerosols (October 2012) in a suburban site (named 'O' in the following) at the Office of Mine Safety and Health Research, CDC, NIOSH (Pittsburgh, PA, USA), ~500 m away from a suburban road (Cochrans Mill Road, Pittsburgh, PA);
- (2) Indoor aerosols in a 50-m<sup>3</sup> room of the EA LAMI at the University of Cassino and Southern Lazio (Italy) (named 'I1' in the following)—the room has no windows and has a dedicated mechanical ventilation system (air exchange rate equal to 2.2 h<sup>-1</sup>) that guarantees temperature and relative humidity values equal to 20 ± 1°C and 50 ± 10%, respectively;
- (3) Indoor aerosols in a 150-m<sup>3</sup> room of the LAMI when no indoor sources were in operation (named 'I2' in the following)—the room has two windows that were closed during the experiments, and an ordinary mechanical ventilation system was in operation (air exchange rate equal to 0.3 h<sup>-1</sup>);
- (4) Indoor aerosols in the 150-m<sup>3</sup> room of the LAMI during incense-burning events (named 'I-Inc' in the following)—windows closed, with a mechanical ventilation system in operation;
- (5) Indoor aerosols in the 150-m<sup>3</sup> room of the LAMI during grilling bacon events (named 'I-Grill' in the following)—windows closed, mechanical ventilation in operation;
- (6) Urban aerosols on a weekday along a busy road in Cassino (Italy) with a traffic density of >20 vehicles min<sup>-1</sup>, having a mean velocity of ~30–40 km h<sup>-1</sup> (named 'Urb' in the following).

Major details on the measurement sites are provided in [Buonanno \*et al.\* \(2011b\)](#). A summary of the microenvironments tested is reported in [Table 2](#). In the authors' opinion, the analysis of the selected microenvironments and particle sources will allow to estimate the instruments performance in industrial occupational environments (e.g. welding, [Buonanno \*et al.\*, 2011c](#), and mines, [Cauda \*et al.\*, 2012](#)) since those aerosols are similar, in terms of size and morphology, to DPM and cooking-generated aerosols.

In order to achieve a proper comparison between the laboratory SMPS and the Nanoscan SMPS, steady-state concentration measurements were considered: the authors point out that the present experimental analysis is focused on the evaluation of the Nanoscan in terms of metrological accuracy and precision. Since in I1 and I2 microenvironments no sources were present, constant concentrations were easily detected. Similarly, during the O test (outdoor at NIOSH), aerosol was found to be constant because of the distance from the road. Therefore, during I1, I2, and O tests, the 11-min samples in steady-state conditions (5 SMPS samples, 11 Nanoscan samples) were performed. Particular attention was paid to tests

**Table 2. Summary of the field test experiments performed in different microenvironments**

Label	Aerosol tested	Experimental apparatus	Notes
O	Outdoor aerosol at NIOSH	SMPS 3936 (CPC 3010), Nanoscan SMPS 3910	No sources in proximity of the sampling point
I1	Indoor aerosol in Lab with no windows (Cassino)	SMPS 3936 (CPC 3775), Nanoscan SMPS 3910, CPC 3775	No indoor sources in operation, thermo-hygrometric conditions continuously monitored ( $T = 20 \pm 1^\circ\text{C}$ , relative humidity = $50 \pm 10\%$ )
I2	Indoor aerosol in Lab with windows (Cassino)		No indoor sources in operation, windows closed
I-Inc	Aerosol produced during incense burning in Lab with windows (Cassino)		Smoke produced by three incense sticks (citronella fragrance), windows closed
I-Grill	Cooking-generated aerosol in Lab with windows (Cassino)		Grilling of 100 g of bacon, windows closed
Urb	Urban aerosol along a busy road of Cassino		>20 vehicles $\text{min}^{-1}$ at 30–40 km $\text{h}^{-1}$ ; highly dynamic aerosol; major details in (Buonanno <i>et al.</i> , 2011a)

with indoor sources in operation (cooking, incense), and for these experiments, the particle concentration increased as the source was turned on. During the incense-burning experiment ('I-Inc'), three incense sticks (citronella fragrance) were equispatially distributed on the floor of the room, then lit by a flame and fanned out so that the glowing ember on the incense continued to smolder and burn away the rest of the materials. The incense-burning event lasted ~30 min. Concentrations and distributions were continuously monitored by the CPC 3775 (1-s sampling frequency), SMPS 3936, and Nanoscan 3910 during the entire experiments. The 11-min samples (5 SMPS samples, 11 Nanoscan samples), characterized by near constant concentrations and distributions, were post-processed. Major details on particle emission of incense sticks (in terms of emission factors and particle distributions) are reported in [Stabile \*et al.\* \(2012\)](#).

Similarly, the cooking experiment ('I-Grill') was performed setting up a domestic gas stove kitchen inside the 150-m<sup>3</sup> room. The cooking experiment was performed by grilling 100 g of bacon on a stove at medium power

for 10 min. The experiment lasted ~40 min. The concentration suddenly increased as the source was turned on, reaching a maximum value after ~10 min; then a concentration decay was detected as the source was turned off. The initial background concentration was reached in ~30 min. For the incense-burning experiment, particle concentrations and distributions were continuously monitored by the CPC 3775 (1-s sampling frequency), SMPS 3936, and Nanoscan 3910 throughout the experiment. In order to perform a comparison during the fresh particle emission from grilling, the 11-min samples (5 SMPS samples, 11 Nanoscan samples) over the particle concentration peak, characterized by relatively constant concentrations and distributions, were considered during the data post-processing. Major details on particle emission from cooking events (in terms of emission factors, particle distributions, particles' volatility) are reported in [Buonanno \*et al.\* \(2009b\)](#) and [Buonanno \*et al.\* \(2011b\)](#).

During the urban microenvironment testing (Urb), the SMPS 3936, Nanoscan 3910, and CPC 3775 continuously collected data for 1 h. The experiment

was performed to monitor the performance of the Nanoscan 3910 in measuring a highly dynamic aerosol typical of urban roads; therefore, steady-state conditions were not reached.

### Data post-processing

The authors debated the necessity to post-process the SMPS 3936 data by applying the multiple charge (Hoppel, 1978; Fissan *et al.*, 1983), diffusion losses (Gormley and Kennedy, 1949), and aggregate corrections (Lall *et al.*, 2008). Since none of the aerosols tested present a mode  $<20$  nm, the diffusion losses correction was found to increase the concentration of the first channels of the distribution, leading to an unusual second mode at very low diameters even if aerosols that were not fresh were monitored. On the contrary, multiple charge correction is only important in the case of exceptional amounts of very large particles (Birmili *et al.*, 2008), and this should not be the case since the particle cut-point diameter ( $D_{50}$ ) of the 0.0457-cm impactor used for the laboratory SMPS measurements ranged from 480 nm (diesel-generated particles with a density of  $1.8 \text{ g cm}^{-3}$ ) to 679 nm (particles with a unit density). Moreover, when multiple charge and aggregate corrections were applied to diesel- and cooking-generated particles (primary particle diameter of 30 nm; Park *et al.*, 2004; Buonanno *et al.*, 2009b), the distributions shifted toward larger diameters, whereas the total concentration was found to be lower than the uncorrected one. In addition, laboratory SMPS data without applying any corrections were closer to the CPC data for experiments during which CPC measurements were simultaneously performed (steady-state tests I1, I2, I-Inc, I-Grill). To demonstrate, in Table 3, average particle number concentration data measured

through the CPC 3775 and the SMPS 3936 with different corrections are reported. The average concentration ratios between the laboratory SMPS and the CPC 3775 were measured equal to 1.14, 0.81, and 0.96 when diffusion and multiple charge correction, multiple charge correction, and no corrections were applied to the laboratory SMPS data, respectively. Results both with and without SMPS corrections are discussed in the following sections. Finally, in the authors' opinion, SMPS data not corrected for multiple charge and diffusion should be considered for a proper comparison with Nanoscan because similar corrections tools are not suitable to post-process Nanoscan data.

### Data analysis

Total particle number concentration and size distribution data during a single test [mode and geometric standard deviation (GSD)] were expressed as average  $\pm$  standard deviation. Differences in total particle number concentration data between two data sets were tested by analysis of variance (ANOVA); a post-hoc Tukey–Kramer test was applied when needed if more than two data sets were compared (i.e. when CPC was also used). A  $P$  value  $<0.01$  (99% confidence level) was considered significant. All the tested data were previously checked for normality in order to realize the pertinence of the ANOVA and Tukey–Kramer tests: a Shapiro–Wilk test was applied.

The metrological compatibility between the Nanoscan and laboratory SMPS in terms of particle size distribution data was evaluated on the basis of the normalized error ( $E_n$ ) introduced by the International Organization for Standardization (1997). The normalized error was evaluated channel by channel ( $E_{n,i}$ ) considering the midpoint channel concentrations of

**Table 3. The 11-min average particle size data measured for aerosols I1, I2, I-Inc, and I-Grill by the SMPS 3936, when different corrections are applied, and the CPC 3775**

Aerosol tested	SMPS 3936 diffusion and multiple charge correction (particle $\text{cm}^{-3}$ )	SMPS 3936 multiple charge correction (particle $\text{cm}^{-3}$ )	SMPS 3936 no corrections (particle $\text{cm}^{-3}$ )	CPC 3775 (particle $\text{cm}^{-3}$ )
I1	$7.79 \times 10^3$	$5.45 \times 10^3$	$6.33 \times 10^3$	$6.33 \times 10^3$
I2	$2.12 \times 10^4$	$1.65 \times 10^4$	$2.02 \times 10^4$	$1.94 \times 10^4$
I-Inc	$3.99 \times 10^4$	$3.05 \times 10^4$	$3.65 \times 10^4$	$4.09 \times 10^4$
I-Grill	$1.63 \times 10^5$	$1.01 \times 10^5$	$1.18 \times 10^5$	$1.29 \times 10^5$

the laboratory SMPS and the Nanoscan. Since the two instruments use different channel widths and midpoints, mode and GSD values measured by the two instruments (Table 5) were used to fit the data through log-normal distributions: a goodness-of-fit value  $<0.002$  was considered acceptable in data-fitting procedure. Fitted size distributions were considered in particle number concentration data provided shown hereinafter (e.g. Table 4). The distributions were fitted considering the midpoint channel of the laboratory SMPS in the range of 14–420 nm. They were normalized to the total number concentration measured for the specific aerosol. The proposed metrological compatibility was focused on the particle size distribution. The channel normalized error ( $E_{n,i}$ ) was evaluated as:

$$E_{n,i} = \frac{|C_{\text{Nanoscan},i} - C_{\text{SMPS},i}|}{\sqrt{U_{\text{Nanoscan},i}^2 + U_{\text{SMPS},i}^2}} \quad (1)$$

where  $C_{\text{Nanoscan},i}$  and  $C_{\text{SMPS},i}$  represent the normalized total particle number concentration of the  $i$ th channel for the Nanoscan and laboratory SMPS, respectively, and  $U_{\text{Nanoscan},i}$  and  $U_{\text{SMPS},i}$  represent corresponding absolute expanded uncertainties (quantity defining an interval about the result of a measurement that may be expected to encompass a large fraction of the distribution of values that could reasonably be attributed to the measurand; International Organization for Standardization, 2008). The uncertainty budget (a statement of measurement uncertainty, of the components of that measurement uncertainty, and of their calculation and combination) of the channel concentration data for both the Nanoscan and laboratory SMPS was performed through the model proposed in Buonanno *et al.* (2009a). The overall metrological compatibility between the Nanoscan and laboratory SMPS distributions was evaluated defining the global normalized error ( $E_n$ ).  $E_n$  was calculated summing the normalized error of all the channels weighted by their particle number concentration, as follows:

$$E_n = \sum_{i=14\text{nm}}^{420\text{nm}} E_{n,i} \frac{C_{\text{average},i}}{C_{\text{average}}} \quad (2)$$

here,  $C_{\text{average},i}$  and  $C_{\text{average}}$  represent the average (between the Nanoscan and laboratory SMPS data)

particle concentration of the single channel and the average (between the Nanoscan and laboratory SMPS data) total particle concentration, respectively. Normalized particle size distributions were considered compatible from a metrological point for  $E_n < 1$  (International Organization for Standardization, 2010).

## RESULTS

In Tables 4 and 5, results for total particle number concentrations and size distributions (mode and GSD), respectively, are reported for both in-chamber and microenvironment test experiments. Data represent the average of the 11-min samples (5 SMPS samples, 11 Nanoscan samples) as reported in the Methods section. Measurements were performed in steady-state conditions; in fact, standard deviations of the Nanoscan, laboratory SMPS, and CPC 3775 total particle number concentration data were measured in the ranges of 2.0–16.0%, 1.9–18.0%, and 2.8–7.0%, respectively. Similarly, standard deviations of the Nanoscan and SMPS 3936 size distribution modes were measured in the ranges of 0–0.2% and 2.1–10.9%, respectively. Both SMPS data with and without corrections are reported.

In order to compare the Nanoscan and laboratory SMPS results, the ratio between total particle number concentration of the Nanoscan and laboratory SMPS was evaluated (Table 4) as well as the potential shift of the Nanoscan mode in comparison to the laboratory SMPS mode (Table 5). When the SMPS data obtained without corrections are considered, the test with DOP particles showed how the Nanoscan and the laboratory SMPS generated similar results in terms of total particle number concentration (ratio 0.98)—in fact; the ANOVA test performed between the Nanoscan and laboratory SMPS data showed no statistically significant differences with a 99% confidence level. This result is in line with the results previously reported by TSI (Tritscher *et al.*, 2013). In contrast, the particle concentration ratios between Nanoscan and SMPS data were measured  $>1.7$  (average 1.94) when measuring DPM.

For the microenvironment tests, in terms of total particle number concentration, the Nanoscan overestimation with respect to the laboratory SMPS decreased as the aerosols became aged and/or mixed with aged particles, which is when the time lag between particle

Table 4. The 11-min average total particle size data measured for both in-chamber and field test experiments

Aerosol tested	Nanoscan 3910 (particle cm <sup>-3</sup> )	Laboratory SMPS no corrections (particle cm <sup>-3</sup> )	Laboratory SMPS diffusion and multiple charge correction (particle cm <sup>-3</sup> )	CPC 3775 (particle cm <sup>-3</sup> )	Nanoscan 3910/ laboratory SMPS average ratio	Nanoscan 3910/laboratory SMPS diffusion and multiple charge correction average ratio
DOP1.28	1.28 ± 0.08 × 10 <sup>6</sup>	1.31 ± 0.04 × 10 <sup>6</sup>	1.41 ± 0.04 × 10 <sup>6</sup>	—	0.98	0.91
DPM5%*	3.02 ± 0.08 × 10 <sup>5</sup>	1.34 ± 0.03 × 10 <sup>5</sup>	1.15 ± 0.02 × 10 <sup>5</sup>	—	2.25	2.62
DPM30%*	4.56 ± 0.07 × 10 <sup>5</sup>	2.35 ± 0.34 × 10 <sup>5</sup>	2.01 ± 0.36 × 10 <sup>5</sup>	—	1.94	2.27
DPM50%*	2.65 ± 0.04 × 10 <sup>5</sup>	1.39 ± 0.21 × 10 <sup>5</sup>	1.15 ± 0.21 × 10 <sup>5</sup>	—	1.91	2.30
DPM70%*	2.97 ± 0.17 × 10 <sup>5</sup>	1.62 ± 0.09 × 10 <sup>5</sup>	1.27 ± 0.07 × 10 <sup>5</sup>	—	1.83	2.34
DPM80%*	4.54 ± 0.35 × 10 <sup>5</sup>	2.57 ± 0.22 × 10 <sup>5</sup>	1.98 ± 0.17 × 10 <sup>5</sup>	—	1.77	2.29
O*	5.39 ± 0.27 × 10 <sup>3</sup>	4.04 ± 0.18 × 10 <sup>3</sup>	3.70 ± 0.17 × 10 <sup>3</sup>	—	1.33	1.46
I1*	6.83 ± 0.14 × 10 <sup>3</sup>	6.33 ± 0.26 × 10 <sup>3</sup>	7.79 ± 0.24 × 10 <sup>3</sup>	6.33 ± 0.26 × 10 <sup>3</sup>	1.08	0.88
I2*	2.43 ± 0.08 × 10 <sup>4</sup>	2.02 ± 0.04 × 10 <sup>4</sup>	2.12 ± 0.04 × 10 <sup>4</sup>	1.94 ± 0.05 × 10 <sup>4</sup>	1.20	1.15
I-Inc*	4.61 ± 0.21 × 10 <sup>4</sup>	3.65 ± 0.21 × 10 <sup>4</sup>	3.99 ± 0.18 × 10 <sup>4</sup>	4.09 ± 0.29 × 10 <sup>4</sup>	1.26	1.16
I-Grill*	1.57 ± 0.08 × 10 <sup>5</sup>	1.18 ± 0.03 × 10 <sup>5</sup>	1.63 ± 0.02 × 10 <sup>5</sup>	1.29 ± 0.05 × 10 <sup>5</sup>	1.33	0.96
Urb <sup>a</sup>	6.32 ± 0.33 × 10 <sup>4</sup>	4.37 ± 1.82 × 10 <sup>4</sup>	6.89 ± 3.79 × 10 <sup>4</sup>	7.00 ± 6.08 × 10 <sup>4</sup>	1.45	0.92

<sup>a</sup>Urban aerosol data were not obtained in steady-state conditions.

\* $P_{(\text{Nanoscan SMPS\_no\_corrections})} < 0.01$ .

Table S. The 11-min average particle size data measured for both in-chamber and field test experiments

Aerosol tested	Nanoscan 3910		Laboratory SMPS no corrections		Laboratory SMPS diffusion and multiple charge correction		Nanoscan 3910 versus laboratory SMPS (no corrections)		Nanoscan 3910 versus laboratory SMPS (diffusion and multiple charge correction)	
	Mode (nm)	GSD	Mode (nm)	GSD	Mode (nm)	GSD	Mode <sub>Nanoscan</sub>	Mode <sub>SMPs</sub>	Mode <sub>Nanoscan</sub>	Mode <sub>SMPs</sub>
							Mode <sub>SMPs</sub>		Mode <sub>SMPs</sub>	
DOP1.28	133.0±0.2	1.60±0.01	145.2±11.5	1.69±0.01	111.2±7.5	1.75±0.01	-8%	0.55	-23%	1.30
DPM5%	86.8±0.0	1.49±0.01	87.6±4.1	1.58±0.01	88.9±5.2	1.59±0.01	-1%	0.46	-3%	0.52
DPM30%	86.8±0.0	1.47±0.01	90.6±4.2	1.56±0.01	95.4±6.4	1.57±0.02	-4%	0.54	-9%	0.76
DPM50%	115.5±0.0	1.46±0.01	107.2±5.1	1.55±0.02	112.7±6.0	1.56±0.02	8%	0.66	2%	0.55
DPM70%	115.5±0.0	1.45±0.01	128.7±2.7	1.59±0.01	132.3±6.0	1.61±0.01	-10%	0.94	-13%	1.09
DPM80%	115.5±0.0	1.47±0.01	131.1±6.7	1.59±0.01	144.9±4.4	1.61±0.01	-12%	0.94	-20%	1.44
O	52.1±0.1	1.99±0.02	49.7±5.4	2.15±0.03	49.2±5.1	2.15±0.06	5%	0.33	6%	0.34
I1	86.8±0.0	1.81±0.01	99.2±8.9	1.98±0.01	100.5±8.8	2.03±0.02	-13%	0.67	-14%	0.75
I2	115.5±0.0	1.71±0.02	120.5±6.9	1.89±0.02	140.8±17.1	1.96±0.02	-4%	0.55	-18%	0.97
I-Inc	101.0±0.2	1.68±0.02	104.0±9.1	1.87±0.02	107.9±11.0	1.92±0.02	-3%	0.61	-6%	0.76
I-Grill	115.5±0.0	2.10±0.02	113.5±5.8	2.23±0.02	113.5±5.8	2.32±0.02	2%	0.24	2%	0.37

Urban aerosol data are not included since particle distributions were not obtained in steady-state conditions.

emission and measurement increased. During I1 test, the particle number concentration ratio is close to 1 because the aerosol is sampled in an environment with no windows not affected by the possible outdoor particle concentration level/source. The ratio increase to 1.2 when the outdoor particle penetration in the room slightly increases and is not anymore negligible (I2 test). All of the other microenvironment test aerosols present concentration ratios  $>1.25$ . In particular, the I-Inc, I-Grill, and O tests present Nanoscan to laboratory SMPS average particle concentration ratios equal to 1.26, 1.33, and 1.33, respectively. All of the tests in different microenvironments as well as those for DPM aerosols reveal metrologically significant differences between the Nanoscan and laboratory SMPS for particle number concentrations ( $P$  value  $< 0.01$ ). Higher differences and ratios were found when SMPS data with correction applied were measured.

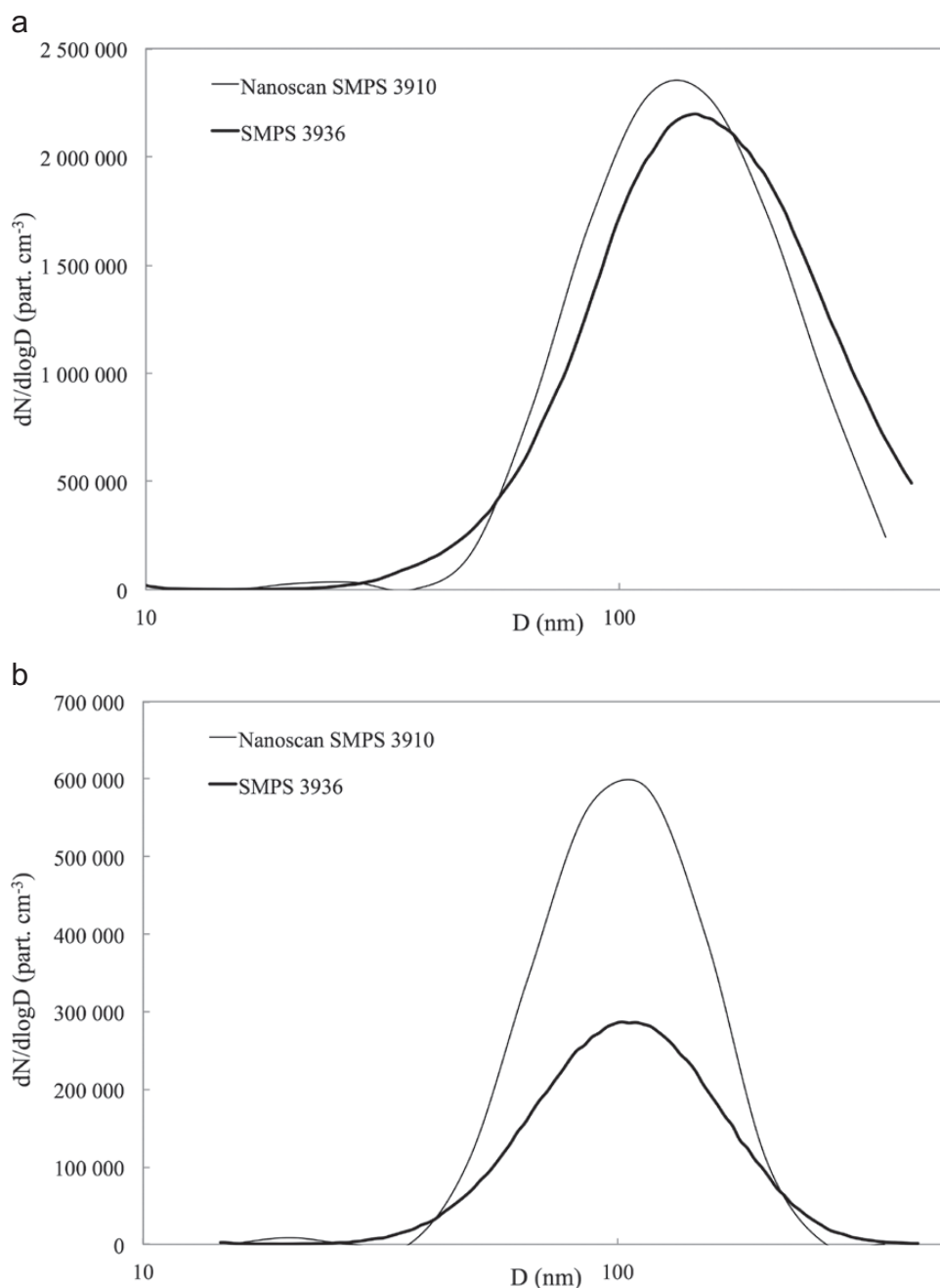
The Nanoscan overestimation in measuring particle number concentration cannot be explained by particle size misclassification in the radial DMA, since a negligible mode shifting was detected. When SMPS data without corrections are considered, the average mode shift was 6%, whereas the maximum one was 13%. However, the maximum shift was found within the channel resolution of the Nanoscan. In particular, the worst Nanoscan particle sizing occurs when the mode detected by the laboratory SMPS corresponds to the lower or upper boundary of the Nanoscan channel; this leads to a maximum shift of  $\sim 16\%$  from the midpoint. All of the Nanoscan distributions measured are centered on the laboratory SMPS modes and slightly shrunk with respect to the laboratory SMPS shapes. As an example, the I1-min average particle size distribution data obtained thorough the Nanoscan and laboratory SMPS during I1 and DPM50% tests are reported in Fig. 1. As shown, the Nanoscan distribution is very close to the laboratory SMPS during the I1 test, whereas the two distributions are significantly different ( $P < 0.01$ ) when diesel particles are monitored. To compare the particle size distributions resulting from the Nanoscan to the laboratory SMPS tests more rigorously, the normalized error ( $E_n$ ) between the two distributions was evaluated as reported in the Data analysis section. Normalized errors were found to be  $<1$  for all the aerosols tested (see Table 5 in Results section) when SMPS data without corrections are considered; therefore, even

if the Nanoscan overestimates the total particle number concentration of certain aerosols, the shape of the distributions (the normalized particle size distributions) it measures is metrologically compatible with the laboratory SMPS results. The higher values of  $E_n$  were measured when larger shifts of the mode as well as higher GSD ratios between the Nanoscan and laboratory SMPS values were detected. When SMPS data with corrections were considered, higher normalized errors were calculated; in particular,  $E_n$  close to or  $>1$  were evaluated. Such metrological incompatibility is due to the fact that corrections are only applied to the SMPS data. Nonetheless, since the aim of the study was to evaluate the metrological performances of the Nanoscan with respect to the SMPS ones, the authors preferred discussing more in depth the SMPS data obtained without corrections. If multiple charge and diffusion correction models were developed for Nanoscan, a comparison with the corrected SMPS data could be performed.

The test on urban aerosol (Urb) provides additional information on the Nanoscan. As described in the Methods section, the test cannot be considered steady state and for this reason, it is more difficult to explain the ratio (1.45) of average particle number concentration monitored by the two instruments. It is possible that the Nanoscan overestimated the particle concentration but the different time resolution (60 s for Nanoscan, 135 s for the laboratory SMPS) is an additional factor. It is known that the laboratory SMPS is not able to perform proper measurements when highly dynamic aerosols are monitored (Manigrasso and Avino, 2012; Manigrasso *et al.*, 2013). With its higher frequency analysis, the Nanoscan provides a particle number concentration trend closer to the CPC data (Fig. 2).

## DISCUSSION

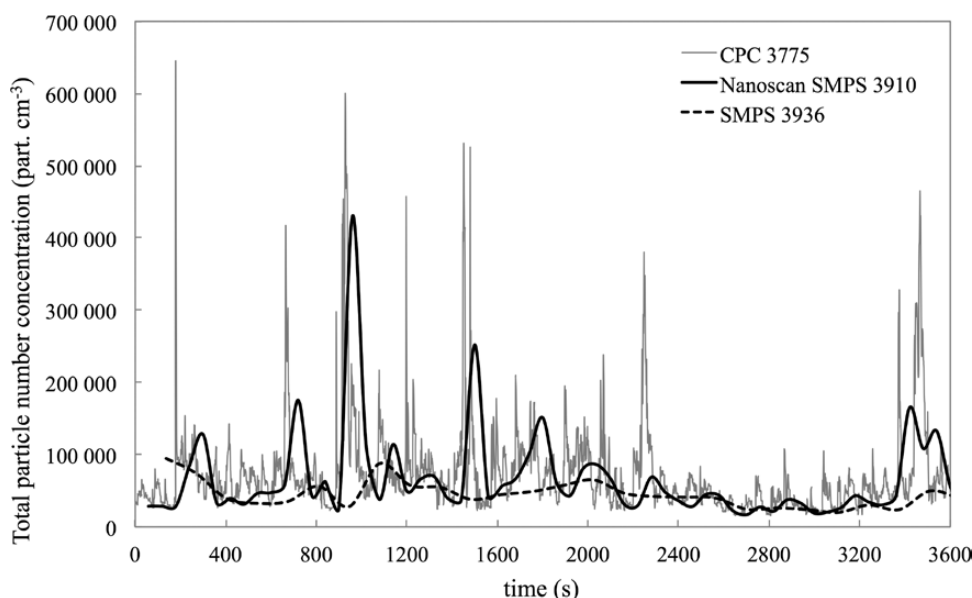
The results reported above suggest that the Nanoscan 3910 has less accurate metrological performance than the laboratory SMPS, which can be considered the best technology commercially available to measure particle size distributions. The difference in total particle number concentration measured for most of the aerosols tested cannot be caused by the lower resolution. It is well-known that the uncertainty related to the total number concentration measured through an integration of several channels increases



1 Comparison of particle size distribution measurements performed through the Nanoscan 3910 and SMPS 3936 during I1 (top) and DPM50% test (bottom).

as the number of channels decreases (Buonanno *et al.*, 2009a) and that this uncertainty is caused by particle misclassification. The modes measured by the Nanoscan 3910 during this study show a good agreement with those measured by the laboratory

SMPS, and the GSDs were just slightly lower. More generally, the normalized error analysis showed that the Nanoscan is able to measure a particle size distribution that 'stays within the error' of the laboratory SMPS distribution when the distributions are



2 Particle number concentration trends measured for 1 h along an urban road of Cassino (Urb test) through CPC 3775, Nanoscan 3910, and SMPS 3936.

normalized to the corresponding total particle number concentration: i.e. Nanoscan provides particle size distribution measurement data metrologically compatible to the SMPS ones. It is possible to propose some explanations for the substantial difference in particle counting for some of the aerosols tested: different particle charging methods, the presence of pre-existing charges on the particle surface, and particle morphology. The Nanoscan 3910 uses an unipolar diffusion charger, which is considered the easiest way to charge particles, but it is also recognized to mischarge particles when they have agglomerate structures (Biskos *et al.*, 2004; Jung and Kittelson, 2005; Asbach *et al.*, 2009b; Ouf and Sillon, 2009; Shin *et al.*, 2010; Wang *et al.*, 2010; Asbach *et al.*, 2012; Leskinen *et al.*, 2012; Kaminski *et al.*, 2013) and/or carry pre-existing charges (Qi *et al.*, 2009; Kaminski *et al.*, 2013). The mischarging phenomenon can be up to 30% in the presence of pre-existing charges (Qi *et al.*, 2009), whereas the morphology itself can lead to a particle overcharge of ~15% (Jung and Kittelson, 2005). Since mobility analyzer measurements are based on a proper aerosol charging technique (i.e. a known particle charge distribution), a mischarging could lead to a different charge distribution, in that case the data inversion cannot be done correctly and

the particle size distribution data result distorted. As an example, several researchers (Asbach *et al.* 2009a, 2012; Leskinen *et al.*, 2012; Kaminski *et al.*, 2013) performed comparisons between fast mobility particle sizers (FMPSs), SMPSs, and CPCs in measuring different aerosols. They recognized that FMPSs overestimated the particle number concentration up to 70%. This is in agreement with the results presented above for the Nanoscan since both the FMPS and the Nanoscan use a similar unipolar diffusion charger.

Summarizing the results, when fresh aerosols made up of aggregated particles (diesel-generated particles) are measured, the Nanoscan is not able to properly neutralize the aerosols then overestimating (up to 2-fold) the actual total particle concentration obtained through the laboratory SMPS. On the contrary, when spherical particles are measured (DOP particles), the Nanoscan agrees with the laboratory SMPS as previously published by Tritscher *et al.* (2013) in their tests on both monodisperse and polydisperse sodium chloride and Emery Oil particles. The authors point out that the pre-existing charge effect could be negligible in the tests here shown since the diesel-generated particles are mixed in a chamber then losing their typical charge distribution. In fact, Maricq (2008) showed that freshly generated particles present a Boltzmann charge distribution

characterized by the flame temperature, but, as soon as the particles leave the flame, the charge temperature (as defined via the Boltzmann distribution) is observed to fall to room/chamber temperature over a period of a few seconds: therefore, the diesel-generated particles sampled inside the chamber present a Boltzmann charge distribution typical of aged aerosol.

The microenvironment testing results provide additional information. In fact, the Nanoscan overestimation reduced to ~1.3-fold the laboratory SMPS concentration when indoor aerosols produced by combustion processes were tested. In particular, the authors tested cooking-generated and incense burning-generated particles showing that these aerosols present aggregated structures (Buonanno *et al.*, 2009b; Chuang *et al.*, 2011). When the combustion source influence is null or negligible, as with the aerosols tested in laboratories with no indoor sources (I1 and I2), the Nanoscan response becomes more similar to the laboratory SMPS response. In that case, the effect of particle morphology and charge is negligible and thus the unipolar charger performance should be better. In particular, in the I1 and I2 tests, the Nanoscan overestimation with respect to the laboratory SMPS in terms of total particle number concentration is within 20%, which represents a worthy metrological performance for a field instrument.

## CONCLUSIONS

In the present work, the metrological performance of a portable differential mobility particle sizer (Nanoscan SMPS 3910, TSI Inc.) was evaluated through a comparison with a calibrated laboratory-based DMA (SMPS 3936, TSI Inc.) in relation to particle size distribution and particle number concentration. In-chamber and microenvironment tests were performed at the Office of Mine Safety and Health Research (CDC, NIOSH, Pittsburgh, PA, USA) and at the European Accredited Laboratory of the University of Cassino and Southern Lazio (Italy) to investigate the Nanoscan SMPS response to different aerosols. Particles produced by typical indoor and outdoor combustion sources (diesel, cooking, incense burning) were tested as well as indoor and outdoor aged aerosols along with atomized DOP particles.

During in-chamber tests with both fresh diesel-generated particles and atomized DOP particles, total concentrations of diesel-generated particles measured

by the Nanoscan SMPS were up to twice those of the laboratory SMPS, whereas particle size distribution measured by the Nanoscan SMPS was metrologically compatible to that of the laboratory SMPS. A possible explanation of the miscounting could be the effect of particle morphology on Nanoscan particle charging technique. In fact, spherical DOP particles were both correctly counted and sized by the Nanoscan SMPS. The effect of particle morphology was still present when outdoor microenvironments and indoor combustion sources were monitored, but it decreased as the fresh aerosol was diluted with ambient particles becoming aged.

The authors acknowledge the potential benefit of using the Nanoscan SMPS for field testing instead of the laboratory SMPS. The microenvironments/sources here analyzed allow extending the findings to other occupational and industrial environments (e.g. welding) since the wide range of particle properties typical of the aerosols examined. During field testing, the use of a condensation particle counter in conjunction with the Nanoscan SMPS is recommended for monitoring the actual total particle concentration and, in case of fresh aerosol sampling, corrected the data. In addition, even if the frequency of monitoring by the Nanoscan SMPS is higher than the laboratory SMPS, its 60-s analysis time might not be sufficient for highly dynamic sources/microenvironments.

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## DISCLAIMER

The mention of any company or product does not constitute an endorsement by the National Institute for Occupational Safety and Health. The findings and conclusions in this paper are those of the authors and do not necessarily represent the views of the National Institute for Occupational Safety and Health.

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