Metrological Performances of a Diffusion Charger Particle Counter for Personal Monitoring

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ABSTRACT

Airborne particles have been shown to be associated with a wide range of adverse health effects, and have thus attracted an increasing amount of attention by medical researchers. However, accurate evaluations of the related exposure-dose-response relationships are highly dependent on the ability to track people's actual exposure to airborne particles. This is quite a complex task, particularly in relation to submicrometer and ultrafine particles, which can vary quite significantly in terms of particle surface area and number concentration. It is thus necessary to develop suitable monitors that can be worn in order to measure personal exposure to these particles.

This paper presents an evaluation of the metrological performance of six diffusion charger sensors, namely NanoTracer (Philips Aerasense) monitors, when measuring particle number and surface area concentrations, as well as particle number distribution mean, when compared to reference instruments. Tests in the laboratory (by generating monodisperse and polydisperse aerosols) and in the field (using natural ambient particles) were designed to evaluate the responses of these devices under both steady-state and dynamics conditions.

The results show that the NanoTracers performed well when measuring steady state aerosols, although they strongly underestimated the actual concentrations during dynamic response testing. The field experiments also showed that, when the majority of the particles were smaller than 20 nm, which occurs during particle formation events in the atmosphere, the NanoTracers underestimated the number concentration quite significantly. Overall, even though NanoTracers can be used for personal monitoring of exposure to ultrafine particles, they have some limitations which need to be considered in order to obtain meaningful results.

Keywords: Ultrafine particles; Alveolar deposited surface area concentration; Personal monitoring; Exposure-risk assessment.

INTRODUCTION

In recent years, a number of studies have focused on aerosol monitoring in order to estimate human exposure to particle concentrations. This increase in technical and scientific attention was motivated by medical studies relating high particle concentration exposure to adverse health effects (Kreyling *et al.*, 2006; Pope and Dockery, 2006; Schmid *et al.*, 2009). However, the view within the medical community is not unanimous when it comes to determining which aerosol physical properties are of most significance in relation to these health effects. Scientific interest has recently shifted from mass concentration (PM) (Loomis, 2000; Pope, 2000) to surface area and number concentration (Giechaskiel *et al.*, 2009; Franck *et al.*, 2011; Cauda *et al.*, 2012), with a focus on smaller particles, such as ultrafine particles (UFPs with a diameter smaller than 100 nm), due to their ability to be deposited in lower regions of the respiratory tract, thus leading to a range of adverse health effects (ICRP, 1994; Wang, 2005; Hofmann, 2011).

Exposure Assessment: State-of-the-Art

Assessing the exposure of urban populations to aerosol particles can be carried out according to five different spatial scales: i) "city scale", the broadest and most common scale used to characterize air quality across several city blocks using remote measurements; ii) "outdoor scale", which is representative of particle exposure outside a building/s of interest; iii) "indoor scale", which is measured within buildings and reflects indoor-based exposure; iv) "individual scale", where the sampling location is within 3 meters of the person, and v) "personal scale", using portable instruments as personal monitors, with a distance of < 30 cm between

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the sampling point and the nasal cavities (Cattaneo *et al.*, 2010).

With regard to urban air quality (city and outdoor scales), regulatory authorities (Directive 2008/50/EC) have only standardized threshold limits for PM_{10} (mass of particulate matter with an aerodynamic diameter less than 10 μm), however these are still stated in terms of average values obtained by time integrated measurements (particles collected over a 24 h period) and do not take into account daily variations in particle concentration, which could be related to atmospheric dynamics, as well as source emission characteristics (Buonanno *et al.*, 2011a). Moreover, the daily PM_{10} concentration is determined at a fixed sampling point (FSP) that is characteristic of the examined area, and it is used to evaluate the exposure of the entire population living nearby. The number of FSPs is determined as a function of population size, without any link to microclimatic characteristics (Council Directive 1999/30/EC). Therefore, the measurements from a FSP cannot be accurately used to characterize real exposure in outdoor or indoor microenvironments.

In reality, the assessment of a person's individual exposure-risk is a complex task, related to a multiplicity of sources, microenvironments and personal lifestyles (Buonanno *et al.*, 2011b, 2012a). For example, in addition to systematically investigated outdoor sources/environments (e.g., traffic-related and heating-related emissions in urban environments, urban background, rural background, sources in proximity to highway and industrial plants), indoor particle exposure should also be considered along with outdoor aerosol sources. In fact, in many countries, people spend most of their time (80–90%) indoors, where cooking represents one of the most significant particle generating activities (Brasche and Bischof, 2009; Chau *et al.*, 2002; Leech *et al.*, 2002).

The adequate evaluation of the exposure-dose-response relationship is even more complex for gases (Kim *et al.*, 2002; Nerriere *et al.*, 2005; Violante *et al.*, 2006) and smaller airborne particles, such as UFPs, whose concentrations depend on the distance from the source (Buonanno *et al.*, 2009a, 2011a). Consequently, in-depth studies aimed to monitor individual exposure to ultrafine particles should be carried out, in order to increase confidence in epidemiologic associations between health outcomes and ambient particle concentrations (Chow *et al.*, 2002). For this purpose, different diffusion charger sensors were recently developed for use in personal monitoring of ultrafine particle exposure (Fierz *et al.*, 2009; Marra *et al.*, 2010; Fierz *et al.*, 2011; Asbach *et al.*, 2012; Fierz, 2012). Philips Aerasense NanoTracer represents one of the diffusion charger sensor recently commercialized. It allows to monitor the personal exposure to both particle number and lung-deposited surface area concentrations.

Aim

The aim of this work was to investigate the metrological characterization of these NanoTracers with respect to: i) monodisperse and polydisperse aerosols generated to reproduce both steady-state and highly dynamic conditions, at the Laboratory of Industrial Measurements (LAMI),

University of Cassino and Southern Lazio (Cassino, Italy); and ii) in the field applications for ambient aerosols at the Queensland University of Technology (Brisbane, Australia). Six identical instruments we used to account, as much as possible, for variation between the characteristics of individual instruments, three of which were used under laboratory conditions, with the remaining three used for ambient air investigations.

In terms of particle number, surface area concentration and particle number distribution modes, calibration was carried out through comparisons with measurements from Condensation Particle Counters (CPCs), Scanning Mobility Particle Sizer (SMPS) spectrometers, Fast Mobility Particle Sizer (FMPS) spectrometers and Nanoparticle Surface Area Monitors (NSAMs).

Nanotracers were previously compared to laboratory-based instruments by Asbach *et al.* (2012) and Monz (2009). In particular, Nanotracer response, in terms of particle number concentrations, was compared to the SMPS one measuring diesel-generated aerosol at the IGF (Institut für Gefahrstoffforschung, Bochum; Monz, 2009): a good agreement amongst the two devices was measured. Asbach *et al.* (2012) performed comparisons of different portable diffusion charger counters measuring different aerosols at different concentration levels: the NanoTracers were found in good agreement with freshly calibrated CPCs when soot and NaCl particles were measured, whereas worse metrological performances were detected during di-ethyl-hexyl-sebacate particle measurements. They also recognized different responses amongst the diffusion charger sensors tested, even if, in principle, there is no technical reason why this should happen as they are very similar in terms of measurement principle (Fierz, 2012). To the authors' knowledge no comparisons amongst NanoTracers and laboratory-based instruments were performed in terms of deposited surface area concentrations, even because previous version of the instrument did not provide lung-deposited surface area data.

According to the specifications of the instrument, the NanoTracer is designed to detect particles down to a size of 10 nm. However, the efficiency of the instrument in the size range 10–20 nm is below 100% (Marra *et al.*, 2010). This has serious implications for accuracy when the majority of particles are in this size range, which is often the case during particle formation events in the atmosphere as well as in indoors (Morawska *et al.*, 2008; Hoang *et al.*, 2011). Such particle bursts have regularly been observed in urban environments close to the middle of the day, when gaseous precursors, such as sulphuric acid from motor vehicles, are oxidized by ozone to produce large concentrations of secondary nanoparticles (Morawska *et al.*, 2009). To this purpose we also tested the performances of the NanoTracer in the field applications for ambient aerosols during nucleation events.

MATERIALS AND METHODS

The NanoTracer: Operating Principle

Operation of the NanoTracer is based on the concept of charging particles and then measuring the total particle

charge per unit of air volume. The NanoTracer can operate in two different modes: fast mode and advance mode. In fast mode, it just measures particle number concentrations in real-time (3 s), while in advance mode, both particle concentration and number-averaged particle size are measured. The NanoTracer provide total particle number concentration in the range 10–300 nm when the aerosol under measurements presents a particle distribution mode in the range 20–120 nm (Marra *et al.*, 2010). A controlled airflow, ϕ, passes through a fan situated on top of the sensor, and then the airborne particles are electrically charged by corona discharge from a needle-tip electrode set at a voltage, V_{cor}, high enough to locally ionize the air then inducing diffusion charging of airborne particles through ion attachment. The needle-tip electrode is bounded by a screen electrode, whereby the screen voltage, V_{scr}, is much lower than the electrode (V_{cor}).

When the NanoTracer runs in advance mode, the particles charged enter the precipitation section of the instrument, where a block-shaped voltage pulse is passed between two parallel electrode plates. Particles are subsequently captured on a filter placed inside an electrically isolated Faraday cage, connected via a sensitive current meter to a reference potential. The current meter records the total particle charge deposited per unit time inside the Faraday cage (I_1), then all remaining particles are subsequently captured inside the Faraday cage, leading to a second sensor signal, I_2.

The total particle number concentration (N) and the number-averaged particle size ($d_{p,av}$) are calculated by:

$$N = S_N(I_1 - I_2) \tag{1}$$

$$d_{p,av} = S_{dp} \frac{I_1}{I_1 - I_2} \tag{2}$$

The NanoTracer is also able to yield information on particle surface area concentration per unit volume of inhaled air that is likely to be deposited in various regions of the respiratory tract. This is a noteworthy aspect since deposited surface area concentration is currently of great interest to the scientific community, for use as an important metric for assessing the relative exposure-induced health risk for a given particle hazard (Giechaskiel *et al.*, 2009;

Cauda *et al.*, 2012). In particular, the alveolar deposited surface area concentration, S_{AL} (namely Fuchs surface area, Jung and Kittelson, 2005; it is expressed in $\mu m^2/cm^3$), is calculated by the NanoTracer according to the following equation:

$$S_{AL} \approx 5.4 \times 10^{-2} \cdot N \cdot d_{p,av} \propto I_1 \tag{3}$$

where N is expressed in part./cm^3 and $d_{p,av}$ in μm.

When running in fast mode, the square-wave voltage is not applied to the electrostatic precipitator, and then only the total current from the charged particles is measured. In this case, since the total current depends on the particle number concentration and the particle sizes (Marra *et al.*, 2010), the instrument assumes a mean particle size equal to 50 nm to obtain the number concentration.

These personal monitors are equipped with an internal rechargeable lithium-ion battery, with a total run time (single battery charge) of about 7 hours, which allows for outdoor trips. All the Nanotracers used in the experimental campaigns were calibrated by the manufacturer approximately one year before the experimental analyses, with the exception of the NanoTracer NT3 which was calibrated two months before the tests.

The main characteristics of the NanoTracer are summarized in Table 1 and a more detailed description of the instrument is reported in Marra *et al.* (2010).

Laboratory Experiments
Experimental Set-up

A submicrometer monodisperse aerosol generation system (TSI Inc. 3940) was used to generate 10–1000 nm particles from a solution of NaCl in distilled water. The system includes a filtered air supply (TSI Inc. 3074B), a constant output atomizer (TSI Inc. 3076), a diffusion dryer (TSI Inc. 3062) and an Electrostatic Classifier (TSI Inc. 3080L). In this work, the atomizer was used to generate a mass of 1.2×10^{-4} grams of NaCl per cm^3 of H_2O (0.002 M solution).

The following instruments were used in the laboratory tests: i) a Butanol-Condensation Particle Counters (CPC 3775, TSI Inc.) to monitor total particle number concentration down to 4 nm; ii) a Scanning Mobility Particle Sizer (SMPS 3936, TSI Inc.) spectrometer to measure particle number

Table 1. Main characteristics of the NanoTracer.

Operating modes	**Fast mode:** measures particle concentrations in real-time. **Advanced mode:** measurement of both particle concentration and average particle size. This requires a slightly longer sample time.
Concentration range	0–10^6 part./cm^3
Range of the particle number distribution mode	20–120 nm
Time resolution	**Fast mode:** user-adjustable (min. 3 s) **Advanced mode:** 16 s
Operating conditions	0–35°C, with optimal performance at room temperature between 0–90% relative humidity (non-condensing)
Dimensions (H×W)	16.5 × 9.5 cm
Weight	0.75 kg
Airflow	0.3–0.4 L/min

distributions in the range 0.015–0.700 µm, with a 64 channel per decade size resolution (recently characterized from a metrological point of view by Buonanno *et al.*, (2009b)); iii) a Fast Mobility Particle Sizer[TM] (FMPS 3091, TSI Inc.) spectrometer, with a one-second time resolution, to measure submicrometer particle concentrations and size distributions in the range 5.6–560 nm; and iv) a Nanoparticle Surface Area Monitor (NSAM 3550, TSI Inc.) to measure the human lung-deposited surface area of particles corresponding to the alveolar (A) regions of the lungs.

The experimental set-up used in the laboratory experiments is presented in Fig. 1. Since the NanoTracer uses a fan to draw air samples into the instrument, the device is sensitive to variations of air flow surrounding the air inlet. Therefore, the NanoTracer was not directly connected to the aerosol flow generated by the atomizer, but placed in the buffer volume (16 L). The buffer volume was the shape of a parallelepiped and it was designed to test only one NanoTracer at a time. Given that the NanoTracer collects airborne particles and exhausts clean air at the air outlet, the clean air was channelled in a duct and removed from the buffer volume in order to avoid the dilution effect. A fan was also inserted in the buffer volume to minimize the formation of particle concentration gradients in the buffer volume. The CPC, NSAM, SMPS and FMPS (for the dynamic test) were connected to the buffer volume through an aerosol splitter, in order to sample the tested aerosol.

The experimental set-up was able to generate the required highly stable and uniform particle concentrations in the buffer volume. In particular, the experimental set-up allowed

the user to set the following parameters: i) particle number concentration by means of the submicrometer polydisperse aerosol generation system (TSI 3940); ii) monodisperse aerosol mode through the Electrostatic Classifier; and iii) flow rate at the inlet of the buffer volume.

Methodology

The aerosol was introduced to the buffer volume directly from the NaCl generator (polydisperse aerosol) or after the Electrostatic Classifier (monodisperse aerosol), in order to obtain the required particle concentrations. Tests were performed according to different generated aerosols and dynamics, as follows:

- polydisperse aerosol at low ($< 2 \times 10^4$ part./cm^3), medium (3×10^4–8×10^4 part./cm^3) and high concentrations ($> 12 \times 10^4$ part./cm^3), under steady-state conditions, during which the stability of the particle number concentration was measured;
- monodisperse aerosol (diameter of 50 nm) at low concentrations, under steady-state conditions, during which the stability of the particle number concentration was measured; and
- the dynamic regime, with several high concentration peaks and consequent dilution. This test was carried out in order to simulate typical exposure in urban areas, which is characterized by peaks and consequent decays in particle concentration due to quick dilution (Buonanno *et al.*, 2011a).

Three NanoTracers were tested in the laboratory. Concerning the fixed concentration (polydisperse and
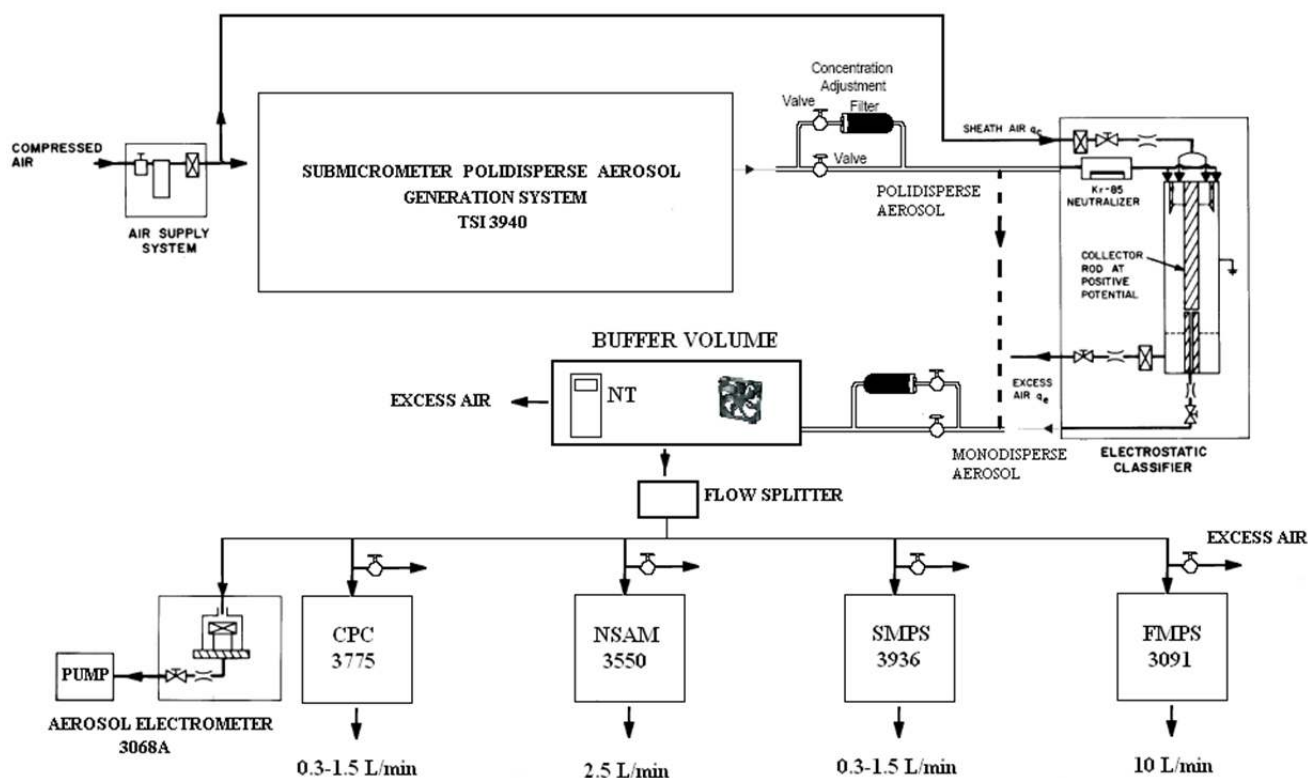
Fig. 1. Scheme of the experimental set-up built up at the European Accreditated Laboratory of the University of Cassino and Southern Lazio.

monodisperse) tests, three tests were conducted for each NanoTracer at a fixed concentration. The time constant of the system (i.e., the time needed by the system to reach 63.2% of its final asymptotic value) was estimated to be equal to about 3 min, and therefore, the following procedure was applied for each test: 10 minutes of measurements were logged after waiting 15 minutes (5 times the constant time), in order to reach a value better than 99% of the final (asymptotic) value.

With regard to the NanoTracer dynamic response testing, particle number evolutions typical of an urban microenvironment were reproduced. In particular, Buonanno *et al.* (2011a) demonstrated that, due to the low ventilation characteristics of a street canyon, peak distribution values need about 15–20 s to decrease to concentration values typical of light duty traffic in that street.

In order to simulate short-term concentrations and dilution in the street canyon, a high concentration aerosol flowrate was introduced to the buffer volume directly from the generator (polydisperse aerosol). The volume was kept closed for a short period, then opened and closed again in order to generate several peaks with a time period of about 20 s. Three tests were conducted for each NanoTracer.

The authors point out that an accurate estimation of short-term exposure is an important issue in personal monitoring. For example, urban microenvironments may significantly contribute to daily total exposure through short-term exposure to elevated particle concentrations. Hence, it is essential to be able to measure particle concentration variations through high-resolution time measurements (Kaur *et al.*, 2009; Buonanno *et al.*, 2011b). In fact, wind and canyon effects influence the ventilation of the street, and hence, the dilution process for aerosols emitted by vehicles. In Buonanno *et al.* (2011a), short-term concentration peaks were measured as heavy duty vehicles (buses) crossed the street. The particle number distribution evolutions presented a constant mode within 30–40 nm, even if the total number concentration suddenly changed as vehicles crossed the measurement point. Consequently, only such a short time resolution allows the instrument to capture the dynamic behaviour of freshly emitted particles.

Ambient Measurements

Three other NanoTracers were placed at each of two outdoor locations and allowed to sample ambient air. Both locations were well away from busy roads, but within the general urban environment of Brisbane, Australia. Measurements were carried out over six hours on each of the six days at each location. The three NanoTracers were placed horizontally on the roof of a mobile trailer, at a height of 2 m above the ground, and lined up longitudinally, as described in the instrument manual, to prevent exhaled air from one instrument being drawn back into a neighbouring instrument. The instruments were operated in the advanced mode, in order to average the particle number concentration and size data at intervals of 16 s. An SMPS, with a TSI 3071 classifier using a TSI 3081 DMA and TSI 3782 CPC, was placed inside the trailer and sampled ambient air drawn through a rubber tube, from a point approximately 60

cm above the roof. Particle size distribution measurements, in the size range 9–414 nm, were continuously logged with a sample scan time of 5 min (plus a 15-s retrace time). All data were corrected for diffusion losses. Correction factors were derived for the particle number concentrations measured by the NanoTracer, and were reported as the average ratio of total particle number concentrations sent by the SMPS to the NanoTracers.

RESULTS

The most important results are calibration correction factors, defined as the ratio between the value measured by the NanoTracer and the reference instrument (CPC, SMPS, NSAM or FMPS), as well as supplementary data as particle number size distribution, stability, etc. In terms of the CPC, which was calibrated by the European Accreditated Laboratory, University of Cassino and Southern Lazio, by comparison with a TSI 3068B Aerosol Electrometer, the accuracy of subsequent measurements was assured through calibration and flow checks conducted at the start of each monitoring period.

NanoTracer Response to Polydisperse and Monodisperse NaCl Aerosol

For polydisperse aerosols, a stability of 2.3%, 3.4% and 4.2% was found at low, medium and high concentrations, respectively. In the case of the monodisperse aerosol, it was equal to 1.8%. For example, in Fig. 2(a), particle number concentrations measured by the CPC during monodisperse and polydisperse (medium concentration) aerosol tests are presented.

Fig. 2(b)–2(d) presents particle number distributions (both for polydisperse and monodisperse aerosols) measured by the SMPS in the buffer volume. For the chosen solution concentration in the atomizer (0.00012 g of NaCl per cm^3 of H_2O; 0.002 M solution), the corresponding mode of the polydisperse particle number distribution was found to range between 39–42 nm.

In Table 2, the calibration correction factors for the three NanoTracers operating in advanced mode are reported for the tested concentrations.

In terms of number concentration, mean calibration correction factor values of 1.25 ± 0.19, 1.00 ± 0.04 and 0.96 ± 0.03 were found for high, medium and low concentrations, respectively. In particular, NanoTracers 1 and 2 show an underestimation at high concentrations.

With regard to the human lung-deposited surface area of particles corresponding to alveolar (A) regions of the lungs, S_A, mean calibration correction factor values of 1.07 ± 0.07, 1.17 ± 0.11 and 0.99 ± 0.04 were found for high, medium and low concentrations, respectively. In light of the NanoTracers consistent performance when measuring this important parameter, Buonanno *et al.* (2012b) confirmed the suitability of this instrument for directly estimating particle dose. Higher differences (within 16%) were found between the NanoTracers and the SMPS when measuring mean particle diameter.

Tests conducted at low concentrations, as was the case

for the monodisperse aerosol (D = 50 nm), showed good agreement between the NanoTracers and the reference instruments, with the exception again of mean diameter. In terms of number and surface area concentrations, the mean calibration correction factor values were found to be 1.02 ± 0.07 and 0.94 ± 0.03, respectively.

Metrological Performance of the NanoTracers in a Dynamic Regime

Fig. 3 shows the particle number distribution trend obtained with the FMPS 1-s measurements during NanoTracer dynamic response testing. The mode of the particle number distribution was set at about 40 nm, in

(a)

(b)
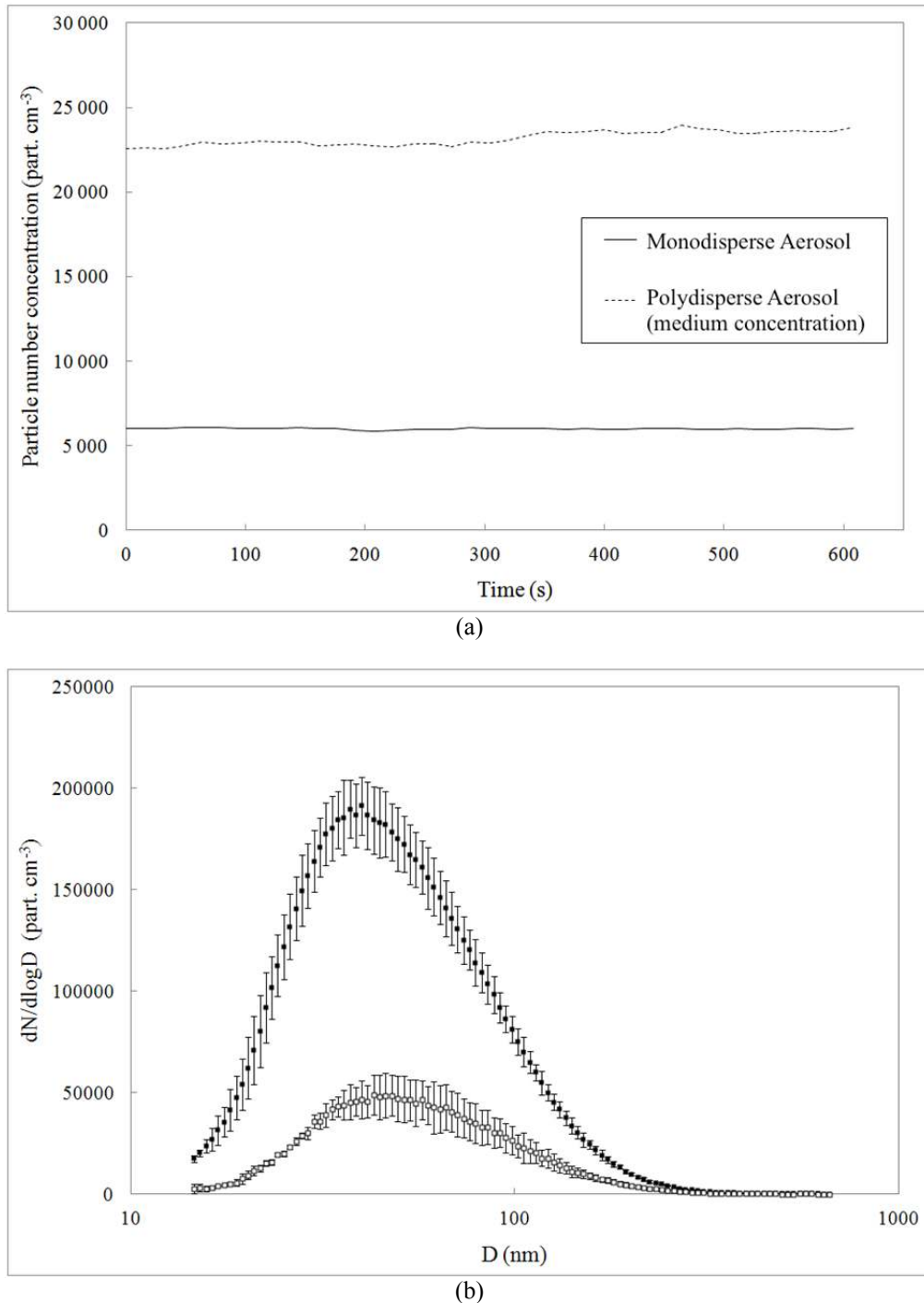
Fig. 2. a) Particle number concentration measured in the buffer volume by the CPC during a monodisperse and polydisperse (medium concentration) during the tests; b) particle number distributions (high and medium concentrations, polydisperse aerosols), c) (monodisperse aerosol) and d) (low concentrations, polydisperse aerosols) measured by the SMPS in the buffer volume. Channel concentration uncertainty values are evaluated through the model reported in Buonanno *et al.* (2009b).

(c)

(d)
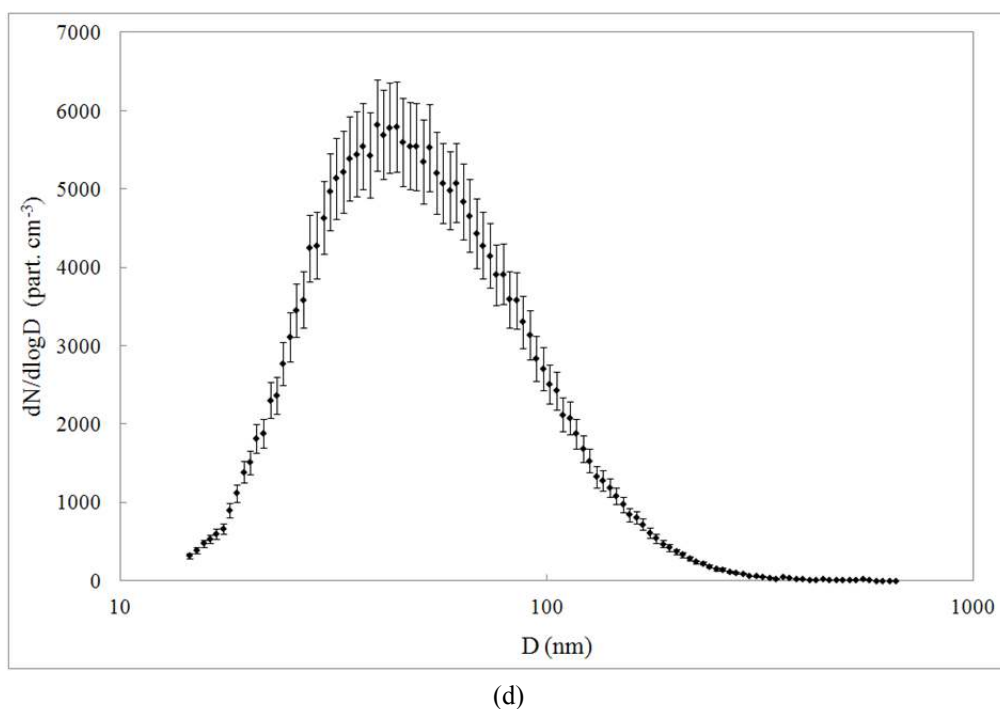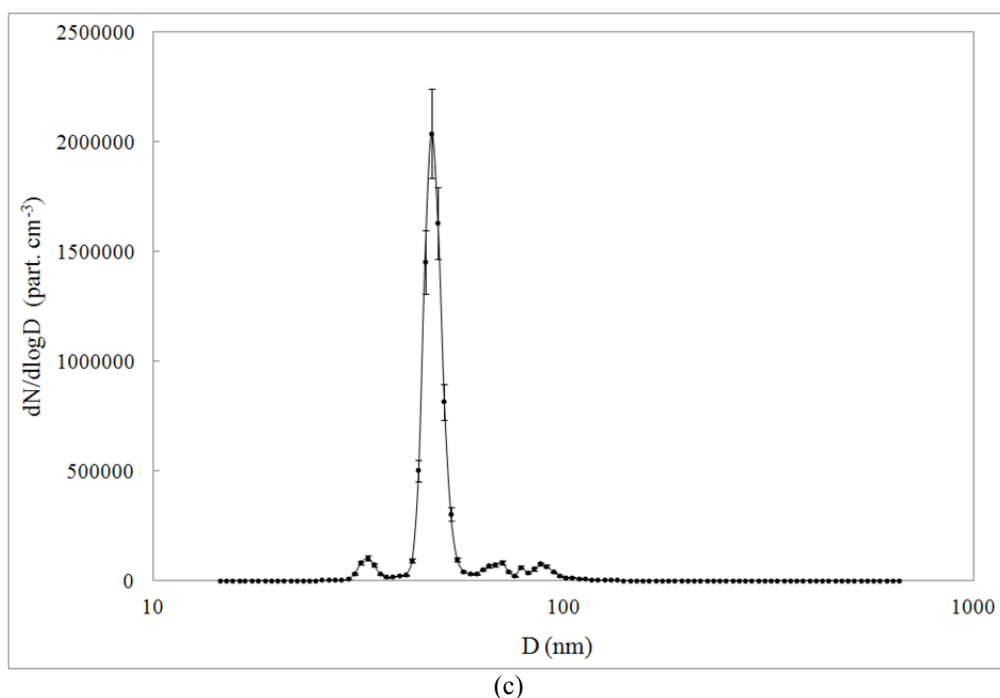
Fig. 2. *(continued).*

order to simulate the short-term exposure in street canyons (Buonanno *et al.*, 2011a). Particle number evolutions with 15–20 s decays were reproduced, as reported in the methodology section.

Table 3 shows the calibration correction factors for the three NanoTracers using a polydisperse NaCl aerosol in the dynamic regime. In this case, the agreement between the NanoTracer and the reference instruments was not so good with respect to the stationary tests, with a strong underestimation of both particle number and alveolar

deposited surface area concentrations. During each test, the sampling time of the reference instruments (CPC and NSAM) was equal to the sampling time of the NanoTracers, for both fast and advanced modes (equal to about 3 and 16 s, respectively).

In terms of number concentration, mean calibration correction factor values of 2.33 ± 1.09 and 2.31 ± 0.76 were found for the fast and advanced modes, respectively.

With regard to the human lung-deposited surface area of particles corresponding to alveolar regions of the lungs,

Table 2. Calibration correction factors in terms of number concentration (N), alveolar-deposited surface area (S_A) and distribution mode (D) for the three NanoTracers operating in advanced mode when testing a polydisperse and moon disperse NaCl aerosol.

Polydisperse aerosol				
		NT1	NT2	NT3
High concentration	N	1.37	1.35	1.03
	S_A	1.15	1.01	1.06
	D	0.78	0.84	0.98
Medium concentration	N	0.98	1.04	0.97
	S_A	1.22	1.25	1.04
	D	0.78	0.86	0.89
Low concentration	N	0.93	0.97	0.98
	S_A	1.04	0.96	0.98
	D	1.22	1.10	1.09
Monodisperse aerosol (50 nm)				
		NT1	NT2	NT3
Low concentration	N	1.05	1.08	0.94
	S_A	0.98	0.92	0.93
	D	0.54	0.69	0.79

mean calibration correction factor values of 2.98 ± 1.36 and 3.00 ± 1.64 were found for the fast and advanced modes, respectively. Consequently, the underestimation by the NanoTracers, highlighted in Fig. 4, could represent a significant difficulty when using these kinds of instruments for personal monitoring. Therefore, NanoTracers could have worse metrological performances with respect to other diffusion charger monitors in terms of alveolar-deposited surface area measurement: in fact, Asbach *et al.* (2012) found good correlation between AeroTrak (TSI Inc. 9000) and MiniDiSC alveolar-deposited surface area measurements and data obtained from FMPS measurements. The authors point out that the correction factors provided for NanoTracer running in fast mode are strictly dependent on the particle size distribution of the aerosol under measurement. This is due to the fact that during fast mode operation the NanoTracer is not able to measure the mean diameter as it assumes a fixed mode of 50 nm.

Ambient Measurements and the Effect of Nucleation

Typically, particle number size distributions obtained by the SMPS showed a modal diameter in the range 20–40 nm, which is representative of an urban background dominated by motor vehicle emissions. Fig. 5 shows a typical example where the count median diameter (CMD) of the particles was 28 nm. The particle number concentration also followed the typical diurnal bimodal distribution, with maximum values corresponding with the morning and late-afternoon peak traffic times. However, a different pattern was observed on three of the twelve days, all at the same location, where the particle number concentration increased by approximately an order of magnitude during the middle of the day. These events occur due to secondary particle
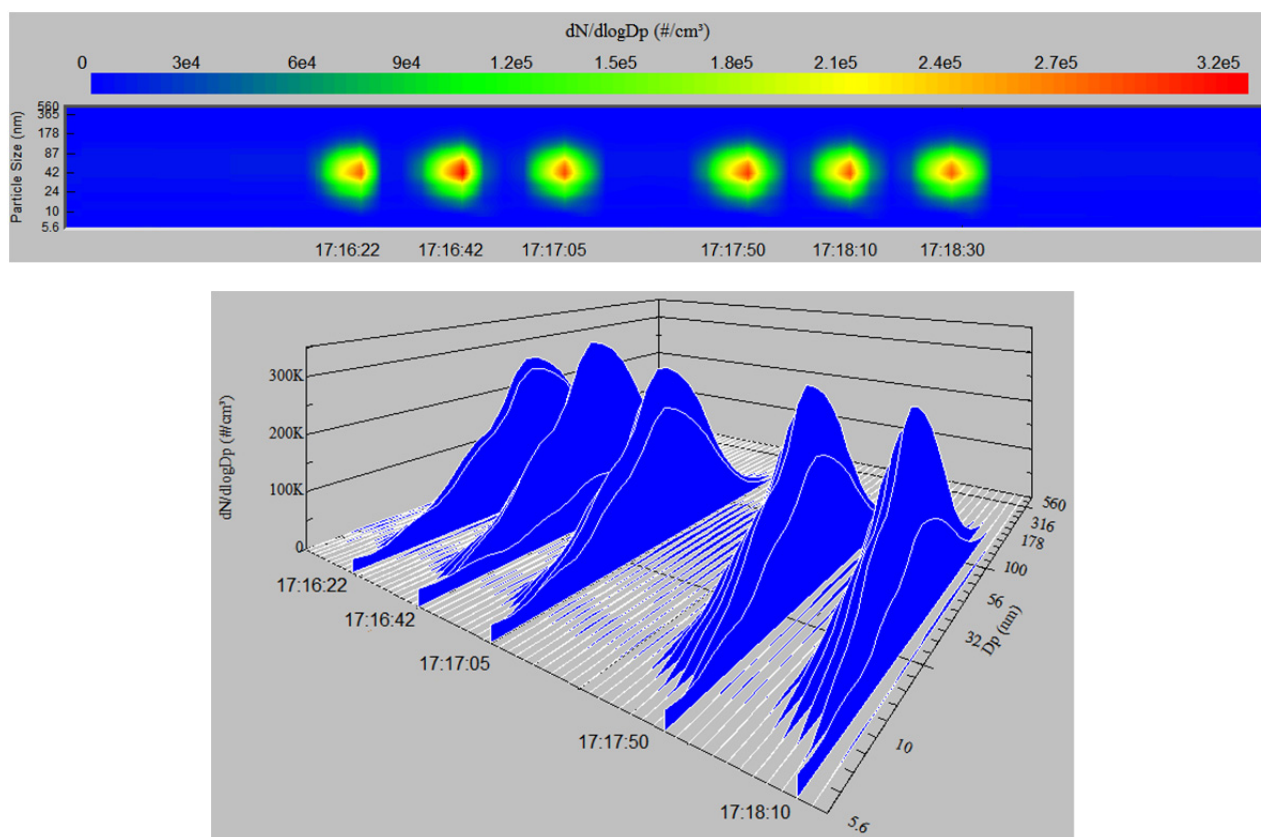
Fig. 3. Particle number distribution evolution obtained with using a reproduction of the FMPS 1-s measurements, to test the dynamic response of the NanoTracer.

Table 3. Calibration correction factors in terms of number concentration (N) and alveolar-deposited surface area (S_A) for the three NanoTracers using a polydisperse NaCl aerosol during dynamic response testing.

|		NT 1	NT 2	NT 3
		Dynamic regime		
Fast mode	N	3.57	1.85	1.56
	S_A	4.55	2.13	2.27
Advanced mode	N	3.13	2.17	1.64
	S_A	4.87	2.33	1.79

formation from the photo-oxidation of gaseous precursors in the atmosphere and are generally observed during the middle of the day when solar radiation is at a maximum.

Fig. 6 shows a typical particle size distribution from the SMPS on one of these three days. Note that the CMD at this time was much smaller than on the day shown in Fig. 5. Note also that the particle number concentrations in Fig. 6 are about an order of magnitude higher than in Fig. 5.

Comparison of the performance of the three NanoTracers against the SMPS on normal days and on days when there was particle formation observed is of interest in this study. It was noted that each of the three particle formation events lasted from 3 to 4 hours. These data (nucleation) were separated from the rest of the data (normal). Under normal conditions, the mean total particle number concentration reported by the SMPS was 5.34×10^3 part./cm^3, whereas for the three NanoTracers they were 6.94×10^3, 5.92×10^3 and 8.64×10^3 part./cm^3, giving correction factors of 0.77,
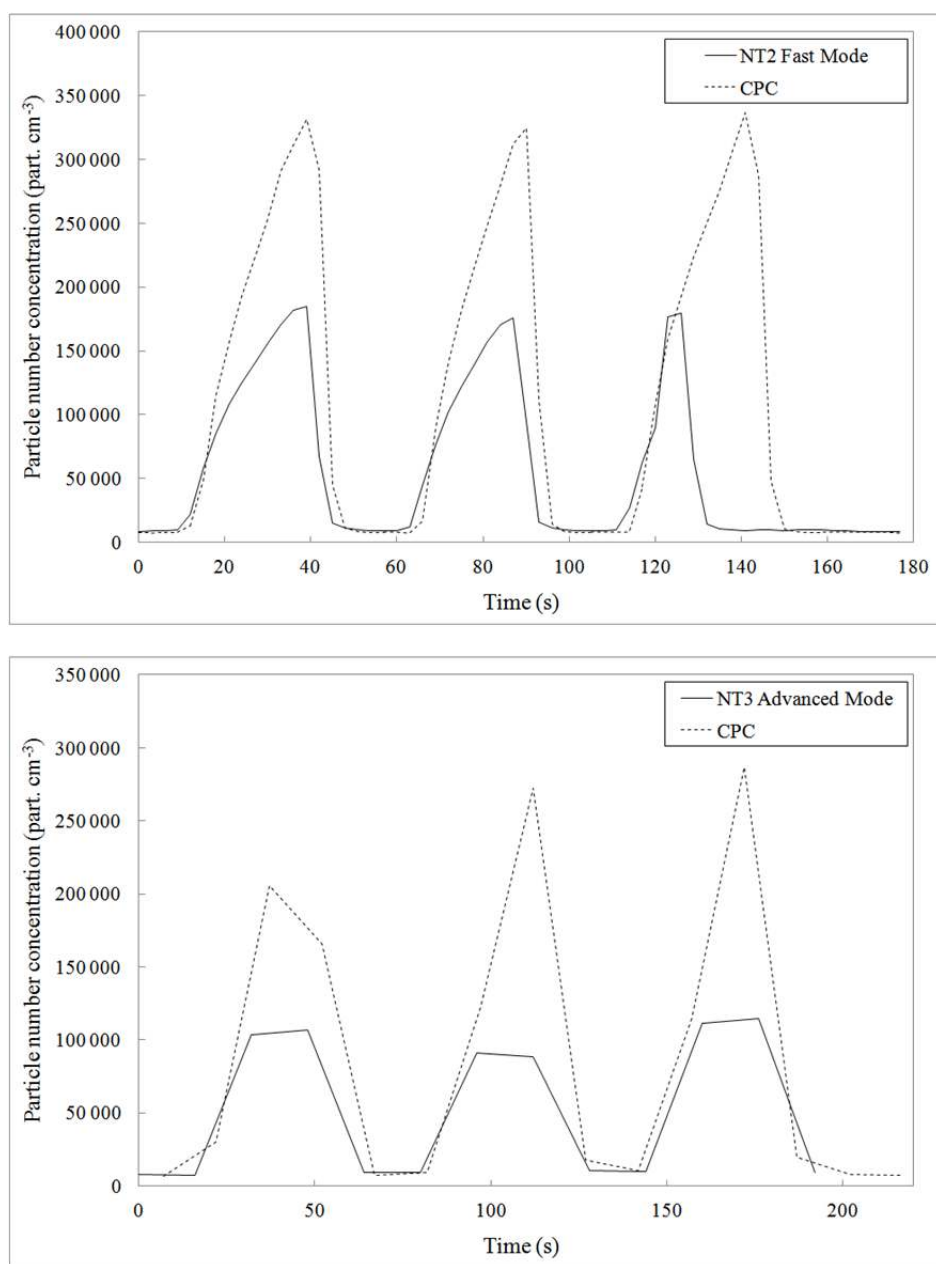
Fig. 4. Particle number concentrations measured by the CPC and NanoTracers during dynamic response testing.
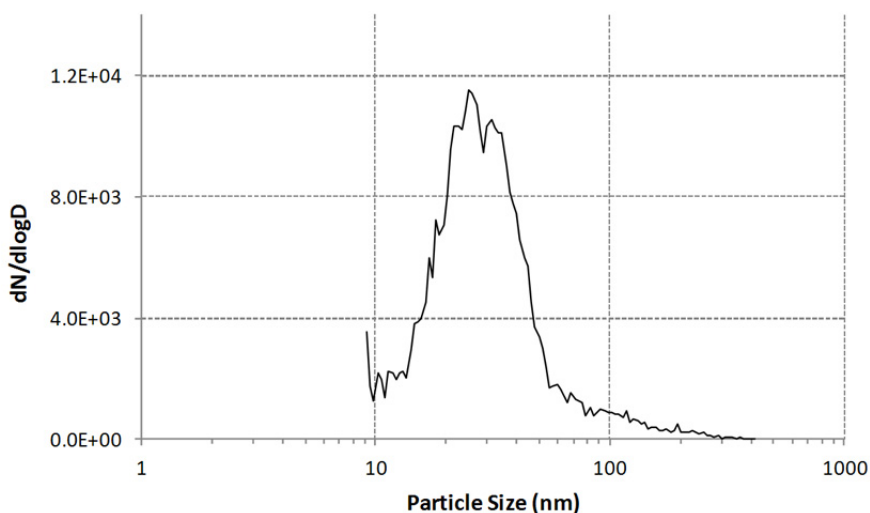
Fig. 5. Particle size distribution from the SMPS on a day when there was no particle formation observed.
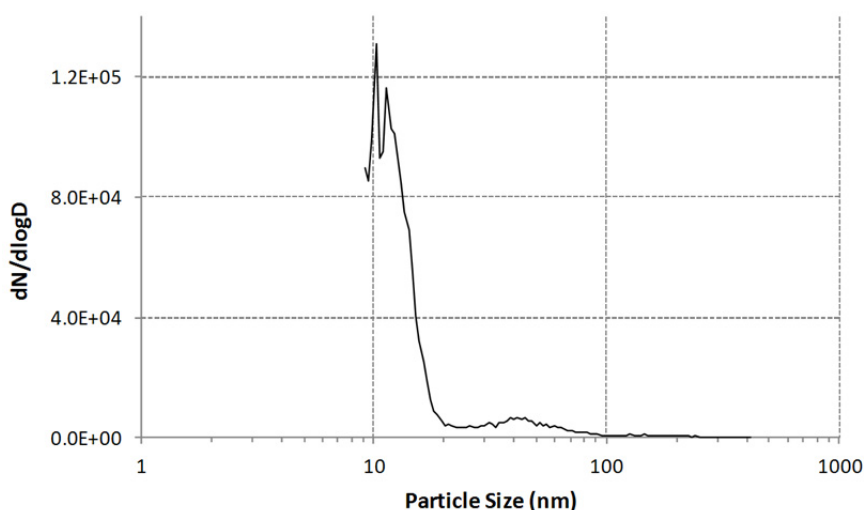
Fig. 6. Particle size distribution from the SMPS at a time when there was a clear particle formation event observed.

0.90 and 0.62, respectively. During the periods when particle formation was observed, the total particle number concentration reported by the SMPS was 2.54×10^4 cm^{-3}, whereas for the three NanoTracers, they were 8.09×10^3, 8.42×10^3 and 12.87×10^3 cm^{-3}, giving correction factors of 3.14, 3.02 and 1.97, respectively. Clearly, the NanoTracers were underestimating number concentration, despite the clear observation from Fig. 6 that the SMPS was not detecting a substantial number of particles that were present at sizes smaller than its lower detectable limit of 9 nm. The vast majority of particles, in this case over 85%, occurred in the size range 10–20 nm, the range where the NanoTracer was not rated at 100% efficiency (Marra *et al.*, 2010; Asbach *et al.*, 2012) and for which no previous metrological characterizations were performed. The average correction factor for the three NanoTracers, over all periods with no particle formation, was 1.1 ± 0.4, while for the three periods with particle formation, it was 1.9 ± 0.3. As above mentioned, the fast mode correction factors are exclusively characteristics of the given aerosol distribution.

CONCLUSIONS

Air pollution is normally monitored through a number of fixed sampling points, chosen as a function of the population size, without any link to microclimatic characteristics. This kind of monitoring data cannot be properly used to accurately characterize real exposure in outdoor or indoor microenvironments, meaning that in-depth studies aimed at monitoring individual exposures should be carried out in order to increase confidence in epidemiologic relationship between health outcomes and ambient concentrations. This work thoroughly analysed, from a metrological point of view, 6 ultrafine particle counters (Philips Aerasense NanoTracers) recently commercialized for individual and personal monitoring, both in the laboratory and in the field. Three main aerosol conditions were analysed according to the following regimes: stationary, dynamic and the presence of nucleation.

Under stationary conditions, particle number concentration measured by the NanoTracers showed good agreement

($\pm10\%$) with respect to calibrated condensation particle counters (CPC), and $\pm20\%$ agreement with regard to alveolar deposited surface area concentration and average particle diameter when compared to a Scanning Mobility Particle Sizer (SMPS) and a Nanoparticle Surface Area Monitor (NSAM). The agreement decreases at very high concentrations ($> 1.5 \times 10^5$ part./cm^3).

In the dynamic regime, designed to simulate the exposure conditions in street canyons, the NanoTracers were found to significantly underestimate particle number and alveolar deposited surface area concentrations in both fast and advanced modes.

Finally, with regard to the field tests, the NanoTracer did not capture a significant number of particles in the size range below 20 nm, and therefore, results obtained under nucleation conditions should be treated with caution since particle number concentration are systematically underestimated.

In summary, the NanoTracer has advantages as an instrument for the personal monitoring of exposure to ultrafine particle, however it also has limitations which need to be considered in order to provide meaningful results.

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