Atmospheric Environment 123 (2015) 220–228

Contents lists available at ScienceDirect

Atmospheric Environment

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

Field comparison of portable and stationary instruments for outdoor urban air exposure assessments

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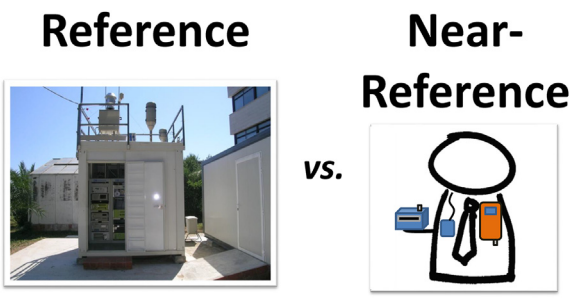
HIGHLIGHTS

- Epidemiological studies highlight the need for exposure assessment studies.
- Portable and stationary monitors were compared for exposure assessment.
- Good agreements were found: R^2 mostly >0.80; relative differences <20%.
- Relative differences were <10% between different units of the same instrument.
- Parameters assessed were BC, N, LDSA and mean particle diameter, in outdoor air.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 2 August 2015
Received in revised form
27 October 2015
Accepted 27 October 2015
Available online 31 October 2015

Keywords:
Intercomparison
Relative error
Uncertainty
Performance
Outdoor
Indoor

ABSTRACT

The performance of three portable monitors (micro-aethalometer AE51, DiscMini, Dusttrak DRX) was assessed for outdoor air exposure assessment in a representative Southern European urban environment. The parameters evaluated were black carbon, particle number concentration, alveolar lung-deposited surface area, mean particle diameter, PM_{10}, $PM_{2.5}$ and PM_1. The performance was tested by comparison with widely used stationary instruments (MAAP, CPC, SMPS, NSAM, GRIMM aerosol spectrometer). Results evidenced a good agreement between most portable and stationary instruments, with R^2 values mostly >0.80. Relative differences between portable and stationary instruments were mostly <20%, and <10% between different units of the same instrument. The only exception was found for the Dusttrak DRX measurements, for which occasional concentration jumps in the time series were detected. Our results validate the performance of the black carbon, particle number concentration, particle surface area and mean particle diameter monitors as indicative instruments (tier 2) for outdoor air exposure assessment studies.

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http://dx.doi.org/10.1016/j.atmosenv.2015.10.076

1. Introduction

Numerous studies have proven the link between air pollution and health (Lim et al., 2012), with outdoor air pollution being classified as carcinogenic to humans by the International Agency for Research on Cancer (IARC, 2013). According to recent studies, adverse effects derived from exposure to air pollution are being observed at ever-lower concentrations of pollutants (Brunekreef et al., 2015; WHO, 2013a, 2013b). In Europe, fine particles ($PM_{2.5}$) in the air are the most important environmental health concern among the major drivers of ill health and premature mortality in the population (Lim et al., 2012).

Degraded air quality is an environmental and health issue affecting citizens around the globe, most of them in urban areas, where approximately 75% of the European population lives (EEA, 2014). To protect the population, Directive 2008/50/CE set the guidelines for monitoring atmospheric pollutants across the EU Member States, as well as the reference methods by which this should be achieved. However, a substantial percentage of the European urban population is still exposed to air pollutant levels exceeding the WHO Air Quality guidelines and the EU Air Quality standards (EEA, 2014). Recent research recommends that current air quality monitoring networks should have a stronger link to health (AirMonTech, 2013; Gao et al., 2015; Snyder et al., 2013; Steinle et al., 2013), which could be achieved by monitoring population exposure. The need for exposure monitoring has been evidenced by numerous works (Buonanno et al., 2011; De Nazelle et al., 2013; Gehring et al., 2013; Gu et al., 2015; Karanasiou et al., 2014; Kaur et al., 2007; Morawska et al., 2013). In addition, current trends in the US suggest that present-day sophisticated ambient air pollution monitoring technology is not economically sustainable as the sole approach and cannot keep up with current needs (Snyder et al., 2013; US-EPA, 2013). Portable monitors and sensors are currently being developed to enable a move towards exposure monitoring (as opposed to background concentration monitoring). One added value of these monitors, because of their smaller size and weight when compared to reference monitors, is that they may also be used in indoor air, e.g. for occupational exposure assessments.

US-EPA advocates a tiered system for different types of air monitors based on cost, application, and end user (US-EPA, 2013): near-reference, monitors/sensors intended for indicative use, and mainly qualitative sensors intended for educational use. The performance of each of these three types of portable monitors must however be tested against reference instrumentation, or against the most widely used instruments in the case of unregulated parameters (e.g., ultrafine particle number concentration, particle surface area) for which no reference is available. The present work focuses on near-reference instruments in indoor and outdoor air applications, with the aim to test their performance against widely used stationary instruments. The use of this kind of instruments is recommended in tier 2 exposure assessment studies within NEAT (nanotechnology emission assessment technique; Methner et al., 2010). As described by Asbach et al. (2012) for workplace air assessments, instruments for tier 2 need to be easy to use, battery operated, portable, and able to deliver a limited but meaningful data set to estimate exposure levels. This is also the case for indoor and outdoor air exposure assessments, which are the focus of this work. The performance of portable monitors for indicative use is assessed for particle number concentration (N), mean particle diameter, lung-deposited particle surface area (LDSA), black carbon (BC) concentration, and particle mass concentration (PM_{10}, $PM_{2.5}$, PM_1). The final goal is to evaluate whether the portable instruments under study are comparable to their reference (or widely accepted) stationary counterparts, for outdoor and indoor air quality studies.

Previous studies have presented comparisons for certain instruments and parameters (Asbach et al., 2012; Fierz et al., 2011; Mills, 2013; Stabile et al., 2014; Tasić et al., 2012). However, most of them tested the instruments under laboratory conditions and challenging the instruments with purposely-generated aerosol types (e.g., NaCl, soot, etc.). In the present work, stationary and portable instruments are compared under real-world operating conditions, measuring ambient urban aerosol and under changing meteorological scenarios, with the aim to assess the performance of portable monitors under the most representative conditions for urban outdoor air monitoring. If their performance is validated, these lower-cost near-reference monitors could be proposed as viable addition to existing air quality monitoring networks, to achieve a broader spatial coverage and a more representative characterisation of population exposure.

2. Materials and methods

All instruments described below were simultaneously co-located inside the air quality monitoring station at Palau Reial at IDAEA-CSIC located in an urban background area in Barcelona (Spain), connected to an inlet or with the sampling tubes through a window to sample outdoor air. Sampling tubes were kept to a minimum to minimise diffusion losses. The portable instruments were placed on a table, with sufficient distance to each other to avoid interferences, and sampled at approximately the same height. The clocks of all instruments were synchronized prior to the first measurement in each intercomparison. The inlets of the stationary instruments for comparison were located within a 1.5 m radius of those of the portable instruments, and at the same height above ground.

The following air quality parameters were evaluated:

- Black carbon (BC) concentration was measured with six Microaeth AE51 (Aethlabs) instruments, and compared with a Thermo Multi-Angle Absorption Photometer (Carusso MAAP). Five of the AE51 instruments were identical, and unit BC5 was a prior version (Magee). The AE51 instruments were operated at a flow of 100 ml m^{-1}, and filter tickets were exchanged every 24 h. No cyclone was used at the inlet. The MAAP instrument was connected directly to outdoor air, with a heated inlet (Müller et al., 2011).
- Particle number concentration (N) was measured with five identical DiscMini (Testo; Fierz et al., 2011) instruments in the range 10–700 nm, and compared with a water condensation particle counter TSI CPC3785 (5–1000 nm). The DiscMini instruments were connected each to an impactor with a cutoff at 700 nm to prevent interference with coarse particles. Anti-static tubing was used during all intercomparison exercises (see details below). The CPC was only available for 2 out of the 4 intercomparison exercises.
- Mean particle diameter was measured with five identical DiscMini (Testo) instruments in the range 10–700 nm, and compared with a scanning mobility particle sizer SMPS system (TSI 3936). The SMPS was comprised of an Electrostatic Classifier (TSI 3080) and a Differential Mobility Analyser (DMA, TSI 3081), connected to a condensation particle counter (CPC TSI 3772). The SMPS provided particle number size distributions between 10.9 and 478.3 nm (N10.9–478.3) in 64 channels/decade, and completed two scans every 5 min. Measurements were corrected for multiple charge and diffusion losses within the system.

- Lung-deposited particle surface area (LDSA) in the alveolar fraction was measured with five identical DiscMini (Testo) instruments in the range 10–700 nm, and compared with a TSI Nanoparticle Surface Area Monitor (NSAM; size range 20–400 nm). As described by (Todea et al., 2015), despite the wider size ranges specified by the manufacturers, LDSA concentrations can only accurately be measured in a size range from 20 to 400 nm. The NSAM inlet was connected to a Nafion dryer. Anti-static tubing was used at all times.
- Particle mass concentration (PM_{10}, $PM_{2.5}$, PM_1) was monitored with five identical TSI Dusttrak DRX instruments, and compared with an aerosol spectrometer Grimm 180 previously calibrated with onsite EU-reference high-volume samplers with PM_{10} and $PM_{2.5}$ cutoff inlets. The PM_1 size fraction was also corrected with regard to a high-volume (non-reference) PM_1 sampler. PM_{10} and $PM_{2.5}$ are the only parameters for which an equivalent to an EU reference instrument is available.

For the purpose of this work the data originating from the stationary instruments (MAAP, CPC, SMPS, GRIMM, NSAM) are considered as internal references given that they are widely used instruments and given that most of the parameters measured are unregulated.

Details of the flows and averaging times of the instruments are reported in the Tables in Supporting Information. Details of the operating principles of the different instruments are not provided in this work.

In total, four intercomparisons were carried out for each type of instrument, prior and posterior to two sampling campaigns (SC) taking place in the framework of the ERC Advanced Grant BREATHE. Thus, the intercomparisons were named pre_ and post_SC1, and pre_ and post_SC2. Each intercomparison had a duration between 2 and 4 days, with a time resolution of 5–10 min depending on the instrument (see Tables in Supporting Information). The 2–3 month periods between each intercomparison allowed identifying potential drifts in the portable instruments. As a result, for each parameter (e.g., BC), the dataset available consisted of 4 sets of 2–4 day intercomparisons between 5 and 6 units of the same instrument (in this example, Microaeth AE51) and one unit of the reference instrument (in this example, MAAP).

Data were processed using a standard spreadsheet application. Negative values were only removed when they coincided with necessary instrument operations (e.g., change of filter ticket in AE51). Specific datapoints were removed when technical errors were reported by the instruments. Data availability is reported in the Results section. The DiscMini units DM2, DM3 and DM4, Dusttrak units DST1, DST2, DST3 and DST4, and Microaeth units BC3 and BC5, were serviced between intercomparisons post_SC1 and pre_SC2.

3. Results and discussion

3.1. Black carbon (BC)

BC concentrations obtained with MAAP were corrected for this work with regard to locally determined EC concentrations (Reche et al., 2011), and should thus be considered as representing equivalent black carbon (EBC) concentrations (Petzold et al., 2013). This, however, only impacts the comparability between the stationary and portable monitors regarding the slope of the regression curve. The technical performance of the portable (AE51) and stationary (MAAP) instruments is summarised in Table S1. Throughout the intercomparisons, the performance of the 6 AE51 units was mostly robust, with data availability >98% for most of the units with the exception of 2 (BC5 and BC3) which failed during two comparison exercises and one unit (BC1) which failed during one exercise. Data availability for MAAP was >90% with only one exception (65% during pre_SC2). Data losses for all instruments were due to specific instrument failures. The AE51 instruments required no periodic maintenance during the intercomparison periods. Filter tickets were exchanged every 24 h to avoid filter loading effects on BC measurements, based on previous tests carried out at the same monitoring station to determine the maximum monitoring duration before changing the filter ticket.

The results from comparing portable and stationary instruments for BC showed a relatively good agreement ($R^2 > 0.75$) which was mostly stable across instruments and intercomparison exercises (Table 1). Ambient air BC concentrations measured were representative of urban environments, ranging between 1529 and 3204 ng m^{-3} (mean standard deviation 1382 ng m^{-3}) (Reche et al., 2011). Coefficients of determination (R^2) between each portable and the stationary instrument ranged typically between 0.75 and 0.85 and were dependent on the exercise, e.g., all units showed similar R^2 values in each intercomparison. The same was true for the slope of the regression equation (ranging between 0.75 and 0.90 in three of the exercises, and between 0.97 and 1.15 in the fourth). This similarity in the performance of the different units is especially relevant for their application in indoor air and exposure studies (Asbach et al., 2012bib_Asbach_and_Kaminski_2012) when two or more units of the same instrument are used to simultaneously monitor background aerosols and concentrations near the process of interest (tiered approach; Methner et al., 2010; OECD, 2015). This applies especially when aiming to compare concentrations between different exposure scenarios, as opposed to assessing compliance with a certain limit value (which is not the objective of this kind of portable instruments). On average, the relative differences between each of the portable units and the stationary instrument ranged between 7% and 11% in terms of black carbon mass concentrations. One example of comparability is shown in Fig. S1, where the specific performance of unit BC4 was tested against the internal reference during pre-SC2 and post_SC2. The Figure shows the absence of drifts between intercomparison exercises (2–4 months apart, as stated above) regarding the performance of the AE51 units (R^2 and slope of the equation), which was also observed for the other units. All of the units (except for BC5, with technical problems during post_SC1 and pre_SC2) performed similarly within each campaign, thus suggesting that at least part of the variability in the comparison with the MAAP was linked to the variability of the stationary instrument data.

According to the instrument's specifications, the measurement range of the AE51 covers between 0 and 1 mgBC·m^{-3}, a broad spectrum including typical urban indoor and outdoor air concentrations. For the concentration range monitored in the present work, mean relative differences between BC from portable and stationary instruments were calculated and are shown in Fig. 1, for 5-min averages. During three of the four intercomparisons the portable instruments overestimated the MAAP BC concentrations on average by <10% in terms of mass. This trend was reversed during post_SC1, when all instruments underestimated BC concentrations and by slightly more than 10%. Unit B6 showed a different performance to the rest of the units, as it underestimated BC concentrations on most occasions. Unit BC5 suffered from instrument failures during two exercises, probably due to the fact that it was an older version of the instrument. With the aim to provide a quantitative estimate of the AE51 uncertainty for the time resolution and the BC concentration range measured, the absolute value of the relative differences with regard to MAAP was calculated (in %) for each unit, and these values were averaged across intercomparison exercises. This was considered as the mean uncertainty of the AE51 instruments, in absolute value (Abs(Mean)).

Table 1
Correlation coefficients and regression equations obtained from the comparison between portable (AE51) and stationary (MAAP) instruments for black carbon (BC) monitoring.

	Pre_SC1	Post_SC1	Pre_SC2	Post_SC2
BC1	Y = 1.15x-239 R^2 = 0.87	Y = 0.80x+254 R^2 = 0.77	Y = 0.92x+258 R^2 = 0.86	Y = 0.80x+355 R^2 = 0.75
BC2	Y = 1.14x-222 R^2 = 0.87	Y = 0.70x+384 R^2 = 0.70	Y = 0.93x+260 R^2 = 0.85	Y = 0.86x+317 R^2 = 0.76
BC3	Y = 1.16x-248 R^2 = 0.87	Y = 0.93x+205 R^2 = 0.79	Y = 0.85x+313 R^2 = 0.89	Y = 0.85x+346 R^2 = 0.75
BC4	Y = 1.13x-204 R^2 = 0.87	Y = 0.72x+321 R^2 = 0.76	Y = 0.89x+272 R^2 = 0.86	Y = 0.74x+378 R^2 = 0.76
BC5	Y = 0.97x-108 R^2 = 0.85	Y = 0.27x+240 R^2 = 0.15	Y = 0.10x+51 R^2 = 0.10	Y = 0.72x+313 R^2 = 0.76
BC6	Y = 1.11x-82 R^2 = 0.82	Y = 0.81x+154 R^2 = 0.79	Y = 0.79x+183 R^2 = 0.83	Y = 0.60x+169 R^2 = 0.76
Time resolution	5-min	5-min	5-min	5-min

Thus, when compared to the stationary MAAP monitor (internal reference), the mean uncertainty for the portable BC monitors ranged between 7 and 12% of the BC mass (in $\mu g \cdot m^{-3}$), for measurements collected with a 5-min time resolution and measuring representative urban aerosol concentrations (Fig. 1).

3.2. Particle number concentration (N)

The technical performance of the portable (DiscMini, DM) and stationary (CPC) instruments is summarised in Table S2. With only two exceptions (DM2 and DM3) during one intercomparison (post_SC1), the performance of the DiscMini units may be considered optimal, with >99% data availability across intercomparisons. The DiscMinis were thus rather robust regarding data capture during this work. CPC data were only available during intercomparisons post_SC1 and pre_SC2. Regarding the DiscMini units, they required inlet cleaning every 24 h to ensure the proper cutoff by the impactor. It should be stated that during preliminary tests several DM units showed technical failures (error message "high voltage", appearing after approximately 4 h of operation, and after which the instruments had to be manually stopped and re-started) which were finally linked to the use of black silicone tubing (TSI, ¼ inch inner diameter, approximately 30 cm in length). When this tubing was replaced by Tygon (Tygon, ¼ inch inner diameter, approximately 30 cm in length), the failures stopped. As also observed by (Asbach et al., 2015), corona voltage increases and LDSA concentration decreases as conductive silicone tubes get attached, and this was interpreted as the cause of the technical failures observed during this work. As a result, the tubing used with DM instruments was identified as a key parameter for their operation.

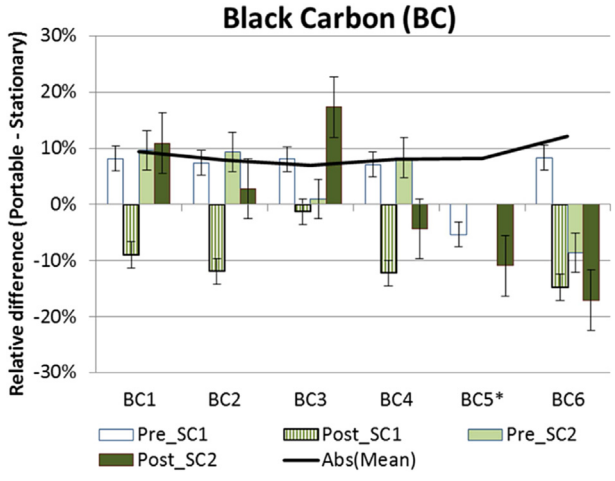

Fig. 1. Relative differences for black carbon concentrations between the portable and stationary instruments for each unit and intercomparison exercise, calculated as 100*(Portable-Stationary)/Stationary. Abs(Mean): average across intercomparison exercises of the absolute value of the differences with regard to MAAP (in %) for each unit. * Unit 5 suffered technical failures during two intercomparison exercises.

Minimum and maximum particle number concentrations (N) measured by the CPC were 9214 $cm^{-3}$and 11,946 cm^{-3}, respectively (means for the respective post_SC1 and pre_SC2 exercises, based on 10-min time resolution). The mean standard deviation was 5182 cm^{-3}. As in the case of BC, these concentrations may be considered representative of the Barcelona urban background (Reche et al., 2011), and within the measurement range of the DiscMini and CPC3785 (10^3-10^6 cm^{-3} and 10^3–$2.5*10^5$ cm^{-3}, respectively). It should be noted that the CPC3785 switches from single-count mode to photometric mode at a concentration of 30.000 cm^{-3}, when the instrument's response becomes highly material-dependent as the eventual droplet size depends on the hygroscopicity of the particles measured. As a result, measurements carried out with the instrument in the photometric and single-count mode would not be directly comparable with each other nor with the DiscMini data. During the study periods <1% of the mean 10-min concentrations measured with the CPC3785 were above the 30.000 cm^{-3} threshold, and therefore this limitation is not expected to impact the results presented in this work.

The comparison between portable and stationary instruments for N showed a good agreement between instruments with correlation coefficients $R^2 > 0.82$ during most intercomparisons (only 2 exceptions, DM3 and DM5 during post_SC1 with $R^2 = 0.72$–0.75). As observed for BC, no drifts in performance were detected over time regarding the R^2 values, and the comparability between instruments seemed to be exercise dependent (with all units obtaining similar R^2 and slope values for each single intercomparison exercise). The slope of the regression equations showed limited variability ranging between 0.90 and 1.8 during the intercomparisons available (post_SC1 and pre_SC2). As stated for BC, the similarity in the performance of the different units is especially relevant for their application in studies comparing different exposure scenarios (Asbach et al., 2012). An example of intra-unit comparability is shown in Fig. S2.

Mean relative differences between N from portable and stationary instruments are shown in Fig. 2. As shown in the Figure, N concentrations measured by the portable instruments were lower and higher than those reported by the CPC, irrespective of the different size ranges measured by both instruments (5–1000 nm by CPC, 10–700 nm by DiscMini): during post_SC1 the stationary instrument reported higher N concentrations, whereas during

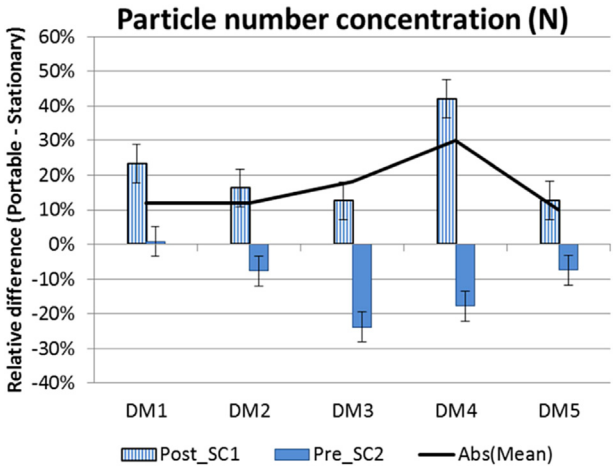

Fig. 2. Relative differences for particle number concentrations between the portable and stationary instruments for each unit and intercomparison exercise, calculated as 100*(Portable-Stationary)/Stationary. Abs(Mean): average across intercomparison exercises of the absolute value of the differences with regard to CPC (in %) for each unit. Data from the CPC were only available for 2 intercomparison exercises.

pre_SC2 the portable instruments measured higher values. This may seem an unusual result, given that higher N concentrations could have been expected to have been measured by the CPC due to the lower particle diameters measured (5 nm as opposed to 10 nm with DiscMini). However, the different DM responses observed when compared to the CPC (Fig. 2) are probably ascribed to varying shapes and geometric standard deviation of the size distributions of the urban aerosol between the different sampling campaigns. The DM is calibrated with monodisperse aerosol, and the instrument response for polydisperse aerosols with a lognormal size distribution with a geometric standard deviation $\sigma = 1.9$ is then calculated (Fierz, 2010). As a result, for aerosols that do not match this size distribution, errors occur both in the number and diameter calculation. The relative difference between the portable and the stationary instruments ranged mostly between 1% and 24%, with one outlier (42% for DM4 during post_SC1). During this work, the mean uncertainty of the DiscMini instruments (calculated as the Abs(Mean), in %) ranged between 10 and 30% (between 10 and 18% if the outlier from DM4 in post_SC1 is excluded) for measurements carried out in outdoor urban air (Fig. 2). These results are well within the instruments' specified accuracy range of ±20–30% (Asbach et al., 2012; Fierz et al., 2011).

3.3. Lung-deposited particle surface area (LDSA)

The same portable instrument as above (DiscMini) was used to quantify alveolar lung-deposited surface area concentrations, and compared to the commercial NSAM monitor. The performance and data availability for the portable instruments are thus the same as in the previous section, and summarised in Table S2. The performance of the NSAM monitor was very robust, with data availability >89% in all the intercomparison exercises (Table S2). Results below (Table 3) suggest a technical failure occurred with the NSAM during pre_SC1 due to its different comparability with regard to the portable instruments in all of the other campaigns, although no specific issues were detected aside from relatively lower (but still good) data availability. The mean LDSA concentrations measured (between 18 and 36 $\mu m^2\ cm^{-3}$) were representative of urban environments under traffic influence across Europe (Reche et al., 2015), and within the measurement ranges of the portable and stationary instruments. The comparability of the results from the

Table 2
Correlation coefficients and regression equations obtained from the comparison between portable (DiscMini) and stationary (CPC3785) instruments for particle number (N) monitoring.

	Post_SC1	Pre_SC2
DM1 (N)	Y = 1.35x-1370 R^2 = 0.82	Y = 1.17x-1527 R^2 = 0.94
DM2 (N)	Y = 1.81x-3799 R^2 = 0.89	Y = 1.07x-1345 R^2 = 0.94
DM3 (N)	Y = 1.30x-1300 R^2 = 0.72	Y = 0.94x-1658 R^2 = 0.93
DM4 (N)	Y = 1.61x-2299 R^2 = 0.85	Y = 0.90x-27 R^2 = 0.88
DM5 (N)	Y = 1.16x-432 R^2 = 0.75	Y = 01.06x-1273 R^2 = 0.93
Time resolution	10-min	10-min

NSAM monitor with surface area concentrations derived from SMPS measurements was also previously verified by Reche et al. (2015).

The portable and stationary instruments showed a good agreement with relatively low variability in the R^2 coefficients obtained, ranging between 0.70 and 0.94. These values are slightly lower than those obtained for the particle number concentrations (N), but similar to those obtained for BC monitoring. As in the cases above, the R^2 values seemed to be intercomparison-dependent, as they were mostly constant across units for each intercomparison exercise. In absolute values, the concentrations reported by the portable instruments were similar or lower than those reported by the stationary monitor (slope of the equation ranging between 0.52 and 0.98) during three of the four intercomparisons (Table 3). However, during pre_SC1 the concentrations reported by the portable monitors differed from the ones from the stationary instrument by a factor of 3, and the cause for this different behaviour is still unclear. As stated above this could be ascribed to a failure in the NSAM instrument, but no specific indications were observed during operation. Because the cause of this difference is unclear the results from pre_SC1 are not included in Fig. 3.

With some exceptions, the portable monitors generally underestimated the LDSA concentrations reported by the stationary monitor. Under the conditions of this work, the relative difference between the portable and the stationary instruments ranged between 7% and 13% (when excluding the results from pre_SC1) (Fig. 3). As in the case of particle number concentrations, these results are within the DM specified accuracy range (±20–30%; Asbach et al., 2012; Fierz et al., 2011).

3.4. Mean particle diameter

The mean particle diameter measured by the DiscMini instruments was compared with the mean particle size measured by the SMPS. These measurements were not carried out simultaneously with the intercomparison exercises described above due to the unavailability of the SMPS instrument. The SMPS data were available from 02/12/2014 to 10/12/2014, with a 5-min time resolution (2270 data points available). Only the DiscMini units 1, 3 and 5 were available during this period. Fig. 4 shows the comparison between the mean diameter measured by both types of instruments, evidencing the lower diameters reported by the portable instruments (ranging between 42 and 45 nm) when compared to the SMPS (51 nm). These differences were however rather constant across instruments, with the portable instruments consistently measuring lower particle diameters than the SMPS. Based on these results, the relative difference between the portable and the stationary instruments regarding mean particle diameter

Table 3
Correlation coefficients and regression equations obtained from the comparison between portable (DiscMini) and stationary (NSAM) instruments for lung-deposited surface area (LDSA) monitoring.

	Pre_SC1	Post_SC1	Pre_SC2	Post_SC2
DM1 (LDSA)	Y = 3.33x+4.22 R^2 = 0.94	Y = 0.95x+1.83 R^2 = 0.86	Y = 0.65x+4.27 R^2 = 0.84	Y = 0.95x+3.94 R^2 = 0.77
DM2 (LDSA)	Y = 3.65x+4.16 R^2 = 0.94	Y = 0.85x+3.73 R^2 = 0.79	Y = 0.68x+4.04 R^2 = 0.80	Y = 0.92x+2.99 R^2 = 0.77
DM3 (LDSA)	Y = 3.31x+3.65 R^2 = 0.94	Y = 0.98x-2.03 R^2 = 0.82	Y = 0.84x+3.47 R^2 = 0.88	Y = 0.74x+3.58 R^2 = 0.70
DM4 (LDSA)	Y = 3.94x+3.92 R^2 = 0.94	Y = 1.02x+3.16 R^2 = 0.83	Y = 0.52x+5.72 R^2 = 0.70	Y = 1.00x+2.52 R^2 = 0.77
DM5 (LDSA)	Y = 3.35x+6.09 R^2 = 0.90	Y = 0.85x+2.64 R^2 = 0.82	Y = 0.62x+4.25 R^2 = 0.83	Y = 0.99x+2.92 R^2 = 0.76
Time resolution	10-min	10-min	10-min	10-min

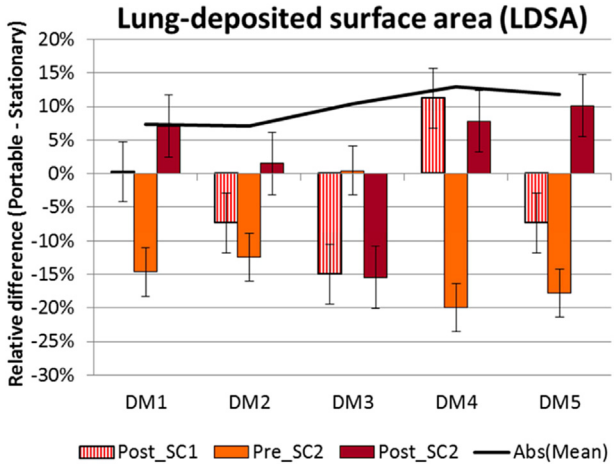

Fig. 3. Relative differences for lung-deposited alveolar surface area (LDSA) concentrations between the portable and stationary instruments for each unit and intercomparison exercise, calculated as 100*(Portable-Stationary)/Stationary. Abs(Mean): average across intercomparison exercises of the absolute value of the differences with regard to NSAM (in %) for each unit.

was quantified as 12–18%. The average sizes of the three DM units were very comparable with each other, with deviations between 2 and 4% which were lower than ±10% reported in under laboratory conditions (Asbach et al., 2012).

3.5. Particle mass concentration

The performance of the portable Dusttrak DRX instruments was assessed in comparison with the EU equivalent data provided by an online laser spectrometer corrected, in turn, by comparison with an EU-reference high-volume sampler. The portable instruments underwent standard maintenance operations between each intercomparison exercise (changing paper filters, cleaning metal mesh and cleaning inlet). The ambient air concentrations measured by the stationary instrument were within usual ranges for a Southern European urban background location and within the measurement ranges of both types of instruments: 14.0–37.1 μgPM_{10} m^{-3}, 8.4–26.2 $\mu gPM_{2.5}$ m^{-3}, and 5.2–23.9 $\mu gPM_1 \cdot m^{-3}$ (Querol et al., 2004).

As shown in Table S3, the operating performance of the Dusttrak DRX instruments was comparable to those of the portable instruments assessed in the previous sections, with data availability >99% in most cases with only one exception (DST3 during post_SC2, which suffered a technical failure). When looking at the comparability with the stationary data, however, the portable instruments showed on certain occasions an anomalous behaviour which remains so far unexplained. An example of this behaviour is shown in Fig. 5. In this example during pre_SC1, the four units being tested showed a consistent behaviour during the first part of the test, whereas at a given point in time one of the units (DST1) showed a clear jump in the time series and continued generating data consistently with the other units, although over what looks like a new background concentration. As shown also in Fig. 5, this re-basing process resulted in two clearly different correlation curves when comparing unit DST1 with another of the more stable units (in this case, DST2). This re-basing was observed on several occasions during the intercomparison exercises described in this work as well as during the actual sampling campaigns in the framework of the BREATHE project. The ultimate causes of the jumps detected in the time series are yet unknown to the authors, given that they appeared randomly and did not seem to be linked to factors such as temperature, humidity, absolute particle concentrations, particle size fraction or even vibrations on the tables where the instruments were located. As shown in Fig. 5, the correlation between the portable and stationary instruments was high in the absence of these jumps. However, because of the apparent randomness of this issue the data required careful post-processing (mainly, screening) in order to identify and correct for re-basing issues.

The mean uncertainty of the portable monitors was calculated as in the sections above. In the absence of re-basing issues, the relative error could be as low as 5% (in terms of $\mu g \cdot m^{-3}$) (Fig. 6). However, when these issues were detected it could reach up to 292%. When the jumps in the time series were relatively small and initially undetected, relative errors calculated were in the range of 40–50%. An in depth analysis as for the other instruments (Tables 1–3) is not shown given that the correlation equations and the R^2 coefficients showed a broad variability when the time series were affected by re-basing problems.

4. Summary and conclusions

The performance of three portable monitors (micro-aethalometer AE51, DiscMini, Dusttrak DRX) was assessed for outdoor (ambient) air exposure assessments in a representative Southern European urban environment. The air quality parameters measured by these monitors were black carbon (micro-aethalometer AE51), particle number concentration (10–700 m), alveolar surface area (20–400 nm) and mean particle diameter (10–700 nm) (DiscMini), and particulate matter mass concentration in the size ranges PM_{10}, $PM_{2.5}$ and PM_1 (Dusttrak DRX). The assessments were carried out based on the assumption that the monitors were indicative (based on the classification by US-EPA, 2013), also considered as tier 2 instruments in the tiered approach (Asbach et al., 2012; Methner et al., 2010), meaning that they should be able to deliver a limited

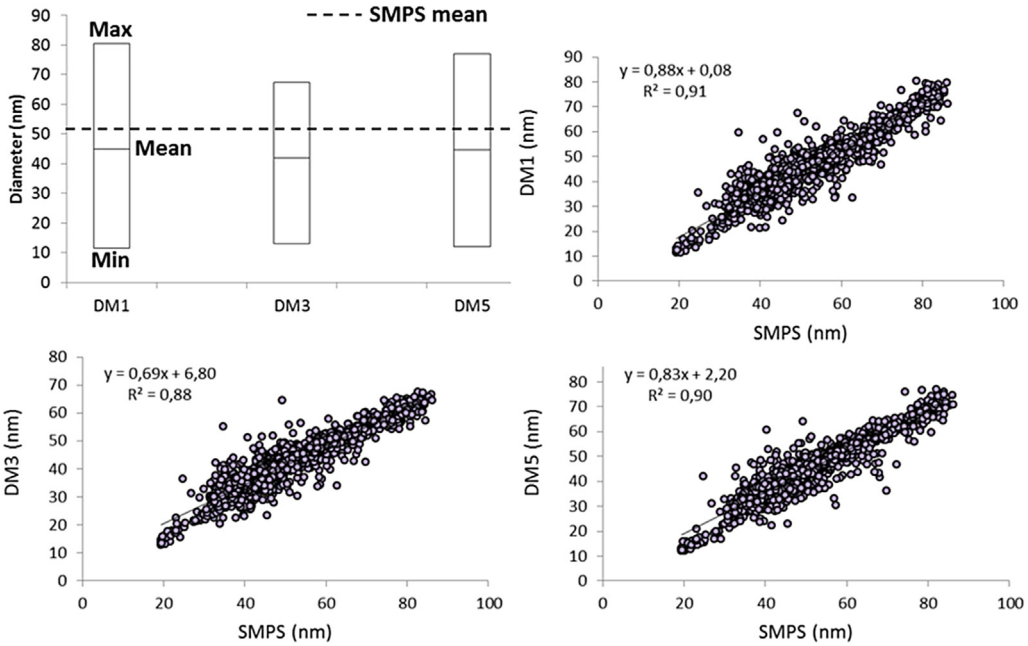

Fig. 4. Mean, minimum and maximum mean particle diameter measured with DiscMini instruments, and comparison with the mean particle diameter measured by the SMPS system. Correlation between the different DM units and the SMPS for mean particle diameter.

but meaningful data set to estimate exposure levels. Their operational requirements (easy to use, portable, battery operation) had been previously confirmed.

The performance of the monitors under study was compared to that of frequently used stationary instruments measuring the same (or comparable) aerosol parameters (MAAP, CPC, SMPS; NSAM, GRIMM laser spectrometer). Overall, all of the portable monitors were robust regarding data capture, with data availability mostly >95%. Technical failures did occur, and they seemed to be linked to specific units (e.g., black carbon monitor BC3, which failed during

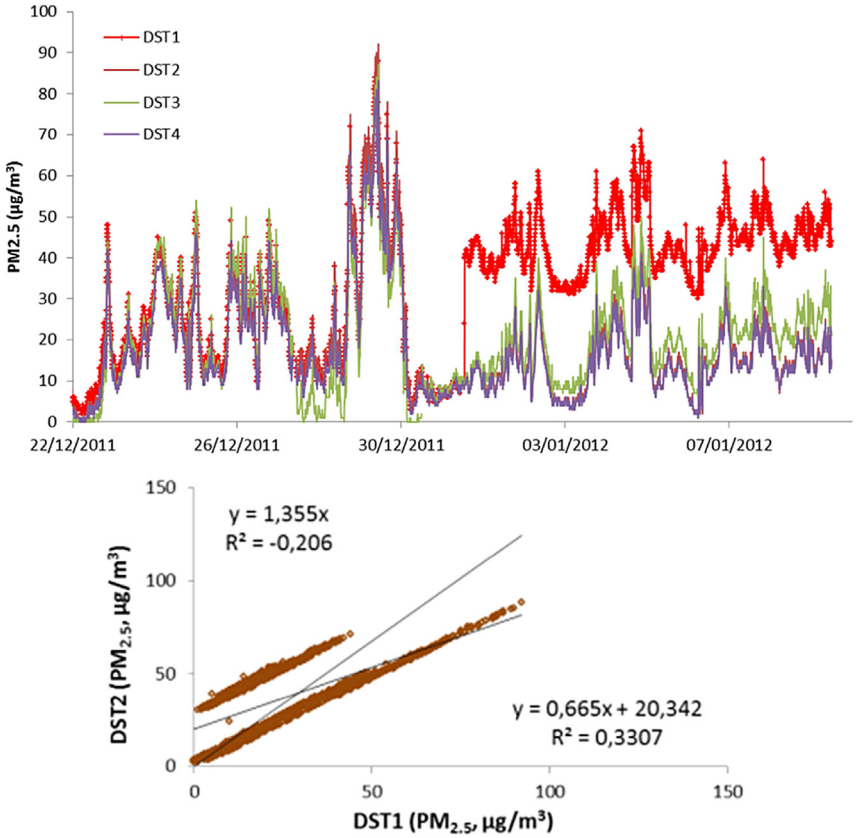

Fig. 5. Example of re-basing in a Dusttrak DRX time series for $PM_{2.5}$ mass concentrations, occurring during pre_SC1. Correlation between the DST1 and DTS2 units during the same period.

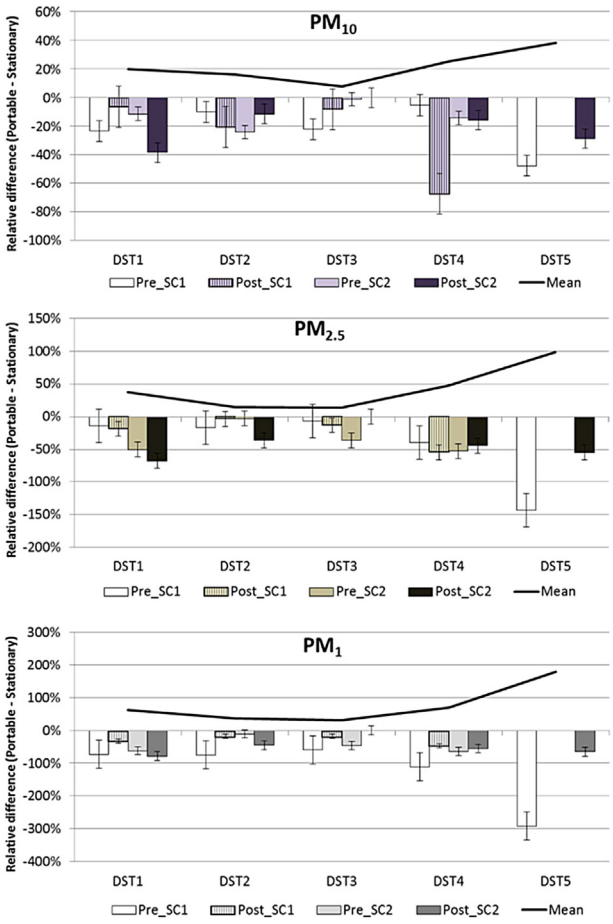

Fig. 6. Relative differences for PM_{10}, $PM_{2.5}$ and PM_1 mass concentrations between the portable and stationary instruments for each unit and intercomparison exercise, calculated as 100*(Portable-Stationary)/Stationary. Abs(Mean): average across intercomparison exercises of the absolute value of the differences with regard to GRIMM1107 (in %) for each unit.

intercomparisons pre_SC2 and post_SC2). This implies that once the technical failure was identified the instrument could be removed and serviced, without further disruption of the monitoring campaigns. This was not the case, however, with the Dusttrak DRX monitors which suffered from apparently random re-basing issues with negative impacts on data quality. The authors were unable, to date, to identify the specific cause of this issue or to provide a technical solution for it. With regard to the portable particle counters, the type of tubing used at the inlet was identified as a key parameter influencing instrument performance.

The portable instruments showed good agreements with their respective stationary counterparts, with R^2 values mostly >0.80. Under the urban aerosol concentrations monitored in this work, the mean relative differences (in %) between the portable and stationary instruments were 7–12% for black carbon (monitored with micro-aethalometer AE51), 10–18% for particle number concentration (with DiscMini), 7–13% for alveolar surface area (with DiscMini), and 12–18% for mean particle diameter (with DiscMini). The relative differences for particle mass concentrations (with Dusttrak DRX) were 5% in absence of re-basing issues, but increased up to 292% when these occurred. Finally, intra-unit comparability was high for the micro-aethalometer AE51 and DiscMini instruments, with relative differences between units of 7–11% for the former and 6–8% for DiscMini particle number concentration, 2–4% for DiscMini surface area, and 2–4% for DiscMini mean particle diameter. The good agreement between units is especially relevant for exposure studies aiming to compare concentrations between different scenarios.

Overall, it may be concluded that a good agreement was found between most of the portable and stationary instruments tested, for outdoor urban background air. Our results validate the performance of the black carbon, particle number concentration, particle surface area and mean particle diameter monitors as indicative instruments (tier 2) for exposure assessment studies These results could be reproducible for indoor air, although a dedicated study is necessary to verify this hypothesis.

Role of the funding sources

The funding sources had no direct involvement in the study design, collection, analysis and interpretation of data, or in the decision to submit the article for publication. They were involved in covering personnel and instrumentation costs.

Acknowledgements

The research leading to these results has received funding from the European Research Council under the ERC Grant Agreement number 268479 – the BREATHE project. Additional funding for specific instrumentation was provided by national project IMPACT (CGL2011-26574), the Spanish Ministry of Agriculture, Food and the Environment (UCA2009020083), and projects VAMOS (CLG2010-19464-CLI) and GRACCIECSD2007-00067. Support is acknowledged to Generalitat de Catalunya 2009 SGR8 and to FP7 Marie Curie ITN HEXACOMM (Nr.315760).

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.10.076.

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