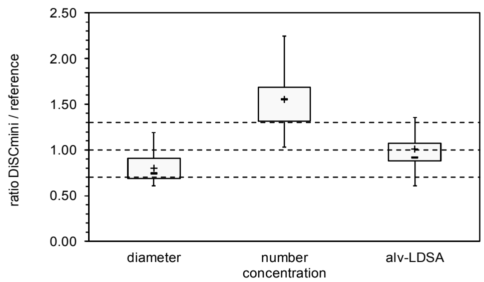
# Environmental Science Processes & Impacts

Accepted Manuscript


Graphical abstract

Comparison of DiSCmini data to reference data for polydisperse test aerosols in terms of diameter, number concentration and alv-LDSA



## Environmental impact statement

This paper deals with the performance characterization of a standalone real-time handheld instrument, the DiSCmini, which provides the number concentration ($10^3$..$10^6$ $cm^{-3}$) and average diameter (20..300 nm) of airborne nanoparticles (particle size below 700 nm as selected by an inlet separator). This device can be used as well for assessing workplace atmospheres (occupational exposure) as for indoor or outdoor air monitoring. Therefore, providing robust characterization of the DiSCmini has become essential, given its potential for use. The present study investigates the response of the instrument with various monodisperse and polydisperse test aerosols over a wide range of sizes and concentrations, under laboratory conditions. Data were compared to data collected in parallel using reference instruments, which complements the information already published.

Journal Name RSCPublishing

ARTICLE

Cite this: DOI: 10.1039/x0xx00000x

Received 00th January 2012,
Accepted 00th January 2012

DOI: 10.1039/x0xx00000x

www.rsc.org/

# Laboratory study of the performance of the handheld Diffusion Size Classifier (DiSCmini) for various aerosols in the 15-400 nm range

S. Bau,[a] B. Zimmermann,[a] R. Payet[a] and O. Witschger[a]

In addition to chemical composition, particle concentration and size are among the main parameters used to characterize exposure to airborne ultrafine or nanoparticles. To assess occupational inhalation exposure, real-time instruments are recommended in the recent strategies published. Among portable devices for personal exposure assessment in the workplace, the DiSCmini (Matter Aerosol AG, Switzerland) has been identified as a potential candidate with its capacity to measure the airborne nanoparticle concentration and average particle size with good time-resolution. Monodisperse and polydisperse test nanoaerosols of varying composition and morphology were produced in the laboratory using the CAIMAN facility. These aerosols covered a range of particle sizes between 15 and 400 nm and number concentrations from 700 to 840,000 $cm^{-3}$. The aerosols were used to investigate the behavior of the DiSCmini, comparing experimental data to reference data. In spite of a slight tendency to underestimate particle size, all particle diameters, number concentrations and surface area concentrations measured were in the same order of magnitude as reference data. Furthermore, no significant effect due to particle composition or morphology was noted.

## Introduction

Atmospheric, indoor and workplace environments contain airborne sub-micrometer particles[1, 2]. For the particular case of industrial aerosols, sources of ultrafine particles can be as various as combustion processes, diesel engine exhausts, printer emissions, metal welding fumes, just to mention a few[3-10].

Contrary to the aforementioned incidental sources of airborne ultrafine particles, the focus has come on nanotechnologies and nanomaterials, where nanoscale particles are intentionally produced. Because of the increasing use of manufactured nanomaterials, it is likely that situations leading to occupational exposure are becoming more numerous at all points in their life cycle, including synthesis, production, downstream use, aging[11], waste treatment and disposal[12, 13], as well as maintenance[14, 15] or accidental scenarios[16, 17]. Workers in research laboratories and industry are also increasingly exposed.

For epidemiological studies seeking to determine the relationship between particulate air pollution and health effects it is essential to have quantitative information on exposure levels[18, 19]. The present study fits into the general framework of increasing what is known about devices devoted to the assessment of occupational exposure to airborne nanomaterials.

Currently, the question of the parameters/characteristics that should be assessed in epidemiological studies remains open. When possible, it has been suggested that a multimetric approach be carried out[20-22]. In addition to chemical composition, particle concentration and size play an important role (*e.g.* Wang *et al.*[23]) in determining the quantity and the region of the respiratory tract where inhaled nanoparticles will be deposited and potentially interact.

To meet these needs, methodologies to assess occupational inhalation exposure to airborne particles during production, handling and use of manufactured nanomaterials have recently been proposed[24-30] and tested in various workplace environments[31-36]. In particular, these strategies emphasize the utility of real-time instruments.

Although time-resolved measurement techniques still need to be improved[37], they can be used to:

(1) characterize airborne nanoparticle sources, either qualitatively (screening) or quantitatively over time and space (see *e.g.* Peters *et al.*[35, 38], Imhof *et al.*[39]);
(2) identify the various tasks associated with emissions (existence of exposure and/or emission peaks), particularly when high sensitivity is sought (*e.g.* Mohr *et al.*[40]);

(3) trigger alarms in case of malfunction of collective protective equipment or accidents[41];
(4) provide a real-time estimate — under specific conditions and assumptions — of workers' occupational exposure[42-45].

As a consequence, it has become essential to provide robust scientific characterization of this type of instrument before they can be widely used in workplace environments[46, 47].

"Gold-standard" instruments such as electrical mobility analyzers (*e.g.* Scanning Mobility Particle Sizer – SMPS, Fast Mobility Particle Sizer – FMPS), or low pressure cascade impactors (*e.g.* Electrical Low Pressure Impactor – ELPI) are not ideally suited to monitoring aerosols in workplaces due to their low time-resolution, lack of field-portability, complexity of use and high cost[48, 49]. Approaches based on simultaneous measurements of an aerosol with different instruments have been proposed (e.g. Woo *et al.*[50], Maynard[51], Bau *et al.*[52]) but these are not suitable for personal monitoring. The latter are more research-grade instruments, which make it is possible to accurately measure aerosol parameters for laboratory studies[53].

A possible solution has recently been developed in the form of time-resolved portable devices based on particle diffusion charging and sequential electrical measurement. Among these, the DiSCmini (Matter Aerosol AG, Switzerland) was developed by Fierz *et al.*[54] based on the meDiSC[55, 56]. It was formerly known and distributed as the miniDiSC, which is why in the remainder of this paper, both miniDiSC (former version) and DiSCmini (actual version) will be used. The DiSCmini is a standalone real-time handheld instrument that provides the airborne nanoparticle number concentration ($10^3$ - $10^6$ $cm^{-3}$) and average diameter (20 - 300 nm) with an accuracy of ± 30% according to the manufacturer[57]. According to Asbach *et al.*[58], we believe that this level of accuracy is sufficient for most monitoring applications. To avoid artifacts due to coarse particles, the DiSCmini is equipped with an inlet separator with a cutoff diameter of 700 nm.

The device functions by aspirating particles at a controlled flow rate of 1.0 $L.min^{-1}$ and imparting a positive charge by means of a diffusion charger (10 nA, 3-5 kV). The instrument principle lies in the measurement of currents due to the collection of charged particles on two separate stages: a diffusion stage ($i_D$ current) and a terminal filtration stage ($i_F$ current). These currents are then converted into a particle size (which is expected to be proportional to the $i_F/i_D$ ratio) and a number concentration (which is expected to be proportional to $i_F + i_D$). Apart from these two key characteristics, the particle lung-deposited surface area concentration for the alveolar region of the respiratory tract can also be determined. This concentration is related to the total current ($i_F + i_D$), as indicated by various authors investigating diffusion chargers[59-62]. It corresponds to the product of the geometric surface area concentration and the probability that a particle will deposit in the alveolar region. This probability can be obtained from models based on experimental data, *e.g.* that provided by the International Commission on Radiological Protection (ICRP)[63].

The DiSCmini is robust, light and battery operated. Moreover, unlike the condensation nucleus counters (CNC), it requires no working fluid. All this makes it an ideal candidate for monitoring personal exposure to airborne nanomaterials. To ensure optimal operation and measurement traceability, the flow rate as well as the charger's voltage and current are recorded every second.

This instrument was previously studied in a laboratory setting by Mills *et al.*[64, 65] for monodisperse and polydisperse aerosols in a range of diameters between 30 and 300 nm and concentrations from 400 to 80,000 $cm^{-3}$. The aerosols used were composed of salt (NaCl) and metal, which are considered to be representative of welding fumes. The results obtained indicated relative discrepancies from measurements with reference instruments of ± 35% in terms of number concentration and particle diameter in a monodisperse configuration. In contrast, the particle size was overestimated by up to 80% with polydisperse aerosols. Relative discrepancies between – 8% and + 25% had previously been reported by Asbach *et al.*[58] between miniDiSC and reference number concentrations for polydisperse test nanoaerosols consisting of NaCl, DEHS (Di-Ethyl-Hexyl-Sebacate) and graphite in a range of concentrations up to 80,000 $cm^{-3}$, while the average particle diameters measured with the miniDiSC were found to be within ± 27% of the reference value.

The behavior of the miniDiSC was also investigated in field conditions by various authors[66-68]. Particle number concentrations as well as diameter profiles measured by the miniDiSC were found to correlate with data from reference instruments. In spite of significantly different concentration levels measured by the different instruments, the miniDiSC is considered to be a useful tool for assessing personal exposure to airborne nanoparticles in workplaces.

However, there is still a need to further our knowledge of the behavior of the DiSCmini[64, 66]. In particular, this study investigated the response of the DiSCmini with various test aerosols over a wider range of sizes and concentrations than previously tested, under laboratory conditions. Both monodisperse and polydisperse test aerosols were used to test the instrument, and data were compared to data collected in parallel using reference instruments.

## Materials and Methods

Monodisperse and polydisperse test aerosols were used to investigate how the DiSCmini performed compared to reference aerosol instruments. Primarily, the Limit of Quantification for both electrometers and the response time were assessed.

### Limit of Quantification and response time for the DiSCmini

Preliminary experiments were performed with HEPA-filtered air, which produce particle-free air, to determine the variability of the electrometer signals ($\sigma$) and thus deduce the corresponding Limit of Quantification ($LOQ$) based on:

$$LOQ = 10 \cdot \sigma \qquad (1)$$

Table 1 presents the mean value and standard deviation obtained after 15 minutes for both diffusion and filtration stages.

Table 1. Electrometer noise levels for the DiSCmini.

| Stage | Mean current (fA) | Standard deviation $\sigma$ (fA) |
|---|---|---|
| Diffusion ($i_D$) | -0.110 | 0.120 |
| Filtration ($i_F$) | -0.100 | 0.097 |

Table 1 indicates an $LOQ$ of 1.2 fA for the diffusion stage, and 1.0 fA for the filtration stage. These values are in agreement with Fierz *et al.*[54]. As indicated in Bau *et al.*[56], these data can then be used to determine the corresponding $LOQ$ in terms of number concentration, depending on particle size, aerosol flow rate, charging efficiency and particle penetration through the system. This roughly corresponds to a concentration of $10^3$ cm$^{-3}$ for 30 nm particles, and $10^2$ cm$^{-3}$ for 250 nm particles, which is in line with the manufacturer's specifications[57]. In the remainder of this paper, all data falling below the $LOQ$ will be disregarded to ensure the robustness of the conclusions drawn.

The instrument response time was assessed by measuring both currents and suddenly increasing particle concentration from 0 (HEPA-filtered air) to a given stable value (concentration step). An example of the instrument response is provided in Figure 1 for the diffusion current in the case of a positive step. Data from run 1 were disregarded due to an automatic zeroing procedure during the corresponding experiment.

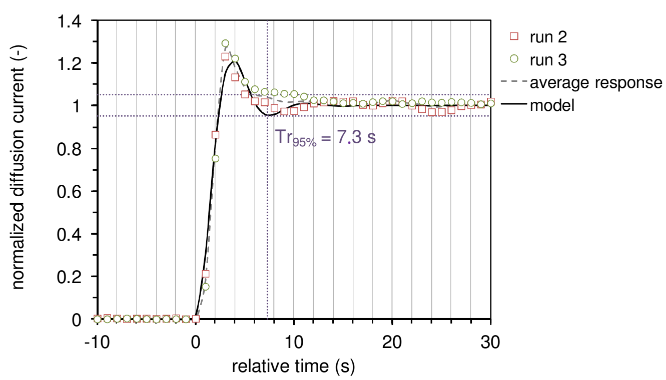



Figure 1. Normalized response of the DiSCmini for a positive concentration step.

The average response was modeled based on a second order system, whose behavior, $y(t)$, was described by:

$$y(t) = 1 - \frac{\exp(-\zeta \cdot \omega \cdot t)}{\sqrt{1-\zeta^2}} \cdot \sin(\omega \cdot t + \varphi) \quad (2)$$

with $\omega = 0.938 \text{ rad} \cdot \text{s}^{-1}$ the pulsation, $\zeta = 0.438$ the damping coefficient, and $\varphi = 1.081$ rad the dephasing angle being the set of parameters obtained by numerical resolution. The response time $Tr$ before the instrument reaches $n\%$ of the expected variation can be determined using the following equation:

$$Tr_n = \frac{1}{\zeta \cdot \omega} \ln\left(\frac{100}{n}\right) \quad (3)$$

The corresponding 95% response time is $Tr_{95\%} = 7.3$ seconds, as shown in Figure 1.

### Aerosols studied (test aerosols)

Experiments were performed with test aerosols produced using the CAIMAN (Characterization of Aerosol Instrumentation used to Measure the Aerosols of Nanoparticles) facility[69]. This setup produces stable and reproducible airborne nanoparticles with controlled properties (size distribution, chemical composition, concentration level).

The CAIMAN facility is composed of a spark discharge generator (PALAS GFG-1000), a bipolar ion generator (EAN 581, TOPAS) and a high-temperature furnace (CARBOLITE BST16 with a maximum operating temperature of 1500 °C). (Nano)particle-free air is introduced into the setup from a purification unit (TSI model 3074B), and excess aerosol is filtered using HEPA filters (CAMFIL, 'BAG filter' model). For this study, neither the ion generator nor the high-temperature furnace were used to modify the properties of the aerosols produced.

Two different electrodes were used in this work: carbon (graphite, pure) and silver (99.99% pure). An alternative experimental configuration, involving a Laskin-type nebulizer[70], was used to produce DEHS (Di-Ethyl-Hexyl-Sebacate, pure) and CsCl (Cesium Chloride, aqueous solutions of 1 and 50 g.L$^{-1}$) aerosols.

While the silver and DEHS particles produced are spherical, carbon-based particles (agglomerates) present a fractal-like morphology, and cesium chloride particles are cubic.

### Experimental setup in monodisperse mode

The DiSCmini's performance was first assessed with monodisperse airborne nanoparticles, in the experimental setup illustrated in Figure 2.

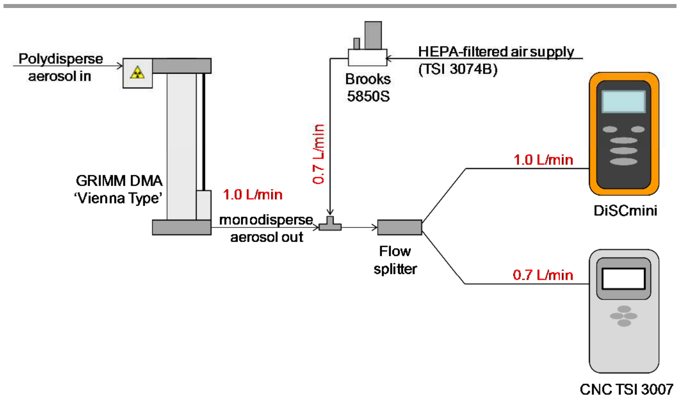



Figure 2. Diagram of the experimental setup for monodisperse test aerosols.

In this configuration, polydisperse aerosols were directed through a DMA (Differential Mobility Analyzer, Grimm Vienna Type, $Q_{aerosol} = 1.0$ L.min$^{-1}$, $Q_{sheath} = 10$ L.min$^{-1}$) equipped with an $Am^{241}$ radioactive source to select airborne particles according to their electrical mobility. The DMA was controlled by means of a DMA-controller (Grimm, device not shown in Figure 2). Assuming that the proportion of multiply-charged particles is negligible (see *e.g.* Bau *et al.*[56]), this results in the production of monodisperse aerosols with a given electrical mobility diameter, the corresponding *GSD* being typically below 1.05 according to previous work where a model for the DMA transfer function was used[71]. The particle diameter selected by the DMA was considered to be the reference diameter.

For the same sample, the reference number concentration was determined using a handheld CNC (Condensation Nucleus Counter, TSI model 3007, $Q$ = 0.7 L.min$^{-1}$). This instrument counts particles between 10 nm and 1 µm in single-count mode for aerosols with up to $10^5$ particles.cm$^{-3}$. Data above $10^5$ cm$^{-3}$ should be considered with caution as they exceed the nominal concentration range of the CNC.

Data measured by the DiSCmini, *i.e.*, particle average diameter and number concentration, were then compared to reference data. The range of particle diameters and number concentrations covered in the different experiments performed are given in Table 2.

Table 2. Characteristics of the monodisperse test aerosols used.

| Material | Range of reference particle diameters (nm) | Range of reference particle number concentrations (cm$^{-3}$) |
|---|---|---|
| Ag | 15 – 120 | 700 – 76,000 |
| C | 20 – 200 | 2,000 – 83,000 |
| CsCl | 60 – 200 | 800 – 3,200 |
| DEHS | 50 – 400 | 900 – 200,000 |

**Experimental setup in polydisperse mode**

In the second part of this study, the DiSCmini was challenged with polydisperse test aerosols, as shown in Figure 3.

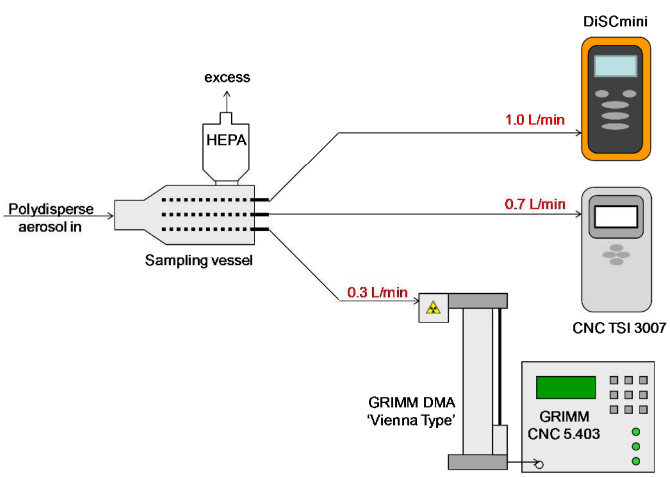



Figure 3. Diagram of the experimental setup with polydisperse test aerosols.

In this setup, the number size distributions of the nanoaerosols produced were measured by a SMPS (Scanning Mobility Particle Sizer), composed of the abovementioned DMA (working at a different flow rate of $Q_{aerosol}$ = 0.3 L.min$^{-1}$) and a CNC (Grimm model 5.403, $Q$ = 0.3 L.min$^{-1}$). This SMPS number size distribution was then fitted with a lognormal curve to determine the modal diameter of the distribution, which is considered to be the reference particle diameter. Examples of lognormal fitting of experimental data are presented in Fig. 4.

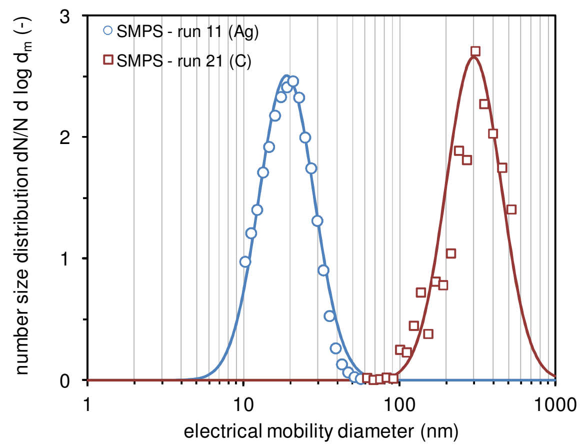



Figure 4. Examples of number size distributions determined for polydisperse test aerosols.

The characteristics of the polydisperse test aerosols used are presented in Table 3. Altogether, the modal diameter ranged from 14 to 300 nm, and the number concentration was between 4,700 and 840,000 cm$^{-3}$. Due to the nominal concentration range of the CNC, higher number concentrations (above $10^5$ cm$^{-3}$) were provided by the SMPS. The geometric standard deviations (*GSD*) of the adjusted lognormal number size distributions were found to be between 1.48 and 2.68.

Table 3. Characteristics of the polydisperse test aerosols used.

| Material | Range of reference modal diameters (nm) | Range of reference particle number concentrations (cm$^{-3}$) |
|---|---|---|
| Ag | 19 – 72 | 7,300 – 710,000 |
| C | 14 – 300 | 4,700 – 440,000 |
| CsCl | 25 – 42 | 27,600 - 840,000 |
| DEHS | 220 – 225 | 61,000 – 154,000 |

Each experimental condition was maintained for 30 to 40 minutes to ensure aerosol stability and sufficient scans to allow the average number size distribution to be determined by the SMPS. The number size distributions for carbon agglomerates were corrected as suggested by Lall & Friedlander[72] using a primary particle diameter of 16 nm[62]. As previously, the reference number concentration was provided by CNC analysis of the same aerosol in parallel.

In half of the cases, a NSAM[73, 74] (Nanoparticle Surface Area Monitor, TSI model 3550, $Q$ = 2.5 L.min$^{-1}$) was added as a fourth sampling line directly connected to the sampling vessel (not depicted in Figure 3) to provide the reference lung-deposited surface area concentration for particles depositing in the alveolar region of the respiratory tract (alv-LDSA). Detailed information on how the NSAM performs is provided elsewhere[62, 75].

As for the monodisperse aerosols, data measured by the DiSCmini were compared to reference data.

## Results and discussion

### Monodisperse aerosols

Figure 5 presents the comparison between measured and reference sizes (*top*) and number concentrations (*bottom*).

This comparison shows a satisfactory agreement between DiSCmini and reference data, with no remarkable effect due to particle composition or morphology.

Taken together, the average relative discrepancies were -16% and +2% for particle diameter and number concentration, respectively. More precisely, the particle diameters provided by the DiSCmini were found to be within ± 38% of the reference particle diameter selected by the DMA. In contrast, the particle number concentrations measured by the DiSCmini were found to be closer to the reference on average, even though they presented a greater variability, with relative discrepancies up to 65%. Similar results were reported by Mills *et al.*[64] in their study using monodisperse test aerosols where relative discrepancies were found to be within ± 29% for particle diameter and ranged from – 21% to + 101% for particle number concentration.

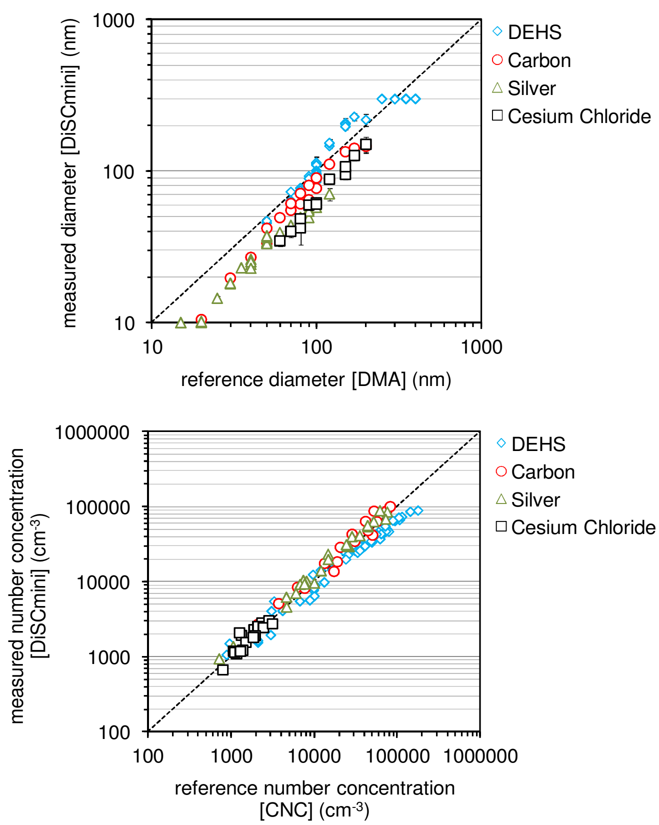



Figure 5. Comparison of average size (*top*) and number concentration (*bottom*) measured by the DiSCmini and reference measurements in monodisperse mode.

The data presented in Figure 5 also show a slight tendency to underestimate particle size for diameters below 40 nm. For this size range ($d_m \leq 40$nm), the average deviation observed between DiSCmini and DMA data is – 37%. This has not been highlighted in previous studies, where both positive and negative deviations were observed, and attributed to random errors[64]. These cases do not correlate with the larger biases observed for number concentrations.

For particles with diameters above ≈ 250 nm, the diameter reported by the DiSCmini was systematically 300 nm, which corresponds to the upper limit of the size range for the instrument. This suggests that the curve describing the ratio of the currents to particle size reaches a plateau. This appears to be confirmed by the data presented later in this paper.

Figure 6 shows a Box & Whiskers plot of the ratios of measured data to reference data. This representation shows that particle diameters and number concentrations agree with the reference within ± 30% in 60% and 70% of cases, respectively.

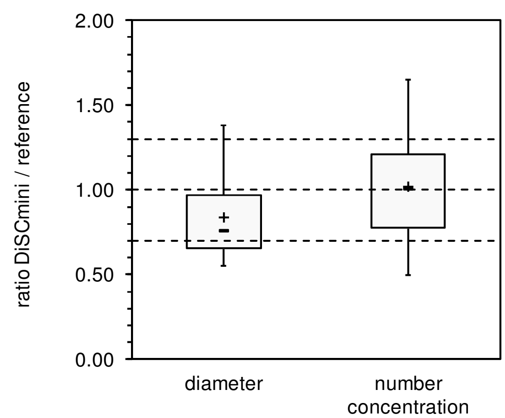



Figure 6. Comparison of the DiSCmini / reference ratio in monodisperse mode ($n$ = 108). Dashed horizontal lines indicate a ratio of one and the ± 30% discrepancy expected based on the manufacturer's specifications.

The experiments carried out in monodisperse mode can also be used to determine the charging law of the unipolar positive corona charger present in the DiSCmini. This is possible as measured currents are related to air flow rate ($Q$), particle number concentration ($C_N$) and the number of elementary charges carried per particle of a given size $p(d_m)$, in the following relationship:

$$C_N = \frac{i_F + i_D}{p(d_m) \cdot e \cdot Q} \tag{4}$$

where $e$ corresponds to the elementary charge ($e = 1.602 \cdot 10^{-19}$ C). In each condition, both currents, $i_F$ and $i_D$, and the reference number concentration were measured; the power charging law can therefore be defined as follows:

$$p(d_m) = a \cdot d_m^b \tag{5}$$

The experimental data (not shown) were found to be independent of particle morphology and chemistry. Taken together, the least-squares power fit of this data ($R^2 = 0.890$) gave $a = 0.016$ and $b = 1.099$. These parameters are in good agreement with those presented by Fierz *et al.*[54] who reported an exponent ($b$) of 1.125 for the miniDiSC upon challenge with NaCl particles from 20 to 240 nm in diameter. This corresponds to a typical value for diffusion chargers, for which the exponent is expected to decrease from 2, in free-molecular regime, to a value tending towards 1 in continuous regime (see *e.g.* Flagan[76] or Ntziachristos *et al.*[77]).

Finally, the correlation between the $i_F/i_D$ ratio and particle diameter was verified (Figure 7). This was essential as the basic design and operation of the instrument rely on this correlation.

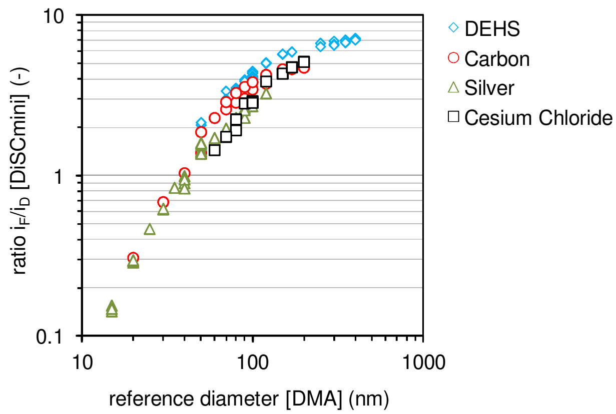



Figure 7. Relationship between current ratio and reference particle size for the different monodisperse test aerosols.

The data presented in Figure 7 highlight a clear correspondence between the ratio of measured currents and particle size, as indicated by the manufacturer[57]. As stated earlier, the ratio of the currents to particle size seems to reach a plateau above ≈ 250 nm. In addition, it can be concluded from Figure 7 that neither particle composition nor particle morphology significantly affect the particle size or current ratio determined by the DiSCmini. Detailed information on the diffusion phenomena governing this behavior are given in Bau *et al.*[56] and Fierz *et al.*[54].

### Polydisperse aerosols

The comparison between DiSCmini and reference data for polydisperse aerosols is proposed in Figure 8 for the average particle diameter (*top*) and number concentration (*bottom*).

Figure 8 suggests that the DiSCmini underestimates particle diameter, leading to an overestimation of the corresponding number concentration. Once again, no significant effect of particle composition or morphology was noted. Relative discrepancies ranged from – 40% to + 19% for particle diameter, with an average of – 20%. There are only three cases where the diameter reported by the DiSCmini was larger than the reference diameter determined from the adjusted SMPS number size distribution. In contrast, the average bias is + 55% for the particle number concentration, ranging from + 3% to + 147%. It should be remembered that a polydisperse aerosol with a *GSD* of 1.7 was considered in the "calibration" of the instrument. When the aerosol measured has a different *GSD* this can cause greater deviations. As indicated by Bau *et al.*[56], the *GSD* of the number size distribution significantly influences the current ratio ($i_F/i_D$) above 1.7, leading to inaccurate particle diameters. Consequently, the resulting number concentration calculated using equation (4) will also be affected because of an incorrect number of elementary charges estimated per particle.

Based on our previous work[52], the count median diameter was also estimated from both number and surface area concentrations (when the NSAM was used). The average size provided by the DiSCmini and this approach, assuming $GSD = 1.8$ (data not shown), produced a linear correlation ($R^2 = 0.949$) with a slope of 0.8, indicating that the DiSCmini underestimates particle size by 20% on average. When considering a $GSD$ of 1.7, the slope becomes 0.75 ($R^2 = 0.951$), *i.e.*, an average 25% underestimation of particle size by the DiSCmini. These findings are in line with previous observations relating to the reference diameter determined by SMPS, as shown in Figure 8.

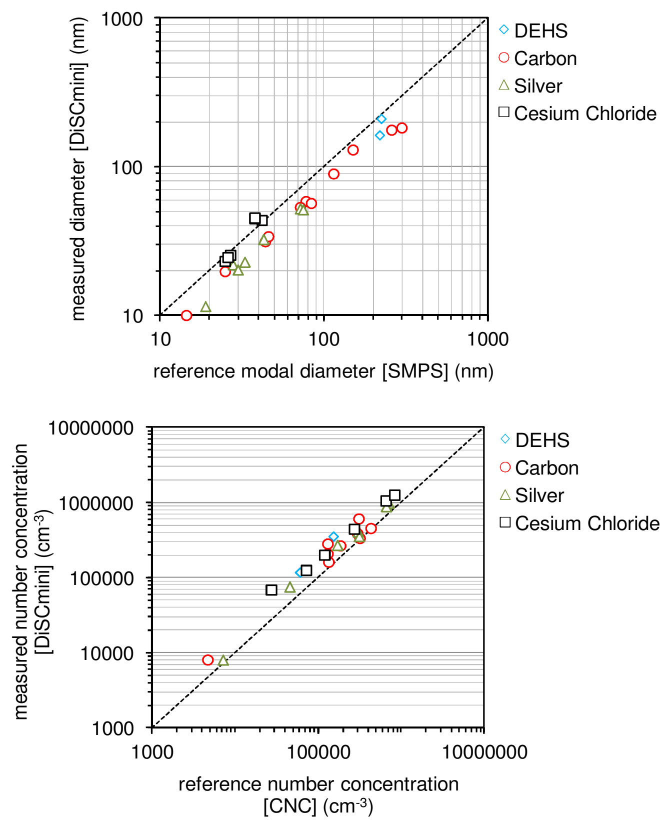



Figure 8. Comparison of average size (*top*) and number concentration (*bottom*) measured by the DiSCmini and reference measurements for polydisperse aerosols.

In their laboratory study, Mills *et al.*[64] reported significant differences — up to 81% — between the particle diameter measured by the DiSCmini and the reference SMPS size in a range from 100 to 200 nm for *GSD*s between 1.5 and 2.3. In contrast to the results presented in Figure 8, they observed ratios greater than one in all cases, *i.e.*, the DiSCmini overestimated particle diameters in their hands. They also found number concentrations to be within 21% of those measured with the SMPS for either NaCl or metal polydisperse aerosols with concentratins between 500 and 50,000 $cm^{-3}$.

Meier *et al.*[66] also highlighted relative discrepancies between the particle diameter determined by miniDiSC and the SMPS-derived geometric mean diameter (from – 23% to + 2%) with their experimental data on stationary aerosol measurements next to a highway. The extent of these discrepancies depended on the SMPS measurement range considered. The aerosols had a geometric mean diameter between 30 and 60 nm; but no information is provided about their *GSD*. These results suggest that the number concentration based on the SMPS data taken between 13 and 311 nm correspond best to those returned by the miniDiSC (1% deviation on average); the concentration range covered was between 8,000 and 50,000 $cm^{-3}$. When the full size range of the SMPS was considered (10 – 1110 nm), the miniDiSC recorded significantly fewer particles.

Aerosols with varying morphologies, from spherical (DEHS) to cubic (NaCl) and fractal-like (soot), were used to challenge two models of miniDiSC in another study[58]. The geometric and arithmetic mean diameters provided by a FMPS (Fast Mobility Particle Sizer, TSI model 3091) were taken as reference values for particle size, ranging between 28 and 177 nm. Whatever the number concentration between < 1,000 and 80,000 $cm^{-3}$ provided by a handheld CPC (TSI model 3007), the data measured by both miniDiSC were found to be between – 8% and + 25% of the reference, in line with the technical specifications. An effect of particle size on the counting accuracy of the miniDiSC was noted. The two miniDiSC were found to be equivalent; the deviations between reported particle diameters and reference values were within ± 27%, with the highest deviations observed for soot particles. Based on these results, the authors suggest that miniDiSC sizing accuracy depends on particle morphology. This appears to contradict the results presented in Figure 8.

As mentioned above, the NSAM was used to provide the reference lung-deposited surface area concentration for particles contained in the aerosol. The average relative discrepancy for this measurement was found to be 1%, even though deviations of up to ± 60% were observed. A similar level of relative bias between miniDiSC and FMPS-derived alv-LDSA has previously been reported[58], particularly for particles larger than 400 nm.

Figure 9 shows a Box & Whiskers plot of the ratios of measured and reference data for the three parameters considered, whatever the nature of the aerosol.

This data shows that the ± 30% deviation tolerated by the manufacturer is satisfied in 70%, 25% and 70% of the cases for particle diameter, number concentration and alv-LDSA, respectively. Again, the underestimation of particle size as well as the assumption that the aerosol *GSD* is 1.7 in the internal operation of the DiSCmini may have caused this overestimation of number concentrations.

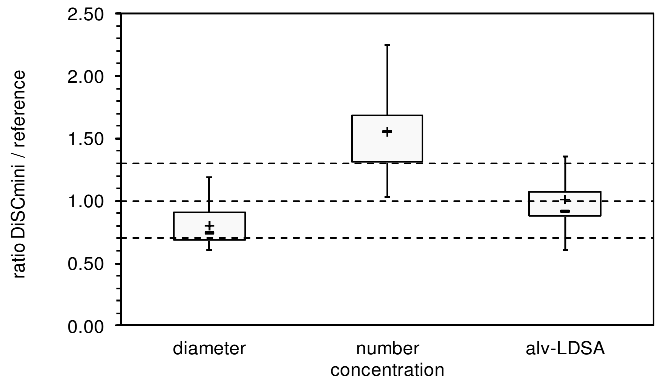



Figure 9. Comparison of the DiSCmini / reference ratio in polydisperse mode (*n* = 24 for diameter and number concentration; *n* = 10 for alv-LDSA). Dashed horizontal lines indicate a ratio of one and the ± 30% discrepancy expected based on the manufacturer's specifications.

## Conclusions

This experimental study aimed to characterize the performances of a miniature diffusion size classifier (DiSCmini). The device is based on diffusion charging and electrical detection to measure the mean diameter and number concentration of airborne particles in real-time. It has been identified as an interesting candidate for personal workplace exposure monitoring.

First, the limit of quantification and the response time of the DiSCmini were experimentally determined. The results suggest that the DiSCmini can only be used when particle number concentration exceeds (roughly) $10^3$ $cm^{-3}$. The response time of 7.3 seconds indicates that, although the DiSCmini is not suitable for monitoring fast changing concentrations, it should be adequate for most of the events observed in workplaces.

Second, investigations were performed with monodisperse aerosols of particles with diameters between 15 and 400 nm composed of four substances. The experimental data indicate a satisfactory correlation with reference diameter, within ± 38%, and a poorer correlation for number concentrations over the whole range (– 51% to + 65%). A slight tendency to underestimate particle sizes below 40 nm was noted. Data recorded by the DiSCmini were found to be independent of particle chemistry, morphology, and concentration. On the whole, the data reported are within ± 30% of reference values in almost 70% of cases. The correlation between current ratio and particle diameter was also verified as the DiSCmini relies on this to determine the parameters of an aerosol.

Third, the experimental results with polydisperse aerosols indicate that, compared to the modal diameter obntained from SMPS number size distributions, the DiSCmini underestimates particle size by – 20% on average, while particle number concentrations are found to be greater than the reference (+ 55% on average). It should be recalled that the aerosols are assumed to be lognormally distributed with a *GSD* of 1.7 in the DiSCmini. This was not necessarily the case for all test aerosols. Finally, the lung-deposited surface area provided by the DiSCmini was found to be within ± 60% of the reference (NSAM).

The experimental results provided in this study help to describe the behavior of the instrument, although only one model was investigated. Further experiments with several models placed in parallel would help to increase the robustness of results. Although not necessarily within the tolerated interval of ± 30%, the data reported by the DiSCmini correlated well with reference data for most cases in this laboratory study: all particle diameters, number concentrations and alv-LDSA measured were found to be in the same order of magnitude as reference data. However, it should be remembered that the DiSCmini is neither a particle counter nor a particle sizer.

Among the major drawbacks of this instrument are its limited size range. Consequently, it is recommended to use the DiSCmini in combination with other devices when seeking to record information over a large size range, *e.g.* up to a few micrometers. This is particularly important in the case of occupational exposure when handling nanomaterials in powder. Furthermore, the case of bimodal aerosols remains open. Finally, like other personal diffusion chargers, the DiSCmini is not specific to airborne particles, *i.e.*, it cannot distinguish them from those present in the background[67]. However, we believe it can be used as a screening tool to locate sources of airborne particles (provided that they have a diameter below 400 nm), and to quantify the emissions related to various tasks (provided that their concentration is above $10^3$ $cm^{-3}$).

From the information provided in this study, which complement those already published[58, 64], it can be now considered that the performance of the DiSCmini under controlled conditions in the laboratory are well known. However, field data using the DiSCmini are still poor[66-68]; thus, we suggest that studies should be henceforth conducted in this direction.

Ultimately, we believe that this handheld instrument is suitable for stationary or personal measurements of ultrafine particles in workplace atmospheres such as welding fumes or diesel emissions. Regarding the workplaces where nanomaterials are fabricated or handled, particularly when aerosols are highly polydisperse and/or composed of micron-sized particles, this instrument shall not be used alone, as already recommended for other types of instruments[28].

## Notes and references

[a] Institut National de Recherche et de Sécurité (INRS), Laboratoire de Métrologie des Aérosols, Rue du morvan CS 60024 54519 Vandoeuvre les Nancy Cedex, France.

1. W. John, in *Aerosol measurement: Principles, techniques and applications*, eds. P. Kulkarni, P. A. Baron and K. Willeke, John Wiley & Sons, Inc., Hoboken, New Jersey, 4th edn., 2011, pp. 41-54.
2. W. C. Hinds, *Aerosol Technology. Properties, behavior and measurement of airborne particles (2nd Edition)*, 1999, New York: John Wiley & Sons, 483 p.
3. D. Bémer, R. Régnier, I. Subra, B. Sutter, M. T. Lecler and Y. Morele, *Ann. Occup. Hyg.*, 2010, **54**, 607-614.
4. D. H. Brouwer, J. H. J. Gijsbers and M. W. M. Lurvink, *Ann. Occup. Hyg.*, 2004, **48**, 439-453.
5. J. Y. Park, P. C. Raynor, A. D. Maynard, L. E. Eberly and G. Ramachandran, *Atmos. Environ.*, 2009, **43**, 502-509.
6. Z. Xu, X. Li, C. Guan and Z. Huang, *J. Aerosol Sci.*, 2014, **76**, 28-38.
7. A. Neer and Ü. Ö. Köylü, *Combust. Flame*, 2006, **146**, 142-154.
8. N. Zimmerman, K. J. Godri Pollitt, C.-H. Jeong, J. M. Wang, T. Jung, J. M. Cooper, J. S. Wallace and G. J. Evans, *Atmos. Environ.*, 2014, **86**, 140-147.
9. A. J. Koivisto, T. Hussein, R. Niemelä, T. Tuomi and K. Hämeri, *Atmos. Environ.*, 2010, **44**, 2140-2146.
10. J. H. Park, I. A. Mundunkotuwa, J. S. Kim, A. Stanam, P. S. Thorne, V. H. Grassian and T. M. Peters, *Aerosol Sci. Technol.*, 2014, **48**, 768-776.
11. L. Jiang, A. Kondo, M. Shigeta, S. Endoh, M. Uejima, I. Ogura and M. Naito, *J. Occup. Environ. Hyg.*, 2014, **11**, 658-664.
12. K. Savolainen, P. Pylkkänen, H. Norppa, G. Falck, H. Lindberg, T. Tuomi, M. Vippola, H. Alenius, K. Hämeri, J. Koivisto, D. Brouwer, D. Mark, D. Bard, M. Berges, E. Jankowska, M. Posniak, P. Farmer, R. Singh, F. Krombach, P. Bihari, G. Kasper and M. Seipenbusch, *Safety Sci.*, 2011, **48**, 957-963.
13. P. C. Raynor, J. I. Cebula, J. S. Spangenberger, B. A. Olson, J. M. Dasch and J. B. D'Arcy, *J. Occup. Environ. Hyg.*, 2011, **9**, 1-13.
14. P. Nilsson, C. Isaxon, A. Eriksson, M. Messing, L. Ludvigsson, J. Rissler, M. Hedmer, H. Tinnerberg, A. Gudmundsson, K. Deppert, M. Bohgard and J. Pagels, *J. Nanopart. Res.*, 2013, **15**, 2052.
15. E. Zimmermann, S. Derrough, D. Locatelli, C. Durand, J. L. Fromaget, E. Lefranc, X. Ravanel and J. Garrione, *J. Nanopart. Res.*, 2012, **14**, 1209.
16. C. Motzkus, C. Chivas-Joly, E. Guillaume, S. Ducourtieux, L. Saragoza, D. Lesenechal, T. Macé, J.-M. Lopez-Cuesta and C. Longuet, *J. Nanopart. Res.*, 2012, **14**, 687.
17. A. Clemente, M. Pilar Lobera, F. Balas and J. Santamaria, *J. Hazard. Mater.*, 2014, **280**, 226-234.
18. G. Ramachandran, *Ann. Occup. Hyg.*, 2008, **52**, 287-301.
19. A. Pietroiusti and A. Magrini, *Occup. Med.*, 2014, **64**, 319-330.
20. M. N. Shepard and S. Brenner, *Ann. Occup. Hyg.*, 2013, **58**, 251-265.
21. P. T. O'Shaughnessy, *Environ. Sci.: Processes Impacts*, 2013, **15**, 49-62.
22. S. C. Gordon, J. H. Butala, J. M. Carter, A. Elder, T. Gordon, G. Gray, P. G. Sayre, P. A. Schulte, C. S. Tsai and J. West, *Regul. Toxicol. Pharm.*, 2014, **68**, 305-311.
23. J. Wang, C. Asbach, H. Fissan, T. Hülser, T. A. J. Kuhlbusch, D. Thompson and D. Y. H. Pui, *J. Nanopart. Res.*, 2011, **13**, 1373-1387.
24. D. Brouwer, B. Van Duuren-Stuurman, M. Berges, E. Jankowska, D. Bard and D. Mark, *J. Nanopart. Res.*, 2009, **11**, 1867-1881.
25. D. Brouwer, *Toxicol.*, 2010, **269**, 120-127.
26. T. Kuhlbusch, C. Asbach, H. Fissan, D. Göhler and M. Stintz, *Part. Fiber Toxicol.*, 2011, **8**, 22.
27. G. Ramachandran, M. Ostraat, D. E. Evans, M. M. Methner, P. O'Shaughnessy, J. D'Arcy, C. L. Geraci, E. Stevenson, A. D.

Maynard and K. Rickabaugh, *J. Occup. Environ. Hyg.*, 2011, **8**, 673-685.
28. O. Witschger, O. LeBihan, M. Reynier, C. Durand, A. Marchetto, E. Zimmermann and D. Charpentier, *Hygiène et Sécurité au Travail*, 2012, **226**, 41-55.
29. M. Methner, L. Hodson and C. Geraci, *J. Occup. Environ. Hyg.*, 2010, **7**, 127-132.
30. M. L. Ostraat, J. W. Thornburg and Q. G. J. Malloy, *Environ. Eng. Sci.*, 2013, **30**, 126-132.
31. M. Methner, L. Hodson, A. Dames and C. Geraci, *J. Occup. Environ. Hyg.*, 2010, **7**, 163-176.
32. M. Methner, C. Beaucham, C. Crawford, L. Hodson and C. Geraci, *J. Occup. Environ. Hyg.*, 2012, **9**, 543-555.
33. P. C. Raynor, J. Ingraham Cebula, J. S. Spangenberger, B. A. Olson, J. M. Dasch and J. B. D'Arcy, *J. Occup. Environ. Hyg.*, 2012, **9**, 1-13.
34. A. J. Koivisto, J. Lyyränen, A. Auvinen, E. Vanhala, K. Hämeri, T. Tuomi and J. Jokiniemi, *Inhal. Toxicol.*, 2012, **24**, 839-849.
35. T. M. Peters, W. A. Heitbrink, D. E. Evans, T. J. Slavin and A. D. Maynard, *Ann. Occup. Hyg.*, 2006, **50**, 249-257.
36. W. A. Heitbrink, L.-M. Lo and K. H. Dunn, *J. Occup. Environ. Health*, 2014, in press, DOI: 10.1080/15459624.15452014.15930559.
37. L. Morawska, H. Wang, Z. Ristovski, R. E. Jayaratne, G. Johnson, H. C. Cheung, X. Ling and C. He, *J. Environ. Monit.*, 2009, **11**, 1758-1773.
38. T. M. Peters, S. Elzey, R. Johnson, H. Park, V. H. Grassian, T. Maher and P. O'Shaughnessy, *J. Occup. Environ. Hyg.*, 2009, **6**, 73-81.
39. D. Imhof, E. Weingartner, U. Vogt, A. Dreiseidler, E. Rosenbohm, V. Scheer, R. Vogt, O. J. Nielsen, R. Kurtenbach, U. Corsmeier, M. Kohler and U. Baltensperger, *Atmos. Environ.*, 2005, **39**.
40. M. Mohr, U. Lehmann and J. Rütter, *Environ. Sci. Technol.*, 2005, **39**, 2229-2238.
41. T. Walser, S. Hellweg, R. Juraske, N. A. Luechinger, J. Wang and M. Fierz, *Sci. Total Environ.*, 2012, **420**, 119-126.
42. J. Binnig, J. Meyer and G. Kasper, *J. Aerosol Sci.*, 2007, **38**, 325-332.
43. CEN, *Workplace exposure-guide for the use of direct-reading instruments for aerosol monitoring - Part 2: Evaluation of airborne particle concentrations using Optical Particle Counters*, 2009, CEN/TR 16103.
44. P. Görner, X. Simon, D. Bémer and G. Lidén, *J. Environ. Monit.*, 2012, **14**, 420-428.
45. T. M. Peters, D. Ott and P. T. O'Shaughnessy, *Ann. Occup. Hyg.*, 2006, **50**, 843-850.
46. G. Buonanno, R. E. Jayaratne, L. Morawska and L. Stabile, *Aerosol Air Qual. Res.*, 2014, **14**, 156-167.
47. D. J. H. Vosburgh, B. K. Ku and T. M. Peters, *Ann. Occup. Hyg.*, 2014, in press.
48. J. Leskinen, J. Joutsensaari, J. Lyyränen, J. Koivisto, J. Ruusunen, M. Järvelä, T. Tuomi, K. Hämeri, A. Auvinen and J. Jokiniemi, *J. Nanopart. Res.*, 2012, **14**, 718-733.
49. L. Stabile, E. Cauda, S. Marini and G. Buonanno, *Ann. Occup. Hyg.*, 2014, **58**, 860-876.
50. K. S. Woo, D. R. Chen, D. Y. H. Pui and W. E. Wilson, *Aerosol Sci. Technol.*, 2001, **34**, 57-65.
51. A. D. Maynard, *Ann. Occup. Hyg.*, 2003, **47**, 123-144.
52. S. Bau, O. Witschger, F. Gensdarmes and D. Thomas, *J. Nanopart. Res.*, 2013, **15**, 2104.
53. O. Witschger, in *Nanoethics and nanotoxicology*, eds. P. Houdy, M. Lahmani and F. Marano, Springer Verlag, Berlin Heidelberg, 4th edn., 2011, pp. 163-199.
54. M. Fierz, C. Houle, P. Steigmeier and H. Burtscher, *Aerosol Sci. Technol.*, 2011, **45**, 1-10.
55. M. Fierz, H. Burtscher, P. Steigmeier and M. Kasper, *SAE Technical Paper*, 2007, 08PFL-484.
56. S. Bau, J. Jacoby and O. Witschger, *J. Environ. Monit.*, 2012, **14**, 1014-1023.
57. Matter Aerosol, *DiSCmini instruction manual Rev. 1.10*, 2012.
58. C. Asbach, H. Kaminski, D. von Barany, T. A. J. Kuhlbusch, C. Monz, N. Dziurowitz, J. Pelzer, K. Vossen, K. Berlin, S. Dietrich, U. Götz, H. J. Kiesling, R. Schierl and D. Dahmann, *Ann. Occup. Hyg.*, 2012, **56**, 606-621.
59. H. Jung and D. B. Kittelson, *Aerosol Sci. Technol.*, 2005, **39**, 902-911.
60. B. K. Ku and A. D. Maynard, *J. Aerosol Sci.*, 2005, **36**, 1108-1124.
61. B. K. Ku, *J. Aerosol Sci.*, 2010, **41**, 835-847.
62. S. Bau, O. Witschger, F. Gensdarmes and D. Thomas, *J. Nanopart. Res.*, 2012, **14**, 1217-1233.
63. ICRP, *Publication 66: Human respiratory tract model for radiological protection*, 1994, Oxford: Pergamon.
64. J. B. Mills, J. H. Park and T. M. Peters, *J. Occup. Environ. Hyg.*, 2013, **10**, 250-258.
65. J. B. Mills, Evaluation of the DiSCmini personal aerosol monitor for submicrometer sodium chloride and metal aerosols, University of Iowa, 2013.
66. R. Meier, K. Clark and M. Riediker, *Aerosol Sci. Technol.*, 2013, **47**, 22-28.
67. S. Oh, B. Kim and H. Kim, *Industrial Health*, 2014, in press.
68. B. Kim, H. Kim and I. J. Yu, *Industrial Health*, 2014, **52**, 152-162.
69. J. Jacoby, S. Bau and O. Witschger, *J. Phys. Conf. Ser.*, 2011, **304**, 012014.
70. K. R. Sabroske, D. A. Hoying and D. C. Rabe, *Laskin nozzle particle generator*, 1996, US patent number 5 498 374.
71. S. Bau, D. Bémer, F. Grippari, J.-C. Appert-Collin and D. Thomas, *J. Nanopart. Res.*, 2014, **16**, 2629.
72. A. A. Lall and S. K. Friedlander, *J. Aerosol Sci.*, 2006, **37**, 260-271.
73. H. Fissan, S. Neumann, A. Trampe, D. Y. H. Pui and W. G. Shin, *J. Nanopart. Res.*, 2007, **9**, 53-59.
74. W. G. Shin, D. Y. H. Pui, H. Fissan, S. Neumann and A. Trampe, *J. Nanopart. Res.*, 2007, **9**, 61-69.
75. C. Asbach, H. Fissan, B. Stahlmecke, T. A. J. Kuhlbusch and D. Y. H. Pui, *J. Nanopart. Res.*, 2009, **11**, 101-109.
76. R. C. Flagan, in *Aerosol measurement: principles, techniques and applications*, eds. P. A. Baron and K. Willeke, 2001.
77. L. Ntziachristos, A. Polidori, H. Phuleria, M. D. Geller and C. Sioutas, *Aerosol Sci. Technol.*, 2007, **41**, 571-580.