Comparability of Portable Nanoparticle Exposure Monitors*

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Five different portable instrument types to monitor exposure to nanoparticles were subject to an intensive intercomparison measurement campaign. Four of them were based on electrical diffusion charging to determine the number concentration or lung deposited surface area (LDSA) concentration of airborne particles. Three out of these four also determined the mean particle size. The fifth instrument type was a handheld condensation particle counter (CPC). The instruments were challenged with three different log-normally distributed test aerosols with modal diameters between 30 and 180 nm, varying in particle concentration and morphology. The CPCs showed the highest comparability with deviations on the order of only ±5%, independent of the particle sizes, but with a strictly limited upper number concentration. The diffusion charger-based instruments showed comparability on the order of ±30% for number concentration, LDSA concentration, and mean particle size, when the specified particle size range of the instruments matched the size range of the aerosol particles, whereas significant deviations were found when a large amount of particles exceeded the upper or lower detection limit. In one case the reported number concentration was even increased by a factor of 6.9 when the modal diameter of the test aerosol exceeded the specified upper limit of the instrument. A general dependence of the measurement accuracy of all devices on particle morphology was not detected.

Keywords: CPC; diffusion charger; exposure; nanoparticle; number concentration; surface area concentration

INTRODUCTION

The use of nanomaterials has significantly increased over the recent years. According to the Woodrow Wilson Database (2011), the number of nanotechnological products has risen by >520% between March 2006 and March 2011. Nanoparticles are considered as important building blocks for nanotechnologies (Rotello, 2004), but concerns have been raised about possible adverse health effects of nanoparticles (Oberdörster et al., 2005). A risk may only arise from nanomaterials, such as nanoparticles, if exposure and hazard exist simultaneously (Krug and Klug, 2008).
Besides toxicity testing, the determination of exposure to nanomaterials is hence crucial in view of risk assessment. Inhalation is considered as the most critical uptake route for nanomaterials, because especially nanoscale particles in air have the highest mobility and can penetrate deep into the lung. Exposure to airborne nanomaterials can generally occur during any stage of the life cycle of a nanomaterial, i.e. during production, handling, and processing of the particles as well as during the use of products containing nanomaterials including recycling. However, exposure to airborne nanomaterials is considered to be most likely in occupational settings during production, handling, and processing. A number of studies concerning exposure to airborne nanomaterials have been conducted in recent years. The American National Institute of Occupational Safety and Health released a nanotechnology emission assessment technique (Methner et al., 2010a,b) that follows a tiered approach for the investigation of a possible release of and consequent exposure to airborne nanomaterials. The German Chemical Industry Association (VCI, 2011) recently also presented a tiered approach for the assessment of exposure to nanomaterials in workplaces. The first tier foresees data gathering for the respective workplace. If exposure to nanomaterials cannot be excluded, the possible presence of nanomaterials in a workplace is assessed in a second tier using portable, easy-to-use devices. If this assessment provides evidence for the presence of nanomaterials in the air, a more detailed study is conducted in the third tier. Most of the studies conducted thus far [summarized in Brouwer et al. (2009) and Kuhlbusch et al. (2011)] focused on the third tier and used a large set of bulky equipment to study the exposure in detail, including different concepts for assessing the ubiquitous background concentration. The equipment used for these studies usually included size distribution measuring devices like electrical mobility particle sizers (Wang and Flagan, 1990; Tammet et al., 2002) for submicron and aerodynamic particle sizers (Baron, 1986) and photometers for micron particles. In addition, particles were commonly sampled on filters, transmission electron microscope (TEM) grids, or scanning electron microscope (SEM) substrates for consecutive chemical and/or morphological analysis. The equipment used for the third tier is well established but commonly designed to be used by trained personnel only and data analysis is usually rather complex. Instruments for exposure assessment in the second tier need to be easy to use, battery operated, portable, and able to deliver a limited but meaningful data set to estimate exposure levels. Requirements for accuracy can be lower than for the bulky equipment, because in Tier 2 the interest is more on the order of magnitude of the concentration level rather than the exact value. Several devices have been developed in the past that fulfill the aforementioned requirements, but no studies have so far been published on their performance and accuracy when challenged with different aerosols. In this study, we compared the reaction of five different portable, battery operated nanoparticle monitors that can be used in Tier 2 exposure assessment.

INSTRUMENTS IN THE STUDY

Five different instrument types were used in the intercomparison study. Four of them are based on unipolar diffusion charging of the particles and consecutive measurement of the particle-induced current. The fifth type was a handheld version of a condensation particle counter (CPC). At least two copies of each device [except for the Aerotrak (9000)] were tested to study their interdevice comparability. The operation principles of the various devices in the test are summarized in the following. An overview of most of the devices used in the test was recently published by Pelzer et al. (2010).

Handheld CPC

Altogether six identical handheld CPCs (model 3007, TSI, Shoreview, MN, USA; named CPC 1–CPC 6 here) were used in the study. The instrument operates at a total flow rate of 0.7 l min⁻¹, out of which only 0.1 l min⁻¹ is used for the measurement, whereas the rest is used as a bypass flow. The sample flow passes a saturator where the air is supersaturated with isopropyl alcohol vapor, followed by a condenser, where the alcohol condenses onto the particles to form droplets that are sufficiently large to scatter the following laser beam. The interruption of the laser beam by the droplets is used to count the particles and determine the particle number concentration. A handheld CPC can measure particles between 10 nm and >1 µm (according to TSI specification sheet) in size with 1-s time resolution. The upper size limit is mainly caused by particle losses inside the CPC, but has never been investigated in detail. For most applications, as in the study presented here, it is however of minor importance because of the usually low number concentrations in the micron size range. The handheld CPC only uses single particle counting and the total number concentration is limited to 100 000 cm⁻³ (Hämeri et al., 2002). Above the upper concentration limit, more than one particle can be present in the measurement cell, causing coincidence errors. From personal experience of several authors of this study, the
upper limit varies significantly between different handheld CPC devices. While some work properly even >200 000 cm−3, others reach the coincidence limit already at only ~80 000 cm−3. A generally good agreement of the handheld CPC (model 3007) with the simpler version P-Trak of the same manufacturer has been reported by Matson et al. (2004), despite different lower size detection limits of the instruments. The specification sheet of the handheld CPC specifies the measurement accuracy to be ±20%.

The nanoTracer (Philips Aerosense, Eindhoven, the Netherlands; Marra et al., 2010) uses a unipolar diffusion charger to electrically charge the aerosol particles. The particle size distribution is manipulated in a low-efficiency electrostatic precipitator (ESP). Intermittently, a low voltage to remove only ions and a higher voltage to manipulate the particle size distribution are applied to the ESP. The current from the particles exiting the ESP is continuously measured with a Faraday cup electrometer. Total particle number concentration and mean particle size are derived from the current values during the two periods with different ESP voltages under the assumption of a log-normal particle size distribution. According to the manual, the total number concentration is limited to 10^6 cm−3 and the modal diameter of the particle size distribution to a range between 20 and 120 nm. The nanoTracer time resolution is 16 s if number concentration and average particle size are determined. It can be shortened to 3 s, but then only the number concentration is measured. The latter mode was not used in this study. The accuracy of the instrument is specified in the nanoTracer handbook to be ±1500 cm−3 for the concentration and ±10 nm for the average particle size. The instrument uses a fan to establish the aerosol flow rate of 0.3–0.4 1 min−1, which on the one hand reduces noise and battery consumption but on the other hand does not allow the use of a pre-separator for large particles. Instead, the nanoTracer sets limitations for size and concentration gradients. Three copies of the nanoTracer were used in this study.

The nanoCheck (model 1.320, Grimm Aerosoltechnik, Ainring, Germany) is based on the same measurement principle as the nanoTracer, but is used as an extension of the Grimm aerosol spectrometer (model 1.109) which measures size distributions in a size range from 0.25 to 32 µm optically. The nanoCheck derives information on the particle number concentration and average particle diameter in a range from 25 to 300 nm assuming log-normal size distribution with a time resolution of 10 s and an accuracy of ±5%, according to the manufacturer’s website. The concentration range is specified by the manufacturer from 5 × 10^2 to 5 × 10^5 cm−3. The instrument operates at a total flow rate of 1.2 l min−1.

Two slightly different models were used in the test, an older version with a nickel electrode and a newer version with gold-coated electrode.

The Aerotrak 9000 (TSI, Shoreview, MN, USA) is a portable and battery operated version of the nanoparticle surface area monitor (NSAM, TSI model 3550; Fissan et al., 2007; Shin et al., 2007) and measures the fraction of airborne particle surface area concentration that would deposit either in the alveolar or the tracheobronchial region of the human lung. Particles...
>1 µm are removed in a cyclone pre-separator and the total incoming flow of 2.5 l min⁻¹ is split into a 1.5 l min⁻¹ aerosol flow and a 1.0 l min⁻¹ ion jet flow. The ion jet flow is completely filtered and passes a corona needle where a high concentration of ions is produced, when a high voltage is applied. The ion jet flow convectively transports these ions through an orifice into a mixing chamber where it meets the aerosol flow. Ions diffusively attach to the particles in the mixing chamber, before the aerosol is led through an ion trap. The ion trap voltage is adjusted to either 100 or 200 V to manipulate the particle size distribution. The eventually measured current is then proportional to the fraction of surface area concentration that is deposited in the tracheobronchial or alveolar region, respectively. The particle size range is specified by the manufacturer to be from 10 to 1000 nm, but Asbach et al. (2009a) showed for NSAM that accurate results can only be obtained between 20 and 400 nm. While the lower end usually does not significantly bias the overall measurement accuracy due to the commonly negligible contribution of sub-20 nm particles to the total surface area concentration, errors caused by particles >400 nm can be significant because of the squared dependency of the particle surface area. The use of a pre-separator with lower cut-off is hence advisable (Asbach et al., 2011). The proposed pre-separator requires a modification of the internal flow control orifices, which was not possible here. The Aerotrac was instead used with a cyclone with 1 µm cut-off, provided by the manufacturer. The concentration range of the Aerotrak is 0–2500 µm² with 1 µm cut-off, provided by the manufacturer. The ion jet flow is completely filtered and passes a corona needle where a high concentration of ions is produced, when a high voltage is applied. The ion jet flow convectively transports these ions through an orifice into a mixing chamber where it meets the aerosol flow. Ions diffusively attach to the particles in the mixing chamber, before the aerosol is led through an ion trap. The ion trap voltage is adjusted to either 100 or 200 V to manipulate the particle size distribution. The eventually measured current is then proportional to the fraction of surface area concentration that is deposited in the tracheobronchial or alveolar region, respectively. The particle size range is specified by the manufacturer to be from 10 to 1000 nm, but Asbach et al. (2009a) showed for NSAM that accurate results can only be obtained between 20 and 400 nm. While the lower end usually does not significantly bias the overall measurement accuracy due to the commonly negligible contribution of sub-20 nm particles to the total surface area concentration, errors caused by particles >400 nm can be significant because of the squared dependency of the particle surface area. The use of a pre-separator with lower cut-off is hence advisable (Asbach et al., 2011). The proposed pre-separator requires a modification of the internal flow control orifices, which was not possible here. The Aerotrac was instead used with a cyclone with 1 µm cut-off, provided by the manufacturer. The concentration range of the Aerotrak is 0–2500 µm² cm⁻³ for tracheobronchial and 0–10 000 µm² cm⁻³ for alveolar deposition. The LDSA concentration is measured with 1-s time resolution and accuracy specified by the manufacturer to be ±20%.

**FMPS**

The FMPS (TSI model 3091) is based on electrical mobility analysis to measure particle size distributions in a size range from 5.6 to 560 nm with 1-s time resolution. The FMPS was used to monitor the size distribution of the test aerosol and to calculate LDSA concentrations for comparison with Aerotrak 9000 and miniDiSC. The FMPS was shown to generally agree well with SMPS (Asbach et al., 2009b; Jeong and Evans, 2009).

**METHODS**

Experiments were conducted at the NanoTestCenter at the Institute for the Research on Hazardous Substances (IGF) in Dortmund, Germany. The set up has been used for numerous intercomparison studies in the past (e.g. Dahmann et al., 2001; Asbach et al., 2009b). In the NanoTestCenter, aerosols can be produced from various sources and are introduced into an ~20-m long wind tunnel with a diameter of 0.7 m, where they mix with clean dilution air. The dilution air flow rate is adjustable between 200 and 1000 m³ h⁻¹ (resulting velocity in wind tunnel 0.44–2.2 m s⁻¹) to provide predefined particle concentration levels. The wind tunnel feeds into a mixing chamber with a volume of ~20 m³, which is ventilated by a large blower that also defines the dilution air flow rate in the wind tunnel. The aerosol has been shown to be homogeneously distributed in the mixing chamber so that all instruments located inside the chamber sample aerosol with identical properties. Further details of the NanoTestCenter are described in a previous publication (Asbach et al., 2009b). In this study, all measurement devices were simultaneously located inside the mixing chamber and sampling directly into their device inlet without any tubes attached, in order to avoid diffusion losses in sampling trains. They were placed on a table, with sufficient distance to each other to avoid interferences, inside the chamber and sampled at approximately the same height. The clocks of all instruments were synchronized prior to the first measurement on each day.

Three different types of aerosols were selected for the intercomparison: sodium chloride (NaCl), di-ethyl-hexyl-sebacate (DEHS, C₂₆H₅₀O₄, obtained from Topas GmbH) and soot particles. NaCl aerosol was produced by spraying an NaCl solution (1 g NaCl per 1 l deionized water) with an atomizer (model ATM 226, Topas GmbH, Dresden, Germany) and dried to RH < 30% in a homemade silica gel dryer before being introduced into the wind tunnel. NaCl is known to produce compact, cubic particles with an average size of ~50 nm with the aforementioned solution. DEHS particles were generated by spraying pure DEHS using the same atomizer. Atomized DEHS forms spherical droplets with sizes of ~200 nm. Soot was produced using a spark generator (GFG 3000, Palas GmbH, Karlsruhe, Germany) with graphite electrodes and chosen because of the fractal-like structure of the produced particles. The spark generator was operated at a total current of 14.6 mA and an argon flow rate of 5 l min⁻¹. Particle sizes with these spark generator settings are commonly ~30 nm. The choice of these three materials hence allowed for the study of the reaction of the different instruments to different particle sizes, morphologies, and—along with different dilution air flow rates—different concentration levels. Particle size distributions, measured with an FMPS are shown in Fig. 1,
normalized with respect to the total particle number concentration. The normalized particle size distributions did not change significantly with different dilution rates in the wind tunnel, proving that the dilution rate is high enough to avoid coagulation effects and massive particle losses in the wind tunnel. All size distributions follow more or less a log-normal shape. Small deviations of log-normal distributions in case of the NaCl and DEHS aerosol in Fig. 1 are likely caused by FMPS artifacts that have been reported several times in the literature (Asbach et al., 2009b; Jeong and Evans, 2009).

Besides the investigation of the reaction of the instruments to different concentration levels, the influence of concentration gradients was also studied by producing concentration profiles as shown in Fig. 2. Before each test run, the mixing chamber was flushed with clean air, until the background concentration in the chamber had reached a level well below 1000 cm⁻³, before the respective aerosol generator was connected to the wind tunnel. Two concentration levels were aimed at, one at ~100 000 and another one at ~20 000 cm⁻³, followed by a quick increase and decrease of the concentration to study whether quick concentration changes affect the measurement accuracy. Initially the DEHS produced significantly higher concentrations, but was consequently reduced to ~100 000 cm⁻³. For the soot particles, only one concentration level of ~80 000 cm⁻³ was chosen. To check whether a possible morphology effect on the unipolar charging efficiency (Shin et al., 2010) affects the different dynamic behavior when exposed to strong concentration gradients, the soot concentration was later quickly increased to ~80 000 cm⁻³ before the generator was switched off and the chamber flushed with clean air again. Experimental runs with NaCl and DEHS aerosol lasted for ~3 h, soot experiments for ~2 h.

RESULTS AND DISCUSSION

**Comparison between handheld CPCs**

Six identical handheld CPCs (model 3007, TSI Inc., Shoreview, MN, USA) were simultaneously tested. They were challenged with NaCl, DEHS, and soot aerosol following concentration profiles as shown in Fig. 2. CPC 1 was freshly calibrated and used as an internal reference for the particle number concentration. Since the handheld CPC may show coincidence errors >80 000 cm⁻³, no higher number concentrations were taken into account for the analysis. Figure 3 shows the correlation of CPC 2–CPC 6 with CPC 1, based on 1-s number concentration data. The black line in Fig. 3 indicates 1:1 correlation, i.e. ideally all data points would fall onto this line. The grey line represents the linear fit to the data points. The corresponding equations and correlation coefficients $R^2$ are also given in each graph. It can be seen that the correlation coefficient is $R^2 > 0.99$.
for all CPCs in the test, independent of the particle material and corresponding size distributions. This shows that the particle detection mechanism used in the handheld CPCs is very reproducible and particle material independent. Figure 3 furthermore shows that the y-intercepts of the fit equations are small and that for CPC 2 and CPC 4–CPC 6 the slope of the fit equations are close to one with maximum ±5% deviation. The small bump in the concentration of CPC 5 at ~60 000 cm$^{-3}$ was caused by improper synchronization of the CPC 5 and CPC 1 clocks. While the lower data points represent measurements during increasing concentration (see Fig. 2), the higher data points represent decreasing concentrations.

These results indicate that the comparability of the handheld CPC was even higher than specified by the manufacturer (±20% accuracy). Only CPC 3 showed a lower agreement with the other CPCs, with a slope on the order of 0.82, hence under-representing the number concentration by ~18%. This would still be in the accuracy range specified by the manufacturer. The correlation coefficient $R^2$ on the order of 0.998, however, indicates a very high correlation. A later check of the instrument revealed that the observed deviation was caused by inaccurate internal splitting of the total flow into the sample and bypass flow inside the instrument, caused by partial clogging of the flow control orifice. This cannot be detected by the user, because the instrument does not generate an error message and the total flow at the inlet was still within the range of specification.

In general the results show that the handheld CPCs measured the particle number concentration in the given size range very accurately. They did not show any dependence on the materials or size distributions used. The main drawback of a handheld CPC is that it uses a liquid that needs to be refilled every 6–8 h and requires the instrument to be maintained in a horizontal orientation during all measurements. Battery operation is usually also only possible for 5–6 h before the batteries need to be recharged.

**Performance of miniDiSC**

Two identical miniDiSCs were challenged with the test aerosol described above. Figure 4 shows the correlation analyses for the number concentration measured with these two devices in comparison with the CPC 1. Very good correlations between the measurement results were obtained with correlation coefficients $R^2$ ~ 0.99. The miniDiSCs did show different levels of agreement with CPC 1 for the three different types of aerosol, however all of them are well in the specified accuracy range of ±30%. Both instruments showed on average a 25% (miniDiSC 1) and 17% (miniDiSC 2), respectively, higher number concentration when challenged with NaCl aerosol and 17% (miniDiSC 1) and 25% (miniDiSC 2) with soot particles, whereas both underestimated the DEHS concentration by ~8%. This shows that the miniDiSC accuracy seems to be more dependent on the particle size than on the particle morphology, because NaCl and soot both showed very similar deviations, although the particles had very different morphologies. The DEHS particles were significantly larger than NaCl and soot (see Fig. 1) and delivered noticeably different deviation from the CPC reading. All regression analyses show a higher y-intersect than the CPC regressions (see Fig. 3).
Fig. 3. Regression analyses for the total number concentration measured with CPC 2–CPC 6 compared with CPC 1, tested with NaCl (left), DEHS (center), and soot (right) particles; all data are 1-s measurement values.
This indicates that the miniDiSC shows decreasing accuracy for lower particle concentrations, which is expected because of a lower signal to noise ratio in the electrometer. This is also reflected by the lower concentration limit specified to be 1000 cm$^{-3}$. Both miniDiSCs followed the steep concentration gradient without problems.

Comparison of the fit equations for the two instruments with each other reveals a good intercomparability of the two devices. This is of high importance, especially when used in the second tier of the abovementioned tiered approach for exposure assessment, where one instrument is used to monitor the particle background whereas the other one is used for measuring the particle number concentration near the process of interest.

The small bumps that can be seen in the regression graphs for soot particles were again caused by improper synchronization of the miniDiSC and CPC 1 clocks.

The mean particle sizes measured by the miniDiSC were furthermore compared with the average particle size calculated from FMPS measurement. Since none of the manufacturers specifies whether their instrument measures the arithmetic or geometric mean diameter, both were calculated and are given in Table 1 in comparison with the mean particle sizes reported by the miniDISCs. The arithmetic mean diameter was calculated as follows:

$$\bar{d}_{p,\text{arithm.}} = \frac{\sum d_{p,i} \cdot n_i}{\sum n_i}$$

### Table 1. Mean particle sizes obtained with miniDiSC in comparison with FMPS.

<table>
<thead>
<tr>
<th></th>
<th>NaCl</th>
<th>DEHS</th>
<th>Soot</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$d_{p,\text{avg}} \pm \text{SD (nm)}$</td>
<td>Number of data points</td>
<td>$d_{p,\text{avg}} \pm \text{SD (nm)}$</td>
</tr>
<tr>
<td>miniDiSC 1</td>
<td>42.8 ± 1.06</td>
<td>1737</td>
<td>181.1 ± 13.95</td>
</tr>
<tr>
<td>miniDiSC 2</td>
<td>40.6 ± 1.01</td>
<td>1737</td>
<td>162.0 ± 12.81</td>
</tr>
<tr>
<td>FMPS (arithm.)</td>
<td>47.9 ± 0.87</td>
<td>1800</td>
<td>176.9 ± 1.83</td>
</tr>
<tr>
<td>FMPS (geom.)</td>
<td>35.7 ± 0.74</td>
<td>1800</td>
<td>143.9 ± 2.53</td>
</tr>
</tbody>
</table>

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**Fig. 4.** Regression analyses for the total number concentration measured with miniDiSC 1 and miniDiSC 2 compared with CPC 1, tested with NaCl (left), DEHS (center), and soot (right) particles; all data are 1-s measurement values.
where \( d_{p,i} \) is the average diameter in the \( i \)th size channel of the FMPS and \( n_i \) is the number concentration measured in the respective channel. The geometric mean was calculated as follows (Hinds, 1999):

\[
\bar{d}_{p,\text{geom.}} = \exp \left( \frac{\sum n_i \cdot \ln(d_{p,i})}{\sum n_i} \right)
\]

Only time periods with very stable concentrations were chosen for the comparison of the average sizes, i.e. the time period with \(~20\,000\,cm^{-3}\) for NaCl and were chosen for the comparison of the average sizes, where the accuracy is lower than specified of the instrument, it should be noted that for higher concentrations the accuracy is lower than specified in case of soot.

The reported average sizes of the two miniDiSCs were very comparable with each other for all three materials with deviations of around \( \pm 10\% \) or below. The agreement with average sizes obtained from FMPS measurements was generally good. For NaCl, the miniDiSC sizes are between the arithmetic and geometric mean with deviations of only a few percent. The average sizes for DEHS are closer to the arithmetic than to the geometric mean, with deviations of \(<10\%\). The highest deviations can be seen for soot particles, where the average sizes reported by the miniDiSCs are 20 and 27% below the arithmetic and 11 and 19% below the geometric mean, respectively. A reason for this difference may be that the FMPS measured an equivalent particle diameter of the agglomerates based on their electrical mobility, whereas in the miniDiSC, size information is derived from mechanical particle deposition on a diffusion screen.

In conclusion, the accuracy of the miniDiSC is well within the specified range of \( \pm 30\% \). While the concentration accuracy was independent of particle material, the sizing accuracy seems to be higher for compact than for fractal-like particles.

**Performance of nanoTracer**

Three identical nanoTracers were compared with the reference handheld CPC. Since the nanoTracer has a time resolution of 16 s, all CPC 1 data were averaged over identical 16-s time periods. As shown in Fig. 5, number concentrations measured with NaCl and soot aerosol were highly correlated with regression coefficients \( R^2 \geq 0.97 \) and showed good agreement with CPC 1 with deviations between 0 and \( +19\% \). Even though this agreement can be considered as quite good, taking into consideration the indirect measurement principle and the small size of the instrument, it should be noted that for higher concentrations the accuracy is lower than specified by the manufacturer, who claims it to be within \( \pm 1500\,cm^{-3} \). The \( y \)-intersects of the nanoTracers were lower (better) than with the miniDiSC during the NaCl and soot measurements, indicating that the nanoTracer may be able to measure lower concentrations with higher accuracy than the miniDiSC. This is not unexpected because of the 16-s time resolution of the nanoTracer, compared with 1 s of the miniDiSC. The electrometer can hence integrate over a longer time to obtain a better signal-to-noise ratio. NanoTracer 3 showed a higher deviation during NaCl experiments, which were most likely caused by the positioning of the instrument on a chamber wall, where the homogeneity of the particle distribution in the chamber may have been affected. During later experiments, the instrument was placed more centrally in the chamber. The results do not show a clear size or morphology dependence of the nanoTracer for the two aerosols with mean particle size \( >100\,nm \) (NaCl and soot). For DEHS with a modal particle diameter of \(~180\,nm\) (see Fig. 1), however, the nanoTracer significantly overestimated the particle number concentrations by a factor of between 3.3 and 6.9. It should, however, be noted that the manufacturer specified the nanoTracer for measuring aerosols with an average diameter between 20 and 120 nm, hence the DEHS size distribution is out of the specification for the instrument. It is anyhow interesting to take a deeper look into the data reported by the nanoTracer for DEHS aerosol. As described above, the nanoTracer uses a limitation for particle size and concentration gradients. When the DEHS aerosol was switched on, both concentration and particle size showed a steep gradient, resulting in a strong increase of the currents measured with the Faraday cup electrometer. It is assumed that the data deconvolution algorithm used in the nanoTracer misinterpreted the strong increase as a very high concentration, resulting in the strong overestimation of the concentration. In addition, the manufacturer, when confronted with the shown DEHS results, responded that ‘…the deeper physical reason underlying the strange results is the quick deposition of an insulating layer of oil (in case of DEHS) on the screen electrode of the diffusion charging section, which effectively raises the electric potential of the screen electrode, leading to a transformation of diffusion charging conditions to more-or-less field charging conditions, thereby much increasing the particle charge. Proper interpretation of the measured signals is then no longer possible...’.
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if the aerosol properties are completely unknown. At least nanoTracers 2 and 3 measured mean particle sizes of 174 nm and 237 nm, respectively (see Table 2), i.e. sizes out of the specified range. To the contrary, nanoTracer 1 showed an average particle size of only 87 nm. It is interesting to note that the instrument that reported the lowest particle size also reported the by far highest number concentration, which even more stresses the assumption of a misinterpretation of the measured current. As summarized in Table 2, the sizing accuracy of the nanoTracer was very good for soot particles, with reported average particle sizes differing only by a few percent from the geometric and arithmetic mean diameter obtained from the FMPS size distribution measurement. The reported average particle sizes for NaCl particles are almost identical from the nanoTracers 1 and 2, nanoTracer 3 reported ~11% larger average diameter than the other two. However, all average sizes reported by the nanoTracers for NaCl are between 29 and 44% higher than the arithmetic mean and between 73 and 93% larger than the geometric mean diameter calculated from FMPS measurements. The good sizing agreement for soot and the worse agreement for NaCl give rise to the assumption that the sizing accuracy of the nanoTracer is probably more

Fig. 5. Regression analyses for the total number concentration measured with nanoTracers 1–3 compared with CPC 1, tested with NaCl (left), DEHS (center), and soot (right) particles; all data are 16-s measurement values, nanoTracer as reported by the device, CPC data averaged over the same 16 s for each data point; note that the average size of the DEHS size distribution was out of the specified range for mean particle sizes; nanoTracer 3 was mounted on a wall during NaCl measurements, which influenced the sample aerosol.
morphology dependent than the accuracy for number concentrations.

In conclusion, the nanoTracers in the test showed a good general agreement with the handheld CPC with at most 19% deviation for the total number concentration, as long as the particle size distribution was within the instruments’ specified size range. Although at higher total concentrations the absolute deviations were higher than the specified accuracy of ±1500 cm\(^{-3}\), it is still a very satisfying result. Care should be taken, when the mode of the particle size distribution exceeds the manufacturer’s specification (20–120 nm), because the reported concentrations can be drastically overestimated.

**Performance of nanoCheck**

Two slightly different versions of the Grimm nanoCheck were used as described above. nanoCheck 1 was the older version. It was later found that the nanoCheck 2 was operated with the electrometer zero-check and control switched off, resulting in a very bad correlation of the device with CPC 1. Results of nanoCheck 2 were therefore omitted. Results of the correlation analyses of nanoCheck 1 in comparison with CPC 1 are shown in Fig. 6.

The correlations between the nanoCheck 1 and CPC 1 were good for NaCl and soot (\(R^2 \geq 0.96\)), but worse for DEHS (\(R^2 = 0.9\)). The overall agreement with CPC 1, however, is only satisfactory for NaCl and DEHS with −18 and +23% deviation, respectively. Even though this deviation is way beyond the 5% accuracy level claimed by the manufacturer, this seems to be the realistic accuracy level of diffusion charger-based number concentration measurements (compare with miniDiSC and nanoTracer). For soot particles, the nanoCheck 1 significantly under-represented the number concentration by ~59%. Unlike the nanoTracer, which otherwise uses the same principle, the nanoCheck removes very small particles on a diffusion screen upstream of the charger. The lower size limit is accordingly specified to be at 25 nm. In case of soot, particles with sizes >25 nm contributed ~42% to the total number concentration (calculated from data in Fig. 1). Bearing in mind that a diffusion screen never shows a sharp cut-off, it is very likely that the discrepancy between CPC and nanoCheck

**Table 2. Mean particle sizes obtained with nanoTracer in comparison with FMPS.**

<table>
<thead>
<tr>
<th></th>
<th>NaCl</th>
<th></th>
<th>DEHS</th>
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<td></td>
<td>(d_{p,\text{avg}} \pm \text{SD (nm)})</td>
<td>Number of data points</td>
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<td>Number of data points</td>
<td>(d_{p,\text{avg}} \pm \text{SD (nm)})</td>
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<tr>
<td>nanoTracer 1</td>
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<td>87.4±3.14</td>
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<td>30.1±0.47</td>
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<td>174.2±8.93</td>
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<td>29.0±0.47</td>
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<tr>
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<td>236.6±17.25</td>
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<td>33.7±0.66</td>
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</tr>
<tr>
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<td>1800</td>
<td>176.9±1.83</td>
<td>1800</td>
<td>31.2±0.43</td>
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<tr>
<td>FMPS (geom.)</td>
<td>35.7±0.74</td>
<td>1800</td>
<td>143.9±2.53</td>
<td>1800</td>
<td>28.2±0.42</td>
<td>1800</td>
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**Fig. 6.** Regression analyses for the total number concentration measured with nanoCheck 1 and 2 compared with CPC 1, tested with NaCl (left), DEHS (center), and soot (right) particles; all data are 10-s measurement values, nanoCheck as reported by the device, CPC data averaged over the same 10 s for each data point.
Comparability of portable nanoparticle exposure monitors is mainly caused by the different size ranges of the instruments. In addition the morphology of the soot particles may also have played a role.

The average particle sizes, reported by the nanoCheck are listed in Table 3 in comparison with results from the FMPS. The delivered average diameters for NaCl and DEHS are quite reasonable and between the arithmetic and geometric mean of the FMPS, but both closer to the arithmetic mean. For soot particles, the nanoCheck consistently reported an average diameter of 25 nm, which is ~25% smaller than the arithmetic mean and 13% smaller than the geometric mean of the FMPS. However, it should be noted that 25 nm is the smallest size reported by the instrument.

In conclusion, the nanoCheck seemed to work reasonably well with a generally similar comparability to CPC measurements like the miniDiSC and nanoTracer, i.e. on the order of ±30%, as long as the particle size range was within the detectable size range of the instrument. The correlation, however, was worse, especially in case of soot particles. Particle morphology did not seem to play a major role for the instrument’s accuracy.

**Performance of miniDiSC and Aerotrak 9000 measuring LDSA**

Two instrument types in the study measured LDSA concentrations directly, Aerotrak 9000 and miniDiSC. The miniDiSC only reports the alveolar surface area concentration, hence the Aerotrak 9000 was also set to alveolar deposition. The number size distributions measured with the FMPS were converted into a surface area size distribution, assuming spherical particles, weighted with the lung deposition curve according to a model published by the International Commission on Radiological Protection (ICRP, publication 66, 1994), and then integrated over the entire size range. The alveolar lung deposition curve according to the ICRP model was obtained using LUDEP software (James et al., 2000). Respiratory input data for the software were identical with those used for the calibration of the Aerotrak, i.e. a reference worker under light exercise, breathing only through the nose with 2200 cm³ functional residual capacity, a breathing rate of 15 breaths per minute and a ventilation rate of 1.3 m³ h⁻¹ (from the instrument’s manual).

The regression analyses of Aerotrak 9000 and the two miniDiSCs against FMPS data are shown in Fig. 7. Again the results were all highly correlated with regression coefficients $R^2 \geq 0.99$. For NaCl, the Aerotrak showed almost perfect agreement with a slope of 1.016. This is not surprising, because the Aerotrak 9000 was originally calibrated with NaCl particles (Shin et al., 2007). For NaCl, both miniDiSCs also showed good agreement with ±6 and −13.5%, respectively, deviation from the FMPS LDSA. The agreement between Aerotrak and FMPS for soot particles was also good with ~10% deviation. Both miniDiSCs reported lower concentrations than the FMPS, miniDiSC 1 by ~10% and miniDiSC 2 by ~17%. It is interesting to note that the miniDiSCs responded to fractal-like agglomerates with an underestimation of the LDSA, whereas the Aerotrak overestimated the LDSA. Both instruments charge the particles to a charge level that is proportional to $d_p^{1.13}$ in case of Aerotrak (Jung and Kittelson, 2005) and proportional to $d_p^{1.1}$ (Fierz et al., 2011), which is approximately the size dependence of the particle surface area weighted with the alveolar or tracheobronchial deposition curve. Both devices therefore derive the LDSA from the total measured current by multiplying it with a constant calibration factor. Differences between Aerotrak and miniDiSC for soot may be caused by the non-negligible fraction of sub-20 nm particles and by different charging efficiencies at the lower particle size end. The assumption of a unique power law for the description of the LDSA only holds for particle sizes up to ~300–400 nm (Asbach et al., 2009a). Above this size, impaction becomes the dominant deposition mechanism in the human lung, whereas below, diffusion is the main deposition mechanism. The combination of the two principles of particle deposition in the human lung, causes the lung deposition curve to show a minimum at ~300–400 nm, which can no longer be described

<table>
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<th>NaCl</th>
<th>DEHS</th>
<th>Soot</th>
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<tr>
<td>nanoCheck 1</td>
<td>46.9 ± 6.81</td>
<td>165.9 ± 30.57</td>
<td>25.0 ± 0.00a</td>
</tr>
<tr>
<td>FMPS (arithm.)</td>
<td>47.9 ± 0.87</td>
<td>176.9 ± 1.83</td>
<td>31.2 ± 0.43</td>
</tr>
<tr>
<td>FMPS (geom.)</td>
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</tr>
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*a25 nm is the minimum diameter reported by the nanoCheck.*

Table 3. Mean particle sizes obtained with nanoCheck in comparison with FMPS.
by the aforementioned power law. For particle sizes above this limit, Aerotrak 9000 and miniDiSC therefore must under-represent the LDSA, which can be seen in the regression analyses for DEHS. Bearing in mind that the particle surface area has a squared dependence on particle diameters, it is obvious that particles >300–400 nm have a significant contribution to the total DEHS surface area (compare with number size distribution shown in Fig. 1). This large surface area contribution of large DEHS particles is likely the reason for the larger discrepancies between Aerotrak and FMPS as well as between miniDiSC and FMPS as shown in Fig. 7. Aerotrak underestimates the total LDSA by ~25%, the two miniDiSCs by 39 and 48%, respectively.

In conclusion, both Aerotrak and miniDiSC are in good agreement with the LDSA concentration obtained from FMPS size distribution measurement for NaCl and soot aerosol. Results for these aerosols are all within the specified accuracy range of ±20 (Aerotrak 9000) and ±30% (miniDiSC). However, both instruments significantly underestimate the LDSA of the DEHS aerosol, caused by the presence of particles >300–400 nm. The particle morphology of soot particles did not seem to affect the measurement.

**SUMMARY AND CONCLUSIONS**

Five different portable instruments that can be used for monitoring particle concentrations, e.g. in workplaces, were subject to an intensive intercomparison study under defined laboratory conditions. Three aerosols with different properties concerning particle sizes and morphologies as well as particle concentrations were used in the study. It was shown...
that the highest comparability for measuring number concentrations was achieved with handheld CPCs, which showed deviations usually below ±5%. Out of the six handheld CPCs in the test, only one produced consistently lower concentrations. This could be tracked back to improper internal flow rate splitting, which could not be detected from outside. The downsides of handheld CPCs are their limitation to a maximum number concentration of 100 000 cm\(^{-3}\) and their need for a working liquid (isopropyl alcohol). On the one hand the liquid needs to be refilled after a few hours and on the other hand requires the instrument to be held in horizontal orientation. Diffusion charger-based instruments (DCs) can be a good alternative to handheld CPCs. They do not require a working liquid and have higher upper concentration limits on the order of 500 000–1 000 000 cm\(^{-3}\). However, DCs use an indirect measurement principle, i.e. charging the particles and measuring the particle induced current following tailored manipulation of the particle size distribution. This indirect measurement principle requires several assumptions on particle size distribution and charging efficiency and therefore DCs may produce less accurate results than handheld CPCs. The manufacturer of the miniDiSC gives an accuracy level of ±30%, which seems to be most realistic. For most monitoring applications, this accuracy is absolutely sufficient. The accuracy level of the nanoTracer was specified by the manufacturer with an absolute value of ±1500 cm\(^{-3}\), which could not always be kept but was rather on the same order as the miniDiSC. The manufacturer of the nanoCheck gives an accuracy level of ±5%, which is certainly too optimistic.

Number concentrations reported by the two miniDiSCs in the test were all within the specified accuracy range with deviations from a freshly calibrated CPC between −8 and +25% for all tested particle materials, sizes, and morphologies. Average particle sizes reported by the miniDiSCs are in line with the defined accuracy range with mean particle sizes calculated from FMPS size distributions. Average particle sizes from miniDiSC generally agreed better with the arithmetic than with the geometric mean of the FMPS measurements. The LDSA concentration measurements agreed similarly well with the LDSA calculated from FMPS measurements for NaCl and soot particles with modal diameter >100 nm. The agreement was significantly worse (−39 and −48%) in the presence of particles >400 nm in case of the DEHS aerosol.

Considering ±30% to be the tolerated accuracy range for DCs rather than the ±1500 cm\(^{-3}\) and ±10 nm, specified by the nanoTracer manufacturer, the three nanoTracers in the test delivered very satisfying results for NaCl and soot particles. For these two aerosols, the nanoTracers showed better agreement with the reference CPC than the miniDiSCs. Deviations were only between −0.6 and +9.3% for NaCl and between 0 and +19% for soot particles. For DEHS, however, which had a modal diameter beyond the specifications of the nanoTracer, all three instruments showed between 250 and 594% higher concentrations, which were likely caused by the presence of too large particles in conjunction with steep concentration gradients. Special attention should therefore be paid to the nanoTracer results when the aerosol size distribution shows a large modal diameter (e.g., indicated by the reported average particle size). The sizing accuracy of the nanoTracer was rather poor for NaCl with deviations between 29 and 42% from the arithmetic and between 73 and 93% from the geometric mean. The agreement was significantly better for soot particles with deviations between −8 and +8% from the arithmetic and +2.8 and +19.5% from the geometric mean. Agreement for DEHS was again worse, likely because of the presence of too large particles, but at least two of the three instruments reported average particle sizes that were out of the specified range and could hence in the future be used for a warning to the user.

The nanoCheck showed deviations of ±19% for compact particles (NaCl and DEHS) but −58% for fractal-like soot. Since the soot particle size distribution showed a large contribution of particles >25 nm, i.e. below the nominal nanoCheck size limit, it is assumed that the rather large discrepancy can be attributed to the unmatched size range. Similarly to the DEHS measurements with the nanoTracer this clearly shows that these devices can only be accurate if the detected size range of the employed instruments is well chosen for the present size range of the aerosol particles. The sizing accuracy of the nanoCheck was in good agreement with the arithmetic mean from FMPS measurements for NaCl and DEHS with −2 and −6.2% deviation, respectively. The reported average size for soot deviated from the arithmetic mean by 19.9%.

Quick concentration changes did not generally affect the measurement accuracy of the instruments. Only the nanoTracers significantly over-represented particle number concentrations following steep gradients of the DEHS concentration, where the mean particle diameter was outside of the specified diameter range.

The LDSA concentration measured with an Aerostrak was in good agreement with the LDSA calculated from FMPS measurements for NaCl and soot.
particles (deviations between +1.6 and +10.2%), but the presence of particles >400 nm caused a larger under-representation (~25%) of the LDSA for DEHS aerosol.

In conclusion, the best accuracy can be achieved with handheld CPCs, which however have some practical shortcomings. DCs can generally be a good alternative, because they are easier to use and offer an extended concentration range, but a lower accuracy has to be accepted. An accuracy of ±30% was shown to be a realistic range for the diffusion chargers in the test. The instruments reacted quite differently to the different aerosols. While the nanoTracer showed large deviations for aerosols with a large modal diameter, the nanoCheck had problems with very small particles. The miniDiSC had no problems with any of the test aerosols when measuring number concentrations and average diameters, but the agreement with the reference CPC for NaCl and soot was lower than with the nanoTracer. The LDSA concentration can only be accurately determined with DCs if the aerosol does not contain large surface area fractions of particles larger than ~400 nm, where the lung deposition curves show their minima. The Aerotrak 9000 and miniDiSC showed good agreement with LDSA calculated from FMPS measurements if such large particles were not present. The particle morphology did not seem to have a major impact on the accuracy of the instruments in the study. Hence each of the instruments in the test had their advantages and disadvantages and therefore represents a compromise. Care should be taken when choosing an instrument, taking into account the expected particle sizes and concentrations as well as the required accuracy levels.

In addition to providing general information on the performance of instrument types the round robin experiment reported here once more shows that the necessity of individual device calibration experiments using well defined aerosols. Only this way the specific instrument’s problems of CPC 3 (internal split) could be identified, corrected for, and finally eliminated.

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