Calibration of optical particle counters: first comprehensive inter-comparison for particle sizes up to 5 µm and number concentrations up to 2 cm$^{-3}$

Konstantina Vasilatou$^1$, KaiDirscherl$^2$, Kenjiro lida$^3$, Hiromu Sakurai$^3$, Stefan Horender$^1$ and Kevin Auderset$^1$

1 Federal Institute of Metrology (METAS), Lindenweg 50, 3003 Bern-Wabern, Switzerland
2 Danish National Metrology Institute (DFM), Kogle Alle 5, 2970 Hørsholm, Denmark
3 National Metrology Institute of Japan (NMIJ), National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Umezono, Tsukuba, Ibaraki, Japan

E-mail: konstantina.vasilatou@metas.ch

Received 7 August 2019, revised 25 November 2019
Accepted for publication 27 November 2019
Published 25 February 2020

Abstract

We report on an inter-laboratory comparison for the calibration of light-scattering optical particle counters measuring the number concentration of airborne particles in the size range 300 nm–5 µm. This comparison, registered as EURAMET pilot study Nr. 1453, is an important first step towards internationally recognized SI traceability in the measurement of low particle number concentrations encountered in clean-room facilities. The participating particle metrology laboratories of NMIJ (Japan), DFM (Denmark) and METAS (Switzerland) each provided a portable optical particle counter as a transfer standard to be circulated amongst the partners. The comparison covered particle number concentrations up to about 2 cm$^{-3}$. The individual measurements for the transfer standards at each laboratory are in good agreement with the reference value, typically within 7%, which supports the applied national primary methods of measurement.

Keywords: aerosol, optical particle counters, intercomparison, counting efficiency, calibration, clean room

(Some figures may appear in colour only in the online journal)

1. Introduction

Optical particle counters (OPCs) have been applied for the past decades as monitoring devices for airborne particle number concentrations in critical environments such as cleanroom production lines. In semiconductor industries, clean rooms are used in manufacturing and servicing of hardware, such as integrated circuits and hard drives. In medical, biotechnology and pharmaceutical environments, clean rooms are necessary to minimize the presence of bacteria, viruses and other pathogens, which could spread diseases or compromise the quality of the manufacturing procedures.

OPCs sample aerosols by means of an internal pump, and subsequently detect, count and determine the particle size based on single particle light scattering. The ISO standard 21501-4 [1] recommends a set-up for calibrating OPCs based on reference aerosols of spherical polystyrene latex (PSL) particles. This set-up consists of an aerosol generator, an aerosol mixing volume and a splitter in order to deliver the aerosol simultaneously to the device under test (DUT-OPC) and the reference (optical or condensation) particle counter. At first sight, the concept seems to be simple and straightforward...
to put into practice; however, generation, homogenisation and sampling of aerosol particles larger than about 0.5 µm becomes increasingly challenging.

The following considerations are important:

– Homogenisation of large particles is difficult due to high inertia and higher deposition rates.
– Isokinetic sampling probes are necessary when sampling with two optical particle counters operating at different flow rates to ensure representative sampling and minimize sampling artefacts of larger particles.
– A turbulent (plug) flow profile is necessary to ensure isokinetic conditions at the inlet of the isokinetic sampling probes. Since the flow rates of the DUTs are high, the dimensions of the inlets are large. A parabolic (laminar) profile cannot therefore ensure isokinetic sampling over the whole inlet area [2].
– Diffusion losses of small particles [3] depend on the OPC sampling flow rate and the length of the connecting tubes. A mismatch in the diffusion losses between DUT and reference counter must be calculated and corrected for.
– Deposition losses of larger particles in the isokinetic probes and/or connecting tubes of the reference counter must be calculated and corrected for [3].
– The sampling flow rate of the reference particle counter must be low enough so that coincidence losses are insignificant.
– The peak of residue particles arising from additives present in the PSL suspension must be well separated from that of the PSL particles or filtered out with a DMA (differential mobility analyser) or AAC (aerodynamic aerosol classifier) [2].

As a result, establishing traceability in the measurement of such low particle number concentrations as those encountered in clean room facilities (typically \( \leq 1 \, \text{cm}^{-3} \)) is a highly challenging task. METAS and DFM have developed primary methods for measuring particle number concentration [2] complying fully with the requirements of the ISO 21501-4 standard [1]. Moreover, Iida et al. have recently demonstrated a novel method for evaluating the counting efficiencies of optical particle counters based on an inkjet aerosol generator (IAG) [4, 5]. The IAG generates monodisperse particles at a constant but tunable rate and, hence, eliminates the need for a reference particle counter. Typical test particles are lactose monohydrate, ionic liquid, and NaCl (sodium chloride) residue particles arising from the evaporation of aqueous solution droplets.

Nevertheless, the metrological basis remains at present incomplete because the degree of equivalence has not been investigated by means of inter-laboratory comparisons. This study presents the results of the very first international comparison in the field of optical particle counters. The participating National Metrology Institutes (NMIs) were NMIJ (Japan), DFM (Denmark) and METAS (Switzerland, Coordinating laboratory). Each institute provided a commercial optical particle counter which was sent as a transfer standard to the other participants. The three instruments were in turn calibrated without adjustments to the instruments at each laboratory, applying the national reference standard and primary method of measurement for particle number concentration. Polystyrene latex particles (DFM, METAS), and NaCl/Lactose monohydrate particles (NMIJ) were used as model aerosols. The comparison covered the particle size range 300 nm to 5 µm and concentrations \( \leq 2 \, \text{cm}^{-3} \).

2. Experimental methods

2.1. Experimental set-up at METAS

The custom-made experimental facility at METAS has been described in detail elsewhere [2] and only the main aspects will be discussed here. Briefly, the experimental set up consists of three distinct sections: the aerosol generation system, a turbulent flow tube (homogeniser) and the particle detection system as illustrated in figure 1.

!!!Figure 1. Schematic representation (simplified) of the experimental facility at METAS. For a more detailed description of the set-up the readers are referred to Horender et al [2].!!!

PSL aerosols in the size range 100 nm–10 µm are generated based on wet or dry dispersion using commercially available generators [2] and, depending on the specific application, they can be size-selected with the use of a DMA or AAC to filter out residue particles.

The homogenizer is a 4 m-long custom-made stainless-steel tube with an inner diameter of 16.4 cm, placed vertically. Dry filtered air at a fixed flow rate of 120 l min\(^{-1}\) enters the
homogenizer from the very top and sweeps the PSL particles down the tube, where they are further mixed by three turbulent air-jets. The air-jet injection tubes are placed symmetrically around the homogenizer tube pointing 60° downwards. The sampling zone is located 3.0 m downstream of the injection position and accommodates two isokinetic sample probes placed just above the outlet of the homogenizer.

The detection system consists of a custom-built optical particle counter placed right at the outlet of the homogenizer to minimize particle losses due to tube bendings. The sampled aerosol enters the detection chamber through a nozzle with an orifice of 0.2 mm and is surrounded by a sheath-air flow, which prevents the particle beam from diverging. The sampling flow is measured with a traceably calibrated mass flow meter. A laser beam of 0.7 mm width is generated by a continuous-wave laser (Verdi V-5, Coherent) at a wavelength of 532 nm, and focused at the point of intersection with the aerosol stream with the use of a cylindrical lens. Particles crossing the laser beam scatter light, which is detected by a photomultiplier tube placed at a 90° angle.

2.2. Experimental set-up at DFM

The set-up at DFM is similar to that of METAS described above, see figure 1. The particle generator of DFM applies pressurized air which has been filtered at several stages to remove any particulate residues, with the final filters being industrial particle filters with a 20 nm cut-off. The commercial Bio-Aerosol Nebulizing Generator (BANG, CH Technologies) is used to generate the aerosol particles from liquid suspension. Subsequently, the aerosol is passed through a diffusion dryer based on silica gel, and injected centrally into a 3 m long vertical sampling pipe with a diameter of 100 mm. Above the point of injection, dilution air can be fed into the sampling pipe which is passed through a mesh of parallel tubes to distribute the dilution air equally across the pipe’s diameter. At the downstream point of aerosol injection into the pipe, three jets within the sampling pipe distribute the aerosol homogeneously across the diameter of the pipe in the dilution air.

The overall air speed is controlled to approx. 0.46 m s\(^{-1}\), corresponding to the typical vertical flow speed of air in cleanroom environments. Close to the bottom of the sampling pipe, the isokinetic probes of the transfer OPC and reference OPC are mounted next to each other as shown in figure 2. The flow field of the pipe has been experimentally verified to be flat at the point of sampling with an average speed of 0.46 m s\(^{-1}\) ± 0.02 m s\(^{-1}\). The lateral aerosol homogeneity in the pipe cross-section at the point of sampling has been determined to be constant within ±1% standard uncertainty.

DFM’s reference OPC is a commercial spectrometric LAS-X II (Particle Measuring Systems, USA). The LAS-X II is connected to the sampling pipe with the instrument’s default sampling tube (Tygon, black, conducting, 1.6 mm inner diameter/orifice, 0.8 mm long). The instrument’s sampling tube is always mounted straight and in direct contact with the DUT’s isokinetic probe within the sampling pipe. Isokinetic sampling for the LAS-X II occurs at an internal volumetric air flow of approx. 60 cm\(^3\) min\(^{-1}\). The airflow of the LAS-X II and its size settings are periodically and SI-traceable calibrated at DFM with a flow meter and reference PSL particle populations, respectively. The commercially available PSL populations are individually measured in-house with a metrological Atomic Force Microscope. In addition to the default sampling software and data output of the LAS-X II, DFM has connected an external 4-channel 2.5 MS s\(^{-1}\) ADC to each of the four internal analogue amplifications stages of the LAS-XII. This external data acquisition records the electric signals of the scattered light intensities for each particle for later offline analysis and data verification if required. Formal traceability for the counting quantity is established through DFM’s measurement procedures in accordance with [8].

The set-up at DFM can operate at number particle concentrations between 0.5 cm\(^{-3}\)–5 cm\(^{-3}\), corresponding to particle rates between 0.5–5 s\(^{-1}\) in the OPC’s air flow. These rates are sufficiently below the instrument’s 5% coincidence error limit of 4000 s\(^{-1}\). However, the uncertainty budget contains a contribution for coincidence, see table 1. In this table, the uncertainty contributions for the particle number
concentration in the generated reference aerosol are given. The values in the table have typically been observed during this comparison with concentrations \(<\ 2 \text{ cm}^{-3}\), where the aerosol was sampled for 30 min. The main uncertainty contribution derives from the temporal stability of the aerosol generator, which includes the Poisson uncertainty of the particle number as counted by the reference OPC. The relative standard uncertainties for this main contribution were typically approx. 2.4% for particles \(<\ 2 \mu\text{m}\), and 2.8% for particles \(>\ 2 \mu\text{m}\), respectively. These are mainly due to the Poisson uncertainties when counting approx. 2000 particles (30 min \(\times\) 66 particles/min, \(<\ 2 \mu\text{m}\)) and approx. 1440 particles (30 min \(\times\) 48 particles/min, \(>\ 2 \mu\text{m}\)).

On top of the uncertainty for particle number concentration in the reference aerosol, Type A contributions of the individual DUT measurements have been added. However, these are of minor significance with relative standard uncertainties of approx. 0.5% and below.

2.3. Experimental setup at NMIJ

The inkjet aerosol generator (IAG [4, 5]) was used to generate monodisperse test particles at a precisely controlled rate. A schematic illustration of the setup is provided in figure 3. The test particles were non-volatile residues of aqueous solutions. In this study, particles made of lactose monohydrate and sodium chloride were used to evaluate the counting efficiency of the OPCs. The rate of particle number being generated by the IAG was 30.00 s\(^{-1}\) (\(U = 0.165\text{s}^{-1}, k = 2\)). The particle generation rate is the product of droplet generation rate and particle generation efficiency. The droplet generation rate is controlled by the inkjet hardware (uDropC, Hantec, Japan). The frequency of the square pulses sent to the inkjet head (MD-K-130-30 \(\mu\text{m}\), microdrop Technologies, Germany) was measured by a traceably calibrated frequency counter (FCA 3000, Tektronix). The uncertainty of the particle generation efficiency is the dominant uncertainty component of the particle generation rate. A schematic illustration of the set-up is shown in the right panel of figure 1. The method simulates the sampling of uniformly mixed aerosol particles by delivering test particles at various places over the inlet planes of the isokinetic probes. The transfer OPC was placed on a motorized XY-stage, which was programmed to bring the tip of the IAG-exit tube to specific points within the inlet plane of the isokinetic probe [4]. In this study, the aerosol flowrate of the IAG was set to 0.3 l min\(^{-1}\), which corresponded to 1%–10% of the sampling flowrates of the transfer OPCs. Filtered air was therefore added through a laminarisation screen to complement the total sampling flowrate.

3. Comparison procedure

3.1. Comparison measurement scheme

Three commercial OPCs, a Solair 1100 (Lighthouse, USA), an Abakus (Markus Klotz GmbH, Germany) and a KC-31 (RION Co. Ltd., Japan) were used as transfer standards. These were shipped to the participants according to the scheme presented in figure 4. In order to evaluate possible drifts in the measurement efficiency of the OPCs due to transport, the transfer standards were shipped to METAS for a recalibration, with the exception of the KC-31 OPC which had to be shipped back to NMIJ for use at another research project.

Table 1. Typical relative standard uncertainty contributions as observed during this comparison.

| Uncertainty contributions for particle number concentration in reference aerosol | Relative standard uncertainties |
|---|---|
| | Particles < 2 \(\mu\text{m}\) | Particles \(\geq\ 2 \mu\text{m}\) |
| Aerosol homogeneity: | | |
| Generator, temporal\(^a\) | 2.4% | 2.8% |
| Cross-section, local | 1.0% | 1.0% |
| Sampling losses, ref. OPC: | | |
| Diffusion, impaction | 0.4% | 0.25% |
| Coincidence | 0.1% | 0.1% |
| Non-isokinetic sample | 0.25% | 1.2% |
| Instrumental source, ref. OPC: | | |
| Airflow stability | 1.2% | 1.2% |
| Electrical signal noise | 0.2% | 0.2% |
| Sample timing | 0.06% | 0.06% |
| Total standard uncertainty | 2.9% | 3.4% |
| Expanded Uncertainty | 5.8% | 6.9% |

\(^a\) The main uncertainty contribution is the temporal variation of the aerosol, on average between 2.4% for particles \(<\ 2 \mu\text{m}\) and 2.8% for particles \(>\ 2 \mu\text{m}\), respectively.
Figure 4. Illustration of the comparison measurement scheme. The number(s) in parentheses designate the order in which the OPC transfer standards were calibrated. The Solair 1100 and Abakus OPCs were shipped back to METAS for a fourth (and final) measurement. The KC-31 OPC, on the contrary, could not be shipped back to METAS due to limited availability.

The comparison was performed with different particle sizes ranging from 300 nm to 5000 nm. DFM and METAS generated PSL particles at a concentration of about 1-2 cm$^{-3}$ (see Table 2). The size of the PSL particles, $d_{PSL}$, had been previously measured by each NMI in-house by means of atomic force microscopy (AFM) traceable to the SI-system of units. The values are listed in Table 2.

NMJJ generated sodium chloride and lactose monohydrate particles at lower concentrations. Here, the PSL equivalent optical diameter, $d_{PSL eq}$, of the test particles was evaluated by the OPC divided by the sampling time. The uncertainty of the PSL equivalent optical diameter was not considered in this study.

Table 2. PSL particle diameter, $d_{PSL}$, reported by METAS and DFM based on traceable AFM measurements, optical equivalent PSL diameter, $d_{PSL eq}$ calculated by NMJJ and particle number concentrations generated for each experiment. The associated uncertainties ($k = 2$) are given in parentheses.

| Nominal particle size/nm | 300 | 500 | 1000 | 5000 |
|-------------------------|-----|-----|------|------|
| $d_{PSL}$, METAS$^a$    | 302(8) | 503(13) | 1005(23) | 5067(47) |
| $d_{PSL}$, DFM / nm     | 296(9) | 493(9) | 986(14) | 5031(70) |
| $d_{PSL eq}$, NMJJ/nm   | — | 500$^b$ | 1000$^b$ | 5000$^b$ |

| Particle number concentration/cm$^{-3}$ |
|-----------------------------------------|
| **Solair 1100 OPC**                     |
| METAS$^a$                               | 0.99/1.27 | 1.47/1.05 | 1.44/2.22 | 1.44/1.81 |
| DFM                                    | 1.68       | 0.93       | 1.33       | 0.75       |
| NMJJ                                   | —          | 0.0636     | 0.0636     | 0.0636     |
| **Abakus OPC**                         |
| METAS$^a$                               | 0.95/1.02  | 1.19/1.12  | 1.70/1.59  | 1.16/0.89  |
| DFM                                    | 0.91       | 1.41       | 1.01       | 0.83       |
| NMJJ                                   | —          | 0.636      | 0.636      | 0.636      |
| **KC-31 OPC**                          |
| METAS                                  | 0.96       | 1.28       | 1.68       | 1.28       |
| DFM                                    | 0.71       | 0.94       | 1.02       | 0.81       |
| NMJJ                                   | —          | 0.0636     | 0.0636     | 0.0636     |

$^a$ The particle number concentration values reported here correspond to the initial and final measurements performed at METAS, respectively.

$^b$ The uncertainty of the PSL equivalent optical diameter was not considered in this study.

The comparison reference value, $E_{ref}$, for each measurement point was determined as the weighted mean of the results reported by the participants ($E_1 \ldots E_N$), with $N = 3$. Here, the weights are given as the inverse square of the associated standard uncertainties $u(E_i)$ [6]:

$$E_{ref} = \frac{E_1}{u^2(E_1)} + \ldots + \frac{E_N}{u^2(E_N)} + \text{frac}.$$  

The result provided by METAS was the mean value of the counting efficiency from the two measurements performed.

(2) the maximum uncertainty of $L_{OPC}$ among the input data of linear regression, (3) the systematic error of $L_{OPC}$ due to the systematic error in $d_{PSL eq}$ since the $d_{PSL eq}$ was measured by using a different OPC. Finally, the particle number concentration, $C_{OPC}$, was calculated as $L_{OPC}$ divided by $Q$.

The concentration generated with the IAG, $C_{primary}$, is given by $L/Q$, where $L$ (s$^{-1}$) is the particle generation rate of the IAG. Since $L$ was set to 30.00 s$^{-1}$ in this study, the value of $C_{primary}$ was 0.0636 cm$^{-3}$ and 0.636 cm$^{-3}$ for OPCs with flow $Q = 28.3$ l min$^{-1}$ (Solair 1100, KC-31) and 2.83 l/min (Abakus), respectively.

A detailed uncertainty budget for $C_{primary}$ and $E_i$ are reported in [2, 4, 5] for the facilities at METAS and NMJJ, respectively. For DFM’s uncertainty budget, see Table 1.

3.2. Calculation of results

The comparison reference value, $E_{ref}$, for each measurement point was determined as the weighted mean of the results reported by the participants ($E_1 \ldots E_N$), with $N = 3$. Here, the weights are given as the inverse square of the associated standard uncertainties $u(E_i)$ [6]:

$$E_{ref} = \frac{E_1}{u^2(E_1)} + \ldots + \frac{E_N}{u^2(E_N)} + \text{frac}.$$  

The result provided by METAS was the mean value of the counting efficiency from the two measurements performed.
at METAS at the beginning and end of the inter-comparison (see measurement scheme depicted in figure 4). An additional factor \( f_{\text{drift}} \) was added in the above equation to account for possible drifts in the OPC measurement efficiency during shipping. The value of \( f_{\text{drift}} \) was set to zero but was assigned an uncertainty \( u_f \). The latter was treated as a Type B uncertainty with a triangular distribution and a halfwidth of limits equal to \( \Delta E_{\text{METAS}}/2 \), with \( \Delta E_{\text{METAS}} = E_{\text{METAS},i} - E_{\text{METAS},f} \) being the deviation between the two values reported by METAS (i.e. the initial and final measurements of the inter-comparison).

\[ \Delta E_{\text{METAS}} \] could not be determined for the measurements with the KC-31 OPC since this transfer standard could not be shipped back to METAS (see caption of figure 4) for repeating the measurement at the end of the inter-comparison campaign. In this case, \( \Delta E_{\text{METAS}} \) was set to zero.

The results of the comparison are also reported in terms of the relative difference \( d_i \) between the result \( E_i \) of laboratory \( i \) and the comparison reference value \( E_{\text{ref}} \):

\[ d_i = E_i - E_{\text{ref}} \]

The standard uncertainty of the reference value \( E_{\text{ref}} \) and the relative difference \( d_i \) were calculated with the software GUM-Workbench Pro which took into account (linear) correlations between the values \( E_i \) and \( E_{\text{ref}} \). The expanded uncertainties \( U \) were calculated as

\[ U = 2u \]

which gives a 95% coverage assuming the results are normally distributed [7].

4. Results and discussion

The three OPcs, which were used as transfer standards during the inter-comparison, had different technical specifications. Nominally, the counters exhibit a 50% cut-off (i.e. 50% measurement efficiency) at their smallest detectable particle size, i.e. at 100 nm for the Solair 1100 and at 300 nm for the Abakus and KC-31 OPcs. At larger particle sizes, the counters exhibit a measurement efficiency of 100%. Unknown to the participants, the size thresholds of the Abakus OPC had been misaligned prior to the inter-comparison in order to challenge the laboratories. The counting efficiencies, \( E_i \), reported by the participating institutes are illustrated in figures 6(a)–(c) for the Solair 1100, Abakus and KC-31 OPC transfer standards, respectively, and summarised in table 3.

The calculation of the reference value, \( E_{\text{ref}} \), and its uncertainty for the Solair 1100 transfer OPC at 500 nm with the use of the GUM Workbench software is shown in table 4 as example.

As mentioned in section 3.1, METAS and DFM generated PSL aerosols in the range 300 nm–5 \( \mu \)m based on dry or wet dispersion. NMIJ generated NaCl and/or lactose monohydrate particles from an aqueous solution. Impurities in the solution...
give rise to solid residue particles upon evaporation of the droplets generated by the IAG [4, 5]. The diameter of the residue particles is in the order of a few hundred nm, thus limiting the controllable size of the generated NaCl and lactose monohydrate particles to above about 500 nm. This explains why NMIJ could not determine the counting efficiency of the transfer OPCs at 300 nm and the values are missing from figure 6 and tables 2, 3 and 5.

During the generation of the reference aerosol for the measurement of the Abakus OPC at 500 nm at DFM, the aerosol

![Figure 6](image)

**Figure 6.** OPC counting efficiencies, $E_i$, reported by the participants and reference value, $E_{ref}$, for nominal particle sizes 300 nm–5 µm obtained using (a) the Solair 1100, (b) the Abakus and (c) the KC-31 OPC transfer standards. The error bars designate the expanded uncertainties ($k = 2$) of the results. In figure 6(b), the counting efficiency $E_i = 1.050$ reported by DFM at 500 nm is biased (see text for more details) and was not taken into account when calculating the reference value. The counting efficiency of the Abakus OPC at 300 nm and 5 µm is significantly higher than the expected 0.5 and 1.0, respectively. This is due to a misalignment of the size thresholds (see text for more details).

**Table 4.** Calculation of the reference value, $E_{ref}$, and its uncertainty for the Solair 1100 transfer OPC at 500 nm with the use of the GUM Workbench software.

| Input parameters | Results (GUM software output) |
|------------------|-------------------------------|
| $E_{METAS}$      | 0.9450 ± 0.0345               |
| $E_{DFM}$        | 1.0097 ± 0.0284               |
| $E_{NMIJ}$       | 1.00600 ± 5.64·10⁻³          |
| $f_{shift}$      | 0.0055 ± 0.0010               |

$E_{ref} = 1.005 ± 0.012$, coverage 95% (normal distribution)

**Table 5.** Relative difference, $d_i$, from the comparison reference value reported by the participants at different particle sizes and a particle number concentration $≤ 2 \text{ cm}^{-3}$. The numbers in parentheses represent the associated uncertainties ($k = 2$).

### Solair 1100 OPC

| Particle size/nm | METAS     | DFM       | NMIJ       |
|------------------|-----------|-----------|------------|
| 300              | 0.050 (0.070) | 0.020 (0.035) | —          |
| 500              | 0.060 (0.068) | 0.005 (0.056) | 0.001 (0.006) |
| 1000             | 0.074 (0.079) | 0.019 (0.050) | 0.001 (0.002) |
| 5000             | 0.055 (0.079) | 0.017 (0.046) | 0.051 (0.053) |

### Abakus OPC

| Particle size/nm | METAS     | DFM       | NMIJ       |
|------------------|-----------|-----------|------------|
| 300              | 0.010 (0.058) | 0.042 (0.058) | —          |
| 500              | 0.042 (0.058) | 0.001 (0.070) | 0.001 (0.002) |
| 1000             | 0.074 (0.079) | 0.019 (0.050) | 0.001 (0.002) |
| 5000             | 0.055 (0.079) | 0.017 (0.046) | 0.051 (0.053) |

### KC-31 OPC

| Particle size/nm | METAS     | DFM       | NMIJ       |
|------------------|-----------|-----------|------------|
| 300              | 0.012 (0.017) | 0.042 (0.066) | —          |
| 500              | 0.042 (0.066) | 0.027 (0.071) | 0.000 (0.004) |
| 1000             | 0.027 (0.071) | 0.057 (0.085) | 0.002 (0.002) |
| 5000             | 0.004 (0.140) | 0.044 (0.064) | −0.006 (0.010) |
contained residual contaminant particles from the particle suspension. These residual particles were clearly distinguishable from the population of the reference particles in DFM’s reference OPC, but they were registered and counted by the Abakus together with the reference PSL particles. The originally calculated and reported $E_{i} = 1.050 \pm 0.054$ by DFM is therefore too high, biased by the additionally registered contaminants. This measurement has been consequently removed from the data analysis in order to allow for an unbiased reference value $E_{\text{ref}}$. After correcting for this error by taking into account the contaminant particles registered by the Abakus, DFM could determine a counting efficiency $E_{i} = 0.972 \pm 0.076$ at 500 nm, in good agreement with the partners’ values.

The uncertainties reported by NMJJ are smaller than those reported by the other participants. Thanks to the regular injection pattern of the IAG system, i.e. generation of single

---

**Figure 7.** Relative difference, $d_{i}$, between the results obtained by participants and the reference value for nominal particle sizes of 300 nm to 5 µm with (a) the Solair 1100, (b) the Abakus and (c) the KC-31 OPC as transfer standard. The error bars designate the expanded uncertainties ($k = 2$) of the results, obtained from the uncertainty of the results reported by each participant and the uncertainty in the reference value.
droplets at controlled rate, uncertainties due to random particle distribution are minimal when the counting efficiencies are close to 100%. On the contrary, the uncertainties reported by METAS and DFM contain a large contribution, typically ≥4%, related to Poisson statistics (random particle distribution) [2]. Moreover, in figures 6(b) and (c) it can be seen that the uncertainties reported by METAS for the 5 µm particles exceed 10%. This can be attributed to technical issues with the use of the fluidised bed generator, which led to instabilities in the aerosol generation. Since the reference value was calculated as a weighted mean of the values reported by the participants, $E_{\text{ref}}$ lies closer to the values reported by NMIJ. The expanded relative uncertainty of $E_{\text{ref}}$ is typically 1%–2% (see table 3) but increases to 4%–5% when uncertainties arising from instrument shipping, $u_p$, become important.

Figure 7 demonstrates that the reported results, with the exception of the measurement reported by DFM at 500 nm for the Abakus DUT-OPC (figure 6(b), agreed with the reference value within the stated uncertainties for all examined particle sizes. The agreement was not compromised above 1 µm in spite of the challenges in the generation and sampling of larger particles.

The results in terms of the relative difference, $d_r$, are plotted in figures 7(a)–(c) for the experiments carried out with the Solar 1100, Abakus and KC-31 OPC transfer standards, respectively, and summarised in table 5.

Overall, the results reveal that the deviations from the reference value are not systematic but random. Moreover, the deviation is typically <7% and lies well within the expanded uncertainty of $d_r$, indicating the good mutual agreement in the measurement of low particle number concentrations between the participating NMIs. As expected, the deviations from $E_{\text{ref}}$ are smallest in the case of NMIJ because of the smaller measurement uncertainties and the larger weighting factor of $E_{\text{NMIJ}}$ when calculating $E_{\text{ref}}$.

Figure 7 reveals that the uncertainties reported by METAS for the measurements with 5 µm particles (see figures 7(b)–(c)) are highly overestimated. As explained in [2], METAS repeats the particle number concentration measurement 25 times and calculates the standard deviation of the mean, which amounts typically to 4%–5% (for a coverage factor $k = 2$) and corresponds to the largest contribution in the METAS uncertainty budget. This uncertainty results from the random particle distribution (Poisson statistics). However, when the aerosol generation is unstable, as often the case when aerosolising large PSL particles from dry powder with a fluidised bed generator, the standard deviation of the mean increases considerably. Nevertheless, the results displayed in figure 7 indicate clearly that possible instabilities in aerosol generation do not compromise the measurement accuracy of the METAS primary method of measurement. This can be explained by the fact that the reference particle counter and DUT-OPC sample aerosol in parallel. Drifts or fluctuations in the PSL concentration are recorded at the same time by both instruments and, hence, do not affect the quality of the calibration procedure. Based on the outcome of this intercomparison, METAS will revise their uncertainty budget. Instead of calculating the standard deviation of the mean (of the 25 number concentration measurements), an uncertainty based on Poisson statistics will be calculated and included in the uncertainty budget. This will account for the random particle distribution but will not include any effects arising from instabilities in aerosol generation.

5. Summary and conclusions

This paper reports on the first comprehensive inter-comparison of primary methods of measurement for low particle number concentrations, such as those encountered in clean room facilities. The comparison covered the particle size range 300 nm–5 µm and concentrations up to about 2 cm$^{-3}$. Since these primary methods of measurement include large-scale facilities which cannot be transported, three commercially available OPCs were used as transfer standards and were shipped to all participants. The participants determined the measurement efficiency of each OPC and the associated uncertainty at each particle size. The analysed results for all particle sizes agreed with the reference value typically within 7%, which is compatible with the stated uncertainties. The agreement was not compromised at larger particle sizes despite the increasing challenges in the generation and sampling of such aerosols.

The quantity ‘particle number concentration’ is calculated by dividing counts (dimensionless) by volume. It is thus a coherent derived unit in the SI system of units. For counting quantities, a formal traceability to the SI can be established through appropriate, validated measurement procedures [8].

The results reported here show that the OPC measurement efficiencies determined by the three participating institutes were equivalent, with the exception of the measurement at 500 nm reported by DFM for the Abakus DUT-OPC (figure 7(b)). The origin of this deviation has been investigated and explained. Furthermore, the results were within their stated uncertainties ($k = 2$) and provide strong evidence for the proficiency of these NMIs in the technical field of OPC calibration, validating their individual measurement procedures.

Acknowledgments

Part of this work was carried out within the 16ENV07 Aeromet project funded by the European Metrology Programme for Innovation and Research (EMPIR). The EMPIR initiative is co-funded by the European Union’s Horizon 2020 research and innovation programme and the EMPIR Participating States. DFM was supported by the 16ENV07 Aeromet project and by funds from the Danish Agency for Institutions and Educational Grants. METAS was supported by the Swiss State Secretariat for Education, Research and Innovation (SERI) under contract number 17.00112 and by internal funds. The opinions expressed and arguments employed herein do not necessarily reflect the official views of the Swiss Government. AIST was partly supported by the international standardization project which is conducted by the Ministry of Economy, Trade, and Industry (METI) of Japan. AIST
borrowed the KC-31 OPC from RION Co., Ltd. during this inter-comparison, and RION’s support is greatly appreciated.

ORCID iDs

Konstantina Vasilatou  https://orcid.org/0000-0002-7771-9596
Kenjiro Iida  https://orcid.org/0000-0001-5364-2232

References

[1] ISO 21501-4:2018 2018 Determination of Particle Size Distribution—Single Particle Light Interaction Methods—Part 4: Light Scattering Airborne Particle Counter for Clean Spaces (International Standardization Organization)

[2] Horender S, Auderset K and Vasilatou K 2019 Facility for calibration of optical and condensation particle counters based on a turbulent aerosol mixing tube and a reference optical particle counter Rev. Sci. Instrum. 90 075111

[3] Hinds W C 1999 Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles (New York: Wiley)

[4] Iida K and Sakurai H 2018 Counting efficiency evaluation of optical particle counters in micrometer range by using an inkjet aerosol generator Aerosol Sci. Technol. 52 1156–66

[5] Iida K, Sakurai H, Saito K and Ebara K 2014 Inkjet aerosol generator as monodisperse particle number standard Aerosol Sci. Technol. 28 789–802

[6] Cox M G 2002 The evaluation of key comparison data Metrologia 39 589–95

[7] BIPM, IEC, IFCC, ILAC, ISO, IUPAC, IUPAP and OIML 2008 Evaluation of Measurement Data—Guide to the Expression of Uncertainty in Measurement (Joint Committee for Guides in Metrology)

[8] BIPM 2019 SI Brochure: The International System of Units (SI) (Sèvres Cedex: Bureau International des Poids et Mesures)