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ABSTRACT

In cleanroom environments, light scattering airborne particle counters are typically employed to monitor particle contamination of production lines and for general classification purposes. In addition to the amount of airborne particles, these instruments also measure the size of each registered particle. This is essential for quality assurance, as different particle sizes have various associated risks connected. Critical parameters for particle sizing are therefore an instrument's size resolution and size setting, and these need regular calibration. The ISO standard 21501-4:2018 describes a calibration method, yet this method requires detailed technical insider knowledge. In this paper, we present an alternative method that allows direct in-use calibration of a particle counter without the need for information, which is typically only available to the instrument manufacturer. In a direct comparison, both methods perform in compliance with the requirements in ISO 21501-4:2018, although our direct approach generally yields lower uncertainties. In addition, our calibration method provides results that are closely related to the instrument's properties during its application, contrary to the ISO method's indirect calibration procedure that requires voltage readouts from internal terminals.

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I. INTRODUCTION

Light scattering airborne particle counters (LSAPCs) operate on the principle that airborne particles scatter light (e.g., from a laser) and by the fact that a nonlinear relationship between the scattered light intensity and particle size exists.¹ By restricting the flow of particles in an airstream of a known volumetric flow rate, the LSAPC can quantify the number of particles as particle number concentration per volume of air in different size range categories. A sketch of the working principle of an LSAPC is depicted in Fig. 1(a). In this simple picture, the light is only collected at a single angle, but more elaborate calibration schemes have also been realized to better determine the particle size,² as well as simultaneous determination of size and refractive index.³

For cleanroom classification, it is only required to monitor the size and not the composition of the airborne particles. Two size categories are of particular interest to the pharmaceutical industry,

namely 0.5 μm and 5 μm . The reason for this is that these size ranges are the ones monitored in cleanrooms for production of medicinal compounds, in accordance with the EU guideline EU GMP Annex 1. Distinguishing small size differences in the vicinity of these delimitation points, i.e., particles smaller or larger than 0.5 μm in diameter, is therefore of great relevance since this directly impacts the classification and permissions for a given cleanroom. In order to facilitate a distinction as precise as possible, careful calibration of the LSAPC is required.

Current procedures of calibration and verification of LSAPCs in order to comply with cleanroom standards (e.g., ISO 14644-1) are detailed in ISO 21501-4: 2018⁴ (hereafter, just ISO 21501-4). This standard requires a variety of certified reference particles (CRPs), e.g., polystyrene (PS) particles of known mean diameter, x_C , and standard deviation, σ_C , with a refractive index of 1.59 at 589 nm. Important features of the ISO standard include estimation of the size setting error (SSE, i.e., where the particle size bin boundary lies

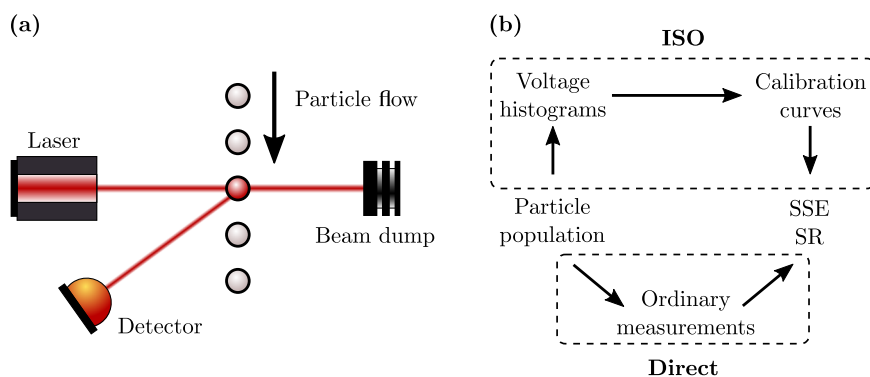


FIG. 1. (a) Sketch of the working principle of a simple light scattering airborne particle counter. Light from a monochromatic source is scattered off the particles being directed into the light path by an air flow. The detector is placed at an angle to the incoming beam to avoid the direct light. (b) Comparison between the measurement pathways of the calibration method recommended by the ISO standard and our direct approach.

relative to the set value), the size resolution (SR, the ability of the instrument to distinguish between particles of similar size, which is limited due to phenomena such as voltage peak broadening), and counting efficiency (the ability of the LSAPC to determine particle number concentration accurately vs a reference counter).

Instrument calibration should be performed on a regular schedule, e.g., annually. The procedure for doing so recommended in ISO 21501-4 requires the ability to read out the detector response using either an internal Pulse Height Analyzer (PHA) of the LSAPC or by a certified technician using an external PHA. Alternative calibration methods are allowed under ISO 21501-4, however, as long as their uncertainties are evaluated and described,⁴ and such methods have been suggested previously in the literature. In one case, oil droplets were generated and classified to efficiently generate a wide range of size distributions of aerosolized particles to use for calibration.⁵ In the other case, the naturally occurring log-normal size distribution of talc powder was used to perform the calibration.⁶ In the first case, it was still necessary to access the PHA of the LSAPC and in neither were the uncertainties evaluated as required by ISO 21501-4.

In this work, we present a direct approach that will allow any experienced user or technician to verify the SSE and SR of an LSAPC using only CRPs and the readout from the counter indicating the particle number in each particle size bin, eliminating the need to access the voltage output of the LSAPC, which in most cases is accessible to the manufacturer only. Furthermore, this method only relies on using two CRP suspensions with sizes close to the size setting to be calibrated, reducing the labor and time required to perform a calibration. We compare this new approach and the uncertainties associated with it to the method recommended in ISO 21501-4. We find that the direct approach, in general, agrees within expanded uncertainties and that the uncertainties of the direct approach are smaller. A schematic overview of the two different methods is shown Fig. 1(b).

II. EXPERIMENTAL SETUP AND PROCEDURES

A. Size determination of CRPs

The particle populations used during this study are commercial off-the-shelf CRPs from Microparticles GmbH and BS-Partikel GmbH. Irrespective of the specified population properties given by the supplier, each population has been re-measured by

Danish Fundamental Metrology (DFM) on our calibrated Metrology Atomic Force Microscope (AFM) NX20 (Park Systems), traceable to the SI system of units. For these measurements, a small sample of each population was taken and applied by drop deposition onto a freshly cleaved and atomically flat mica substrate. The average particle diameter and the populations' standard deviation were subsequently determined from the AFM measurements in accordance with accredited measurement procedures under ISO 17025. The measurement uncertainties have been calculated in accordance with established procedures.^{7,8} Table I lists the determined properties of the particle populations used. An example of a measured sample of a particle population can be seen in Fig. S1.

B. Aerosolization of CRP suspensions

We used a commercial Bio-Aerosol Nebulizing Generator (BANG/CH Technologies) to generate the particle aerosols from suspension. For the generation of aerosols, we used pressurized air

TABLE I. The mean diameter, x_C , of the certified reference particles and their standard deviation, σ_C , used in this work, as well as the measured median voltages, V_M , and bin count ratios, n_L/n . The latter represents the relative number of particles identified by the LAS-X as being larger than the unknown bin boundary size x'_i . The middle particle in each range was used as the boundary value for the evaluation of the size setting error and size resolution. The voltages V_M were measured at different analog amplification stages in the LAS-X and can therefore not be correlated with particle size over all sizes but only within a single size range. All uncertainties are reported with a 2σ interval.

x_C (nm)	σ_C (nm)	V_M (V)	n_L/n
248.0 ± 6.8	5.5 ± 0.3	88 ± 10	0.16 ± 0.06
296.2 ± 9.0	9.0 ± 1.0	192 ± 11	0.37 ± 0.06
319.3 ± 6.9	3.4 ± 0.2	302 ± 11	0.92 ± 0.06
431.4 ± 9.4	8.2 ± 0.7	1066 ± 13	0.10 ± 0.06
493.1 ± 9.0	9.8 ± 0.5	1430 ± 15	0.14 ± 0.06
556.1 ± 9.2	5.5 ± 0.7	1967 ± 25	0.84 ± 0.06
917 ± 14	7.1 ± 0.7	4077 ± 38	0.01 ± 0.06
945 ± 13	17.2 ± 1.2	4674 ± 63	0.37 ± 0.06
986 ± 14	17.6 ± 1.9	5172 ± 65	0.60 ± 0.06
4462 ± 75	106 ± 12	3588 ± 18	0.01 ± 0.06
5031 ± 70	97 ± 9	3810 ± 24	0.37 ± 0.06
5427 ± 97	132 ± 24	4394 ± 21	0.96 ± 0.06

that had been filtered at several stages to remove any particulate residues from the compression process, with the final filter stage consisting of industrial particle filters with a 20 nm cutoff.

We use commercially available CRPs characterized as described above. The raw particle solutions were diluted with Milli-Q water to produce a particle number concentration of $\sim 1/\text{cm}^3$ of air in the final aerosol. Subsequent to the aerosolization of the diluted particle suspension, the aerosol was 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 into the pipe, additional dilution air can be fed in. Before mixing with the aerosol, the dilution air is first passed through a mesh of parallel tubes in order to distribute it equally across the pipe's diameter. At the downstream point of aerosol injection into the pipe, jets are built into the sampling pipe to distribute the aerosol homogeneously across the diameter of the pipe.

The overall air speed was controlled to ~ 0.46 m/s, which corresponds to the typical vertical flow speed of air in cleanroom environments. Close to the bottom of the sampling pipe, DFM's reference particle counter is mounted with the option of adding additional particle counters for calibration purposes. At this cross section of the pipe, 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 ± 0.02 m/s. The lateral aerosol homogeneity in the pipe's cross section at the point of sampling has been determined to be constant within $\pm 1\%$.

C. Particle measurements

LSAPCs count particles in set size ranges called bins. We measured four particle size ranges around $0.3 \mu\text{m}$, $0.5 \mu\text{m}$, $1 \mu\text{m}$, and $5 \mu\text{m}$, which are the common bin boundaries for a commercial LSAPC. An overview of the CRPs used can be seen in Table I.

DFM's reference counter is a LAS-X II particle spectrometer (Particle Measuring Systems). We measured the analog detector signals of the LAS-X directly after the four amplifier stages on the instrument's electronic main board using NI6133 (National Instruments). This 14-bit analog-to-digital-converter allows for sampling of up to four analog voltages in parallel with a sampling rate of 2.5 MHz per channel. This allows for supersampling of the particle detection signal, which is typically a Gaussian shape for a period of ~ 10 to $100 \mu\text{s}$. The recorded signals were offline analyzed for their peak height values. Artifacts such as noisy peaks or non-Gaussian signals were ignored such that each measured peak height could be associated with a single particle. The measurement accuracy of the analog-to-digital converter is ~ 5 mV, corresponding to 0.5% of typical peak height values at around 1 V for each amplifier stage.

For the direct approach, we simply used the LAS-X as a regular LSAPC. For the $0.3 \mu\text{m}$ and $0.5 \mu\text{m}$ ranges, we set the bin range from 90 nm to 1080 nm in steps of 10 nm, for the $1 \mu\text{m}$ range, we used bins from 100 nm to 2550 nm in steps of 25 nm, and for the $5 \mu\text{m}$ range, we used bins from 140 nm to 6080 nm in steps of 60 nm.

The direct approach can be applied to any LSAPC with any size ranges because it only requires the number of particle counts in two neighboring bins in order to be applicable. This information is always available to the user by default, being the counter's main functionality, and it suffices to determine the size setting error and

size resolution at the intersecting bin boundary. While the bin for the largest detectable particle size typically has no upper limit, i.e., all particles $>10 \mu\text{m}$ are registered in the bin " $10 \mu\text{m}$," the bin for the smallest detectable particle size has a well-defined lower boundary, e.g., $0.3 \mu\text{m}$, which requires the same size setting measurements as the other bin boundaries. In order to calibrate the SSE and SR for the lowest bin, the results of ordinary counting efficiency calibrations supply all the information needed. For more details, see the [supplementary material](#).

D. Recommended calibration method

The particle diameter delineating each boundary between size bins is fixed based on the preset bin boundary voltages, V_I . The bin boundaries corresponding to these voltages typically drift over instrumental lifetime and therefore need to be calibrated on an annual basis. A part of this process is to determine the SSE and the SR, which we will focus on here, since these are the two parameters we can calibrate with our new direct approach.

The recommended calibration procedure in ISO 21501-4 is briefly presented here.⁴ In short, it relies on the creation of calibration curves translating the PHA response voltages into particle sizes. For each size range, a calibration curve is made by measuring the voltage distributions of several CRP sizes using the particle counter's internal or an external PHA, yielding several calibration points. These points are fitted to a calibration curve, allowing for interpolation between CRP sizes and thus the determination of the SSE and SR in an indirect manner.

From each measured voltage distribution, the lower and upper bounds, V_L and V_U , are determined to clearly separate the main peak from the rest of the histogram, as illustrated in Fig. 2(a). Within these bounds, the median voltage V_M can be determined and V_M is then ascribed to the mean particle diameter x_C . Using these pairs of values (x_C , V_M) obtained from each measured CRP distribution, the calibration curve is determined by fitting a suitable function, as indicated in Fig. 2(b).

Equipped with the calibration curve, it is subsequently possible to determine the SSE and the SR. This is done by determining the size x'_I corresponding to the current bin boundary voltage setting V_I of the LSAPC, as shown in Fig. 2(b). The SSE can then be calculated as

$$\text{SSE} = \frac{x'_I - x_I}{x_I}, \quad (1)$$

where x_I is the size setting (i.e., nominal bin boundary value) specified for the LSAPC. The SR is defined as

$$\text{SR} = \frac{\sqrt{\sigma_1^2 - \sigma_C^2}}{x_C}, \quad (2)$$

where x_C and σ_C are the CRP mean diameter and standard deviation, respectively, and σ_1 is the apparent standard deviation of the distribution as measured by the LSAPC. From the measured voltage distribution, the lower and upper bounds, V_1 and V_2 , are obtained as the values at 61% of the peak value of the histogram, i.e., these bounds are different from V_L and V_U . These voltages $V_{1,2}$ are then converted to the size values $x_{1,2}$ using the calibration curve. The apparent standard deviation σ_1 is finally given by the greater of the two differences $|x_C - x_1|$ and $|x_2 - x_C|$, which allows for the calculation of the SR.

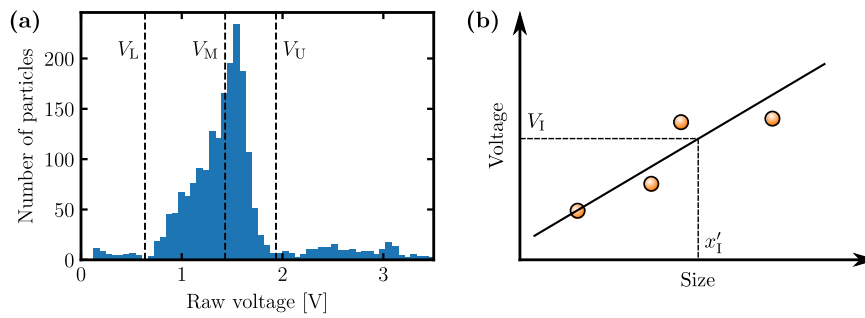


FIG. 2. (a) Voltage distribution obtained directly from the LAS-X for certified reference particles with a mean diameter of 493.1 nm. The lower and upper bounds along with the median voltage (V_L , V_U , and V_M , respectively) are indicated by the dashed lines. (b) Sketch of a calibration curve obtained from the voltage histograms of several certified reference particle distributions. The dashed lines represent the determination of the bin boundary size setting x'_I from the bin boundary voltage V_I .

When using conventional particle counters for monitoring cleanrooms, the bin boundaries are preset to certain values, in many cases $0.3 \mu\text{m}$, $0.5 \mu\text{m}$, $1 \mu\text{m}$, and $5 \mu\text{m}$. In the case of our LAS-X, however, the bin widths and boundaries can be individually programmed to fit a particular measurement. As such, we do not have a specific bin boundary to calibrate, and we therefore use CRP sizes close to the values of $0.3 \mu\text{m}$, $0.5 \mu\text{m}$, $1 \mu\text{m}$, and $5 \mu\text{m}$ to represent our bin boundary values. By using two particle populations, one to either side of the boundary value, we can create a calibration curve and calculate the SSE and the SR relative to this. The CRPs used are reported in Table I, where the middle size in each of the four ranges serves as the boundary value and the two others to create the calibration curve. This means that the used values of x_I are $0.2962 \mu\text{m}$, $0.4931 \mu\text{m}$, $0.945 \mu\text{m}$, and $5.031 \mu\text{m}$, respectively. Voltage histograms for these four sizes are shown in Fig. S2. Finally, since we use two CRP populations generating two points for each particle range, we employ a linear model for the calibration curves.

E. Procedure for the direct approach

We now present our new proposed method for calibrating an LSAPC without the need to access the internal PHA of the instrument, i.e., a direct approach. This method can be used to simultaneously calibrate the SSE and the SR with a smaller workload than the ISO method by using only two CRPs with known x_C and σ_C and their uncertainties. For the best results, the mean sizes of the CRPs should be chosen within 15% on either side of x_I , corresponding to a typical size resolution.

Our direct approach is based on two assumptions. First, the response of the LSAPC to a normally distributed particle size population can be approximated by a normal signal voltage distribution. This is also the underlying assumption in the method recommended in ISO 21501-4, which can be seen from the figures and methods used (e.g., the 61% threshold for the voltage distribution width when determining the SR). Secondly, both particle populations are assumed to be subject to the same signal broadening by the LSAPC. Even though they have distinct mean diameters, they can hardly be resolved as they were chosen to be close to, or within, the instrumental size resolution.

Light scattered off a particle results in a voltage signal of a certain magnitude. The LSAPC internally sorts this signal into counts being smaller (n_S) or larger (n_L) than the bin boundary voltage V_I (which corresponds to the unknown boundary value x'_I), with a total amount of particles measured equal to $n = n_S + n_L$ (see Fig. 3).

Applying the assumption of a normal voltage distribution, the bin count ratio n_S/n of particles smaller than x'_I corresponds to the value of the cumulative distribution function (CDF) of the normally distributed particles with mean x_C and enlarged standard deviation σ_I at the point x'_I . For computational reasons, we consider the complementary ratio $n_L/n = 1 - n_S/n$, corresponding to $1 - \text{CDF}$, instead, which we use in the following.

By using two bin count ratios $n_{L,(1,2)}/n_{(1,2)}$ from two populations, the two parameters x'_I and σ_I can be determined unambiguously. This is done by an iterative nonlinear least squares fitting routine,⁹ which adjusts x'_I and σ_I so that the calculated values of the CDFs of the two normal distributions $N(x_{C,1}, \sigma_{I,1})$ and $N(x_{C,2}, \sigma_{I,2})$

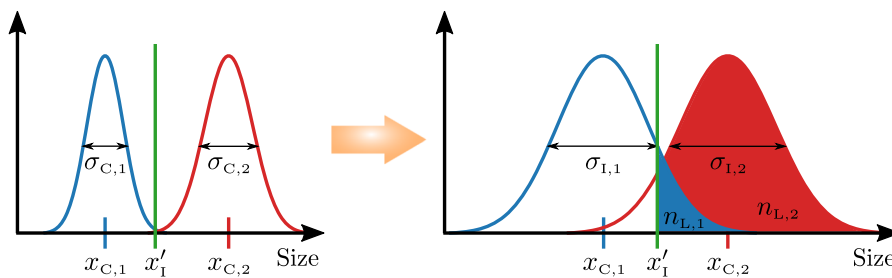


FIG. 3. Sketch of the new proposed direct calibration scheme. The initial particle populations (left) are convoluted with the instrument's resolution (right), increasing their widths from σ_C to σ_I . The amount of particles, $n_{L,(1,2)}$, larger than the unknown boundary value are used to iteratively fit the unknown values of $\sigma_{I,(1,2)}$ and x'_I using a least squares routine.

match the observed values of the ratios $n_{L,(1,2)}/n_{(1,2)}$. For all particle size ranges in Table I, we use the two particle populations with the largest means to obtain the SSE and the SR. As the SSE and SR are relative values, we use the middle value from each size range as the boundary value, i.e., as the denominator in Eqs. (1) and (2) for the calculation, as we do when employing the ISO method. For a typical LSAPC, the boundary values are naturally given by the nominal specified bins. For the determination of SSE and SR at the lowest boundary, the LSAPCs counting efficiency CE_1 serves as the ratio $n_{L,1}/n_1$. A second calibration of counting efficiency with a second different particle population yields CE_2 , which serves as the ratio $n_{L,2}/n_2$.

From the definition of the SR in Eq. (2), we have $\sigma_{I,(1,2)}^2 = x_C^2 \text{SR}^2 + \sigma_{C,(1,2)}^2$, where x_C , here, is the chosen boundary value as described earlier. The assumption that the instrument broadening introduced by the LSAPC is the same for both particle distributions therefore translates into the relation $x_{C,1}^2 \text{SR}^2 = x_{C,2}^2 \text{SR}^2$. As such, we impose the constraint $\sqrt{\sigma_{I,1}^2 - \sigma_{C,1}^2} - \sqrt{\sigma_{I,2}^2 - \sigma_{C,2}^2} = 0$ on the result from the fitting routine to maintain the same instrument broadening for both particle distributions.

The initial conditions used are x_C and $2\sigma_C$ taken from the particle distribution selected as the boundary value; for a regular LSAPC, the first would translate to the nominal bin boundary value to be calibrated, while the second is the same. These values are fed to a regular least squares routine, and the results from here are then used as the basis for the iterative scheme.⁹

The least squares routine directly outputs the uncertainty in the estimated parameters reducing the need for further calculations, making it compliant to ISO 21501-4 inasmuch as uncertainties are evaluated and described.

It should be noted that, since the applied least squares routine requires uncertainties in all parameters, the uncertainty in the population width σ_C must be known. This can be calculated by following the *Guide to the expression of uncertainty in measurement*.⁸ If the uncertainty cannot be readily determined, we recommend using a value of $\sigma_C/10$ to avoid underestimation.

F. Uncertainty estimation

The uncertainties in the median voltages were estimated using the fact that the distribution of medians from a population with density function $f(x)$ is asymptotically normal with variance $1/(4nf(V_M)^2)$, where n is the sample size.¹⁰ We used Freedman–Diaconis' rule to determine the number of bins in the histogram in order to obtain $f(V_M)$. The calculated variance was combined with the 5 mV uncertainty from NI6133 in a sum of squares.

For the voltage limits to determine the SR, we estimate an uncertainty of one bin width corresponding to $\sim 3\%$.

In the direct approach, we use a standard uncertainty of 0.03 on the calculated ratios $n_{L,(1,2)}/n$. This number stems from an evaluation of the counting efficiency of the LAS-X in different size ranges, where we have used the maximum of the obtained uncertainties.

Where necessary, all uncertainties in the two methods were propagated using the *uncertainties* Python package to correctly take the covariance between correlated terms into account.¹¹

III. RESULTS AND DISCUSSION

Following the experimental procedures described in Secs. II D and II E, we obtain the median voltages and bin count ratios reported in Table I, leading to the values in Table II. For each of the particle ranges 0.3 μm , 0.5 μm , 1 μm , and 5 μm , we report the obtained SSE as well as the SR for both the ISO calibration method and the direct approach. Since uncertainties have been evaluated for the results from both the ISO method and our direct approach, we stress that they are both equally accepted calibration methods in determining the SSE and SR according to ISO 21501-4. The standard furthermore requires that the SSE should be $\leq 10\%$ and the SR should be $\leq 15\%$ for an acceptable calibration, and we can therefore conclude that with both methods, our LSAPC meets the requirements. In most cases, we see that the values overlap within the stated 2σ -intervals, but there are some systematic discrepancies between the results obtained with the two methods, which we will discuss in the following. It should be noted that although the LAS-X is our in-house reference particle counter at DFM, it is not special in this regard. For the experiments performed here, we have used the LAS-X as a regular particle counter, and even though it has higher resolution than regular LSAPCs, the direct approach can be used with any LSAPC.

Regarding the methodology of the ISO method, there is no recommendation in the standard as to how a technician should construct the voltage histograms. Depending on the software used for data treatment, various options will be used as per the program's default setting if not considered (or if unchangeable). This can lead to different results for the distribution widths in the ISO method when determining the SR, since changing the number of bins can result in different positions for the 61% threshold for calculating σ_I , as well as potentially changing the voltage boundaries V_L and V_U when determining the median voltage V_M . In this work, we have used Freedman–Diaconis' rule since this is resilient to outliers and takes data variability and data size into account, but without consensus, this is a source for uncertainties, possibly introduced unintentionally by the specific software used.

A. Effects of voltage distribution skewness

We next pay attention to the seemingly low bin count ratios for the middle particle population in each range, where especially

TABLE II. Summary of the main results comparing the calibration method recommended in ISO 21501-4 and the direct approach. All uncertainties are reported with a 2σ interval. SSE: size setting error. SR: size resolution.

	x_C (nm)	SSE (%)	SR (%)
ISO	296.2 \pm 9.0	-4.5 \pm 6.2	5.7 \pm 7.9
Direct		1.7 \pm 1.0	4.3 \pm 2.8
ISO	493.1 \pm 9.0	-2.3 \pm 4.2	2.2 \pm 7.3
Direct		6.8 \pm 1.8	6.0 \pm 1.8
ISO	945 \pm 13	1.1 \pm 3.9	6.4 \pm 5.4
Direct		2.4 \pm 1.4	7.1 \pm 4.6
ISO	5031 \pm 70	-6.0 \pm 3.9	10.3 \pm 4.7
Direct		1.1 \pm 0.8	2.9 \pm 2.4

the one at the 0.5 μm range stands out at 0.14 (see Table I). Ideally, with an SSE of 0%, this ratio should be 0.5 since half of the particles should fall on either side of the bin boundary represented by the particle mean diameters used as boundary values. This could be potentially explained by a drift over time of the LAS-X over all the particle size ranges, but inspecting an obtained voltage histogram, like in Fig. 2(a), and the corresponding size histogram of a particle population alludes to a different explanation. As we see from Fig. S1, the initial particle population is very close to a normal distribution albeit with a small tail toward smaller sizes. Mie theory predicts a well-known nonlinear and nonmonotonic relationship between particle size and scattered light intensity, where several sizes can result in the same intensity (see Fig. S3a).

In an LSAPC, however, a unique relationship between a given signal intensity and a particle size must exist, which means smoothening of the curve by linear or polynomial fitting. This can lead to an otherwise normally distributed particle population being skewed to the low side both in the voltage and size histogram as we show in the right panel of Fig. S3b, which is a violation of the underlying assumption of normal voltage distributions. The ISO method alleviates this to some extent by using the median voltage for the calculation of the SSE, which yields a slightly lower voltage than the peak position. However, for the SR, the peak position of the voltage histogram is used, which in the case of a skewed distribution has a tendency to overestimate the SR due to the low-side tail. In the direct approach, we see the effect of the skewed distributions expressed in the ratios in Table I. However, the SSE and SR resulting from the direct method are impacted to a smaller degree than in the ISO method. Although the ratio for the middle particle population is lower than expected, the ratio of the higher mean population is also lower, and these offsets will to some extent cancel each other out in the case of the SR. On the other hand, the SSE is generally pushed to higher values for the direct approach by the change in the bin count ratios, and we do also observe that the SSE across the size ranges can be higher than the ISO method.

B. Impact from choice of calibration curve

In addition to the effect discussed above, the specific choice of calibration curve will impact the SSE for the ISO method, which again is a consequence of the Mie nonlinearities. The relationship between the particle size and signal intensity is generally superlinear (e.g., goes as radius cubed in the small-particle limit¹²). Since we have used a linear model for the calibration curve, the curve will always underestimate the actual particle size from a given median voltage when compared to a superlinear curve. This problem is further amplified when the internal calibration curve translating signal voltages to particle sizes in the LSAPC becomes steeper. In addition to this, the ISO standard permits the calibrating party to select an appropriate shape of the curve,⁴ and as such, errors are prone to occur at this step.

Since the created calibration curve is also used for determining the SR through the conversion of the voltages $V_{1,2}$ to the sizes $x_{1,2}$, any errors at that step from the Mie nonlinearities will be inherited by the calculation of the SR. This is especially evident in the case of the 5 μm particle range. There is a significant SSE of -6.0% , meaning that the predicted bin boundary lies around 4730 nm,

which again leads to x_1 being calculated smaller than it actually is. Since the SR must be evaluated against the mean of the CRPs, this leads to the large SR of 10.3% reported here. Examining the size histogram of the 5031 nm population, we can estimate an SR of about 4% as the actual instrument broadening; the remainder comes from the errors induced by the specific choice of calibration curve.

A potential remedy for the challenges associated with the shape of the calibration curve would be to map it out with more CRP populations. However, simply increasing the workload required for each calibration is unfeasible from an application's point of view. In contrast to this, the direct approach always only requires two CRP populations sufficiently close to the bin boundary to give the desired result. Furthermore, our direct approach has no need for any user input regarding the specific form of calibration curves or histograms, instead only relying on the manufacturer's internal calibration curve to translate signal voltages to particle sizes.

Finally, we observe that the uncertainty in all cases of both the SSE and the SR is smaller using the direct approach than in the ISO method. This is most likely due to the reduced number of variables and steps from the measurement to the final result used in our approach. While all errors are propagated and the covariances between terms are taken into account in the ISO method, we altogether avoid the uncertainty contributions from the voltage measurements in the direct approach.

IV. CONCLUSIONS

We have presented a calibration approach that outperforms the calibration method described in ISO 21501-4 in terms of simplicity and workload. Only two measurements are needed in order to confidently determine the SSE and the SR. Furthermore, the fitting routine directly outputs the uncertainties in the result in the process, thus eliminating the need for any further calculations on the user's part, and we also showed that we obtain lower uncertainties with the direct approach compared to the ISO method.

As discussed, there are advantages and disadvantages with each calibration scheme related to the nonlinear origin of the scattered light. The direct approach provides a method that is highly simplified, using only the output values from the instrument. This makes the method accessible to daily users without having to gain access to the sometimes unavailable raw instrument data such as voltage distributions and calibration curves in order to perform calibration with uncertainty evaluations. In addition, the direct approach relies on binned particle data only, providing results that are closely related to its actual modus operandi.

SUPPLEMENTARY MATERIAL

See the [supplementary material](#) for example data from our determination of the normal size distribution of the particles by AFM, detailed description of how to calibrate the lowest bin boundary setting, discussion on how the conversion from normally distributed particle sizes to detector voltage in the particle counter can cause a transformation into a non-normal voltage distribution and how this is caused by the nonlinear Mie response of the particles, and also the presentation of a simple model that reproduces the presented experimental data well.

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