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# Statistical signal analysis for optical aerosol spectrometers: Closing the gap between single particle counting and signal fluctuation analysis



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# ABSTRACT

Light scattering aerosol spectrometers (also known as optical particle counters, OPCs) are widely used for aerosol quantification. The single particle counting method, which is based on light scattering, can measure the size distribution and the number concentration of the sampled aerosol. However, this method is limited to low concentrations due to coincidence error. At higher concentrations, the particle pulses overlap and cannot be counted individually. It was recently shown that the detector signal of an optical aerosol spectrometer can also be evaluated by fluctuation analysis if the concentration is significantly higher than the coincidence limit of the device. This new mode of operation cannot yet provide a detailed size distribution but itis feasible to measure the median particle size and number concentration independently. The measurement information required for fluctuation analysis is drawn from the intensity distribution of the detector signal instead of individual pulses. Therefore, fluctuation analysis requires a certain average number of particles inside the measuring volume so that the detector output continuously leaves the baseline. Theminimum number concentration of the fluctuation analysis is around a factor of 20 higher than the coincidence limit for single particle counting. Consequently, there is a concentration range where neither single particle counting, nor fluctuation analysis can be used.

This work introduces a new statistical signal analysis to bridge this gap. The new measurement method was experimentally verified using a monodisperse di-ethyl-hexyl-sebacat aerosol with a particle size range of  $0.3\,\mu m$  to  $2.2\,\mu m$  and a number concentration range of  $1\times10^4\,cm^{-3}$  to  $2\times10^5\,cm^{-3}$ . An accuracy of  $2\,\%$  with respect to median particle size and  $5\,\%$  with respect to number concentration was achieved. The new method finally closes the gap between single particle counting and fluctuation analysis, enabling light scattering aerosol spectrometers to quantify aerosols at any given concentration.

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Fig. 1. The maximum number concentration for single particle counting is smaller than the minimum particle concentration required for fluctuation analysis. In this work, statistical signal analysis is proposed to close the previously inaccessible concentration gap in the transition regime.

#### 1. Introduction

Many industrial applications require the in-situ quantification of aerosols. Quite often, the number concentration c and a representative size parameter (e.g., number median diameter  $d_{50}$ ) are the quantifies of interest. The light scattering aerosol spectrometer (also known as optical particle counter, OPC) is a popular and versatile instrument, which can be used to quantify aerosols with particle sizes between 60 nm and 45 µm (International Organization for Standardization, 2009). The aerosol is sucked through an illuminated measuring volume, where the light is scattered by each individual particle. The intensity of the scattered light, which depends on the size, shape, and optical properties of the particle, can be converted into an electrical signal. Therefore, every single particle causes a peak on the detector signal, which can be counted and assigned into a specific size class by a pulse height analyzer. Since the relationship between the peak amplitudes and the particle size is defined by a device specific calibration curve (International Organization for Standardization, 2009), the result is the number-based size distribution of the sampled aerosol. Furthermore, the number concentration of the aerosol can be calculated from the number of counted particles, as long as the sampling time and sample flow rate are known. Recently, there have been several approaches introduced to retrieve additional information such as refractive index, sphericity, and fractal dimensions of the particles (Nagy, Szymanski, Gál, Golczewski, & Czitrovszky, 2007; Pitz, Hellmann, Ripperger, & Antonyuk, 2018; Weirich, Misiulia, & Antonyuk, 2024).

Although light scattering aerosol spectrometers are widely used, the method is limited to low number concentrations c. For single particle counting it is crucial that only a single particle passes at a time through the measuring volume V. For higher number concentrations, the average number of particles inside the measuring volume  $c \cdot V$  increases. Therefore, it becomes more likely that the signal of two particles will overlap each other and cannot be separated by the detector anymore. Those coincidence events reduce the counting efficiency  $\eta$ 

$$\eta = e^{-c \cdot V} \tag{1}$$

and shift the measured size distribution towards coarser particles (International Organization for Standardization, 2009; Oeser et al., 2023; Raasch, 1984; Sachweh, Umhauer, Ebert, Büttner, & Friehmelt, 1998). A common approach to measuring higher number concentrations is to reduce the measuring volume (Sachweh et al., 1998). But due to physical or application specific requirements, this method is limited. Until a certain extent, the effects of the coincidence error can also be corrected (Oeser et al., 2023; Raasch, 1984; TSI Incorporated, 2023). However, an average number of particles inside the measuring volume of  $c \cdot V = 0.1$  is generally considered as coincidence limit for light scattering aerosol spectrometers (International Organization for Standardization, 2009). Even though it was shown that this limit can be extended up to  $c \cdot V = 0.3$  by advanced peak detection algorithms (Oeser, Samala, Hillemann, Rudolph, & Lienig, 2022), the approach of single particle counting is not suitable for high number concentrations.

If the average amount of particles inside the measuring volume is large ( $c \cdot V \gg 1$ ), the detector signal deviates from the baseline almost continuously, as there is nearly always at least one particle present in the measuring volume (Jani et al., 2002; Oeser et al., 2024). In this mode of operation, the instrument becomes a photometer (Sorensen, Gebhart, O'Hern, & Rader, 2011). Photometers can be implemented either in a scattering or extinction-based setup. Nevertheless, the drawback of this method is that conventional photometers can only provide one measurand (typically mass concentration), since only the average light intensity is evaluated. The independent measurement of both size and concentration requires additional information. For liquid-borne extinction photometers it is known that the required information can be found in the standard deviation of the detector signal (Karlsson & Pettersson, 1982). Based on the Beer–Lambert law (Beer, 1852) an analytical relationship between the probability distribution of the amplitude of the detector signal and particle size and concentration was found (Wessely, 1999). With the extinction-based fluctuation analysis an accuracy of about 5% with regards to particle size and 12% with regards to volume concentration can be achieved (Dannigkeit, Steinke, & Ripperger, 2010). At high concentrations, the errors increase due to parasitic effects such as multiple scattering. This can be minimized by using spatial frequency filters (Schwarz, Ripperger, & Antonyuk, 2018).

Since the Beer-Lambert law applies exclusively to extinction-based setups, it is not suitable for angularly scattered light measurements. Nevertheless, it is known that also for scattering setups a unique assignment between the probability distribution of the amplitude of the detector signal and size and concentration of the aerosol exists (Jani et al., 2002; Oeser et al., 2024). Although

the relationship cannot be fully described analytically, a calibration function found by polynomial regression can be used for the simultaneous measurement of median particle size and number concentration (Oeser et al., 2024).

Scattered-light fluctuation analysis, described in Oeser et al. (2024), might be integrated into an light scattering aerosol spectrometer as a high-concentration mode. The user would then be able to choose between single particle counting for measurements at lower, and fluctuation analysis at higher, number concentrations. In theory, this feature would expand the range of application of an light scattering aerosol spectrometer, to any number concentration, which would result in an universal aerosol instrument. However, the combination of single particle counting and fluctuation analysis would not really cover any number concentration. An average amount of particles inside the measuring volume above  $c \cdot V > 2$  was found to be required for the detector signal to leave the baseline continuously (Oeser et al., 2024). Otherwise, the probability distribution would not be suitable for evaluation.

Assuming a scattered light setup with a given measuring volume, there is a concentration gap of a factor 20 where neither single particle counting, nor the fluctuation analysis is suitable (refer to Fig. 1). This work demonstrates how the characteristics of the detector signal can be utilized to measure number concentration and median particle size of a monodisperse aerosol in the transition regime. The fundamental idea for the number concentration measurement is based on a patent that describes the measurement of number concentration and volumetric flow for an extinction setup (Altmann & Wessely, 2006).

Our approach is based on the continuous sampling of the detector signal. For each measurement, the average signal amplitude and the proportion of time the signal remains below a predefined threshold value are calculated. These two quantities serve as the basis for estimating the number concentration and the median particle size. The output parameters are calculated using a two-dimensional regression function which is determined by calibration against a reference aerosol.

In Section 2.1 the theoretical foundation of this approach is first developed using rectangular-shaped particle pulses, as this simplification provides a clearer understanding of the mathematical background of our method. Subsequently, the theory is verified by a computer simulation and extended to Gaussian-shaped particle pulses, which are commonly encountered in light scattering aerosol spectrometers (Section 2.2). Finally, Section 2.3 presents the experimental setup used to validate the method on real aerosol measurements (Section 3).

#### 2. Materials and methods

#### 2.1. Theory

Within the concentration range between  $c \cdot V = 0.1$  and  $c \cdot V = 2$ , a significant fraction of the particle pulses overlap with each other, so it is no longer feasible to count single particles. The amplitude of the detector signal as a function of time U(t) leaves the baseline, depending on the actual number concentration, for a moderate to considerable proportion of time (around 5% to 95%), but not permanently as required for fluctuation analysis.

Consider an ideal monodisperse aerosol moving through a measuring volume of a scattered light setup with a completely homogeneous light intensity and a constant velocity. If the particle size is negligible compared to the geometry of the measuring volume, each particle will generate a rectangular-shaped pulse with a constant height and width. During a coincidence event, the total amplitude of the detector signal is assumed to be the sum of the amplitudes of all individual particles inside the measuring volume. The time discrete detector signal, sampled by an analog-to-digital converter (ADC), might then look like the one shown in Fig. 2. For every sample k, the measured amplitude of the detector signal  $U_k$  is compared to a threshold  $U_{th}$ , which is set to a value between the baseline and the amplitude of a single particle peak (dashed line at 1 V in Fig. 2). The total number of samples n and the number of samples above the threshold  $n_{th}$  are counted. Assuming a large number of samples, the proportion of time of the detector signal below the threshold is given by

$$P = \frac{n - n_{\rm th}}{n} \tag{2}$$

(cf. Altmann and Wessely (2006)) and corresponds to the probability of no particle within the measuring volume

$$P = \frac{(c \cdot V)^0}{0!} \cdot e^{-c \cdot V} = e^{-c \cdot V}$$
(3)

Raasch (1984) which is, according to Poisson statistics, given by number concentration c and measuring volume V. Eqs. (2) and (3) can be rearranged as

$$c = -\frac{1}{V} \cdot \ln\left(\frac{n - n_{\rm th}}{n}\right) \tag{4}$$

to calculate the concentration of the sampled aerosol when the measuring volume is known. Note that, for the simplification of rectangular-shaped particle pulses, the concentration can be calculated completely independent of the particle size as long as the amplitude of the particle pulses is higher than the selected threshold. Note that Eq. (4) is only valid for rectangular shaped particle pulses.

In single particle counting mode, the amplitude  $U_i$  of a particle pulse

$$U_{i} = f(d) \tag{5}$$

is a function f of its size d (optical equivalent diameter). Although the function can be calculated by the Mie theory, it is more common to determine the relationship between particle size and peak amplitude by calibration with reference particles (International



**Fig. 2.** Principle of statistical fluctuation analysis on a simulated detector signal with rectangular shaped particle pulses and constant amplitude. The 0.5 V baseline offset is caused by the dark current of the photo detector. The stems show the measured amplitudes  $U_k$  for each time step k of the detector signal (solid curve). The dashed line represents the selected threshold value  $U_{th}$ . For the shown sequence, the total number of samples is n = 21, the number of samples above the threshold is  $n_{th} = 13$ . This would give a probability of P = 38.1% for no particle inside the measuring volume. The average value of the detector signal is  $\overline{U} = 1.21$  V.

Organization for Standardization, 2009). For an ideal monodisperse aerosol (each particle causes a peak with the same height) the average amplitude  $\overline{U}$  of the detector signal can be written as

$$\overline{U} = c \cdot V \cdot U_{i} = c \cdot V \cdot f(d) \tag{6}$$

since the average amount of particles within the measuring volume is given by  $c \cdot V$ . The average amplitude of the detector signal

$$\overline{U} = \frac{1}{n} \cdot \sum_{k=0}^{n} U_k \tag{7}$$

can be easily measured by calculating the sum of each measured amplitude  $U_k$  and dividing it by the number of samples *n*. Subsequently, Eqs. (6) and (7) can be rearranged as

$$d = f^{-1}(U_{i}) = f^{-1}\left(\frac{\overline{U}}{c \cdot V}\right)$$
(8)

to calculate the particle size, on the basis of the calibration function from single particle counting mode. According to Eq. (8), the result of the particle size measurement depends not only on the average amplitude of the detector signal, but also on the number concentration of the sampled aerosol, which is given by Eq. (4).

#### 2.2. Simulation

The theoretical model can be verified by numerical simulation. This requires a geometric definition of the measuring volume and the definition of the aerosol flow that passes through it. Consequently, the width of an individual particle pulse can be calculated. The simulation step size is then chosen to be significantly smaller (e.g., by a factor 100), so that it is ensured that the form of the detector signal is well described. The simulation, like a real measurement can only be performed over a finite period of time. The detector signal is initialized with a constant value. Then, individual pulses with a given amplitude are added at randomly selected positions one by one. The total amount of particle pulses is defined by the number concentration, but follows a Poisson distribution. Coincidence events may occur within the considered time frame, also in a random manner. Both effects contribute to statistical uncertainties in the parameters extracted from the simulated detector signal, such as the proportion of time below the threshold and the average amplitude. It can be shown that these statistical deviations decrease for longer simulated measurement times. In practice, the simulated measuring time is a compromise between a sufficient number of particle pulses on the detector signal to keep the statistical errors low and keeping the computational effort reasonable.

The simulation allows to generate different synthetic signals for a variety of number concentrations and peak amplitudes. From these, the parameters proportion of time where the detector signal remains below the threshold P and the average signal amplitude  $\overline{U}$  can be extracted. Now, it is possible to compute for rectangular and Gaussian-shaped pulses how these two parameters behave as a function of number concentration and particle size.

Fig. 3 shows the simulation result for the proportion of time where the detector signal is below the threshold P in dependency of the number concentration. The simulation is in good agreement with Eq. (3). The small deviations of the black dots are due to statistical errors, as only a short measurement interval of 100 ms was analyzed in order to keep the computation time manageable.

(12)



**Fig. 3.** Simulated proportion of time of the detector signal below the threshold P for different number concentrations. For a given particle size, the relationship is consistent with the theory (exponential decay, Eq. (3)) for rectangular shaped particle pulses. As expected, the Gaussian-shaped particle pulses cause a size dependency. The proportion of time below the threshold decreases for coarser particles.

Increasing the measuring interval would cause the simulation result to approach even further. The colored dots, on the other hand, represent the simulation result for Gaussian-shaped particle pulses. The peaks were modeled by

$$U(t) = U_{i} \cdot \exp\left(-2 \cdot \left(\frac{t}{\Delta t/2}\right)^{2}\right)$$
(9)

so that the peak with  $\Delta t$  is defined as the period of time where the detector signal is above  $1/e^2$  (approx. 13.5%) of the maximum amplitude, since this convention is commonly used to describe the spot size of a Gaussian-shaped laser beam (Siegman, 1986). According to Fig. 3, the Gaussian-shaped particle pulses cause the theoretical model of Eq. (3) to be significantly affected by the peak amplitude. The reason for this behavior is that the period of time when the detector signal is above the threshold  $\Delta t_{th}$  is now a function of the ratio between amplitude of the particle pulse  $U_i$  and threshold value  $U_{th}$ . The ratio of the time above the threshold  $\Delta t_{th}$  to the initial peak width  $\Delta t$  is described by

$$\frac{\Delta t_{\rm th}}{\Delta t} = \frac{1}{\sqrt{2}} \sqrt{\ln(U_{\rm i}) - \ln(U_{\rm th})} \tag{10}$$

for Gaussian-shaped pulses, while

$$\frac{\Delta t_{\rm th}}{\Delta t} = 1 \tag{11}$$

is independent of the peak amplitude  $U_i$  for the rectangular shaped particle pulses. Eq. (10) might be used to derive an alternative model to calculate the probability for no particle within the measuring volume (cf. Eq. (2)) that is more suitable for Gaussian peak shapes. When doing so, the amplitude of different coincidence scenarios must be considered, since the added signal of two particle pulses might be substantially longer above the threshold than it would be for separated particles. This makes it challenging to find a proper analytical model. However, the amplitude dependency is not the only effect that increases the time where the detector signal is above the threshold. The model described above, considers the particles to be infinitely small. Therefore, the peak width is only as long as the particle needs to pass the measuring volume. Light scattering aerosol spectrometers can focus the laser beam, so that the measuring volume has a height of only a few micrometers, i.e., is in the same order of magnitude of the particles being measured (International Organization for Standardization, 2009). When particle size is not negligible, the detector signal would start to rise as soon as the edge of the particle enters the measuring volume and falls until the particle has completely passed through. If a particle is approximately the same size as the height of the measuring volume, the detector signal remains above the baseline for roughly three times longer.

Fig. 4 shows the simulation results for the average amplitude of the detector signal  $\overline{U}$ , which is intended to be used to calculate the particle size from the number concentration (cf. Eq. (6)). The black lines indicate the theoretical model for rectangular shaped particle pulses for an average number of particles within the measuring volume of  $c \cdot V = 0.1$ ,  $c \cdot V = 0.3$ , and  $c \cdot V = 2$  respectively. Despite minor statistical fluctuations caused by the short simulation time, the simulation results for the rectangular shaped particle pulses (not shown in Fig. 4) match the theoretical model. The colored dots represent the simulation result for Gaussian-shaped particle pulses. The Gaussian pulse shape does neither affect the linear dependency of the amplitude of the particle pulses are just added up. The integral of a single rectangular shaped particle pulse

$$I_{\rm r} = U_{\rm i} \cdot \Delta t$$



**Fig. 4.** The average amplitude of the detector signal  $\overline{U}$  is linear dependent of both, peak amplitude and number concentration (Eq. (6)). The black lines show the theoretical result for rectangular shaped pulses at a concentration of  $c \cdot V = 0.1$ ,  $c \cdot V = 0.3$ , and  $c \cdot V = 2$  respectively (bottom to top). The Gaussian peak shape does not affect the general relationship, but causes a shift on the logarithmic scale.

differs from the integral of a single Gaussian-shaped particle pulse

$$I_{g} = \int_{-\infty}^{\infty} U_{i} \cdot \exp\left(-2 \cdot \left(\frac{t}{\Delta t/2}\right)^{2}\right) dt.$$
(13)

Therefore, the average amplitude of the detector signal for Gaussian-shaped pulses is a constant factor

$$\frac{I_{\rm r}}{I_{\rm g}} = \frac{2 \cdot \sqrt{2}}{\sqrt{\pi}} \approx 1.6 \tag{14}$$

smaller than for rectangular shaped pulses, which causes a shift on the logarithmic scale (Fig. 4).

Although the simulation is not capable of accounting every detail of a real-world light scattering aerosol spectrometer, some basic conclusions can be derived. The proportion of time where the detector signal is below the threshold P does not only depend on the number concentration as assumed, but also on the particle size. The question arises whether there is still a unique relationship to determine particle size and number concentration independently of each other (similar to Eqs. (4) and (8)).

For Gaussian-shaped particle pulses, the average amplitude of the detector signal

$$\overline{U} = \sqrt{\frac{\pi}{8}} \cdot c \cdot V \cdot U_{\rm i} \tag{15}$$

scales linearly with both the pulse amplitude of a single particle  $U_i$  and number concentration c, provided that multiple scattering and extinction from neighboring particles can be neglected. Finding and exact analytical expression

$$P = f(c, U_i) \tag{16}$$

for Gaussian-shaped particle pulses is quite challenging (refer to Eq. (4) for rectangular shaped particle pulses). However, it is known that the proportion of time in which the detector signal remains below the threshold is strictly monotonically decreasing with increasing pulse amplitude

$$\frac{\partial f(c, U_i)}{\partial U_i} < 0 \tag{17}$$

for a given concentration. Furthermore, the proportion of time in which the detector signal remains below the threshold is strictly monotonically decreasing with increasing number concentration

$$\frac{\partial f(c, U_i)}{\partial c} < 0 \tag{18}$$

for a given pulse amplitude. Eq. (15) can be rearranged with respect to concentration or amplitude and substituted into Eq. (16). This yields

$$P = f(\sqrt{\frac{8}{\pi}} \cdot \frac{\overline{U}}{V \cdot U_{\rm i}}, U_{\rm i})$$
<sup>(19)</sup>

or

1

$$P = f(c, \sqrt{\frac{8}{\pi}} \cdot \frac{\overline{U}}{V \cdot c}).$$
(20)

2 f(. II)

.

Eqs. (19) and (20) each have a unique solution if P is strictly monotonic in  $U_i$  or c. Using the chain rule, it follows that

$$\frac{\partial P}{\partial U_{i}} = \frac{\partial f(c, U_{i})}{\partial c} \cdot \frac{dc}{dU_{i}} + \frac{\partial f(c, U_{i})}{\partial U_{i}}$$

$$= -\sqrt{\frac{8}{\pi}} \cdot \frac{\overline{U}}{V \cdot U_{i}^{2}} \cdot \frac{\partial f(c, U_{i})}{\partial c} + \frac{\partial f(c, U_{i})}{\partial U_{i}}$$
(21)
(22)

and

$$\frac{\partial P}{\partial c} = \frac{\partial f(c, U_{\rm i})}{\partial c} + \frac{\partial f(c, U_{\rm i})}{\partial U_{\rm i}} \cdot \frac{\mathrm{d}U_{\rm i}}{\mathrm{d}c}$$
(23)

$$=\frac{\partial f(c,U_{\rm i})}{\partial c} - \sqrt{\frac{8}{\pi}} \cdot \frac{\overline{U}}{V \cdot c^2} \cdot \frac{\partial f(c,U_{\rm i})}{\partial U_{\rm i}}.$$
(24)

Due to the sign change introduced by the derivative, the inequalities (17) and (18) do not ensure that

36(. 11)

$$-\sqrt{\frac{8}{\pi}} \cdot \frac{\overline{U}}{V \cdot U_{i}^{2}} \cdot \frac{\partial f(c, U_{i})}{\partial c} + \frac{\partial f(c, U_{i})}{\partial U_{i}}$$
(25)

or

$$\frac{\partial f(c, U_{\rm i})}{\partial c} - \sqrt{\frac{8}{\pi}} \cdot \frac{\overline{U}}{V \cdot c^2} \cdot \frac{\partial f(c, U_{\rm i})}{\partial U_{\rm i}}$$
(26)

are consistently negative or positive for all combinations of c and  $U_i$ . Therefore, P is not necessarily strictly monotonic in  $U_i$  or c. Without a proper analytical expression of Eq. (16), it cannot be mathematically proven that both concentration and size can be uniquely determined. However, no ambiguities were observed in the subsequent measurements.

#### 2.3. Experiment

The theory as well as the simulation served for a general understanding of statistical signal analysis and showed that the models do not cover all relationships in perfect detail. Therefore, just like single particle counting, it becomes necessary to calibrate the device, so that number concentration and particle size of the aerosol can be measured. For single particle counting, light scattering aerosol spectrometers are usually calibrated against a set of certified reference particles like spherical polystyrene (PSL) particles. Here, the number concentration of the calibration aerosol is not of interest, as long as it is well below the coincidence limit. The proposed statistical signal analysis however, does not calculate the number concentration based on the amount of events which occur during a specified interval, but on the statistics of the detector signal itself. Hence, it becomes necessary to calibrate the device against a size and a concentration standard. This can be done with a monodisperse aerosol which is varied in size and number concentration.

The experimental investigations in this work were done with the same setup as presented in Oeser et al. (2024) and which is shown in Fig. 5. A monodisperse aerosol (containing droplets generated from DEHS) is generated by a Sinclair-La Mer-Type particle generator ① (SLG 270, Topas GmbH, Dresden, Germany). The number concentration and the particle size of the produced aerosol mainly depends on the number of primary particles that act as condensation nuclei, as well as the amount of DEHS evaporated in the reheater (Altmann & Peters, 1992; Sinclair & La Mer, 1949). Since a change in the number of primary particles would change the amount of DEHS available for the condensation process on each single particle, this parameter does effect both, particle size and number concentration. So, the number of primary particles, and therefore also the number concentration of the generated aerosol, was held constant during the experiments, while the particle size was set by the saturator temperature. The aerosol was diluted in a mixing chamber ②. A controlled blower ③ (RFU 564, Topas GmbH, Dresden, Germany) with an internal and an external high-efficiency particulate air (HEPA)-filter was used in closed loop configuration, to remove particles and set the number concentration of the final test aerosol, depending on the chosen flow rate.

The statistical signal analysis was performed with a modified light scattering aerosol spectrometer ④ (LAP 323, Topas GmbH, Dresden, Germany). Since the used light scattering aerosol spectrometer splits the sample aerosol flow of  $3 \text{ L} \text{min}^{-1}$  internally into the effective detection flow rate of  $0.1 \text{ L} \text{min}^{-1}$  and a bypass volume flow of  $2.9 \text{ L} \text{min}^{-1}$  (GmbH, 2020), the capillary of the measuring cell was directly connected to the mixing chamber by a short tube, to avoid an unnecessary DEHS contamination of the device. Further, the light scattering aerosol spectrometer was equipped with a field programmable gate array platform (FPGA, Eclypse Z7, Digilent Inc., Pullman, USA) and an analog front end (Zmod Scope 1410-125, Digilent Inc., Pullman, USA) to probe the detector signal directly from the preamplifier with a sampling rate of 5 MHz. The field programmable gate array (FPGA) was programmed to measure the average amplitude of the detector signal as well as the proportion of time the signal remained below a threshold. The threshold value was set as close as possible above the baseline of the detector signal with the boundary condition that the number of samples above the threshold is  $n_{\text{th}} = 0$  for a 5 min measurement interval in clean air.

The reference measurement of number median particle size  $d_{50}$  and number concentration *c* was carried out with a second light scattering aerosol spectrometer (5) (OPS 3330, TSI Inc., Shoreview, USA). The reference instrument has a maximum measurable number concentration of  $3000 \text{ cm}^{-1}$  (TSI Inc., 2024). Since the experiments were carried out at significantly higher number concentrations (refer to Fig. 7) two dilution systems (6) (DIL 550, Topas GmbH, Dresden, Germany) were connected in series to achieve a dilution factor of around 16000. As can be seen in Fig. 6, this factor is independent of the particle size and therefore allows for the calculation of the actual number concentration of the test aerosol from the number concentration of the reference instrument. An additional pump (7) with a flow rate of  $3 \text{ L} \text{ min}^{-1}$  was used to keep the setting time through the mixing chamber and the dilution system minimal.

![](_page_7_Figure_2.jpeg)

Fig. 5. Setup for the statistical signal analysis. The light scattering aerosol spectrometer for the measurement of high concentrations is calibrated against a reference light scattering aerosol spectrometer, which quantifies the diluted aerosol.

![](_page_7_Figure_4.jpeg)

Fig. 6. Scaled dilution factor (i.e., ratio of actual value to set value) of a DIL 550 dilution system as function of particle size. Results based on two simultaneously operated condensation particle counters (CPC, Model 3772, TSI Inc., Shoreview, USA) and monodisperse DEHS aerosols composed of electrostatically classified DEHS droplets.

#### 3. Results and discussion

For each measurement, the concentration and particle size are varied by the saturator temperature of the generator and the flow rate of the blower. A lead time of 30 s was accounted before each measurement. For each datapoint, a 30 s measurement of number concentration c and number median particle size  $d_{50}$  was performed by the reference spectrometer. At the same time, the probability for no particle within the measuring volume P and the average signal amplitude  $\overline{U}$  was recorded by the FPGA. Fig. 7 shows the obtained dataset which is used for the statistical signal analysis. It can be deduced that the median particle size of the monodisperse DEHS aerosol (Oeser et al., 2024) was varied within a range of approximately  $0.3 \,\mu\text{m} < d_{50} < 2.2 \,\mu\text{m}$ . The 0.3  $\mu\text{m}$  limit is equal to the lower detection limit of the reference spectrometer (TSI Inc., 2024). Note that statistical signal analysis can also be used to measure finer or coarser aerosols, depending of the specific setup. The number concentration was varied within a range of approximately  $1 \times 10^4 \,\text{cm}^{-3} < d_{50} < 2 \times 10^5 \,\text{cm}^{-3}$ . This corresponds to  $0.1 < c \cdot V < 2$  of the light scattering spectrometer used in this work. The vertical lines in Fig. 7 represent the limits for single particle counting (red), single particle counting with dead time reduction (dashed red), and fluctuation analysis (blue) respectively. Accordingly, the complete transition regime between single particle counting and fluctuation analysis is covered by the dataset.

Each data-point contains information about the median particle size  $d_{50}$  and c of the aerosol measured by the reference spectrometer, as well as the parameters P and  $\overline{U}$  measured by the device under test (DUT). Fig. 8 shows a scattering plot of the proportion of time where the detector signal is below the threshold P against number concentration c. The color of the points indicates the particle size. It can be deduced that the parameter P decreases at higher number concentrations as expected (Eq. (3)). The observed behavior is in good agreement with the simulation result for Gaussian-shaped particle pulses (Fig. 3). As predicted by the simulation, for a given number concentration, the time of the detector signal below the threshold decreases for coarser particles, because of the higher pulse amplitudes.

![](_page_8_Figure_2.jpeg)

**Fig. 7.** Dataset for statistical fluctuation analysis. The solid red line represents the coincidence limit for single particle counting of the modified light scattering spectrometer ( $c \cdot V = 0.1$ ) with a threshold based peak detection algorithm. The dashed red line takes dead time reduction into account ( $c \cdot V = 0.3$ ) (Oeser et al., 2022). The blue line represents the minimum number concentration for fluctuation analysis ( $c \cdot V = 2$ ) (Oeser et al., 2024).

![](_page_8_Figure_4.jpeg)

Fig. 8. Measured proportion of time of the detector signal below the threshold *P* for different number concentrations. As predicted by the simulation, the Gaussian-shaped particle pulses cause a size dependency. The proportion of time below the threshold decreases for coarser particles.

Fig. 9 shows the scattering plot of the average signal amplitude  $\overline{U}$  against the median particle size. At first glance, the measured relationship seems to differ from the simulation results in Fig. 4. Note that the abscissa does not represent the peak amplitude, but the particle size measured by the reference instrument. According to Mie theory, the transfer function between particle size and pulse amplitude is nonlinear due to electromagnetic resonances (International Organization for Standardization, 2009), which explains the observations. For a given number concentration, the datapoints show a relationship similar to the calibration function of the aerosol spectrometer. If the particle size is significantly smaller than the wavelength, the theoretical calibration function would exhibit a steep gradient ( $U_i \propto d^6$ ) (International Organization for Standardization, 2009). In contrast, for particles significantly larger than the wavelength, the peak amplitude would be proportional to the square of the size ( $U_i \propto d^2$ ). In between, if the particle size is close to the wavelength, the transfer function of an light scattering spectrometer can, in general, even have some oscillations, which make the exact measurement of the particle size impossible (International Organization for Standardization, 2009). However, the light scattering spectrometer used in this work utilizes two laser sources with different wavelengths (blue 450 nm, red 660 nm) which are matched, so that an unique assignment between peak amplitude and particle size can be guaranteed (GmbH, 2020).

Since the device specific calibration function (Eq. (5)) of the spectrometer is known, it can be used to calculate the peak amplitude of a single particle pulse. Therefore, the abscissa in Fig. 9 can be substituted which results in Fig. 10. This allows a direct comparison to the predictions of the theoretical model. The dependency of the average signal amplitude and the number concentration does fairly match the results of the simulation (Fig. 4) and seems quite linear. Obviously, the effects of multiple scattering can be neglected if on average less than two particles are within the measuring volume.

The statistical signal analysis requires two transfer functions to calculate particle size  $d_{50} = f_1(P, \overline{U})$  and number concentration  $c = f_2(P, \overline{U})$  from the proportion of time where the detector signal is below the threshold, and the average value of the detector signal. Both functions are found by two-dimensional least-squares polynomial regression on the dataset. The resulting surfaces can be used to calculate size or concentration for a given set of *P* and  $\overline{U}$ . A proper definition of the polynomial order in the direction of *P* and  $\overline{U}$  is crucial. If the polynomial order is chosen too low, the surface is not able to describe more detailed features. In contrast,

![](_page_9_Figure_2.jpeg)

Fig. 9. Average signal amplitude of the detector signal  $\overline{U}$  in dependency of the median particle size of the test aerosol. For a given number concentration, the relationship is similar to the calibration function of an aerosol spectrometer.

![](_page_9_Figure_4.jpeg)

Fig. 10. Average signal amplitude of the detector signal  $\overline{U}$  in dependency of the peak amplitude. Same data as in Fig. 9. However, the abscissa has been substituted by the device specific calibration function. Now the result matches the simulation (cf. Fig. 4).

if the polynomial order is chosen too high, the regression function over-fits, which means that the surface does approximate the measured data well, but performs worse on new data. To find a proper choice for the polynomial order, the data is randomly split into three datasets, which contain 80%, 10%, and 10% of the data. The regression functions are then calculated exclusively on the first dataset for different sets of polynomial orders. The regression functions are tested with the second dataset to find the best-regularized regression surface for size and concentration respectively, which can now be used for statistical signal analysis. The actual model is tested on the third dataset. So that the evaluation is performed only with data which is neither used to calculate the surface nor to find the best polynomial order.

The histograms in Figs. 11 and 12 show the absolute frequencies of the relative errors for estimation of median particle size and number concentration on the test data. The average and maximum relative errors are given in Table 1. For the measurement of the median particle size an accuracy of about 2% can be expected. The number concentration can be measured with an accuracy of about 5%. Hence, the statistical signal analysis in the transition regime appears to be more precise than the fluctuation analysis for higher number concentrations. Here, an accuracy of about 5% with respect to size and 10% with respect to concentration can be achieved (Oeser et al., 2024). Similar to the fluctuation analysis, the statistical signal analysis can estimate the size better than the number concentration. Table 1 shows also the average and maximum relative errors for the complete dataset, including the data used for regression and selection of polynomial degree. The values are close to the errors obtained for the raw test data. This indicates that the regression functions generalize the dataset well. The difference of the average relative error for the number concentration is caused by a single outlier (cf. Fig. 13). Due to the small amount of test datapoints (37), this value has a significant influence. When the outlier is excluded, the average relative error for the measurement of the test data is 3.6%.

Fig. 13 shows a scattering plot of the absolute relative errors for the number concentration measurement in dependency of the actual number concentration. It appears that the uncertainty is higher for smaller concentrations. A pure statistical error seems unlikely to explain this behavior, since even for concentrations of only  $1 \times 10^4$  cm<sup>-3</sup> approximately half a million particles contributed to the detector signal for each measurement. Even for concentrations below  $2 \times 10^4$  cm<sup>-3</sup>, the error is mostly below 10% and exhibits 15% only a few times. This accuracy might be sufficient for some applications. Especially in combination with the improvements proposed in Oeser et al. (2022), single particle counting can be used for this lower number concentration range. The relative errors

![](_page_10_Figure_2.jpeg)

Fig. 11. Relative measurement error for the median particle size  $d_{50}$  of the statistical signal analysis on the test data.

![](_page_10_Figure_4.jpeg)

Fig. 12. Relative measurement error for the number concentration c of the statistical signal analysis on the test data.

Table 1

Absolute relative error of the statistical signal analysis in the transition regime between single particle counting and fluctuation analysis.

Parameter	Test data		Complete	
	Max.	Avg.	Max.	Avg.
d <sub>50</sub>	6.3%	1.9%	10.3%	2.0%
с	28.1%	4.6%	28.1%	3.6%

for the number concentration measurement did not show a dependency on the particle size. The relative errors for the measurement of the median particle size show a negligible dependency of the particle size, while there was no dependency of the number concentration. This proves that the proposed statistical signal analysis is suitable to measure both median particle size  $d_{50}$  and number concentration c independently of each other within the complete transition regime between single particle counting and fluctuation analysis. This enables the light scattering spectrometer, or other scattering-based setups, to quantify aerosols of any number concentration.

### 4. Conclusion

Scattered light setups can determine the particle size and number concentration of an aerosol either by single particle counting, which is limited to low concentrations only, or by fluctuation analysis, which is limited to high concentrations only. For a given measuring volume, the minimum number concentration of the fluctuation analysis is approximately 20 times higher than the maximum concentration for single particle counting. Consequently, there is a concentration range where neither single particle counting, nor fluctuation analysis is applicable. In this work the method of statistical signal analysis was proposed to specifically address the transition regime. After theoretical investigations and numerical simulations for rectangular-shaped and Gaussian-shaped particle pulses, the method was experimentally validated using a monodisperse DEHS-aerosol with a median particle size range of

![](_page_11_Figure_2.jpeg)

Fig. 13. Absolute relative error for the measurement of particle size in dependency of the number concentration of the aerosol.

 $0.3 \,\mu\text{m} < d_{50} < 2.2 \,\mu\text{m}$  and a number concentration range of  $1 \times 10^4 \,\text{cm}^{-3} < d_{50} < 2 \times 10^5 \,\text{cm}^{-3}$ . The accuracy with respect to median particle size was found to be around 2%, and 5% with respect to number concentration. This makes the new measurement method suitable for several aerosol monitoring applications. Although it may be possible to develop a dedicated instrument for statistical signal analysis, the method can also be implemented as an add-on to light scattering spectrometers alongside single particle counting and fluctuation analysis. Such an instrument would, in principle, be capable of measuring aerosols at any number concentration. In this work, the statistical signal fluctuation analysis was performed with a monodisperse aerosol only. Further investigations might be necessary to adapt the method for polydisperse aerosols. As with all optical measurement techniques, potential inaccuracies can arise due to variations in the refractive index or deviations from the assumption of spherical particle shape. These factors may influence the accuracy of the determined particle size and concentration. Future studies should systematically evaluate these effects and explore potential correction methods.

#### CRediT authorship contribution statement

Lukas Oeser: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. Benno Wessely: Writing – review & editing, Validation, Formal analysis, Conceptualization. Nakul Samala: Writing – review & editing, Writing – original draft, Formal analysis, Data curation. Lars Hillemann: Writing – review & editing, Supervision, Project administration, Formal analysis. Daniel Göhler: Writing – review & editing, Visualization. Jan Müller: Supervision, Project administration. Claudia Jahn-Wolf: Supervision, Project administration. Andreas Rudolph: Supervision, Project administration, Funding acquisition. Jens Lienig: Supervision, Project administration.

#### Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Lukas Oeser, Benno Wessely, Nakul Samala, Lars Hillemann, Daniel Göhler, Jan Müller, Claudia Jahn-Wolf, Andreas Rudolph, and Jens Lienig have no conflict of interest. Lukas Oeser, Nakul Samala, Lars Hillemann, Daniel Göhler, Jan Müller, Claudia Jahn-Wolf, and Andreas Rudolph are employees of the Topas GmbH, a company that develops, produces and markets aerosol technologies for research and industrial applications.

#### Data availability

The data that has been used is confidential.

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