Drop size measurement techniques for sprays: Comparison of image analysis, phase Doppler particle analysis, and laser diffraction

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ABSTRACT

Four different methods for measuring droplet size distributions are evaluated: the Image Analysis VisiSizer technique, a stroboscopic imaging method developed in-house, phase Doppler particle analysis (PDPA), and laser diffraction (Malvern Spraytec). We find that the larger the droplets, the bigger the differences between the results obtained by the different methods. The Image Analysis VisiSizer technique yields results that are comparable with those of the stroboscopic imaging method, provided that the raw Visisizer data are used, as the VisiSizer software makes corrections that can skew the results. Our measurements confirm how the limitations of PDPA can influence its outcomes; the presence of air bubbles inside droplets will cause PDPA to mistake them for smaller droplets. The fact that PDPA reports no droplets larger than 1200 μm might be caused by large drops often not being spherical. The results of the laser diffraction technique are influenced by its fitting method to obtain the droplet size distribution and by overestimation of the number of small droplets due to their low velocity and thus higher concentration in the sample volume. Our results emphasize the need for selecting the size measurement technique to fit the physical nature and expected range of droplet parameters.

I. INTRODUCTION

The size distribution of droplets is important in many applications and everyday life events. One example of current interest is the contribution of droplets to the spread of infectious diseases, such as influenza and severe acute respiratory syndrome (SARS), where the droplet size plays a key role in the travel distance of virus containing droplets[1] and hence their ability to reach the respiratory system.[2–5] The droplet size is also of particular importance to spray applications such as pesticide spraying in agriculture,[6] inkjet printing,[7] de-icing, spray painting, firefighting, and drug administration.[8,9]

While there are various techniques for measuring droplet size distributions,[10,11] information on their accuracy and on how they compare with each other is limited and scattered, especially for droplet diameters of over 10 μm. In this paper, we compare three widely used measurement principles to determine droplet size distributions: (1) image analysis, (2) phase Doppler particle analysis (PDPA), and (3) laser diffraction.

The first technique uses light to image an ensemble of droplets, followed by a software analysis of the snapshot to determine the droplet sizes. By taking subsequent snapshots, the droplet velocities can also be determined.

In the PDPA technique, two laser beams are focused such that they intersect each other. The measurement point is defined by this intersect, where the laser beams interfere and generate a set of parallel equidistant fringes. As a droplet passes the fringes, it scatters light. The receiving optics placed at a well-chosen off-axis location project a portion of the scattered light onto multiple detectors. Each detector converts the optical signal into a Doppler burst with a frequency proportional to the particle velocity. The phase shift between the Doppler signals from different detectors is proportional to the particle's diameter. PDPA is highly suited to measure the velocities and local structure of sprays. However, complications are known to occur when droplets are inhomogeneous, for instance, when they contain an internal structure, such as air inclusions, caused by, for example, surfactants, or emulsion droplets. The light passing

through the droplets will interfere internally and cause an erroneous calculation of the droplet diameter.[12,13]

Finally, with the laser diffraction technique, a laser beam hits the droplets, followed by reflection, diffraction, or absorption. The diffraction angle is inversely proportional to the size of the droplet, and so the light diffraction pattern allows us to obtain the droplet size distribution using Mie theory or Fraunhofer diffraction light theory, and assuming that the droplet has a spherical shape.[14,15] Laser diffraction is widely applied as it has a wide dynamic range, allows fast measurements, and is repeatable with a high degree of precision.

When discussing drop size distributions, it is important to distinguish spatial and temporal distributions. With spatial distributions, typically, a snapshot is made from the spray and all drops within the snapshot are processed. Spatial distributions are related to, for instance, taking (real) photographs or determining particle concentrations. Image analysis and laser diffraction are examples of techniques that yield spatial distributions. With temporal distributions, typically the flux of drops across an imaginary surface is observed during a certain time interval. In that case, not only the concentration of drops is important but also the velocity of the particles in the spray. PDPA is an example of a technique producing temporal distributions.[8,10]

A lot of research has been done to compare laser diffraction and PDPA.[16–20] However, all comparisons were done in high turbidity for small diameter sprays to a maximum of 10 μm, of which it is known that the multiple scattering affects the laser diffraction measurements.[21] Corcoran *et al.*[22] found that PDPA results agreed well with the laser diffraction results for water sprays but less so for propylene glycol sprays. They used a nebulizer to produce the droplets, resulting in a high turbidity, small diameter spray.

Less research has been done into droplet sizes from 10 μm up to 1000 μm. Tuck *et al.*[12] compared PDPA with droplet imaging using a particle measuring system for agricultural sprays with a droplet diameter between 10 μm and 900 μm and concluded that PDPA yields a larger proportion of small droplets and consistently lower values for the volume median diameter.

Herbst[23] compared the PDPA, laser diffraction, and the image analysis to classify agricultural nozzles to the international spray classification system created by the British Crop Protection Council[24] for nozzle sprays with a droplet diameter between 10 μm and 1000 μm. This comparison found that the three methods yielded the same classification of nozzles, although the cumulative droplet volume distributions were different between the methods. The volume mean diameter and droplet size spectra of the image analysis technique and laser diffraction agreed very well, while the PDPA was found to deviate. The differences became larger for coarser spectra.[13,23] Herbst mainly focused on the classification of the different nozzles and did not go into the details of the differences between the methods.

In this paper, we compare these three techniques for droplet sizes in the range from 10 μm up to 2000 μm and determine under which conditions and for which physical systems each technique can be safely applied. We specifically look into the implicit assumptions made by the software of commercial instruments and the impact thereof on the results.

II. EXPERIMENTS

A. Nozzles and droplet size ranges

To vary the droplet size, we apply multiple nozzles, a technique commonly used in agricultural sprays.[25] We use a reference nozzle at 3 bars,[26] a pre-orifice nozzle that reduces the number of droplets smaller than 100 μm with 50%, and two air-induced nozzles that reduce the number of droplets smaller than 100 μm with 75% and 90%.[27] This reduction in the number of droplets smaller than 100 μm results in a shift of the droplet size distribution to larger droplets.

The nozzles used for this comparative study allow us to vary the droplet sizes between 10 μm and 2000 μm, with the volume mean diameter between 140 μm and 1000 μm, as summarized in Table I.

B. Liquids

We investigate water droplets and droplets of a polymer surfactant based adjuvant that is commonly used to control the deposition of droplets on leaves.[29] This polymer surfactant solution increases the average droplet size in order to prevent spray drift. (The presence of surfactants causes a slight decrease in the mean diameter,[30,31] but the average droplet size will increase because the polymer suppresses small droplets.[32]) The polymer surfactant solution creates larger droplets that also contain air inclusions.

C. Measurement and analysis techniques

The droplet sizes are measured at a distance of 40 cm directly below the nozzle. It is known that the droplet sizes are bigger on the edge of the sheet,[25] but for this comparison, we focus on the middle of the sheet. For each technique and nozzle, we measure at least 60 000 droplets.

To investigate the performance of the three measurement techniques, we use four setups, as shown in Fig. 1.

1. Image analysis

First, we use the commercially available and widely used Oxford Laser VisiSizer P15. It uses a short, double light pulse to

TABLE I. Nozzles used to produce droplets of various sizes.

Nozzle	Pressure (bars)	Size classification	Approximate mean diameter range (μm)[28]	Type of nozzle
Teejet XR 11004 VS	3	Fine	150–250	Flat fan
Teejet DG 11004 VS	3	Medium	250–350	Pre-orifice
Agrotop Airmix 11003	2	Very coarse	450–550	Air-induced
Agrotop TDXL 11006	3	Ultra coarse	>550	Pre-orifice + Air-induced

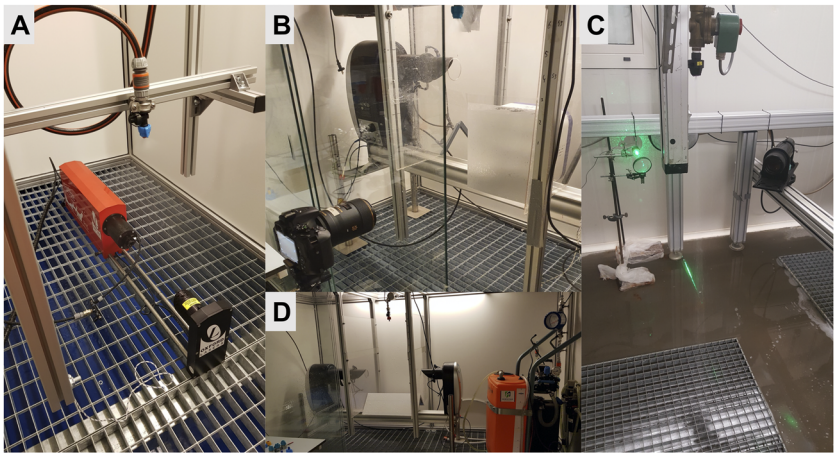

FIG. 1. Setups of the Image Analysis VisiSizer technique (a), stroboscopic imaging technique developed in-house (b), TSI® PDPA technique (c), and Malvern Spraytec® laser diffraction technique (d).

illuminate a screen that is photographed such that droplets show up as dark spots against a bright background (see Fig. 2). A digital camera is used to capture two snapshots of the particles. The Oxford VisiSizer software analyzes the images and uses image thresholds to identify the in-focus droplets in the image and determine their sizes and velocities. Depending on the droplet sizes, a 2× (10 μm –1000 μm), 1× (21 μm–2039 μm), or 0.56× (41 μm–3543 μm) magnification lens is used.

Second, we use a so-called stroboscopic imaging technique developed in-house, with a comparable working principle as the VisiSizer. It uses a Nikon D5600 digital camera with a Sigma 105 mm 1:2.8 DG Macro HSM lens and a shutter speed of 1/2.5 s, F13 and ISO400 in a dark room (see Fig. 2). It measures droplets in a range from 20 μm to 3000 μm. A time machine opens the shutter of the camera and simultaneously triggers a short light pulse of 0.5 μs with a Vela One flash light apparatus. This light pulse illuminates a diffuser that is situated behind the spray. The droplets from the spray are halfway between the diffuser and the camera. In-house developed software uses the sharpness of the droplet edges to automatically decide which droplets are in focus, taking into account that this effect depends on the droplet size. The data of the in-focus droplets of all photographs are translated into droplet size distributions.

2. PDPA

The PDPA instrument from TSI is equipped with front lenses with 1000 mm focus length on both transmitter and detector. A beam contractor is used in the transmitter to allow the measurement of large drops. The detector is set up in a forward refraction mode at an angle of 40°. The laser power in the measurement area is adjusted to 20 mW and checked before each measurement. The light signal from the detector passes on to a photomultiplier, supplied with a voltage of 520 V. In signal processing, a threshold of 70 mV is used.

3. Laser diffraction

For the laser diffraction technique, we use a Malvern Spraytec with a 750 mm lens that can detect droplets from 2 μm to 2000 μm. The Spraytec measures the droplet size distribution constantly for

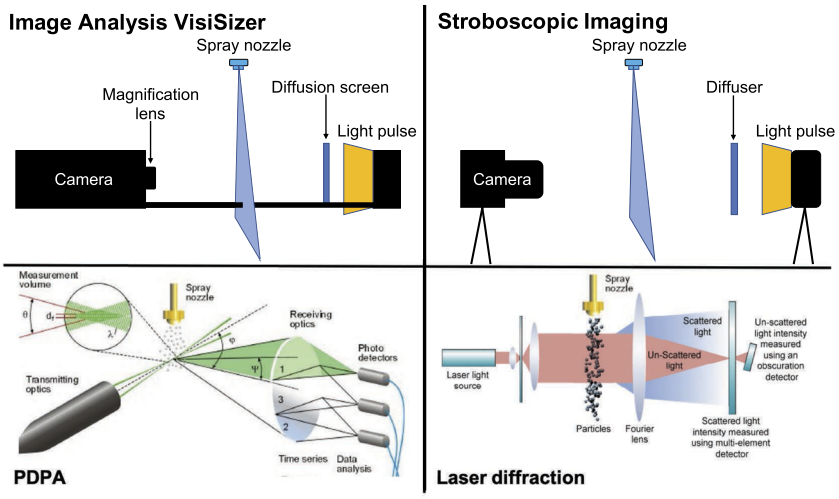

FIG. 2. Working principle of the four droplet size measurement techniques (lower panels courtesy of Dantec Dynamics and Malvern Instruments Ltd.).

TABLE II. Volume mean diameter of water droplets produced by different nozzles as determined by the four methods.

Size classification	Expected diameter range (μm)	Image Analysis VisiSizer	Measured volume mean diameter (μm)		
			Stroboscopic image	PDPA	Laser diffraction
Fine	150–250	200	188	200	170
Medium	250–350	284	306	282	231
Very coarse	450–550	502	471	432	375
Ultra coarse	>550	655	692	533	636

a given time and averages the results. The software of the Spraytec also employs the Lorentz–Mie theory to account for the contribution to the angular light energy distribution of scattering through small droplets.[21]

III. RESULTS AND DISCUSSION

Table II gives the volume mean diameter of water droplets as determined by all methods for all nozzle classifications. Figure 3 shows all droplet size distributions for water. The droplet size distributions are plotted as probability density distributions by volume, where a correction is made for the bin size. Most methods use an exponential bin size, meaning that the larger the droplets, the larger the bin size. By dividing the volume distribution by the bin size and normalizing this distribution, one obtains the probability density distribution of drop sizes. The data show that for the finest droplets, all methods are comparable. The most significant deviations are in the tail of the distribution, involving the largest droplets. These big droplets are also responsible for a large part of the volume fraction and therefore have a large effect on the measured volume

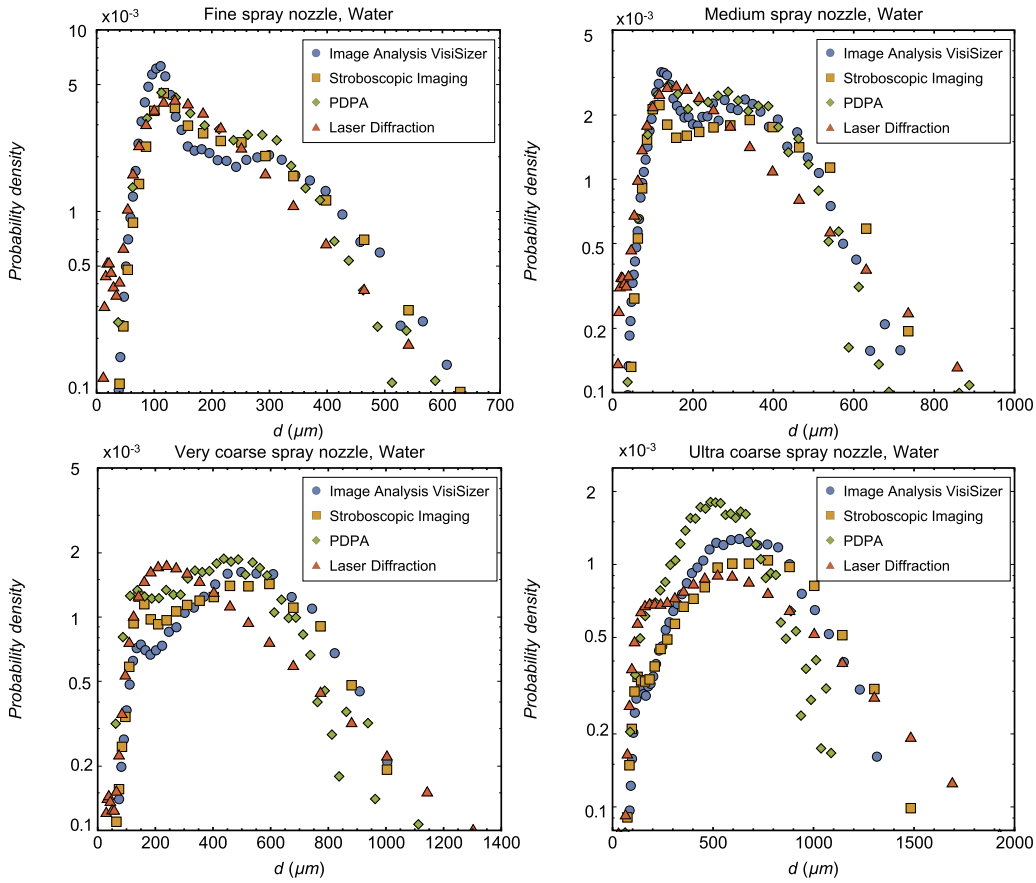

FIG. 3. Water droplet size distributions for sprays produced by four types of nozzles as determined by the four imaging/analysis methods. The Visisizer data correspond to the raw data.

mean diameter, which is the parameter most often extracted from this type of measurement.

A. Image analysis VisiSizer vs stroboscopic imaging

First, we compare the Image Analysis VisiSizer technique with the stroboscopic imaging method, as they are conceptually very similar. Figure 4 shows typical images from both techniques.

Figure 5 compares the probability density distributions as obtained from the two techniques, where we show two distributions for the VisiSizer technique: (1) the droplet size distribution as calculated by the software and (2) the raw data that include the size of each single droplet that has been measured. By manually moving every single droplet from the raw data into bin sizes, we calculated the droplet size distribution ourselves. It can be seen that the two outputs from the VisiSizer deviate considerably over the full range of droplet sizes. On the other hand, the distribution as obtained from the raw data of the VisiSizer is similar to the stroboscopic imaging data. This points to possible corrections by the VisiSizer software leading to skewed results. We will discuss this in the Appendix. In our main article, we will use the distribution from the raw VisiSizer data.

B. PDPA vs stroboscopic imaging

Figure 6(a) shows the distribution of droplet sizes in terms of cumulative volume percentages as determined by the stroboscopic imaging and PDPA techniques for sprays produced by the fine and ultra coarse nozzles. It can be seen that for the fine nozzle, with droplets up to around 500 μm, both methods yield a comparable droplet size distribution. For the ultra coarse spray nozzle, they deviate. One reason for this deviation might be that the working principle of PDPA is based on the assumption of spherical droplets. However, large drops (>1000 μm) may not be spherical but at best ellipsoidally shaped. This means that these drops could be misinterpreted, depending on which part of the drop the effective radius is measured. Thus, large drops may be interpreted as smaller ones, while other signals of large drops may be considered inconclusive by the processing software and hence discarded.

On average, this shifts the cumulative droplet volume percentage to smaller droplet sizes because droplets between 1000 μm and 1500 μm weigh heavily in the cumulative volume. This shift can also be observed in the probability density function of the same

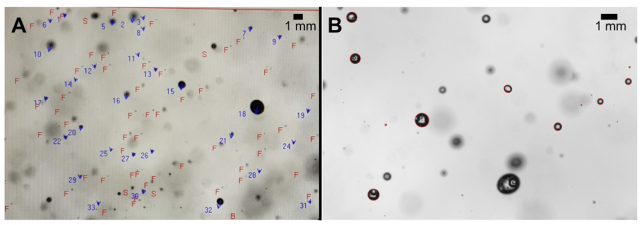

FIG. 4. Typical images produced by the Image Analysis VisiSizer technique (a) and stroboscopic imaging method (b). Scale bars: 1 mm. In the VisiSizer image, all droplets that are counted are labeled with blue numbers, while the out-of-focus droplets are labeled with a red F. For the stroboscopic imaging method, the edge of each in-focus droplet is traced by a red circle, and the droplet size is determined from the dimensions of this circle.

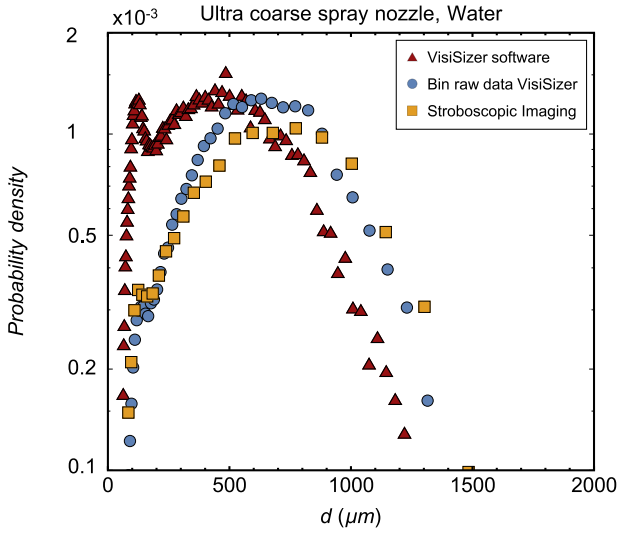

FIG. 5. Droplet size distribution (ultra coarse nozzle, water droplets) as determined by the stroboscopic imaging technique (yellow squares) compared to the results of the VisiSizer (red triangles) and those recalculated from the VisiSizer raw data (blue circles).

data shown in Fig. 6(b), where we fit the data with a two-parameter compound gamma function,[33]

$$\mathscr{P}_{m,n}\left(x=\frac{d}{\langle d\rangle}\right)=\frac{2(mn)^{\frac{m+n}{2}}x^{\frac{m+n}{2}-1}}{\Gamma(m)\Gamma(n)}\mathscr{K}_{m-n}\left(2\sqrt{mnx}\right), \quad (1)$$

with $\mathscr{K}$ being the modified Bessel function of the second kind and $\langle d\rangle$ being the mean droplet size. The parameter m sets the order of the ligament size distribution and n the ligament corrugation. This formula is known to describe the drop size distribution of agricultural sprays.[25,34] The two distributions can be fitted with the same values for m and n; the only parameter that changes between the two datasets is the mean droplet size $\langle d\rangle$ of the spray due to the filtering out of large droplets.

C. Laser diffraction vs stroboscopic imaging

Figure 7 shows that the droplet size distribution obtained from the laser diffraction method deviates for both small and large droplet sizes from the one obtained using the stroboscopic imaging technique. The small droplet population is overestimated for both laser diffraction and stroboscopic imaging, resulting in a peak at small droplet sizes (Fig. 7). This is because they both yield a spatial distribution and they measure continuously for a given time frame, meaning that small droplets traveling at a slow speed will appear in a higher concentration in the sample volume. This is further discussed in Sec. III E. The deviation for large droplets is due to the fitting method of the Spraytec, which assumes a certain shape of the droplet size distribution.[21] In fact, for the ultra coarse spray nozzle data in Fig. 7, analysis of the raw images reveals that there are no droplets greater than 1700 μm. This means that the laser diffraction software "creates" droplets of diameters larger than 1700 μm to meet the expected shape of the distribution. This correction of the

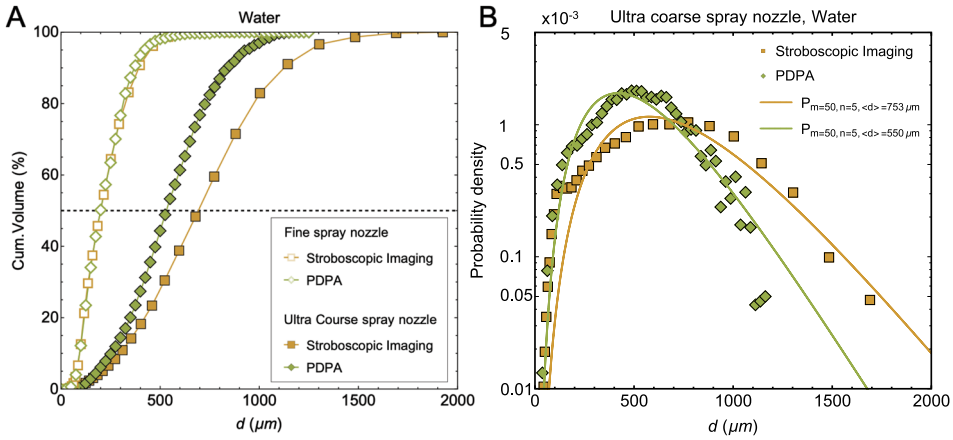

FIG. 6. (a) Cumulative volume percentage per droplet size as determined by the stroboscopic imaging technique (yellow symbols) and the PDPA technique (green symbols). For the fine nozzle (open markers), the cumulative results of the two methods are comparable, but for the ultra coarse spray nozzle (solid markers), they deviate. (b) Droplet size distribution (ultra coarse nozzle, water droplets) as determined by stroboscopic imaging (yellow squares) and PDPA (green diamonds). Solids lines are compound gamma functions [Eq. (1)] with parameters as shown. The fits only differ in their mean droplet size $\langle d \rangle$.

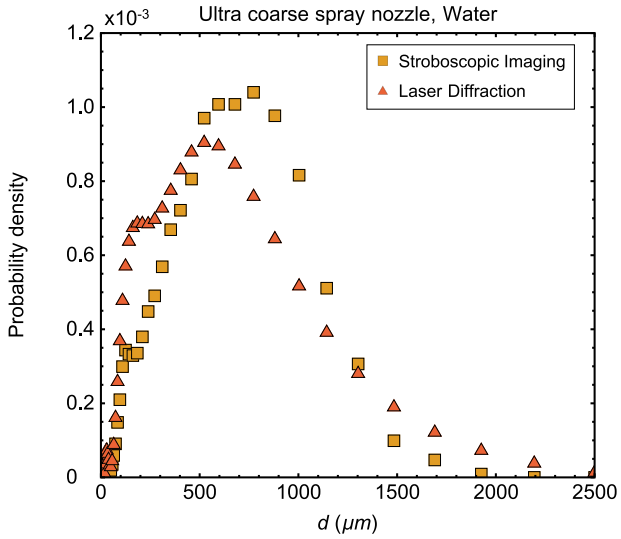

FIG. 7. Droplet size distribution (ultra coarse spray nozzle, water droplets) as determined by laser diffraction (red triangles) and stroboscopic imaging (yellow squares). The laser diffraction method yields different results for both small and large droplet sizes due to, respectively, the low speed of small droplets and the fitting method of the software.

raw data is shown in Fig. 8, which compares the raw scatter data with the fit-corrected data. The largest deviations are at the first five detectors (smallest diffraction angles), where the largest droplets are detected. Indeed, in Fig. 3, the laser diffraction data for all droplet size classifications appear more smooth than the results from the other methods.

D. Distorted droplet distributions

All results hitherto are from homogeneous water sprays. The effect of inhomogeneities is expected to specifically affect the PDPA method, as this technique requires drops to be transparent and homogeneous. To check the influence of non-homogeneous droplets, we also study sprays of polymer surfactant solutions. We use long-chain poly(ethylene oxide) (PEO) polymers to suppress small droplets, resulting in a shift of the size distribution, which is indeed reflected in the results obtained by stroboscopic imaging (Fig. 9). The PDPA technique does not yield the same shift when switching from water to polymer solution. In fact, the addition of surfactants causes a shift in the distribution to smaller sizes, and the emergence of an extra peak around $d = 150\mu$m. This is because the combination of surfactant and an air-induced nozzle causes air to become trapped inside the droplets, as can be visually confirmed (Fig. 10). For drops containing air bubbles, the path of the refracted laser beam is not clear and may lead to unexpected signals at the

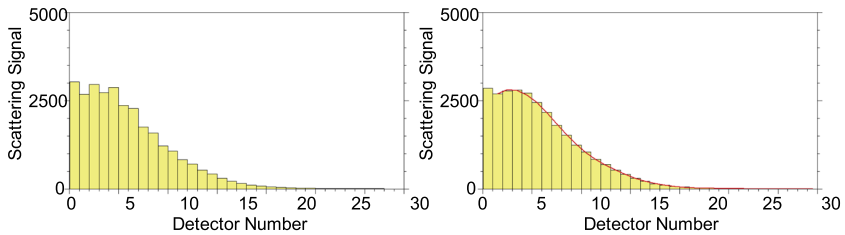

FIG. 8. Raw scattering data of the laser diffraction detectors (left) and corrected data (right), including the fit from the Spraytec software (red solid line). These scattering data correspond to the droplet size distribution of the laser diffraction in Fig. 7.

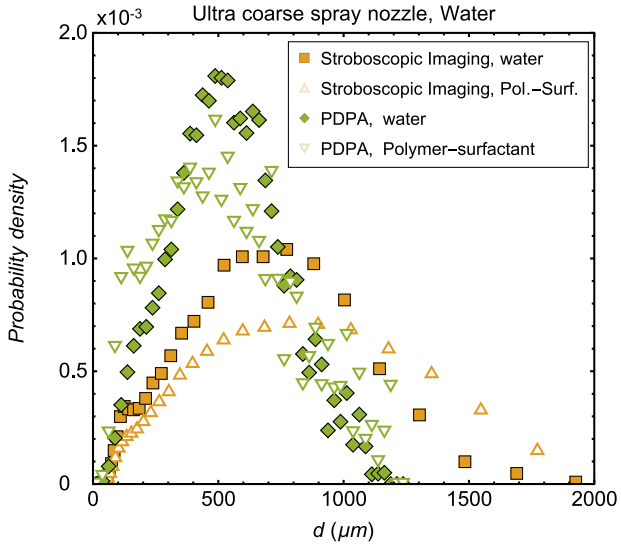

FIG. 9. Droplet size distributions (ultra coarse nozzle) for water (filled symbols) and polymer surfactant solutions (open symbols) as determined by stroboscopic imaging (yellow) and PDPA (green).

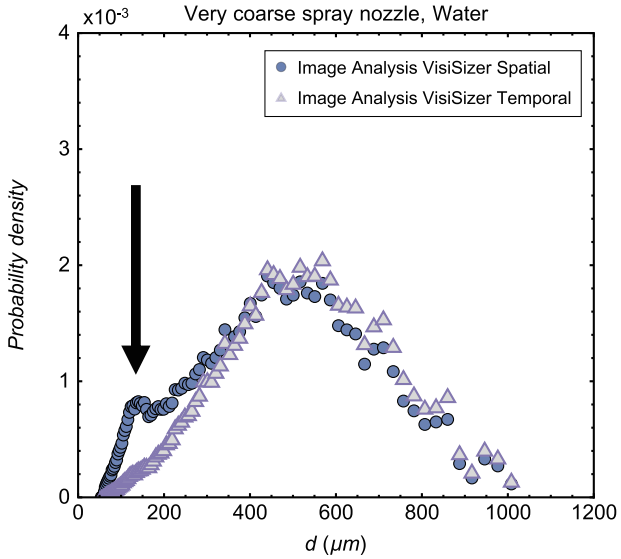

FIG. 11. Raw VisiSizer data (blue circles), representing the spatial distribution of droplets and temporal distribution (purple triangles) as calculated using Eqs. (2) and (3). The black arrow points to a peak in the raw data around 100 μm that disappears due to the low velocity of these small droplets.

detector. When the detected signal is linked to the curvature of the liquid–air surface, it can be misinterpreted as coming from a smaller droplet than the actual drop.

E. Difference between spatial and temporal distributions

The Spraytec and the two imaging techniques produce spatial drop size distributions, whereas the PDPA yields temporal drop size distributions. To move from one to the other, one needs to know the velocity of each drop. The problem is that not all spatial techniques provide the velocities of each drop. In principle, the velocity of a drop in a spray cone depends on the droplet size and its distance from the nozzle outlet. Drops of equal size have the same velocity, approximately. Then, the average velocity of each drop size class can be used for each drop in that class. In temporal distributions, slowly moving drops are less likely to reach the measurement area in the measurement time interval than high speed drops. Therefore, the droplet velocity is the weighting factor to turn spatial distributions into temporal distributions. In that case, the following equations can be used:[8,10]

$$N_{tmp}(d_i) = \frac{N_{spat}(d_i)v(d_i)}{\sum N_{spat}(d_i)v(d_i)} \quad (2)$$

and

$$V_{tmp}(d_i) = \frac{N_{tmp}(d_i)d_i^3}{\sum N_{tmp}(d_i)d_i^3}, \quad (3)$$

with N_{tmp} being the temporal droplet count, N_{spat} being the spatial droplet count, v being the average velocity of droplets of size d_i, V_{tmp} being the temporal volume percentage, and d_i being the diameter of the droplet. In Fig. 11, we convert the spatial distribution obtained by the VisiSizer technique into a temporal distribution using Eqs. (2) and (3). It can be seen that the spatial data of the Image Analysis VisiSizer contain more small particles when compared to the temporal data. This is as expected because small droplets have lower velocities so they are contained in the sample volume for a longer period. When converting the spatial data into the temporal data, the only difference is that the peak around 100 μm disappears. This also explains the peak we see for the laser diffraction technique.

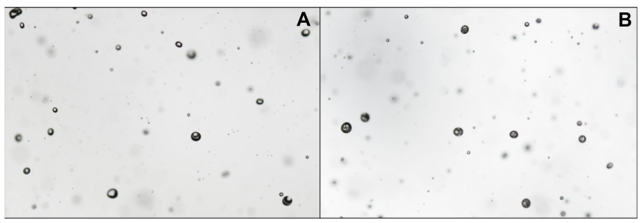

FIG. 10. Typical photographs of droplets of water (a) and polymer surfactant solution (b) produced by the ultra coarse, air-induced spray nozzle. In (b), more trapped air bubbles are visible.

IV. CONCLUSION

We compared three techniques to determine droplet size distributions in sprays: image analysis (VisiSizer and stroboscopic imaging), phase Doppler particle analysis (PDPA), and laser diffraction. We showed that the coarser the droplets, the bigger the deviations between the methods. The VisiSizer technique is based on the same image analysis principle as the stroboscopic imaging method. When using the raw data of the VisiSizer, the two techniques indeed match.

However, the distribution provided by the VisiSizer software is different, as will be discussed in the Appendix. For the PDPA technique, droplets need to be homogeneous, transparent, and spherical. Non-spherical drops may be interpreted as slightly smaller drops, thus resulting in a finer drop size spectrum. Inhomogeneous droplets due to the presence of air bubbles resulted in PDPA misinterpreting them as smaller droplets, shifting the distribution to smaller sizes. The disadvantage of the laser diffraction technique is that it uses a fitting method during measurements to obtain the droplet size distribution. Droplet size distributions that deviate from the expected shape are therefore misinterpreted. The laser diffraction technique also overestimates the contribution of small droplets due to the low velocity of these small droplets, with the result that a small droplet will be in higher concentration in the sample volume. For the image analysis methods, an extra peak is also visible at small droplets. However, when these spatial distributions are converted into temporal distributions, so when droplet velocity is included, this peak disappears.

Herbst[23] also found that deviations between measurement methods become larger for coarser nozzles. However, quantitatively, for the nozzles tested here, the results of the laser diffraction (medium and very coarse nozzle) and the PDPA (very and ultra coarse nozzle) do not fall into the same anti-drift classification. Since the work by Herbst, more and more attention has been paid to environmental pollution by drift of droplets smaller than 100 μm. To minimize this drift, increasingly coarser nozzles are used. The deviations between the techniques for coarser nozzles are larger, with the result that nozzles end up in different classifications. This requires a critical look at the techniques for measuring droplet sizes.

Overall, we conclude that for droplets up to ~400 μm, the image analysis and PDPA techniques agree very well. For these droplet sizes, one can choose the most appropriate method; the stroboscopic imaging system is easy to develop in-house without expensive equipment and the Image Analysis VisiSizer is easy to move, but one should be aware of the built-in software corrections. PDPA is an expensive and difficult to setup method, but robust when installed. When spraying coarser, non-spherical, or inhomogeneous droplets, PDPA is no longer reliable and the image analysis methods are more appropriate. The Spraytec is the most simple method to measure droplet sizes in a fast and easy way, but the results should be taken with some caution, as this technique may give results that are different from the other techniques depending on the experimental conditions.

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APPENDIX: VisiSizer CALCULATION

As stated in the main article, the VisiSizer provides two outputs. One output is from the VisiSizer software, which, among other things, provides the volume percentage of the droplets for each bin size. This volume percentage is used to calculate the probability density distribution. The other output is the raw data that include the size of each single droplet that has been measured. When the size of each drop is known, the droplet size distribution can be calculated and compared to the software-generated data, as done in Fig. 12.

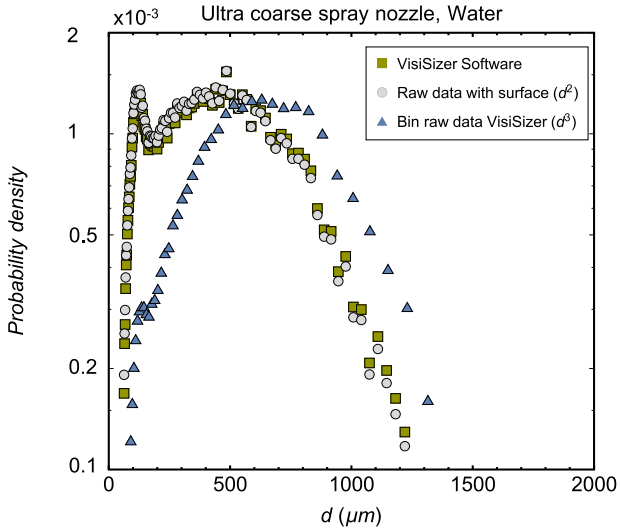

FIG. 12. Droplet size distributions (ultra coarse nozzle and water droplets) as obtained from the VisiSizer software (green squares) and calculated from the raw data using the surface (d^2) of the droplets (gray dots) and using the volume (d^3) of the droplets (blue triangles).

In order to explain the differences, it is important to note that the VisiSizer software assumes that there is a bias toward counting bigger droplets because the larger the droplet size, the more likely it is to be in focus. This has to do with the fact that only droplets within the depth of field are included. The heavier a drop is, the greater the chance that it will fall straight down and thus enter the depth of field. The VisiSizer makes the assumption that this effect is (roughly) proportional to d, the droplet size itself. To correct for this, the software divides the number of droplets by d. To go from droplet count to droplet volume, a multiplication by d^3 should be used. The result is mathematically the same as multiplying by the area, as $d^3/d = d^2$. Indeed, if we look at Fig. 12, we see that when we multiply the raw data by the surface, so d^2 (white circles), it matches the distribution that the VisiSizer software provides. However, the droplet size distribution should make use of the volume, d^3, and when we do this with the raw VisiSizer data, we see in Fig. 5 that this is consistent with the stroboscopic imaging method, which, in turn, corresponds well to the other methods.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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