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Review

Drop Size Measurement Techniques for Agricultural Sprays:A State-of-The-Art Review

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Abstract: Plant protection control based on the spray application of plant protection products is a very complex task depending on a series of factors, among which droplet size is the most influential for deposition and pesticide effectiveness. In fact, the adoption of the correct droplet size can ensure that the required dose reaches the target area and is not wasted, minimizes the off-target losses due to evaporation, drift and run-off and, at the same time, enhances the operator's safety in terms of inhalation, ingestion and dermal exposure. In this paper, after defining some mean characteristic diameters helpful for a description of a drop population and focusing on the main drop size distribution functions for the statistical characterization of sprays, a critical analysis of known methods, both intrusive and non-intrusive, for drop size measurement is carried out by reviewing the literature. Among intrusive methods, the liquid immersion method and the use of water-sensitive papers are discussed, whereas, among non-intrusive methods, laser-based systems (laser diffraction, phase Doppler particle analysis) and high-speed imaging (shadowgrapy) are presented. Both types of method, intrusive and non-intrusive, can be used in machine-learning-based approaches exploiting regression techniques and neural network analysis.

Keywords: droplet size distribution; image analysis; laser diffraction; PDPA; spray quality; spray characterization system

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1. Introduction

Despite the promotion of non-chemical methods of crop protection within the framework of integrated pest management, plant protection products (PPPs) are still used on a large scale. The environmental impact of the inappropriate use of the PPPs is a major public concern [1,2], so European Directive 2009/128/EC [3] recognizes their application in agriculture as an important issue to be properly managed to avoid undesirable effects on humans and the natural environment; in fact, in some cases, PPPs may not reach the target, causing serious economic losses and environmental risks [4]. The occupational exposure, environmental effects and efficacy of PPPs are affected by many factors, including the active substances and their formulations, the type of packaging, the task to be performed, the amount of pesticide to be handled, the duration of activity, the personal protective equipment (PPE) used, the type and quality of nozzles, the type of sprayers, their maintenance status and their operating parameters, the target structure (canopy, fruit, leaves, soil), the environmental conditions (temperature, relative humidity, wind speed) and the operator's expertise [5–7].

Within spray liquid applications, the drop size spectrum plays a key role in determining the spray behavior as it affects the biological efficacy of PPP treatment due to target coverage, the environmental pollution due to evaporation, drift and run-off and the operator's safety due to ingestion, inhalation and dermal exposure. Therefore, agricultural nozzles have a remarkable impact on the treatment efficiency, and the types that provide a more homogeneous droplet spectrum give a higher quality of application. This means that an optimal droplet spectrum is crucial for obtaining desired spray performance and reducing spray drift in order to ensure the deposition of the required dose to the target (leaf, fruit, etc.), minimize the off-target losses and reduce the operator's exposure. In agricultural PPP applications, small droplets, carried by air currents, can more easily reach the inner parts of the canopy, increasing coverage on leaves; but, on the other hand, if they become too small, they are more often subjected to wind drift and evaporation before reaching their target, especially in hot conditions. Large droplets, being heavier and having higher kinetic energy, are less prone to deflection by air currents and reach the external parts of the canopy more easily, but run-off becomes high, and the risk of soil contamination increases [8–10].

Drop size primarily depends on the atomization device and on the operating parameters. Agricultural nozzles for PPP application are mainly based on hydraulic, pneumatic and centrifugal atomization. In hydraulic nozzles, pulverization is accomplished by forcing the liquid under pressure through a small opening or orifice. Drop size primarily depends on orifice diameter and spraying pressure; increasing the nozzle openings results in larger drops (and vice versa), and increasing the spraying pressure results in smaller drops (and vice versa). Another aspect that affects the drop size is the liquid viscosity; high-viscosity solutions tend to produce larger drops, whereas low-viscosity solutions produce smaller drops. Pneumatic atomization is obtained when the liquid is exposed to a stream of air flowing at high velocity. For a given air flow rate, the higher the liquid flow rate, the higher the drop diameter (and vice versa). In rotary atomizers, the liquid is released near the center of a high-speed rotating disk; the liquid flows radially outward across the disk and is discharged at high velocity in the form of droplets from its periphery due to the centrifugal force. Drop diameter depends on disk rotation speed and liquid flow rate; for a given liquid flow rate, increasing the speed results in smaller drops (and vice versa), and, for a given rotating speed, increasing the liquid flow rate results in larger drops (and vice versa).

The effects of droplet size on treatment efficacy can also depend on the mechanism of action of the active substance. Pesticides with systemic action do not require complete and uniform coverage of the plant, and translocation in plant tissues may be ensured by larger droplets. They control pests when applied to one area of a plant or animal. In fact, a systemic herbicide, absorbed through a plant's roots or surfaces, moves throughout all parts of the plant and kills the entire plant. Conversely, drop size is of great importance for contact pesticides which control a pest because of direct contact with the plant's outside layer (epidermis). They require good coverage of the target, and pests are killed when enough of the leaf surface area is covered with a contact PPP, ensured by smaller droplets on the target. In the Czech research of Prokop and Veverka (2006) [11], the authors studied the influence of different droplet spectra on the efficiency of contact and systemic fungicides. They found that, with the same volume of sprayed mixture, smaller droplets allow coverage of a larger area, increasing efficacy with a higher droplet density per unit of leaf area (droplets/cm^2) with products containing a contact active substance.

On this basis, an efficient spray application in terms of the droplet spectrum plays an important role in increasing the benefits of PPPs and in reducing the risk of environmental and human contamination, as well as in producing high-quality and safe food in a more sustainable way [12–17].

All these aspects are typical of precision agriculture (PA), which has become an increasingly important topic. The importance that accurate and correct PPP spraying could have for a precise agronomic management of soil and plants is very high due to the use of the most modern technologies which meet the specific needs of individual crops. Precision spraying is an approach used within this context to control the amount of mixture distributed across the field only where needed. Many researchers across the world have contributed to smart technologies for precision spraying, developing innovative pesticide spraying systems (drone systems, robotic sprayer platforms) in order to enhance the soil productivity, reduce the wastage of pesticide and control dangerous effects on the environment [18–23]. Research involving the use of drone systems (unmanned aerial vehicle (UAV) models) was reviewed in the Indian study of Mogili and Deepak (2018) [19], where specific informations for crop monitoring and pesticide spraying were summarized. A UAV is an aircraft able to fly without a human pilot, and its integration with sprayer systems, multispectral cameras and sensors enables accurate, site-specific application in the field. A related Indian study for the development of pesticide-spraying robotic systems was conducted by Meshram et al. (2021) [20]. The review paper described the robotic system categories (platform mobility and steering, localization and navigation control, sensing and target detection and pesticide spraying arrangement). Baltazar et al., 2021 (Portugal) [21], developed a smart electric sprayer (precision robotic sprayer, PRySM) that can be assembled on a robot capable of operating autonomously on uneven ground and equipped with a crop perception system to calculate the leaf density. In the Italian study of Cantelli et al. (2019) [23], a system based on a reconfigurable vehicle with a high degree of automation for PPP application in environments where standard sprayers cannot operate (greenhouses and mountain areas) was presented.

Precision spraying in agriculture requires a proper choice and control of the spraying system, for which the correct drop size is fundamental. According to the International Standard ISO 25358:2018 [24], drop size measurement is of particular importance for pesticide spraying in agriculture, because it makes the description of the spray easier and, therefore, enhances biological efficacy and spray drift management. However, the adopted measurement systems may produce different values for a given droplet spectrum with variations in the analyzed parameters, especially due to sampling effects and dynamic size range capabilities [25,26].

Different methods to measure droplet size have been discussed in the literature, and several instruments exploiting different principles are available on the market. The droplet size spectrum can be measured with intrusive or non-intrusive methods [27]. Examples of non-intrusive systems are phase Doppler particle analysis (PDPA) [28], laser diffraction (LD) [29] and imaging principles, such as high-speed imaging (HSI), better known as shadowgraphy methods [30–33]. PDPA and HSI methods also allow the measurement of drop velocity at the same time [34]. Among intrusive techniques, based on digital image analysis, the most popular available in the literature include the use of water-sensitive papers (WSPs) [35] and the liquid immersion (LI) method [36]. In addition, machine learning (ML) methods have recently been used to classify sprays and evaluate droplet size and deposition [37,38].

All the aforesaid techniques are widespread for the evaluation of spray droplet size, and all have a strong influence on the results, which may be significantly different depending on the settings and type of measuring equipment. For this reason, the ISO 25358:2018 [24] standard recommends classification of spray droplet spectra by using reference sprays to define reference categories to increase uniformity in relative measures and classifications among different measurement systems and laboratories.

The main objective of this paper is to carry out a critical analysis of the most widespread methods for drop size measurement in the context of PPP application in agricultural systems by reviewing the literature. The paper is organized as follows:

1. The first section discusses the most important characteristic mean diameters and the main drop size distribution functions used for the description of sprays;
2. In the second section, non-intrusive measurement methods are presented, namely, laser diffraction, phase Doppler particle analysis and high-speed imaging;
3. The third section discusses two widespread intrusive methods, namely water-sensitive papers and liquid immersion;
4. The fourth section presents some methods for drop sizing based on machine learning approaches;

5. In the last section, error sources common to all the measurement methods are briefly discussed.

For each measuring technique, the main characteristics, peculiarities and limitations are highlighted, and some applicative examples are presented. Issues regarding the applications of the techniques and possible ways to overcome the problems are discussed.

2. Mean Characteristic Diameters and Statistical Characterization of Sprays

Considering the importance of droplet diameter (usually expressed in µm) in spray performance and drift minimization, a classification of sprays based on droplet size is used to classify nozzles in relation to spray efficacy and drift potential. Each nozzle, depending on its features, liquid properties and working pressure, produces a range of drop sizes which can be described by several mean characteristic diameter values. For this reason, it is necessary to know the technical characteristics of the spray nozzles to select those that produce droplets of the appropriate size under certain working pressure [39,40].

In light of this, the specific standard S572 was developed and approved by the ASABE (American Society of Agricultural and Biological Engineers) for the purpose to measure and interpret the spray quality in terms of volumetric diameters and to aid in the selection of the most appropriate spray size [41]. According to this standard, aligned with the ISO 25358:2018 standard [24], spray droplets are classified into eight quality categories in increasing order as follows: extremely fine (XF), very fine (VF), fine (F), medium (M), coarse (C), very coarse (VC), extremely coarse (XC) and ultra coarse (UC). The definition of the spray droplet size classes is based on a set of certified reference nozzles operated at given spray pressures while spraying clean water (free of particulates) (Table 1).

Table 1. Reference nozzles and pressure combination for class boundary definition (adapted from ISO 25358:2018 [24]).

Reference Nozzle	Pressure (MPa)	Flow Rate (mL/s)	Class Boundary
Mee Fog IP-16 impaction pin	0.550	8.1	XF/VF
TeeJet TP 11001-SS	0.450	8.2	VF/F
TeeJet TP 11003-SS	0.300	19.6	F/M
TeeJet TP 11006-SS	0.200	32.3	M/C
TeeJet TP 8008-SS	0.220	45.1	C/VC
TeeJet TP 6510-SS	0.120	42.2	VC/XC
TeeJet TP 6515-SS	0.100	56.8	XC/UC

Classification is based upon volumetric diameters. In general terms, the volumetric diameter $D_{v\alpha}$ is the diameter at which the α fraction of the total volume is carried by droplets with a diameter lower than $D_{v\alpha}$. The most used parameter to evaluate the droplet size is the volumetric diameter $D_{v0.5}$, also known as the "volume median diameter" (VMD), which divides the spray into two equal parts in terms of sprayed volume; 50% of the total volume is carried by droplets lower than the VMD and the other 50% by droplets larger than the VMD. The volume median diameter is used to define the spray quality of a nozzle in terms of its average droplet size in relation to the volume sprayed. According to the S572 ASABE standard [41], VMD values (µm) vary as follows: XF: <60; VF: 60–105; F: 106–235; M: 236–340; C: 341–403; VC: 404–502; XC: 503–665, UC: >665. These VMD ranges may vary widely based upon the type of measuring equipment used.

The information given by the VMD gives an indication of the risk of drift and of the nozzle to select for a particular application. As an example, very fine droplets are collected efficiently by flying insects, but they tend to remain in the air stream, which carries them around the stems and leaves of weeds. Fine and medium-sized droplets deposit efficiently on stems and narrow vertical leaves if applied when there is some air movement (fungicide in foliar protective or curative applications, insecticide in foliar contact and herbicide in foliar/post-emergent contact applications). Coarse or larger droplets deposit efficiently on large, flat surfaces such as the leaves of broad-leaved weeds or the soil (insecticide in foliar and soil-applied systemic applications, herbicide in foliar/post-emergent and soil-applied/pre-emergent systemic applications).

Two additional diameters that are commonly used are $D_{v0.1}$ and $D_{v0.9}$: 10% and 90%, respectively, of the total volume consisting of drops with diameters smaller or equal to these values [39,42]. The $D_{v0.1}$ diameter refers to the drift potential of individual drops and then it can be used for nozzle classification for drift reduction purposes. If the value of $D_{v0.1}$ is large, then the spray contains many droplets that are prone to drift. Conversely, the $D_{v0.9}$ diameter is best suited when complete evaporation of the spray is required. In addition, high values of $D_{v0.9}$ indicate that too much of the volume of spray may be taken up by a few large droplets. Spray coverage and efficiency may be reduced as there are not enough droplets to cover all treated surfaces, leading to an increased risk of reduced effectiveness due to poor coverage.

A dimensionless parameter indicative of the uniformity (width) of the drop size distribution is the relative span factor (RSF), defined by:

$$RSF = \frac{D_{v0.9} - D_{v0.1}}{D_{v0.5}} \tag{1}$$

The higher the RSF value, the wider the drop size distribution and vice versa. Then, the lower the RSF value, the lower the variation among drop sizes, resulting in less drift potential and greater coverage. The RSF provides a practical means for comparing various drop size distributions and should be used when possible.

Other mean diameters of special interest in agricultural spray application and their equations are listed below, with D_i representing the droplet diameter and N the total number of droplets.

$$D_{10} = \frac{\sum_{i=1}^{N} D_i}{N} \tag{2}$$

- D_{20}: surface mean diameter, best suited for surface controlling applications such as absorption. It is the diameter of a drop, the surface of which, multiplied by the total droplet number, equals the sum of all droplet surfaces:

$$D_{20} = \sqrt{\frac{\sum_{i=1}^{N} D_i^2}{N}} \tag{3}$$

- D_{30}: volume mean diameter, best suited for volume-controlling applications such as hydrology. It is the diameter of a drop, the volume of which, multiplied by the total droplet number, equals the sum of all droplet volumes:

$$D_{30} = \sqrt[3]{\frac{\sum_{i=1}^{N} D_i^3}{N}} \tag{4}$$

- D_{32}: Sauter mean diameter (also known as SMD), best suited to calculating the efficiency and mass transfer rates in chemical reactions, such as the fuel injection in combustors. It is the diameter of a drop with the same volume/surface area ratio as the total volume of all the drops to the total surface area of all the drops:

$$D_{32} = \frac{\sum_{i=1}^{N} D_i^3}{\sum_{i=1}^{N} D_i^2} \tag{5}$$

Considering the definition of the SMD, the same equation can be written in a simpler way as follows:

$$D_{32} = \frac{(D_{30})^3}{(D_{20})^2} \tag{6}$$

Finally, a diameter referring to the number of drops is the number median diameter (NMD or $D_{n0.5}$), i.e., the diameter at which 50% of the total number of droplets is smaller than this value. The NMD is usually smaller than the VMD because most PPP sprays often contain a large number of very small droplets. The VMD is influenced by relatively few large droplets, whereas the NMD is more affected by small droplets. So, the more uniform the size of the droplets, the closer to unity is the ratio of VMD and NMD.

All mean and characteristic diameters discussed above can be calculated by knowing the drop size distribution (DSD) function. According to Déchelette et al. (2011) [43], there are two categories of droplet distribution functions for describing droplet characteristics in different process, such as atomization or spraying: analytical and empirical functions. The atomization process consists of the disintegration of a liquid mixture into a multiplicity of small drops from a specific nozzle which has different shaped orifices and produces various spray patterns. A simple explanation of this process is that the potential energy (measured as liquid pressure for hydraulic nozzles or air and liquid pressure for pneumatic nozzles) of the spray liquid, along with the geometry of the nozzle, causes the liquid to emerge like small ligaments and then the breakup of these ligaments leads to the formation of further small drops [39,44]. Such a process is particularly important in agricultural "spraying" since it refers to an effective method where drops, resulting from atomization, are applied to the target by means of sprayers in order to obtain an adequate deposition, allow security against diseases and pests and increase productivity.

In the book of Lefebvre and McDonell (2017) [45], the authors provided a summary of the main probability distribution in each group (analytical and empirical). Theoretical atomization models for agricultural spray nozzles used for PPP applications are rare in the literature; conversely, according to the empirical approach, fitting theoretical curves to measured data is much more common. A wide variety of distribution functions have been proposed in the literature to describe agricultural sprays, such as the normal, log-normal, gamma, Nukiyama–Tanasawa and Rosin–Rammler distributions, able to fit experimental data with different levels of accuracy. The equations usually depend on two parameters, the first representing the mean diameter and the second indicating a measure of the droplet size range; in the empirical approach, their values are determined by fitting the function to the experimental data [42,46–48]. They have also been used to simulate the DSD as a function of the particle size to produce data on the droplet characteristics.

The normal distribution function is based on the random occurrence of a given drop size, and it is usually expressed in terms of a number distribution function $f_0(D)$ that gives the number of particles of a given diameter D:

$$f_0(D) = \frac{1}{\sqrt{2\pi\sigma}} e^{-\frac{(D-\overline{D})^2}{2\sigma^2}} \tag{7}$$

where σ is the standard deviation, σ^2 is the variance and $\overline{D}$ is the mean value. The normal distribution is considered when the range of drops is narrow, but it does not usually show a good fit to experimental results since natural distributions are rarely symmetrical.

If the logarithm of the particle diameter is used as the variable, the corresponding distribution function is the log normal:

$$f_0(D) = \frac{1}{\sqrt{2\pi D\sigma_g}} e^{-\frac{(\ln D - \ln \overline{D}_g)^2}{2\sigma_g^2}} \tag{8}$$

where $\overline{D}_g$ is the geometric mean drop size, and σ_g is the geometric standard deviation. The log-normal distribution is used because it provides a satisfactory correlation with experimental data, even if it is less popular than the normal distribution.

The probability density function of the gamma distribution has the general form:

$$f_0(D) = \frac{1}{\beta^\alpha \Gamma(\alpha)} D^{\alpha-1} e^{-D/\beta} \tag{9}$$

where α is the shape parameter, β is the scale parameter and $\Gamma(\alpha)$ is the gamma function. According to the French study of Villermaux et al. (2004) [49], the atomization process involves the fragmentation of ligaments into droplets, and the gamma probability distribution function reasonably fits experimental data distribution.

The Nukiyama–Tanasawa probability density function has the general form:

$$f_0(D) = aD^p e^{-(bD)^q} \tag{10}$$

This expression contains four independent constants, the simultaneous optimization of which is often difficult to apply and can lead to measurement errors. Most of the commonly used size distribution functions represent either simplifications or modifications of this function.

Finally, the Rosin–Rammler distribution, also known as the Weibull distribution, is one of the most well-known and -used empirical functions for droplet size distribution analysis. The general equation, expressed as cumulative volume, is the following:

$$F_3(D) = 1 - e^{-\left(\frac{D}{D_m}\right)^n} \tag{11}$$

where $F_3(D)$ is the fraction of the total volume contained in drops with a diameter of less than D, D_m is a representative diameter where 63.2% of the total liquid volume is in drops of smaller diameter and n is an adjustable parameter characteristic of the distribution that provides a measure of the spread of the drop. The higher the value of n, the more uniform the spray. The advantage of this expression is its simplicity, and it enables data to be extrapolated into the range of very fine drops, where measurements are typically more difficult. These features are the reason for its popularity [48,50].

Regardless of which drop size distribution function is used, all functions perform the same task. However, the discussion as to why a certain distribution function might fit better than another is still open for further study by researchers [45–51].

3. Measuring Techniques for Spray Characterization

Spray drop size measurement is a very complex task that has been faced in many ways over the years. In this sense, knowledge of the drop size distribution has for a long time been known as a fundamental factor for the research and development of new techniques relating to measuring drop size since a precise droplet size has a direct impact on biological efficacy and reduction of environmental contamination [45].

According to the study of Schick from Spraying Systems Co. (USA, 2008) [39], when discussing drop size distributions, it is preliminarily necessary to distinguish spatial and flux (or temporal) sampling methods. Typically, with spatial techniques, a collection of drops occupying a given volume is sampled instantaneously by using holographic means (for example, high-speed photography or light-scattering instruments). Drop size analyzers that implement a spatial technique include image analysis and laser diffraction. Conversely, with temporal techniques, the flux of drops is examined during an interval of time with the aid of optical instruments that are capable of detecting individual drop characteristics. Thus, not only the density of drops is important but also the velocity of the particles in the spray; in fact, it is used to obtain the number of droplets per cubic meter per second. Phase Doppler particle analysis (PDPA) is an example of a technique producing temporal distribution [39,52].

In more detail, Figure 1 shows a diagram of the various methods used to measure drop size and distributions in the agricultural sector, classified as non-intrusive and intrusive (or mechanical). The first category gathers methods based on light-scattering properties, which do not interfere with the droplet flow, while the second involves the collection of drops on slides (Petri dishes) or artificial collectors, such as water-sensitive papers. Some methods of both categories are based upon digital image analysis (DIA) principles. In addition, more recent approaches are based upon machine learning methods that exploit regression techniques and neural network analysis. All measuring methods, whether simple or sophisticated, are susceptible to various errors and ambiguities, some of which are debated below. Cited references have been systematized, as reported in Table 2, according to the measurement method treated.

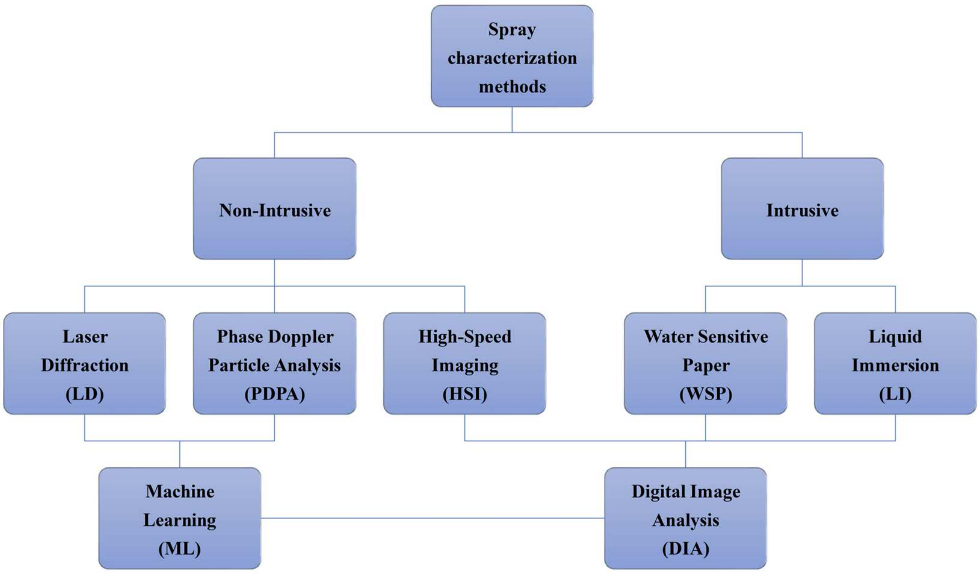

Figure 1. Scheme of the main spray characterization methods.

Table 2. Systematization of cited bibliographic references according to the drop size measurement method treated.

			DIA		
LD	**PDPA**	**HSI**	**WSP**	**LI**	**ML**
26, 27, 29, 39, 52, 53, 58, 59, 60, 61, 63, 89	26, 27, 28, 39, 52, 54, 55, 62, 63	26, 27, 30, 31, 32, 33, 39, 52, 56, 57, 64, 65, 66, 67, 68, 80, 89	27, 35, 68, 69, 70, 71, 72,73, 74, 75, 76, 77, 78, 79, 80, 88	27, 36, 67, 80, 81, 82, 83	37, 38, 84, 85, 86, 87, 88, 89, 90, 91
Total = 12	Total = 9	Total = 17	Total = 16	Total = 7	Total = 10

4. Non-Intrusive Measurement Methods

The development of modern technologies such as powerful computers and laser systems has led to the introduction of new techniques for obtaining non-intrusive spray characterization information. Over recent years, several laser-based techniques have been developed to determine droplet features, such as laser diffraction [53] and phase Doppler particle analysis [54,55]. The recent improvements in digital image processing have increased the interest in high-speed imaging methods for agricultural applications in general, specifically for PPP application [56,57].

In general terms, a potential feature of these methods is that they do not influence spray behavior during tests, allowing full analysis of the spray in a non-intrusive way.

4.1. Laser Diffraction (LD)

Laser diffraction devices have been largely adopted by the agricultural application community for spray droplet analysis [29,58]. The most widely used commercial laser diffraction instrument used today by researchers is the Malvern analyzer, which consists of a transmitter unit, a receiving system and a computer. The principle of diffraction by laser beam is applied; when the droplets intersect the laser beam, they scatter the direction of the laser rays, creating a diffraction pattern. The laser beam is crossed by the drops within the working zone of a lens, and a multi-element detector gathers the light diffracted by the spray droplets (Figure 2). Both the angle and intensity of scattering are strictly connected to the droplet size. The scattered light intensity is measured using a series of semi-circular photodiodes housed in the receiver unit. Later, a specific curve-fitting software is used to convert the light intensity into any of several empirical function, typically the Rosin–Rammler distribution function. In particular, the scattering angle is inversely proportional to the size of the droplet, and so it increases as droplet size decreases and vice versa [26,27,39,59].

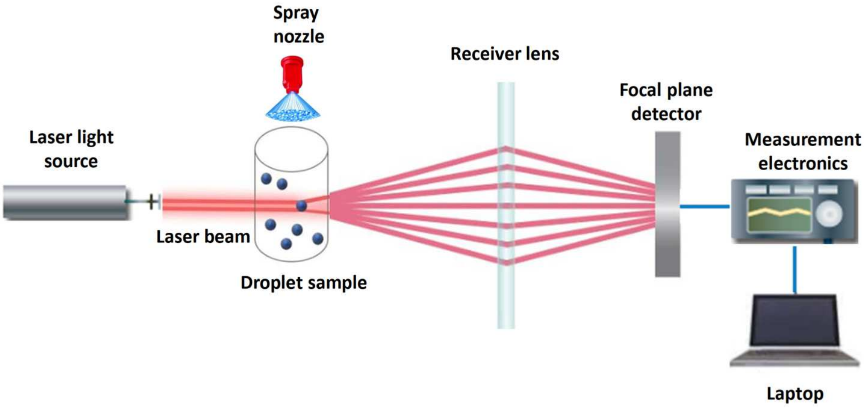

Figure 2. Scheme of the laser diffraction measurement system.

Other manufacturers of laser diffraction devices have emerged. As an example, a Sympatec analyzer was used in the study of US researchers Fritz et al. (2012) [60] for investigating a set of reference nozzles under three concurrent air flow velocities (0.7, 3.1 and 6.7 m/s). The system used a 623 nm He–Ne laser and was fitted with a lens capable of detecting particles in the range between 0.5 and 3500 µm.

The study of Fritz and Hoffmann (2016) [29] described the methods used in making spray droplet size measurements with laser diffraction equipment for both ground and aerial application scenarios. The authors configured the equipment in such a way as to have a dynamic size range of 18–3500 µm for ground nozzle testing and 9 to 1750 µm for aerial nozzle testing. The authors provided some guidelines that can be used to ensure inter- and intra-laboratory precision while minimizing the sampling bias associated with laser diffraction systems. A Sympatec Helios laser diffraction particle size analyzer was also used in a study by Dorr et al. (2013) [53] to compare the initial spray characteristics, including drop size, produced by several types of agricultural nozzles. Finally, in the Indian study of Sirmour and Verma (2019) [58], a Malvern Spraytec laser diffraction system was used to detect the drop size distribution of agricultural nozzles in the range of 1–2500 µm.

The measurement of the size distribution of spray droplets is commonly based on Lorenz–Mie theory or Fraunhofer diffraction light theory. They both assume that particles have a spherical shape. The Fraunhofer theory is based on the approximation that the laser beam is parallel, and the detector is at a distance that is very large compared with the size of the diffracting particle. The Lorenz–Mie principle can be understood by considering that the drops scatter the incident light, producing, due to the wave interference, a central forward scattering lobe flanked by a succession of lateral lobes of decreasing amplitude. In addition, it requires the proper choice of refractive index for ensuring the accuracy of the computed size distributions [61].

Even if they require a little knowledge of their basic principles for operation, laser diffraction instruments have been widely used by researchers for drop size measurements due to their ease of use, capacity to rapidly measure high-number density sprays with a high degree of precision and the large measurement range. Despite that, their use is not free from problems and can produce considerable errors in results, affecting the size measurements. The most significant limitation associated with this method is called multiple scattering; this effect occurs when the concentration of the drops in the spray is remarkably high, and, consequently, there is a dispersion of light before it reaches the detector since the light that is scattered by a drop may be scattered by another drop. This introduces errors in computing the drop size distribution and should be corrected empirically. Another limitation to consider concerns the correct alignment of the laser and the detector, which introduces significant complexity in droplet size distribution measurement. About this, experience has shown that a good alignment of the laser is crucial, and, to obtain accurate measurements, it should be perpendicular to the spray axis at a suitable distance from the spraying point [29,39,52]. Although many non-intrusive techniques have been developed for the measurement of drop size distribution, laser diffraction system and the Malvern instrument remain a reference for many researchers.

4.2. Phase Doppler Particle Analysis (PDPA)

PDPA is another commonly used method for droplet size measurement. It falls into the category of flux-sampling instruments and, together with the laser diffraction method, is considered a powerful tool for non-intrusive investigations of sprays. In comparison to laser diffraction, PDPA is a single-droplet technique in which the phase difference of scattered light at multiple angles is measured and inverted to obtain the size and velocity of individually detected drops [26]. In general terms, it is equipped with transmitter and receiver units, a signal processor and a computer. The principle of operation is based on the creation of an unbroken laser that is split through a beam splitter into two beams with the same wavelength; these laser beams intersect again at a point named "the probe volume", and the measurement point is defined by this intersection, producing a set of parallel equidistant interference fringes. When a droplet passes through the intersection zone of the two beams (sampling area), a diffused light modulated in space and time is produced. The spatial frequency is proportional to the size of the droplet, whereas the temporal one is proportional to its speed. Hence, the method measures droplet size one by one and also the speed of the droplets at the same time (Figure 3).

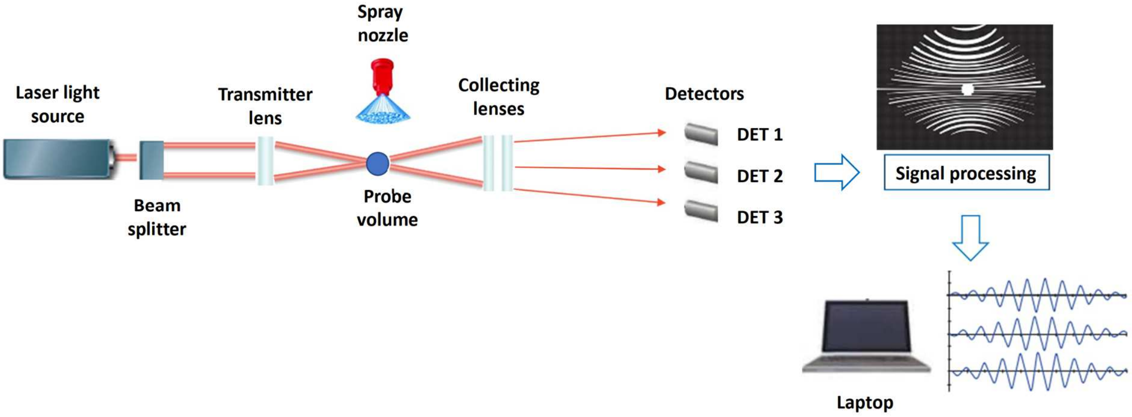

Figure 3. Scheme of PDPA measurement system.

The light diffusion angle is usually 30°, and three detectors collect the scattered light from the individual particles that pass through the sample volume, assessing their phase shifts. Ad hoc software, capable of providing real-time displays of droplets spectra, constantly evaluates and adjusts the sample volume to be analyzed, which is affected by the probability that larger droplets cross the edge, producing an inadequate signal. The range for these instruments can vary from 1 µm even to 8000 µm. However, the PDPA technique is strictly limited to homogeneous and spherical droplets since the measured signal depends on the radius of curvature of the droplet. Therefore, the presence of inhomogeneous droplets, due to air inclusions inside droplets, may be interpreted as smaller drops and can adversely affect the results [27,28,39,62].

In the US study of Bachalo and Houser (1984) [54], a theoretical analysis of the method was carried out, and experimental verification of the theory was obtained by using monodisperse droplet streams. Simulated spray environments and fuel spray nozzles were used in the evaluation of the method. The measurements of the monodispersed drops showed complete agreement with the theoretical predictions.

A particular comparison of drop sizing results obtained using an Aerometrics phase Doppler particle analyzer and a Malvern laser diffraction instrument was proposed in the study of US researchers Dodge et al. (1987) [63], where, after an appropriate conversion procedure from temporal to spatial sampling, they demonstrated that average drop sizes measured in different points of a sample showed similar trends during spraying with both systems, but those produced by the phase Doppler values were generally larger.

A detailed description of the PDPA measuring system was given in a more recent study by Nuyttens et al. (2006) [28]. The laser used in this study was an Aerometrics PDPA 1D system, which was used to test 32 nozzle–pressure combinations for a total of 288 measurements. The experiment was performed in a controlled climate room with defined dimensions and was provided with temperature and humidity control. The results showed the effect of nozzle type and flow rate on droplet size and velocity.

4.3. High-Speed Imaging (HSI)

Over the years, the rapid development of imaging techniques has made high-speed imaging even easier to use and a cheaper alternative to scatter- or diffraction-based measurement methods for the characterization of agricultural sprays. In general terms, high-speed imaging analyzers are spatial sampling techniques consisting of a light source (generally a strobe light), a high-speed camera and a computer with image processing software able to identify the in-focus droplets in the image and determine their sizes and velocities (Figure 4).

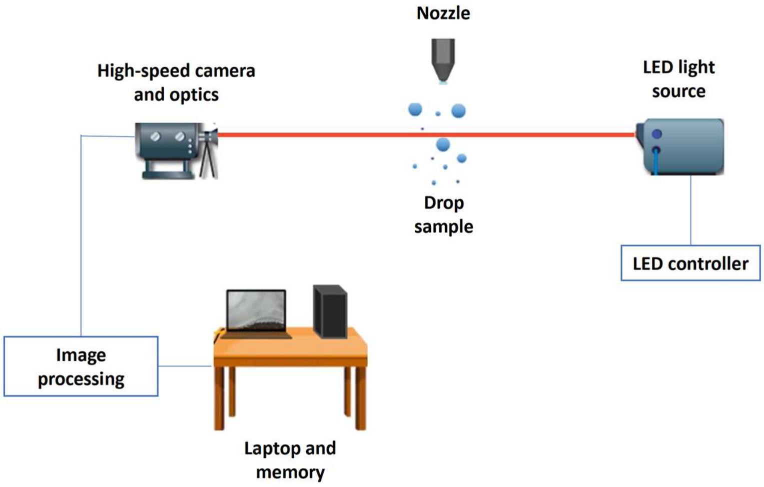

Figure 4. Scheme of HSI measurement system.

The illumination source spreads uniform light on the spray, and, consequently, part of the light is refracted by the spray, leaving a shadow on the bright background [33,56,57,64]. This methodology, usually performed in a back-lighted arrangement for image acquisition, is often known as the shadowgraph technique [31]. In detail, the system consists of a camera equipped with high-magnification optics used to acquire images of the drops and synchronized with a light-emitting diode (LED) light source for irradiating the spray. In addition, an LED controller provides intensity control of the light source. The droplets appear on the images as darker shadows on a brighter background, so, for each acquisition, two frames are recorded within a small time lapse. The droplet diameter and velocity are retrieved by using an advanced image analysis algorithm on each pair of frames. Then, the drop size distribution is obtained by gathering the data retrieved from all the images that are visible on the monitor of the computer.

In the work conducted by the Belgian researchers De Cock et al. (2016) [57], the drop size distribution of a set of reference nozzle/pressure combinations defined in the ISO

25358 [24] was measured by using a high-speed imaging method. The results proved how the method can be a real alternative to the laser-based technique, emphasizing the fact that the spray quality categories were well distinguished. An investigation conducted by the French researchers Castanet et al. (2013) [65], aimed at studying drop impact on heated surfaces, showed many advantages resulting from the combination of size and velocity measurements.

In absolute terms, the shadowgraphy method stands out among the other droplet sizing techniques as a powerful and versatile tool with a low cost. The main source of error in this technique relates to the limited depth of field (DOF); in fact, during the imaging processing, not all droplets appear sharp or in focus, making those that are unfocused larger than their real size. This leads to an overestimation of the droplet size distribution [66].

Many researchers have proposed some corrections to improve this limitation. For instance, the Franco-Belgian study of Minov et al. (2016) [33] applied shadowgraph imaging for measuring droplet size and velocity from different nozzle types used in spray applications. To minimize the error due to unfocused droplets, the authors introduced an in-focus parameter as a function of the gray level gradient at the droplet edge, droplet diameter and gray level intensities of the background and droplet. Consequently, an in-focus criterion based on the droplet diameter was established to correct the measurement of droplet diameter and reject unfocused droplets. The US researchers Kumar et al. (2019) [64] proposed a different approach based on digital inline holography (DIH), able to ensure high-resolution imaging of the sample over an extended depth of field typically several orders of magnitude larger than that of traditional imaging.

In an exploratory study by Sijs et al. (the Netherlands, 2021) [26], the authors compared three methods for measuring droplet size in the range from 10 µm up to 2000 µm, namely, image analysis (commercial VisiSizer and developed in-house stroboscopic imaging), a phase Doppler particle analyzer and a laser diffraction tool (Malvern Spraytec), using several nozzles and a surfactant-based adjuvant in such a way as to obtain fine, medium, very coarse and ultra-coarse sprays. They found that the larger the droplets, the bigger the differences between the results obtained by the different methods (Figure 5).

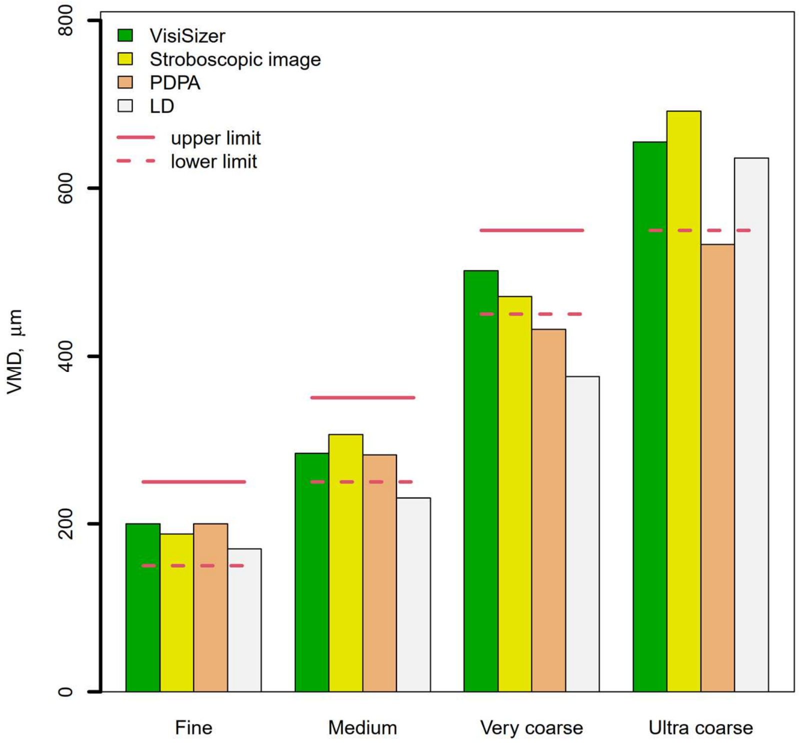

Figure 5. Comparison between non-intrusive measurement methods based on the research of Sijs et al. (2021) [26]. Upper and lower limits refer to the expected range for each spray quality.

In more detail, the commercial VisiSizer technique provided the same results as the in-house stroboscopic imaging when using the raw data (diameter of each single droplet) to calculate the probability density distribution, but differences were observed when the default software output was used, which suggests that there is a bias toward counting bigger droplets due to their greater probability of being in focus. The PDPA technique requires homogeneous, transparent and spherical droplets. Non-spherical and inhomogeneous droplets due to the presence of air bubbles may be interpreted as slightly smaller drops, thus, resulting in a finer drop size spectrum. The laser diffraction technique overestimates the contribution of small droplets due to their low velocity and then higher concentration in the sample volume, resulting in lower VMD measurements (Figure 5). In addition, since it uses a fitting method during measurements to obtain the drop size spectrum, distributions that deviate from the expected shape are misinterpreted.

Overall, the results emphasized how the limitations of each method can influence the droplet size measurements and also the need for selecting the size measurement method to fit the expected range of droplet parameters. In the Belgian study of Nuyttens et al. (2007) [8], the authors compared their PDPA measurements with results from 17 references for the British Crop Protection Council (BCPC) reference nozzles and deduced that absolute results vary considerably between different investigators depending on the settings and type of measuring tool.

5. Intrusive Measurement Methods

Various intrusive methods based on image analysis have been used in the literature for spray characterization in agricultural treatments in order to provide accurate information on droplet size. Among these, in this section, we focus our attention on the use of water-sensitive papers (WSPs) and the liquid immersion (LI) method, which appear to be the most useful. They usually involve the capture of a droplet sample on a solid surface or in a cell containing a special liquid. Then, the drops are photographed or observed with the aid of a camera or microscope. In general, these methods use a light source that is as diffused as possible to illuminate a collection of droplets, followed by software analysis of the images to determine all the droplet spray parameters. These methods, even if intrusive, have the virtues of simplicity and low cost, but the extraction and collection of representative droplet samples should be considered. Another aspect to be considered regards the resolution of the image, to be chosen according to the size of the particles to be extracted [67,68].

5.1. Water-Sensitive Papers (WSPs)

The use of WSPs is considered an important tool for the evaluation of agricultural spray parameters. They are semi-rigid papers of various sizes with one side covered with a yellow surface layer that changes its color to dark blue when in contact with water droplets, allowing the assessment of the droplet stains in spray tests. Being the most popular artificial targets used in agricultural spray applications, WSPs have been on the market for over 30 years. Due to their quick response in field, they are commonly used by farmers to evaluate the amount of covered surface (spray coverage) and the quality of deposition in terms of number of droplets per unit area (droplet density). They are normally placed at specific points for a given crop to check the spray coverage of a certain treatment. In addition, it is also recommended to place a large number of these targets to obtain a representative sample [31,69,70].

Calculation of droplet size by using WSPs is heavily affected by the spread factor, i.e., the ratio between the stain diameter and the droplet diameter, which varies with physical properties of the liquid such as surface tension and with the direction and energy of the impact on the cards. Moreover, with a high degree of coverage, a halo grows around the stains that increases the area processed during the image analysis.

Several studies have been carried out to determine the characteristic diameters of spray by analyzing drop stains on WSP through image analysis techniques using commercial or freeware software based on the image processing of scanned WSPs or an app for smartphones [71–74]. All these tools produce significant indicators related to the spray quality. However, as demonstrated by Cunha et al. [72], comparing the results obtained from seven software packages after the analysis of nine water-sensitive papers, there was a great difference among the results in terms of the calculated spray parameters. In the US study of Salyani et al. (2013) [35], the authors, operating in a citrus orchard, assessed spray distribution and deposition using WSPs and analyzed them with three different image analysis systems. They found a good correlation among the three imaging systems in measuring the amount of area coverage of WSPs but concluded that WSPs cannot be used to quantify the amount of spray deposit in most field applications.

Although it is possible to visually determine if a treatment has been either insufficient or correct by counting the stains using a lens, it is advisable to analyze the cards using an image analyzer to obtain reliable results and to accurately verify the effectiveness of the treatment. ImageJ software, an easily accessible package, is one of the most used during experimental tests because it can operate on any operating system, it is easy to use and it can perform a full set of imaging manipulations [75] (Figure 6).

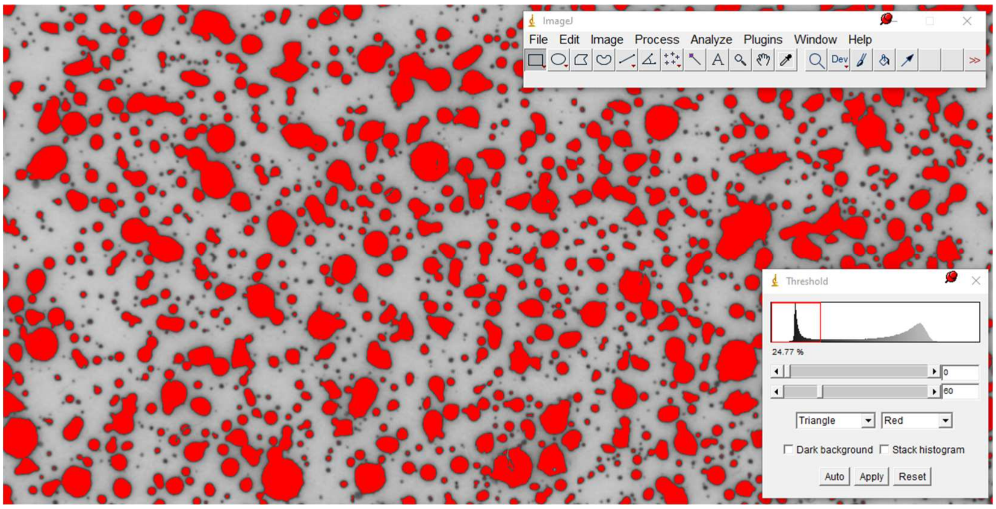

Figure 6. Example of a WSP image processed with ImageJ software. Overlap between stains must be properly managed during drop size measurement.

When WSPs are used, some negative aspects should be considered, such as the difficulty and slowness of analyzing them manually or in high-humidity environments (>85%), because, in this condition, the cards become unreadable, and there is a limitation in detecting drops with a diameter of less than 50 µm as they do not generally contain enough water to create a detectable stain. Furthermore, the droplet spread varies with the physical properties of the spray liquid, such as the surface tension and temperature, and kinematics of the impact, such as the angle and energy of impact. Finally, using WSPs to determine droplet size may not be as accurate as other techniques, e.g., laser diffraction, because larger droplets may coalesce after they impact on the cards, while small droplets may not be deposited. In general terms, WSPs show a quite good applicability for evaluations carried out in the field, representing a simple and low-cost tool for improving PPP application [76–79].

5.2. Liquid Immersion (LI)

The liquid immersion method is considered one of the most popular mechanical techniques to measure droplet sizes and their distributions. According to this method, droplets are captured within a glass plate (or slide), usually a Petri dish, covered with a suitable fresh mixture of lightly viscose liquids, for instance, Vaseline, light mineral oil or silicone oil, which, due to their hydrophobic nature, cause drops to form almost perfectly spherical shapes. After Petri dishes are exposed to the spray, they are immediately photographed in situ with high-resolution cameras or observed with a microscope for subsequent scanning, allowing drop counting and size measurement. This step is performed to obtain magnified photographs, and, due to use of a camera and microscope, the method does not require special equipment [80,81].

The method was applied by Eigel and More (1983) [81] to develop a direct technique for measuring raindrop size and distribution for both laboratory and field use. Another application is described by Hurlburt and Hanratty (2002) [82] to obtain measurements of drop diameter in a horizontal annular flow of air and water in a pipe. Drop samples were captured in a high-viscosity oil, and photographic images of the samples were used to measure the distribution of diameters. A more recent application of this method was described in an experimental study carried out in Italy by Longo et al. (2020) [83] to construct a low-cost test bench useful for evaluating agricultural spray nozzles with hydraulic atomization under effective work conditions. In this trial, the droplets were collected within three Petri dishes containing silicone oil of suitable density and viscosity, photographed with a digital single-lens reflex (DSLR) camera and then their images were analyzed with ImageJ software to calculate the usual spray parameters (Figure 7). The results showed good agreement with other measurement systems, even if some aspects still needed improvements.

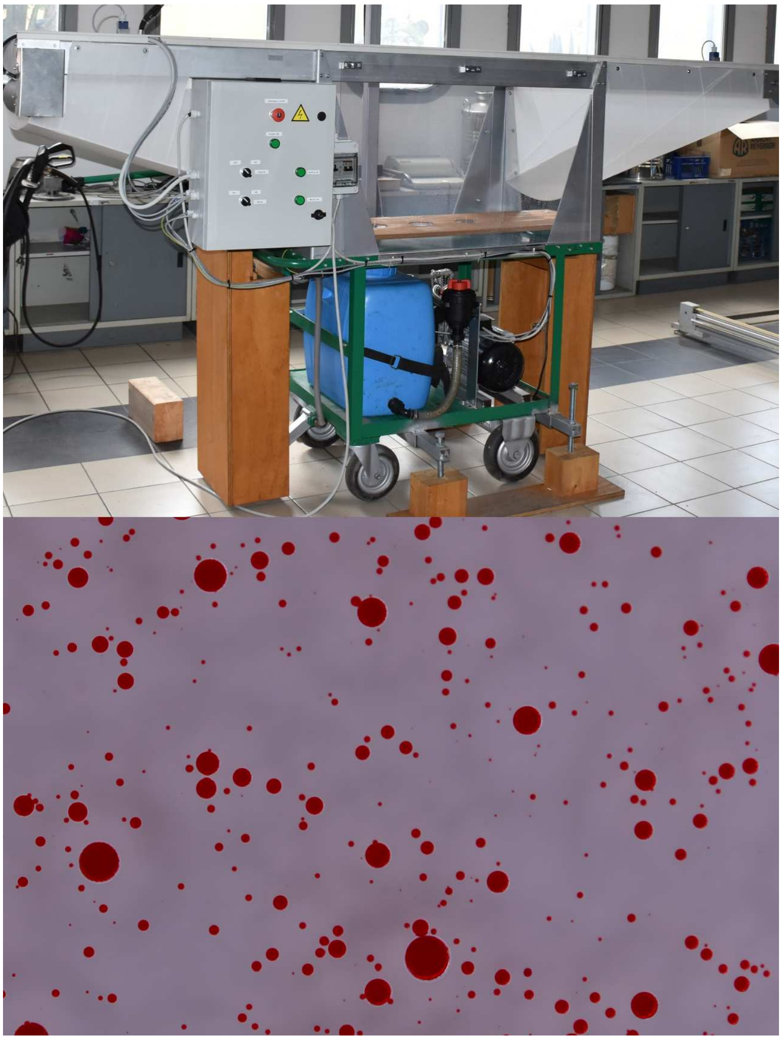

Figure 7. Test bench exploiting the liquid immersion method and example of drops (water solution with soluble red tracer) captured in Petri dishes containing silicone oil [83]. Red color simplifies drop feature extraction during image processing with ImageJ.

When using the liquid immersion method, it must be said that there are several associated problems. Among these, one common limitation regards the determination of what fraction of the liquid surface area should be covered by droplets. In fact, if too many drops are collected, the probability of error due to overlap is high, and, consequently, drop counting is difficult; alternatively, if too few drops are collected, the sample may not be representative of the spray. Other important considerations are related to drop evaporation and drop coalescence. The evaporation effects are very significant in the measurement of fine droplets because, being too small to break the surface tension of the oil, they evaporate. Instead, regarding drop coalescence phenomenon, a solution could be the use of an immersion liquid with low viscosity and surface tension. This remedy may be applied satisfactorily for the problem of droplet breakup since the risk of disintegration of the largest droplets is relevant to the impact with the liquid. In addition, the liquid immersion method, even if it falls within the category of intrusive methods, is efficient, easy to use and does not require complex or high-cost equipment; it is also adopted to confirm the adequacy of the data obtained by optical methods, such as phase Doppler particle analysis [80,81].

Based on the research of Cerruto et al. (2019) [76], the percentage of covered surface and stain density on WSPs were compared to the deposit and drop density on Petri dishes with silicone oil (Figure 8). Both targets (WSPs and Petri dishes) were simultaneously sprayed by means of an ATR 80 orange hollow cone nozzle (Albuz, France) at the pressures of 0.3, 0.5, 1.0 and 1.5 MPa. Images of WSPs were acquired using a scanner at 600 dpi resolution, and images of drops on Petri dishes were acquired using a DSLR camera at 6000 × 4000 pixels; both images were processed using ImageJ software [75].

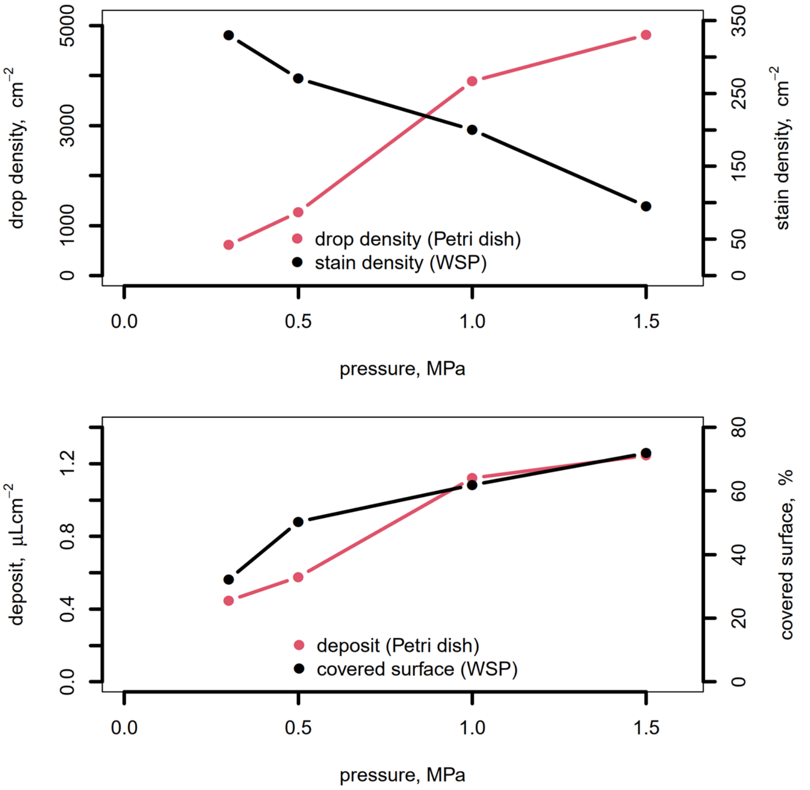

Figure 8. Comparison between Petri dishes and WSPs in terms of drop density, stain density, deposit and covered surface.

The results showed that the droplet density and unit deposit measured on Petri dishes increased when the pressures increased due to the increase in both the flow rate and in droplet pulverization. On the contrary, the analysis of WSPs showed that, when the pressure increased, the mean percentage of covered surface also increased, but mean stain density decreased due to the overlap of stains. The authors concluded that the spray qualities of the ATR 80 orange hollow cone nozzle at the four pressures measured by adopting the liquid immersion method were in accordance with nozzle producer information. In addition, the spray deposit measured on Petri dishes was found to be highly significantly correlated to the covered surface measured on the WSPs.

6. Machine Learning (ML)

In simple terms, the main task of machine learning is to develop learning algorithms that build models from data. Modern digital agriculture is characterized by great availability of data from a variety of sources (satellites, drones, cameras, sensors), and these data can be properly analyzed (artificial intelligence, deep learning, machine learning, Internet of Things) to build models able to guide decisions to optimize many agricultural practices. As an example, the study of Rehman et al. (2019) [84] surveyed the current applications of statistical machine learning techniques for agricultural machine vision systems able to ensure an increase in crop production by using automated, non-destructive and cost-effective techniques. The Indian study of Talaviya et al. (2020) [85] discussed the implementation of artificial intelligence in agriculture for optimizing some agricultural tasks, such as irrigation, application of pesticides and herbicides, crop monitoring and guidance of drones and robots. Another study by Australian researchers Chlingaryan et al. (2018) [86] carried out a review on ML approaches for crop yield prediction and nitrogen status estimation. The authors concluded that the rapid progress in sensing technologies and ML techniques provides cost-effective and comprehensive solutions for better crop yielding and decision making. A review of applications of artificial intelligence in agriculture was also reported in the Nigerian study of Eli-Chukwu (2019) [87]. The paper discussed the potentialities of artificial intelligence in soil, crop, disease and weed management.

Applications of ML methods to measurements and classifications of spray drop size are also common in the literature. The Iranian researchers Gargari et al. (2019) [88] developed a piezoelectric sensor for detecting the vibration signals from the impact of droplets onto the active surface of the sensor. Vibration signal analysis allowed the building of a model able to classify spray droplets based on their VMD, previously detected by using WSPs. Another Chinese study by Guo et al. (2020) [37] discussed different machine learning methods used to build mathematical models able to predict droplet size (VMD) and deposition in function of sampling location coordinates and for different types of nozzles. In the study of Pieloth et al. (2023) [89], convolutional neuronal networks (CNN) were used to categorize sprays by analyzing the spray cone images. The authors reported that deviation from laser diffraction was less than 1.5%. Multiple regression models were used in the study of Liao et al. (2019) [90] to predict several spray parameters (volumetric diameters, RSF, coefficient of variations) for different air-induction nozzles in the function of nozzle flow rate and spray pressure. Finally, a Chinese study by Dong et al. (2023) [91] on electrohydrodynamic atomization systems proposed the use of an artificial neural network model to efficiently and accurately correlate the relationship between the process variables (nozzle diameter, flow rate, liquid properties, distance between the nozzle and the grounding electrode, applied voltage) and droplet diameter, reaching determination coefficients near to unity.

All these recent techniques have been increasingly used in digital agriculture to address challenges in terms of productivity and sustainability. Their application in several sectors is expected to revolutionize many modern agricultural practices.

7. Final Considerations

Drop size measurement is a complex process that can be affected by various factors, and all measuring techniques are susceptible to various errors and ambiguities, the nature and importance of which depend on the particular method employed. A number of potential sources of error are common to almost all methods and include the sampling method, the sample size, the sampling location, the drop saturation, evaporation and coalescence and environmental conditions.

The sampling method (spatial or temporal) and sampling location affect results due to the different drop velocity. Pressure atomizers produce sprays with a high concentration of smaller drops near the atomizer because the smaller drops decelerate more quickly than larger ones. As a result, measuring the mean drop diameter using spatial sampling in this area produces smaller values than measurements taken using flux-based sampling. In sampling regions where all the drops are moving at the same velocity, the results obtained by both methods should be the same.

A spray from an agricultural nozzle contains a fraction of small drops much higher than that of large drops, but a few large drops predominate in determining the average volumetric diameter. Thus, if a sample of drops must be representative of the whole spray, the inclusion of large drops is vitally important. The ISO standard 5682-1 [92] recommends computation of the spray parameters on at least 2000 droplets, but, in the absence of a stated confidence interval on a particular mean value, the sample size can even be greater.

Drop saturation occurs when the drop flux exceeds the capability of the sizing instrument or method. The problem is most evident when drops are collected on WSPs or Petri dishes. If the sample is too large, the probability of error due to overlapping and/or coalescence of drops (or their stains) is high. The evaporation effect, depending on environmental conditions, is very important in the measurement of fine sprays, as the lifetime of small drops is extremely short. This leads to an increase or decrease in mean drop size in the function of the initial drop size distribution.

While instrument designers strive to minimize these errors, it is important to recognize that all measuring techniques come with some level of uncertainty. To accurately quantify the size of drops, it is necessary to consider these potential sources of error and their impact on the results.

In general, by controlling the VMD, farmers can reduce the amount of spray that is wasted or deposited in unintended areas, minimizing harm to the environment and surrounding crops. In addition, precision spraying with an optimized VMD can help farmers to reduce the amount of chemical they need to apply, leading to cost savings. Overall, the use of the VMD in precision spraying helps farmers to achieve more effective and efficient application of their sprays while reducing waste and potential harm to the environment.

New approaches based on artificial intelligence and machine learning methods may have great impact on precision spraying. Applications of artificial intelligence in agriculture for irrigation, weeding, crop spraying and monitoring with the help of sensors and other means embedded in robots and drones allow the saving of water, pesticides and herbicides, maintenance of soil fertility and improved productivity and quality.

8. Conclusions

This review paper illustrated several techniques used for agricultural spray measurements, highlighting the importance of droplet size in PPP application. The droplet size spectrum has to be as uniform as possible to reach the target, avoiding very coarse or very fine droplets. Overall, in light of the recent introduction of precision agriculture principles, it helps to have a better deposit on the target to reduce both environmental contamination and population risks (risks to farmers, consumers, bystanders). Measurement techniques were conveniently grouped into two main categories, namely, non-intrusive and intrusive systems. Some methods of both categories are based on digital image processing and on sophisticated machine learning techniques (multiple regression, neural network). Laser systems provide fast measurements but are unable to give reliable droplet size and distribution values. On the other hand, image analysis techniques can give accurate measurements of droplet size, but more time is needed to analyze droplets from a lot of images. Machine learning methods have proven to be very reliable in providing deeper insights and precise predictions but require large amount of complex data during the training stages, with which it is impossible to cope with all the problems arising from agricultural cropping systems.

It has been shown in the literature that each measuring technique produces different results in terms of range of droplet sizes, depending primarily on type of nozzle and its orifice size, spraying pressure, mixture properties and, not least, the technique itself. All these factors should be considered when the atomizing capabilities of a nozzle are studied, and the measuring technique should be appropriate for the expected range of drop size and velocities.

Although there are various techniques able to measure droplet size distributions, information on their accuracy and on how they compare with each other is limited yet and has to be deepened. Hence, further studies and measuring campaigns should be aimed at comparing different working conditions while using the same technique, thus, providing relative results. Absolute results are affected by the limiting factors specific to each technique, whereas relative sizes with respect to the reference nozzles always allow correct classifications. Finally, the potentialities of the latest available technological solutions (machine learning, artificial intelligence) should be better exploited and included in the precision agriculture principles to intensify agricultural production while limiting input requirements.

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