



Article

Characterization of the Droplet Population Generated by Centrifugal Atomization Nozzles of UAV Sprayers

Fábio Henrique Rojo Baio ¹, Job Teixeira de Oliveira ^{1,*}, Marcos Eduardo Miranda Alves ¹,
Larissa Pereira Ribeiro Teodoro ¹, Fernando França da Cunha ² and Paulo Eduardo Teodoro ¹

¹ Chapadão do Sul Campus, Federal University of Mato Grosso do Sul (UFMS), Chapadão do Sul 79560-000, MS, Brazil; fabio.baio@ufms.br (F.H.R.B.); marcos.eduardo@ufms.br (M.E.M.A.); larissa_ribeiro@ufms.br (L.P.R.T.); paulo.teodoro@ufms.br (P.E.T.)

² Viçosa Campus, Federal University of Viçosa (UFV), Viçosa 36570-900, MG, Brazil; fernando.cunha@ufv.br

* Correspondence: job.oliveira@ufms.br; Tel.: +55-31971230145

Abstract: The use of unmanned aerial spraying systems is currently being explored and applied worldwide. The objective of this study was to characterize the droplet population generated by hydraulic nozzles and centrifugal atomization nozzles used in sprayers mounted on remotely piloted aircraft (RPA). Two spray nozzle technologies were tested using a Malvern SprayTech laser particle size meter. The hydraulic nozzle evaluated was model 11001, which generates a wide-use fan spray. The centrifugal atomization nozzle, used in RPA sprayers, was manufactured by Yuenhoang, model DC12V. The experimental design was implemented in a completely randomized scheme, containing variations in the nozzles (hydraulic nozzle and centrifugal atomization nozzle) and application rate (AR) (5, 10, and 15 L ha⁻¹ in the test with the hydraulic nozzle; and 9.2, 12.8, and 15.6 L ha⁻¹ in the test with the centrifugal nozzle), with five replicates per treatment. The hydraulic nozzle test data showed a coefficient of variation of 6.8% VMD for all treatments, with droplet sizes within the fine classification ranging from 132.8 to 163.2 μm. It is noteworthy that the average relative span (span) of the droplet population generated by the hydraulic nozzle was 1.2, i.e., 20% higher than the desired reference value of 1. This value exceeds the general average reported for the centrifugal atomization nozzle, which has a span of 1.1. The relative span of the droplet size distribution for the hydraulic nozzles is greater than that observed with the centrifugal atomization nozzles. Excluding the extreme rotational speeds of the centrifugal atomization nozzle, the percentage of droplets generated with a volume smaller than 100 μm is lower compared to those produced by the hydraulic nozzle.

Keywords: application technology; spraying drones; application quality; pressurization; volumetric median diameter



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1. Introduction

Remotely piloted aircrafts (RPAs), popularly known as drones, or even as UAVs (unmanned aerial vehicles), are being widely used for various agricultural applications, mainly in collecting data through remote sensing, to monitor the vegetative vigor of plants, pest, or disease attacks, and in mapping weeds [1,2]. More recently, this equipment is being used for aerial spraying [3]. These machines are being adapted and still lack the technological development necessary to achieve the quality standards required for pesticide application, according to the principles of application technology [1]. The use of RPA in aerial spraying of small areas involves simple hydraulic circuits, with most available models

configured with conventional hydraulic nozzles. However, there is limited knowledge regarding proper droplet deposition when using this technique [4].

RPAs used in aerial spraying are primarily applied in rice crops [5] and have been less studied in the spraying of other crops [6]. RPAs are becoming popular in three basic structural aircraft designs: fixed-wing, commonly known as airplane, mobile wing, including helicopters, and multi-rotors [1]. Fixed-wing aircrafts offer greater flight autonomy, enabling applications in larger fields compared to other types. However, this type of aircraft requires a suitable take-off and landing strip, whereas multi-rotors can take-off vertically and can operate in any environment [7]. RPAs have a limited payload capacity compared to conventional agricultural aircraft. Therefore, they are used in situations with application rates below 25 L ha^{-1} in annual crops [4]. Their flight time is determined by the size of the energy source, which in most cases is an electric battery. As a result, the application rates used in RPA sprayers are lower than those in conventional aerial spraying [5].

The droplet population generated in agricultural spraying can be produced using various technologies, with the most notable being hydraulic nozzles, centrifugal atomization, thermal atomization, and gas atomization [3,8]. The most widely utilized technology in agricultural sprayers is hydraulic atomization, where droplets are generated via the pressurized flow of liquid through an orifice at the hydraulic nozzle outlet [9]. Technically, this process is relatively simple, and hydraulic nozzles have a lower acquisition cost. However, the process tends to produce droplets with a wide range of diameters, resulting in a droplet size distribution similar to a normal distribution, with droplets both significantly smaller and larger than the median size. The variation in droplet size generated by hydraulic nozzles is directly influenced by the application pressure within the hydraulic system, as well as the specific nozzle model employed. Different nozzle designs result in distinct droplet size distributions and varying degrees of homogeneity [10].

In contrast to hydraulic atomization, centrifugal atomization nozzles, commonly used in conventional aerial spraying, are being increasingly adapted for use in remotely piloted aircraft (RPA) sprayers [3]. Initially, remotely piloted aircrafts (RPAs) used for spraying were equipped with hydraulic nozzles; however, centrifugal spray nozzles have recently been introduced to the market [7]. In this system, a low flow rate of the spray solution is delivered hydraulically through a fine-mesh sieve or a conical, ridged disk rotating at high angular speeds, breaking the hydraulic flow into small droplets [3,8]. The variation in droplet size is mainly associated with the rotational speed of the device and the hydraulic flow rate. In contrast to hydraulic nozzles, this system offers the significant advantage of producing droplets with a smaller relative span, indicating reduced variability in droplet sizes compared to the median. The study of the droplet population is primarily based on determining the volumetric median diameter (VMD) of the spray, the relative span of the droplet size distribution, and the percentage of droplets with diameters less than $100 \mu\text{m}$. The relative span, also referred to as the span index, reports the homogeneity of the droplet population. A lower span index indicates a more homogeneous droplet size distribution, with greater consistency in droplet sizes throughout the spray spectrum. [10]. Droplet size is essential for accurate spray solution deposition. It is noteworthy that hydraulic nozzles exhibit lower uniformity in droplet size distribution compared to rotary nozzles [8]. The size of the droplets generated during spraying is directly correlated with both coverage and deposition on the target, which in turn significantly affects the efficacy of the product [11]. Droplet size also interferes with the potential for drift losses. In theory, fine and very fine droplet classes enhance target coverage but pose a higher risk of drift. Very fine droplets are highly susceptible to wind transport, particularly because a significant portion of the spray volume consists of droplets smaller than $100 \mu\text{m}$. Conversely, extremely coarse

droplets, while less prone to drift, are more likely to result in leaf runoff and provide reduced coverage of the treated area.

It is extremely important to elucidate the characteristics of the droplet populations produced by centrifugal atomization nozzles that have recently been incorporated into RPA sprayers. Recent research utilizing this technology has primarily focused on examining parameters such as flight height, effective spray width (or application swath), and the flight speed of the RPA. However, there has been limited investigation into the quality of the droplet population generated by these nozzles, particularly given their recent development. Chen et al. [12] reviewed the subject, highlighting the characteristics of unmanned aerial spraying systems, spray drift, and factors affecting drift, as well as additional research that still needs to be developed.

When UAV sprayers were developed, manufacturers initially used common hydraulic boom sprayer nozzles. But is this really the best choice? The objective of this study was to characterize the droplet populations produced by hydraulic nozzles and centrifugal atomization nozzles employed in sprayers integrated in remotely piloted aircrafts (RPAs).

2. Materials and Methods

2.1. Study Condition

The experiment was conducted in the Application Technology Laboratory at the Federal University of Mato Grosso do Sul (UFMS), Chapadão do Sul, Brazil, and consisted of two distinct test sessions. The first test was conducted using the most widely utilized hydraulic nozzle in RPA spraying, while the second test employed a centrifugal atomization nozzle. The data from both tests were analyzed separately because, although both nozzles are designed to generate sprayed droplets, they operate through distinct physical principles [3]. This difference occurs either due to the passage of the spray solution under pressure through a restrictor orifice at the nozzle outlet, which causes the spray solution to accelerate and break into droplets due to the generated hydraulic film (hydraulic nozzle), or by the atomization of the hydraulic film facilitated by the centrifugal energy produced by a rotating fluted disk (centrifugal atomization nozzle).

2.2. Experimental Design

The experimental design was structured as a completely randomized scheme, containing variations in the nozzle types (hydraulic nozzle and centrifugal atomization nozzle) and application rates (AR). The application rates tested were 5, 10, and 15 L ha⁻¹ for the hydraulic nozzle and 9.2, 12.8, and 15.6 L ha⁻¹ for the centrifugal nozzle, with five replicates per treatment. A single model of hydraulic nozzle cannot achieve the same flow range as the centrifugal nozzle, which accounts for the differences in the evaluated application rates. A standardized speed of 20 km h⁻¹ (5.56 m s⁻¹) was employed to calculate the spray application rate. The variation in flow rate, and consequently the application rate, was achieved by adjusting the spray pressure. Additionally, the rotation of the centrifugal atomization nozzle was varied to evaluate the characteristics of the resulting droplet population. The eleven evaluated rotational speeds were 2000, 3000, 4000, 5000, 6000, 7000, 8000, 9000, 10,000, 11,000, and 12,000 rpm (rotations per minute). The greater the flight speed and the higher the flight altitude, the farther the diffusion distance of the wake vortex [12]. When the drone flew at excessive speeds (more than 20 km/h), the direction of the downwash airstream produced by the rotor changed from vertically downward to obliquely downward due to the relative external wind, which weakened the pressure effect on the sprayed droplets.

Throughout the experimental period, the laboratory temperature was continuously monitored and kept constant at 24 °C to ensure optimal conditions for evaluations, specifi-

cally targeting moments when the relative humidity of the air exceeded 70%. The variations in the application rate were achieved by adjusting the flow rate at the nozzle tip. The evaluated nozzle model produced fine droplet sizes, which are recommended for low spray volume conditions [11], and was consistent with aerial applications conducted by conventional agricultural aircraft, with the volumetric median diameter (VMD) averaging approximately 150 μm [13].

2.3. Sprayer Nozzles Evaluated and Experimental Development

The hydraulic nozzle utilized in the evaluation was the Teejet model XR 11001, while the centrifugal atomization nozzle, model DC12V, was manufactured by Yuenhoang (Figure 1). Spraying at application rates below 20 L ha^{-1} is not suitable for the use of medium or larger droplets as it compromises the overall coverage target in the treated area [11].

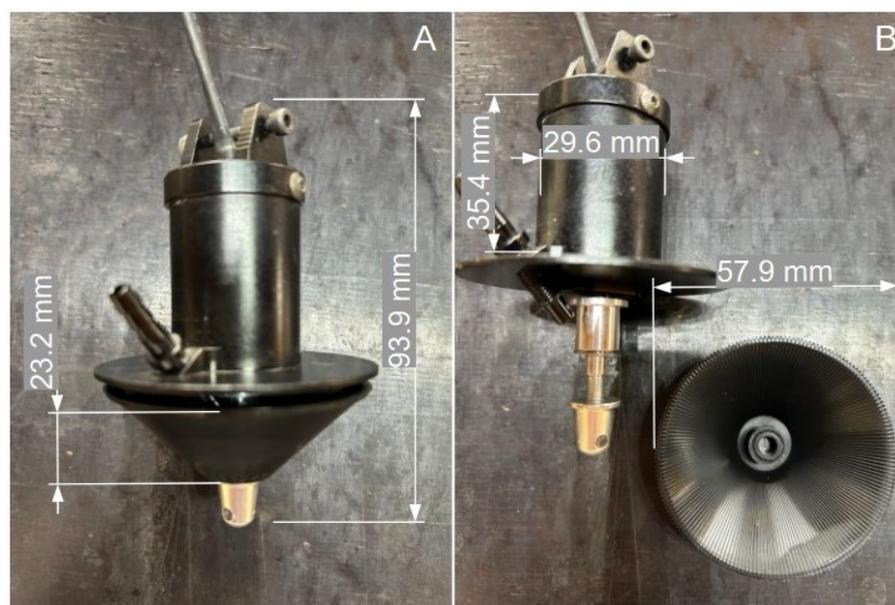


Figure 1. Dimensions of the centrifugal atomization nozzle used in the RPA, shown assembled (A) and with the droplet generation cone disassembled (B). Source: authors (2024).

A stationary sprayer, the Combat model (Micron), equipped with an 80 L tank, was used to supply the hydraulic flow of the spray solution (water only). The flow rate of each sample was measured using a graduated cylinder over a period of 60 s. The centrifugal atomization nozzle can modulate droplet size based on the rotational speed of the disk, operating within a flow rate range of 1 to 2 L min^{-1} . The atomizer's rotation was achieved using a custom-built DIY 12 V, 900 kV model 2210 electric motor, powered by a generic pulse generator (model ZK-MG) (Figure 2). This generator controlled the nozzle's rotation by varying the pulse width modulation (PWM) frequency between 1 Hz and 100 kHz. This device enables the precise adjustment of the atomizing nozzle's electric motor rotation while maintaining a consistent 12 V direct current (DC) power supply.

The droplet population spectrum (VMD, relative span, and percentage of the spray volume containing droplets smaller than 100 μm) was directly determined using a real-time laser particle analyzer (Figure 3), which is capable of measuring the droplet spectrum passing through a laser beam that undergoes refraction during the sampling period. The droplet size classification followed the ASAE S572.1 standard [14]. The particle size analyzer used was the Spraytec model (Malvern Spraytec). The spray nozzle was positioned 0.5 m above the optical beam of the measuring instrument.



Figure 2. Control of nozzle rotation through a pulse width modulation (PWM) pulse generator. Source: authors (2024).

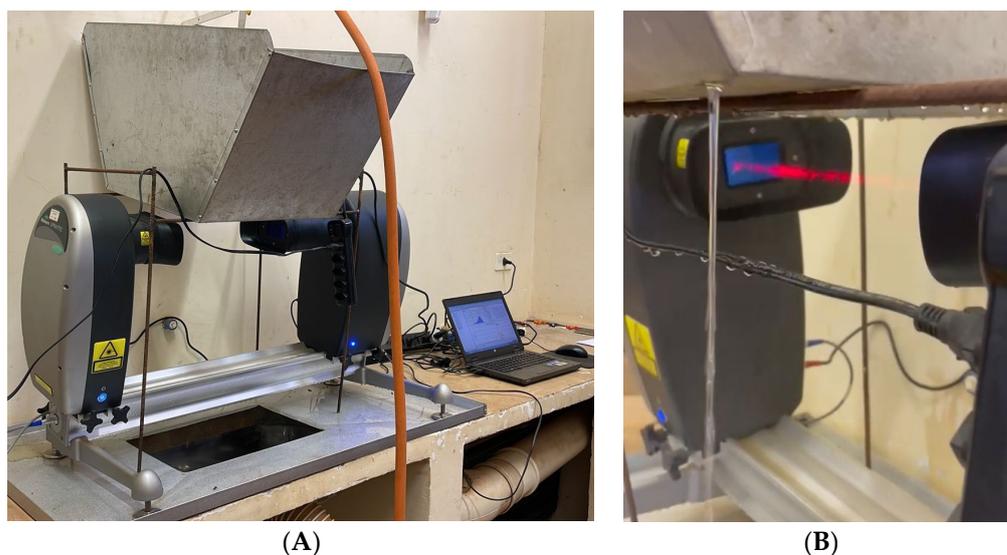


Figure 3. Diagram of the Malvern Spraytec particle sizer (A) and the laser beam in operation (B). Source: authors (2024).

2.4. Statistical Analysis

The data were subjected to analysis for variance using the RBio program [15]. The response surface models were fitted using the SigmaPlot program for each test (test F (Anova), normality test (Shapiro–Wilk), and constant variance test). The best-fitting equation was selected based on the coefficient of determination. Subsequently, principal component analysis (PCA) was performed on all treatments to assess for similarity between variances and to identify which variables contributed most to the observed similarities or disparities.

3. Results

The PWM pulse generator was calibrated by varying the pulse variation percentage from 0 to 100%, resulting in a proportional and linear variation (Figure 4) in the rotational speed of the centrifugal atomization nozzle. This procedure allowed for the consistent repetition of the desired rotational speed throughout the laboratory testing, since the

equipment display provided accurate feedback based on potentiometer adjustments. The calibration of the curve resulted in a linear equation with a high coefficient of determination, ensuring precise and reliable control of the nozzle’s rotational speed.

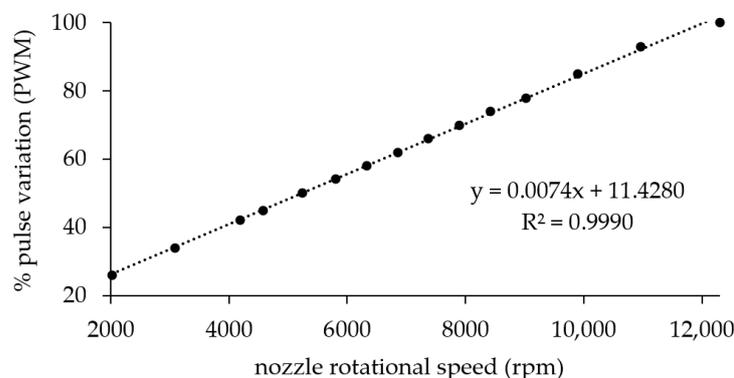


Figure 4. Calibration curve of the rotary atomizer rotation as a function of the PWM pulse variation. Source: authors (2024).

The data from the hydraulic nozzle showed a coefficient of variation for the volumetric mean diameter (VMD) for all treatments of 6.8%, with droplet sizes varying within the fine class from 132.8 to 163.2 μm (Table 1). In contrast, the variation in VMD observed with the centrifugal atomization nozzle was significantly greater (91.9%), enabling the adjustment of several droplet size classes. These classes ranged from very fine droplets, with a VMD of 77.5 μm at a treatment rate of 10 L ha⁻¹ and a rotation speed of 12,000 rpm, to extremely coarse droplets, measuring 1339.0 μm at a treatment rate of 15 L ha⁻¹ and a rotation speed of 2000 rpm.

Table 1. Characterization of the droplet population generated by the hydraulic nozzle and centrifugal atomization nozzle, considering all treatments together.

	Hydraulic Nozzle			Centrifugal Nozzle		
	VMD (μm)	Span	% < 100 μm	VMD (μm)	Span	% < 100 μm
minimum	132.8	1.1	13.0%	77.5	0.2	0.0%
maximum	163.2	1.4	27.3%	1339.0	1.8	48.3%
average	147.8	1.2	20.4%	304.2	1.1	9.7%
coefficient of variation	6.8%	6.5%	20.5%	91.9%	26.5%	147.2%

It is noteworthy that the average relative span (span) of the droplet population generated by the hydraulic nozzle was 1.2, representing a 20% increase over the desired reference value of 1. This value exceeds the overall average observed with the centrifugal atomization nozzle, which had a span of 1.1. The highest span index values measured with the centrifugal nozzle were achieved in treatments with a higher nozzle rotation, where a significant reduction in droplet size was observed (Figure 5).

The variation in pressure of the hydraulic nozzle influences both the flow rate (and consequently the application rate) and the droplet size (Figure 6A). Although droplet sizes exhibit some variation, this variation is relatively minor (remaining within the same class of fine droplet sizes) compared to the variation produced by the centrifugal atomization nozzle (Figure 6B). Therefore, substantial modifications in droplet sizes require the selection of an alternative hydraulic nozzle model. In contrast, the tested centrifugal atomization nozzle enables variation in the volumetric mean diameter (VMD) in response to changes in its rotational speed. This characteristic is advantageous as it allows for rapid adjustments to spraying conditions without requiring any alterations to the device.

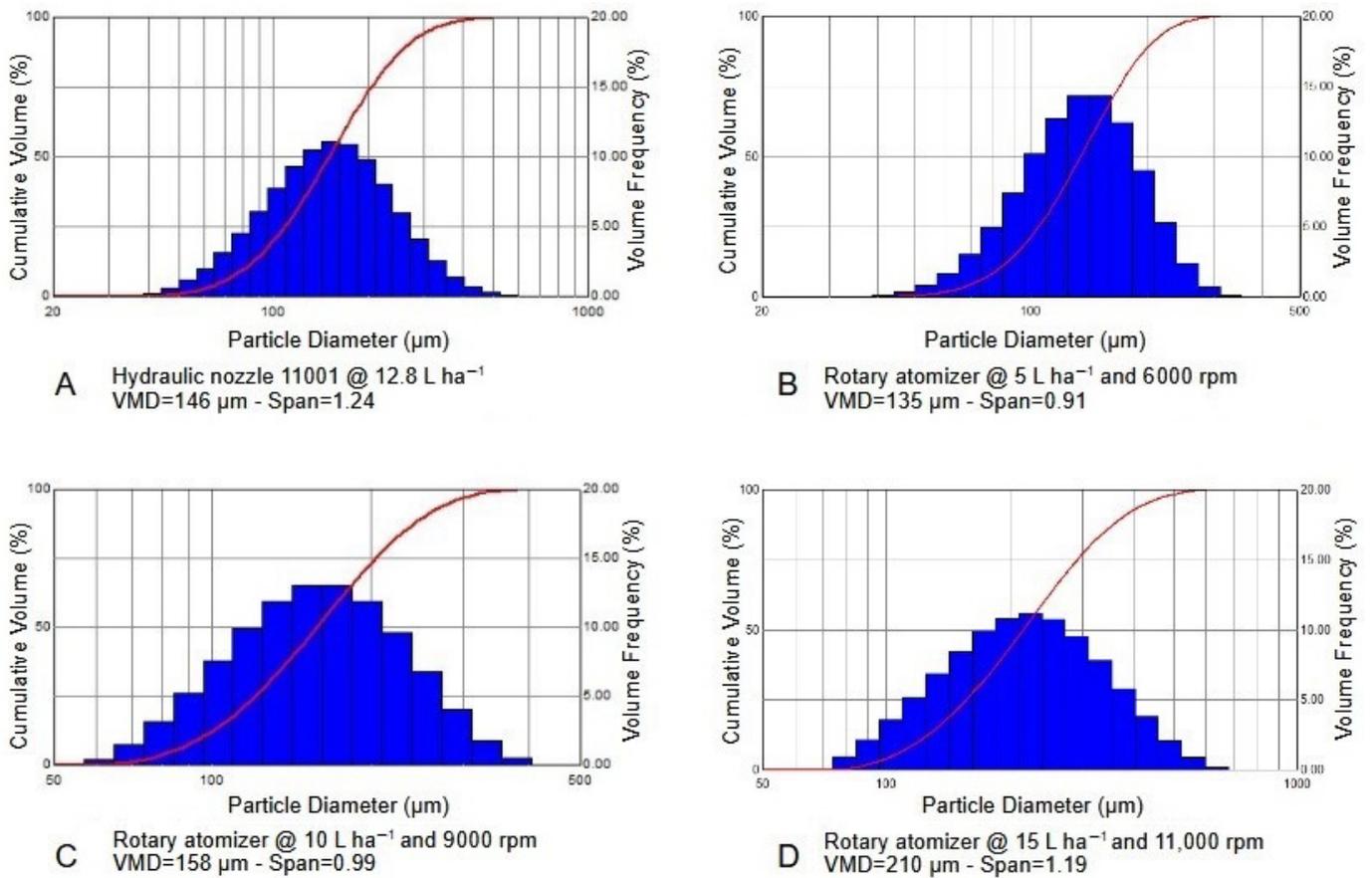


Figure 5. Cumulative and relative frequency as a function of the droplet sizes generated from samples analyzed with the hydraulic tip 11001 (A) and centrifugal atomization nozzle (B–D), the latter varying the rotation and application rate (B–D). Source: authors (2024).

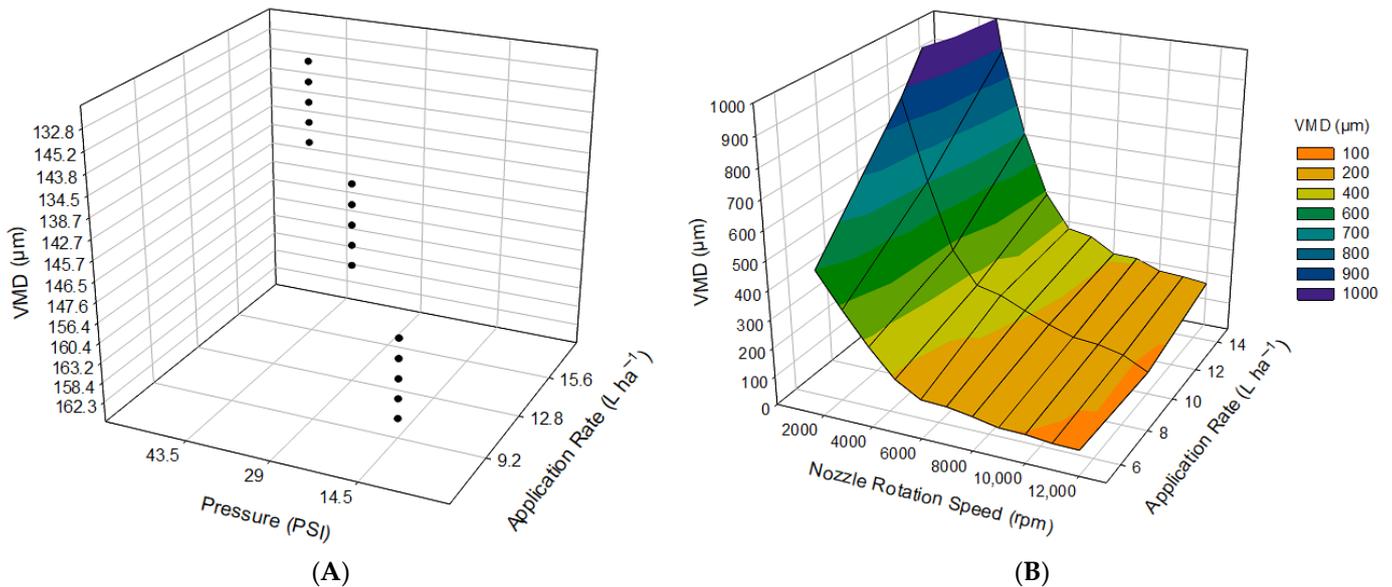


Figure 6. The relationship between the volumetric median diameter (VMD) of the generated drops and the spray application rate as a function of variations in hydraulic nozzle pressure (A) or centrifugal nozzle rotational speed (B). Source: authors (2024).

Another notable feature of this nozzle model is its ability to maintain a consistent droplet size while allowing for variations in the application rate. Specifically, if the operator needs to apply a spray pattern with a droplet size similar to a conventional aerial spraying

of 150 μm , this spray pattern can be achieved regardless of the application rate. However, when using a hydraulic nozzle, the variation in the application rate affects the droplet size, thereby influencing other droplet population parameters, such as the percentage of the spray volume composed of droplets smaller than 100 μm (Figure 7) and the relative span (Figure 8).

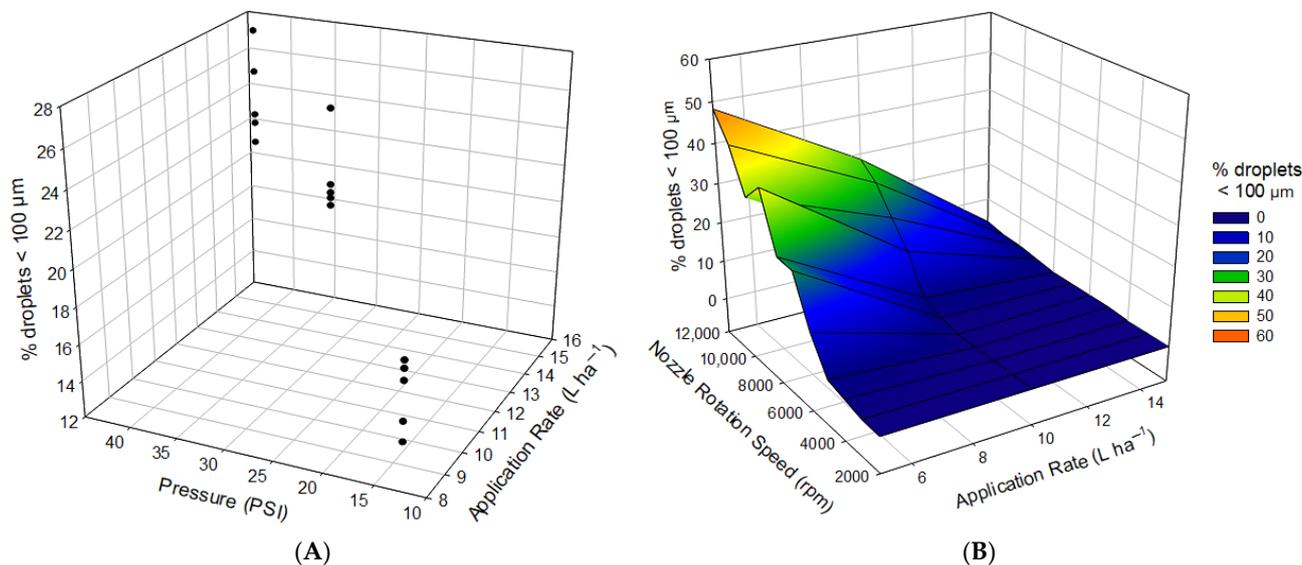


Figure 7. The relationship between the percentage of droplet volume smaller than 100 μm and the spray application rate as a function of variations in hydraulic nozzle pressure (A) or centrifugal nozzle rotational speed (B). Source: authors (2024).

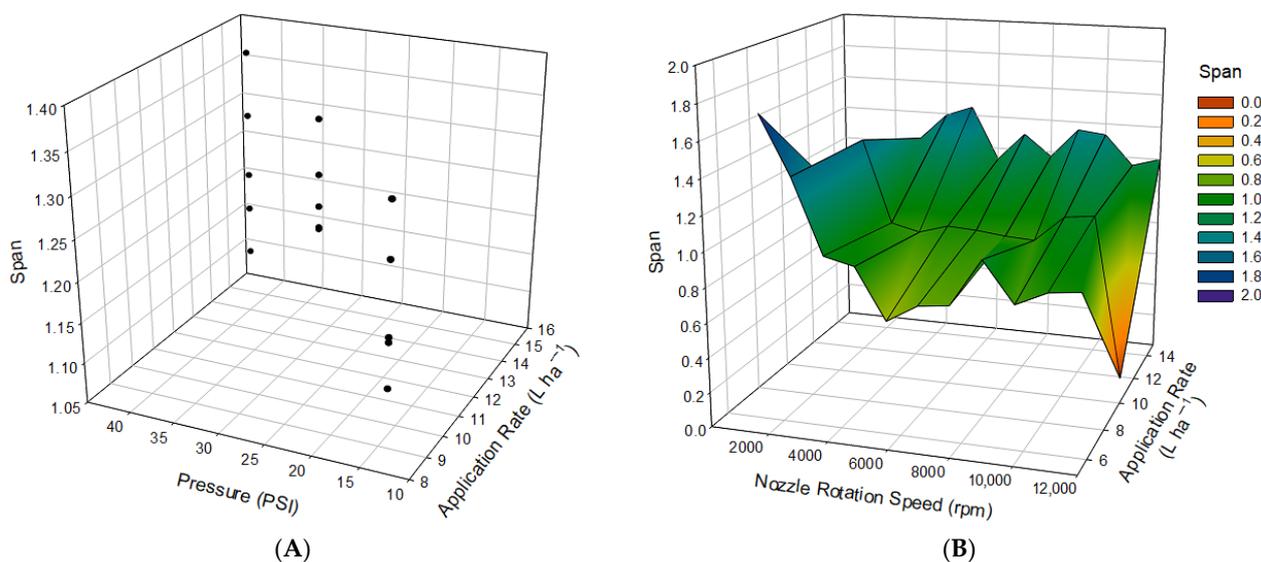


Figure 8. The relationship between the relative span of the droplet population (dimensionless) and the spray application rate as a function of variations in hydraulic nozzle pressure (A) or centrifugal nozzle rotational speed (B). Source: authors (2024).

Pearson correlation analysis was performed for both study conditions, as shown in Figure 9. Positive correlations are represented by green lines, while negative correlations are indicated by red lines. The thickness of each line reflects the strength of correlation between the variables, with thicker lines denoting stronger associations. By analyzing the principal components of the variables from the hydraulic nozzle test (Figure 10A), it was observed that, as expected, hydraulic pressure exerts a strong, proportional, and positive influence

on the variation in application rate. This phenomenon can be observed by the near overlap of the vectors for these two variables, both in direction and intensity (vector length). This increase in hydraulic pressure also exerts a direct and proportional effect on the percentage of droplet volume smaller than 100 μm . Conversely, the increase in hydraulic pressure on the nozzle has an inversely proportional effect on droplet size, as indicated by a reduction in the volumetric median diameter (VMD). The relative span (span) of the hydraulic nozzle did not exhibit a clear trend of variation in response to changes in pressure. Consistent with the previous analyses of the surface graphs, an increase in the rotation of the centrifugal atomization nozzle (Figure 10B) significantly affects droplet size (VMD) in an inversely proportional manner. Additionally, while the increase in rotation also inversely affects the variability of the relative span, this effect is less pronounced than those observed in VMD. The analysis revealed that the variability of the measured data between the relative span and the percentage of droplets smaller than 100 μm exhibit an inversely proportional relationship. Furthermore, the increase in nozzle rotation is directly proportional to the volume of droplets smaller than 100 μm . This increase is undesirable, as it correlates with a greater quantity of droplets that are more susceptible to drift losses.

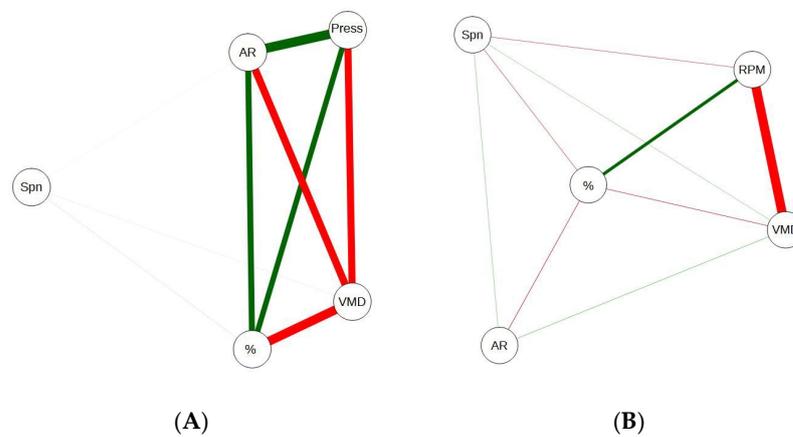


Figure 9. Pearson correlation analysis for the hydraulic nozzle (A) and the centrifugal atomization nozzle (B). The variables include application rate (AR), volumetric median diameter (VMD), revolutions per minute (RPM), span (SPN), pressure (Press), and the percentage of the volume of droplets smaller than 100 microns (%). Source: authors (2024).

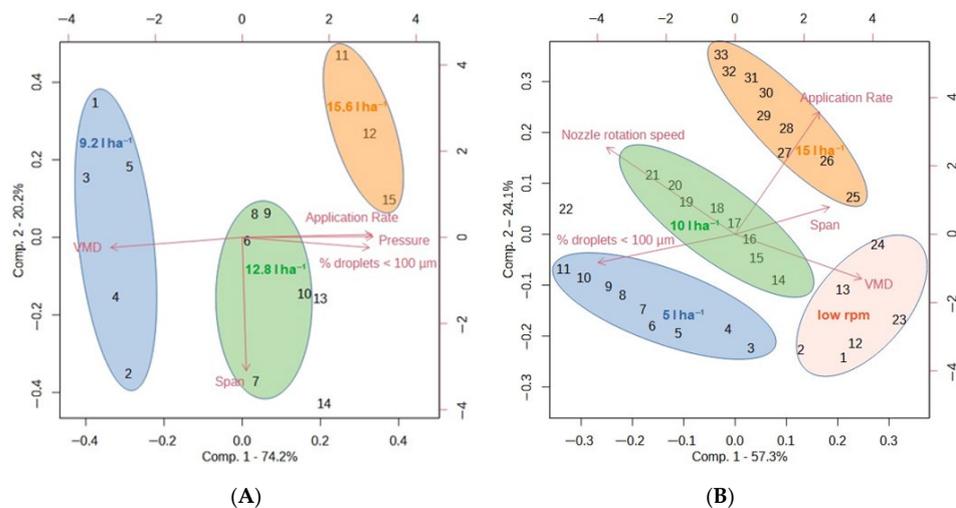


Figure 10. Analysis of the principal components for the variables assessed in the tests conducted with the hydraulic nozzle (A) and the centrifugal atomization nozzle (B). Source: authors (2024). $R_{sq} - DMV = 0.8452$, $span = 0.3071$, and $\% = 0.7341$.

It is important to highlight some premises of this study to evaluate the results of the centrifugal atomization nozzle in comparison with those of the hydraulic nozzle. The variability in the results from the population of droplets generated by the hydraulic nozzle cannot be directly compared to those from the centrifugal atomization nozzle through a statistical analysis. The droplet size produced by the hydraulic nozzle varies with pressure, while the centrifugal atomization nozzle's droplet size changes with the nozzle rotation speed. These represent different operational characteristics, which make direct comparison through statistical tests difficult. However, the differentiating results are evident. Additionally, this is important to note as a consideration during future evaluations of principal component analysis, specifically regarding the possible reduction in the number of treatments. This could lead to better results in the principal component analysis evaluation, as indicated by the Kaiser–Meyer–Olkin (KMO) test. Nevertheless, all treatments have been presented here to highlight the variability of the results, even when considering all possible treatments.

4. Discussion

Calibration adjustments, both in practice and in the field, are of paramount importance. According to existing reports, the use of unmanned aerial spraying systems for chemical applications is widespread globally [12]. In East Asia, where agricultural field conditions are limited, and original plant protection equipment remains prevalent, there is a significant demand for unmanned aerial spraying systems in the market [16]. By the end of 2020, the number of drones operating in China reached 106,000, covering a total annual working area of 64 million hectares [17]. Moreover, calibration is critical in the cultivation of soybean and corn crops, which typically require a minimum of five or six spray applications throughout their growing cycles. Furthermore, the use of pesticides mixtures in these applications is common, which can alter the physical characteristics of the sprayed solutions [18].

Droplet size is inversely correlated with evaporation losses; smaller droplets evaporate faster than larger ones [19]. Altitude and flight speed can extend the time droplets remain airborne, but they also increase their susceptibility to natural crosswinds and environmental weather conditions, which contribute to drift [20]. Although the market for unmanned aerial spraying systems is expanding, the risk of environmental drift associated with drone applications remains a significant concern [21,22]. The risk of spray drift can be closely related to the operational efficiency and various operational parameters [12].

The water discharged by emitters is dependent on the pressure applied to the system, which generates the force necessary to push the water through the nozzle or emitter. For a standard nozzle or emitter, an increase in pressure is directly correlated with an increase in discharge volume [23].

The variation in droplet size as a function of changing the rotation of the centrifugal atomization nozzle is significantly greater than that observed with alterations in the spray application rate at a constant nozzle rotation. Likewise, an increase in the spray application rate, while maintaining a constant rotation, is accompanied by a corresponding increase in VMD. This scenario is particularly pronounced when the nozzle operates at lower rotational speeds (Figure 6B). The results obtained from the hydraulic nozzle test (Figures 6A–8A) closely resemble those reported in previous studies [13], since the extended-use fan tip 11001 is widely recognized and commonly employed in applications involving ground boom sprayers [10]. The variation in the volume of droplets with a diameter smaller than 100 μm , as measured using the centrifugal atomization nozzle, exhibited greater variability in response to changes in nozzle rotation compared to variations in the application rate (Figure 7B). This proportional variation was significantly more pronounced at higher nozzle rotations, as evidenced by a substantial decrease in droplet size as a function of this

increase in rotation. This indicates a greater breakdown of the sprayed volume into smaller droplets. No clear trend in the relative span of the data measured with the centrifugal nozzle was observed, as it neither consistently increased nor decreased in relation to variations in rotation (Figure 8B). Ninety percent of the treatments tested showed a relative span variation ranging from 0.925 to 1.265. Notably, more expressive variations in relative span were observed at both the lower and upper extremes of the centrifugal atomization nozzle rotation.

The spectrum of atomized droplets can be adjusted by controlling the rotational speed of the spray disk to meet the different droplet size requirements. At different voltage levels, the nozzle rotational speed can range from 0 to 17,000 revolutions per minute (RPM) [21].

In Figure 9, Pearson correlations derived from the analyses conducted separately for each nozzle type are presented. One analysis for the rotary nozzle and another for the hydraulic nozzle. Each analysis is independent, highlighting the requirement of a minimum hydraulic pressure for the proper operation of hydraulic nozzles, which is essential for initiating spray and generating droplets. Changes in this variable significantly influence the relative span (spray homogeneity) and the percentage of droplets that are more susceptible to drift. In contrast, centrifugal atomization nozzles do not require a specific working pressure, making it easier for the spraying equipment to control the application rate without changing the droplet size. Furthermore, these devices enable enhanced uniformity in the spectrum of the generated droplet population. Centrifugal atomization nozzles are more suitable for low-volume spraying and especially for spraying that employs variable rate technology [3]. On the other hand, hydraulic nozzles are not suitable for these technologies. According to Gong et al. [3], given that variable rate spraying is an important function in precision agriculture, the remotely piloted aircraft (RPA) sprayer configured with a rotary atomization nozzle is expected to serve as a guiding reference for future RPA sprayer projects and the automation of this activity. Adapting the VMD and controlling other variables of the droplet population to the conditions of aerial applications using remotely piloted aircraft (RPA) sprayers are of crucial importance. This is primarily due to the very low application rates employed by these devices, which can be as low as 5 to 10 L ha⁻¹. Spraying under such conditions is extremely technical and can compromise the effectiveness of pests, diseases, and weed control. Therefore, the selection of rotary atomizing nozzles for RPA sprayers can enhance the efficacy of controlling these targets. Consistent with the increasing preference for centrifugal atomization nozzles in conventional aerial spraying in Brazil, particularly for low and ultra-low volume applications, this approach should also be adopted by operators utilizing RPA sprayers. This choice will likely enhance the effectiveness of aerial spraying with this technology and consequently promote the wider adoption of this type of equipment. Furthermore, the rapid adjustability of droplet size generated by centrifugal atomization nozzles, even during flight, makes them particularly suitable for RPA sprayers equipped with weather sensors. This feature allows for real-time adjustments in droplet size in response to variations in wind speed or atmospheric humidity conditions.

5. Conclusions

Increasing the pressure of the hydraulic nozzle and the rotational speed of the centrifugal atomization nozzle results in a reduction in droplet size. This effect is more pronounced in the centrifugal nozzle, leading to a shift in the classification of the generated droplet sizes.

The pressure within the hydraulic nozzle circuit directly affects both the application rate and droplet size. In contrast, the centrifugal atomization nozzle allows for variation in the application rate without modifying the droplet size.

The relative span of the hydraulic nozzle is greater than that observed with the centrifugal atomization nozzle. When disregarding the extreme rotation rates of the centrifugal atomization nozzle, which are unusual in practice, the percentage of the volume of droplets generated that are smaller than 100 μm is lower than those produced by the hydraulic nozzle.

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Data Availability Statement: The raw data supporting the conclusions of this article will be made available by the authors on request.

Conflicts of Interest: The authors declare no conflicts of interest.

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