## PARTICLE SIZE CHARACTERISATION OF AGRICULTURAL SPRAYS USING LASER DIFFRACTION

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#### Abstract

Control of agricultural spray deposition is vital if pesticides and other agrochemicals are to be delivered safely and effectively to the intended target. One of the most significant parameters in controlling the behaviour of these sprays is the droplet size. Research has shown that the spray droplet size, and the factors which effect it, is one of the most important variables affecting spray deposition levels downwind of the application area (spray drift). The droplet size also defines how the spray is accumulated by the target species.

A variety of different particle sizing techniques have been used to characterise agricultural sprays in the past. Of those available, laser diffraction provides a flexible and rapid method for the assessment of the delivered particle size. Modern laser diffraction equipment allows data acquisition rates of up to 2500Hz to be achieved (one measurement every 0.4ms), permitting both the average particle size delivered by the spray system and the spray dynamics to be assessed. This allows for an increased understanding of the atomisation process and the effect of formulation changes on the delivered particle size, leading to improved efficiency of delivery to the intended target.

In this paper we present some new results obtained for agricultural spray nozzle systems and formations, showing how it is possible to rapidly produce a profile of how the particle size varies across the entire spray plume using laser diffraction. The question of how laser diffraction is affected by the presence of air bubbles within the spray droplets is also explored. The results of both theoretical modelling and practical work show that laser diffraction provides a robust method for particle size determination for agricultural sprays.

#### Introduction

The particle size and size distribution of the droplets produced by nozzles is important in determining the performance of agrochemical application systems, both in terms of treatment efficiency and environmental impact. The use of fine particle sprays can lead to increased surface deposition on leaves. However, fine particles are more likely to be subject to spray drift away from the target area (the settling velocity of particles varies approximately as the square of the droplet's diameter). The use of coarse particle sprays can help eliminate drift and can also penetrate into the plant canopy, thus allowing the lower parts of the target to be treated. However, the risk of soil contamination and run-off becomes high. Thus, the particle size must be controlled in order to provide the correct performance, dependant upon the target species and the environmental conditions.

#### **Spray Particle Size Measurements**

A variety of different particle sizing techniques have been used for the characterisation of agrochemical sprays in the past. These vary from those based on intrusive collection methods, to imaging techniques and those using laser light scattering or absorption. Of those available, laser diffraction provides a flexible and rapid method for the assessment of the delivered particle size. Modern laser diffraction equipment allows data acquisition rates of up to 2500Hz to be achieved (one measurement every 0.4ms), permitting both the average particle size delivered by the spray system and the spray dynamics to be assessed. This allows for an increased understanding of the atomisation process and the effect of formulation changes on the delivered particle size.

Laser diffraction as a technique for particle sizing has been around since the late 1970's [1]. It is widely used for particle sizing in many different applications. Its success is based on the fact that it can be applied to a variety of different particulate systems. The technique is well established with the recent ISO standard "Particle Size Analysis – Laser Diffraction Measurements" [2], the stated purpose which is to "provide a methodology for adequate quality control in particle size analysis." The ISO standard provides a clear description of the general principles of Laser Diffraction particle sizing and provides the user with a guide explaining the performance expected from an instrument of this type.

The laser diffraction measurement principle is based on the fact that a particle passing through a collimated laser beam will scatter light at an angle which is inversely proportional to its size. A typical experimental set-up is shown in figure 1. The particles being measured are passed through a parallel laser beam. Any scattered light is focused onto a radial array of silicon diode detectors using a Fourier transform lens. This lens has the property of imaging the scatter from particles of the same size to the same part of the detector array, regardless of their position and velocity as they pass thought the laser beam and their speed. Consequently at any given moment, there is a light energy distribution across the detector which directly corresponds to the particle size distribution of the droplets which are present in the laser beam.



Figure 1. A schematic of a typical laser diffraction system.

The particle size distribution relating to a given scattering pattern is obtained by fitting the data obtained to an appropriate scattering model. Historically, the Fraunhofer approximation was employed for calculating particle size distributions. However, this model incorrectly predicts the scattering seen for particles smaller than 50 microns in size [2]. For this reason it is necessary to use the Mie scattering model. The Mie model correctly accounts for the scattering seen from small and transparent particles and can therefore be used to accurately assess the fine particle fraction present within sprays. This is important in the case of agrochemical sprays, as it is the fine fraction that is responsible for spray drift. Mie theory requires the refractive index of the particles, the refractive index of the medium (air) and a parameter relating to the transparency of the particles is input.

#### Theoretical Considerations – The measurement of Droplets Containing Bubbles

The measurement of spray droplets containing internal scatterers such as bubbles can represent a significant challenge for light scattering techniques. This is particularly true of techniques such as Phase Doppler Anemometry (PDA) where the measurement of particle size is dependent on the retrieval of phase information. The presence of secondary scattering species within the particle being measured changes the phase of the observed scattering, thus making it virtually impossible to obtain size as well as velocity information. This has been a significant obstacle to the acceptance of phase-based techniques within industries such as the agricultural industry where bubbles are often present within the droplets being measured. It has often been assumed that laser diffraction particle sizing is subject to the same errors as are observed with techniques such as PDA when droplets containing bubbles are measured, as refraction of light within the particle can provide a significant contribution to the observed scattering.

#### **Modelling Droplet Scattering**

The scattering from droplets containing bubbles can be modelled using the Mie scattering model for layered particles [3]. This model is available within Malvern Instrument's Zetasizer software and allows the scattering intensity at any angle to be predicted for droplets containing a second phase. The scattering intensity values can then be input into Malvern Instrument's laser diffraction software in order to calculate the associated size distribution using a Mie model which assumes that only as single droplet phase is present. The layered particle model assumes that the droplet structure is similar to that shown in figure 2 and requires the following parameters as input:

- The refractive index (RI) and absorption of the droplet phase (in this case water: RI = 1.33, Absorption = 0).
- The RI of the internal phase (an air bubble in this case: RI = 1).
- The RI of the medium surrounding the droplet (air in this case: RI=1)
- The proportion of the droplet volume which is occupied by the internal phase.
- The size of the droplet (R) in microns.
- The scattering angle at which the scattering intensity is to be calculated.



Figure 2. Droplet structure assumed in the core-shell scattering model used in this study.

Mie theory predicts that refraction within the droplet only contributes significantly to the observed scattering when the droplet diameter is less that  $40\lambda$  [2]. This relates to a limit of around 25 to 30 microns in the case of the Spraytec system, which uses a 670nm laser source. Calculations of the scattering from bubble-filled droplets were therefore limited to under 25 microns in this case. Figure 3 shows the distribution used in this study, as calculated by the Malvern laser diffraction software for the case where no bubbles were present within the droplet phase.



Figure 3. Size distribution used in the polydisperse distribution calculations.

The scattering patterns calculated as a function of bubble size for the polydisperse distribution defined above are shown in figure 4.



**Figure 4.** Scattering patterns calculated for polydisperse distribution centred at 12 microns containing air bubbles. Bubble proportions of between 0% and 90% by volume are shown. The scattering angle increases as the detector number increases.

Hardly any variation is observed in the magnitude and position of the primary scattering peak as the bubble proportion is increased. Some variation is seen in the secondary scattering response (detectors 29-36). This is as expected as the presence of an internal scatterer will effect refraction of light through the droplet.

The Malvern software reports a D[4,3] of 12.6 microns for the droplets containing no air bubbles. The reported particle size remains relatively constant as the bubble proportion is increased (figure 5). A decease in the result obtained is observed at a bubble proportion of around 80%. The reason for this is unclear. The result variation is excellent, with only a 0.7% variation being observed in the reported D[4,3] across all bubble proportions (table 3). These results suggest that the effect of bubble-filled droplets on the laser diffraction result is negligible, even for small droplets. The effect would be expected to become smaller as the droplet size increases due to the decrease in importance of the secondary scattering component. However, a greater effect would be expected for very narrow, sub-25 micron size distributions where the secondary scattering component will be both significant and well-resolved.



Figure 5. D[4,3] reported by the Mastersizer program as a function of the air bubble volume percentage.

Bubble Proportion (%)	Dv10 / Microns	Dv50 / Microns	Dv90 / Microns	D[4,3] / Microns
0	8.80	12.30	16.90	12.63
10	8.83	12.31	16.94	12.65
20	8.75	12.29	16.95	12.62
30	8.58	12.30	17.12	12.62
40	8.62	12.31	17.06	12.62
50	8.72	12.30	16.97	12.63
60	8.72	12.25	16.94	12.60
70	8.48	12.16	17.11	12.53
80	8.55	11.96	16.62	12.33
90	8.74	12.19	16.97	12.58
95	8.51	12.29	17.25	12.63
Average	8.66	12.24	16.98	12.59
Variation (%)	1.40	0.88	0.94	0.72

Table 1. Particle size distribution statistics reported for droplets containing different bubble proportions.

#### Application of Laser Diffraction to Agrochemical Spray Measurements

The particle size distribution delivered by a two agrochemical spray nozzles (one air inclusion (AI) nozzle and one standard anvil nozzle) was measured using the Spraytec laser diffraction system (Malvern Instruments, Malvern, Worcs, UK) [4]. This system allows real time measurements to be carried out at speeds of up to 2500Hz, allowing the dynamics of spray evolution to be assessed. In this case measurements were acquired every second as the spray was passed through the measurement zone, with the spray fan set at right angles to the laser beam.

#### **Air Inclusion Nozzle Measurements**

Figure 6 shows the "time history" recorded for an AI nozzle as the spray was scanned across the laser diffraction measurement zone.



Figure 6. Time history recorded for the movement of an AI fan spray through the Spraytec measurement zone.

The transmission value recorded here relates to the concentration of particles within the spray plume. This increases towards the centre of the plume. The recorded particle size is relatively small at the extremes of the spray plume. In the centre a roughly constant particle size is observed. Some large fluctuations are seen in the recorded values. This relates to the sensitivity of the laser diffraction technique to the presence of coarse particles (the scattering strength increases according to the volume of the particle). The appearance or disappearance of a few large particles can therefore have a large effect of the calculated results.

The average particle size distribution calculated for the entire spray plume is shown in figure 7. As can be seen, the particle size is large, partly due to the air inclusions with the droplet phase. No artefacts relating to the bubbles within the droplet phase are observed. These results are in broad agreement with those obtained using image-based measurements.



Figure 7. Average size distribution calculated for the AI nozzle.

#### **Anvil Nozzle Measurements**

The time history profile recorded for passage of the spray produced by a standard anvil nozzle through the Spraytec measurement zone is shown in figure 8.



Figure 8. Time history recorded for the movement of an anvil nozzle spray through the Spraytec measurement zone.

The profile obtained in this case is completely different to the AI nozzle. The concentration of the spray is much higher. The concentration at the centre of the spray is also lower than towards the edge of the spray plume. This is as expected for an anvil-type nozzle. The particle size at the extremes of the spray plume is large in this case, moving to smaller particle sizes towards the centre of the plume. Again, these results are in broad agreement with those obtained using image-based measurements.

#### Conclusions

The technique of laser diffraction provides a robust means of measuring the particle size of agrochemical sprays, allowing both the spray dynamics and the spray-plume profile to be assessed. The results obtained using laser diffraction techniques have, in the past, been questioned due to the unknown effect of air inclusions on the obtained results. Theoretical modelling based on the Mie layered particle model shows that for large particles and polydisperse distributions the presence of bubbles has a negligible effect on the results. Both AI and standard agrochemical sprays can therefore be characterised using the technique.

#### References

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