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Viscosity, surface tension and droplet size of sprays of different formulations of insecticides and fungicides

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The influence of three types of commercial formulation of insecticides and fungicides, emulsifiable concentrate (EC) formulation, suspension concentrate (SC) formulation and water dispersible granule (WG) formulation on the surface tension, viscosity and the droplet size spectra of sprays was evaluated using thirty commercial insecticide and fungicide products. The concentration of sprays was based on application water spray volume of 50 L ha^{-1} using an XR 8003VS flat fan nozzle, operated at 200 kPa pressure. The lowest surface tensions were obtained with EC formulations, while the SCs had the highest viscosities. Emulsions were the most effective at decreasing the percentage of droplets smaller than 100 μm and relative span, while increasing the $DV_{0.1}$ and $DV_{0.5}$ values than the dispersion-type formulations, represented by WG and SC formulations. These factors should be considered for planning spray applications and reducing drift.

1. Introduction

Spray drift is one of the major concerns during the application of pesticides, as it may affect the environment and human health, and the efficacy of controlling insects, fungal diseases and weeds (Newsom, 1967; De Schampheleire et al., 2008; Hilz and Vermeer, 2013).

Fungicides and insecticides are commonly sprayed using similar spray qualities in Brazil. Because of the need for good coverage and penetration in the canopy, most applications are done using smaller droplets (usually from very fine to medium spray quality), most frequently using standard flat fan nozzles. Dual flat fan and hollow cone nozzles are also used to spray these pesticides, but because of the higher risk of spray drift (Chechetto et al., 2014; Carvalho et al., 2017) the adoption of these nozzles is decreasing.

Pesticide formulations affect parameters such as viscosity, surface tension and droplet size, which influence the drift potential of sprays atomized with conventional flat fan nozzles (Miller and Butler Ellis, 2000).

The presence of emulsion droplets present in sprays using EC formulations, or the small particles in SC or WG formulations, affect the liquid sheet formed at the nozzle, influencing the droplet spectra, and consequently, the drift potential (Miller and Butler Ellis, 2000; Hilz and Vermeer, 2013). According to these authors, emulsions or dispersions may lead to formation of larger droplets than those formed when spraying water-soluble formulations.

Nevertheless, most spray application research is conducted using "blank" formulations, often water, where there are no active ingredients, or using just adjuvants, trying to simulate the characteristics of commercial products. This is done to avoids contamination of the laboratory, or exposure of people to pesticide residues. This is also due in part to the prohibitions of usage of active ingredients by law in some locations (Hoffmann et al., 2007; Nuyttens et al., 2009; Fritz et al., 2010). Some researchers also have evaluated commercial formulations, but for comparing different formulations of the same active ingredient (Sanderson et al., 1997; Kirk, 2000).

It is not clear, however, how commercial formulations of

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pesticides affect surface tension, viscosity and droplet size spectra of sprays. For instance, there is no consensus whether the impact of EC, WG, and SC formulations on droplet size is the same for insecticides and fungicides. This represents an important area of study for improving spray applications quality, as well as for mitigating spray drift.

Thus, the aim of this study was to evaluate the effects of EC, SC and WG formulations of insecticide and fungicide products, represented by five commercial products for surface tension, viscosity and droplet size spectra of spray.

2. Material and methods

Five samples of commercial products comprising EC, SC and WG formulations of insecticides and fungicides were selected for evaluation in a 2 (insecticide versus fungicide) x 3 (EC versus SC versus WG) factorial design, in a completely randomized experiment. This selection of products was based on their availability in the Brazilian market.

The concentration of the pesticide in water was determined according to the label recommendations, prioritizing those for controlling Asian soybean rust (*Phakopsora pachyrhizi* Sydow & P. Sydow), to fungicides, and velvetbean caterpillar (*Anticarsia gemmatalis* Hübner), to insecticides, both for the use in soybeans (*Glycine max* (L.) Merril) with application volumes of 50 L ha^{-1}. This volume application rate is commonly used in some regions of Brazil for ground applications (Chechetto et al., 2014; Carvalho et al., 2017).

A flat fan nozzle XR 8003VS (TeeJet, Spraying Systems, Wheaton, Illinois, USA) was used at an operating pressure of 200 kPa (2 bar), according to earlier trials by Moreira Júnior and Antuniassi (2010), and to represent the nozzle type commonly used for fungicides and insecticides applications in Brazil (Chechetto et al., 2014). Environmental condition during these studies were 20.5° C (±0.74° C) and relative humidity of 73.51% (±3.47%).

The droplet spectra were measured with a VisiSizer P15 equipment (Oxford Lasers, Imaging Division, Oxford, U.K.), Particle/Droplet Image Analyses (PDIA), according to the methodology described by Guler et al. (2007). This method for measuring droplets, ranging from 21 μm up to 3490 μm is detailed by Kashdan et al. (2003). A CO_2 propellant was used to pressurize the spray system.

Among the available options offered by the equipment used to characterize droplet size spectra, $DV_{0.1}$, $DV_{0.5}$ (or Volume Median Diameter, VMD) and $DV_{0.9}$ (the diameter of droplets representing 10%, 50% and 90% of the sprayed volume), % < 100 μm (percentage by volume composed of droplets smaller than 100 μm) and relative span (difference in diameter for $DV_{0.9}$ and $DV_{0.1}$ of the sprayed volume divided by the $DV_{0.5}$) were selected (Mugele and Evans, 1951; Tate and Janssen, 1966; Goering and Smith, 1978; Hewitt, 2007; Ferguson et al., 2015; Al Heidary et al., 2014).

Relative span is a dimensionless parameter used to indicate uniformity of the droplet size spectra, where smaller values indicate a narrower spectrum (Hewitt, 2007). The %<100 μm has a positive correlation with spray drift potential, while $DV_{0.5}$ has a negative correlation (Courshee, 1959; Miller, 1998; Antuniassi et al., 2011; Oliveira et al., 2015). Droplet spectra data was replicated three times.

A Brookfield DV-II + Pro viscometer measured solution viscosity (Oliveira et al., 2015). The instrument was equipped with a cylinder of 100 mm external diameter (*spindle* # S-00) at 60-rpm rotation, according to the manufacturer's recommendations. The surface tension of the solutions was determined using the drop-weight method (Gans and Harkins, 1930; Saad et al., 2011; Oliveira et al., 2015). This method uses the weight of droplets generated at the end of a capillarity tip to indicate the surface tension. Five replications were used to characterize both parameters.

All the evaluations were completed using two spray solutions for the same treatment, to ensure that analytical mistakes during the evaluations were minimized. The treatments are described in Tables 1 and 2.

The data was subjected to analyses of variance (ANOVA) and, when significant differences were observed, the average of the results were compared by the Tukey's test at 5% level of significance using SAS (SAS, Cary, NC, USA) software.

3. Results and discussion

There was no interaction between formulations and pesticides ($P > 0.05$). Furthermore, there were no statistical differences between the classes of pesticides, but there were between the types of formulations for all the evaluated parameters (Table 3). However, the results for relative Span, $DV_{0.1}$, $DV_{0.5}$, and %<100 μm were similar between fungicides and insecticides for each evaluated formulation. This shows that the type of formulation was the determinant factor for the results obtained for those parameters.

The SC and WG formulations resulted in the relative span being about 34% and 47%, respectively, higher than for EC formulations, that was close to 1.2. The %<100 μm was also higher for those formulations, by approximately 120% and 250%, respectively, than that observed for EC formulations, which resulted in %<100 μm smaller than 6%.

The viscosity of SC formulations was higher than observed with EC and WG. According to Knowles (2008), Paul and Robeson (2008) and Zhang et al. (2011) this is explained by the components, usually polymers, used to keep the solid active ingredient particles in suspension in SC formulations, increasing the viscosity of spray liquid. According to the authors, these substances also have the capability of altering parameters as such as $DV_{0.5}$ and %<100 μm. However, as observed in this study, the effects of emulsions on increasing $DV_{0.1}$, $DV_{0.5}$ and decreasing %<100 μm has exceeded the effect of higher viscosity of SC formulations.

The SC and WG formulations resulted in $DV_{0.5}$ approximately 20% and 34%, smaller than observed to EC formulations, respectively. Hilz and Vermeer (2012) observed that an oil dispersion formulation (OD) of an imidacloprid insecticide had a $DV_{0.5}$ around 20% higher than that obtained for WG and SC formulations of that insecticide. These authors used an XR 11003VS nozzle, at 300 kPa, simulating a volume rate of 200 L ha^{-1} to conduct that research. The %<100 μm for the OD formulation was about 50% smaller than the observed to the other evaluated formulations. These data are in congruence with the observed in the present study.

When liquids being atomized are forced through the orifice of flat fan nozzles, a sheet is formed that spreads out as it breaks up with perforations forming within the sheet that forms ligaments at its edge and then as individual droplets that create the spray. As some droplets are formed there is often a smaller satellite droplet also formed (Matthews et al., 2014a).

Hewitt et al. (2002) explained that droplet spectra is affected principally by the physical characteristics of spray liquids, and do not depend on active ingredients. The presence, particularly the proportion of the spray liquid in droplets smaller than 100μm, including the very small satellite droplets determines the drift potential. However, the nature of some active ingredients may determine the formulation type, thus whether it is soluble in a suitable solvent or is a solid more suited to a particulate formulation. And thus indirectly affects physical characteristics of the spray liquids (Matthews et al., 2014b).

The EC formulations had the lowest surface tension results, about 37 mN m^{-1}, while for the other treatments it ranged from 44.42 mN m^{-1}, for SC fungicides, to 63.43 mN m^{-1}, for WG

Table 1
Active ingredient (a.i.), commercial products (c.p), percentage of the a.i. (% a.i.) and rate of the commercial products (mL or g c.p. 50 L^{-1}), for EC, emulsifiable concentrate, SC, suspension concentrate, and WG, water dispersible granules, formulations of insecticides.

Active ingredient	Commercial product	% a.i.	Rate (mL or g c.p. 50 L^{-1})
Insecticides			
EC			
chlorpyrifos	Lorsban® 480 BR	48	1000
deltamethrin	Decis® 25 EC	2.5	300
prophenophos + lufenuron	Curyon® 550 EC	50 + 5	150
abamectin	Vertimec® 18 EC	1.8	600
bifenthrin	Talstar® 100 EC	10	160
SC			
thiamethoxam + lambda-cyhalothrin	Engeo™ Pleno	14.1 + 10.6	200
flubendiamide	Belt®	48	70
methoxyfenozide	Intrepid® 240 SC	24	150
imidacloprid + beta-cyfluthrin	Connect®	10 + 1.25	1000
teflubenzuron	Nomolt® 150	15	50
WG			
fipronil	Fipronil 800 WG	80	40
thiamethoxan	Actara® 250 WG	25	200
imidacloprid	Evidence® 700 WG	70	250
thiodicarb	Larvin® WG	80	70
imidacloprid	Imidagold 700 WG	70	250

Table 2
Active ingredient (a.i.), commercial product (c.p), percentage of the a.i. (% a.i.) and rate of commercial products (mL or g c.p 50 L^{-1}) for EC, emulsifiable concentrate, SC, suspension concentrate, and WG, water dispersible granules, formulations of fungicides.

Active ingredient	Commercial product	% a.i.	Rate (mL or g c.p. 50 L^{-1})
Fungicides			
EC			
tebuconazole	Folicur® 200 EC	21.3	500
tebuconazole	Orius® 250 EC	25	400
pyraclostrobin	Comet®	25	300
difenoconazole	Score® 200	25	200
difenoconazole	Tilt®	25	400
SC			
difenoconazole + difenoconazole	Aproach® Prima	20 + 8	300
trifloxystrobin + prothioconazole	Fox®	15 + 17.5	400
azoxystrobin + cyproconazole	Priori Xtra®	20 + 8	300
trifloxystrobin + tebuconazole	Nativo®	10 + 20	500
fluxapyroxad + pyraclostrobin	Orkestra®	16.7 + 33.3	350
WG			
azoxystrobin	Amistar WG	50	120
azoxystrobin + benzovindiflupir	Elatus™	30 + 15	300
mancozeb	Manzate® WG	75	3000
mancozeb	Unizeb® Gold	75	3200
metiran	Polyran® DF	70	3000

insecticides. According to Miller and Butler Ellis (2000) the presence of surfactants in formulations will affect surface tension, but it is not the unique parameter affecting droplet size.

Knowledge of the droplet spectrum, especially the proportion by volume of the smallest droplets, often regarded as those below 100 μm for different formulations and adjuvants, if used, is important for different nozzles and operating pressures to be able to choose most suitable nozzle to minimize spray drift (Hilz and Vermeer, 2013; Gandolfo et al., 2014).

The tests recorded here refer to one specific flat fan nozzle and provide data relevant to current practice in Brazil and countries where similar practices are used. However, with greater concern about downwind spray drift, Nuyttens et al. (2007) compared several different nozzles and confirmed that low-drift flat-fan nozzles and air-inclusion nozzles applied significantly less volume in the smallest droplets liable to drift. Further study is needed to see if these nozzles should be used in Brazil, especially as they vary in design and in some cases, produce a very coarse spray with high VMD, that is less likely to provide adequate coverage with some pesticides to achieve adequate control of some pests, when using low volume application rates.

In conclusion, this study provides data of the droplet spectra and properties of certain pesticide formulations, relevant to current practices in Brazil. Such data is important for planning spray applications and to adopt drift reduction technologies (DRTs).

At the low volume application rate considered (50 L ha^{-1}) and use of one nozzle (XR 8003VS) and operating pressure (200 kPa), the emulsifiable concentrate formulations of both insecticides and

Table 3
Means of the evaluated parameters for spray solutions composed by fungicides and insecticides of EC, SC and WG formulations.

Class	Evaluated parameters[a]		
	EC[b]	SC	WG
ST (mN m^{-1})			
Insecticide	39.23cA[c]	61.53bA	63.43aA
Fungicide	35.73cA	44.42bB	51.29aB
Viscosity (mPa s)			
Insecticide	1.15bB	1.27aA	1.10cB
Fungicide	1.12cA	1.19aB	1.17bA
Span			
Insecticide	1.25cA	1.59bA	1.77aA
Fungicide	1.16cA	1.63bA	1.76aA
$DV_{0.1}$ (μm)			
Insecticide	124.41aA	102.83bA	90.01cA
Fungicide	126.13aA	101.17bA	91.73cA
$DV_{0.5}$ or VMD (μm)			
Insecticide	255.20aA	200.95bA	165.10cA
Fungicide	247.41aA	200.21bA	166.68cA
$DV_{0.9}$ (μm)			
Insecticide	440.78aA	415.80bA	382.50cA
Fungicide	412.58bB	424.51aA	383.23cA
%<100 μm (%)			
Insecticide	5.91cA	11.37bA	18.05aA
Fungicide	4.73cA	12.08bA	16.81aA

[a] ST: surface tension; Span: relative Span; $DV_{0.1}$, $DV_{0.5}$ and $DV_{0.9}$ (10%, 50% and 90% of the sprayed volume contains droplets smaller than a droplet whose diameter is the $DV_{0.1}$, $DV_{0.5}$ or $DV_{0.9,}$ respectively), %<100 μm (percentage by volume composed by droplets smaller than 100 μm).
[b] EC: Emulsifiable concentrate; SC: concentrate suspension; WG: water dispersible granules.
[c] Different lowercase letters on the lines, and capital letters on the columns, statistically differ according to the Tukey's test ($\alpha = 0.05$).

fungicides increased the volume median diameter of the spray with less spray prone to drift compared with the SC and WG formulations.

Small differences detected in surface tension and viscosity of the three types of formulation were not reflected in the overall droplet size data.

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