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RESEARCH ARTICLE

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The effect of coarse-droplet spraying with double flat fan air induction nozzle and spray volume adjustment model on the efficiency of fungicides and residues in processing tomato

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Abstract

The study was conducted for the purpose of improving the application of fungicides against potato late blight (*Phytophthora infestans* (Mont.) de Bary) (PLB) in processing tomato. The usability of coarse spray quality with double flat fan air induction IDKT12003 nozzle and the impact of fixed and variable spray volume and adjuvants during alternate application of azoxystrobin and chlorothalonil were analysed on the basis of plant infestation and fungicide residues. The variable spray volume was calculated based on the number of leaves on a plant. The study was conducted during three vegetation seasons. Spraying of plants with significantly flattened canopies during the peak of the fructification season using an IDKT12003 nozzle was as effective as in the case of fine spraying performed with an XR11003 nozzle and facilitated the increase of fungicides residue. In the case of plants with high-spreading canopy at the beginning of fructification, XR11003 nozzle favoured the reduction of PLB infestation. Both spray volume adjustment systems enabled the same level of protection of tomato against PLB, which could result from alternate application of systemic and contact fungicides. Polyalkyleneoxide modified heptamethyltrisiloxane adjuvant, which causes siginificant increase in wetting and droplet spreading, facilitated the reduction of tomato PLB infestation during the application of fungicides using an IDKT12003 nozzle.

Additional keywords: azoxystrobin; chlorothalonil; *Phytophthora infestans*; spray deposit; QuEChERS.

Abbreviations used: GS (growth stage); LAI (leaf area index); OS (organosilicone surfactant); PLB (potato late blight); PMH (polyalkyleneoxide modified heptamethyltrisiloxane); PSV (proportionate spray volume); QuEChERS (quick, easy, cheap, effective, rugged, and safe); RSF (relative span factor); SV (spray volume); SVAM (spray volume adjustment model); SV300 (spraying 300 L/ha of fungicide suspension).

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Introduction

The effectiveness of pesticides applied with hydraulic nozzles depend on precise spraying (Ozkan *et al.*, 2006; Nuyttens *et al.*, 2007; Fritz *et al.*, 2016; Gonzalez-de-Soto *et al.*, 2016). A lot of studies have analysed the influence of the properties of the sprayed liquid and the type of the spray nozzle used on retention and the surface covered with the sprayed droplets (Lebeau, 2004; Zhu *et al.*, 2004; Hewitt, 2008; van Zyl *et al.*, 2010). Theoretically, assuming that droplets of the same size are produced out of the same liquid volume, the coverage of the flat surface increasing in inverse proportion to the decrease of the droplet diameter. However, the spray deposition in the plant canopy depends, to a great extent, on the location of the analysed fragment. During spraying with horizontal boom, the application is significantly greater in the upper part of the canopy than in the middle and lower parts (Zhu *et al.*, 2004; Lipiński *et al.*, 2007). Aside from the air assistance, the coverage of the lower leaves in potatoes was increased using a double flat fan nozzle. Coverage of these leaves with droplets from fine spray was still low, despite yielding greater deposition than in the case of coarse spray quality (Kierzek & Wachowiak, 2009).

The laboratory and field studies on droplet deposition show that the effectiveness of contact fungicide is positively correlated with the leaves coverage (Grinstein *et al.*, 1997; Washington, 1997) including potato (Prokop & Veverka, 2006). The biological activity of systemic fungicides may be less dependent on the coverage of the surface with droplets. The biological effect of the application of a systemic pesticide depends on its amount in the target site according to the deposition, penetration and translocation of the active substance (Green & Hazen, 1998; Kierzek & Wachowiak, 2005; Wang & Liu, 2007; Taylor, 2011). It is well known that coarser droplets facilitate the absorption of glyphosate (Feng *et al.*, 2003), although their potential of covering surface is lower than in case with smaller droplets produced out of the same volume of liquid. In fact, little is known about other pesticides (Wang & Liu, 2007).

The droplets of the sprayed liquid are influenced by weather conditions. A lot of studies focused on droplet evaporation and drift, usually within the context of environmental contamination (Holterman *et al.*, 1997; Nuyttens *et al.*, 2007; Farnham *et al.*, 2015). Both phenomena may have adverse effect on the effectiveness of the treatment, resulting in significant loss of active substance. Taking the above into consideration, the use of nozzle produced coarse spray during application of systemic fungicides may result in satisfactory effectiveness of the treatment and not only in windy conditions.

As it has already been mentioned, the effectiveness of contact fungicides depends on the coverage of the surface by the liquid droplets (Grinstein *et al.*, 1997; Washington, 1997) which, in turn, is correlated with spray volume (SV). Various authors have achieved better protection of potatoes and tomatoes against potato late blight (PLB), using higher SV (Prokop & Veverka, 2006; Ratajkiewicz *et al.*, 2016). However, Jensen & Nielsen (2008) have obtained satisfactory results after applying only 160 L/ha against PLB. On the other hand systemics are less influenced than contact fungicides (Ratajkiewicz *et al.*, 2009; 2016; Wise *et al.*, 2010). Although only part of the active substance is absorbed and further transported within the plant (Bartlett *et al.*, 2002), these phenomena may explain similar residues of the fungicide (azoxystrobin) after its application with highly diversified SV (Ratajkiewicz *et al.*, 2016). In agriculture, systemic and contact fungicides are usually applied alternately. In such conditions, higher SV may also facilitate lower plant infestation with the disease, which may be associated with higher SV rate inside canopy. Both SV rate and fungicide rate on a plant depend on the type of crop and the canopy characteristics (Walklate *et al.*, 2003; Dammer *et al.*, 2008; Cooke *et al.*, 2011; Llorens *et al.*, 2011). The spray deposit into the canopy is proportional to the SV applied (Walklate & Cross, 2013) and is inversely proportional to the leaf area index (LAI) (Zhu *et al.*, 2004). As a consequence, during spraying with horizontal boom the high interception of fungicide was on external part of the plant canopy (potato) and decreased from top to bottom (Bruhn & Fry 1982; Hamm & Clough, 1999). The development of vegetable-dedicated SV adjustment models is relatively poor. However, when SV was calculated on the basis of the number of leaves on a plant, it had significant influence on the effectiveness of the protection of tomato against PLB when the application was performed by using standard fine-droplet spray nozzle (Ratajkiewicz *et al.*, 2016). It seems that further research should be conducted on this topic.

Various types of physical interactions can improve the biological action of pesticide: adhesiveness, wetting, spreading, penetrating, retention, rainfastness and extension of duration of action (Holloway *et al.*, 2000; Ryckaert *et al.*, 2007; Hunsche, 2008; Taylor, 2011). Optimization of the properties of the adjuvant in relation to the physical effects of the pesticide is of key importance (Schönherr *et al.*, 1999; Ramsdale & Messersmith, 2001).

It is believed that increasing the coverage of leaves with liquid through the use of surfactants, increases the effectiveness of the contact fungicide (Prokop & Veverka, 2006). Organosilicone surfactants (OS) are one of the most effective wetting agents (Nikolov *et al.*, 1998). They also facilitates the infiltration of stomata and capillaries and influence on foliar penetration (Stevens *et al.*, 1991; Schönherr *et al.*, 1999). One of the widely-known examples of a highly effective mix of contact fungicide and a surfactant against PLB is cyazofamid and OS (Mitani *et al.*, 2001).

Adjuvants increasing retention, wetting, infiltration, and rainfastness are considered suitable for systemic pesticides (Field & Bishop, 1988; Stevens *et al.*, 1988; Manthey *et al.*, 1998; Schönherr *et al.*, 1999; Wang & Liu, 2007). Many factors influence the infiltration of pesticide through the cuticle in the presence of an adjuvant (Holloway, 1995; Schönherr *et al.*, 1999; Wang & Liu, 2007; Taylor, 2011). However the fungicidal activity may depend on the chemical group and specific properties of the adjuvant (Grayson *et al.*, 1996). Schönherr *et al.* (1999) divided adjuvants into two groups: passive and active, the latter being called accelerators. Passive adjuvants solubilise the pesticide deposit on a surface, while accelerators not only solve it, but also penetrate into cuticles and increase the solute mobility (Wang & Liu, 2007; Schönherr *et al.*, 1999). Some surfactants, in particular OS, may increase the infiltration of liquids through stomata, as well as to leaf sheaths and, as a result, may accelerate pesticide penetration through the leaf cuticle and the rainfastness (Field & Bishop, 1988; Stevens *et al.*, 1991). Some adjuvant may also activate the active substance (Thelen *et al.*, 1995; Green & Hazen, 1998); however, little is known about such effects in the case of fungicides.

It is believed that the influence of an adjuvant on the effectiveness of a pesticide is greater when SV is lower (Schönherr *et al.*, 1999; Ramsdale & Messersmith, 2001). This can be explained by higher ratio between the amount of the adjuvant and the pesticide and higher concentration of the adjuvant in water (Ramsdale & Messersmith, 2001; Wang & Liu, 2007). Finally, adjuvants may also be used in combination with reduced doses of active substances (Gaskin *et al.*, 2004; Ryckaert *et al.*, 2007). In practice, reduction of the pesticide doses with tank-mix adjuvants is most widely practised in case of systemic herbicides (Kierzek & Wachowiak, 2005), especially with sulfonylurea group (Stagnari *et al.*, 2007; Barros *et al.*, 2009).

Adjuvants that modify the physical properties of liquids may also influence the formation of droplets during spraying and the deposit of droplets on a plant. Similarly, the physical properties of the preparation, being a resultant of interaction between different substances, may influence the droplet formation (Miller & Ellis, 2000). Droplet formation during spraying depends mainly on viscosity and surface tension. In turn, both of these properties, especially liquid viscosity, depend on temperature (Lefebvre, 1989; Miller & Tuck, 2005; Farnham *et al.*, 2015). Moreover, both the shear and extentional viscosity influences the size of the produced droplets (Różańska *et al.*, 2012; Broniarz-Press *et al.*, 2013). Droplet retention on the surface of the leaves also depends on physical properties of the liquid, as well as on the size and speed of the droplet (Spillman, 1984; Stevens *et al.*, 1993; Rioboo *et al.*, 2002; Dorr *et al.*, 2015). At this stage, three elements of physical properties play crucial role: surface tension, viscosity and liquid density. Finally, the effect of adjuvants on the effectiveness of fungicides is multidirectional and is gaining recognition.

Taking into consideration the importance of the adjustment of SV for the protection of processing tomato against PLB and the residues of azoxystrobin and chlorothalonil in fruits that have been found and shown in the studies where fine-droplet spray nozzle was used (Ratajkiewicz *et al.*, 2016), this study aims at presenting the results of a parallel study where double flat fan air induction nozzle produced coarse spray was used. The aim of this work was to assess the usability of IDKT12003 nozzle and the spray volume adjustment models (SVAM) in the protection of processing tomato against PLB on the basis of infestation of the plants with the disease and fungicide residues.

Material and methods

Experimental design

The experimental model compares two spray volume adjustment models (SVAM), two nozzles and two adjuvants used to apply fungicides against PLB (Table 1). The study was conducted using the setup, location, time and other output data consistent with the information presented in Ratajkiewicz *et al.* (2016).

The adjustment of SV to the spraying of tomatoes with fungicides was based on two models. From the first to the last application, either fixed SV (300 L/ha, SV300) or proportionate spray volume (PSV), calculated on the basis of the number of the leaves on a plant, were used, according to the model presented in the previous study (Ratajkiewicz *et al.*, 2016). The appropriate SV for spraying with PSV model and order of fungicides is shown in Table 2.

The double flat fan air induction IDKT12003 (Lechler GmbH, Metzingen, Germany) and as standard extended range flat fan XR11003VP (TeeJet Spraying Systems Co., Wheaton, USA) nozzles were used to spraying at 300 kPa pressure. Both nozzles has got identical flow rates (size “03” according to ISO) but produce coarse and fine spray quality, respectively, according to BCPC specification (Tomlin, 2000). Plots were sprayed with a precision field knapsack sprayer equipped with horizontal spray boom with 4 nozzles. The boom was 40 cm over the top of canopy.

In the study, two fungicides were used interchangeably: azoxystrobin (methyl (E)-2-[2-[6-(2-cyanophenoxy) pyrimidin-4-yl]oxyphenyl]-3-methoxyprop-2-enoate, Amistar 250 SC; Syngenta Ltd, Guildford, UK) at a dose of 125 and 250 g/ha (50% and 100% of the dose recommended) or chlorothalonil (2,4,5,6-tetrachlorobenzene-1,3-dicarbonitrile - Gwarant 500 SC; Arysta LifeScience S.A.S., Noguères, France) at a dose of 625 g/ha and 1250 g/ha (50% and 100% of the dose recommended) (Table 2).

Table 1. The experimental model with incomplete factor structure of the treatments

Treatment	Nozzle	SV[a] (L/ha)	Adjuvant	Fungicide dose[b] (%)
T1	XR11003	300	–	100
T2	IDKT12003	300	–	100
T3	IDKT12003	300	–	50
T4	IDKT12003	300	PMH	50
T5	IDKT12003	300	multi-ingredient adjuvant	50
T6	XR11003	PSV	–	100
T7	IDKT12003	PSV	–	100
T8	IDKT12003	PSV	–	50
T9	IDKT12003	PSV	PMH	50
T10	IDKT12003	PSV	multi-ingredient adjuvant	50
T11 (control)		–	–	

[a]SV, spray volume; PSV, proportionate spray volume. [b]Azoxystrobin at dose of 125 and 250 g/ha (50% and 100% of the dose recommended, respectively) or chlorothalonil at dose of 625 and 1250 g/ha (50% and 100% of the dose recommended, respectively)

Table 2. The order of application of fungicides and the number of leaves per plant and the spray volume (SV) for specific dates for proportionate spray volume (PSV) model of application

Spraying no.	2009			2010			2011		
	Fungicide and application date [a]	Leaves numbers (GS) [b]	SV (L/ha)	Fungicide and application date	Leaves numbers (GS)	SV (L/ha)	Fungicide and application date	Leaves numbers (GS)	SV (L/ha)
1	A 20 Jul	26.9 (65-71)	300	C 06 Aug	33.4 (67-72)	372	A* 13 Jul	46.7 (68-71)	521
2	A 24 Jul	30.5 (66-72)	340	A 13 Aug	47.9 (71-75)	535	C 20 Jul	55.2 (69-73)	615
3	C 31 Jul	41.7 (71-75)	465	C 24 Aug	49.9 (73-81)	565	A 25 Jul	61.5 (71-75)	686
4	A 07 Aug	55.7 (71-75)	622	A 03 Sept	53.5 (81-85)	597	C 02 Aug	50.3 (71-75)	686
5	C 14 Aug	58.2 (73-81)	668	C 18 Sept	- (85-89)	597	A 09 Aug	- (73-81)	686
6	A 25 Aug	67.6 (75-83)	754	A [c] 18 Sept; 20 Sept	- (85-89)	597	C 12 Aug	- (75-82)	686
7	C 07 Sept	63.0 (85-88)	754	-	-	-	A 16 Aug	- (75-82)	686
8	A 11 Sept	63.0 (85-88)	754	-	-	-	-	-	-
Sample collection	14 Sept			23 Sept			19 Aug		

[a] A, azoxystrobin; C, chlorothalonil. [b]GS, growth stages of tomato according to BBCH scale. [c]The azoxystrobin application was repeated (20 September) due to local short heavy rainfall.

Every year, the fungicide treatment started with azoxystrobin. The first spraying was conducted after five and four days of observing the first small pale green lesion on leaf, characteristic to PLB, in 2009 and 2010, respectively (Table 2). In July 2010, no PLB symptoms were observed and the fungicide treatments started in August due to the increased risk of the disease.

One of the two adjuvants, Slippa (Interagro Ltd., Great Notley, UK), containing polyalkyleneoxide modified heptamethyltrisiloxane (PMH) (655 g/L), or Torpedo II (De Sangosse Ltd, Swaffham Bulbeck, UK), were added to the fungicide suspension. The last adjuvant contains alkoxylated tallow amine 210 g/kg; alcohol alkoxylate 380 g/kg; natural fatty acids 75 g/

kg and polyalkylene glycol 210 g/kg, which is why it was described in this work as a multi-ingredient adjuvant. Both substances were applied at the concentration of 1 mL/L and added to the suspension only when the dose of fungicide equalled half of the recommended dose.

Experimental set-up

Field studies were carried out in Poznań, Poland (16.858092 52.408529 decimal degrees) in 2009, 2010 and 2011. The processing tomato (*Solanum lycopersicum* L.) cv. Polset F1 was cultivated on a sandy loam soil in a twin-row planting system (0.5 m × 0.5 m × 1.5 m). The tomatoes were planted in the first decade of June in the number of 14 plants per plot. Standard fertilization and weed control with herbicides were used. No fungicides other than the above, insecticides and acaricides were used in the study.

Fungicides extraction and chemical analysis

The samples of tomato fruits for study of the residues were collected in the amount of no less than 1 kg from the plot according to the schedule presented in Table 2 and frozen after pre-processing.

The modified Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) method was used for extraction of azoxystrobin and chlorothalonil residue. The optimization process and method validation were presented in a previous work (Ratajkiewicz *et al.*, 2016). Gas chromatography with a nitrogen and phosphorus detector and an electron capture detector (GC–NPD/ECD 6890 – N Agilent Technologies, USA; column: DB-5, 30 m × 0.53 mm × 0.88 µm, Agilent J&W Scientific, USA) was used for the analysis of fungicides.

PLB evaluation

The degree of leaf infestation with PLB was assessed using the modified Horsfall-Barrat rating scale of 1 to 12, where 1 is 0% and 12 is 100% disease severity (Berger, 1980). The fruit infestation with PLB was presented as percentage of fruits with disease symptoms, with 20 fruits observed per plant. The degree of infestation was assessed on the basis of 10 plants on each plot on the day of the last spraying in the season and two weeks later.

Meteorological conditions

HOBO weather station (ONSET Comp. Corp., Bourn, USA) and dedicated sensors were used to registered meteorological data presented in Table S1 [suppl].

Droplet size measurement

Droplet size distributions for the XR11003 and IDKT12003 nozzle was conducted using the droplet laser image analysis system Malvern Spraytec. The spraying medium was water and water solutions of adjuvants.

Droplet samples were taken 50 cm below the nozzle orifice and across centerline along the long axis of the spray pattern by scanning within the entire width of the droplet stream. Measurement was replicated three times for each condition. The spraying spectrum was then defined by volume median $D_{V0.5}$ and diameters $D_{V0.1}$ and $D_{V0.9}$. The relative span factor (RSF) was also calculated using the formula RSF= $(D_{V0.9} - D_{V0.1}) / D_{V0.5}$.

Statistical analysis

The field study was performed using completely randomised block design, with 3 blocks in 2009 and 4 blocks in 2010 and 2011.

In the experiment, 11 treatments, comprising an incomplete structure consisting of four factors and zero control, were analysed (Table 1). Therefore, the study was treated as a one-factor experiment. As the results of Bartlet test for some of the variables, a logarithmic transformation or non-parametric Kruskal-Wallis test had to be performed.

An appropriate one way variance analysis (ANOVA) was also conducted on the basis of the variables divided into group contrasts between the treatments (so called basic contrasts). The coefficients of the contrasts obtained after normalisation and orthogonalisation of the data conducted in R gmodels package (Warnes *et al.*, 2015) were presented in Table 3. Tukey's *post hoc* test was used for the comparison of the studied treatments. In case of non-parametric analysis Dunn *post hoc* test (package dunn.test in R stats package) was necessary.

In order to determine the relationship between data vectors the Pearson linear correlation was used.

Results

Droplets characteristics

Characteristic droplet diameters was approximately twice as big in the case of the IDKT12003 spray nozzle than the XR11003 nozzle (Table 4). The adjuvants had slightly bigger influence on the droplet diameter produced by IDKT12003 nozzle than the XR11003 nozzle. However, $D_{V0.1}$, $D_{V0.5}$ and $D_{V0.9}$ were similar in both cases and depended mainly on the nozzle type. RSF decreased under the influence of PMH in the case

Table 3. The coefficients of basic contrast applied in the variance analysis

Contrast	T1	T2	T3	T4	T5	T6	T7	T8	T9	T10	T11
Control	0.009	0.009	0.009	0.009	0.009	0.009	0.009	0.009	0.009	0.009	–0.091
Dose	0.083	0.083	-0.167	0.000	0.000	0.083	0.083	-0.167	0.000	0.000	0.000
SVAM	-0.100	-0.100	-0.100	-0.100	-0.100	0.100	0.100	0.100	0.100	0.100	0.000
Nozzle	0.250	-0.250	0.000	0.000	0.000	0.250	-0.250	0.000	0.000	0.000	0.000
With or without adjuvant	-0.033	-0.033	-0.033	0.050	0.050	-0.033	-0.033	-0.033	0.050	0.050	0.000
Adjuvant type	0.000	0.000	0.000	-0.250	0.250	0.000	0.000	0.000	-0.250	0.250	0.000
Interaction_1	0.286	-0.214	0.071	-0.071	-0.071	-0.286	0.214	-0.071	0.071	0.071	0.000
Interaction_2	-0.071	-0.071	-0.143	0.143	0.143	0.071	0.071	0.143	-0.143	-0.143	0.000
Interaction_3	0.291	0.291	-0.436	-0.328	0.183	-0.291	-0.291	0.436	0.328	-0.183	0.000
Interaction	0.173	0.173	-0.259	0.387	-0.473	-0.173	-0.173	0.259	-0.387	0.473	0.000

SVAM, spray volume adjustment model. Treatments T1–T11 according to description in Table 1.

of both nozzle types, which means that the uniformity of the droplet size distribution increased.

Infestation of tomato with PLB

Dose response

The spraying of fungicides reduced tomato infestation with PLB (Table 5) in each year (Table 6). A full dose of the fungicide without adjuvant resulted in lower infestation of tomato than half the dose in 2009 (leaves) and in 2011 (leaves and fruits) (Table 5 and 6). Two weeks after last spraying of fungicide the corresponding contrast value was –6.2 in 2009 while in 2011 it was –4.0 on leaves and –8.1 on fruits (Table 5). In 2009, two weeks after last spraying of fungicide without adjuvant using IDKT12003 nozzle the mean infestation of tomato by PLB, independently on SVAM, was 42.1% in the leaves area and 15.3% in the fruits number for full dose while 60.0% for leaves and 15.3% for fruits for half the dose. Thus after application half of the fungicide dose the corresponding increase in PLB severity was only on leaves by mean of 42.6%. Two weeks after last spraying in 2010 increase in infestation of fruits with PLB was 19.4% while in 2011 it was 45.1%. In case of leaves in 2011 an appropriate increase amounted 12.9%. However, post-hoc test proved that only in 2011 the recommended dose of the fungicide sprayed with XR11003 nozzle decreased significantly fruits infestation with PLB in comparison to half a fungicide dose applied with IDKT12003 nozzle (Table 6). Due to significant damage of the plants by *Aculops lycopersicae* in 2010, there is no data on the leaf infestation with PLB (Table 6).

Table 4. Spray quality parameters for the nozzles used in field trials

Nozzle	Parameter	Sprayed mixture		
		Water alone	PMH	Multi-ingredient adjuvant
IDKT12003	$D_{v0.1}$	213.3	222.2	217.4
	$D_{v0.5}$	462.3	433.6	454.6
	$D_{v0.9}$	791.2	695.2	743.8
	Span	1.25	1.09	1.16
XR11003	$D_{v0.1}$	109.03	112.4	111.3
	$D_{v0.5}$	221.9	224.7	230
	$D_{v0.9}$	370.3	350.5	385.5
	Span	1.18	1.06	1.19

SVAM response

Negative contrast values (Table 5) show that the plant infestation with PLB was lower in the case of PSV than in the case of SV300. However, only in 2011, PSV was found to be a better system. That year, plants had higher canopies and were younger (growth stage GS = 75-82) than in the previous years. However, the contrast analysis proved that the SVAM did not influence significantly the infestation of tomato by PLB (Table 5) in any year. It should be emphasized that the both nozzles, IDKT12003 and XR11003, were used to construct the basic contrast. In the case of treatments where only IDKT 12003 nozzle was used, two weeks after the last spraying using the PSV system in 2011, the average tomato infestation with PLB was lower by only 3.2% on leaves and 1.6% on fruits. In the case where the plant canopy was significantly flattened (2009 and 2010), the average infestation of tomato fruits after spraying using the PSV system was even higher, *i.e.* by 2.3% and 21.7% in 2009 and 2010, respectively (Table 6). The leaf infestation was lower by only 1.9% (2009).

Table 5. Values of the basic contrasts and their significance for the infestation of leaves and fruits by potato late blight (PLB) throughout the years. The data in the table columns are presented only in years, date and for leaves and fruits if could be done one-way analysis of variance

Basic contrast	2009				2010	2011			
	Leaves		Fruits		Fruits (log)	Leaves		Fruits	
	11 Sept	2 weeks later	11 Sept	2 weeks later	2 weeks later	16 Aug	2 weeks later	16 Aug	2 weeks later
Control	–1.96 **	–4.2 **	–0.25 **	–4.4 **	–0.05 **	–4.6 **	–2.5 **	–4.5 **	–4.3 **
Dose	–0.96	–6.2 **	–0.03	–0.1	–0.04	–6.0 **	–4.0 **	–7.9 **	–8.1 **
SVAM	0.28	–0.1	0.25	–0.1	0.04	–3.3	–1.0	–3.3	–0.7
Nozzle	–0.40	–0.8	–0.75	–0.2	0.02	–5.1	–3.9 .	–4.4	–4.9
With or without adjuvant	–0.09	–0.6	–0.06	0.4	0.05 *	1.4 .	0.9	2.1 *	2.4 **
Adjuvant type	–0.3	1.7	–0.03	2.0	–0.08	6.2 .	2.9	6.9 .	6.1 .
Interaction_1	0.60	0.5	0.34	1.0	0.01	–0.2	0.0	0.2	0.8
Interaction_2	0.20	–2.1	0.13	–1.8	–0.05	3.4	1.4	2.6	1.2
Interaction_3	1.20	–1.6	0.57	1.8	–0.15	4.5	2.8	8.4	6.1
Interaction	–1.90	–8.1	1.38	–3.6	–0.25	–2.0	–7.4	–4.7	–8.7

[a]SVAM, spray volume adjustment model. *p*-value significance codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1.

Table 6. Percentages of the infestation of tomato leaves and number of fruits with potato late blight (PLB) symptoms. The trial was performed with 3 blocks in 2009 and 4 blocks in the other two years

Treatment	2009				2010[a]		2011			
	Leaves		Fruits		Fruits		Leaves		Fruits	
	11 Sept	2 weeks later	11 Sept	2 weeks later	21 Sept [b]	2 weeks later[c]	16 Aug	2 weeks later	16 Aug	2 weeks later
T1	17.1 b	39.2 b	2.30 a	16.0 b	1.0	5.8 a	39.3 bc	56.0 c	36.5 bc	35.0 cd
T2	16.4 b	41.9 b	3.00 a	16.0 b	0.0	6.5 a	46.3 bc	62.5 bc	42.3 bc	42.0 bcd
T3	19.5 b	65.0 ab	1.67 a	17.3 b	2.3	11.5 a	56.5 bc	71.8 bc	57.8 bc	61.5 bcd
T4	16.8 b	38.3 b	2.53 a	11.33 b	2.9	13.8 a	51.5 bc	63.8 bc	48.3 bc	48.5 bcd
T5	18.3 b	47.8 b	1.58 a	19.3 b	1.3	9.3 a	68.0 b	77.4 ab	70.5 ab	71.3 ab
T6	16.4 b	41.7 b	1.31 a	14.0 b	0.4	8.3 a	31.0 c	55.0 c	26.3 c	31.5 d
T7	18.6 b	42.2 b	3.62 a	14.7 b	0.4	9.0 a	44.3 bc	64.3 bc	38.0 bc	44.0 bcd
T8	20.5 b	54.9 b	3.63 a	13.3 b	2.1	7.0 a	59.8 bc	71.3 bc	61.3 b	63.3 bc
T9	19.2 b	47.5 b	2.06 a	18.7 b	0.6	23.1 a	42.5 bc	66.5 bc	45.8 bc	55.5 bcd
T10	16.3 b	44.7 b	2.92 a	18.7 b	0.8	10.9 a	51.0 bc	64.5 bc	51.3 bc	57.0 bcd
T11 (control)	39.4 a	92.4 a	5.16 a	64.7 a	18.0	47.1 a	99.0 a	93.1 a	97.8 a	98.5 a

[a]Infestation of tomato leaves by PLB in 2010 was not showed due to serious damage to leaves caused by unpredictable appearance of *Aculops lycopersici* (Massee). [b]The data failed to meet criteria necessary for both ANOVA and non-parametric analysis. [c]Results subject to statistical analysis with non-parametric test. Means within columns with the same letter are not significantly different by $p<0.05$.

Nozzle response

The nozzle type did not significantly influence the infestation of tomato with PLB when fungicides at full dose were applied (Table 5). However, the negative contrast values show that the fine spray quality (XR11003) provided lower tomato leaf and fruit infestation with PLB than the coarse spray produced by IDKT12003 nozzle in 2011 and 2009. After using the XR11003 nozzle, the average leaf infestation during the last assessment was lower by 14.2% and 4% in 2011 and 2009, respectively, whereas the number of infested fruits was lower by 29.3% and 2.3% in 2011 and 2009, respectively.

Adjuvants

The examined adjuvants did not improve significantly tomato protection against PLB (Table 5 and 6). The infestation of tomato did not differ between plots with the fungicide applied with or without adjuvant in any year (Table 6). In 2011 the infestation of tomato by PLB was even insignificantly higher by mean of 7.8% for leaves and 15.9% for fruits when multi-ingredient adjuvant was applied with SV300 in comparison to fungicide alone at full dose. However both adjuvants gave a satisfactory results of tomato protection with PSV.

The positive values of the basic contrasts show that the PMH had potential to decrease tomato infestation by PLB more than multi-ingredient adjuvant in 2009 and 2011. (Table 5). Two weeks after the last treatment, the average infestation of leaves and fruits by PLB after spraying with fungicide with PMH was lower, respectively, by 7.2% and 21% in 2009 and 8.2% and 18.9% in 2011. In comparison with fungicide applied at half dose without this surfactant, the infestation of leaves and fruits by PLB was lower, respectively by 28.4% and 1.9% in 2009 and 8.9% and 16.7% in 2011. High infestation of fruits in 2010 after the application of fungicides with PMH, especially when performed according to the PSV model, in comparison with the treatment using multi-ingredient adjuvant, as well as without the adjuvant, might have been connected with significant damage of the fruit epiderm by *Aculops lycopersicae* which infested the plants throughout the experiment.

Fungicide residue

General remarks

The mean chlorothalonil residue was lower in 2011 (0.07 mg/kg) than 2009 and 2010 (0.28 and 0.27 mg/kg, respectively). It resulted from much more frequent occluding of fruits by leaves in 2011 than in the remaining years. The reason for that were more numerous stems in upright or slightly inclined position in the day of last spraying due to younger GS in 2011 (GS 75-82) than in 2009 and 2010 (GS ≥85) (Table 2). On the other hand the azoxystrobin residue in fruits was the highest in 2010 (0.43 mg/kg) due to repeated application in the last period (18 and 20 September) because of local short heavy rainfall. Apart from that, the average azoxystrobin residue was slightly higher in 2011 (0.15 mg/kg) than in 2009 (0.12 mg/kg).

The analysis of the Pearson correlation coefficient for the data from all of the treatments from three years of study (n=30) showed that the fungicide residues in fruits was negatively correlated with their infestation by PLB. The correlation coefficients (r) used in the assessment of the infestation performed two weeks after the last treatment in the season equalled –0.6 for chlorothalonil and –0.51 for azoxystrobin. In the case of leaf infestation (data from 2009 and 2011, n=20), r= –0.65 for chlorothalonil and only –0.1 for azoxystrobin.

Dose response

The contrast analysis showed that the full dose of the fungicide resulted in higher residues of azoxystrobin and chlorothalonil in fruits than half of the dose (Table 7).

The reduction of chlorothalonil residue after spraying half of the dose with IDKT12003 nozzle independently on SVAM was 55.9%, 60.4% and 21.4% in 2009, 2010 and 2011, respectively. The corresponding reduction of azoxystrobin residue was 59%, 27.3% and 47.6% in 2009, 2010 and 2011, respectively.

However, the post-hoc test proved that only azoxystrobin residue was significantly lower with half of the dose only after application with SV300 in 2009 and 2011 (Table 8).

SVAM response

The PSV significantly decreased the chlorothalonil residue only in 2009 (contrast value –0.11) (Table 7) in comparison to SV300. However the post-hoc test showed that there was no important differentiation between the treatments. The contrast analysis showed that azoxystrobin residue was not affected by SVAM.

The chlorothalonil residue after application with an IDKT12003 nozzle equalled, on average, 0.32 mg/kg with SV300 and 0.18 mg/kg with PSV in 2009, and 0.25 mg/kg with SV300 and 0.20 mg/kg with PSV in 2010. In 2011, the average chlorothalonil residue equalled 0.05 mg/kg and 0.06 mg/kg for SV300 and PSV, respectively. In the case of azoxystrobin, the average residue after application with an IDKT12003 nozzle was very similar between both SVAM in 2009 (0.11 mg/kg) and 2010 (0.41 with SV300 and 0.43 mg/kg with PSV). In 2011, the azoxystrobin residue equalled 0.15 mg/kg for SV300 and 0.13 mg/kg for PSV, which means that it was slightly higher (14.7%) after application with SV300. It is worth noting that

Table 7. Values of the basic contrasts and their significance for the azoxystrobin and chlorothalonil residues throughout the years. The data in the table columns are presented only in years and fungicides if could be done one-way analysis of variance

Basic contrast	2009		2010	2011
	A[b]	C	A	A
Control	0.009 **	0.086 **	0.030 **	0.006 **
Dose	0.035 **	0.111 **	0.037 *	0.030 **
SVAM[a]	–0.007	–0.111 *	0.023	–0.011 ·
Nozzle	–0.008	–0.014	–0.04	–0.013
With or without adjuvant	–0.015 **	–0.101 **	–0.022 ***	–0.009 **
Adjuvant type	0.003	0.004	0.021	0.005
Interaction_1	0.010	–0.096	–0.021	–0.009
Interaction_2	0.006	0.090	0.02	0.003
Interaction_3	0.048	0.070	–0.032	0.006
Interaction	–0.001	–0.030	–0.067	0.004

[a]SVAM, spray volume adjustment model. [b]A, azoxystrobin; C, chlorothalonil. *p*-value significance codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1.

the plants were younger and taller during the last treatment in 2011 (GS 75-82) than in 2009 and 2010 (GS≥85).

The following correlation coefficients between fungicide residues in fruits after application with coarse spray obtained with IDKT12003 and their infestation with PLB two weeks after the last treatment in the season were achieved: for SV300 system r=–0.47 for azoxystrobin and –0.54 for chlorothalonil and –0.51 and –0.58, respectively, for PSV (with n=12). The leaf infestation was correlated with fungicide residue in the following way: in the case of SV300 r=0.04 for azoxystrobin and r=–0.63 for chlorothalonil, whereas in the case of PSV r=0.08 and –0.56, respectively (for n=8).

Nozzle response

The contrast analysis proved that nozzle type did not influence significantly the fungicides residue in fruits (Table 7). However, except for chlorothalonil in 2011, higher average residues of fungicides applied at full dose, independently of SVAM, were found in the case of the IDKT12003 nozzle than in the case of the XR11003. The average residue of azoxystrobin was higher by 7.7%, 4.5% and 11.9% in 2009, 2010 and 2011, respectively, whereas the chlorothalonil residue was higher by 17.6% and 11.9% in 2009 and 2010. In 2011, the chlorothalonil residue after spraying with IDKT12003 was lower than in the case of XR11003 even by 64%, which resulted from the PSV model. The reasons for this situation are unknown.

The fungicide residue in fruits after application of coarse droplets being produced by IDKT12003 nozzle were negatively correlated with PLB infestation. Two weeks after the last treatment in the season r=–0.49 for azoxystrobin and – 0.54 for chlorothalonil (n=24). In

Table 8. Residues of azoxystrobin and chlorothalonil in tomato fruits (mg/kg). The trial was performed with 3 blocks in 2009 and 4 blocks in the other two years

Treatment	2009		2010		2011	
	A[a]	C	A	C[b]	A	C[c]
T1	0.21 a	0.45 a	0.40 ab	0.45	0.19 ab	0.06 a
T2	0.20 a	0.67 a	0.51 ab	0.57	0.23 a	0.07 a
T3	0.06 c	0.22 ab	0.39 ab	0.15	0.11 bc	0.05 a
T4	0.07 bc	0.16 ab	0.32 bc	0.12	0.13 bc	0.04 a
T5	0.10 bc	0.21 ab	0.41 ab	0.15	0.14 bc	0.05 a
T6	0.15 ab	0.39 a	0.54 ab	0.44	0.18 ab	0.17 a
T7	0.19 a	0.35 a	0.59 a	0.44	0.19 ab	0.07 a
T8	0.10 bc	0.23 ab	0.41 ab	0.25	0.11 bc	0.06 a
T9	0.08 bc	0.07 bc	0.36 ab	0.05	0.10 bc	0.05 a
T10	0.06 c	0.06 bc	0.36 ab	0.07	0.12 bc	0.05 a
T11 (control)	0.02 c	0.03 c	0.03 c	0.02	0.03 c	0.03 a

[a]A, azoxystrobin; C, chlorothalonil. [b]The analysis was conducted using non-parametric Dunn *post hoc* test, results are presented in Table S2 [suppl]. [c]Results subject to statistical analysis with non-parametric Kruskal-Wallis test. Means within columns with the same letter are not significantly different by *p*<0.05.

the case of leaf infestation, r=0.06 for azoxystrobin and r=–0.57 for chlorothalonil (n=16).

Adjuvants

The contrast analysis showed that adjuvants decreased residues of fungicides (Table 7). The post-hoc test did not show any significant differences in the results between treatments using fungicide applied at half dose with and without an adjuvant (Table 8). However, after averaging the results for the adjuvants, the chlorothalonil residue was lower by 44.4% in 2009, 51.3% in 2010 and 13.6% in 2011 in comparison with the same fungicide applied at half of the dose. The reduction of residues using adjuvants was more effective when tomatoes were sprayed at the peak of the fructification season (GS ≥85 in 2009 and 2010) than at the beginning of that season (GS 75-82 w 2011). PMH resulted in significantly high reduction of chlorothalonil residue which equalled 48.9% in 2009, 57.5% in 2010, and 18.8% in 2011. The adjuvants also allowed for a slight reduction of azoxystrobin residues in 2009 (3.2%) and 2010 (9.4%), taking into account an average of the two adjuvants. However, in 2011, an increase in azoxystrobin residues was observed, by 11.4%. The azoxystrobin residue obtained using PMH was lower by 6.3% and 15% in 2009 and 2010, respectively. In 2011, it was higher by 4.6% in comparison to the fungicide alone.

The contrast analysis showed that adjuvant type had no effect on fungicide residue (Table 7). However, the PMH contributed to slightly higher reduction of residues of both fungicides than a multi-ingredient adjuvant. Independently of SVAM, the chlorothalonil residues were lower by 14.8%, 22,7% and 10% in 2009, 2010 and 2011, respectively, whereas in the case of azoxystrobin, the residues were lower by 6.3%, 11.7% and 11.5% in the same years.

Discussion

Fungicide rate

The studies confirmed that when the environmental conditions were conducive to disease development, half dose of the fungicide was less effective in the protection of plants against PLB (Bain *et al.*, 2014; Ratajkiewicz *et al.*, 2016). The risk of disease development increased also as a result of lower fungicide rate inside canopy thus the present study showed that the tomato infestation by PLB was the highest in 2011. The chlorothalonil rate inside canopy estimated by its residue was the lowest in this year, resulting from the fact that the droplets could not adhere to the surface of the fruits which were covered by the leaves. This could be explained by the high-spreading tomato canopy habit at GS 75-82 in 2011. However, the systemic azoxystrobin residue did not depend on the characteristics of tomato canopy. Similar results were obtained when the fine spray produced by XR11003 nozzle was used (Ratajkiewicz *et al.*, 2016). In case of other crops, the characteristics of the plant canopy influenced the fungicide rate per leaf surface area unit and fungicide efficiency (Walklate *et al.*, 2003; Dammer *et al.*, 2008; Cooke *et al.*, 2011; Llorens *et al.*, 2011).

SVAM response

In the case of plants with high-spreading canopy and numerous elevated stems (GS 75-82), during the last treatments in 2011, a higher spray deposition was expected for PSV than for SV300, because according to Walklate & Cross (2013) spray deposition inside canopy should increase along with the increase of SV. The above has been indirectly proved on the basis of the residues of contact chlorothalonil in fruits. As the chlorothalonil residues were very similar and the SV applied on 1-ha significantly higher for the PSV system than the SV300 system, spray deposition on and near fruits had to be correspondingly higher for PSV. Despite the infestation of tomato leaves and fruits by PLB did not differ in favour of PSV even on plants sprayed with half of the fungicide dose. It could result from the fact that a coarse spray with IDKT12003 nozzle was applied. In the same conditions, the XR11003 nozzle, that allows for fine-droplet spraying, led to a decreased infestation of tomato with PLB on plots sprayed using the PSV system rather than SV300 (Ratajkiewicz *et al.*, 2016).

Significantly flattened tomato canopy with numerous uncovered fruits (GS≥85) in 2009 and 2010 contributed to obtaining lower average residue of chlorothalonil using PSV than SV300 after spraying with an IDKT12003 nozzle. The corresponding reduction in residue was 43.7% in 2009 and 18.2% in 2010. In the same conditions, the chlorothalonil residues when using an XR11003 nozzle were also lower for PSV than for SV300, on average by 36% in 2009 and 35% in 2010 (Ratajkiewicz *et al.*, 2016). It is worth noting that fruits located in the flattened tomato canopy were poorly covered by leaves and exposed to contact with droplets. Lower SV facilitated higher pesticide or a spray deposition outside of the plant canopy in other crops. The above conclusion was drawn from the studies conducted with the use of chlorothalonil (Bruhn & Fry, 1982; Hamm & Clough, 1999), as well as studies on spray deposition in potatoes (Gajtkowski *et al.*, 2005; Lipiński *et al.*, 2007; Kierzek & Wachowiak, 2009).

The effectiveness of systemic fungicides is far less dependent on SV applied on 1-ha and SV rate (Ratajkiewicz et al, 2009; Wise *et al.*, 2010). Current study show that the azoxystrobin residue did not depend on SVAM. Therefore, the application of azoxystrobin using an IDKT12003 nozzle could have led to a lack of differentiation between both SVAM in terms of infestation of tomato by PLB. To conclude, the same degree of protection of tomato against PLB using SV300 and PSV system resulted from alternate application of systemic and contact fungicides.

Nozzle response

Coarse-droplet spraying using double flat fan IDKT12003 nozzle resulted in a slightly higher residues of fungicides applied at full dose than in the case of fine-droplet spray (XR11003), with the exception of chlorothalonil in 2011. Despite the fungicide residue did not provide explanation for the infestation of plants by PLB which was slightly lower with the XR11003 nozzle in 2011 and 2009. It could have been connected with the coverage of surface with droplets which varied from nozzle to nozzle. We may assume that considering significant difference in $D_{V0.5}$ between the two nozzles, the droplets produced by IDKT12003 ($D_{V0.5}$ =433.6-462.3 µm) had far lower leaf and fruit coverage potential than in the case of XR11003 ($D_{V0.5}$ =221.9-230 µm). Many authors have noted that as a result of higher coverage of the surface, the fungicides, especially contact fungicides, were more effective (Grinstein *et al.*, 1997; Washington, 1997; Prokop & Veverka, 2006). However, own studies showed that spraying with an XR11003 nozzle (with fungicide at full dose) allowed for lower tomato leaf and fruit infestation, in particular when the plant canopy was still high (GS 75-82 in 2011). More numerous and finer droplets produced using XR11003 may have better penetrated the inside of such canopy, what was confirmed in studies on potatoes (Kierzek & Wachowiak 2009). At the end of the spraying season, in 2009, the canopy was flattened (GS ≥85) and easier to penetrate by less numerous droplets produced using IDKT12003. In these conditions, the average infestation of leaves and fruits by PLB was usually slightly lower for XR11003 than for IDKT12003, although the chlorothalonil residue with IDKT12003 was higher by 18% on average.

The studies seem to show that the distribution of fungicides, related to the droplet size distributions, may have influenced the durability of fungicides. It is possible that the active substance collected in larger deposits under droplets was less affected by UV radiation and might evaporate longer. Both physical factors have more adverse effect on the durability of chlorothalonil than azoxystrobin (Tomlin, 2000; Bartlett *et al.*, 2002; Lichiheb *et al.*, 2014). Higher average fungicide residues achieved with coarse spray than fine spray in case of plants with flattened canopies in 2009 and 2010 may support the above thesis.

On the other hand, higher fungicide deposit under the droplet could have resulted in improved systemic effectiveness of azoxystrobin. The ratio between the volume of the droplet and the contact surface of the leaves, which is higher in the case of bigger droplets, is correlated with the absorption and translocation of the systemic substance (Liu, 2003). In the case of azoxystrobin, the above statement can be supported by the fact that the residues after the application of 250 g of this active substance on 1-ha were in each year higher using coarse spray than fine spray.

To sum up, the IDKT12003 nozzle produced coarse spray was as beneficial to the effectiveness of fungicides in plants with flattened canopies as the fine-droplet XR11003 nozzle. Fine-droplet spraying of plants with high-spreading canopy contributed to lower infestation by PLB and higher fungicide residue than in the case of coarse-droplet spraying. Moreover, the IDKT12003 nozzle can provide better use of the potential of systemic azoxystrobin than the XR11003 nozzle.

Adjuvants

Fungicides with the addition of PMH had greater biological potential than the fungicide alone or with the addition of a multi-ingredient adjuvant. A positive impact of OS on the effectiveness of fungicides, including products used for protection of plants against PLB, was already known (Mitani *et al.*, 2001; Gent *et al.*, 2003). PMH, by increasing the coverage of the plant surface with liquid and the infiltration of cuticle, could have increased the fungicide effectiveness (Stevens *et al.*, 1991; Gent *et al.*, 2003). However, the PMH also led to a significant reduction of the average chlorothalonil residue in plants with flattened canopy in 2009 and 2010. It is possible that high interception of sunlight in the flattened tomato canopy could have accelerated the photodegradation of fungicide on the surface of the plant. The half-life of chlorothalonil due to photodegradation is relatively short and according to Monadjemi *et al.* (2011) equals 5.3 days. Additionally, faster evaporation of chlorothalonil, with vapor pressure of 0.076 mPa at 25 °C (Tomlin, 2000), may have occurred. In comparison with contact fungicide, the average azoxystrobin residue did not decrease that much under the influence of PMH, and in 2011 even slightly increased. Photodegradation could have had significant impact on the disappearance of azoxystrobin (Braunschweiler & Koivisto, 2000) which can explain, at least partially, the slightly increased reduction of residues in plants with flattened canopy in comparison to plants with high-spreading canopy. On the other hand, the same results can point to the positive role played by OS in the absorption of azoxystrobin (Gent *et al.*, 2003). This probably depended on age and physiological condition of the plants. It was previously known that the absorption is more intensive in younger parts of the plants (Bartlett *et al.*, 2002). The results obtained may point to the possibility of more intensive absorption of azoxystrobin in younger plants and in the presence of the surfactant under study. It seems that the addition of OS to the fungicide spray liquid applied using a coarse droplets has positive impact on the protection of tomato against PLB. In the case of previously published studies where fine-droplet spraying was conducted (Ratajkiewicz *et al.*, 2016) a significantly better effectiveness of fungicides applied with PMH against PLB was not found. However, the average infestation of plants by PLB during the last assessment every year was slightly lower when the fungicide suspension contained that surfactant.

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