

# Effects of sprayer operating parameters on airborne drift from citrus air-carrier sprayers

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**Abstract:** Florida citrus is mostly sprayed with various types of air-carrier sprayers. These sprayers differ substantially in design features and are normally operated at different volume rates and ground speeds, during day and night applications. The main objective of this study was to characterize drift potential (not total drift) of several commonly used citrus sprayers when operated under typical application conditions (different operating variables). Drift potential of the applications was assessed by capturing samples of airborne spray droplets with two high-volume air samplers, positioned above tree canopies at two sides of the spray course. For most applications, higher spray volumes (larger droplets) showed significantly reduced drift potential than lower volumes. Higher ground speed appeared to have more drift potential compared to lower speed but the effect of speed was not significant. Nozzles with comparatively lower flow rates (smaller droplets) were generally more drift-prone than the ones with higher flow rates (larger droplets) and spray from the upper nozzle bank had higher drift potential than spray from lower nozzles. These results are comparative and could show the importance of optimizing spray variables to reduce drift from typical citrus applications.

**Keywords:** spray drift, spray volume rate, sprayer ground speed, air sampler, fluorometry

**Citation:** Salyani, M., D. R. Miller, M. Farooq, and R. D. Sweeb. 2013. Effects of sprayer operating parameters on airborne drift from citrus air-carrier sprayers. *Agric Eng Int: CIGR Journal*, 15(1): 27–36.

## 1 Introduction

Air-carrier sprayers are the main type of spray equipment in Florida citrus applications (Salyani, 1997). They differ distinctively in size, shape, air delivery system, nozzle arrangement, and other features and are normally operated at different volume rates and ground speeds, during day and night applications (Whitney et al., 1986). Such sprayers are usually drift-prone and spray drift is a matter of concern in most citrus operations (Salyani and Farooq, 2004; Salyani et al., 2007). Apart from adverse effects of drifted pesticides on neighboring crops, animals, and surface water resources, the proximity of residential areas to citrus orchards has increased the

chance of public exposure to drifted pesticides. Therefore, the concern about spray drift has become more critical than ever and any effort to mitigate the problem is highly desirable.

For a given sprayer and its operating variables, the degree of spray drift mostly depends on droplet size and meteorological conditions (Bouse, 1994; Fox et al., 2000; Miller et al., 2000). Generally speaking, smaller droplets are more drift-prone than larger droplets and, at a constant pressure, nozzles with lower flow rates generate finer droplets (Womac et al., 1998). The British Crop Protection Council (BCPC) has proposed a spray classification system which divides the quality of spray into five categories (Doble et al., 1985; Van de Zande et al., 2000). These categories (very fine, fine, medium, coarse, and very coarse) provide an indication of drift potential for a given spray. Increasing spray droplet size by adjuvants can be effective in drift reduction (Sanderson et al., 1991;

**Received date:** 2012-10-24 **Accepted date:** 2013-01-24

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Salyani and Cromwell, 1993; Miller et al., 2001); however, Fritz et al. (2012) did not find a pronounced effect of adjuvant on droplet size in a wind tunnel study simulating high speed aerial applications (due to high shear rate). Air-inclusion nozzles, producing larger droplets for a given nozzle capacity, have been found effective in reducing ground losses but were not useful in eliminating downwind deposits (Derksen et al., 2000). In orchard applications, proper orientation of the sprayer air jet and matching spray volume to tree canopy size and shape can also reduce drift potential of orchard sprays to some extent (Holownicki et al., 2000; Van de Zande et al., 2002; Balsari et al., 2005).

In spraying the edge row of dwarf apple trees, Fox et al. (1990) reported that ground deposits of drifted spray decrease greatly beyond 120 m. Using ground and aerial sprayers in citrus, Salyani and Cromwell (1992) found that more than 70% of downwind ground deposit and airborne drift could originate from sprays applied to the last two rows of a grove. Studying the effects of spray volume and airflow rate on deposition, Pergher and Gubiani (1995) estimated the drift of vineyard sprays at 7% - 20% of the applied rate. For Florida citrus, spray drift was estimated at 6% - 14% (Salyani et al., 2007).

Several sampling methods have been adopted to quantify spray drift from various applications. Salyani and Cromwell (1992, 1993) used plastic sheets and high volume air samplers to assess ground fallout and airborne drift, respectively, at several distances from the spray line. Ganzelmeier (1993), Van de Zande et al. (2006), and a few other researchers have also sampled drift deposit at various downwind distances using absorbent or non-absorbent targets. Wind tunnel studies have shown that differences in the drift potential of various nozzles could be related to the quantity of the airborne spray (Southcombe et al., 1997). Richardson et al. (2000) and Salyani et al. (2007, 2009) installed vertical and horizontal sampling lines to quantify drift and ground losses from apple and citrus sprays, respectively. Fox et al. (2004) found that the collection efficiency of monofilament nylon screens depends on droplet size and air velocity. Miller et al. (2003) used a light detection and ranging (LIDAR) system to sample the drift cloud

generated from various citrus applications remotely. Balsari et al. (2005) assembled a sampling structure over two adjacent rows to assess spray mass balance in tree crop or vineyard applications. Vanella et al. (2011) used a special sampling device (drift test bench (Balsari et al., 2007)) to quantify drift potential of a citrus herbicide applicator.

In this study, high volume air samplers were deployed on two sides of the spray line to sample the airborne spray cloud of various applications. Wind tunnel studies by Southcombe et al. (1997) have shown that differences in the drift potential of various nozzles could be related to the quantity of the airborne spray. The main objective of this project was to compare drift potential (not total drift) of several commonly used citrus sprayers when they are operated under typical application conditions (different operating variables). The results could be used as a general guideline for reducing drift from typical citrus applications. Specific objective of the study was to determine the effects of spray volume rate (nozzle size), sprayer ground speed, and nozzle position on the relative magnitudes of airborne spray deposits from different applications. Koo et al. (2000), Farooq et al. (2003, 2005), and Salyani et al. (2002, 2006) have reported on deposition characteristics of these sprayers under various application conditions.

## 2 Materials and methods

The study involved five commonly used citrus air-carrier sprayers, including: Curtec<sup>®</sup> 648 (BEI Inc., South Haven, MI), Titan<sup>®</sup> 1093 and FMC 9100 (John Bean Sprayers, Hogansville, GA), DW AF500 (Durand-Wayland Inc., LaGrange, GA), and PowerBlast<sup>®</sup> (PB) 500 (Rears Manufacturing Co., Eugene, OR). Figure 1 shows the schematic views of the sprayers during the spray applications. Except for the Curtec, all other sprayers were equipped with a single axial-flow fan. The Curtec had three pairs of vertically stacked cross-flow fans which could be adjusted to conform to the shape of canopy boundary (Farooq et al., 2002). The Titan was equipped with an air tower attachment to discharge the spray along the tree height (Salyani et al., 2002). Both Curtec and Titan sprayers as well as the

FMC were engine-driven. The latter had a large fan with an elevated (modified radial) air outlet (Salyani and Whitney, 1991). DW and PB sprayers were PTO-powered and had conventional low profile radial outlets (Salyani and Hoffmann, 1996; Salyani and Farooq, 2003). These sprayers were equipped with various types/numbers of hydraulic nozzles or rotary atomizers (Table 1).

Spray solutions contained Pyranine-10G fluorescent dye (Keystone Aniline Corp., Chicago, IL) at tank concentrations of 250-300 mg L<sup>-1</sup>. They were applied to 4.5 - 5.5-m tall sweet orange trees at ground speeds of 2.4 or 4.8 km h<sup>-1</sup> and volume rates of 301- 4,381 L ha<sup>-1</sup> (based on 6.1 m row spacing) as different treatments (Table 1). The PB sprayer was operated with two fan types (4-blade/18° and 9-blade/32°) to obtain low (L) and high (H) airflow rates (11.4 and 16.4 m<sup>3</sup> s<sup>-1</sup>), respectively. Within each group of sprayer tests (Table 1), the treatments were applied in a randomized block design with five replications, except for the Curtec-night and PB tests which were made with three and four replications, respectively.

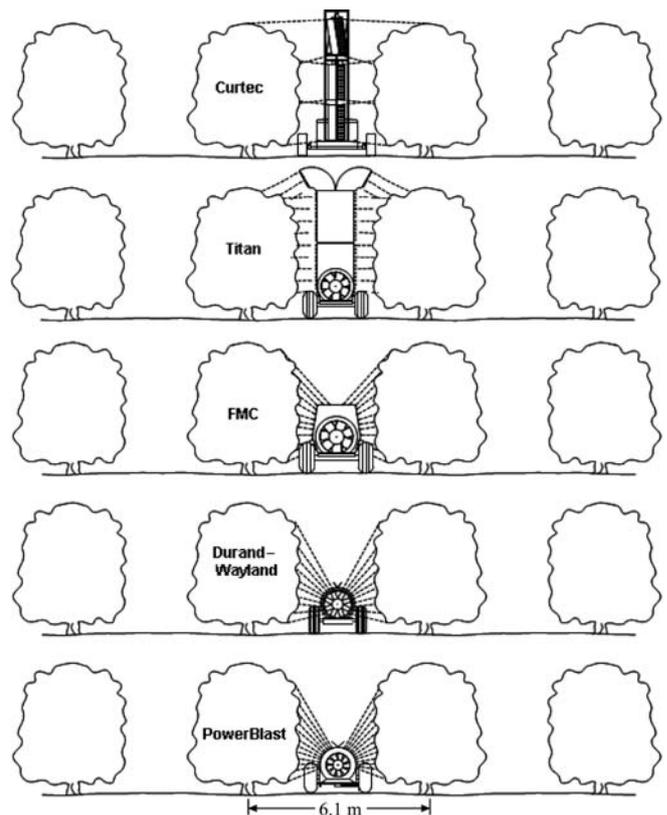


Figure 1 End views of the test sprayers (drawn to scale) during the spray applications

Table 1 Application variables for different sprayers

Sprayer	Treatment code <sup>a</sup>	Nozzle Size <sup>b</sup>	Nozzle No.	Nozzle Press /kPa	Fan Type <sup>c</sup>	Fan Speed /min <sup>-1</sup>	Fan Airflow /m <sup>3</sup> s <sup>-1</sup>	Flow rate /L min <sup>-1</sup>	Ground speed /km h <sup>-1</sup>	Volume rate <sup>d</sup> /L ha <sup>-1</sup>
Curtec <sup>e</sup>	C-LL	Rotary	2x6	*	Cr	*	*	14.7 (L)	2.4 (L)	602
	C-LH	Rotary	2x6	*	Cr	*	*	14.7 (L)	4.8 (H)	301
	C-HL	Rotary	2x6	*	Cr	*	*	41.0 (H)	2.4 (L)	1680
	C-HH	Rotary	2x6	*	Cr	*	*	41.0 (H)	4.8 (H)	840
Titan	T-LL	TXVK-6	2X34	*	Ax	*	*	47.4 (L)	2.4 (L)	1943
	T-LH	TXVK-6	2X34	*	Ax	*	*	47.4 (L)	4.8 (H)	971
	T-HL	DC 5/25	2X34	*	Ax	*	*	103.0 (H)	2.4 (L)	4221
	T-HH	DC 5/25	2X34	*	Ax	*	*	103.0 (H)	4.8 (H)	2111
FMC-1	F-3B	FDC 3/1	2x6	*	Ax	*	*	14.0	2.4	574
	F-3T	FDC 3/1	2x6	*	Ax	*	*	14.0	2.4	574
	F-6B	FDC 6/3	2x6	*	Ax	*	*	92.3	2.4	3783
	F-6T	FDC 6/3	2x6	*	Ax	*	*	92.3	2.4	3783
FMC-2	F-4L	FDC 4/2	2x12	*	Ax	1600 (L)	*	39.7	2.4	1627
	F-4H	FDC 4/2	2x12	*	Ax	2250 (H)	*	54.9	2.4	2250
DW	D-4L	DC 4/23	2x10	715 (L)	Ax	*	*	23.8	2.4	975
	D-4H	DC 4/23	2x10	950 (H)	Ax	*	*	27.3	2.4	1119
	D-5L	DC 5/25	2x10	715 (L)	Ax	*	*	40.9	2.4	1676
	D-5H	DC 5/25	2x10	950 (H)	Ax	*	*	46.9	2.4	1922
PB-1	P-LL	Lilac	2X12	*	Ax	*	11.4 (L)	14.7 (L)	2.4	602
	P-LH	Blue	2X12	*	Ax	*	11.4 (L)	106.9 (H)	2.4	4381
PB-2	P-HL	Lilac	2X12	*	Ax	*	16.4 (H)	14.7 (L)	2.4	602
	P-HH	Blue	2X12	*	Ax	*	16.4 (H)	106.9 (H)	2.4	4381

Nate: <sup>a</sup> Treatment codes: C=Curtec® 648, T=Titan® 1093, D=Durand-Wayland (DW) AF500, P=PowerBlast®; (PB) 500, F= FMC 9100, L=Low, H=High, B=Bottom six nozzles open, T=Top six nozzles open. <sup>b</sup> Nozzles: TXVK=Spraying Systems (SS) Co. conejet, DC=SS ceramic disc-core, Lilac/Blue = Albus; APT conejet, and FDC=FMC ceramic disc core. <sup>c</sup> Fan type: Cr= Cross-flow, Ax= Axial-flow. <sup>d</sup> Based on row spacing of 6.1 m. <sup>e</sup> Curtec sprayer treatments were repeated in night applications. \* Not pertinent to the experiments.

Drift potential of the applications was assessed by sampling the drifting spray cloud (accumulated over the measurement time) at the sampler location. It was accomplished by capturing airborne spray droplets with two high-volume air samplers (model TFIA, Staplex Co., Brooklyn, NY), using Staplex TFA41 filter papers. The samplers were located in the third row at two sides of the spray course. They were positioned above the tree canopy, at 7.3 m height with the filter surface facing the sky. Clean filter papers were loaded before each sprayer run and sprayed filters were collected about 2 - 4 min after stopping the spray. The samples were placed in sealable plastic bags, stored in a cooler, and later transported to the laboratory where they were stored in a refrigerator until analyzed. A few days after sample collection, spray deposits on the filters were quantified by fluorometry (Salyani, 2000). Earlier tests had shown negligible degradation of Pyranine-10G deposits under cold storage and short solar exposure (Salyani, 2003; Khot et al., 2011). Therefore, the captured spray (filter deposit) data, expressed as percentage of the applied rate of each treatment, were not corrected for the potential

minute dye degradation. Within each test, the data pertaining to the North and South samplers as well as their combined data were analyzed separately and the means were separated by the Duncan multiple range test. The significance of the differences was assessed at the 5% level.

Weather parameters including air temperature, wind velocity, and wind direction were recorded during the applications. Table 2 shows the average weather data for each treatment. The measurements were made with two 3-D sonic anemometers at 2.5 and 6.0 m above ground. The lower anemometer was located below the tree crowns in the gap between two adjacent trees whereas the upper anemometer was above the tree canopy. For those two levels, the stability parameter was expressed by unitless  $z/L$  and  $(z-d)/L$ , respectively; where  $z$  is height above the ground,  $d$  is the zero plane displacement due to the tree canopy, and  $L$  is the Monin-Obokov length (Miller et al., 2012). Figure 2 shows the schematic view of the test site and locations of the air samplers and meteorological instrumentation.

**Table 2 Meteorological data during spray applications**

Treatment code	Wind speed		Wind direction <sup>b</sup>	Air temperature		Stability parameter <sup>c</sup>	
	Top <sup>a</sup> /m s <sup>-1</sup>	Bot/m s <sup>-1</sup>		Top/°C	Bot/°C	Top	Bot
C-LL	2.535	0.695	67.5	19.2	20.2	-0.525	-0.533
C-LH	1.593	0.663	93.5	18.9	19.8	-0.359	-0.418
C-HL	2.563	0.807	104.5	18.5	19.3	-0.314	-0.163
C-HH	2.021	0.751	142.9	18.5	19.3	-0.274	-0.192
C-LL (N) <sup>d</sup>	2.226	0.308	73.7	18.6	18.3	0.675	0.415
C-LH (N)	1.247	0.308	88.6	18.4	18.1	1.060	0.514
C-HL (N)	1.152	0.274	71.5	18.3	17.9	0.988	0.383
C-HH (N)	1.308	0.304	71.5	18.2	17.8	0.966	0.690
T-LL	1.334	0.310	182.3	29.6	30.3	-6.320	-8.496
T-LH	1.024	0.250	306.2	29.8	30.4	-8.236	-3.222
T-HL	0.602	0.289	315.3	29.9	30.5	-11.537	-4.590
T-HH	1.036	0.373	311.6	29.8	30.4	-6.360	-2.355
F-3B	2.967	0.718	188.4	28.2	28.8	-1.027	-5.023
F-3T	2.281	0.593	198.4	28.4	29.0	-0.927	-3.819
F-6B	2.092	0.556	208.7	28.6	29.2	-0.850	-3.406
F-6T	1.751	0.621	163.0	28.7	29.3	-0.937	-1.978
F-4L	4.161	0.585	272.5	26.8	27.7	-0.236	-0.638
F-4H	4.253	0.606	236.0	26.6	27.4	-0.235	-0.349
D-4L	1.461	0.462	203.2	26.3	26.9	-2.776	-15.558
D-4H	1.475	0.465	167.5	25.9	26.5	-1.961	-12.473
D-5L	1.459	0.489	229.1	26.4	27.1	-2.392	-13.190
D-5H	1.332	0.445	156.6	26.5	27.1	-1.794	-11.116

Treatment code	Wind speed		Wind direction <sup>b</sup>	Air temperature		Stability parameter <sup>c</sup>	
	Top <sup>a</sup> /m s <sup>-1</sup>	Bot/m s <sup>-1</sup>		Top/°C	Bot/°C	Top	Bot
P-LL	1.461	0.318	49.7	22.9	23.2	0.008	-0.073
P-LH	0.902	0.331	29.6	22.8	23.2	0.052	-0.112
P-HL	2.585	0.585	69.5	25.1	-0.126	-1.049	-0.821
P-HH	2.262	0.627	73.4	25.4	-0.131	-0.926	-0.729

Note: <sup>a</sup> Top and Bot denote the data pertinent to the 3-D sonic anemometers at 6 m and 2.5 m heights.

<sup>b</sup> Direction of the winds coming from: North = 0°, East=90°, South=180° and West=270°.

<sup>c</sup> The stability parameters for the Top and Bot (above and below the tree canopy) are expressed by unitless (z-d)/L and z/L, respectively; where z is the anemometer height, d is the zero plane displacement due to the tree canopy, and L is the Monin-Obokov length (Miller et al., 2012). Negative and positive values indicate unstable and stable atmospheric conditions, respectively.

<sup>d</sup> Night-time applications with the Curtec sprayer during stable weather conditions.

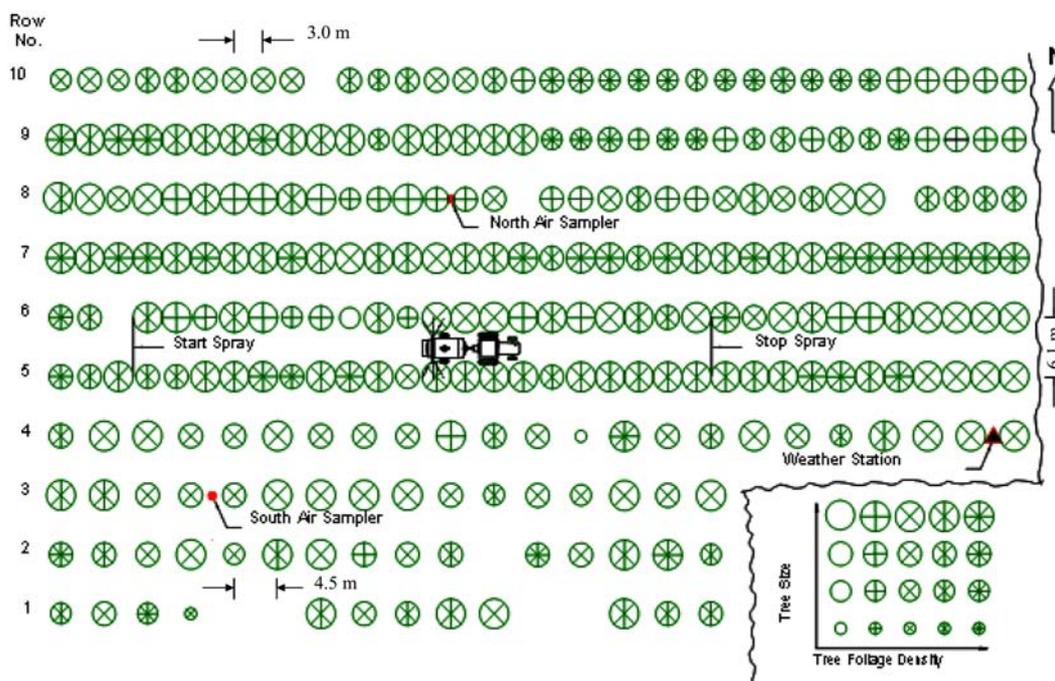


Figure 2 Schematic view of the test site, showing the locations of the air samplers and weather station

### 3 Results and discussion

For most tests, weather conditions were consistently unstable, as indicated by the negative stability parameter (SP) values of <-0.1 (Table 2). However, during the night-time application with the Curtec weather conditions became stable (SP >0.1), as expected. During the PB-1 sprayer tests stability conditions moved toward neutral and most of the replications were made under neutral stability conditions (-0.1 < SP < 0.1).

Figures 3 - 8 show the amounts of drifted droplet deposit on the filter of each sampler. These comparative deposits could provide a measure of drift potential (not total drift) from various applications. Evidently, the amounts of deposits on each sampler were mostly dependent on the prevailing wind direction. Usually,

winds coming from a northerly direction (N, NW, NE) gave higher deposits on the South sampler (downwind) compared to deposits on the North sampler (upwind) and vice versa. In Figures 3-8, the inset plots show the combined deposits of both samplers and the significance of the treatment variable effects.

In both day- and night-time Curtec applications, spray volume had a significant effect (\*) on the sampler filter deposition but the effect of ground speed was not significant (ns) (Figure 3). Overall, the higher volume, which involved larger droplets, gave less drift deposit than the lower volume (smaller droplets). Within each volume rate, faster ground speed (4.8 km.h<sup>-1</sup>) appeared to numerically increase the drift deposit of the applications to some extent. Visual comparison of the day and night plots indicated that the latter has generated more drift

deposits. This observation may be attributed to the difference in the atmospheric stability during those applications (Table 2). The more intense vertical mixing and dispersion of the drift cloud in day-time (negative SP, i.e., unstable conditions) has resulted in less droplet capture with the air sampler. This is due to the fact that droplets that move out (above the tree boundary layer) in stable conditions do not rise vertically more than a few meters whereas they may rise to much higher levels during convective conditions (Miller et al., 2003). Therefore, drift cloud concentrations above the canopy (i.e., at the sampler locations) will be higher at night than during the day-time. It should be noted that this comparison is not pertinent to the total amount of material that could potentially drift (flux) out of the orchard.

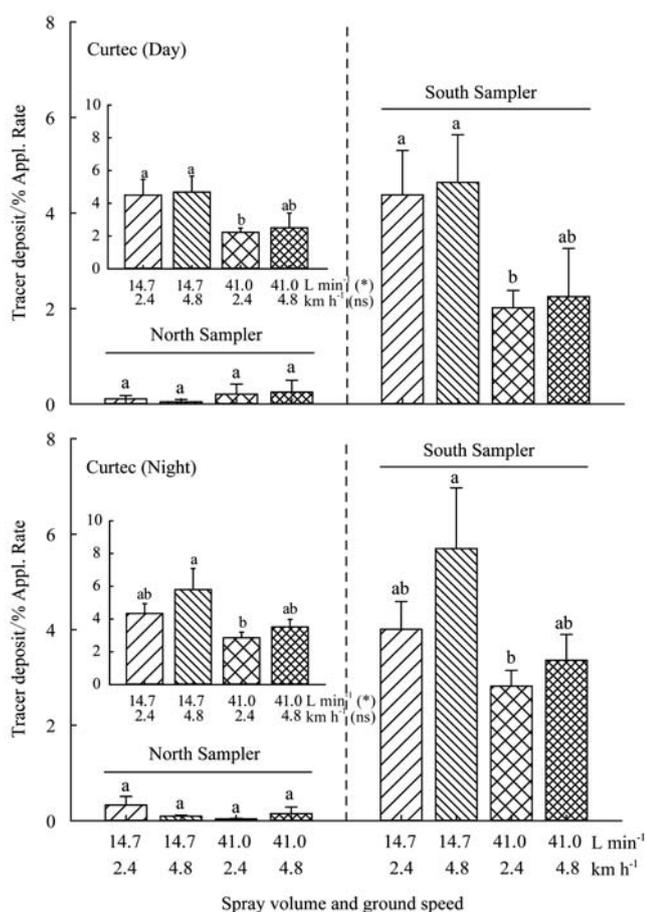


Figure 3 Deposit on the air sampler filters during the day (top) and night-time (bottom) Curtec® sprayer applications. The inset plots show the combined data of North and South samplers and the bars indicate standard error of the deposition

During Titan sprayer applications, both North and South samplers captured a substantial amount of tracer

deposits (Figure 4). Similar to the Curtec, spray volume (droplet size) had a significant effect on drift potential of the applications but the effect of ground speed was not significant. Higher volume (103.0 L min<sup>-1</sup>) reduced the drifted deposits significantly compared to the lower volume (47.4 L min<sup>-1</sup>). The higher drift deposit of the latter could be associated with the use of smaller droplets ( $D_{v0.5} = 134.6 \mu\text{m} @ 1,050 \text{ kPa}$ ) generated by TXVK-6 nozzles versus larger droplets ( $D_{v0.5} = 250.5 \mu\text{m} @ 700 \text{ kPa}$ ) of DC 5/25 nozzles during the higher volume rate applications. The effect of ground speed was more pronounced at the lower volume, i.e., ground speed of 2.4 km h<sup>-1</sup> resulted in significantly reduced drift deposit compared to 4.8 km.h<sup>-1</sup> applications (Figure 4 inset plot).

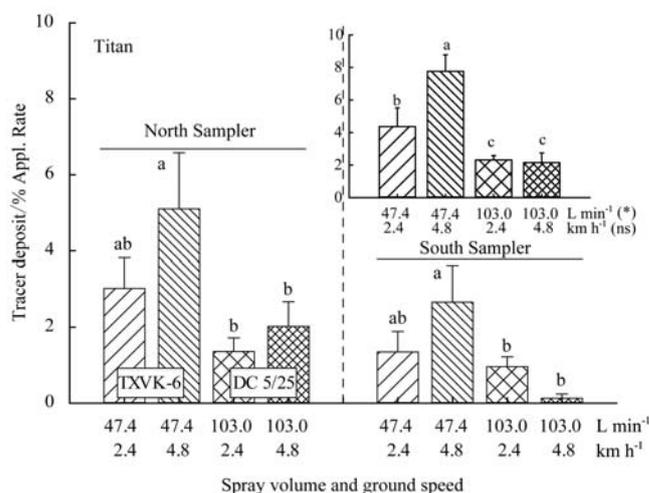


Figure 4 Deposit on the air sampler filters during the Titan® sprayer applications. The inset plot shows the combined data of North and South samplers and the bars indicate standard error of the deposition

In the FMC-1 test, the effect of spray volume (nozzle size) was not significant; however, nozzle position (top/bottom bank) showed a significant effect on sampler deposition (Figure 5). For both spray volumes (nozzle sizes), the placement of nozzles on the lower half of the manifold (bottom) decreased the drift deposit of the application as compared to the nozzle position on the upper manifold (top). The lower drift deposit with the former nozzle arrangement may be attributed to the reduction of droplet movement over the top of trees or to higher droplet deposition on the canopy. This result confirms the earlier results of Holownicki et al. (2000) and Van de Zande et al. (2002) and reveals the

importance of the proper nozzle arrangement in matching the sprayer output with the size and shape of the canopy. The FMC-2 test did not show a significant effect for spray volume and fan speed although there was somewhat reduced sampler deposit with the reduced fan speed (Figure 6).

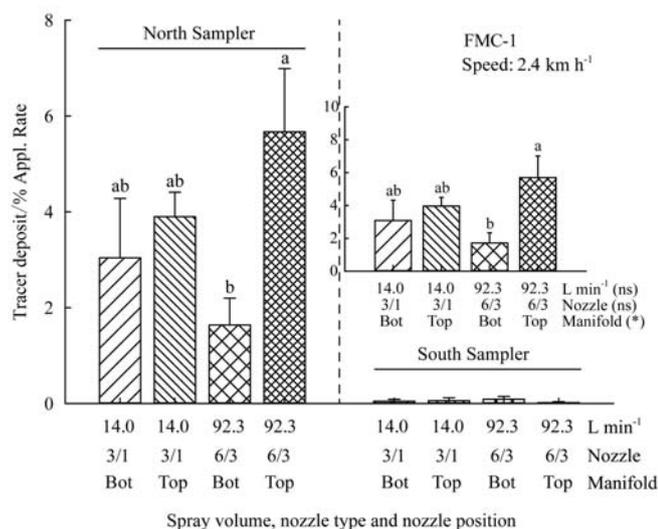


Figure 5 Deposit on the air sampler filters during the FMC-1 sprayer applications. The inset plot shows the combined data of North and South samplers and the bars indicate standard error of the deposition

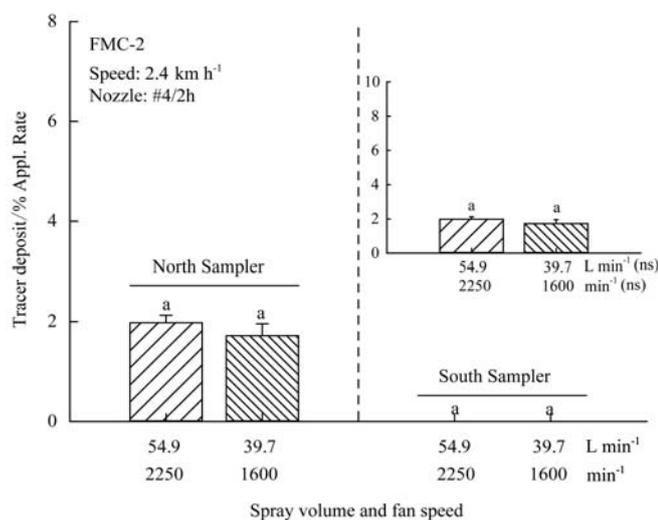


Figure 6 Deposit on the air sampler filters during the FMC-2 spray applications. The inset plot shows the combined data of North and South samplers and the bars indicate standard error of the deposition

With the Durand-Wayland sprayer (Figure 7), nozzle size (DC 4/23 versus DC 5/25) did not have a significant effect on drift deposit of the applications. Considering the volume median diameter ( $D_{v0.5}$ ) of their droplets

(187.1 and 250.5  $\mu\text{m}$  @ 700 kPa, respectively), the nozzle with a smaller orifice (DC 4/23) was expected to give somewhat higher drift deposit as has been reported earlier by Salyani and Farooq (2004). Although increasing nozzle pressure could ordinarily reduce the droplet size range, and thereby generate more drift-prone sprays, the corresponding increase in spray volume apparently had masked that effect and resulted in a lower percentage for the captured droplets.

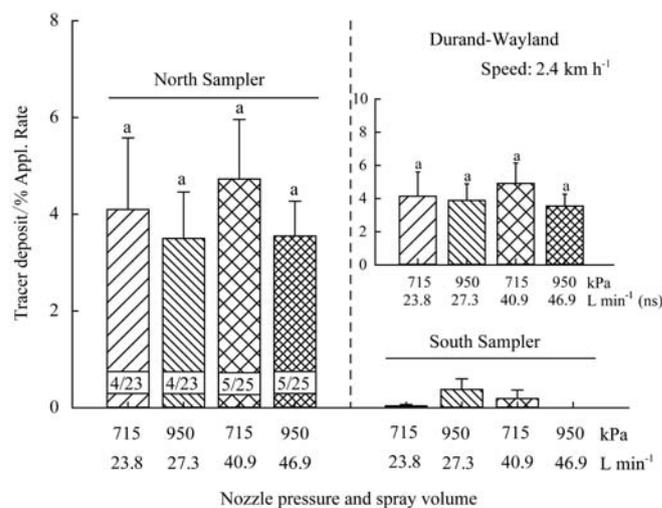


Figure 7 Deposit on the air sampler filters during the Durand-Wayland sprayer applications. The inset plot shows the combined data of North and South samplers and the bars indicate standard error of the deposition

During the PB tests (PB-1 and PB-2), using larger nozzles (Blue) versus smaller ones (Lilac) increased spray volume from 14.7 to 106.9 L min<sup>-1</sup> (Figure 8). This increase in spray volume resulted in significantly lower drift deposit from the applications as was the case with other sprayers. The trends were similar for both lower and higher sprayer airflow rates (11.4 and 16.4 m<sup>3</sup> s<sup>-1</sup>). Using this sprayer in an earlier study, Salyani and Farooq (2003) had found that higher sprayer air volume rates may not give significant increase in spray deposition or canopy penetration.

Overall, lower spray volumes and smaller droplets showed significantly more drift deposits than higher volumes. These results are consistent with the findings of Salyani and Cromwell (1992) and Cross et al. (2001) who reported higher airborne drift from lower spray flow rates used in citrus and apple orchards, respectively.

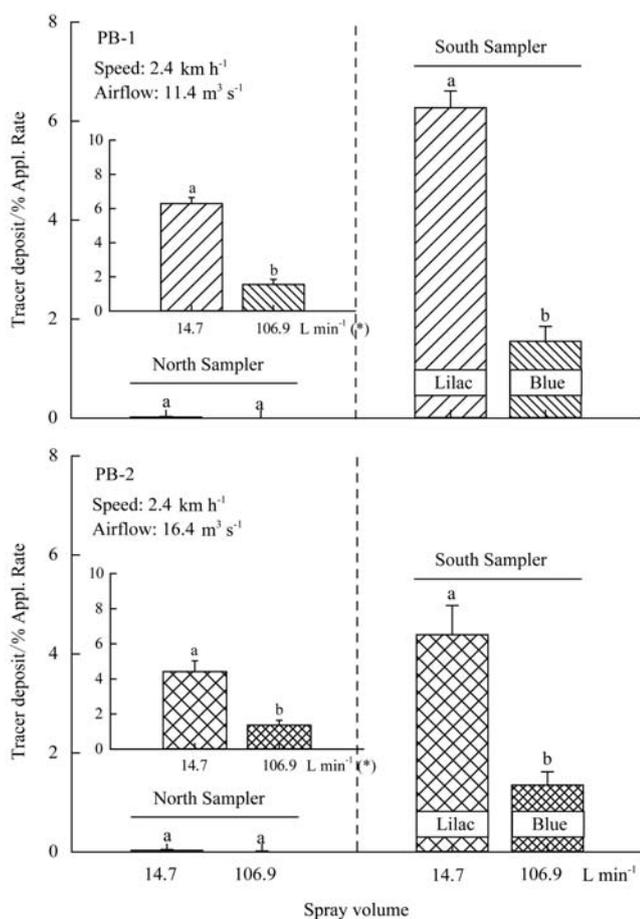


Figure 8 Deposit on the air sampler filters during the PowerBlast® sprayer applications with low airflow (top) and high airflow (bottom). The inset plots show the combined data of North and South samplers and the bars indicate standard error of the deposition

Higher ground speed appeared to have more drift potential compared to the lower speed but the effect of speed was not statistically significant. These results are in line with the report of Salyani et al. (2009), using similar airblast sprayer in citrus applications. Nozzles with smaller orifices, generating smaller droplet size ranges, were generally more drift-prone than larger ones as observed in the Titan sprayer test. Mounting nozzles on the upper manifold of the sprayer generated more drift deposits than those mounted on the lower manifold. Again, this result is in line with the findings of Holownicki et al. (2000). Finally, the effect of the

sprayer airflow rate on its drift potential was not clear in these tests.

## 4 Conclusions

These field studies revealed the drift potential (not total drift) of several sprayers, as they are typically used in Florida citrus applications. The comparative results could show the importance of optimizing spray variables in reducing drift from typical citrus applications. The following bullet points outline the conclusions.

- The results revealed measureable off-target movement of airborne spray droplets from all tested citrus applications.
- For the Curtec, day- and night-time applications gave similar drift deposit trends; however, the latter appeared to be more drift-prone when operated under stable weather conditions.
- Higher spray volumes, normally associated with larger droplets, resulted in lower drift potential with Curtec, Titan, and PB sprayers.
- Lower ground speed ( $2.4 \text{ km.h}^{-1}$ ) appeared to give less drift deposit than the  $4.8 \text{ km.h}^{-1}$  speed in Curtec and Titan applications.
- Nozzles mounted on the upper manifold of FMC sprayer gave more drift deposits than those mounted on the lower manifold.
- Nozzle pressure, fan speed, and sprayer airflow rate did not appear to affect drift potential of the DW, FMC, and PB sprays, respectively.

## Acknowledgements

This research was supported by the Florida Agricultural Experiment Station and a grant from Florida Citrus Production Research Advisory Council. The mention of trade names and commercial products is solely for providing specific information and does not imply any recommendation by the University of Florida and cooperating institutions.

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