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Author manuscript *J Expo Sci Environ Epidemiol.* Author manuscript; available in PMC 2019 October 01.

Published in final edited form as:

J Expo Sci Environ Epidemiol. 2019 April; 29(3): 397-405. doi:10.1038/s41370-018-0090-5.

## Real-Time Particle Monitoring of Pesticide Drift from an Axial Fan Airblast Orchard Sprayer

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#### Keywords

pesticides; particulate matter; environmental monitoring; exposure modeling; empirical models; statistical models

In Washington State, a majority of reported pesticide-related illnesses and applicationrelated complaints involve drift. We employed real-time particle monitors (Dylos) during a series of experimental spray events investigating drift. Sections of an orchard block were randomly sprayed by an axial fan airblast sprayer while monitors sampled particulate matter above and below the canopy at various downwind locations. We found elevated particle mass concentrations (PMC) at all distances (16-74 m). The 75<sup>th</sup> percentile PMC while spraying was significantly greater than the control periods by 107 (95% CI 94–121)  $\mu$ g/m<sup>3</sup>, after adjusting for sampler height and wind speed. The 75<sup>th</sup> percentile PMC below the canopy was significantly greater than above the canopy by 9.4 (95% CI 5.2–12)  $\mu$ g/m<sup>3</sup>, after adjusting for spraying and wind speed. In a restricted analysis of the spray events, the 75<sup>th</sup> percentile PMC significantly decreased by 2.6 (95% CI -3.2 - 1.7) µg/m<sup>3</sup> for every additional meter away from the edge of the spray quadrant, after adjusting for canopy height and wind speed. Our results were consistent with a larger study that performed passive sampling during the same spray events, suggesting that real-time monitoring can be used as a screening tool for pesticide drift. Compared to traditional methods of drift sampling, realtime monitoring is overall an easily employed, affordable sampling technique, and it can provide minute-by-minute measurements that can be coupled with meteorological measurements to better understand how changes in wind speed and direction affect drift.

Supplementary Information

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Conflict of Interest

The authors declare no conflict of interest.

Supplementary information is available at the Journal of Exposure Science & Environmental Epidemiology website (http://www.nature.com/jes).

## Introduction

Pesticide exposures among farm workers lead to more chemically-related injuries and morbidities than for any other workforce (1). In Washington State, most pesticide-related illnesses and application-related complaints involve drift and orchard airblast applications, making this a significant public health concern for agricultural communities (2). Many studies have focused on pesticide applicator exposures, but there has been less attention to occupational drift exposures in neighboring areas (3–6). Axial fan airblast sprayers (AFA) have been widely used in Washington tree fruit orchards since the 1950s (7). Over time, however, changes in tree shape and reduced tree heights have made this technology more likely to spray above the canopy, increasing the potential to produce spray drift and reducing the amount of pesticide that reaches the target crop. Spray drift is defined by the U.S. Environmental Protection Agency (U.S. EPA) as the movement of pesticide particles in air during or soon after an application to an unintended area (8). Some studies estimate that 45% of airblast spray misses the intended target and instead becomes drift or deposits at ground level (9,10). Recent studies have evaluated bystander and resident spray drift exposures, but have not focused specifically on occupational exposures (11-15). To the best of our knowledge, there is currently limited research looking at the real-time drift potential of AFA sprayers, despite their ubiquitous use in agriculture.

The U.S. EPA has recently added Application Exclusion Zones (AEZ) to the Worker Protection Standard (WPS) in an effort to limit pesticide drift (16). The AEZ requires that the immediate area surrounding pesticide application equipment be free of individuals not properly trained or equipped to handle pesticides during an application. The AEZ has been set to 100 ft (30 m) for airblast sprayers, though it is unclear how protective this buffer zone is in terms of reducing potential pesticide drift exposure.

The purpose of this study was to use real-time particle monitors to characterize the spray drift produced by a traditional AFA sprayer in order to evaluate this technology's potential to cause occupational pesticide drift exposures in a neighboring orchard block. Additionally, we wanted to describe drift differences above and below the canopy and with increasing distance from a sprayer. This study was nested in a larger study that measured micronutrient drift using accepted methods of passive sampling for spray drift characterization (17).

## Materials and Methods

#### Study design

This research took place in a Washington State University (WSU) research orchard on June 10, 2016 and September 28–30, 2016. The prevailing winds from the north in this area made it possible to sample for drift in a southern field. Two on-site meteorological stations collected wind speed, wind direction, temperature and relative humidity throughout our study period. The use of two meteorological stations followed applicable protocols for the International Organization for Standardization (ISO) and the American Society of Agricultural (and Biological) Engineers (ASAE) (18,19). The first was a permanent AgWeatherNet station, which was 2 meters high and 70 meters from the nearest corner (southwest) of the sprayed block (20). This regularly-maintained station took 5 second

meteorological measurements and integrated these into 15 minute summaries. This station's 15 minute wind direction and speed measurements were monitored to ensure that wind at the beginning of each spray event was blowing in the general direction of the neighboring field (wind rose direction from  $281-360^{\circ}$  or  $0-56^{\circ}$ ) and at wind speeds within the US EPA's drift-reducing recommendations of 1.3-4.5 m/s (3-10 MPH) (21). The second was a temporary station 10 meters high and 190 meters from the nearest corner (northeast) of the sprayed block. This station took more precise one-minute wind-speed and wind direction measurements which we used to afterwards control for wind speed and direction during spray events in our analyses. The study design is further detailed in Kasner et al., 2017 (17).

A 0.4 hectare (1 acre) orchard block (28 tree rows, each 49 m long) was divided into four quadrants that were randomly sprayed each day with micronutrients by a certified pesticide applicator using an AFA sprayer (Figure 1). We sprayed each of the four quadrants four times, which yielded a total of 16 "spray events." The sprayer was calibrated running at 1.6 km/hr (3 mi/hr) and outputting 935 L/ha (100 gal/ac). It used a 14 bar (205 PSI) operating pressure, approximately 566–850 m<sup>3</sup>/min (20,000–30,000 ft<sup>3</sup>/min) air volume flow rate, and disc-core nozzles (D3-D5 discs with size 25 cores) that created hollow cone spray patterns of fine droplets estimated to range from 110 to 125  $\mu$ m. We placed a global positioning system (GPS) data logger on the sprayer to verify its location and spray times. Spraying began early in the morning, between 8 AM and 10 AM, as is typical of agricultural work, and ended around noon.

We placed Dylos DC1100 Pro (Dylos Corporation; Riverside, CA) real-time optical particle monitors above and below the canopy at five distinct locations in a neighboring southern block with their inlets facing towards the spray field in order to sample for particulate matter (PM), including spray droplets, throughout our study period. Dylos monitors sampled four size measurements of PM (aerodynamic diameter greater than 0.5 µm, 1.0 µm, 2.5 µm and 10 µm) every second and integrated these into one-minute particle number concentrations (PNC). The samplers placed at a height of 2-meters were meant to capture potential worker exposure from drift below the top of the orchard canopy while the samplers placed at a height of 6-meters were meant to capture drift above the top of the orchard canopy. We aligned samplers at 11, 40 and 67 meters south of the spray field, measuring from the spray field's southernmost tree (Location A, C and E) to better characterize drift decay. Additional samplers were placed 21 meters east and west of sampler C to better capture overall drift (Location B and D). Samplers used a flow rate of 1,700 cm<sup>3</sup>/min (0.06 ft<sup>3</sup>/min) and ran from approximately an hour before the first spray of the day to an hour after the last spray of the day. During each spray day, each sampler collected one-minute observations during a "control period" which consisted of the 15 minutes preceding the first spray event and the 15 minutes following the last spray event. Each control period thus consisted of 15 one-minute observations.

#### **Data Analysis**

We used our temporary station's cup anemometer at three meters above ground for oneminute wind speed, temperature, relative humidity measurements, and the temporary station's ultrasonic anemometer 10 meters above ground for wind direction values since they

We used our on-site field measurements to calculate the length between each of our samplers and the central proximal edge of each spray quadrant, as is conventional in the drift literature (Figure 1) (18,19). Since the sprayer could travel each spray row within a minute, this length represented the shortest distance that the sprayer would have been from a sampler. For summary statistics, distances were categorized within our sampling range of 16–74 m into three categories: 16–33 m, 34–53 m and 54–74 m.

All of our data were analyzed using R (RStudio 0.99.903 using R 3.3.1; Boston, MA). We created four particle bin sizes ( $\mu$ m) (0.5  $b_1 < 1.0, 1.0$   $b_2 < 2.5, 2.5$   $b_3 < 10.0, b_4$  10.0) from the raw Dylos data. In order to adjust for diurnal trends in PNC, we calculated a fifth percentile, eight-minute rolling average of particle PNC that represented the changing ambient PM concentrations over time and subtracted these from raw PNCs to create flat baselines. An eight-minute rolling average was selected to account for our short spray durations, which lasted a similar length of time, and because it characterized steadily changing background concentrations well. The resulting departures from background represented short term transient changes in PNC attributable to our spray events. Similar methods of background adjustment for real-time air quality sampling of moving sources have been used in other studies (22,23). Using the equation below, we converted each bin's PNCs to particle mass concentrations (PMC) assuming an aerosol density () of 1 µg/m<sup>3</sup> and each bin's () geometric mean diameter (), except for the largest bin where the lowest size cut was used ( $d_1 = 0.71 \ \mu$ m,  $d_2 = 1.58 \ \mu$ m,  $d_3 = 5.00 \ \mu$ m and  $d_4 = 10.00 \ \mu$ m). All bin PMCs were added to estimate a total PMC (Supplementary Information Table S1).

$$PMC_{\text{Total}} = \sum_{b=1}^{4} \left( PNC_b x \frac{\pi}{6} x d_b^3 x \rho \right) \quad 1$$

We used a quantile regression in this analysis because it is a semi-parametric regression technique that has two advantages in this case, as compared to least-squares mean regression. First, we did not have to assume that the data was normally distributed. Second, we were able to investigate the shape of tails of the distribution by modeling the higher quantiles of the concentration distribution. Unlike least squares regression, a quantile regression allowed us to better look at spray drift peaks rather than the mean; was more robust at handling extreme values and outliers (which we expected to see); and had no distribution assumptions so we did not have to log-transform our data (24,25). Quantile regression has previously been used in Exposure Assessment studies in order to better understand the shape of the exposure distribution curve (26–28). Moreover, we calculated

geometric means for descriptive statistics because we expected our data to be right skewed. The R "quantreg" package was used while applying the Barrodale and Roberts algorithm for datasets of up to several thousands of observations, and a default tolerance parameter for convergence of the algorithm of  $10^{-6}$ . We performed a 75<sup>th</sup> quantile regression to predict PMC (µg/m<sup>3</sup>) using the following predictors: AFA spray periods (compared to nonspray, control periods), one-minute wind speeds (m/s), and sampler height above or below the canopy. Spray periods and sampler height were treated as categorical variables, whereas wind speed was treated as a continuous variable:

$$Q_{\Upsilon|X}(\tau = 0.75) = \beta_0 + X_{AFA}\beta_{\tau_{AFA}} + X_{windspeed}\beta_{\tau_{wind}speed} + X_{height}\beta_{\tau height}$$
 2

Furthermore, we completed a restricted analysis looking at the effect of sprayer distance (m) on PMCs, adjusting for wind speed and sampler height. Distance and wind speed were treated as continuous variables, whereas sampler height was treated as a categorical variable:

$$Q_{\gamma|X}(\tau = 0.75) = \beta_0 + X_{distance} \beta_{\tau_{distance}} + X_{windspeed} \beta_{\tau_{windspeed}} + X_{height} \beta_{\tau height} \quad 3$$

This analysis was restricted to spray periods where we had distance measurements between the active AFA sprayer and each sampler, and it excluded control periods when the AFA sprayer was not spraying.

#### Results

Ten Dylos samplers collected real-time samples during each spray event, with the following exceptions: 1) one sampler was not placed at location E below the canopy during the first three sprays due to a protocol modification; and 2) two samplers failed at locations B and D above the canopy during four other spray events. Moreover, the entire first spray event as well as some additional one-minute measurements were dropped from our analyses since they did not meet our wind direction inclusion criteria (see Methods, about 12% of our one-minute measurements). For 15 spray events we thus had 72 samples and 18 controls from below the canopy, as well as 67 samples and 19 controls from above the canopy, for a total of 139 spray samples and 37 controls (Table 1, Supplementary Information Table S2a,b). Each spray event lasted a mean of 7.0 (SD = 0.8) minutes. We had 493 and 535 one-minute spray observations above and below the canopy, respectively, for a total of 1,028 measurements. In addition, we had 404 and 445 one-minute control observations above and below the canopy, respectively, for a total of 849 measurements. Supplementary Information Figure S1 shows an example of a time series plot of one-minute PMCs during one of our study days.

We observed elevated arithmetic and geometric mean PMCs during spray events compared to the control periods for every sampling distance (16–74 m), with closer distances having the highest PMCs (Table 1). PMCs were also greater below the canopy than above the canopy. Compared to background (control) levels, the lowest geometric mean (GM) PMC

The median wind speed at the beginning of each spray quadrant was 3.5 (Interquartile range, IQR = 1.2) m/s with a minimum and a maximum wind speed of 2.6 m/s and 4.3 m/s, respectively (Table 2).

The 75<sup>th</sup> percentile of PMC during spray events was significantly greater than the control periods by 107 (95% CI 94–121)  $\mu$ g/m<sup>3</sup>, after adjusting for sampler height and wind speed (Table 3). The 75<sup>th</sup> percentile of PMC below the canopy was significantly greater than above the canopy by 9.4 (95% CI 5.2–12)  $\mu$ g/m<sup>3</sup>, after adjusting for spray events and wind speed. The 75<sup>th</sup> percentile of PMC significantly decreased by 2.0 (95% CI –2.8 – -0.9)  $\mu$ g/m<sup>3</sup> for every meter per second increase in wind speed, after adjusting for spray events and sampler height.

Our restricted analysis looking at the effect of distance on PMC showed that the 75<sup>th</sup> percentile of PMC significantly decreased by 2.6 (95% CI -3.2 - -1.7) µg/m<sup>3</sup> for every additional meter away from the central proximal edge of the spray quadrant, after adjusting for canopy height and wind speed (Table 4). The 75<sup>th</sup> percentile of PMC significantly increased by 76 (95% CI 36–114) µg/m<sup>3</sup> below the canopy compared to above the canopy, after adjusting for wind speed and distance. Finally, the 75<sup>th</sup> percentile of PMC significantly decreased by 24 (95% CI -34 - -15) µg/m<sup>3</sup> for every meter per second increase in wind speed, after adjusting for distance and canopy height.

## Discussion

Numerous studies have been conducted to characterize potential human exposure to drift from agricultural spraying (14,29–31). To our knowledge, however, this study is the first to use real-time particle monitoring to sample water aerosols to document agricultural spray drift. We used real-time instrumentation as an alternative to traditional methods of drift sampling that are more laborious, result in smaller sample sizes and provide limited information on variability. Our use of real-time instruments provided us with a large sample size that indicated substantial variability in spray drift and showed a clear distinction between control and spray periods.

We observed significantly higher PMCs during spray events compared to non-spray periods after adjusting for sampler height and wind speed. These results are in line with other studies that have reported poor target crop application accuracy by AFA sprayers (9,10,32). We also measured significantly greater PMCs at all of our measured distances (16–74 m), with closer distances having significantly higher PMCs. These results are in agreement with past studies that have found elevated pesticide concentrations closer to orchard blocks during and after applications (33).

We observed elevated PMCs at all of our measured distances, well beyond the US EPA's Worker Protection Standard (WPS) Application Exclusion Zone (AEZ) of 100 ft (30 m) established for airblast sprayers. Study conditions conformed to the application wind speeds recommended by the EPA of 1.34.5 m/s (3–10 MPH) (34). Below the canopy, given a

distance of 31 m from the central proximal edge of a spray field and wind speeds of 2.9 m/s, our model found a 4.3-fold increase in PMC above background. This finding indicates that drift can extend beyond 30 m under these study conditions. Moreover, had we measured distance from a more central point in the spray quadrant rather than the proximal edge, reported distances for PMCs would have been even greater. Our results thus suggest that the current AEZ may not completely protect workers from nearby drift, and that further work needs to be done to identify an appropriate AEZ for airblast applications.

Previous studies have found that orchard structures can greatly affect drift (35). In particular, vegetation barriers parallel to wind direction may funnel pollutants below the canopy downwind, creating what is known as the street canyon effect (36). In our study, spray below the canopy would have been directed towards our lower samplers in the neighboring field, while spray above the canopy would have dispersed more readily before reaching our samplers downwind. In line with this explanation, PMCs were significantly higher below the canopy than above the canopy when we adjusted for wind speed and spraying. These findings are particularly relevant for workers whose tasks are conducted below the canopy.

While higher wind speeds are a known contributing factor for drift (37), our results showed slightly lower PMCs at higher wind speeds. These results are consistent with a Gaussian plume (mass balance) model (38). They imply that the sprayer emission rate is constant, and that as wind speed increases, the amount of dilution also increases, leading to lower PMCs.

Our overall results were in concordance with our parent study that used accepted methods of passive sampling for drift characterization (17). These findings demonstrate the capability of the Dylos monitors to characterize drift and support their use more generally as exposure assessment tools. As low-cost monitors, these instruments could be deployed within a community to warn individuals of elevated PM levels when evacuation, ventilation, personal protective equipment or other safety measures may be required (39). Their data could be accessed remotely in real-time if they were customized (40). Customized Dylos could also be used to collect more frequent observations, rather than the one-minute summary measures that are recorded by default by this instrument in order to further characterizing quickly moving drift plumes. Moreover, the Dylos monitors could be used as indicators to perform further analyses with more sophisticated instrumentation.

This study had several limitations. First, we did not calibrate the instruments in our own laboratory, but rather used manufacturer-calibrated instruments to perform area sampling. High correlations, however, have been seen between manufacturer-calibrated Dylos DC1100 and three well-characterized reference instruments: the Grim 1.109 (Grim Technologies; Ainring, Germany) APS 3321 (TSI Inc.; Shoreview, MN) and FMPS 3091 (TSI Inc.) (41). Other studies have found strong correlations between the Dylos DC1700 (an updated monitor that uses the same sensor as the DC1100) and the DustTrak 8520, DustTrak II for PM<sub>2.5</sub>, and Sidepack AM510 (TSI Inc.) for PM<sub>2.5</sub> (42–44). These high correlations suggest that there is a predictable, linear relationship between the Dylos and other reference instrument readings, and that higher Dylos readings are indicative of higher reference method estimates. As noted earlier, the Dylos monitors could be used as screening tools prior to performing further analyses with more sophisticated instrumentation. Future studies

could deploy reference instrumentation alongside the Dylos as a quality control measure. It is also important to note that the Dylos monitors collected particles ranging from 0.5  $\mu$ m to "greater than" 10  $\mu$ m, making it difficult to more precisely determine the particle size distribution of larger particles.

Second, our samplers collected some extraneous PM, not just the AFA spray. In particular, dust, as well as water aerosols from early day humidity may have contributed to elevated PNCs. As indicated in the Methods section, we adjusted for these factors, and then compared our estimated PMC values during spray periods to PMC values during control periods. The elevated levels of PMC that we observed during spray periods can thus be attributed to AFA spraying.

Third, autocorrelation may have affected our results, since we would expect that spray events near one another in time would be most similar. We minimized this factor by randomizing our spray quadrants and by allocating time between spray events to allow for any potential residual drift to clear out. Because environmental conditions known to affect drift have predictable diurnal patterns (e.g., temperature and relative humidity), randomization was particularly important in our study since we started spraying at the beginning of each day when environmental conditions are known to be quickly changing (45).

## Conclusions

This study was nested in a larger study in which micronutrient drift was measured using accepted methods of passive sampling for drift characterization (17). In line with that study, we observed aerosol drift during AFA spray events at all of our measured distances from 16-74 m, with higher PMC levels below the canopy. These findings are particularly concerning for workers who may be in nearby orchard blocks or fields. They also demonstrate that the US EPA's 100-ft (30 m) AEZ for airblast sprayers may not be sufficiently protective under field conditions similar to those in this study. Our results underscore the value of US EPA efforts to support drift-reduction technologies, including those designed for orchard settings, and encourage well-designed empirical studies to determine their true potential to reduce drift. The US EPA's Voluntary Drift Reduction Technology Program encourages the development of driftreducing technologies but does not currently include orchards and is voluntary. Finally, real-time particle monitors appear to be useful screening instruments for drift. Compared to traditional methods of drift sampling, real-time monitoring is overall an easily employed, affordable sampling technique, and it can provide minute-by-minute measurements that can be coupled with meteorological measurements to better understand how changes in wind speed and direction affect drift.

## **Supplementary Material**

Refer to Web version on PubMed Central for supplementary material.

## Acknowledgements

This study would not have been possible without the Pacific Northwest Agricultural Safety and Health (PNASH) field team's (Pablo Palmandez, Maria Negrete, Maria Tchong-French, Jane Pouzou, Jose Carmona, Ryan Babadi, Christine Perez Delgado) expertise and time on this project. We would also like to thank Gwen A. Hoheisel from the Center for Precision & Automated Agricultural Systems at Washington State University (WSU) for her contribution to the design of this study. The WSU Tree Fruit Research & Extension Center, Washington Tree Fruit Research Commission and Vine Tech & Equipment were also involved in the logistics of this study.

This study was supported by the University of Washington's (UW) Department of Environmental & Occupational Health Sciences (DEOHS), including their Pacific Northwest Agricultural Safety and Health (PNASH) Center (CDC/NIOSH Cooperative Agreement #5 U54 OH007544), Medical Aid and Accident Fund Initiative, Award Number 5P30 ES007033–23 from the National Institute of Environmental Health Sciences, Award Number 83618501–0 from the US Environmental Protection Agency and Russel L. Castner Endowed Student Research Fund. UW's Graduate Opportunities Minority Achievement Program (GO-MAP) also supported this study.

FUNDING: This research would not have been possible without the support of the Graduate Opportunities and Minorities Achievement Program (GO-MAP); the University of Washington's Department of Environmental and Occupational Health Sciences (DEOHS); the Pacific Northwest Agricultural Safety and Health Center (PNASH; CDC/NIOSH Cooperative Agreement #5 U54 OH007544); the DEOHS Washington Medical Aid and Accident Fund (MAAF) Award; and the Russel L. Castner Endowed Student Research Fund.

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280 ft 84 m Page 30 of 35 Spray Quadrants ling wind from N 160 ft 49 m るなな 70 ft 21 m 37 ft З 40 ω 3 \$ D B 220 67 m # KEY **Sampling Location Sprayer Route** Distance

#### Figure 1.

Field setup diagram showing a northern orchard block with four randomly sprayed quadrants, and an unsprayed southern block that was used for sampling. Each sampling location had two Dylos collecting one-minute particle number concentrations (PNC) of four bin sizes (0–1.0  $\mu$ m, 1.0–2.5  $\mu$ m, 2.510  $\mu$ m, >10  $\mu$ m) throughout a spray day, above and below the canopy. Distance was measured as the length between each sampler and the central proximal edge of each spray quadrant. The dashed line shows an example of the

distance measurement between samplers at location A and quadrant 2 (16 m). Figure is not to scale.

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## Figure 2.

Boxplot using arithmetic mean PMC levels ( $\mu g/m^3$ ) above and below the canopy at increasing distances from the central proximal edge of the spray quadrant. PMC is on the log scale.

#### Table 1.

Mean one-minute PMC ( $\mu$ g/m<sup>3</sup>) per spray event above and below the canopy. Distances were grouped into three distance categories. Event samples are the total number of Dylos samplers within each distance category for all spray events (n = 15). One non-spray control period was assigned for each sampler (9–10) per sampling day (4).

Distance (m)	Distance (ft)	Samples	AM <sup>1</sup>	ASD <sup>2</sup>	GM <sup>3</sup>	GSD <sup>4</sup>
Above Canopy						
Control	Control	18	10	5.1	8.3	1.7
16 – 33	51 - 109	7	106	45	96	1.6
34 - 53	110 - 175	39	70	69	40	3.1
54 - 74	176 – 244	21	54	53	31	3.3
Below Canopy						
Control	Control	19	14	8.3	11	1.8
16 - 33	51 - 109	7	417	239	341	2.1
34 - 53	110 - 175	45	161	151	97	3.1
54 - 74	176 – 244	20	70	64	41	3.5
Total Control Samples		37				
Total Spray Samples		139				

<sup>1</sup>AM: arithmetic mean;

<sup>2</sup>ASD: arithmetic standard deviation;

<sup>3</sup>GM: geometric mean;

<sup>4</sup>GSD: geometric standard deviation.

Summary of wind speed measurements at the beginning of each spray event (n = 15). Q1: first quantile; Q3: third quartile.

Wind Speed	Min	Q1	Median	Mean	Q3	Max
m/s	2.6	3.0	3.5	3.6	4.2	4.3
MPH <sup>1</sup>	5.8	6.7	7.8	8.0	9.4	9.6

 $^{I}$ Wind speeds are provided in imperial units, as regulations in the United States are expressed in these units.

#### Table 3.

75th quantile regression of PMC ( $\mu$ g/m<sup>3</sup>) using the following predictors: spray events ("AFA Spraying") versus control periods, sampler height above or below the canopy ("Below Canopy"), wind speed per minute ("Wind (m/s)") (1,028 spray and 849 control one-minute observations). The intercept represents control periods, samplers above the canopy and no wind.

Covariate	Coefficient $(95\% \text{ CI})^{1}$		
(Intercept)	15 (11, 19)		
AFA Spraying	107 (94, 121)		
Below Canopy	9.4 (5.2, 12)		
Wind (m/s)	-2.0 (-2.8, -0.9)		

<sup>1</sup> 95% confidence interval

#### Table 4.

Restricted 75th quantile regression of PMC ( $\mu$ g/m<sup>3</sup>) during spray events using the following predictors: sampler height above or below the canopy ("Below Canopy"), wind speed per minute ("Wind (m/s)"), and sampler distance ("Distance (m)") (1,028 one-minute spray observations). This analysis excludes control period data (see Methods). The intercept represents samplers above the canopy, no wind and a 0 m distance.

Covariate	Coefficient (95% CI)		
(Intercept)	330 (264, 393)		
Below Canopy	76 (36, 114)		
Wind (m/s)	-24 (-34, -15)		
Distance (m)	-2.6 (-3.2, -1.7)		

<sup>1</sup>95% confidence interval