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Deposition and Air Concentrations of Permethrin and Naled Used for Adult Mosquito Management

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Abstract One of the most effective ways of managing adult mosquitoes that vector human and animal pathogens is the use of ultra-low-volume (ULV) insecticides. Because of the lack of environmental fate studies and concerns about the safety of the insecticides used for the management of adult mosquitoes, we conducted an environmental fate study after truck-mounted applications of permethrin and naled. One hour after application, concentrations of permethrin on cotton dosimeters placed at ground level 25, 50, and 75 m from the spray source were 2, 4, and 1 ng/cm^2 in 2007 and 5, 2, and 0.9 ng/cm^2 in 2008, respectively. One hour after application, concentrations of naled 25, 50, and 75 m were 47, 66, and 67 ng/cm^2 in 2007 and 15, 6.1, and 0 (nondetectable) ng/cm^2 in 2008, respectively. Deposition concentrations 12 h after application were not significantly different than 1 h after application for permethrin and naled either year. During 2007 and 2008 permethrin applications, two quantifiable air concentrations of 375 and 397 ng/m^3 were observed 1 h after application. In 2007 and 2008, naled air concentrations ranged from 2300 to 4000 ng/m^3 1 h after application. There were no quantifiable air concentrations between 1 and 12 h after application in either 2007 or 2008 for both naled and permethrin. Environmental concentrations observed in this study demonstrate that models used in previous risk assessments were sufficiently conservative (i.e., the models overestimated environmental concentrations). However, we also demonstrate inadequacies of models such as AgDrift® and AGDISP, which currently are used by the US Environmental Protection Agency to estimate environmental concentrations of ULV insecticides.

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West Nile virus (WNV) has now become endemic to North America and disease cases occur throughout the virus transmission season. Since the arrival of WNV, more areas of the country have been experiencing large-scale insecticide applications for mosquito-borne pathogens like WNV. To effectively manage infection rates, morbidity, and mortality due to mosquito-borne pathogens like WNV, there must be a reduction in contact between infected mosquitoes and humans and other nonhuman animals (Marfin and Gubler 2001). One of the most effective ways of managing high densities of adult mosquitoes that vector human and nonhuman animal pathogens is ultra-low-volume (ULV) aerosol applications of insecticides (Mount 1998; Mount et al. 1996). ULV utilizes small droplets from 5 to 25 μm in diameter, which are the optimum size to impinge on and knock down flying adult mosquitoes (Haile et al. 1982; Lofgren et al. 1973; Weidhaas et al. 1970).

Since the majority (>60%) of the American public is not concerned about the threat of WNV (Ho et al. 2007), there has been greater public attention on the human-health and environmental risks associated with ULV insecticide applications (Peterson et al. 2006; Roche 2002; Thier 2001). In response to these concerns, risk assessments have been performed to quantify reasonable worst-case estimates of risk. Peterson et al. (2006) performed a reasonable worst-case human-health risk assessment for six active ingredients used for mosquito management, including permethrin ([3-phenoxyphenyl]methyl 3-[2,2-dichloroethenyl]-2,2-dimethylcyclopropane carboxylate) and naled (1,2-dibromo-2,2-dichloroethyl dimethyl phosphate), after

truck-mounted ULV applications. Schleier et al. (2009b) performed a probabilistic human-health risk assessment of the same insecticides as Peterson et al. (2006) and supported their findings that the risks after mosquito adulticide treatments most likely would not exceed regulatory thresholds. Davis et al. (2007) and Schleier et al. (2008a) examined ecological risks for permethrin and naled as well and found similar results.

Currently, there are no publicly available data concerning the terrestrial deposition of naled and there are only two studies examining the terrestrial deposition of permethrin after truck-mounted ULV applications (Knepper et al. 1996; Pierce et al. 2005). In addition, there are no publicly available studies that have measured air concentrations of permethrin and naled after truck-mounted ULV applications. Knepper et al. (1996) measured concentrations of permethrin on turf grass ranging from nondetectable (ND) to 14 ng/cm^2 after truck-mounted ULV application in a suburban neighborhood. Pierce et al. (2005) measured concentrations of permethrin ranging from 0.05 to 5 ng/cm^2 in the Florida Keys National Marine Sanctuary, 2–4 h after application. Jensen et al. (1999) found no detectable concentrations of permethrin in water samples from wetlands before and after truck-mounted ULV applications. The majority of the studies examining the fate of insecticides after truck-mounted ULV applications have been performed with malathion (*O*,*O*-dimethyl dithiophosphate of diethyl mercaptosuccinate). These studies have found that 1–22% of the insecticide sprayed settled onto the ground within 500 m from the spray source, with the amount substantially decreasing 24–36 h after application (Moore et al. 1993; Tietze et al. 1994, 1996).

Currently, very little is known about the deposition and drift of small droplets like those utilized in ULV (Teske et al. 2000). Although risk assessments have been performed, they have relied on estimates of environmental concentrations of adulticides from models that do not have algorithms for ULV application methods. These models are designed for industrial and agricultural applications, not ULV. In addition, the probabilistic risk assessment of Schleier et al. (2009b) demonstrated that the estimated deposition and air concentration of insecticides are contributing the largest amount of variance to exposure estimates. Because of the concerns about the safety of insecticides used for the adult mosquito management and because of the lack of actual environmental concentration data and uncertainties associated with the fate of the ULV insecticides, we conducted an environmental fate study in 2007 and 2008. The objectives of our study were to characterize terrestrial surface residues and air concentrations of permethrin and naled after truck-mounted ULV applications.

Materials and Methods

The study occurred near Cascade (N47°13.489′, W111°42.040′) and Ulm (N47°25.402′, W111°29.767′), Montana, USA during the summers of 2007 and 2008, respectively. Because of considerations of cost and logistical issues, naled and permethrin were applied on separate evenings. Therefore, four field experiments were conducted: one for naled and one for permethrin each year.

The applications took place in open fields with no vegetation taller than 20 cm, to represent a worst-case assessment of ground deposition and air concentrations. Within each study site, surface residue and air concentration samples were taken. Surface residue sample collectors were placed 25, 50, and 75 m from the spray source and air concentration samplers were placed 25 m from the spray source (Fig. 1). There were three sample replicates with 200-m buffer zones between replicates (Fig. 1).

Insecticides were applied by a truck equipped with a Bison (VecTec Inc., Orlando, FL, USA) ULV generator. The source of permethrin was Permanone® 10% EC (Bayer Environmental Science, Research Triangle Park, NC, USA) mixed 1:1 with BVA oil (BVA Inc. Wixom, MI, USA) applied at the maximum application rate of 7.85 g/ha with a flow rate of 205 mL/min. The source of naled was Trumpet® EC (AMVAC, Los Angeles, CA, USA) applied undiluted at the maximum application rate of 22.42 g/ha with a flow rate of 44.36 mL/min. Truck speed was 16.1 km/h and spraying began and ended 100 m on each side of the sample collectors (Fig. 1). Applications occurred when the prevailing wind was blowing perpendicular to the collection site. Temperature, wind speed, and relative humidity at 1.5 m above ground level were recorded with a Kestrel® 4000 pocket weather tracker (Nielsen-Kellerman, Boothwyn, PA, USA).

In 2007, permethrin was applied on August 12 at 2030 h MDT, and naled was applied at 2020 h on August 27. Winds were out of the northeast at 8 km/h, with wind gusts to 17.7 km/h at the time of the permethrin application. Wind speed was 2.4 km/h out of the north, with wind gusts to 4.8 km/h at the time of the naled application. Average temperature and relative humidity were 22°C and 35% and 21°C and 33% at the time of the permethrin and naled applications, respectively.

In 2008, permethrin was applied at 2015 h on July 25 and naled was applied at 2000 h on August 12. Winds were out of the northeast at 7 km/h, with wind gusts to 12.9 km/h at the time of the permethrin application. Wind speed was 8 km/h out of the southwest, with wind gusts to 12.9 km/h during the application of naled. Average temperature and relative humidity was 27°C and 27.5% and 24°C and 23% at the time of the permethrin and naled applications, respectively.

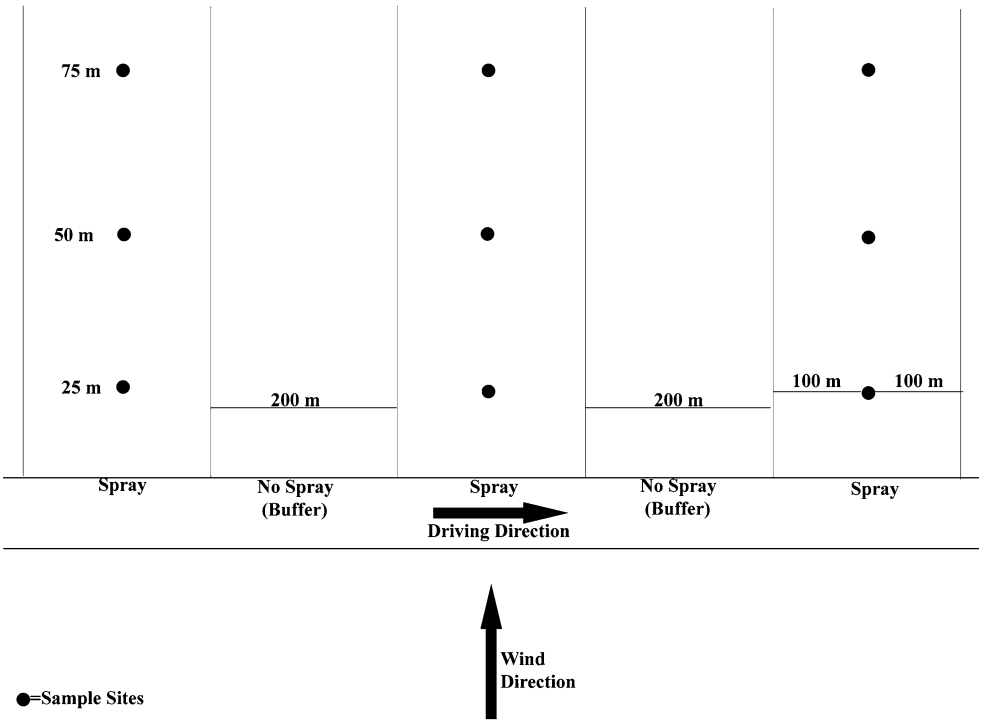

Fig. 1 Site layout with wind direction, spray zone, buffer zones, driving direction, and sampling locations for each application

Collections of surface residues at ground level and 1.25 m above the ground were taken on 10 cm × 10 cm (100 cm^2) cotton dosimeters pinned to a piece of cardboard (Ross et al. 1990). The cotton dosimeters at 1.25 m were perpendicular to the ground. Cotton dosimeters were separated by 15 cm at the edges of the dosimeters on the piece of cardboard. The cardboard was covered with plastic wrap to prevent contact between the cardboard and dosimeters. Before each application, dosimeters were placed 25 m from the spray line at ground level 1.5 h before spraying and were collected just before applications began.

For permethrin, two dosimeters were pinned on each piece of cardboard at ground level before the application and were collected 1 and 12 h after application. At 25 m, a third dosimeter was placed on the board before the application and collected at 24 h after application. At 1.25 m above the ground, three dosimeters were pinned on each piece of cardboard before the application in the treated areas and were collected 1, 12, and 24 h after application. Three blank and three spiked dosimeters were placed in a control (untreated) area, with one of each collected at 1, 12, and 24 h after application. For naled, two dosimeters were pinned on each piece of cardboard at ground level and 1.25 m above ground level before the application and were collected 1 and 12 h after application. Two blank and two spiked cotton pads were placed in a control area for naled and one of each was collected 1 and 12 h after application.

The control areas were located where no spraying or drift could occur, but they were subject to the same meteorological conditions as the residue samples. For the positive controls, cotton pads were dosed with 750 ng of technical-grade insecticide.

Cotton dosimeters were collected with tweezers. The tweezers were rinsed with pesticide-grade acetone between dosimeters to prevent cross-contamination. Individual samples of naled and permethrin were stored in separate 60-mL I-Chem™ glass jars with Teflon® lids (Chase Scientific Glass, Vineland, NJ, USA). Jars were placed in a cooler with dry ice for transport from the field site to the lab. Jars were stored in a freezer at $< 4°C$ to prevent degradation of insecticide until analysis (Lewis 1999).

Air samples were continuously drawn by an SKC® Model 224-PCXR4KDB universal pump (SKC Inc. Eighty Four, PA, USA) through an SKC sorbent polyurethane foam (PUF SKC Catalog No. 226-92) cartridge for both permethrin and naled. Polyurethane foam cartridges were kept in aluminum packing during transport to and from the field. The cartridges were removed from the aluminum package and attached to the pump via a 1.5-m piece of flexible plastic tubing. The flow rate was set at 2 L/min in 2007 and 5 L/min in 2008. The flow rate was adjusted in 2008 to sample more air because of the high percentage of ND concentrations the previous year. Cartridges were 1.25 m off the ground and attached to a stake. After the 1- and 12-h sampling periods, PUF cartridges were wrapped in the original aluminum foil and placed in a cooler on dry ice for transport to the lab. PUF cartridges were stored in a freezer at $< 4°C$ until analysis (Lewis

1999). One field blank and one spiked PUF for each collection time were treated exactly the same as the other cartridges, except no air was pulled through the cartridges (Lewis 1999). The PUFs were spiked with 750 ng of insecticide and were treated like the field blanks.

Extraction of all samples occurred within 7 days of sampling to prevent degradation (Lewis 1999). PUF cartridges were removed from their glass containers using tweezers and cut into six pieces and placed into a 60-mL I-Chem glass jars with Teflon lids. Dosimeters and PUF cartridges were extracted using 45 mL of high-pressure liquid chromatography-grade hexane (Fischer Scientific, Pittsburgh, PA, USA). Jars were placed on a shaker table for 2 h. A 13-mL aliquot was concentrated to 1 mL using nitrogen evaporator at 30–35°C. Extraction efficiency and recovery corrections were performed in the laboratory using methyl chlorpyrifos (*O*,*O*-dimethyl *O*-3,5,6-trichloro-2-pyridyl phosphorothioate).

Chemical analyses were performed by the Montana State Department of Agriculture's Chemical Analytical Laboratory in Bozeman, Montana, USA. The reported detection limits were 30 ng for *cis*- and *trans*-permethrin and 1.5 ng for naled for both dosimeters and PUFs. Quality assurance measures included the analysis of reagent blanks, matrix blanks (dosimeters and PUFs), duplicates, and spiked samples (laboratory and field).

Permethrin analysis was performed on an Agilent 6890 gas chromatograph–electron capture detector (Agilent Technologies, Inc., Santa Clara, CA, USA) equipped with a Restek RTX-5 column with Intraguard (30 m × 0.25 mm × 0.25 μm) (Restek U.S., Bellefonte, PA, USA). The temperature program used to separate *cis*- and *trans*-permethrin started at 60°C, increased at 25°C/min to 280°C, and held at that temperature for 4 min. Naled analysis was performed using an Agilent 5973 gas chromatograph–mass spectroscopy detector equipped with a Restek RTX-5 column (30 m × 0.25 mm × 0.25 μm). The temperature program for naled started at 80°C, increased at 20°C/min to 280°C, and held at that temperature for 2 min.

BoxCox transformations were performed on the raw data to determine appropriate transformation. We used Statistical Analysis System 9.1 (SAS Institute, Cary, NC, USA) to run repeated measures analysis of variance ($\alpha = 0.05$; PROC GLM) on log-transformed concentrations to determine differences between times, distances, heights, and year sprayed.

For ND concentrations in the deposition data, we substituted half of the detection limit when the number of NDs was less than 10% of the data points (Lubin et al. 2004). If the number of NDs was greater than 10% of the data, they were quantified using the maximum-likelihood estimate (MLE) (Helsel 2005). The MLE was determined using

$$\mathrm{NDC} = \exp\left(\mu_{\ln} + \sigma^2_{\ln}/2\right), \quad (1)$$

where NDC is the mean concentration for nondetects, μ is the mean of detected concentrations, and σ^2 is the variance of the detected concentrations (Helsel 1990, 2005).

Results

Deposition

Permethrin was not detected on the control or background dosimeters. Recovery of methyl chlorpyrifos ranged from 78% to 110%, and recovery of both field and laboratory spikes for *cis*- and *trans*-permethrin ranged from 94% to 130% in both 2007 and 2008. There was no significant difference between concentrations at ground level and at 1.25 m above the ground ($F = 0.11$, $p = 0.74$), so data for the two heights were combined for analysis (Table 1). There was a significant difference between the 2007 and 2008 concentrations ($F = 5.95$, $p = 0.02$) and a significant effect of distance from the spray source ($F = 16$, $p < 0.0001$; Table 1). Concentrations at 24 h were significantly lower than concentrations measured 1 h ($F = 5.72$, $p = 0.02$) after application. However, concentrations at 12 h were not significantly different from the 1-h samples ($F = 0.67$, $p = 0.41$).

There were no detectable concentrations of naled on the control or background dosimeters. Recovery of methyl chlorpyrifos ranged from 64% to 108%, and recovery of both field and laboratory spikes of naled ranged from 107% to 130% in both 2007 and 2008. There was a significant difference between concentrations at ground level and

Table 1 Mean permethrin deposition 25, 50, and 75 m from the spray source in ng/cm^2 ± standard error 1, 12, and 24 h after application in 2007 and 2008

Distance	2007	2008
1 h		
25 m	2.3 ± 1.2	4.6 ± 0.67
50 m	3.8 ± 1.1	2.3 ± 1
75 m	1.1 ± 0.63	0.94 ± 0.18
12 h		
25 m	2 ± 1.2	3.7 ± 0.69
50 m	3.1 ± 0.46	1.3 ± 0.39
75 m	0.8 ± 0.5	0.86 ± 0.2
24 h		
25 m	1.8 ± 1.2	3.9 ± 0.78
50 m	1.3 ± 0.4	0.7 ± 0.32
75 m	0.2 ± 0.1	0.42 ± 0.36

1.25 m above the ground ($F = 4$, $p = 0.05$; Tables 2 and 3). There was a significant difference between the 2007 and 2008 concentrations ($F = 20.42$, $p < 0.0001$). There also was a significant effect of distance from the spray source ($F = 19.83$, $p < 0.0001$). Concentrations at 1 h were not significantly different than concentrations measured 12 h after application ($F = 0.45$, $p = 0.51$).

Air Concentrations

In 2007, there was one quantifiable air concentration of 375 ng/m^3 1 h after the permethrin application. In 2008, there was one quantifiable air concentration of 397 ng/m^3 and one detection below the limit of quantification measured 1 h after the permethrin application. In 2007, there was one quantifiable air concentration of 3910 ng/m^3 1 h after application, and in 2008 there were three quantifiable concentrations of 2300, 2900, and 4000 ng/m^3 1 h after the naled applications. There were no quantifiable air concentrations measured between 1 and 12 h after application in either 2007 or 2008 for either permethrin or naled.

Discussion

Deposition of permethrin and naled did not decrease significantly from 1 to 12 h after application, which is similar to what has been observed with aerial applications of ULV insecticides for adult mosquito management (Schleier et al. 2008b). This is most likely because the main route of degradation for both permethrin and naled is photolysis (USEPA 2002, 2006) and all field applications were after 2000 h. In addition to photolysis, volatilization off the dosimeters could also result in a decline of residues for permethrin and naled. There were no detectable air concentrations of permethrin or naled between 1 and 12 h after application. These results suggest that the insecticide movement exceeded sample collectors within 1 h after application. Additionally, air exposures to ULV insecticides might be limited to within 1 h after application.

Table 2 Mean naled deposition at ground level 25, 50, and 75 m from the spray source in ng/cm^2 ± standard error 1 and 12 h after application in 2007 and 2008

Distance	2007	2008
1 h		
25 m	47 ± 0.10	15 ± 2.9
50 m	66 ± 9.6	6.1 ± 2.1
75 m	67 ± 11	ND[a]
12 h		
25 m	51 ± 6.7	20 ± 2.1
50 m	74 ± 7	7.7 ± 2.9
75 m	71 ± 5.8	0.57 ± 0.56

[a] Nondetectable concentrations

Table 3 Mean naled deposition 1.25 m above the ground 25, 50, and 75 m from the spray source in ng/cm^2 ± standard error 1 and 12 h after application in 2007 and 2008

Distance	2007	2008
1 h		
25 m	11 ± 2.2	23 ± 5
50 m	6.5 ± 1.5	13 ± 5.4
75 m	4.8 ± 3.7	0.54 ± 0.53
12 h		
25 m	9.7 ± 1.2	14 ± 1.2
50 m	4.9 ± 0.57	12 ± 3.3
75 m	5.2 ± 3.9	1.6 ± 0.95

The concentrations generally decreased as distance from the spray source increased, which is similar to the finding of Knepper et al. (1996), but in 2007 the concentration of permethrin was greater at 50 than at 25 m. Additionally, ~ 4.4% of the permethrin and 28% of the naled sprayed settled onto the ground 25 m from the spray source. The significant difference between ground level and 1.25 m above the ground for naled, but not permethrin, and the higher concentrations of naled settling onto the ground could be due to the heavier oil used in the formulation of Trumpet. In the present study, we did not assess the collector efficiency of the samplers located 1.25 m above the ground; thus, our measured values could be underestimating the actual amount of insecticide that deposited (Duan et al. 1994; Miller et al. 1992). Previous studies of truck-mounted ULV applications have found 1–22% of the insecticide sprayed settled onto the ground (Knepper et al. 1996; Moore et al. 1993; Tietze et al. 1994, 1996; Tucker et al. 1987). Knepper et al. (1996) measured ~ 3 ng/cm^2 of permethrin at 15 min after application 30.5 m from the spray source in a domestic setting. Their results and the results of Pierce et al. (2005) were similar to what we measured 1 h after application at 25 m in 2007 and 2008.

Previous risk and regulatory assessments have used models like ISCST3 (http://www.epa.gov/scram001/tt22.htm#screen), AERMOD (http://www.epa.gov/scram001/dispersion_prefrec.htm#aermod), AgDrift® (Stewart Agricultural Research Services, Macon, MO, USA) (Teske et al. 2002), and AGDISP (http://www.continuum-dynamics.com/pr-agdisp.html) (Bilanin et al. 1989) to estimate environmental concentrations of insecticides (Davis et al. 2007; Gosselin et al. 2008; Macedo et al. 2007; Mickle et al. 2005; Peterson et al. 2006; Schleier et al. 2008a, 2009a, b; USEPA 2008; Valcke et al. 2008).

Using the modeling assumptions of Peterson et al. (2006), the estimated environmental concentrations using ISCST3 at 25 m for permethrin and naled were 31 and 6.4 times greater, respectively, than what were measured in this study. The values predicted by ISCST3 were 1.7 times greater than what was found in other studies with permethrin (Knepper et al. 1996; Pierce et al. 2005). The estimated environmental concentrations using AERMOD at 25 m for permethrin and naled were 21 and 5 times greater than the highest air concentrations measured in this study.

Using the model assumptions of Schleier et al. (2008a), concentrations estimated by AgDrift at 25 m from the spray source for permethrin and naled were one-eighth and one-twelfth, respectively, of what was measured in the field. The US Environmental Protection Agency (USEPA) used AGDISP to estimate environmental concentrations of permethrin based on the model inputs of Mickle et al. (2005) for the reregistration eligibility document of permethrin (USEPA 2006). Deposition concentrations estimated by AGDISP at 25 m using the assumptions outlined by Mickle et al. (2005), and weather conditions and application rate for the permethrin sprays in 2007 and 2008 were one-sixth and one-fourth, respectively, of what was measured in the present study.

Current and past environmental fate studies and existing models demonstrate that a separate model needs to be developed that can more accurately predict concentrations of ULV applications. Based on our measurements of environmental concentrations of permethrin and naled, both ISCST3 and AERMOD were sufficiently conservative models for conducting lower tiered risk assessments (Davis et al. 2007; Macedo et al. 2007; Peterson et al. 2006; Schleier et al. 2009a, b); however, AgDrift and AGDISP seem to be underestimating environmental concentrations and, thus, risks.

The year effect most likely indicates that environmental conditions significantly affect the deposition of permethrin and naled. Concentrations of naled differed drastically between the two years, which is most likely due to the difference in wind speed between years. A more extensive study with additional temporal and spatial replicates should be conducted to examine the effect of wind speed, air temperature, humidity, and other factors that influence the behavior of ULV aerosols. The results of our study demonstrate that a more accurate model needs to be developed so that federal, state, and local officials can more accurately estimate the risks of ULV insecticides for use in regulatory documents and communications with the public.

Beyond constructing more accurate models for ULV insecticides, the actual environmental concentrations measured from this study could be used to refine previous risk assessments, which were based on estimated environmental concentrations. Concentrations of permethrin and naled measured in the current study were lower than what Peterson et al. (2006), Davis et al. (2007), and Schleier et al. (2009b) estimated using ISCST3 and AERMOD. This suggests that exposures and concomitant human-health and ecological risks from truck-mounted ULV applications most likely are lower than what was estimated by previous assessments. However, the use of either AGDISP or AgDrift to estimate environmental concentrations of insecticides after ULV applications could result in an underestimation of exposures and, thus, risks associated with adult mosquito management.

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