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Environmental fate model for ultra-low-volume insecticide applications used for adult mosquito management

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HIGHLIGHTS

- ► No model can accurately predict deposition of insecticides applied with ultra-low-volume technology for mosquito management.
- ► We perform field studies to measure actual environmental concentrations of insecticides.
- ► We develop a validated model to predict the deposition of insecticides after ultra-low-volume applications.
- ► The model demonstrated good predictive ability and was validated.

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ABSTRACT

One of the more effective ways of managing high densities of adult mosquitoes that vector human and animal pathogens is ultra-low-volume (ULV) aerosol applications of insecticides. The U.S. Environmental Protection Agency uses models that are not validated for ULV insecticide applications and exposure assumptions to perform their human and ecological risk assessments. Currently, there is no validated model that can accurately predict deposition of insecticides applied using ULV technology for adult mosquito management. In addition, little is known about the deposition and drift of small droplets like those used under conditions encountered during ULV applications. The objective of this study was to perform field studies to measure environmental concentrations of insecticides and to develop a validated model to predict the deposition of ULV insecticides. The final regression model was selected by minimizing the Bayesian Information Criterion and its prediction performance was evaluated using *k*-fold cross validation. Density of the formulation and the density and CMD interaction coefficients were the largest in the model. The results showed that as density of the formulation decreases, deposition increases. The interaction of density and CMD showed that higher density formulations and larger droplets resulted in greater deposition. These results are supported by the aerosol physics literature. A *k*-fold cross validation demonstrated that the mean square error of the selected regression model is not biased, and the mean square error and mean square prediction error indicated good predictive ability.

1. Introduction

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 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 virus-impacted animals (Marfin and Gubler, 2001).

One of the more effective ways of managing high densities of adult mosquitoes that vector human and animal pathogens is ultra-low-volume (ULV) aerosol applications of insecticides (Mount, 1998; Mount et al., 1996). Ultra-low-volume applications utilize small droplets from 5 to 25 μm, which are the optimum size to impinge on and cause knock down of flying adult mosquitoes through intoxication (Haile et al., 1982; Lofgren et al., 1973; Weidhaas et al., 1970).

Ground-based ULV applications used for adult mosquito management are very different than agricultural pesticide applications because the nozzles produce an aerosol (droplets <100 μm) and are pointed at a +45° angle from the horizon. Ultra-low-volume applications used for adult mosquito management are most effective when the insecticide remains airborne and moves through the target area; in contrast,

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applications for agricultural pests are designed to minimize the movement of droplets (Hiscox et al., 2006). Droplet spectra for ULV applications used during adult mosquito control operations have a volume median diameter (VMD) between 8 and 30 μm (VMD<30 μm) and 90% of the droplet spectrum should be smaller than 50 μm (VMD 0.9<50 μm). The droplet spectrum used for adult mosquito management is well below those classified as "very fine" to "fine" (VMD<137 μm) by the American Society of Agricultural Engineers, which is considered to be a high drift hazard (Hewitt, 2008; Teske et al., 2000).

Little is currently known about the deposition and drift of small droplets such as those used during ULV applications for adult mosquito management (Teske et al., 2000). Droplets smaller than 50 μm have very low settling velocities, and have similar transport characteristics to those of gaseous mixtures (Thistle, 2000). Currently, there is no validated model that can accurately predict deposition of insecticides applied using ULV technology for adult mosquito management.

Computer models of pesticide drift are widely used tools by regulatory agencies for predicting the deposition of spray particles beyond the intended target area (Felsot et al., 2011). The U.S. Environmental Protection Agency uses different models and assumptions to assess the risks of ULV insecticides (USEPA, 2002, 2006a, 2006b, 2006c, 2006d, 2006e, 2006f). Previous risk and regulatory assessments have used models like ISCST3, AgDrift® (Stewart Agricultural Research Services, Macon, MO, USA) (Teske et al., 2002), and AGDISP (Bilanin et al., 1989) to estimate environmental concentrations of insecticides (Davis et al., 2007; Macedo et al., 2007; Peterson et al., 2006; Schleier et al., 2008, 2009a, 2009b; USEPA, 2008). The ISCST3, AERMOD, AgDrift, and AGDISP models use a steady-state Gaussian plume algorithm, and are applicable for estimating anthropogenic compound concentrations from point, area, and volume sources with coarse droplet sizes and applications that are 10 to 100 m above ground level.

A reliable model that can predict environmental concentrations of ULV insecticides is needed because previous probabilistic risk assessments have shown that the deposition of the insecticide contributes the largest amount of variance to the estimated exposure (Schleier et al., 2009a, 2009b). In addition, a model is needed because of the limited amount of knowledge about which environmental and physicochemical factors have the largest effect on the movement of pesticide aerosols.

Because of public concerns about the safety of adulticides used for the control of adult mosquitoes (Peterson et al., 2006; Roche, 2002; Thier, 2001), the lack of actual environmental concentration data (Schleier and Peterson, 2010), and uncertainties associated with the fate of the ULV insecticides, we conducted environmental fate studies during the summers of 2009 to 2011 in California, Montana, and Louisiana to develop a predictive model for ULV insecticide deposition. For wide applicability, we validate the model with respect to predictive ability and a range of environmental variables.

2. Materials and methods

Ground-based ULV field experiments were conducted near Elk Grove, California (38°27′17.27″N, 121°27′9.25″W), Bozeman, Montana (45°38′ 47.09″N, 111°24′8.18″W), and Baton Rouge, Louisiana (30°31′1.57″N, 91° 9′20.32″W) during the summers of 2009 to 2011. Sites with little vegetative structure and a flat topography were chosen for all experiments because vegetation affects air movement and subsequent deposition of insecticides and we were interested in high depositions for conservative estimates of exposure. Sites were 200 m long with two lines of horizontal drift collectors positioned 25 m to the left and right of the center of the plot to capture any variability of deposition within the spray plot (Fig. 1). Because the two lines of deposition samplers are sub-samples they were averaged together at each distance from the spray source for statistical analysis. During each spray event, 11 receptors on the two sampling lines were placed in the field at different distances from the spray source (Fig. 1). Sampling occurred at distances of 5, 10, 15, 20, 30, 35, 40, 50, 60, 65, 70, 75, 80, 90, 95, 100, 110, 120, 125, 130, 135, 140, 155, 160, and 180 m from the spray source.

During all applications, the truck speed was 16.1 km/h. Applications occurred when the prevailing wind was blowing perpendicular to the collection site (Fig. 1). Sprays were conducted using a Guardian 95 ES (ADAPCO, Sanford, FL, USA) in Montana and a London Fogger model 18 (London Fog Inc., Long Lake, MN, USA) in California and Louisiana. Nozzle orientation of the sprayers was a +45° angle compared to the horizon which is the most commonly used angle for mosquito management. Between each spray replication the nozzle, pump, and hoses were rinsed with 300 ml of D.I. H_2O followed by 300 ml of a 1:1 mixture of high pressure liquid chromatography acetone (99.7% purity; EMD Chemicals, Gibbstown, NJ, USA) and American Chemical Society (ACS) grade toluene (99.5% purity, Mallinckrodt Baker, Inc., Phillipsburg, NJ, USA) (Schleier et al., 2010).

The oil-based insecticides Permanone® 30–30 (30% permethrin), Scourge® 18 + 54 (18% resmethrin), Permanone® 31–66 (31% permethrin) (Bayer Environmental Science, Research Triangle Park, NC, USA), Zenivex® E20 (20% etofenprox) (Central Life Sciences®, Schaumburg, IL, USA), and Pyronyl™ Crop Spray (6% pyrethrins) (Prentiss Inc., Alpharetta, GA, USA) were used. The water-based formulations Aqua-Reslin® (20% permethrin) (Bayer Environmental Science, Research Triangle Park, NC, USA) and Aqua-Kontrol (20% permethrin) (Univar®, Redmond, WA, USA) were used. The active ingredients were applied at the maximum rate of 7.85 g/ha of active ingredient according to label for all insecticides, except for Pyronyl Crop Spray which was applied at the maximum rate of 2.8 g/ha of active ingredient.

The experimental design was completely randomized with each formulation randomly selected for the order it was sprayed. Replications were performed over time within the same night and over different nights with a total of 96 spray events occurring during the three field seasons. Applications began no earlier than 18:00 h at all locations, but most applications occurred after 20:00 h.

Between June 21 and 26, 2009 a total of nine and eight sprays of Aqua-Reslin and Permanone 31–66 occurred in California, respectively. Aqua-Reslin was mixed 1:1.5 with deionized (D.I.) H_2O and applied at the flow rate of 240 ml/min. Permanone 31–66 was mixed 1:0.25 with ACS grade toluene and applied at a flow rate of 74 ml/min.

Between July 16 and August 5, 2009, a total of 13, 12, and 4 sprays of Aqua-Reslin, Permanone 30–30, and Scourge 18 + 54 occurred in Montana, respectively. Aqua-Reslin was mixed 1:1 with D.I. H_2O and applied at a flow rate of 192 ml/min. Permanone 30–30 was mixed 1:2:1 with Crystal Plus 70 T light mineral oil (STE Oil Company, Inc., San Marcos, TX, USA) and ACS grade toluene and applied at a flow rate of 192 ml/min. Scourge 18 + 54 was mixed at 1:0.4:0.4 with Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min.

Between June 7 and 22, 2010 a total of seven sprays of Aqua-Reslin and Pyronyl Crop Spray occurred in California, respectively. Aqua-Reslin was mixed 1:1 with D.I. H_2O and applied at a flow rate of 192 ml/min. Pyronyl Crop Spray was mixed at 1:0.2 with ACS grade toluene and applied at a flow rate of 163 ml/min.

Between July 19 and August 12, 2010, a total of eight, seven, two, seven, and six sprays of Aqua-Reslin, Permanone 30–30, Scourge 18 + 54, Zenivex E20, and Aqua-Kontrol occurred in Montana, respectively. Aqua-Reslin was mixed 1:1 with D.I. H_2O and applied at a flow rate of 192 ml/min. Permanone 30–30 was mixed 1:2:1 with Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min. Scourge 18 + 54 mixed at 1:0.4:0.4 to Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min. Zenivex E20 was mixed 1:0.4:0.4 with Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min. Aqua-Kontrol was mixed 1:1 with D.I. H_2O and applied at a flow rate of 192 ml/min.

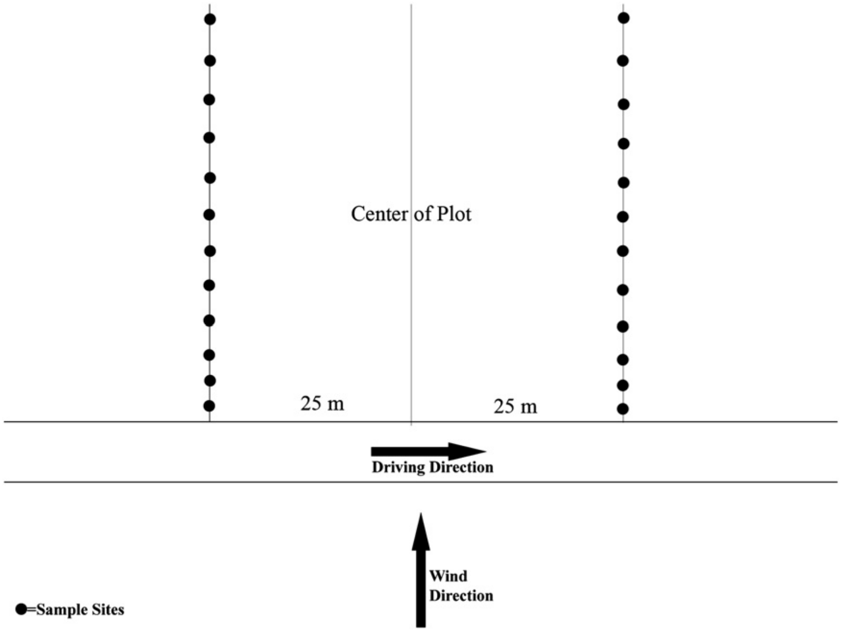

Fig. 1. Site layout for the 11 receptors that were selected from distances of 5, 10, 15, 20, 30, 35, 40, 50, 60, 65, 70, 75, 80, 90, 95, 100, 110, 120, 125, 130, 135, 140, 155, 160, and 180 m from the spray source.

Between July 17 and 28, 2011, a total of two and five sprays of Aqua-Reslin and Permanone 30–30 occurred in Louisiana, respectively. Aqua-Reslin was mixed 1:1 with D.I. H_2O and applied at a flow rate of 192 ml/min. Permanone 30–30 was mixed 1:2:1 with Crystal Plus 70 T light mineral oil and ACS grade toluene and was applied at a flow rate of 192 ml/min.

Between July 19 and August 12, 2011, a total of two, two, one, and four sprays of Aqua-Reslin, Permanone 30–30, Permanone 31–66, Zenivex E20 occurred in Montana, respectively. Aqua-Reslin was mixed 1:1 with D.I. H_2O and applied at a flow rate of 192 ml/min. Permanone 30–30 was mixed 1:2:1 with Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min. Permanone 31–66 mixed 1:2.25:1 to Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min. Zenivex E20 was mixed 1:0.4:0.4 with Crystal Plus 70 T light mineral oil and ACS grade toluene and applied at a flow rate of 192 ml/min.

Collection of deposition at ground level was with 15-cm diameter glass Petri dishes (Sundaram et al., 1987). Cardboard was staked to the ground to provide a level surface for the glass Petri dishes. One glass Petri dish was placed on each piece of cardboard at ground level and collected 10 min after the application. Fluorescent tracers such as Fluorescein and Tinopal OB have been commonly used in agricultural spray drift tests to determine the concentrations of insecticide (Barber and Parkin, 2003; Cadogan et al., 2005; Cai and Stark, 1997; Cooke and Hislop, 1993; Davis and Elliott, 1953; Longley et al., 1997; Parkin and Merritt, 1988; Peng et al., 2005; Pergher, 2001; Sharp, 1974, 1976; Staniland, 1959; Sundaram and Sundaram, 1992; Yates and Akesson, 1963). The oil-soluble tracer Tinopal OB (BASF Corp., Florham Park, NJ, USA) was mixed with all oil-based formulations at a rate of 11 g/l and the water-soluble tracer Fluorescein (Aqua Solutions, Deer Park, TX, USA) was mixed with all water-based formulations at a rate of 14 g/l. Previous research has shown that the addition of fluorescent tracers to ULV pesticide formulations does not alter the density, viscosity, or droplet spectrum of ULV insecticides (Schleier et al., 2010). For all applications, a maximum of 1-L of formulation was mixed with the tracer in a Nalgene 1-L narrow mouth high density polyethylene bottle (Fischer Scientific, Pittsburgh, PA, USA) to ensure that the tracer was adequately mixed into the formulation.

Petri dishes were extracted with 15 ml of toluene or D.I. H_2O for Tinopal OB and Fluorescence, respectively. The liquid was decanted into analysis vials which were wiped with KimWipes (Kimberly-Clark® Global Sales, LLC, Roswell, GA, USA) to remove fingerprints and dry the vials before analysis. After extraction, Petri dishes were rinsed with toluene or D.I. H_2O depending on the tracer used then rinsed with high pressure liquid chromatography acetone.

A GFL-1A fluorometer (Opti-Sciences, Inc., Hudson, NH, USA) was used to detect the amount of light absorbed at a specific wavelength which represented the amount of tracer present in the sample. For the detection of Fluorescence, the source filter was 465 nm and the detection filter was 530 nm. For the detection of Tinopal OB, the source filter was 370 nm and the detection filter was 430 nm. Standard curves were prepared using serial dilutions prepared in toluene and D.I. H_2O for Tinopal OB and fluorescence, respectively. Absorbance was recorded for each sample representing the concentration of the insecticide.

The detection limit was determined by measuring the absorbance of 20 blank samples and adding two standard deviations to the mean. The detection limit for Tinopal OB and Fluorescence is 0.00012 and 0.000015 $\mu g/cm^2$, respectively. Therefore the detection limit for water- and oil-based insecticides was 0.00076 and 0.0002 $\mu g/cm^2$, respectively. For non-detectable (ND) concentrations in the data, we substituted half of the detection limit because the number of NDs was less than 10% of the data points (Lubin et al., 2004).

On August 4, 2010 in Montana we sprayed Aqua-Reslin and Permanone 30–30 three consecutive times over the collectors to determine if our technique was correctly measuring the fluorescent tracers. The three sprays were applied by spraying the 200-m block turning the truck around and spraying the site two more times in the same method using the same application parameters used during 2010 listed above. These results demonstrated that the concentration of the tracers was three times greater than a single spray in similar environmental conditions.

A DC-III portable droplet measurement system (KLD Labs, Inc., Huntington Station, NY, USA) was used to measure droplet spectra of each formulation at all locations. The DC-III probe was held 2 m from the nozzle in the center of the spray plume and sampling was terminated at 15 s or when 10,000 droplets were sampled (Schleier et al., 2010). Two subsamples were taken for each spray formulation and combined. Distributions were fit using MATLAB® R2010a distribution fitting tool to determine the count median diameter (CMD; μm) and the volume median diameter (VMD; μm) using the Hatch-Choate conversion equations (Cooper, 2001; Hinds, 1982). Distributions for droplet spectra were determined based on the chi-square goodness of fit test, which tests if a sample of data came from a population with a specified distribution (Cooper, 2001; Hinds, 1982; Neter et al., 1996). The log-normal distribution fit all spectra obtained, which is consistent with previous studies of droplet distributions for ULV sprays and aerosols other than ULV (Hinds, 1982; Schleier et al., 2010). Volume and count median diameters by year, location, and formulation mixture are listed in Table 1.

Wind speed, air temperature, wet-bulb depression, and relative humidity were measured with a Hobo Micro Station Data Logger (Onset Computer Corporation, Bourne, MA) attached to 12-bit temperature and relative humidity sensors with a solar radiation shield and a wind speed and direction smart sensor positioned 2.5 m above the ground. Temperature and relative humidity readings were also taken 10 m above the ground using a Hobo temperature and relative humidity data logger (Model H08-032-08, Onset Computer Corporation, Bourne, MA). Meteorological measurements were taken upwind of the spray site. Wind speed, wet bulb depression, temperature, and relative humidity at both 2.5 and 10 m above the ground were averaged over a 5 minute interval.

Temperature readings taken 2.5 and 10 m and the mean wind speed at 2.5 m above the ground were used to calculate the stability ratio. The stability ratio is defined as:

$$SR = \frac{T_{10} - T_{2.5}}{\mu_W} \times 10^5 \qquad (1)$$

where SR is the stability ratio, T_{10} and $T_{2.5}$ is air temperature (°C) at 10 and 2.5 m respectively and μ_W is the mean wind velocity (cm/s) at 2.5 m (Armstrong, 1979; Yates et al., 1981). A stable atmospheric condition (termed an inversion) is defined as a warm air layer overlying a cold air layer. An unstable atmospheric condition (termed a lapse) is defined as a cold air layer overlying a warm air layer. For model development, atmospheric stability was categorized into four categories as defined by Yates et al. (1981) (Table 2). A categorical variable was used to simplify the model inputs and because the continuous stability ratio variable did not substantially increase the predictive ability of the selected model.

Table 1
Volume median diameter (VMD), count median diameter (CMD), and number of each formulations and number of replications for each year, location, and formulation.

Year	Location	Formulation	VMD (μm)	CMD (μm)	Number of replications
2009	California	Aqua-Reslin	21.3	1.3	9
2009	California	Permanone 31–66	24.56	1.36	8
2009	Montana	Aqua-Reslin	21.19	2.2	13
2009	Montana	Permanone 30–30	18.72	2.3	12
2009	Montana	Scourge	11.63	1.43	4
2010	California	Aqua-Reslin	22.56	1.75	7
2010	California	Pyronyl Crop Spray	31.3	2.1	7
2010	Montana	Aqua-Reslin	19.76	1.25	8
2010	Montana	Permanone 30–30	13.4	1.3	7
2010	Montana	Zenivex E20	13.96	1.3	7
2010	Montana	Aqua-Kontrol	24.75	1.2	6
2010	Montana	Scourge	12.37	1.25	2
2011	Louisiana	Aqua-Reslin	6.99	1.19	2
2011	Louisiana	Permanone 30–30	8.79	1.24	5
2011	Montana	Permanone 31–66	20.8	1.57	1
2011	Montana	Permanone 30–30	16.1	1.43	2
2011	Montana	Zenivex E20	12.6	1.47	4
2011	Montana	Aqua-Reslin	22.95	1.36	2

Formulation density was determined by averaging the weight of five 1 ml samples of each formulation on a calibrated Mettler AM100 analytical balance (Mettler Toledo AG, Switzerland). The density of Aqua-Reslin mixed 1:1 with D.I. H_2O, Aqua-Reslin mixed 1:0.75 with D.I. H_20, Aqua-Reslin mixed 1:1.5 with D.I. H_2O, Permanone 30–30, Permanone 31–66, Scourge, Aqua-Kontrol, Zenivex E20, and Pyronyl Crop Spray was 0.99, 0.99, 1.02, 0.88, 1.01, 0.93, 1.02, 0.88, and 0.89 g/l, respectively.

Linear regression analysis and analysis of variance (ANOVA) were run using R Statistical Package version 2.12.2 (The R Foundation for Statistical Computing, Vienna, Austria). Diagnostic plots of the selected model showed that concentrations required log transformation to meet the assumptions of normality and constant variance. Correlation analysis between predictor variables was assessed, and correlation values greater than 0.5 were excluded because collinearity inflates the standard errors. Variables that were excluded were temperature and relative humidity measured at 10 m above the ground, wet bulb depression, and dew point because they were all correlated with temperature and relative humidity measured 2.5 m above the ground. In addition, measurements of temperature and relative humidity at ground level are easier to obtain than the excluded variables.

Linear regression variables that were incorporated into the model were wind speed (cm/s), air temperature (°C), relative humidity (%), stability category, distance from spray source (m), application rate (g/l), flow rate (ml/min), density (g/ml), CMD (μm), and VMD (μm). Interactions were allowed based on aerosol physics and included: distance∗CMD, distance∗density, distance∗wind speed, distance∗temperature, distance∗relative humidity, distance∗stability category, distance∗CMD, distance∗VMD, distance∗flow rate, density∗CMD, density∗VMD, density∗wind speed, density∗stability category, CMD∗wind speed, CMD∗temperature, CMD∗relative humidity, CMD∗stability category, CMD∗VMD, VMD∗wind speed, VMD∗temperature, VMD∗relative humidity, VMD∗stability category, wind speed∗temperature, wind speed∗relative humidity, wind speed∗stability category, and temperature∗relative humidity. These interactions were included because they indirectly take into account variables that cannot be directly measured. For example, temperature∗relative humidity can indirectly estimate the effects evaporation. In addition, multicollinearity with interaction variables does not affect the predictive ability of multiple linear regression models (Neter et al., 1996).

Model selection was performed using the Bayesian information criterion (BIC) with the R library MASS because of the higher penalization for the number of coefficients parameters which is appropriate for large sample sizes (Hastie et al., 2009). Analysis of variance was performed to determine if there were significant differences in mean concentrations between years and location ($\alpha = 0.05$). An extra sum of squares *F*-test was carried out to determine if there was a reduction in the error sums of squares between the model selected with BIC and the full model with all interactions listed above ($\alpha = 0.05$) (Neter et al., 1996). In addition, we performed an extra sum of squares *F*-test to determine if adding location to the selected model would reduce the error sum of squares.

Because the selected model mean square error may understate the inherent variability in making future predictions, we performed model validation using *k*-fold cross validation using the R library DAAG to estimate the mean square prediction error. *k*-fold cross validation was used because it reduces the bias associated with other model validation techniques (Olson and Delen, 2008). The *k*-fold technique works by splitting

Table 2
Atmospheric turbulence, stability ratio calculation as defined by Eq. (1), as defined by Yates et al. (1981), and the linear regression category.

Atmospheric turbulence	Stability ratio (SR)	Linear regression category
Unstable	−1.7 to −0.1	1
Neutral	−0.1 to 0.1	2
Stable	0.1 to 1.2	3
Very stable	1.2 to 7.0	4

the complete data set into *k* mutually exclusive subsets of approximately equal size with the model being trained *k* times and compared to the held out set of data (Olson and Delen, 2008). The data were split into 10 data sets (*k*), which is the most commonly used number of data sets for *k*-fold cross validation (Olson and Delen, 2008). If the mean square prediction error is close to the selected model mean square error, then the fit of the selected regression model is not seriously biased and gives an appropriate indication of the predictive ability (Neter et al., 1996).

3. Results

A total of 1067 data points collected from the three states and years were used for statistical analysis. The regression model that was selected is,

$$\begin{aligned}\mathrm{C} = {} & -3.71 + 0.008{*}\mathrm{D} - 1.15{*}\mathrm{AR} - 0.005{*}\mathrm{FR} - 24.57{*}\mathrm{DEN} \\ & +3.95{*}CMD + 1.13{*}VMD \\ & -0.014{*}\mathrm{WS} + 0.34{*}\mathrm{T} + 0.15{*}\mathrm{RH} + 1.44{*}\mathrm{SC} \\ & -0.007(\mathrm{D}{*}\mathrm{CMD}) - 0.00002(\mathrm{D}{*}\mathrm{WS}) + 18.95(\mathrm{DEN}{*}\mathrm{CMD}) \\ & -0.007(\mathrm{DEN}{*}\mathrm{WS}) - 1.66(\mathrm{DEN}{*}\mathrm{SC}) - 0.16(\mathrm{CMD}{*}\mathrm{T}) \\ & -0.064(\mathrm{CMD}{*}\mathrm{RH}) - 0.32(\mathrm{CMD}{*}\mathrm{SC}) - 0.78(\mathrm{CMD}{*}\mathrm{VMD}) \\ & -0.004(\mathrm{VMD}{*}\mathrm{T}) + 0.034(\mathrm{VMD}{*}\mathrm{SC}) + 0.001(\mathrm{WS}{*}\mathrm{T}) \\ & +0.0001(\mathrm{WS}{*}\mathrm{RH}) + 0.0004(\mathrm{WS}{*}\mathrm{SC}) - 0.003(\mathrm{T}{*}\mathrm{RH})\end{aligned} \tag{2}$$

where C is the log of the concentration (μg/cm^2), WS is wind speed (cm/s), T is temperature (°C), RH is relative humidity (%), SC is stability category, D is distance from spray source (m), AR is application rate (g/l), FR is flow rate (ml/min), DEN is density (g/ml), CMD is count median diameter (μm), and VMD is volume median diameter (μm).

The model with the lowest BIC value coefficient estimates and standard errors are listed in Table 4. There was no evidence that a richer model that included all interactions provided a better fit (extra sum of squares F-test; $F_{10,\ 1041} = 1.01$, $p = 0.43$) suggesting that the simpler model was sufficient. Diagnostic plots for the selected model demonstrate that there were no significant outliers and the residuals were consistent with the assumption of normality. The selected model (Eq. (2)) adjusted R^2 is 0.4 and the model BIC value was −140. The full model with all interactions has an adjusted R^2 of 0.42 and BIC value of 2919. The selected regression model (Eq. (2)) mean square error was 0.904 which was similar to the average mean square prediction error of 1.24 (range of mean square prediction errors for the 10 folds was 1 to 1.4) estimated by the *k*-fold cross validation. The similarity between the mean square prediction error and the mean square error of the model indicates that the selected regression model was not seriously biased, demonstrating good predictive ability (Neter et al., 1996). In addition, the coefficients estimated for each fold differed less than 10% from the selected model using BIC, demonstrating the accuracy of the coefficients for predicting concentrations.

Analysis of variance showed that there was no significant effect of year ($F_{2,\ 1062} = 1.72$, $p = 0.19$; Fig. 3), but there were significant differences in mean concentrations measured between Montana, California, and Louisiana ($F_{2,\ 1062} = 11.18$, $p = 0.0009$). However, there was no evidence to support location in the model with the lowest BIC value (full model included location, reduced model excluded location $F_{2,\ 1041} = 0.37$, $p = 0.54$) and location was not a significant categorical coefficient within the selected model ($t = -0.89$, $p = 0.38$). In addition, Fig. 2 shows significant overlap for the three locations and demonstrates that the significant difference may be due to outliers, the larger number of samples, and the greater variety of formulations in Montana. Therefore, because there was no difference between years, and location did not significantly improve the selected model, we pooled the data to use the largest data set for generating the predictive model so that it has the largest range of environmental and application scenarios (Table 3).

Fig. 4 shows the estimated prediction line and the 95% prediction interval using Eq. (2) for the log of concentration (μg/cm^2) using application rate, flow rate, density, count median diameter, volume median diameter, wind speed, temperature, relative humidity, and stability category of 7.85, 192, 0.996, 2.2, 19.76, 215.06, 24.86, 29.68, and 1, respectively. The selected input values are observed from the one replication (field trial) randomly selected from the data set to demonstrate the predictive ability of the model (Fig. 4). Fig. 4 demonstrates that the predictive accuracy of the model is good with all data points within the 95% prediction interval. The model is robust with respect to a wide range of applications (flow rate, density of formulation etc.) and environmental explanatory factors used for its development (Table 3). Because we grouped over years and the model is representative of a wide range of environmental conditions and application equipment scenarios.

Density of the formulation and the density ∗ CMD interaction coefficients were the largest in the model when all coefficients were scaled (Table 4). The results showed that a 0.1 g/ml decrease in density results in a 2.46 times greater concentration over all distances when all other variables are held constant. This is most likely because greater density formulations have greater deposition within 75 m of the spray source while lower density formulations have greater deposition beyond 75 m. Schleier and Peterson (2010) observed the same pattern with an increase in concentration between 25 and 50 m and a subsequent decrease in the concentration. Therefore, because we fit a linear model to the data it resulted in a negative density coefficient. Larger CMD results in greater deposition of the insecticide. The interaction of density and CMD shows that greater density formulations and larger droplets result in greater deposition.

Distance did not have a large influence on the deposition of ULV insecticides, which is most likely due to the large number of droplets between 1 and 10 μm produced by ULV technology (Schleier et al., 2010) which have low settling velocities and therefore settle out uniformly across the spray area. However, Fig. 4 shows that the model predicts decreasing concentrations of insecticide with increasing distance from the spray source. Higher temperatures and relative humidity lead to greater deposition most likely because these conditions lead to less dense air allowing the droplets to settle out faster (Davis, 1992). Higher wind speeds resulted in lower deposition of ULV insecticides because higher wind speeds push the aerosol through the spray area allowing less to settle out. Stable atmospheric conditions resulted in greater deposition than unstable conditions. During unstable atmospheric conditions thermal currents from the warmer ground keep the aerosol in the air column longer thus leading to lower deposition of the insecticides.

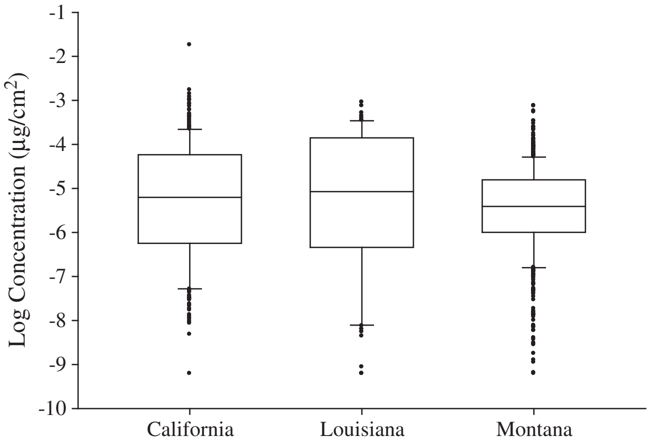

Fig. 2. Boxplot of log concentrations (μg/cm^2) in California, Louisiana, and Montana.

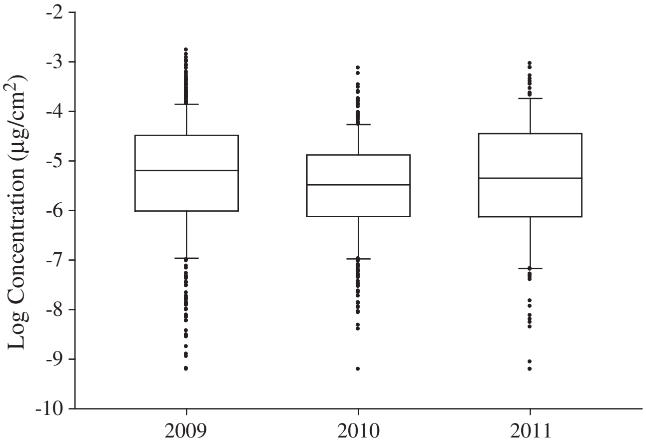

Fig. 3. Boxplot of log concentrations (μg/cm^2) in 2009, 2010, and 2011.

4. Discussion

Previous studies of ground-based ULV applications using both pyrethroid and organophosphate insecticides have found 1 to 30% of the insecticide sprayed deposits on the ground within 100 m of the spray source (Knepper et al., 1996; Moore et al., 1993; Schleier and Peterson, 2010; Tietze et al., 1994). We observed an average of 10.4% (SE = 0.4%) of the insecticides sprayed settled out within 180 m of the spray source. Concentrations of insecticide measured in the present study are similar to concentrations measured by previous studies that used gas chromatography to quantify concentrations (Knepper et al., 1996; Moore et al., 1993; Schleier and Peterson, 2010; Tietze et al., 1994). The model is robust with regard to environmental factors and locations because we were able to group all three locations to develop the model. Therefore, the model can be extrapolated to different locations provided that the input parameters are within the values show in Table 3.

The model we have developed and validated is also supported by theory and other previous empirical results, which is another important line of evidence for model validation (Neter et al., 1996). The transport and deposition of aerosols in the environment are due to gravity, diffusion, inertia, and electrostatic mechanisms (Baron and Willeke, 2001a; Baron and Willeke, 2001b). Although aerosol particles follow the overall wind direction, the trajectories can deviate due to external forces such as wind direction and velocity (Baron and Willeke, 2001b). The density of a formulation affects how particles gravitationally settle out of the air column (Baron and Willeke, 2001a; Hinds, 1982). To describe the gravitational settling of aerosols, the aerodynamic diameter is used to characterize particles that move primarily by settling as opposed to diffusion in still air (Hinds, 1982). The aerodynamic diameter therefore characterizes the terminal settling velocity of a droplet of a given size which determines the distance a droplet travels. Therefore, the settling velocity modeled by the CMD and density interaction confirms that the distance a droplet travels is significantly influenced by the settling velocity of the droplets.

A statistical approach to modeling aerosols like those used for ULV applications is more appropriate for model development than physics-based modeling techniques because of the difficulties in estimating the evaporation of droplets within the aerosol cloud and the large spectrum of droplet sizes produced within the cloud (Teske et al., 2003). Physics based models for pesticide drift use wet-bulb depression to

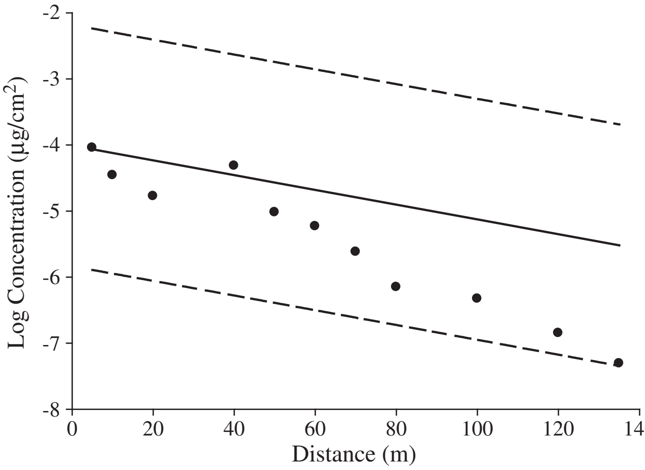

Fig. 4. The prediction line (solid line) and 95% prediction interval (dashed lines) using Eq. (2) for the log of concentration (μg/cm^2) with a application rate, flow rate, density, count median diameter, volume median diameter, wind speed, temperature, relative humidity, and stability category of 7.85, 192, 0.996, 2.2, 19.76, 215.06, 24.86, 29.68, 1, respectively. The input parameters represent the explanatory factors for the data points from one trial randomly selected from the entire data set to demonstrate the predictive ability of the model.

Table 3
Minimum and maximum values for explanatory variables used for model selection.

Response	Minimum	Maximum
Distance (m)	5	180
Application rate (g/l)	2.8	7.85
Flow rate (ml/min)	74	240
Density (g/ml)	0.88	1.02
CMD (μm)	1.19	2.3
VMD (μm)	6.99	31.3
Wind speed (cm/s)	31.76	1267.24
Temperature (°C)	13.94	32.17
Relative humidity (%)	13.06	77.97
Stability category	1	4

Table 4
Coefficients, coefficient estimates, standard error of the coefficient estimates (SE), *t*-values, and *p*-values for the selected model using the Bayesian Information Criterion.

Coefficients:	Coefficient estimate	SE	*t*-Value	*p*-Value
Intercept	−3.71	3.71	−1.0	0.32
Distance	0.008	0.003	3.01	0.003
Application rate	−1.15	0.18	−6.45	<0.0001
Flow rate	−0.005	0.001	−5.46	<0.0001
Density	−24.57	4.73	−5.2	<0.0001
CMD[a]	3.95	1.59	2.48	0.013
VMD[b]	1.13	0.13	8.54	<0.0001
Wind speed	−0.014	0.004	−3.7	<0.0001
Temperature	0.34	0.058	5.79	<0.0001
Relative humidity	0.15	0.026	5.57	<0.0001
Stability category	1.44	0.63	2.3	0.022
Distance ∗ CMD	−0.007	0.001	−5.09	<0.0001
Distance ∗ wind speed	−0.00002	0.000004	−3.67	0.0003
Density ∗ CMD	18.95	2.64	7.18	<0.0001
Density ∗ wind speed	−0.007	0.003	−2.11	0.035
Density ∗ stability category	−1.66	0.65	−2.58	0.010
CMD ∗ temperature	−0.16	0.031	−5.06	<0.0001
CMD ∗ relative humidity	−0.064	0.009	−6.89	<0.0001
CMD ∗ stability category	−0.32	0.10	−3.11	0.002
CMD ∗ VMD	−0.78	0.088	−8.78	<0.0001
VMD ∗ temperature	−0.004	0.002	−2.38	0.018
VMD ∗ stability category	0.034	0.007	4.73	<0.0001
Wind speed ∗ temperature	0.001	0.0001	8.28	<0.0001
Wind speed ∗ relative humidity	0.0001	0.00002	7.90	<0.0001
Wind speed ∗ stability category	0.0004	0.0002	2.03	<0.0001
Temperature ∗ relative humidity	−0.003	0.001	−3.83	<0.0001

[a] CMD is count median diameter.
[b] VMD is volume median diameter.

estimate droplet evaporation (Teske et al., 2003). We excluded wet-bulb depression from the model because it is highly correlated with both temperature and relative humidity. This is because temperature and relative humidity are used to determine the wet-bulb depression. However, our statistical approach allows the model to indirectly take into account the evaporation of droplets through temperature and relative humidity and the interaction between temperature and relative humidity, thus overcoming the complexities of droplet evaporation within the spray cloud.

AGDISP and AgDrift have been validated as an accurate means of estimating the drift of pesticides after aerial agricultural applications, but it has not been validated for ground-based sprayers (Baetens et al., 2009; Bilanin et al., 1989; Duan et al., 1992; Teske and Barry, 1993). Additionally, there has been little work done on validating or calibrating other Gaussian models for the estimation of spray drift from ground-based sprayers (Baetens et al., 2009). Furthermore, the minimum values for input parameters in AGDISP are greater than those used for ground-based ULV applications. For example, the minimum flow rate in AGDISP is 11,690 ml/min which is 50 times higher than the highest flow rate used during our study or those recommend by the label. Therefore, the model we developed and validated provides a better estimation of deposition based on the application parameters for ground-based ULV applications than current agricultural models.

Studies of agricultural spray drift have found that the deposition of insecticides is independent of the active ingredient properties (molecular weight, structure, etc.), but is dependent on environmental factors and formulation properties (Klein and Johnson, 2002; Majewski and Capel, 1995; Reichenberger et al., 2007; SDTF, 2001). Our results similarly showed significant overlap of concentrations for each formulation used (Fig. 5); therefore, the model developed can be applied to classes of insecticides other than pyrethroids.

The model can also be used to estimate bystander exposure because Preftakes et al. (2011) observed similar concentration range of permethrin depositing on mannequins within the spray zone using the formulations Aqua-Reslin and Permanone 30–30 at the application rates, flow rates, droplet spectrum, and dilutions that were used in the current study. In addition to human-health risk assessment, the model can be applied to ecological risk assessment including exposure to aquatic organisms. Therefore, the model can be used by regulatory agencies and researchers to standardize their estimation of human-health and ecological exposures associated with ULV technology.

We have created a validated model for predicting deposition concentrations of insecticides applied with ground-based ULV technology for adult mosquito management using, to our knowledge, one of the largest data set yet generated to develop and validate a spray drift model. Because of the inherent differences between coarse and fine droplet sizes, agricultural and industrial models are not appropriate for ULV applications because smaller droplets travel farther from the spray source. The model that we developed is robust with respect to environmental and application scenarios and demonstrates a good predictive performance. The model coefficients of the selected model reported here are supported by the aerosol physics literature showing that the density of the formulation and diameter of the droplets are the most important determinants of the movement and subsequent deposition of ULV insecticides. Because the model has been validated and is specific to ULV applications used for mosquito management, regulatory agencies such as the USEPA could use it to improve and standardize their risk assessments for registration and re-registration of relevant insecticides.

The model may be useful for mosquito control districts to mitigate risk because they are now required to obtain National Pollutant Discharge Elimination System permits for insecticide applications which require the monitoring of insecticide concentrations in aquatic environments. The model can be used to predict concentrations in water bodies and can allow users to predict when the greatest amount of deposition will occur, thus allowing them to determine if applications should occur. In addition, the findings of our study can be used for other anthropogenic aerosols emitted at ground level.

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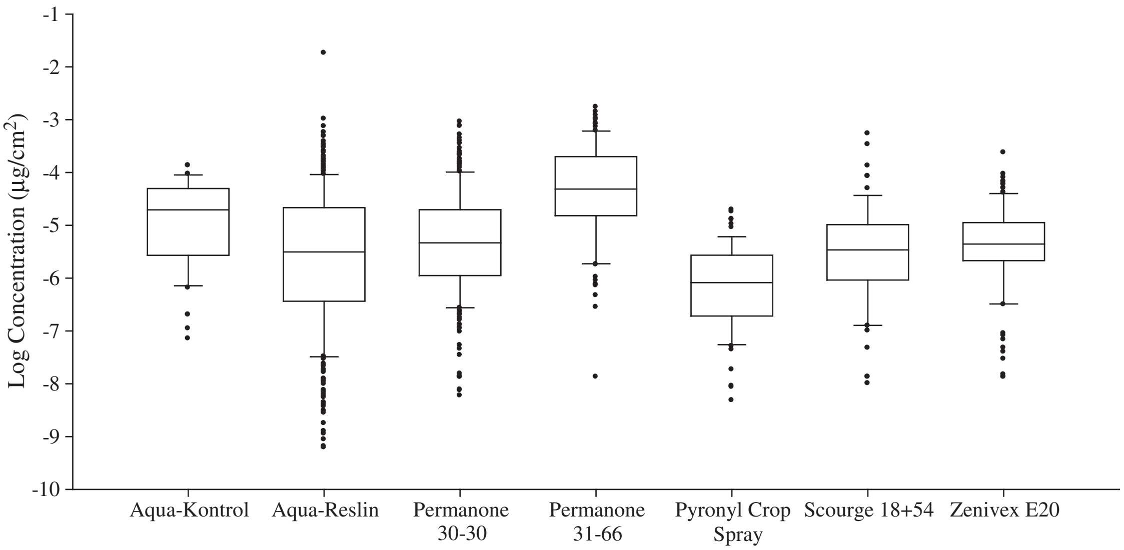

Fig. 5. Boxplots of log concentrations (μg/cm^2) for each formulation.

or firm names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

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