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Hazard/Risk Assessment

A REFINED AQUATIC ECOLOGICAL RISK ASSESSMENT FOR A PYRETHROID INSECTICIDE USED FOR ADULT MOSQUITO MANAGEMENT

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Abstract—The use of pyrethroid insecticides has increased substantially throughout the world over the past few decades as the use of organophorous, carbamate, and organochlorine insecticides is being phased out. Pyrethroids are the most common class of insecticides for ultralow-volume (ULV) aerosol applications used to manage high densities of adult mosquitoes. Pyrethroids are highly toxic to nontarget organisms such as certain aquatic organisms, and there have been concerns about the effect of applications of ULV insecticides on these organisms. To address the uncertainties associated with the risks of ULV applications and the contradictory findings of other ecological risk assessments, the authors performed a probabilistic aquatic ecological risk assessment for permethrin using actual environmental deposition on surfaces to estimate permethrin concentrations in water. The present study is the first ecological risk assessment for pyrethroids to quantitatively integrate the reduction in bioavailability resulting from the presence of dissolved organic matter. As part of the risk assessment, the authors incorporated a species sensitivity distribution to take into account the differences in toxicity for different species. The 95th percentile estimated concentration would result in less than 0.0001% of the potentially affected fraction of species reaching the lethal concentration that kills 50% of a population. The results of the present study are supported by the weight of evidence that pyrethroids applied by ground-based ULV equipment will not result in deleterious effects on aquatic organisms. Environ. Toxicol. Chem. 2013;32:948–953. © 2013 SETAC

Keywords—Mosquito management Pest management Pyrethroid Risk analysis Ultralow volume

INTRODUCTION

The use of pyrethroid insecticides has increased substantially throughout the world over the past few decades as organophorous, carbamate, and organochlorine insecticides are being phased out [1–3]. Pyrethroids are the most common class of insecticides for ultralow-volume (ULV) aerosol applications that are used to manage high densities of adult mosquitoes [4,5].

Pyrethroids are highly toxic to nontarget organisms such as invertebrates and aquatic organisms, and there have been concerns about the effect of ULV insecticide applications on these organisms [6–11]. Pyrethrins and pyrethroids are highly nonpolar chemicals that have low water solubility and volatility, high octanol:water partition coefficients, and a high affinity to bind to sediment and dissolved organic matter [12]. Studies have shown that the presence of dissolved organic material significantly decreases the bioavailable concentration of pyrethroids and the toxicity to aquatic organisms that are not sediment dwellers [13–21].

Davis et al. [22] performed a tier 1 deterministic risk assessment on ground-based ULV applications using the pesticide root zone model (PRZM) and the exposure analysis modeling system (EXAMS) [23] to estimate concentrations of pyrethrins, permethrin, resmethrin, and phenothrin in water. They found that acute and chronic risks to aquatic vertebrates and invertebrates most likely would not result in deleterious impacts on populations after ground-based ULV applications.

To assess the risks of ground-based ULV applications of permethrin, the U.S. Environmental Protection Agency (U.S. EPA) used the agricultural dispersion model (AGDISP) [24] to estimate concentrations of permethrin in a standard 2-m deep farm pond [25]. The U.S. EPA estimated that acute risk to freshwater and estuarine or marine fish would be below regulatory levels of concern [25]. In contrast, the U.S. EPA found that acute risks to freshwater and estuarine or marine invertebrates would exceed regulatory levels of concern [25]. However, the U.S. EPA did not refine its tier 1 assessment.

Ultralow-volume applications of insecticides used for adult mosquito management are most effective when the insecticide remains airborne and moves through the target area; in contrast, applications for agricultural pests are designed to minimize the movement of droplets [26]. To address the lack of a model specific to ULV applications for adult mosquito management, Schleier et al. [27] developed a validated statistical model for predicting deposition of insecticides applied with ground-based ULV technology for adult mosquito management. The data set that Schleier et al. [27] generated of actual deposition in the environment is one of the largest systematic studies on pesticide drift to date. The data set and model are robust with respect to environmental and application scenarios that are typically used for adult mosquito management [27].

To date, only deterministic ecological risk assessments have been conducted for insecticides used for the management of adult mosquitoes, and these have used models that are not validated or appropriate for estimating environmental concentrations [22,25,28–34]. To address the uncertainties associated with the risks of ULV applications and the contradictory findings of other ecological risk assessments, we performed a probabilistic aquatic ecological risk assessment for permethrin using the measured actual environmental deposition from Schleier et al. [27]. The present study is the first ecological risk assessment for pyrethroids to integrate quantitatively the reduction in bioavailability resulting from the presence of

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dissolved organic matter. As part of the risk assessment, we incorporated a species sensitivity distribution to take into account the differences in toxicity for different species.

MATERIALS AND METHODS

Problem formulation

We performed a probabilistic acute ecological risk assessment using actual environmental deposition of permethrin on surfaces after ground-based ULV applications [27]. We chose an acute risk assessment because the presence of suspended sediment substantially reduces the freely dissolved concentration of pyrethroids, thereby greatly reducing the bioavailability [13–21]. Acute exposures were defined in the present study as a single-day exposure after a single application of permethrin.

Hazard identification

Permethrin and pyrethroids, in general, are highly nonpolar chemicals that have low water solubility and volatility, high octanol:water partition coefficients, and high affinity to bind to soil and sediment particles [12,13]. Pyrethroids are broad-spectrum insecticides, so they can have impacts on nontarget organisms [35]. Pyrethroids are highly toxic to certain aquatic organisms, which typically are much more susceptible to pyrethroids than terrestrial organisms [36,37]. We performed a risk assessment for permethrin because it is one of the more widely used insecticides for management of adult mosquitoes and has toxicity similar to that of the other pyrethroids used for mosquito management.

Toxicity and concentration response

To estimate the risk to aquatic organisms, we created a species sensitivity distribution using the U.S. EPA's Ecotox database [38] for permethrin with 40 aquatic species based on 96-h median lethal concentration (LC50) values (Table 1). In the case of more than one entry for a species, we used the lowest LC50 value to be conservative. We used both freshwater and saltwater receptors to construct a species sensitivity distribution, which reflects the diversity of habitats where ULV insecticides may be applied (Table 1). We used data from the Ecotox database because the U.S. EPA used a standard guideline and accepts these studies so that they can be used in regulatory risk assessments.

Table 1. The 96-h median lethal concentration (LC50) values for permethrin obtained from the U.S. Environmental Protection Agency's Ecotox database for both vertebrate and invertebrate species used to model the species sensitivity distribution

Species	Vertebrate or invertebrate	LC50 (μg/L)
Menippe mercenaria	Invertebrate	0.018
Hyalella azteca	Invertebrate	0.021
Palaemonetes pugio	Invertebrate	0.05
Chironomus dilutes	Invertebrate	0.059
Americamysis bahia	Invertebrate	0.075
Crangon septemspinosa	Invertebrate	0.13
Gammarus pseudolimnaeus	Invertebrate	0.17
Penaeus duorarum	Invertebrate	0.22
Procambarus clarkii	Invertebrate	0.28
Daphnia magna	Invertebrate	0.3
Penaeus aztecus	Invertebrate	0.34
Gammarus pulex	Invertebrate	0.44
Ceriodaphnia dubia	Invertebrate	0.57
Nitocra spinipes	Invertebrate	0.6
Homarus americanus	Invertebrate	0.73
Salmo salar	Vertebrate	1.5
Oncorhynchus clarki ssp. *Henshawi*	Vertebrate	1.58
Erimonax monachus	Vertebrate	1.7
Oncorhynchus gilae ssp. *Apache*	Vertebrate	1.71
Salvelinus fontinalis	Vertebrate	2.3
Uca pugilator	Invertebrate	2.39
Etheostoma lepidum	Vertebrate	2.71
Chironomus riparius	Invertebrate	2.89
Oncorhynchus mykiss	Vertebrate	2.9
Pimephales promelas	Vertebrate	3
Etheostoma fonticola	Vertebrate	3.34
Notropis mekistocholas	Vertebrate	4.16
Xyrauchen texanus	Vertebrate	5.95
Ictalurus punctatus	Vertebrate	7.2
Micropterus salmoides	Vertebrate	8.5
Micropterus sp.	Vertebrate	8.5
Chironomus tentans	Invertebrate	10.45
Gambusia affinis	Vertebrate	12
Lepomis macrochirus	Vertebrate	13
Cyprinodon variegates	Vertebrate	17
Cyprinodon bovines	Vertebrate	21
Ptychocheilus lucius	Vertebrate	24
Atherinops affinis	Vertebrate	25.3
Menidia beryllina	Vertebrate	27.5

Species sensitivity distributions are used to estimate the concentrations at which a specified fraction or proportion of species could be affected (also known as the potentially affected fraction). Species sensitivity distributions can also estimate the concentration that may result in the percentage of species reaching their LC50, which is referred to as the hazardous concentration (HCp) [39]. The HCp typically used for regulatory purposes is the HC5, which represents 5% of the potentially affected fraction of species reaching their LC50 [39].

To estimate the species sensitivity, the distribution was fit using the MATLAB R2010a (The MathWorks) distribution fitting tool [39]. The best fit distribution was selected based on the χ^2 goodness of fit test [40–42]. The distribution parameters for the species sensitivity distribution are shown in Table 2.

Table 2. Distributions for deposition on water and the species sensitivity distribution

Input	Distribution type	Parameter	Concentration	Units
Deposition on water	Gamma (Truncated)	Location	0.00009	μg/cm^2
		Scale	0.01	
		Shape	0.81	
Species sensitivity distribution	Log-normal (Truncated)	Mean	11.56	μg/L
		Standard deviation	92.19	

Exposure assessment

We used the actual measured permethrin deposition (μg/cm^2) from the data on ground deposition from Schleier et al. [27] to develop the model. Schleier et al. [27] collected deposition samples after ground-based ULV insecticide applications in field experiments conducted near Elk Grove, California, USA (38°27′17.27″N, 121°27′9.25″W); Bozeman, Montana, USA (45°38′47.09″N, 111°24′8.18″W); and Baton Rouge, Louisiana, USA (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 the greatest depositions for conservative estimates of exposure. The ground deposition for the formulations Permanone 30–30 (30% permethrin), Permanone 31–66 (31% permethrin), Aqua-Reslin (20%

permethrin; Bayer Environmental Science), and Aqua-Kontrol (20% permethrin; Univar) between the distances of 5 and 180 m was used to model the environmental concentrations. Schleier et al. [27] showed that the densities of the formulations had the largest effect on the predicted deposition of insecticide depositing on surfaces; therefore, we modeled all deposition values for permethrin using the different formulations to reflect the variability in the densities of different formulations. All formulations of permethrin were applied at the maximum application rate of 7.85 g active ingredient/ha as listed on the label. Formulations and the order in which they were sprayed were randomly selected. The experimental design was completely randomized, with each formulation randomly selected for the order in which it was sprayed. Replications were performed over time within the same night and over different nights, with a total of 826 deposition data points taken over 82 spray events, which were modeled as a distribution of deposition. MATLAB R2010a was used to fit a distribution to all permethrin deposition values measured 5 to 180 m from the spray source (Table 2).

To estimate the concentrations of permethrin in water, we used a static pond (no inflow or outflow of water). We modeled two water depths representing the standard farm pond with a depth of 2 m and semiaquatic habitats with a depth 0.15 m [23,43]. We used the following equation to estimate the concentration of insecticide in the water.

$$C_t = D \times WD \times CF \tag{1}$$

where C_t is the estimated concentration of permethrin in water (μg/L), D is the deposition on the water surface (μg/cm^2; Table 1), WD is the water depth (2 or 0.15 m), and CF is the conversion from μg/m^3 to μg/L. Schulz et al. [44] demonstrated that this conversion produces concentrations similar to actual measured concentrations in water.

For chemicals that have lipophilicity similar to that of pyrethroids, such as organochlorine insecticides, the dissolved organic content in water is the most significant factor influencing the partitioning of the chemicals [45,46]. To model the bioavailable fraction of permethrin in the presence of dissolved organic matter, we used the equation experimentally derived by Yang et al. [18]

$$C_w = \frac{C_t}{1 + K_{doc}(DOC)} \tag{2}$$

where C_w is the bioavailable concentration of permethrin (μg/L), C_t is the total aqueous concentration of permethrin (μg/L) from equation 1, K_{doc} is the partition coefficient for dissolved organic carbon [18], and DOC is the dissolved organic content [18,21,47]. The range of the K_{doc} values from Yang et al. [18] was modeled with a uniform distribution, the minimum and maximum values being 16,000 and 79,000, respectively, to incorporate the differences in measured values. We used a uniform distribution from 3 to 20 mg/L to model the dissolved organic carbon content, which is representative of ponds, lakes, streams, rivers, and semiaquatic habitats [17,18,48,49].

The model for estimating concentrations of permethrin in water has four key assumptions. First, when the insecticide deposits on the water, it will disperse instantly into the 2- or 0.15-m deep water column. Second, there will be no dilution from water movement. Third, the application will occur immediately adjacent to the pond, and the prevailing wind direction will be over the pond. Fourth, the insecticide will be applied at the maximum application rate of 7.846 g/ha.

Table 3. Percentiles of estimated concentrations of permethrin in water (μg/L) modeled using Equations 1 and 2 with water body depths of 2 or 0.15 m

Percentile	2-m-deep water body concentration (μg/L)	0.15-m-deep water body concentration (μg/L)
5	3.3×10^{-9}	4.39×10^{-8}
10	6.7×10^{-9}	8.91×10^{-8}
15	1.1×10^{-8}	1.46×10^{-7}
20	1.5×10^{-8}	2.00×10^{-7}
25	2.0×10^{-8}	2.66×10^{-7}
30	2.6×10^{-8}	3.46×10^{-7}
35	3.3×10^{-8}	4.39×10^{-7}
40	4.0×10^{-8}	5.32×10^{-7}
45	4.9×10^{-8}	6.52×10^{-7}
50	5.9×10^{-8}	7.85×10^{-7}
55	7.0×10^{-8}	9.31×10^{-7}
60	8.3×10^{-8}	1.10×10^{-6}
65	9.9×10^{-8}	1.32×10^{-6}
70	1.2×10^{-7}	1.60×10^{-6}
75	1.4×10^{-7}	1.86×10^{-6}
80	1.8×10^{-7}	2.39×10^{-6}
85	2.2×10^{-7}	2.93×10^{-6}
90	2.9×10^{-7}	3.86×10^{-6}
95	4.5×10^{-7}	5.99×10^{-6}

Probabilistic risk assessment

To generate the percentiles of water concentrations, we used Monte Carlo simulation (Crystal Ball 7.3; Oracle) with 20,000 iterations using equations 1 and 2 and the distribution for deposition in Table 2. We compared the modeled percentile concentrations to the species sensitivity distribution to determine the potentially affected fraction of species.

RESULTS AND DISCUSSION

The percentiles of estimated concentrations of permethrin in water are presented in Table 3. The HC5 for the species is 0.05 μg/L. The 95th percentile estimated concentration for both the standard farm pond (2 m deep) and the semiaquatic habitat (0.15 m deep) would result in less than 0.0001% of the potentially affected fraction of species reaching their LC50 (Table 3 and Figure 1). If the concentrations were not modeled with the

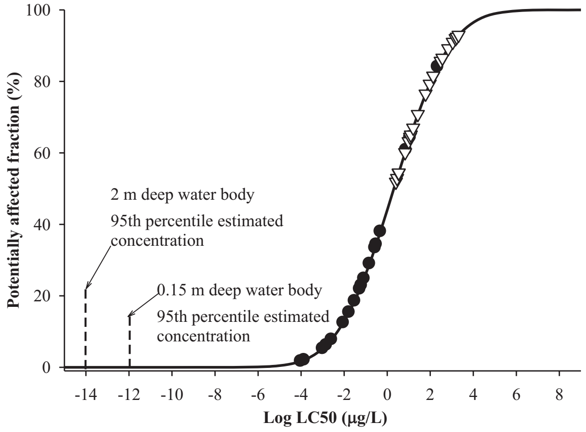

Fig. 1. Acute species sensitivity distribution estimated from the 96-h log of the median lethal concentration (LC50) values, demonstrating the proportion of species affected (aquatic organisms) at the 24-h 95th percentile estimated permethrin concentration with water depths of 2 and 0.15 m. Triangles represent vertebrate species, and circles represent invertebrate species from Table 1.

incorporation of dissolved organic content, the estimated concentrations for the standard farm pond at the 50th and 95th percentiles would be 0.03 and 0.14 μg/L, which would result in approximately 3 and 13% of the potentially affected fraction species reaching their LC50, respectively. If the concentrations were not modeled with the incorporation of dissolved organic content, the estimated concentrations for the semiaquatic habitat at the 50th and 95th percentiles would be 0.39 and 1.86 μg/L, which would result in approximately 25 and 55% of the potentially affected fraction species reaching their LC50, respectively.

The U.S. National Marine Fisheries Service is currently examining the direct and indirect effects of pesticides on endangered salmonids [50]. It suggests that aerial applications of ULV insecticides could adversely affect salmonid prey species [50]. However, Bogen and Reiss [51] showed that the risk was overestimated by the National Marine Fisheries Service because flowing water in a riparian–aquatic scenario reduces the concentration of insecticide between 50- and 300-fold, depending on the water depth and flow rate. Therefore, the concentration incorporating dilution without incorporating the effect of DOC at the 95th percentile from our data would be between 0.003 and 0.0005 μg/L, which would result in less than 0.1 and 0.009% of the potentially affected fraction of species reaching their LC50, respectively. Our results are protective of aerial ULV applications because the concentrations deposited on surfaces and water are lower than those observed after ground-based ULV applications [52–54].

Davis et al. [22] estimated that the concentration of permethrin in water was approximately 0.0004 μg/L using PRZM EXAMS. The concentration estimated by Davis et al. [22] is greater than the 95th percentile concentration in the present study. Therefore, the risk estimate of Davis et al. [22] is conservative, based on our results.

Although pyrethroids are highly toxic to certain aquatic organisms when in the aqueous phase, the presence of suspended sediment substantially reduces the freely dissolved concentration of pyrethroids and therefore their bioavailability [13–15]. Pyrethroids have little mobility in soils and are associated with sediments in natural water; consequently, they will be in the water phase for only a relatively short time, limiting the exposure of many organisms [17–19]. In addition, the half-life of many pyrethroids in aquatic systems that are not bound to sediment is 1 to 5 d [12,13]. Therefore, chronic exposures of organisms that do not have a benthic component in their life cycle most likely will not result in observed effects because pyrethroids dissipate rapidly (dissipation half-life in the water column is generally less than 1 d) [12]. Our results suggest that the bioavailable permethrin after ground-based ULV applications would not result in concentrations above the detection limit in aquatic systems. Furthermore, the rapid dissipation of pyrethroids makes it difficult to reconcile field exposures with those used in laboratory studies that maintain constant concentrations without dissolved organic content.

Experiments have shown that the toxicity of cypermethrin to *Daphnia magna* and *Chironomus tentans* decreases as the dissolved organic carbon content of the water increases [21]. Acute toxicity of pyrethroids decreases 56 to 92%, depending on the concentration of suspended sediments [17,48]. Yang et al. [19] found that pyrethroids adsorbed on suspended sediment or dissolved organic matter were completely unavailable for uptake by *D. galeata mendotae* after a 24-h exposure period. Therefore, because of the physicochemical properties and the use of the 96-h LC50 values for permethrin, the estimated species sensitivity distribution most likely overestimates the toxicity in the environment [17–21,48,55].

In measurements of actual water concentrations of pyrethrins and permethrin, Jensen et al. [56] found no detectable concentrations in wetlands before and after ground-based ULV applications. Weston et al. [10] found no detectable concentrations of pyrethrins in suburban streams 10 and 34 h after aerial ULV applications. Schleier et al. [52] found no detectable concentrations of pyrethrins one hour after aerial ULV applications over irrigation ditches and static ponds. Concentrations of ULV resmethrin in Suffolk County, New York, USA, after ground-based applications were below the limit of detection [57]. These studies support our findings that the concentrations would be below the detection limit in water, which is approximately 5 ng/L [57]. The average detection limits for pyrethrins, permethrin, and resmethrin was 5, 3, and 406 ng/L, respectively.

Several studies on the effects of both aerial and ground-based ULV applications on aquatic organisms have been performed. Davis and Peterson [58] demonstrated no significant impact of pyrethroids on aquatic and terrestrial invertebrates sampled after single and multiple ground-based ULV applications. Lawler et al. [59] found that ground-based ULV applications of pyrethrins synergized with piperonyl butoxide did not cause significant mortality of the aquatic invertebrates *D. magna* and *Callibaetis californicus*. Ground-based applications of ULV permethrin had no significant impact on aquatic macroinvertebrates and *Gambusia affinis* when used near wetlands [56].

After agricultural applications of pyrethroids, reductions of populations in aquatic communities have been observed at concentrations of 5 to 10 μg/L of pyrethroid in the water, with populations recovering within two weeks [13,60–62]. Agricultural applications use as much as 100-fold greater concentrations of active ingredient compared with ULV applications for mosquito management. Hill [63] reviewed approximately 70 freshwater field studies in natural ponds, farm ponds, streams, rivers, rice paddies, and microcosms and mesocosms and found that there were few acute effects of agricultural-use pyrethroids on fish and aquatic invertebrates. Aerial agricultural applications of cypermethrin adjacent to a farm pond showed that dipterans were the most affected in the water, but the populations quickly recovered after the application [64]. Sediment-dwelling invertebrates in the families Gammaridae and Asellidae were adversely affected by direct agricultural sprays of cypermethrin and lambda-cyhalothrin in experimental ponds, but increases in Planorbidae, Chironomidae, and Lymnaeidae were also observed [65]. The effects on sediment-dwelling invertebrates can also be accounted for because type II pyrethroids such as cypermethrin and lambda-cyhalothrin have a greater toxicity than type I pyrethroids (permethrin) to both aquatic and terrestrial invertebrates [11,36,66].

Our exposure model is most likely conservative because most mosquito control districts typically apply the insecticides at one-half or one-quarter of the maximum application rate of 7.845 g/ha that we assumed in our analysis [5]. We also assumed that there would be no buffer, the prevailing wind direction would be over the water body, the truck would travel along the water's edge, the insecticide would instantly disperse into the water column, and the water body would be static. Bogen and Reiss [51] incorporated the flow of water to estimate the exposure to insecticides after pesticide applications and found that dilution reduced the exposure by about 50- to 300-fold from the initial concentration depending on the water depth and flow rate.

Supporting our exposure model are field studies on the effects of both ground-based and aerial applications of ULV insecticides, which showed no significant effects on aquatic organisms. In addition, agricultural applications, which often use 100-fold greater concentrations of pyrethroids and greater toxicity type II pyrethroids, do not have significant effects on aquatic communities until the concentrations reach substantially greater concentrations than the estimated 95th percentile concentration in the current study.

Our study is the first to estimate the aquatic risks from ground-based ULV applications for adult mosquito management using a species sensitivity distribution and actual environmental concentrations deposited on surfaces. It is also the first study to integrate the effect of dissolved organic matter to estimate the bioavailable concentration of pyrethroids in the environment into a risk assessment framework. The data used to estimate the deposition of ULV insecticides on water were obtained from the data set used to generate the validated model developed by Schleier et al. [27], which more accurately represents environmental concentrations. We found that the estimated 95th percentile concentration of permethrin for water depths of 2 and 0.15 m would result in less than 0.0001% of the potentially affected species being exposed to their LC50 when the physicochemical properties of pyrethroids are incorporated into our analysis. Our results are supported by the weight of evidence that pyrethroids disseminated by ground-based ULV applications will not result in detectable concentrations of insecticides or deleterious effects on aquatic organisms.

Because our exposure model estimates concentrations of permethrin in water based on ground deposition data and dissolved organic matter, further research could systematically measure actual concentrations in water after ground-based ULV applications. This research not only would test the conservatism of our assessment but also could be used to refine our exposure model by incorporating actual water concentrations.

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