Aquatic Toxicity Due to Residential Use of Pyrethroid Insecticides

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Pyrethroids are the active ingredients in most insecticides available to consumers for residential use in the United States. Yet despite their dominance in the marketplace, there has been no attempt to analyze for most of these compounds in watercourses draining residential areas. Roseville, California was selected as a typical suburban development, and several creeks that drain subdivisions of single-family homes were examined. Nearly all creek sediments collected caused toxicity in laboratory exposures to an aquatic species, the amphipod *Hyalella azteca*, and about half the samples caused nearly complete mortality. This same species was also found as a resident in the system, but its presence was limited to areas where residential influence was least. The pyrethroid bifenthrin is implicated as the primary cause of the toxicity, with additional contributions to toxicity from the pyrethroids cyfluthrin and cypermethrin. The dominant sources of these pyrethroids are structural pest control by professional applicators and/or homeowner use of insecticides, particularly lawn care products. The suburbs of Roseville are unlikely to be unique, and similar sediment quality degradation is likely in other suburban areas, particularly in dry regions where landscape irrigation can dominate seasonal flow in some water bodies.

Introduction

Pyrethroid insecticides now fill most of the residential needs previously met by organophosphates. Use of organophosphates was drastically curtailed in the United States by the recent withdrawal of nearly all products for residential use that contain chlorpyrifos or diazinon. The vast majority of insecticides sold for consumer use now contain pyrethroids, and they are widely used around homes by professional pest control applicators as well. Agricultural use of pyrethroids has resulted in residues in runoff (*1*), with resulting contamination of creeks receiving return flow from irrigated fields (*2*). Similarly, the pyrethroid bifenthrin has been found in runoff from a commercial nursery (*3, 4*). Landscape irrigation or stormwater runoff could play similar roles in transporting residentially used pyrethroids into urban water bodies. However, there is no monitoring for most pyrethroids in urban environments. The U.S. Geological Survey's National Water-Quality Assessment (NAWQA) Program, the largest effort to monitor urban drainages, monitors sediments for permethrin, only one of many residential use pyrethroids and the one with the lowest aquatic toxicity (*5*). Given the minimal monitoring that has been done for these pesticides with widespread use, there is a need to determine the following: (1) if residential use of pyrethroids results in residues in nearby aquatic systems; (2) if concentrations reach levels that cause mortality in sediment toxicity tests; and (3) if the presence of pyrethroids is a factor controlling the distribution of resident aquatic invertebrates.

Materials and Methods

Study Area. The area surrounding Sacramento, California has experienced rapid population growth, and within the past few years, thousands of homes have been built on land that was historically open grassland. Roseville is one of many such suburban communities surrounding Sacramento. The western portion of Roseville is characterized by numerous contiguous subdivisions of single-family homes, most of which are less than 10 years old. There is no industry in the area and only minimal commercial development and agriculture. The area was selected as a candidate for a case study on residential pesticide use because of the few pesticide sources other than residential application, and the fact that historical data had indicated the presence of *Hyalella azteca* (Arthropoda: Crustacea) in streams in the area, a species of particular interest in this study.

The main watercourse west of Roseville is Pleasant Grove Creek, a slow-moving stream 2–4 m wide and 0.5–1 m deep in most reaches. Kaseberg Creek and the South Branch of Pleasant Grove Creek (hereafter referred to as the South Branch) are the main tributaries (Figure 1). Precipitation of typically 40–60 cm/yr occurs primarily from November through March. During the summer, the primary source of water to the system is runoff from residences from over-irrigation of landscapes and lawns. Many stormwater drains from the housing subdivisions discharge to Pleasant Grove Creek, and particularly its tributaries, along much of their lengths.

Sampling Procedures. Sampling sites were established at 3–6 locations along the mainstem of each of the three creeks, and in 2–3 secondary tributaries entering each creek. These smaller tributaries originate at the outfall of storm drains serving the residential areas, and carry water from the outfalls to the main creeks.

Pyrethroids are rapidly adsorbed to soil particles, so sediments would expected to be the main repository for these compounds (*4*). Bottom sediments were collected from most sampling sites in September 2004, with the remainder of the sites sampled in either the preceding or following month. There were rain events between each of these sampling occasions, though 1–3 sites were resampled before and after each rain. No appreciable change in toxicity or pyrethroid concentrations was observed, and results from these few sites with multiple samples are sometimes averaged in the data presented.

All sites were sampled from the bank or by wading into the creek, using a steel scoop to skim the upper 1 cm of the sediment column. Approximately 3 L of sediment was collected at each site, placed in pre-cleaned glass jars, and held on ice until return to the laboratory. All sediments were

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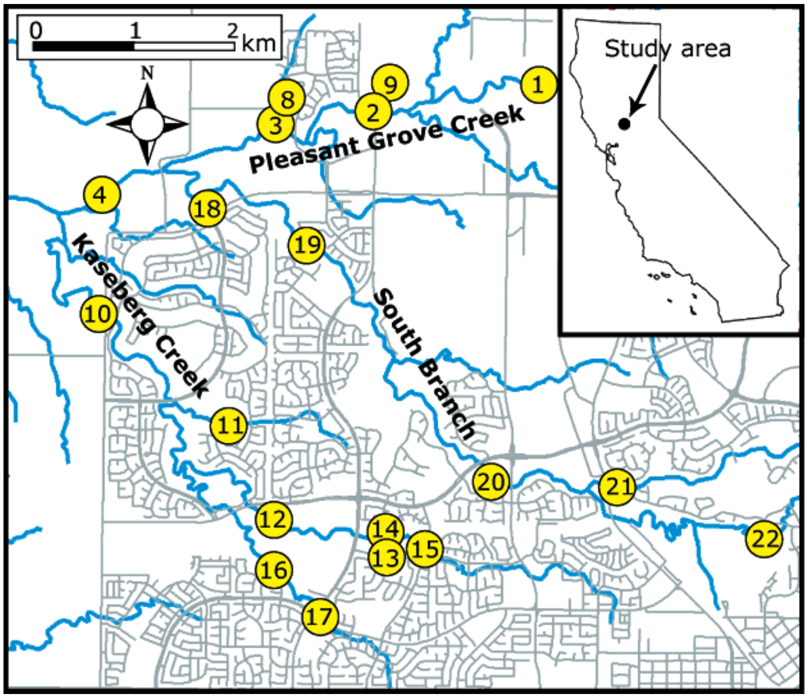

FIGURE 1. Map of study area with sampling sites shown. Inset map shows location of study area within California. Areas of housing development can be inferred from density of roads. Water flow in all creeks shown is from east to west. Stations 5, 6, and 7 are in Pleasant Grove Creek off the left side of the map, approximately 7, 10, and 13 km downstream of station 4, respectively. They are not shown because doing so would substantially reduce the detail visible in the map.

homogenized in the laboratory by hand mixing, then held at 4 °C (toxicity samples) or −20 °C (chemistry samples).

A physical habitat assessment was conducted at each site to document any heterogeneity among sites that could affect availability of *H. azteca* habitat. Physical habitat assessments consisted of collection of the standardized Habitat Assessment Field Data Sheet (*6*) for low-gradient wadeable streams. Site habitat data included estimates of epifaunal substrate/available cover, pool substrate characterization, pool variability, sediment deposition, channel alteration, channel sinuousity, bank stability, vegetation protection, and riparian vegetative zone width. Data are not presented, though the assessment documented comparable habitat throughout the study area. Water quality measurements were taken for dissolved oxygen, temperature, pH, and specific conductivity.

Field sampling for resident *H. azteca* was conducted using a low-gradient modification of the California Stream Bioassessment Procedure (*7*). All of the sample sites were low gradient (slope <0.2) and did not contain riffle habitat. Each sampling site consisted of a relatively homogeneous 100-m sampling reach. The reach was divided into three equal segments and each segment was sampled by approximately 20 jabs followed by a sweeping motion using a 0.5-mm mesh D-frame kick net. Sampling included aquatic macrophytes and overhanging riparian vegetation along the banks, as well as scraping along the surface of the bottom sediments. The sample from each segment was preserved with 10% formalin and later transferred to 70% ethanol. Laboratory processing included enumeration of only the *H. azteca* in each sample.

Analytical Methods. Chemical analytes included seven pyrethroids, 20 organochlorine pesticides or their degradation products, and one organophosphate (chlorpyrifos). Individual pyrethroid isomers were quantified, though they are summed in all data presented. Analysis followed the methods described by You et al. (*8*), differing only in quantification of 3 additional pyrethroid analytes. Briefly, sediment samples were sonicated with a solution of acetone and methylene chloride and the extracts were cleaned by column chromatography with deactivated Florisil. Analysis was performed on an Agilent 6890 series gas chromatograph with an Agilent 7683 autosampler, an electron capture detector, and two columns, an HP-5MS and a DB-608 (Agilent Technologies, Palo Alto, CA). Qualitative identity was established using a retention window of 1% with confirmation on a second column, and calibration was based on area using external standards at concentrations ranging from 10 to 100 μg/L diluted from stock solutions. Analytical grade standards were used throughout the study. The pyrethroids were purchased from Chem Service (West Chester, PA). Organochlorines, organophosphates, and surrogate standards were purchased from Supelco (Bellefonte, PA). Detection limits for the individual pyrethroids ranged from 0.1 to 0.6 ng/g, though a consistent reporting limit of 1.0 ng/g was used for all analytes. Recovery of pyrethroids from fortified samples analyzed blind ranged from 61 to 105%.

Two samples (sites 13 and 15) were also analyzed by a second laboratory for quality assurance purposes. This second laboratory extracted the sediments using pressurized fluid extraction (Dionex 200 Accelerated Solvent Extractor, Dionex, Sunnyvale, CA). Gel permeation chromatography followed by Florisil column chromatography were used for extract cleanup. Analysis was done with an Agilent 6890plus gas chromatograph with autosampler, equipped with two ^{63}Ni micro-electron capture detectors and dual 60-m capillary columns (DB-5 and DB-17MS, Agilent Technologies). Positively identified pyrethroids were confirmed using gas chromatography with mass spectrometry-ion trap detection (GC/MS-ITD) when possible. A Varian GC/MS-ITD, Saturn 2000 (Varian, Palo Alto, CA) was used with a 30-m DB-5MS column (Agilent Technologies). The GC/MS-ITD was used in select ion storage (SIS) and/or MS−MS mode. All concentration data presented were derived from analyses by the primary laboratory, rather than the second laboratory that was used primarily for confirmation of analyte identity by GC/MS. However, results from the second lab confirmed both the identity and quantification of analytes as reported herein.

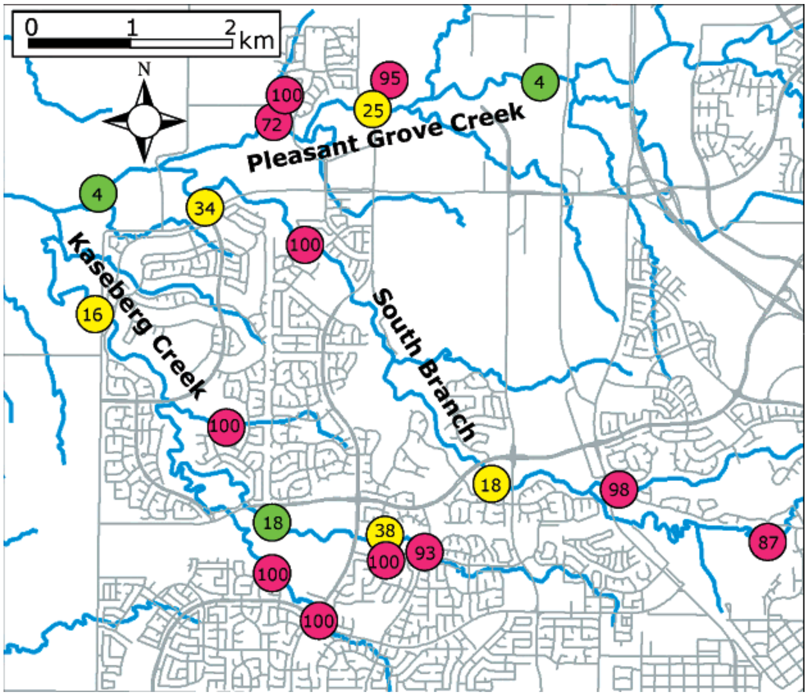

FIGURE 2. Distribution of sediment toxicity among the study sites. The numerical values at each site indicate the percent mortality of *H. azteca* in 10-d toxicity tests. Results are also illustrated by color coding (red = high toxicity with >70% mortality; yellow = moderate toxicity with mortality significantly greater than control but <70%; green = nontoxic with mortality not significantly different than control). Two stations (sites 5 and 6) not shown, but located on Pleasant Grove Creek 7 and 10 km, respectively, further downstream of station 4 were also nontoxic.

Total organic carbon was determined on a CE-440 elemental analyzer from Exeter Analytical (Chelmsford, MA), following acid vapor treatment to remove inorganic carbon.

Toxicity Testing. Toxicity testing was performed using 7–10-d old *H. azteca*, following standard methods (*9*). Testing was done in 400-mL beakers containing about 75 mL of sediment, with eight replicate beakers per sample. Test protocols included use of moderately hard water reconstituted by addition of salts to Milli-Q purified water (Millipore, Billerica, MA), a temperature of 23 °C, a 16:8-h light/dark cycle, and daily feeding with YCT (yeast, cerophyll, trout chow). Two volume additions of water were supplied daily to each testing chamber by an automatic water delivery system. This rate of water renewal was sufficient to keep dissolved oxygen levels high (5–7 mg/L) in most instances. However, three sediments required gentle aeration. Sediment from site 8 was aerated for the full test duration. Sediments from sites 21 and 22 received aeration beginning on day 5 when dissolved oxygen had declined to about 3 mg/L. After a 10-d exposure period, the amphipods were recovered, survival rate was determined, and biomass was measured after drying at 70 °C to determine growth.

All test batches included control sediment containing 1.87% organic carbon, collected from the South Fork of the American River in Placer County, CA near Folsom Lake. Sediment from this location was one of three sediments that had previously been amended with pyrethroids to determine the LC_{50} values used herein (American River sediment of Amweg et al. (*10*)).

Toxicity data were analyzed using ToxCalc Version 5.0 (Tidepool Scientific Software, McKinleyville, CA). Dunnett's Multiple Comparison test was used to identify stations with significantly greater mortality than the control. Arcsin squareroot transformation was used when necessary to meet assumptions of normality and homogeneity of variance. If these assumptions were not met even after transformation, comparison to control was done using Steel's test.

Results and Discussion

Toxicity Testing. Sediments throughout Pleasant Grove Creek and its tributaries were tested for acute toxicity to the amphipod, *H. azteca*, a species widely used for freshwater sediment testing. Sediment from 9 of the 21 sites caused total or nearly total (>90%) mortality of *H. azteca* in a 10-d exposure (Figure 2). Sediments from the smaller secondary tributaries, all of which originate at storm drain outfalls and carry runoff to the three creeks, were particularly toxic with mortalities ranging from 34 to 100% (mean = 90%). Growth data are provided in the Supporting Information (Table S1), but are not discussed herein as they do not substantially alter the results gained from the mortality endpoint alone.

Sediments from most of the mainstem of Pleasant Grove Creek showed no toxicity (~4% mortality). However, sediments were acutely toxic (25–72% mortality) in Pleasant Grove Creek at the confluence with two tributaries that drain housing developments to the north. Sediments collected within these two tributaries caused total or nearly total mortality.

Sediments throughout most of Kaseberg Creek showed mortality rates greater than the control, and mortality rates tended to increase from its confluence with Pleasant Grove Creek (16% mortality) to the most upstream sites (93 and 100% mortality). Similarly, every site in the South Branch showed significant mortality (18–100%).

Sediment Chemistry. To help identify the cause of the observed toxicity, sediments were analyzed for 28 pesticides including one organophosphate (chlorpyrifos), 20 organochlorines, and 7 pyrethroids. The concentrations of chlorpyrifos and the organochlorines were below levels associated with toxicity to *H. azteca* (*2*). Those results are presented in the Supporting Information (Table S2) but not discussed here. All seven of the pyrethroid analytes were detected in sediments from at least some sites, but esfenvalerate and lambda-cyhalothrin were found infrequently and at low concentrations (Table 1).

TABLE 1. Pyrethroid Concentrations in Creek Sediments (ng/g Dry Weight)[a] (Sites with a and b Designations Indicate Two Samples Taken at the Same Location Approximately One Month Apart)

sampling site	Compound[b]						
	bif	cyfl	cyper	delta	esf	lam	perm
Pleasant Grove Creek							
mainstem							
1	3.3	u	u	u	u	u	14
2	9.0	u	u	u	u	u	24
3a	17	5.2	2.4	5.1	u	u	17
3b	14	3.5	2.6	2.8	u	2.5	11
4	1.7	u	u	u	u	u	8.2
5	1.5	u	u	u	u	u	3.1
6	1.2	u	u	u	u	u	2.1
tributaries							
9	40	8.3	14	2.0	u	1.6	19
8	77	70	18	5.1	u	2.0	22
South Branch							
mainstem							
21	36	27	23	8.7	2.5	3.4	57
20	5.8	u	u	u	u	u	7.4
19a	146	11	8.0	4.9	u	1.6	54
19b	78	12	3.7	3.1	1.6	1.6	29
tributaries							
22	74	48	40	u	u	3.4	154
18	11	u	4.0	u	u	u	u
Kaseberg Creek							
mainstem							
17	340	161	64	46	u	9.3	188
15	201	96	30	17	5.8	12	117
14	30	6.5	u	1.8	u	1.3	22
12	13	u	u	u	u	1.2	u
10a	6.1	u	u	1.6	u	1.6	u
10b	7.4	3.1	1.3	u	u	1.2	4.5
tributaries							
13	413	167	736	5.7	u	13	225
16	217	90	36	11	5.8	3.5	100
11	437	169	38	15	5.3	8.7	335

[a] u indicates concentration below reporting limit (<1 ng/g). [b] bif = bifenthrin, cyfl = cyfluthrin, cyper = cypermethrin, delta = deltamethrin, esf = esfenvalerate, lam = lambda-cyhalothrin, perm = permethrin.

Pleasant Grove Creek generally had no detectable pyrethroids except for small quantities of permethrin and bifenthrin. However, an exception was the region around stations 2 and 3, the only portion of Pleasant Grove Creek within the study area where there is housing immediately adjacent to the creek. This region contained moderate concentrations of bifenthrin (9–15 ng/g), probably from two small tributaries draining the developed area to the north. Sediments in the two tributaries (stations 8 and 9) contained 40–77 ng/g bifenthrin and up to 70 ng/g cyfluthrin.

Kaseberg Creek and the South Branch, both of which pass through extensive housing developments, contained far higher concentrations of pyrethroids than Pleasant Grove Creek, which borders the northern fringe of the developed area. Secondary tributaries of Kaseberg Creek had the highest concentrations of pyrethroids, particularly bifenthrin, cyfluthrin, cypermethrin, and permethrin. Contamination in the Kaseburg Creek mainstem was less severe, with the exception of the most upstream sites (stations 15, 17).

The extent of pyrethroid contamination in these suburban sediments is remarkable, particularly in comparison to the lesser levels of contamination for some of the same compounds reported in water bodies affected by agriculture. Bifenthrin concentrations in the secondary tributaries reached 437 ng/g, about 15 times greater than the highest bifenthrin concentration reported from about 70 samples from creeks and drains in areas of intensive agriculture in California (*2*). Peak concentrations of permethrin and lambda-cyhalothrin were comparable in the suburban and agricultural areas. These comparisons, however, may be distorted by the fact that samples with the highest concentrations in the current study were often collected near the point of storm drain inputs, whereas agriculture-related samples have been farther from individual outfalls.

The data suggest that sediment contamination was localized near storm drain outfalls and at points where the secondary tributaries enter the main creeks. For example, station 14 in Kaseberg Creek contained 62 ng/g total pyrethroids, far less than at a site only 0.2 km upstream (station 13, 1560 ng/g) or another site 0.5 km upstream (station 15, 479 ng/g). The fact that Pleasant Grove Creek is relatively unaffected despite the widespread contamination in Kaseberg Creek and the South Branch also suggests minimal transport of contaminated sediments, probably due to the low current speeds in the creeks. Overall, it appears that any given outfall affects sediment quality for a distance of tens to hundreds of meters downstream. However, given the numerous outfalls scattered throughout the system, the result is a patchwork of highly contaminated reaches.

Pyrethroid concentrations were used to calculate toxicity units (TU) in the sediments as follows:

$$\mathrm{TU} = \frac{\text{Actual concentration (organic carbon normalized)}}{\text{Reported } H.\ azteca\ \mathrm{LC}_{50} \text{ concentration (organic carbon normalized)}}$$

LC_{50} concentrations for a 10-d exposure of *H. azteca* to pyrethroid-contaminated sediments have been determined for 3 sediments (*10*). The LC_{50} values used in the TU analysis are the means from these 3 sediments: bifenthrin = 0.52 μg/g organic carbon (oc), cyfluthrin = 1.08 μg/g oc, cypermethrin = 0.38 μg/g oc, deltamethrin = 0.79 μg/g oc, esfenvalerate = 1.54 μg/g oc, lambda-cyhalothrin = 0.45 μg/g oc, and permethrin = 10.83 μg/g oc. All pyrethroids are extremely hydrophobic, thus, LC_{50} values are more consistent and the TU analysis is improved by expressing concentrations normalized to sediment organic carbon (*10*).

When the pyrethroid concentrations are expressed as TUs, it is apparent that nearly all of the sites had concentrations that would be expected to be acutely toxic (Figure 3). All sites but one (station 20) in Kaseberg Creek and the South Branch had at least 1 TU, indicating that *H. azteca* would be expected to show high mortality in sediment toxicity tests due to pyrethroids nearly anywhere in either creek. The tributaries of Kaseberg Creek are particularly noteworthy because their sediments had 14–41 times the acutely lethal concentrations of pyrethroids.

Pleasant Grove Creek was the only creek where TU analysis suggests pyrethroids concentrations were low enough to allow survival of *H. azteca*. Sediments in its two small northern tributaries (stations 8 and 9) had 7–13 TU, and Pleasant Grove Creek had 2–3 TU in the region where these tributaries enter. However, the remainder of Pleasant Grove Creek was well below 1 TU.

When the pyrethroid concentration data are weighted by toxicity of the individual compounds, as in the TU analysis, it is apparent that most of the expected toxicity is attributable to bifenthrin. Bifenthrin alone comprised an average of 70% of the TUs among all sites. While bifenthrin is the dominant contributor to the toxicity, it is not the sole contributor. Other pyrethroids, particularly cyfluthrin and cypermethrin, were found in some locations at concentrations expected to be toxic to *H. azteca*. Of the 16 sites with one or more TU, 11 sites would still have more than 1 TU if bifenthrin were excluded from the analysis.

Permethrin was commonly found in creek sediments, having the highest or second highest concentration of all

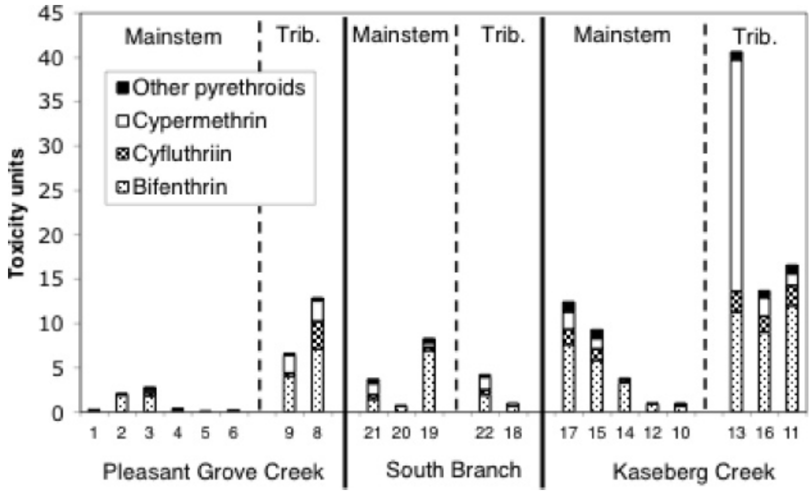

FIGURE 3. ***H. azteca*** **TUs in the sediments at each sampling site, and the contribution of the various pyrethroids to the total TU. At one TU, data in the literature (*10*), suggests that the concentration of pyrethroids would be sufficient to cause 50% mortality to *H. azteca* in 10-d sediment toxicity tests.**

pyrethroid analytes in over $^3/_4$ of the samples. However, it contributed little to the pyrethroid TUs. It is among the least toxic of the pyrethroids to *H. azteca* (*10*) and to aquatic life in general (*5*).

The TU analysis identifies sites where *H. azteca* mortality would be expected based on chemical concentrations and previously reported toxicity thresholds. It was a good predictor of the actual toxicity testing results (Figure 4a) with the correlation between pyrethroid TUs and observed *H. azteca* mortality being highly significant ($p < 0.001$; Spearman rank correlation). At sites with less than 1 TU, little mortality would be expected, and little was observed. At all sites with more than 4 TUs there was, as would be expected, little or no survival. Between 1 and 4 TUs there was some divergence between expected and observed mortality. Mortality of 50% would be expected at 1 TU, but only 15−30% mortality was observed. This discrepancy is not unusual since pyrethroid sediment LC_{50} values can vary somewhat among sediments even after adjustment for organic carbon content (*10*, *11*).

Resident Macroinvertebrates. *H. azteca* is resident in the Pleasant Grove Creek system, and its distribution was studied to determine if patterns were correlated with pyrethroid concentrations and toxicity test results. Populations were present at all sites in Pleasant Grove Creek, although densities were reduced at the mouths of the two northern tributaries and sampling sites nearest to and downstream of the South Branch and Kaseberg Creek tributaries (Table S3). The species was completely absent from both the South Branch and Kaseberg Creek.

The abundance of resident *H. azteca* was inversely correlated with pyrethroid TUs (Figure 4b; $p < 0.05$; Spearman rank correlation). Sediments containing more than 1 TU of pyrethroids had few or no resident *H. azteca*. Densities were variable at sites having less than 1 TU, presumably due to factors other than pyrethroid concentrations. The distribution of resident *H. azteca* was consistent with the patterns of sediment pyrethroid concentrations and toxicity test results, but the patterns were confounded by other habitat factors, for example, the low dissolved oxygen concentrations in some regions of the system. The low input of water in the summer results in low current speeds, and with the accumulation of decaying riparian vegetation in the bottom of the creeks, dissolved oxygen levels can be low (measured at 1.0−7.6 mg/L in Pleasant Grove Creek, 3.6−7.8 mg/L in the South Branch, and 0.5−4.5 mg/L in Kaseberg Creek).

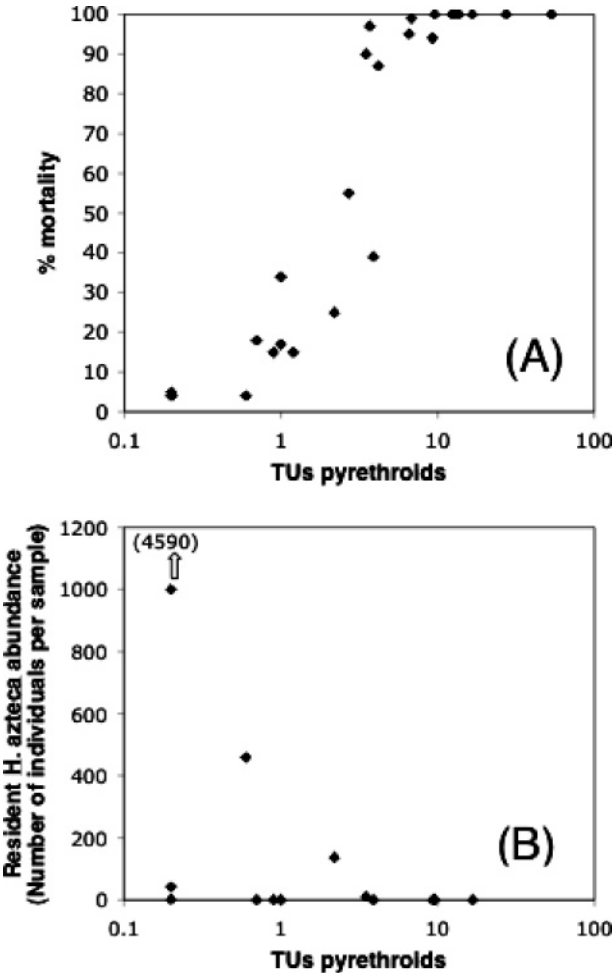

FIGURE 4. Relationship between the number of TUs of pyrethroids in creek sediments and the biological measures: (A) the toxicity to *H. azteca* in 10-d laboratory exposures to the sediments; (B) the density of resident *H. azteca* in Pleasant Grove Creek, South Branch, and Kaseberg Creek. When tested by Spearman rank correlation, both relationships were significant ($p < 0.001$ and $p < 0.05$ for (A) and (B), respectively).

Identifying the Source of pyrethroids. The strong relationship between pyrethroid TUs and observed sediment toxicity, and the fact that *H. azteca* mortality appeared when

TABLE 2. Reported Pyrethroid Use (kg/year) in Placer County, California in 2003 (Reported Data Include Only Commercial Applications, Not Use by Homeowners)

pyrethroid	agricultural use	structural pest control	landscape maintenance
bifenthrin	0.01	141.4	6.2
cyfluthrin	0	275.1	3.9
cypermethrin	0	3337.9	0.05
deltamethrin	0	32.1	0.83
esfenvalerate	17.8	0.02	0
lambda-cyhalothrin	22.6	2.3	0
permethrin	0	673.5	157.5
other	0	1.2	0

sediment pyrethroid concentrations reached levels at which the literature suggests it should if pyrethroids were the responsible agent (at or slightly below 1 TU; Figure 4a), provide strong evidence implicating pyrethroids as the cause of toxicity. The potential sources of the pyrethroids observed in the creek sediments include the following: (1) agriculture; (2) mosquito control; (3) landscape treatment by professional applicators; (4) structural pest control by professional applicators; and (5) landscape application by homeowners or their gardening services. It is possible to eliminate some of these potential sources using statistics taken from the Pesticide Use Reporting database maintained by the California Department of Pesticide Regulation (www.cdpr.ca.gov). All commercial pesticide applications in California require an entry into the database, including information on product used, active ingredient, amount used, date, and place of application. The database includes all agricultural pesticide use and residential applications by licensed pest control firms, but it does not include products purchased from retail stores and used by homeowners or gardening services they may employ.

It is unlikely that the pyrethroids observed were of agricultural origin. There is very little agriculture in the watershed studied or in the county as a whole. In Placer County in 2003 only 1% of the reported pyrethroid use was agricultural (Table 2). Bifenthrin appeared most widespread of all the pyrethroids in Pleasant Grove Creek and its tributaries and made the greatest contribution to the toxicity, but county-wide agricultural use of bifenthrin was only 0.01 kg compared to 147.6 kg of reported nonagricultural use.

Mosquito control spraying can also be eliminated as a source of the contamination observed. The Placer County Mosquito Abatement District controls adult mosquitoes using Scourge, a product containing the pyrethroid resmethrin and the synergist piperonyl butoxide (J. Scott, Placer County Mosquito Abatement District, personal communication). Thus, none of the seven pyrethroid analytes in this study could have originated from mosquito spraying. Two sediment samples (sites 13 and 15) were analyzed specifically for resmethrin with no detectable residues found (GC/MS-ITD screening, 10 ng/g detection limit).

Landscape treatment by professional applicators may have contributed to permethrin in the creeks, as permethrin is the primary compound used for this purpose. However, landscape treatment was unlikely to have been the major bifenthrin source. Reported landscape use of bifenthrin by professional applicators was very low, with only 6.2 kg used county-wide in 2003.

In 2003, 4463.5 kg of pyrethroids was used by professional applicators in Placer County for structural pest control (i.e., in or around the exterior perimeter of homes and other structures). Cypermethrin comprised 75% of the total, followed by permethrin (15%), cyfluthrin (6%), and bifenthrin (3%). The cypermethrin and permethrin products used have substantial below-ground use as termiticides where they would be less prone to runoff, though the product labeling does permit above-ground application as well. Reported structural pest control use of bifenthrin and cyfluthrin were primarily in products intended for above-ground treatment (bifenthrin = Talstar CA granular insecticide, Talstar lawn and tree flowable insecticide; cyfluthrin = Tempo 20 WP, Prescription Treatment brand Cy-Kick CS). Twice as much cyfluthrin was used as bifenthrin for structural pest control, but cyfluthrin concentrations in creek sediments were much lower than bifenthrin. The dominance of bifenthrin in the creeks is not consistent with structural pest control as its dominant source, however, differences in environmental persistence among the pyrethroids may confound this comparison. Bifenthrin half-life in sediments is about a year (*12*), but sediment persistence data on most other pyrethroids are lacking. Thus, it is uncertain if the dominance of bifenthrin in the sediments reflects greater input or greater persistence. Structural pest control could be a significant source for many of the pyrethroids (bifenthrin, cyfluthrin, cypermethrin, and permethrin), but it is not possible with available data to determine its relative magnitude, particularly for the bifenthrin that appears to be the major contributor to toxicity.

An alternative potential source of bifenthrin to the Pleasant Grove Creek system is landscape use by homeowners or their gardening services. Retail pesticide sales data are not publicly available, so it is not possible to quantify usage as was done for the other potential sources. However, the majority of bifenthrin-containing products available in retail outlets are granular products that are broadcast onto lawns using a spreader. Consumer surveys in California have found that about half of retail pesticide purchases are made at large home supply stores (*13*). In a shelf survey of a Home Depot store in the Roseville area, six insecticide products intended for lawn application were found, three of which contained bifenthrin as the active ingredient. One of these three products (Scott's Turf Builder with SummerGuard) is a mixture of bifenthrin and fertilizer; the other two (Ortho Basic Solutions Lawn and Garden Insect Killer, Ortho Bug-B-Gon Max Insect Killer for Lawns) are intended solely for use as insecticides for control of pests such as ants, mole crickets, ticks, and fleas. The remaining three available lawn products contained lambda-cyhalothrin (Spectracide Triazide Soil and Turf Insect Killer (granular)), esfenvalerate (Ortho Bug-B-Gon Max Lawn and Garden Insect Killer (liquid)), or permethrin (Ortho Basic Solutions Lawn and Garden Insect Killer (liquid)).

Using the bifenthrin-containing Ortho Bug-B-Gon Max Insect Killer for Lawns product as an example, if the product were applied to a 100-m^2 lawn at the recommended application rate (738 g product/100 m^2, containing 0.115% bifenthrin), off-site transport of a hypothetical 1% of the applied amount would equate to 8.5 mg of bifenthrin. This amount of bifenthrin would have to be dispersed in over 0.8 metric tons of sediment (dry weight) before the concentration would decrease below the *H. azteca* 10-d LC_{50} (*10*; assuming 2% oc), and even further dilution would be necessary to reach nontoxic concentrations. If the bifenthrin were in the dissolved phase, dilution with at least 2.2 million L of water would be required to reduce the concentration below that acutely lethal to sensitive aquatic species (*5*; given a 5th percentile LC_{50} of all aquatic species tested of <3.8 ng/L). These values used for lawn area and off-site transport are hypothetical, but they illustrate it is plausible that even a very small amount of product carried by irrigation runoff from the lawn to which it was applied could adversely affect sensitive aquatic life in nearby creeks.

This study documented the presence of pyrethroids in the sediments of creeks within a residential neighborhood, and it is possible to identify likely sources, though further work will be necessary to determine their relative magnitudes.

The compounds of greatest concern are bifenthrin, and secondarily, cyfluthrin and cypermethrin, and their potential sources appear to be limited to structural applications by professional pest control applicators and homeowner use of insecticides, particularly lawn care products. The question arises as to whether these results are unique to Roseville, California or representative of suburban systems in general. Factors such as high-density housing, the cultural emphasis on intensive lawn and landscape care, and efficient storm drain systems that carry irrigation runoff directly to nearby creeks all undoubtedly play a role. However, none of these factors are unique to Roseville, but are typical of countless suburban communities across the United States.

One factor that may exacerbate conditions in the study area is the low rainfall from May through October. During this period, much of the water and suspended sediment entering suburban creeks consists of runoff from landscape irrigation. This situation exists in much of California and other relatively dry regions in the western U.S., and suggests that degradation of sediment quality in suburban watersheds may be more severe in these areas. Other urban and suburban creeks in several additional California cities are under study, and acutely toxic concentrations of bifenthrin have been found in creeks in many of these communities as well (D. Weston, unpublished data). These results indicate that monitoring for pyrethroids in urban and suburban streams is overdue, and that the public, regulators, and scientific community should give greater consideration to the potential effects of residential use of pyrethroid pesticides on aquatic systems.

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Supporting Information Available

Mortality and growth data for the toxicity tests (Table S1), chemistry data for analytes other than pyrethroids (Table S2), and abundance data for resident *H. azteca* (Table S3). This material is available free of charge via the Internet at http://pubs.acs.org.

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