Aquatic Life Water Quality Criteria Derived via the UC Davis Method: II. Pyrethroid Insecticides

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1 Introduction

Pyrethroid insecticides are broad spectrum agents that have been widely detected in sediments and surface waters in the USA (Amweg et al. 2006; Budd et al. 2007; Gan et al. 2005; Hladik and Kuivila 2009; Weston et al. 2004). They are hydrophobic compounds that primarily partition to sediments and solid materials in the water column, and exposure to pyrethroid-contaminated sediments has been demonstrated to produce toxicity in the environment (Anderson et al. 2006; Holmes et al. 2008; Phillips et al. 2010; Weston et al. 2004; Weston et al. 2005). Only very low concentrations are found freely dissolved in the aqueous phase, but these pesticides are still of concern to water quality managers because they exhibit toxicity to aquatic organisms at very low concentrations (<1 μg/L). Water quality regulators in the USA are required, under the Clean Water Act (section 303(c)(2)(B)), to provide numeric water quality criteria for priority pollutants that could reasonably be expected to interfere with the designated uses of a state's waters. Numeric water quality criteria are chemical concentrations in water bodies that should protect aquatic wildlife from the toxic effects of those chemicals, if these concentrations are not exceeded. Numeric criteria are derived using existing toxicity data; consequently, criteria calculation is dependent on the availability of these data. In the USA, there are currently no numeric criteria available for the pyrethroids, and many of the available pyrethroid data sets do not meet the requirements of the 1985 US Environmental Protection Agency (USEPA) criteria derivation methodology (USEPA 1985). One of the goals of developing the UC Davis methodology (UCDM) was to be able to derive criteria for compounds that do not meet all of the USEPA (1985) data requirements, such as the pyrethroids.

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The UCDM is an updated water quality criteria derivation methodology that was designed to be more flexible than the USEPA (1985) methodology, and to incorporate the results from new research in environmental toxicology and risk assessment. Like the USEPA (1985) method, the UCDM continues to recommend the use of a species sensitivity distribution (SSD) for criteria calculation, and an acute-to-chronic ratio (ACR) when chronic data are limited. The main procedures of the UCDM that differ from those of the USEPA method are that the UCDM provides for a thorough and transparent study evaluation procedure, a more advanced SSD, alternate procedures if data requirements for the SSD or ACR cannot be met, and a consideration for the toxicity of chemical mixtures. Previous publications have described why there was a need for a new methodology (TenBrook et al. 2009), the rationale behind the development of this new methodology, and detailed instructions for UCDM criteria derivation (TenBrook et al. 2010).

This paper is the second in a series in which water quality criteria were derived for nine pesticides: chlorpyrifos, diazinon, malathion, bifenthrin, cyfluthrin, cypermethrin, λ-cyhalothrin, permethrin, and diuron. In this article, we describe the derivation of water quality criteria for five pyrethroid insecticides (bifenthrin, cyfluthrin, cypermethrin, λ-cyhalothrin, and permethrin) according to the UCDM; we have also extended this review to render it wide ranging and useful as a review of the current knowledge regarding the risk to aqueous ecosystems of the pyrethroids' toxicity.

2 Data Collection and Evaluation

Bifenthrin ((2-methyl[1,1′-biphenyl]-3-yl)methyl (1*R*,3*R*)-rel-3-[(1*Z*)-2-chloro-3,3,3-trifluoro-1-propenyl]-2,2-dimethylcyclopropanecarboxylate), cyfluthrin (cyano(4-fluoro-3-phenoxyphenyl)methyl 3-(2,2-dichloroethenyl)-2,2-dimethylcyclopropanecarboxylate (unstated stereochemistry)), cypermethrin (cyano(3-phenoxyphenyl) methyl 3-(2,2-dichloroethenyl)-2,2-dimethylcyclopropanecarboxylate), λ-cyhalothrin ([1α(*S**), 3α (*Z*)]-(±)-cyano-(3-phenoxyphenyl)methyl 3-(2-chloro-3,3,3-trifluoro-1-propenyl)-2,2-dimethylcyclopropanecarboxylate), and permethrin ((3-phenoxyphenyl) methyl 3-(2,2-dichloroethenyl)-2,2-dimethylcyclopropanecarboxylate) are widely applied pyrethroid insecticides. Bifenthrin and permethrin are type I pyrethroids while cyfluthrin, cypermethrin, and λ-cyhalothrin are type II pyrethroids (containing an α-cyano moiety); the two types are distinguished by slightly different toxicological mechanisms (Breckenridge et al. 2009). These pyrethroids are hydrophobic organic compounds that are moderately persistent (see Tables S1 and S2 of the Supporting Material http://extras.springer.com/). Based on their physical–chemical properties (Table 1), they are likely to partition to sediments from the aqueous phase, and are not likely to volatilize.

Aquatic toxicity effects studies were identified in the peer-reviewed open literature and from unpublished studies submitted to the USEPA and California Department of Pesticide Regulation (CDPR) for bifenthrin (~40), cyfluthrin (~53),

Table 1 Physical–chemical properties of five selected pyrethroids

	Bifenthrin	Cyfluthrin	Cypermethrin	λ-Cyhalothrin	Permethrin
Molecular weight	422.87	434.3	416.3	449.850	391.288
Density (g/mL)	1.21 (geomean, $n = 2$)	1.28[d] (20°C)	1.24[d] (20°C)	1.33 (25°C[d,f])	1.23 (geomean, $n = 2$)
Water solubility (mg/L)	0.001 (geomean, $n = 2$)	0.0023[e] (20°C)	0.004 (geomean, $n = 2$)	0.0047 (geomean, $n = 4$)	0.0057 (geomean, $n = 2$)
Melting point (°C)	69.3 (geomean, $n = 2$)	60[d]	71.2 (geomean of extremes)	48.3 (geomean of extremes)	36.4 (geomean of extremes)
Vapor pressure (Pa)	2.41×10^{-5} (geomean, $n = 2$)	2×10^{-6}[e]	2.87×10^{-7} (geomean, $n = 2$)	2.0×10^{-7} (20°C) (geomean, $n = 3$)	3.74×10^{-6} (geomean, $n = 4$)
Henry's law constant (K_H) (Pa m^3 mol^{-1})	0.24 (geomean, $n = 2$)	0.37[e]	0.0238 (geomean, $n = 3$)	1.96×10^{-2} (geomean, $n = 2$)	0.12 (geomean, $n = 2$)
Log K_{oc}[a]	5.29 (geomean, $n = 7$)	5.09[e] (mean, $n = 4$)	5.49[e] (mean, $n = 3$)	5.52 (geomean, $n = 2$)	5.12 (geomean, $n = 2$)
Log K_{ow}[b]	6.00[c]	5.97[e] (mean, $n = 4$)	6.57 (geomean, $n = 2$)	7.0[d,e,f]	6.3 (geomean, $n = 2$)

[a] Log-normalized organic carbon–water partition coefficient
[b] Log-normalized octanol–water partition coefficient
[c] Sangster Research Laboratories (2010)
[d] Tomlin (2003)
[e] Laskowski (2002)
[f] Mackay et al. (2006)

cypermethrin (~108), λ-cyhalothrin (~65), and permethrin (~155). Each study was reviewed according to the UCDM paradigm to determine the usefulness of these studies for criteria derivation. Studies were divided into three categories to be rated: (1) single-species effects, (2) ecosystem-level studies, and (3) terrestrial wildlife studies.

The UCDM provides a detailed numeric rating scheme for single-species effects studies that assigns (1) a relevance score and (2) a reliability score, which is summarized in the first chapter of this volume (Palumbo et al. (2012)). The possible relevance scores were relevant (R), less relevant (L), or not relevant (N). The studies rated N were deemed irrelevant for criteria derivation, and only the relevant (R) and less relevant (L) studies were evaluated for reliability. For all studies, study details and scoring were summarized in data summary sheets (Supporting Material http://extras.springer.com/). The reliability evaluation assigned possible scores of reliable (R), less reliable (L), or not reliable (N) so that each single-species study is described by a two-letter code, corresponding to the relevance and reliability ratings. The only studies used directly in criteria

calculation were those rated as relevant and reliable (RR), which are summarized in Table 11. Studies that were rated as relevant and less reliable (RL), less relevant and reliable (LR), or less relevant and less reliable (LL) were used to evaluate the derived criteria against data for any particularly sensitive, threatened, or endangered species found in these data sets. Studies that were rated N for either relevance or reliability were not considered in any aspect of criteria derivation.

Multispecies studies conducted in mesocosms, microcosms, and other field and laboratory ecosystems were rated for reliability. The results of the studies that were rated reliable (R) or less reliable (L) were compared to the derived criteria to ensure that they are protective of ecosystems. Studies of the effects of pyrethroids on mallard ducks were rated for reliability using the terrestrial wildlife evaluation. Mallard studies rated as reliable (R) or less reliable (L) were used to consider bioaccumulation of pyrethroids.

3 Data Reduction

As described in Palumbo et al. (2012), multiple toxicity values for a given species in the acceptable data set were combined into one species mean acute value (SMAV) or one species mean chronic value (SMCV). Some data that were rated RR were excluded from the final data set for one or more of the following reasons: flow-through tests are preferred over static tests, a test with a more sensitive life stage of the same species was available, more appropriate exposure durations were available, and tests with more sensitive end points were available (Tables S3–S6, Supporting Material http://extras.springer.com/). For bifenthrin, the final acceptable data sets contain 8 SMAVs and 2 SMCVs (Tables 2 and 3), the final cyfluthrin data sets contain 8 SMAVs and 3 SMCVs (Tables 4 and 5), the final cypermethrin data sets contain 14 SMAVs and 1 SMCV (Tables 6 and 7), the final λ-cyhalothrin data sets contain 20 SMAVs and 2 SMCVs (Tables 8 and 9), and the final permethrin data sets contain 19 SMAVs and 3 SMCVs (Tables 10 and 11).

4 Acute Criterion Calculations

An acute data set must have species representing five taxa to use a SSD to calculate the acute criterion; the five taxa are a warm water fish, a species in the family Salmonidae, a planktonic crustacean, a benthic crustacean, and an insect. The final acute data sets for each of the five pyrethroids (Tables 2, 4, 6, 8, and 10) met the five taxa requirement. Log-logistic distributions were fit to the bifenthrin and cyfluthrin acute data sets using the ETX 1.3 software (Aldenberg 1993) because there were between five and eight SMAVs in each of these data sets. The Burr Type III distribution was fit to the acute λ-cyhalothrin and permethrin data sets because there were more than eight SMAVs in these data sets. Of the three related distributions in the Burr Type III SSD, the Burr III

Table 2 Final acute toxicity data set for bifenthrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC/ EC_{50} (µg/L)	Reference
Ceriodaphnia dubia	SR	Est	97.8	96	24.0–24.7	Mortality	<24 h	0.078	Guy (2000a)
C. dubia	S	Nom	97.0	48	25	Mortality	<24 h	0.142	Wheelock et al. (2004)
Geometric mean								0.105	
Chironomus dilutes	FT	Nom	100	96	23 ± 1	Mortality	Third instar	2.615	Anderson et al. (2006)
Daphnia magna	FT	Nom	88.4	48	20–21	Mortality	<24 h	1.6	Surprenant (1983)
Hyalella azteca	S	Nom	100.0	96	23 ± 1	Mortality	7–14 days	0.0093	Anderson et al. (2006)
H. azteca	SR	Est	98	96	23 ± 1	Mortality	7–14 days	0.0027	Weston and Jackson (2009)
H. azteca	SR	Est	98	96	23 ± 1	Mortality	7–14 days	0.0073	Weston and Jackson (2009)
H. azteca	SR	Est	98	96	23 ± 1	Mortality	7–14 days	0.0080	Weston and Jackson (2009)
H. azteca	SR	Est	98	96	23 ± 1	Mortality	7–14 days	0.0082	Weston and Jackson (2009)
Geometric mean								0.0065	
Lepomis macrochirus	FT	Nom	88.4	96	21–22	Mortality	2.5 g, 8 mm	0.35	Hoberg (1983a)
Oncorhynchus mykiss	FT	Nom	88.4	96	11–12	Mortality	1.0 g, 46 mm	0.15	Hoberg (1983b)
Pimephales promelas	S	Meas	96.2	96	25 ± 1	Mortality	40 days, 0.059 g	0.21	McAllister (1988)
P. promelas	SR	Est	97.8	96	24.0–24.5	Mortality	8 days, 0.0039–0.0052 g	0.78	Guy (2000b)
Geometric mean								0.405	
Procloeon sp.	S	Nom	100.0	48	23 ± 1	Mortality	0.5–1.0 cm	0.0843	Anderson et al. (2006)

All studies were rated relevant and reliable (RR)

Est Toxicity values were calculated based on estimated concentrations (calculated from the recovery of some concentrations), *S* static, *SR* static renewal, *FT* flow through

Table 3 Final chronic toxicity data set for bifenthrin

Species	Test type	Meas/Nom	Chemical grade (%)	Duration (days)	Temp (°C)	End point	Age/size	NOEC (µg/L)	LOEC (µg/L)	MATC (µg/L)	Reference
Daphnia magna	FT	Meas	97.0	21	19–22	Reproduction	<24 h	0.0013	0.0029	0.0019	Burgess (1989)
Pimephales promelas	FT	Meas	96.2	92	25	Mortality	<48 h	0.040	0.090	0.060	McAllister (1988)

All studies were rated relevant and reliable (RR)
FT flow through

Table 4 Final acute toxicity data set for cyfluthrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC/EC_{50} (μg/L) (95% CI)	Reference
Aedes aegypti Rockefellar	S	Nom	93.0	24	25	Mortality	Early fourth instar	1 (1–2)	Rodriguez et al. (2007)
A. aegypti Nicaragua	S	Nom	93.0	24	25	Mortality	Early fourth instar	0.5 (0.5–0.6)	Rodriguez et al. (2007)
A. aegypti Peru	S	Nom	93.0	24	25	Mortality	Early fourth instar	0.3 (0.1–0.4)	Rodriguez et al. (2007)
A. aegypti Geometric mean								0.5	
Ceriodaphnia dubia	S	Nom	97.0	48	25	Mortality	<24 h	0.344 ± 0.041	Wheelock et al. (2004)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.093 (0.050–0.146)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.136 (0.103–0.185)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.189 (0.112–0.292)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.134 (0.097–0.194)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.170 (0.121–0.229)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.145 (0.105–0.185)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.102 (0.027–0.395)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.159 (0.105–0.234)	Yang et al. (2007)
C. dubia	S	Nom	99.0	96	21	Mortality	<24 h	0.180 (0.127–0.280)	Yang et al. (2007)
Geometric mean								0.155	
Daphnia magna	FT	Meas	98.6	48	19	Mortality	<24 h (first instar)	0.16 (0.14–0.18)	Burgess (1990)
Hyalella azteca	SR	Est	98.0	96	23	Mortality	7–14 days	0.0017 (0.0011–0.0023)	Weston and Jackson (2009)
H. azteca	SR	Est	98.0	96	23	Mortality	7–14 days	0.0023 (0.0009–0.0028)	Weston and Jackson (2009)

(continued)

Table 4 (continued)

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC/EC_{50} (μg/L) (95% CI)	Reference
H. azteca	SR	Est	98.0	96	23	Mortality	7–14 days	0.0031 (0.0021–0.0046)	Weston and Jackson (2009)
Geometric mean								0.0023	
Lepomis macrochirus	FT	Meas	97.6	96	22	Mortality	0.82 g, 31.8 mm	0.998	Gagliano (1994)
Oncorhynchus mykiss	FT	Meas	97.6	96	11	Mortality	0.92 g, 39 mm	0.209	Gagliano and Bowers (1994)
O. mykiss	FT	Meas	97.6	96	12	Mortality	1.4 g, 43.3 mm	0.302 (0.240–0.432)	Bowers (1994)
Geometric mean								0.2512	
Pimephales promelas	FT	Meas	99.0	96	25	Mortality	30-day old	2.49	Rhodes et al. (1990)
Procambarus clarkii	FT	Meas	97.0	96	20	Mortality	0.59 g, 29 mm	0.062	Surprenant (1990)

All studies were rated relevant and reliable (RR). *Est* Toxicity values were calculated based on estimated concentrations (calculated from the recovery of some concentrations)

S static, *SR* static renewal, *FT* flow through, *95% CI* 95% confidence interval

Table 5 Final chronic toxicity data set for cyfluthrin

Species	Test type	Meas/ Nom	Chemical (%)	Duration (days)	Temp (°C)	End point	Age/ size	NOEC (µg/L)	LOEC (µg/L)	MATC (µg/L)	Reference
Daphnia magna	FT	Meas	94.7	21	20	Reproduction (young/ female/day)	<24 h	0.020	0.041	0.02864	Forbis et al. (1984)
D. magna	FT	Meas	94.7	21	20	Length	<24 h	0.020	0.041	0.02864	Forbis et al. (1984)
Geometric mean										0.02864	
Oncorhynchus mykiss	FT	Meas	96.0	58	9.4	Biomass/chamber	Eggs	0.01	0.0177	0.0133	Carlisle (1985)
O. mykiss	FT	Meas	96.0	58	9.4	Mean weight/fish	Eggs	0.01	0.0177	0.0133	Carlisle (1985)
Geometric mean										0.0133	
Pimephales promelas	FT	Meas	99.0	7–61	25	F_0 survival	Eggs	0.14	0.29	0.20	Rhodes et al. (1990)
P. promelas	FT	Meas	99.0	61–120	25	F_0 survival	Eggs	0.14	0.29	0.20	Rhodes et al. (1990)
P. promelas	FT	Meas	99.0	90	25	F_1 % hatch	Eggs	0.14	0.29	0.20	Rhodes et al. (1990)
P. promelas	FT	Meas	99.0	60	25	F_1 survival	Eggs	0.14	0.29	0.20	Rhodes et al. (1990)
Geometric mean										0.20	

All studies were rated relevant and reliable (RR)
S static, *SR* static renewal, *FT* flow through

Table 6 Final acute toxicity data set for cypermethin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC/EC_{50} (μg/L) (95% CI)	Reference
Aedes aegypti	S	Nom	>85	24	18	Mortality	Larvae	1 (0.4–4)	Stephenson (1982)
Asellus aquaticus	S	Nom	>85	24	15	Mortality	3–8 mm	0.2 (0.1–0.4)	Stephenson (1982)
Ceriodaphnia dubia	SR	Nom	>90	48	25	Mortality	<24 h	0.683 ± 0.072	Wheelock et al. (2004)
Chaoborus crystallinus	S	Nom	>85	24	15	Mortality	Larvae	0.2 (0.03–0.4)	Stephenson (1982)
Chironomus thummi	S	Nom	>85	24	15	Immobility	Larvae	0.2 (0.1–0.3)	Stephenson (1982)
Cloeon dipterum	S	Nom	>85	24	15	Mortality	Larvae	0.6 (0.3–1)	Stephenson (1982)
Corixa punctata	S	Nom	>85	24	15	Immobility	Adults	0.7 (0.4–2)	Stephenson (1982)
Daphnia magna	SR	Meas	92.3	48	20	Mortality	<24-h old	0.134 (0.114–0.157)	Ward and Boeri (1991)
D. magna	FT	Nom	95.7	48	20	Mortality	<24-h old	0.1615 (0.1344–0.1917)	Wheat and Evans (1994)
Geometric mean								0.147	
Gammarus pulex	S	Nom	>85	24	15	Mortality	3–8 mm	0.1 (0.08–0.2)	Stephenson (1982)

Gyrinus natator	S	Nom	>85	24	15	Immobility	Adults	0.07 (0.04–0.2)	Stephenson (1982)
Hyalella azteca	SR	Meas	>98	96	23	Mortality	7–14 days	0.0021 (0.0017–0.0025)	Weston and Jackson (2009)
H. azteca	SR	Meas	>98	96	23	Mortality	7–14 days	0.0023 (0.0013–0.0035)	Weston and Jackson (2009)
H. azteca	SR	Meas	>98	96	23	Mortality	7–14 days	0.0031 (0.0020–0.0044)	Weston and Jackson (2009)
H. azteca	SR	Nom	97.0	96	23	Mortality	Adults	0.0036 (0.002–0.0049)	Hamer (1997)
Geometric mean								0.0027	
Oncorhynchus mykiss	FT	Meas	91.5	96	12	Mortality	83-day-old juvenile	0.90 (0.72–1.35)	Vaishnav and Yurk (1990)
Oreochromis niloticus	FT	Meas	98.4	96	25	Mortality	0.6–3.0 g	2	Stephenson et al. (1984)
Piona carnea	S	Nom	>85	24	15	Mortality	Adults	0.05 (0.03–0.08)	Stephenson (1982)

All studies were rated relevant and reliable (RR)

S static, *SR* static renewal, *FT* flow through, *95% CI* 95% confidence interval

Table 7 Final chronic toxicity data set for cypermethrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (days)	Temp (°C)	End point	Age/size	NOEC (µg/L)	LOEC (µg/L)	MATC (µg/L)	Reference
Pimephales promelas	FT	Meas	93.1	60	25	Mortality	<48 h	0.077	0.15	0.11	Tapp et al. (1988)

All studies were rated relevant and reliable (RR)

S static, *SR* static renewal, *FT* flow through, *NR* not reported

Table 8 Final acute toxicity data set for λ-cyhalothrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC_{50}/EC_{50} (μg/L) (95% CI)	Reference
Asellus aquaticus	S	Nom	88.0	48	20	Immobility	NR	0.026 (0.018–0.036)	Hamer et al. (1998)
Brachydanio rerio	FT	Meas	88.7	96	25	Mortality	0.70 g, 36 mm	0.64 (0.48–0.90)	Kent and Shillabeer (1997c)
Ceriodaphnia dubia	S	Nom	97.0	48	25	Mortality	<24	0.200 ± 0.090	Wheelock et al. (2004)
Chaoborus sp.	S	Nom	88.0	48	20	Maintenance of body shape/ equilibrium	Larvae	0.0028 (0.0018–0.0041)	Hamer et al. (1998)
Cloeon dipterum	S	Nom	88.0	48	20	Immobility	Nymph	0.038 (0.023–0.093)	Hamer et al. (1998)
Corixa sp.	S	Nom	88.0	48	20	Immobility	NR	0.030 (0.021–0.042)	Hamer et al. (1998)
Cyclops sp.	S	Nom	88.0	48	20	Immobility	NR	0.300 (0.200–0.460)	Hamer et al. (1998)
Daphnia magna	FT	Meas	94.3	72	20	Mortality	<24 h	0.013 (0.010–0.017)	Farrelly and Hamer (1989)
Gammarus pulex	FT	Meas	99.2	96	15	Immobility	5 mm, >3 weeks	0.0059	Hamer et al. (1985a)
Gasterosteus aculeatus	FT	Meas	87.7	96	12	Mortality	0.41 g, 34 mm	0.40 (0.33–0.50)	Long and Shillabeer (1997a)
Hyalella azteca	S	Nom	88.0	48	20	Immobility	NR	0.0023 (0.0010–0.0078)	Hamer et al. (1998)
Hydracarina (Class)	S	Nom	88.0	48	20	Immobility	NR	0.047 (0.033–0.062)	Hamer et al. (1998)
Ictalurus punctatus	FT	Meas	87.7	96	17	Mortality	1.57 g, 48 mm	0.16 (0.13–0.20)	Long and Shillabeer (1997b)

(continued)

Table 8 (continued)

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC_{50}/EC_{50} (μg/L) (95% CI)	Reference
Lepomis macrochirus Rafinesque	FT	Meas	99.0	96	21.9	Mortality	Juvenile	0.106 (0.0855–0.140)	Marino and Rick (2001)
L. macrochirus	FT	Meas	98.0	96	22	Mortality	1.51 g, 38.2 mm	0.21 (0.18–0.25)	Hill (1984b)
Geometric mean								0.15	
Leuciscus idus	FT	Meas	88.7	96	12	Mortality	2.15 g, 53 mm	0.078 (0.056–0.11)	Kent and Shillabeer (1997a)
Oncorhynchus mykiss	FT	Meas	99.0	96	12	Mortality	39 mm, 0.52 g	0.19 (0.16–0.20)	Machado (2001)
O. mykiss	FT	Meas	81.5	96	12	Mortality	43 mm, 1.12 g	0.44 (0.38–0.51)	Tapp et al. (1989)
O. mykiss	FT	Meas	98.0	96	12	Mortality	38.3 mm, 0.83 g	0.24 (0.08–0.70)	Hill (1984a)
Geometric mean								0.27	
Ostracoda (class)	S	Nom	88.0	48	20	Immobility	NR	3.300 (2.100–6.600)	Hamer et al. (1998)
Pimephales promelas	FT	Meas	97.0	96	25	Mortality	Larvae	0.360 (0.252–0.765)	Tapp et al. (1990)
P. promelas	FT	Meas	88.7	96	25	Mortality	0.37 g, 28 mm	0.70 (0.38–1.3)	Kent and Shillabeer (1997d)
Geometric mean								0.50	
Poecilia reticulata	FT	Meas	88.7	96	25	Mortality	0.62 g, 33 mm	2.3 (1.8–3.1)	Kent and Shillabeer (1997b)
Procambarus clarkii	SR	Nom	99.1	96	21.7	Mortality	3-month old	0.16 (0.06–0.27)	Barbee and Stout (2009)

All studies were rated relevant and reliable (RR)
S static, *SR* static renewal, *FT* flow through

Table 9 Final chronic toxicity data set for λ-cyhalothrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (days)	Temp (°C)	End point	Age/size	NOEC (μg/L)	LOEC (μg/L)	MATC (μg/L)	Reference
Daphnia magna	FT	Meas	94.3	21	20	Reproduction (young/ female/ day)	<24 h	0.00198	0.00350	0.00263	Farrelly and Hamer (1989)
D. magna	SR	Meas	94.3	21	20	Reproduction (young/ female/ day)	<24 h	0.00375	0.00490	0.00429	Hamer et al. (1985b)
Geometric mean										0.00336	
Pimephales promelas	FT	Meas	97.0	56	25	F_1 survival	F_1 larvae	0.031	0.062	0.044	Tapp et al. (1990)

All studies were rated relevant and reliable (RR)
SR static renewal, *FT* flow through

Table 10 Final acute toxicity data set for permethrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (h)	Temp (°C)	End point	Age/size	LC/EC_{50} (μg/L) (95% CI)	Reference
Ceriodaphnia dubia	S	Nom	99.0	48	25	Mortality	<24 h	0.250 (±119)	Wheelock et al. (2004)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.652 (0.484–0.856)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.788 (0.545–1.040)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.622 (0.427–0.824)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.772 (0.574–1.013)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.745 (0.568–0.957)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.858 (0.591–1.138)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.571 (0.427–0.740)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.580 (0.407–0.718)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.609 (0.486–0.747)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.570 (0.459–0.689)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.827 (0.669–1.012)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.585 (0.677–0.793)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.849 (0.655–1.085)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.889 (0.666–1.120)	Yang et al. (2007)
C. dubia	S	Nom	99.3	96	21	Mortality	<24 h	0.865 (0.672–1.098)	Yang et al. (2007)
Geometric mean						Mortality		0.664	
Chironomus dilutus	S	Meas	>96	96	23	Mortality	Fourth instar	0.189 (0.131–0.295)	Harwood et al. (2009)
Danio rerio	SR	Nom	90.0	96	23	Mortality	3.0 cm, 0.3 g	2.5 (1.7–3.2)	Zhang et al. (2010)
Daphnia magna	S	Nom	Technical	48	22	Immobility	<24 h	0.32 (0.24–0.44)	LeBlanc (1976)
Erimonax monachus	S	Nom	95.2	96	17	Mortality	NR	1.7	Dwyer et al. (2005)
Etheostoma fonticola	S	Nom	95.2	96	22	Mortality	62 mg, 20.2 mm	3.34 (2.75–4.16)	Dwyer et al. (1999, 2005)
Etheostoma lepidum	S	Nom	95.2	96	22	Mortality	NR	2.71 (2.36–3.13)	Dwyer et al. (1999, 2005)
Hyalella azteca	S	Nom	100.0	96	23	Mortality	Third instar	0.0211	Anderson et al. (2006)

Ictalurus punctatus	S	Nom	92.4	96	21	Mortality	1.2 g, 35 mm	5.4 (3.9–7.4)	Buccafusco (1976a)
Notropis mekistocholas	S	Nom	95.2	96	17	Mortality	NR	4.16	Dwyer et al. (2005)
Oncorhynchus apache	S	Nom	95.2	96	12	Mortality	0.615 g	1.71 (1.3–2.2)	Dwyer et al. (1995, 2005), Sappington et al. (2001)
Oncorhynchus clarki henshawi	S	Nom	95.2	96	12	Mortality	0.46 g	1.58 (1.1–2.2)	Dwyer et al. (1995, 2005), Sappington et al. (2001)
Oncorhynchus mykiss	FT	Meas	91.9	96	15.6	Mortality	Juvenile	7.0	Holcombe et al. (1982)
Orconectes immunis	S	Nom	92.0	96	16.5	Mortality	Juvenile 2 g	0.21 (0.17–0.25)	Paul and Simonin (2006)
Pimephales promelas	S	Nom	95.2	96	22	Mortality	0.41 g	9.38 (6.7–16)	Dwyer et al. (1995, 2005), Sappington et al. (2001)
Procambarus blandingi	FT	Nom	89.1	96	22	Mortality	24 g, 48 mm	0.21 (0.13–0.33)	Buccafusco (1977)
Procloeon sp.	S	Nom	100.0	48	23	Mortality	0.5–1 cm	0.0896	Anderson et al. (2006)
Salmo salar	S	Nom	92.4	96	12	Mortality	1 g, 35 mm	1.5 (1.1–2.0)	Buccafusco (1976b)
Xyrauchen texanus	S	Nom	95.2	96	22	Mortality	0.32 g	5.95 (4.6–7.7)	Dwyer et al. (1995, 2005), Sappington et al. (2001)

All studies were rated relevant and reliable (RR)
S static, *SR* static renewal, *FT* flow through

Table 11 Final chronic toxicity data set for permethrin

Species	Test type	Meas/ Nom	Chemical grade (%)	Duration (days)	Temp (°C)	End point	Age/size	NOEC (μg/L)	LOEC (μg/L)	MATC (μg/L)	Reference
Brachycentrus americanus	FT	Meas	Technical	21	15	Mortality	Larvae	–	–	LC_{50}: 0.17 (0.09–0.34)	Anderson (1982)
Daphnia magna	FT	Meas	98.6	21	20	Reproduction	<24 h	0.039	0.084	0.057	Kent et al. (1995a)
D. magna	FT	Meas	98.6	21	20	Length	<24 h	0.039	0.084	0.057	Kent et al. (1995a)
D. magna										0.057	
Pimephales promelas	FT	Meas	92.0	32	25	Mortality	4–5-day-old larvae	0.66	1.4	0.96	Spehar et al. (1983)

All studies were rated relevant and reliable (RR)
S static, *SR* static renewal, *FT* flow through, *NR* not reported

distribution was selected as the best fit for both λ-cyhalothrin and permethrin based on maximum likelihood estimation using the BurrliOZ software (CSIRO 2001). Fit tests based on cross validation and Fisher's combined test found no significant lack of fit for bifenthrin, cyfluthrin, λ-cyhalothrin, or permethrin, with $X^2_{2n} > 0.199$ for these four compounds (calculations shown in the Supporting Material http://extras.springer.com/). The Burr III distribution was initially selected as the best fit for the cypermethrin data set, but this distribution did not provide a satisfactory fit based on the fit test ($\chi^2_{2n} = 0.000014$; calculations shown in the Supporting Material http://extras.springer.com/); so a log-logistic distribution, which is less likely to overfit the data, was fit to the cypermethrin data set instead.

Acute values were derived from the distributions, including fifth percentiles (median and lower 95% confidence limit), as well as first percentiles (median and lower 95% confidence limit). The median fifth percentile is the most robust of the four distributional estimates, and is therefore the estimate recommended for criteria calculation.

Bifenthrin Log-Logistic Distribution

HC5 fitting parameters: $\alpha = -0.661$; β (median) = 0.4872, β (lower 95% CI) = 0.9328

Fifth percentile, 50% confidence limit: 0.00803 μg/L
Fifth percentile, 95% confidence limit: 0.000391 μg/L
First percentile, 50% confidence limit: 0.00126 μg/L
First percentile, 95% confidence limit: 0.0000113 μg/L
Recommended acute value: 0.00803 μg/L (median fifth percentile)

$$\text{Acute criterion} = \frac{\text{Acute value}}{2}. \quad (1)$$

Bifenthrin acute criterion = 0.004 μg/L

Cyfluthrin Log-Logistic Distribution

HC5 fitting parameters: $\alpha = -0.7446$; β (median) = 0.5478; β (lower 95% CI) = 1.04898

Fifth percentile, 50% confidence limit: 0.00439 μg/L
Fifth percentile, 95% confidence limit: 0.000147 μg/L
First percentile, 50% confidence limit: 0.000547 μg/L
First percentile, 95% confidence limit: 0.0000027 μg/L
Recommended acute value: 0.00439 μg/L (median fifth percentile)
Cyfluthrin acute criterion = 0.002 μg/L

Cypermethrin Log-Logistic Distribution

HC_5 fitting parameters: $\alpha = -0.6601$, β(median) = 0.4199, β(lower 95% CI) = 0.6768

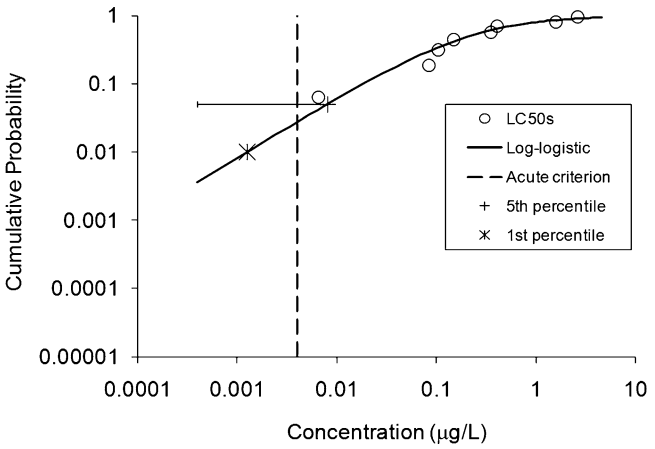

Fig. 1 Plot of bifenthrin species mean acute values and fit of the log-logistic distribution. The graph shows the median fifth and first percentiles with the lower 95% confidence limit on the fifth percentile and the acute criterion at 0.004 µg/L

Fifth percentile, 50% confidence limit: 0.0127 µg/L
Fifth percentile, 95% confidence limit: 0.00222 µg/L
First percentile, 50% confidence limit: 0.00257 µg/L
First percentile, 95% confidence limit: 0.000170 µg/L
Recommended acute value: 0.0127 µg/L (median fifth percentile)
Cypermethrin acute criterion = 0.006 µg/L

λ-Cyhalothrin Burr III Distribution

Fit parameters: $b = 0.232356$; $c = 1.100750$; $k = 0.596085$ (likelihood = −4.987264)

Fifth percentile, 50% confidence limit: 0.00243 µg/L
Fifth percentile, 95% confidence limit: 0.000501 µg/L
First percentile, 50% confidence limit: 0.000208 µg/L
Recommended acute value: 0.002432 µg/L (median fifth percentile)
λ-Cyhalothrin acute criterion = 0.001 µg/L

Permethrin Burr III Distribution

Fit parameters: $b = 7.80465$; $c = 6.599725$; $k = 0.07608$ (likelihood = 35.742158)

Fifth percentile, 50% confidence limit: 0.020008 µg/L
First percentile, 50% confidence limit: 0.000811 µg/L
Recommended acute value: 0.020008 µg/L (median fifth percentile)
Permethrin acute criterion = 0.01 µg/L

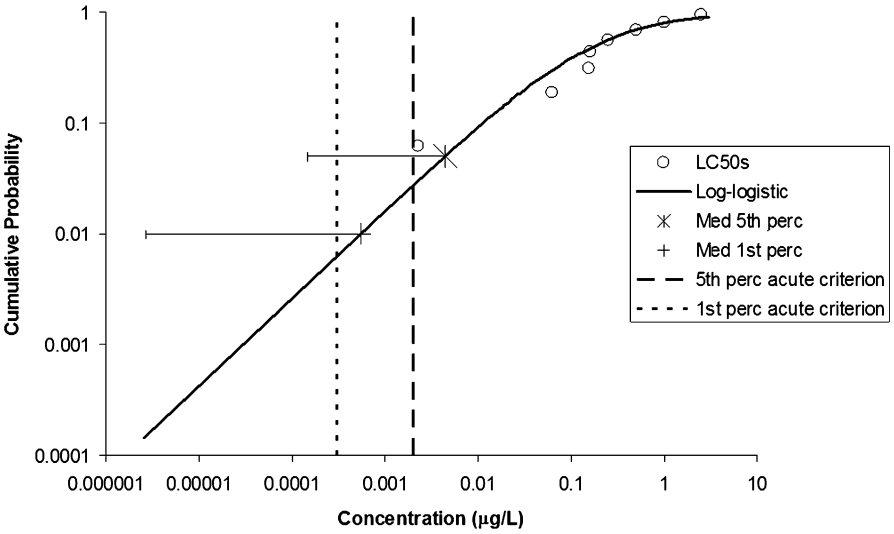

Fig. 2 Plot of cyfluthrin species mean acute values and fit of the log-logistic distribution. The graph shows the median fifth and first percentile values with the lower 95% confidence limits and the acute criteria calculated using both the median fifth percentile value and the median first percentile value

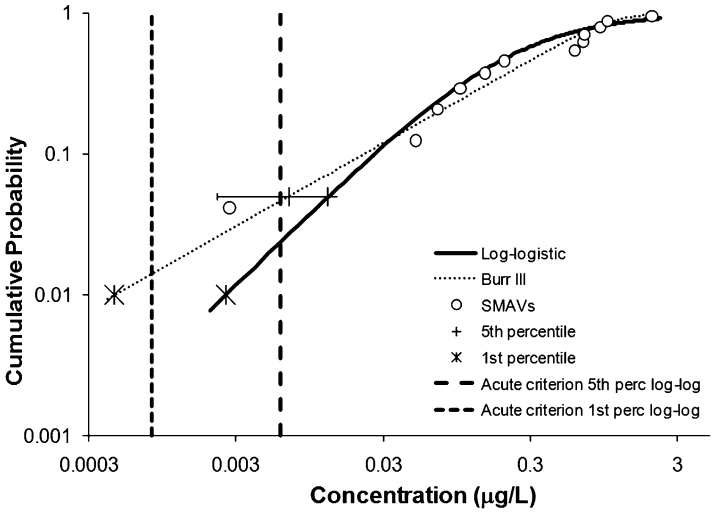

Fig. 3 Plot of species mean acute values for cypermethrin and fit of the log-logistic and Burr Type III distributions. The graph shows the median fifth and first percentiles for both distributions with the lower 95% confidence limit for the median fifth percentile on the log-logistic, and the acute criteria calculated using both the median fifth and first percentiles of the log-logistic distribution

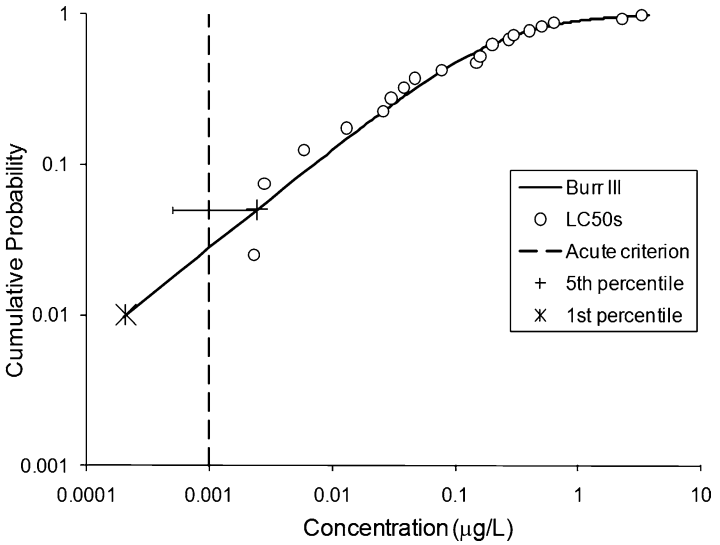

Fig. 4 Plot of species mean acute values for λ-cyhalothrin and fit of the Burr III distribution. The graph shows the median fifth and first percentile values with the lower 95% confidence limit of the fifth percentile and the acute criterion at 0.001 μg/L

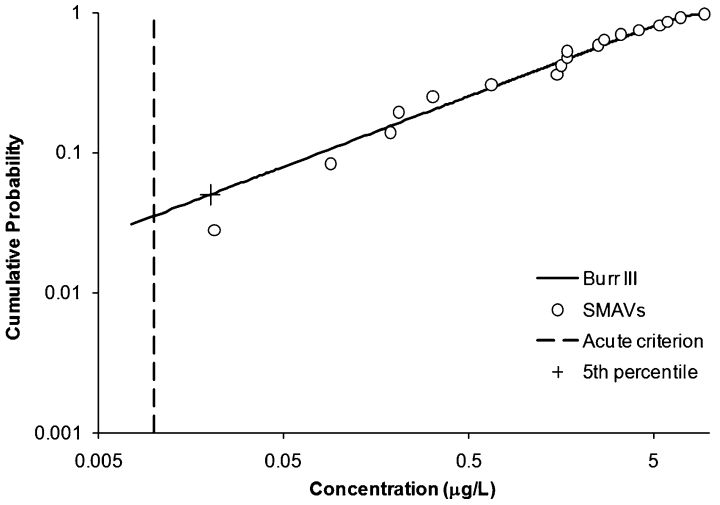

Fig. 5 Plot of species mean acute values for permethrin and fit of the Burr III distribution. The graph shows the median fifth percentile and the acute criterion at 0.01 μg/L

The fits of the distributions to the acute data sets are shown in Figs. 1–5, in cumulative probability plots. Because there is variability between the first significant digits of the median and lower 95% confidence limit estimates, the final criteria are reported with one significant figure. Although a lower 95% confidence limit could not be calculated for the permethrin distribution for comparison, the permethrin acute criterion is also reported with one significant digit because there was variability in the first digit of the fifth percentiles generated in the fit test. Later in this chapter, the acute criteria for cyfluthrin and cypermethrin are recalculated using lower percentile estimates because the criteria for these compounds calculated with the median fifth percentile acute values were not protective when compared to data for sensitive species, threatened and endangered species, and multispecies ecosystem-level studies.

5 Chronic Criterion Calculations

The chronic data sets of all five pyrethroids were limited and did not include data that met the five taxa requirements of the SSD procedure. Instead, the ACR procedure (TenBrook et al. 2010) was used for chronic criteria calculation, which is based on the ACR procedure in the USEPA (1985) method, but also includes a default ACR, when ACR data is also limited. For bifenthrin and cypermethrin, only one or two of the five SSD taxa requirements were satisfied (Tables 3 and 7), and none of these values could be paired with an appropriate corresponding acute toxicity value to calculate ACRs. There were also no appropriate saltwater data to use for ACR calculation; thus, these chronic criteria were calculated with the default ACR of 12.4 (TenBrook et al. 2010). Three of the five cyfluthrin taxa requirements were satisfied: a species in the family Salmonidae (*Oncorhynchus mykiss*), a warm water fish (*Pimephales promelas*), and a planktonic crustacean (*Daphnia magna*). Each of these three chronic values were paired with appropriate corresponding acute toxicity values, which satisfied the three family requirements (a fish, an invertebrate, and another sensitive species) for the ACR procedure with measured toxicity data. There were two λ-cyhalothrin chronic toxicity values, satisfying two of the five taxa requirements: a warm water fish (*P. promelas*) and a planktonic crustacean (*D. magna*). Both freshwater chronic values were paired with corresponding acute toxicity values to calculate ACRs, and paired data for the saltwater species *Cyprinodon variegatus* was used to complete the third family requirement for the ACR procedure. While two chronic values were available for permethrin, neither could be paired with appropriate acute data, but paired data for the saltwater species *Americamysis bahia* was available. This ACR was combined with two default ACRs to complete the three ACR requirements, allowing for the calculation of a final ACR for permethrin.

To calculate species mean ACRs (SMACRs) for each species, the acute LC_{50} was divided by the chronic maximum acceptable toxicant concentration (MATC)

Table 12 Calculation of the final acute-to-chronic ratio for cyfluthrin

Species	LC_{50} (μg/L)	Acute reference	Chronic end point	MATC (μg/L)	Chronic reference	ACR (LC_{50}/MATC)
Daphnia magna	0.160	Burgess (1990)	Reproduction/length	0.02864	Forbis et al. (1984)	5.58659
Oncorhynchus mykiss	0.2512	Bowers (1994), Gagliano and Bowers (1994)	Biomass/weight	0.0133	Carlisle (1985)	18.88970
Pimephales promelas	2.49	Rhodes et al. (1990)	Various	0.20149	Rhodes et al. (1990)	12.35793
Multispecies ACR = geomean (individual ACRs)						10.27

Table 13 Calculation of the species mean acute-to-chronic ratios for λ-cyhalothrin

Species	LC_{50} (μg/L)	Chronic end point	MATC (μg/L)	Reference	ACR (LC_{50}/MATC)
Cyprinodon variegatus	0.81	Weight	0.31	Hill et al. (1985)	2.6129
Daphnia magna	0.013	Reproduction (young/female/day)	0.00263	Farrelly and Hamer (1989)	4.9430
Pimephales promelas	0.36	FI survival	0.044	Tapp et al. (1990)	8.1818
Multispecies ACR = geomean (individual ACRs)					4.73

Table 14 Acute-to-chronic ratios used for derivation of the permethrin chronic criterion

Species	LC_{50} (μg/L)	Acute reference	Chronic end point	MATC (μg/L)	Chronic reference	SMACR (LC_{50}/MATC)
Americamysis bahia	0.075	Thompson (1986)	Mortality	0.016	Thompson et al. (1989)	4.6875
Default						12.4[a]
Default						12.4[a]
Multispecies ACR = geomean (individual ACRs)						8.96592

[a] The derivation and source data of the default ACR are described in the UCDM (TenBrook et al. 2010)

for a given species. The final ACRs for cyfluthrin (10.27) and λ-cyhalothrin (4.73) were calculated as the geometric mean of all of the SMACRs in each ACR data set and the final ACR for permethrin (8.96592) was calculated with one SMACR and two default ACRs (Tables 12–14) while the default ACR of 12.4 was used for bifenthrin and cypermethrin. Chronic criteria were calculated by dividing the recommended acute value (median fifth percentile) by the final ACR. Later in this chapter, the cyfluthrin and cypermethrin are adjusted downward to be protective based on comparisons to data for sensitive, threatened, and endangered species and ecosystem-level studies.

Bifenthrin chronic criterion calculated with the acute median fifth percentile estimate:

Fifth percentile, 50% confidence limit: 0.00803 μg/L

$$\begin{aligned}\text{Chronic criterion} &= \frac{\text{Recommended acute value}}{\text{ACR}}, \\ &= \frac{0.0243\ \mu g/L}{12.4}, \\ &= 0.0006\ \ \mu g/L, \\ &= 0.6\ ng/L.\end{aligned} \tag{2}$$

Cyfluthrin chronic criterion calculated with the acute median fifth percentile estimate:

Fifth percentile, 50% confidence limit: 0.00439 μg/L

$$\begin{aligned}\text{Chronic criterion} &= \frac{0.00439\ \mu g/L}{10.27}, \\ &= 0.0004\ \ \mu g/L, \\ &= 0.4\ ng/L.\end{aligned}$$

Cypermethrin chronic criterion calculated with the acute median fifth percentile estimate:

Fifth percentile, 50% confidence limit: 0.0126904 μg/L

$$\begin{aligned}\text{Chronic criterion} &= \frac{0.0126904\ \mu g/L}{12.4}, \\ &= 0.001\ \ \mu g/L, \\ &= 1\ ng/L.\end{aligned}$$

λ-Cyhalothrin chronic criterion calculated with the acute median fifth percentile estimate:

Fifth percentile, 50% confidence limit: 0.00243 μg/L

$$\begin{aligned}\text{Chronic criterion} &= \frac{0.00243\ \mu g/L}{4.73}, \\ &= 0.0005\ \ \mu g/L, \\ &= 0.5\ ng/L.\end{aligned}$$

Permethrin chronic criterion calculated with the acute median fifth percentile estimate:

Fifth percentile, 50% confidence limit: 0.020008 μg/L

$$\begin{aligned}\text{Chronic criterion} &= \frac{0.020008\ \mu g/L}{8.96592}, \\ &= 0.002\ \ \mu g/L, \\ &= 2\ ng/L.\end{aligned}$$

6 Bioavailability

Although pyrethroids are not very soluble in water, aquatic organisms are very sensitive to the pyrethroids and toxicity does occur. Pyrethroids have been found as the cause of toxicity in surface waters in the California Central Valley (Phillips et al. 2007; Weston et al. 2009a; Weston and Lydy 2010). This toxicity is believed to occur primarily from the fraction of the compound that is dissolved in the water, not from the compound that is associated with particulate phases. For example, Surprenant (1988) demonstrated that bifenthrin from spiked soil samples was available at concentrations sufficient to cause toxicity to *D. magna* that were housed in a separate container from the sediment, but shared the same recirculating water (however, dissolved particles could have been involved in this exposure).

Numerous studies demonstrate that the uptake and toxicity of pyrethroids are greatly reduced when solids or dissolved organic matter (DOM) are present (Day 1991; DeLorenzo et al. 2006; Lajmanovich et al. 2003; Muir et al. 1985, 1994; Smith and Lizotte 2007; Yang et al. 2006a, b, c, 2007). These studies indicate that bound pyrethroids are unavailable, and thus nontoxic to aquatic organisms. It has been presumed that the pyrethroids primarily sorb to the organic carbon phase of solids or DOM, and Hunter et al. (2008) demonstrated that sediment OC-normalized concentrations of permethrin were highly correlated with the uptake of permethrin by *Chironomus dilutus* (formerly *C. tentans*), which supports this assumption. Yet Yang et al. (2007) did not find a direct correlation between dissolved organic carbon (DOC) content and uptake or toxicity of pyrethroids, indicating that partitioning is not solely dependent on the quantity of DOC, but is also dependent on the quality of the DOC. Consequently, to accurately estimate pyrethroid sorption to DOC and particulate OC in whole water, site-specific partition coefficients would be preferred.

Alternately, the bioavailable fraction of pyrethroids can be estimated by measuring only the freely dissolved concentration using solid-phase microextraction (SPME). Yang et al. (2006a, 2007) reported that organism uptake was closely mimicked by SPME results and that the aqueous concentration of pyrethroids measured by SPME correlated well with the variations in uptake and toxicity with various DOM. Xu et al. (2007) clearly demonstrated that it is the freely dissolved aqueous concentration of pyrethroid that is bioavailable when they tested bifenthrin and cyfluthrin toxicity to *C. tentans* in 10-day sediment exposures with three types of sediment. The researchers reported LC_{50}s for five phases: bulk sediment, OC-normalized sediment, bulk pore water, DOC-normalized pore water, and freely dissolved pyrethroid. The LC_{50}s calculated for each of the five phases varied greatly, and varied between sediments for all phases tested except the freely dissolved, indicating that toxicity of the freely dissolved phase is independent of site-specific characteristics. The LC_{50}s based on the freely dissolved concentrations were at least an order of magnitude lower than those based on bulk pore water concentrations that included DOC, indicating that toxicity may be greatly underestimated if bioavailability is not taken into account. Based on these myriad studies, it can be concluded that the freely dissolved concentration is the most accurate predictor of toxicity and that bound pyrethroids were unavailable to the studied organisms.

However, bound pyrethroids can continue to desorb into the water column for long periods of time because pyrethroids have long equilibration times (~30 days; Bondarenko et al. 2006) and environmental systems are usually not at true equilibrium. The fraction of chemical that is potentially available to an organism is known as the bioaccessible fraction, and it has been linked to biological effects (Semple et al. 2004; You et al. 2011). Benthic organisms, such as *Hyalella azteca*, may be at greater risk because of their exposure to pore water and close proximity to sediments, where dissolved concentrations may persist.

Additionally, the role of dietary exposure on bioavailability of pyrethroids has not been considered. In the tests performed by Yang et al. (2006a, b) with *Ceriodaphnia dubia* and *D. magna*, organisms were not fed during the test. Organisms living in contaminated waters may also be ingesting food with sorbed hydrophobic compounds that can be desorbed by digestive juices (Mayer et al. 2001). The effects of dietary exposure may also be species specific, depending on typical food sources; some species may have greater interaction with particles, increasing their exposure. Palmquist et al. (2008) examined the effects due to dietary exposure of the pyrethroid esfenvalerate on three aqueous insects with different feeding functions: a grazing scraper (*Cinygmula reticulata* McDunnough), an omnivore filter feeder (*Brachycentrus americanus* Banks), and a predator (*Hesperoperla pacifica* Banks). The researchers observed adverse effects in *C. reticulata* and *B. americanus* after feeding on esfenvalerate-laced food sources and that none of the three insects avoided the contaminated food. The effects included reduced growth and egg production of *C. reticulata* and abandonment and mortality in *B. americanus*. Stratton and Corke (1981) tested toxicity of permethrin to *D. magna* with and without feeding of algae, and found that mortality at 24 h was significantly increased when daphnids were fed, although mortality at 48 h was not affected. The authors proposed that permethrin may have been ingested by the daphnids if it was sorbed on the algal cells, and caused increased toxicity, although the same effect was not seen when bacteria were provided as a food source. These limited studies indicate that ingestion may be an exposure route, but it is not currently possible to incorporate this exposure route into criteria compliance assessment.

The studies above suggest that the freely dissolved fraction of pyrethroids is the primary bioavailable fraction and that this concentration is the best indicator of toxicity; thus, it is recommended that the freely dissolved fraction is directly measured or calculated based on site-specific information for compliance assessment. The most direct way to determine compliance would be to measure the pyrethroid concentration in the dissolved phase to determine the total bioavailable concentration. SPME has shown to be a good predictor of pyrethroid toxicity in many studies (Bondarenko et al. 2007; Bondarenko and Gan 2009; Hunter et al. 2008; Xu et al. 2007; Yang et al. 2006a, b, c, 2007). Bondarenko and Gan (2009) reported method detection limits of 1.0 ng/L for bifenthrin, 2.0 ng/L for cyfluthrin, 2.0 ng/L for cypermethrin, 2.4 ng/L for λ-cyhalothrin, 2.0 ng/L for cis-permethrin, and 3.0 for trans-permethrin, and Li et al. (2009) reported method detection limits of 0.2 ng/L for bifenthrin, 0.2 for cyhalothrin, 0.9 ng/L for cyfluthrin, 1.0 ng/L for cypermethrin,

and 1.2 ng/L for permethrin using SPME. Analytical detection limits may create a problem for criteria compliance because most of these reported detection limits are above the derived criteria, meaning it is possible that one of these pyrethroids could be present in toxic amounts, yet below the detection limit so that an excursion is not identified. Filtration of suspended solids is not recommended for determining criteria compliance because pyrethroids have been demonstrated to adsorb to glass fiber filters by Gomez-Gutierrez et al. (2007). They found that on average 58% of a 50 ng/L solution of permethrin was lost on the filter; this magnitude of loss may be critical for determining compliance at environmental concentrations.

If the freely dissolved concentration is not directly measured, the following equation can be used to translate total pyrethroid concentrations measured in whole water to the associated dissolved pyrethroid concentrations:

$$C_{\text{dissolved}} = \frac{C_{\text{total}}}{1 + ((K_{\text{OC}} \times [\text{SS}])/f_{\text{oc}}) + (K_{\text{DOC}} \times [\text{DOC}])}, \tag{3}$$

where $C_{\text{dissolved}}$ is the concentration of chemical in dissolved phase (μg/L), C_{total} is the total concentration of chemical in water (μg/L), K_{OC} is the OC–water partition coefficient (L/kg), [SS] is the concentration of suspended solids in water (kg/L), f_{oc} is the fraction of OC in suspended sediment in water, [DOC] is the concentration of dissolved organic carbon in water (kg/L), and K_{DOC} is the OC–water partition coefficient (L/kg) for DOC.

To determine compliance by this calculation, site-specific data are necessary, including K_{OC}, K_{DOC}, concentration of suspended solids, concentration of DOC, and fraction of OC in the suspended solids. If all of these site-specific data, including the partition coefficients, are not available, then this equation should not be used for compliance determination. Site-specific data are required because the sorption of pyrethroid to suspended solids and DOM depends on the physical and chemical properties of the suspended solids resulting in a range of K_{OC} and K_{DOC} values, as discussed earlier in this section.

The freely dissolved pyrethroid concentration is recommended for determination of criteria compliance because the literature suggests that the freely dissolved concentrations are the most accurate predictor of toxicity. Environmental managers may choose an appropriate method for determining the concentration of freely dissolved pyrethroid. If environmental managers choose to measure whole water concentrations for criteria compliance assessment, the bioavailable fraction will likely be overestimated.

7 Chemical Mixtures

Pyrethroids often co-occur in the environment (Trimble et al. 2009; Werner and Moran 2008), and various other chemical mixtures are ubiquitous in surface waters. Because the presence of other chemicals can add to or alter the toxicity of another

given chemical, it is important to examine the effects of chemical mixtures on individual pyrethroid toxicity. Although chemical interactions are rarely straightforward, the concentration addition model is recommended for chemicals with the same toxicological mode of action. All pyrethroids have a similar mode of action in that they bind to and prolong the opening of voltage-dependent ion channels, causing convulsions, paralysis, and death (Brander et al. 2009). The three studies that tested toxicity of pyrethroid mixtures found that the effects were generally well-predicted by the concentration addition model (Barata et al. 2006; Brander et al. 2009; Trimble et al. 2009). Overall, the concentration addition model should be used by following either the toxic unit or relative potency factor approach to determine criteria compliance when multiple pyrethroids are present.

Barata et al. (2006) observed slight antagonism for *D. magna* survival for λ-cyhalothrin—deltamethrin mixtures, but the deviation from additivity was attributed to a few unexpected extreme values for joint survival effects, as most observed effects were within a factor of 2 of the effects predicted by the concentration addition model. Brander et al. (2009) tested mixture toxicity of cyfluthrin and permethrin, and found slight antagonism for the binary mixture, but additivity was demonstrated when piperonyl butoxide (PBO) was added. Brander et al. (2009) offered several explanations for the observed antagonism between the two pyrethroids. Permethrin is a type I pyrethroid and cyfluthrin is a type II pyrethroid, and type II pyrethroids may be able to outcompete type I pyrethroids for binding sites, which is known as competitive agonism; or binding sites may be saturated so that complete additivity is not observed. They also note that cyfluthrin is metabolized more slowly than permethrin, so cyfluthrin can bind longer. PBO may remove this effect because the rate of metabolism of both pyrethroids is reduced in its presence. To examine if pyrethroid mixture toxicity is additive with a more comprehensive study design, Trimble et al. (2009) performed sediment toxicity tests with *H. azteca* in three binary combinations: type I–type I (permethrin–bifenthrin), type II–type II (cypermethrin–λ-cyhalothrin), and type I–type II (bifenthrin–cypermethrin). The toxicity of these combinations was predicted with the concentration addition model, with model deviations within a factor of 2, indicating that in general pyrethroid mixture toxicity is additive.

PBO is commonly added to pyrethroid insecticide treatments because it is known to increase the toxic effects of pyrethroids (Weston et al. 2006). Many studies have demonstrated that the addition of PBO at a concentration that would be nonlethal on its own increases the toxicity of pyrethroids (Brander et al. 2009; Brausch and Smith 2009; Hardstone et al. 2007, 2008; Kasai et al. 1998; Paul and Simonin 2006; Paul et al. 2005, 2006; Rodriguez et al. 2005; Singh and Agarwal 1986; Xu et al. 2005). Several of these studies report single-species interaction coefficients (K; also called synergistic ratios) for pyrethroids and PBO ranging from 1.35 (*D. magna*; Brausch and Smith 2009) to 60 (snails; Singh and Agarwal 1986). While many studies report interaction coefficients for synergism of PBO, none of them reported interaction coefficients for multiple PBO concentrations; so a relationship between PBO concentration and K cannot be determined for any given species. In addition, no multispecies interaction coefficients are available; thus, there is no accurate way to account for synergism with PBO in compliance determination.

Mixture effects with pyrethroids and various other chemicals have also been studied and are summarized here, but there are currently no multispecies interaction coefficients available for these combinations. Binary mixtures of λ-cyhalothrin with deltamethrin and cadmium demonstrated additivity (Barata et al. 2006, 2007). Mixtures with various fungicides have been investigated and some synergism has been demonstrated. Norgaard and Cedergreen (2010) reported synergism with equitoxic mixtures of the fungicides and α-cypermethrin, yielding interaction coefficients ranging from 1.4 to 27, while other ratios tested resulted in interaction coefficients ranging from 0.41 to 37. Adam et al. (2009) also reported synergism for mixtures of fungicides and cypermethrin, which are often found in combination in wood preservatives. Permethrin in combination with propoxur, a carbamate, demonstrated synergism, which the authors propose is due to the complementary modes of action acting on different parts of the nervous system (Corbel et al. 2003). The thiocarbamate pesticide cartap appears to be antagonistic when combined with cypermethrin as no toxicity was observed in tests with *D. magna* and *Oryzias latipes*, when the concentrations of each chemical tested in combination were higher than the reported EC/LC_{50} values for the single chemicals (Kim et al. 2008). Gartenstein et al. (2006) reported synergism for cypermethrin in binary combinations with diflubenzuron and diazinon, but the combination of all three compounds produced an antagonistic effect. Zhang et al. (2010) tested mixtures of permethrin with the organophosphates dichlorvos or phoxim and reported that the toxicity of binary combinations was additive.

No studies on aquatic organisms were found in the literature that could provide a quantitative means to consider mixtures of pyrethroids with other classes of pesticides. Although there are examples of nonadditive toxicity, multispecies interaction coefficients are not available for any pyrethroid, and therefore the concentrations of nonadditive chemicals cannot be used for criteria compliance.

8 Water Quality Effects

Temperature has been reported to be inversely proportional to the aquatic toxicity and bioavailability of pyrethroids (Miller and Salgado 1985; Werner and Moran 2008). In fact, the increase of toxicity of pyrethroids with decreasing temperature has been used to implicate pyrethroids as the source of toxicity in environmental samples (Phillips et al. 2004; Weston et al. 2009b). The inverse relationship between temperature and pyrethroid toxicity is likely due to the increased sensitivity of an organism's sodium channels at lower temperatures (Narahashi et al. 1998).

Enhanced toxicity of cyfluthrin to larval fathead minnows (*P. promelas*) at lower temperatures was demonstrated by Heath et al. (1994). Sublethal cyfluthrin concentrations reduced the ability of fish to tolerate temperatures both higher and lower than standard conditions. The toxicities of six aqueous pyrethroids were 1.33- to 3.63-fold greater at 20°C compared to 30°C for mosquito larvae (Cutkomp and Subramanyam 1986). Harwood et al. (2009) tested permethrin toxicity to *C. dilutus* in an aqueous exposure at 13 and 23°C, and reported a

3.2-fold decrease of the 96-h LC_{50} at the lower temperature. Kumaraguru and Beamish (1981) reported that for small trout the toxicity of permethrin increased by a factor of 10 with a decrease in temperature from 20 to 5°C, but showed little change from 10 to 5°C. These studies indicate that the enhanced toxic effects of pyrethroids at lower temperature may not be as accurately represented by the results of typical laboratory toxicity tests, which tend to be run at warmer temperatures, 20–23°C (USEPA 1996a, b, 2000) than those of the habitats of coldwater fishes, about 15°C or lower (Sullivan et al. 2000).

The toxicity of sediments contaminated with pyrethroids was more than twice as toxic when tested at 18°C compared to 23°C (Weston et al. 2008). Weston et al. (2008) used a toxicity identification evaluation (TIE) procedure to determine the effect of temperature reduction (18 vs. 23°C) on toxicity of a particular environmental sediment sample to *H. azteca*. These results are not directly applicable for use in water quality criteria compliance because they were sediment exposures and used environmental samples, instead of an exposure to a pure compound.

Unfortunately, there are limited data in which aquatic exposures with relevant species were used, making it unfeasible to quantify the relationship between the toxicity of these five pyrethroids and temperature for water quality criteria at this time. Information regarding the effects of pH or other water quality parameters on pyrethroid toxicity was not identified, but based on the physical–chemical properties of these compounds they are not expected to be affected by these parameters.

9 Sensitive Species

Data for particularly sensitive species found in the acceptable (RR) and supplemental (RL, LR, LL) data sets (Tables S8–S12, Supporting Material http://extras.springer.com/) were compared to the criteria. There are some species represented in the supplemental data set that are not represented in the acceptable data set, and it is possible that data at the extreme sensitive end of the data set could be below the criteria derived using the median fifth percentiles. The bifenthrin acute criterion of 4 ng/L is below the lowest freshwater SMAV in the bifenthrin data sets (6.5 ng/L for *H. azteca*), and the chronic criterion of 0.6 ng/L is below the lowest freshwater SMCV in the data sets (1.9 ng/L for *D. magna*), so these criteria appear to be protective based on the available data. For λ-cyhalothrin, the acute and chronic criteria calculated with the acute median fifth percentile (1 and 0.5 ng/L, respectively) are both below all of the freshwater toxicity values in the respective acute and chronic data sets. The lowest LC_{50} is 2.3 ng/L for *H. azteca* while the lowest freshwater MATC is 2.63 ng/L for *D. magna*. For bifenthrin and λ-cyhalothrin, there are toxicity values equal to or below the derived criteria for the saltwater species *A. bahia*, but the criteria were not adjusted because they are only intended to protect freshwater species. The permethrin acute criterion (10 ng/L) is below the lowest acute value in the acute data sets (21.1 ng/L for *H. azteca*; Anderson et al. 2006). The permethrin

chronic criterion (2 ng/L) is below all of the chronic values in the available data sets (16 ng/L for *A. bahia*; Thompson et al. 1989).

The lowest SMAV in the cyfluthrin RR data set (Table 4) was 2.3 ng/L for *H. azteca*, which is approximately equal to the derived acute criterion of 2 ng/L. Based on the available data, the criterion derived using the median fifth percentile acute value is not protective of *H. azteca*; therefore, the next lowest acute value was used to calculate the cyfluthrin criteria. The acute and chronic cyfluthrin criteria calculations using the median first percentile acute value are as follows:

Recommended acute value: 0.000547 μg/L (median first percentile)

$$\begin{aligned}\text{Cyfluthrin acute criterion} &= \frac{0.000547\ \mu\text{g/L}}{2},\\ &= 0.0003\ \mu\text{g/L}\ (0.3\ \text{ng/L}).\\ \text{Cyfluthrin chronic criterion} &= \frac{0.000547\ \mu\text{g/L}}{10.27},\\ &= 0.00005\ \mu\text{g/L}\ (0.05\ \text{ng/L}).\end{aligned}$$

The cyfluthrin chronic criterion calculated with the median first percentile (0.05 ng/L) is below the lowest MATC in the data sets of 0.27 ng/L for *A. bahia*.

The derived cypermethrin acute criterion (0.006 μg/L) is higher than one SMAV in the RR acute data set, 0.0027 μg/L for *H. azteca* (Table 6). The *H. azteca* SMAV is the geometric mean of four values, three from a study in which concentrations were measured (Weston and Jackson 2009), all of which are lower than the acute criterion of 0.006 μg/L. Thus, the next lowest estimate from the log-logistic distribution (median first percentile) was used to derive the cypermethrin acute and chronic criteria as follows:

Recommended acute value: 0.0025723 μg/L (median first percentile)

$$\begin{aligned}\text{Cypermethrin acute criterion} &= \frac{0.0025723\ \mu\text{g/L}}{2},\\ &= 0.001\ \mu\text{g/L}\ (1\ \text{ng/L}).\\ \text{Cypermethrin chronic criterion} &= \frac{0.0025723\ \mu\text{g/L}}{12.4},\\ &= 0.0002\ \mu\text{g/L}\ (0.2\ \text{ng/L}).\end{aligned}$$

There is one supplemental datum (96-h EC_{50} = 0.6 ng/L for *D. magna*) that is below the adjusted cypermethrin acute criterion, but this toxicity value was not based on measured concentrations, and this species is represented in the RR data set with an SMAV that indicates that it is protected by the acute criterion. There are two supplemental MATCs that are below the adjusted chronic criterion of 0.2 ng/L (MATCs of 0.00063 and 0.063 ng/L for *D. magna*; Kim et al. 2008), but they are based on nominal concentrations, and it is recommended that criteria should only be adjusted based on toxicity values calculated with measured concentrations.

10 Ecosystem-Level Studies

Toxicity data from multispecies studies that more closely mimic ecosystems can yield different results than single-species effect studies, so community-level study data were compared to the criteria derived from single-species studies to ensure that the criteria are protective of ecosystems. A total of 28 studies addressing effects on microcosms, mesocosms, and model ecosystems were rated acceptable (R or L reliability rating) for the five selected pyrethroids (ratings listed in Table S13, Supporting Material http://extras.springer.com/). None of the bifenthrin, cyfluthrin, or permethrin studies reported ecosystem-level NOECs or no-effect concentrations (NECs) to which the chronic criteria could be directly compared.

Data in three of the bifenthrin studies (Drenner et al. 1993; Hoagland et al. 1993; Surprenant 1988) included toxic effects at concentrations ranging from 20 to 3,150 ng/L, which are well above the derived chronic criterion (0.6 ng/L). Sherman (1989) reported toxic effects for several invertebrates and fish in a pond receiving runoff contaminated with bifenthrin, but the effects did not correlate well with aqueous bifenthrin concentrations. Average pond concentrations fluctuated from slightly above 1 to 10 ng/L, but could not be linked to the occurrence of toxicity.

Authors of all the cyfluthrin studies reported toxic effects at applied or measured concentrations that were far above the chronic criterion (Gunther and Herrmann 1986; Johnson 1992; Johnson et al. 1994; Kennedy et al. 1990; Morris 1991; Morris et al. 1994). Toxic effects were observed in all of the studies, especially on aquatic macroinvertebrates, but it is not possible to assess if effects would have occurred if lower concentrations were tested, closer to the chronic criterion of 0.05 ng/L.

Several studies consisted of single concentrations of cypermethrin (0.01–24,000 μg/L) that were well above the chronic criterion in pond or marine mesocosms, followed by measurement of the recovery of the invertebrate communities. Toxic effects were observed particularly for insects and crustaceans, and some populations did not recover during the posttreatment observation periods (Crossland 1982; Farmer et al. 1995; Maund et al. 2009; Medina et al. 2004). The study by Maund et al. (2009) simulated natural reinvasion in some microcosms by adding invertebrates to the enclosures post treatment; in these microcosms, there was a general recovery of invertebrate populations in approximately 100 days. In contrast, the microcosms that received no additional organisms showed only limited recovery after 16 weeks of observation. These results indicate that small, isolated, or heavily impacted waterbodies will likely recover more slowly than waterbodies that are only partially impacted or are near other unimpacted waterbodies from which organisms can immigrate.

Friberg-Jensen et al. (2003) calculated cypermethrin NECs for crustaceans, copepods, and cladocerans ranging from 0.02 to 0.07 μg/L in enclosures set in a lake. These NECs are all significantly higher than the chronic criterion of 0.0004 μg/L. They also reported that rotifers, protozoans, bacteria, periphyton plankton, and periphytic algae all proliferated after treatment with cypermethrin, in response to the decreased populations of grazers. A sister paper, describing

effects for the same experiment, reported an NEC of 0.01 μg/L for copepod nauplii (Wendt-Rasch et al. 2003). This paper also reported significant changes to species composition of the aforementioned communities at nominal concentrations greater than 0.13 μg/L.

Several λ-cyhalothrin studies reported community NOECs to which the calculated criteria may be compared. Van Wijngaarden et al. (2006) and Roessink et al. (2005) reported various community-level NOECs that were season- and trophic-system-dependent, the lowest being <10 μg/L, and Schroer et al. (2004) reported a community-level NOEC of 10 ng/L. Schroer et al. (2004) also calculated a community-level criterion of 4.1 ng/L while the criterion calculated based on laboratory single-species data was 2.7 ng/L. The UCDM chronic criterion (0.5 ng/L) is below the reported NOECs for this set of studies by at least a factor of 20.

Hill et al. (1994) investigated the effects of λ-cyhalothrin on artificial pond mesocosms containing microbes, algae, macrophytes, zooplankton, macroinvertebrates, and fish. λ-cyhalothrin was applied at three rates as a spray and as a soil–water slurry to simulate runoff. Few effects were observed for most taxa, but macroinvertebrates and zooplankton were adversely affected at the highest rate; macroinvertebrates experienced some effects at the middle rate as well. Measured aqueous concentrations of λ-cyhalothrin ranged from 3 to 98 ng/L, in the mesocosms treated at the highest rate, and 2 to 10 ng/L in those treated at the middle rate. λ-cyhalothrin was not detected in the ponds treated at the lowest rate. The method detection limit reported in this study ranges from 2 to 3 ng/L, so it is possible that λ-cyhalothrin was present at lower concentrations when reported as nondetects. This study indicates that the derived chronic criterion of 0.5 ng/L should be protective of macroinvertebrates and zooplankton because it is likely similar to the actual concentrations in the ponds treated at the lowest rate.

Several study authors reported significant macroinvertebrate mortality and drift due to exposure to λ-cyhalothrin (Farmer et al. 1995; Lauridsen and Friberg 2005; Rasmussen et al. 2008; Wendt-Rasch et al. 2004), particularly for *Gammarus* species. Farmer et al. (1995) sprayed pond mesocosms with λ-cyhalothrin (measured at 2 ng/L, 1 h post treatment for the lower rate) and reported that *Gammarus* spp. abundance was significantly reduced compared to controls. Rasmussen et al. (2008) demonstrated that *Gammarus pulex* exposed to 10.65 ng/L λ-cyhalothrin (nominal) for 90 min and then transferred to clean water drifted significantly more than controls ($p<0.0001$). Phytoplankton and algae productivity increased in response to λ-cyhalothrin exposure (Farmer et al. 1995; Rasmussen et al. 2008; Wendt-Rasch et al. 2004) likely due to the decrease in macroinvertebrate populations, as macroinvertebrates are known to graze on algae. Lauridsen and Friberg (2005) examined macroinvertebrate drift in outdoor experimental channels with two insect species and *G. pulex*. Catastrophic drift was observed for all three species during the 1-h pulse exposure and 2–3-h post exposure. Drift of *G. pulex* was significantly affected at 1 ng/L (nominal), and it should be noted that the measured concentrations may have been even lower. While several studies indicate that *Gammarus* species experience lethal and sublethal effects due to λ-cyhalothrin exposures at concentrations near the chronic criterion, none of them reported toxicity values

(e.g., NOEC, EC_x) or measured concentrations at or below the derived chronic criterion; thus, the chronic criterion is not adjusted downward at this time.

In all permethrin studies, adverse effects were reported on aquatic organisms, but they all used formulations and test concentrations (0.02–100 μg/L) that were significantly higher than the chronic criterion of 0.002 μg/L. In two studies, increased drifting in model riverine systems was reported after exposure to permethrin for some invertebrate species (Poirier and Surgeoner 1988; Werner and Hilgert 1992), and another model riverine study reported that snails and water thyme (*Elodea*) were both adversely affected at permethrin concentrations of 4 and 20 μg/L (Lutnicka et al. 1999). Several pond exposures also demonstrated adverse effects on various aquatic invertebrates, including some populations that did not recover during the posttreatment observation period (Conrad et al. 1999; Coulon 1982; Yasuno et al. 1988). Conrad et al. (1999) dosed small artificial ponds with permethrin (1–100 μg/L nominal Picket® formulation) and conducted bioassays with chironomids, which were compared to laboratory sediment toxicity tests with *Chironomus riparius*. The chironomid responses of reduced larval density and adult emergence were not predicted by bulk sediment chemistry, sediment toxicity tests, or laboratory bioassay results—all three measurements underestimated the acute effects. Toxicity to *C. riparius* in the field was best predicted by acute water-only toxicity test data, indicating that the primary exposure route is via the water column. This study supports measurement of the truly dissolved fraction for criteria compliance and indicates the relevance of water quality criteria for protection of aquatic life.

11 Threatened and Endangered Species

Data for species listed as threatened or endangered were examined to ensure that the criteria are protective of these species. Both the US Fish and Wildlife Service federal list of threatened and endangered plant and animal species (USFWS 2010) and the California state list of threatened and endangered plant and animal species (the California Department of Fish and Game (CDFG) 2010a, 2010b) were consulted for this evaluation.

There are ten evolutionarily significant units of *O. mykiss* listed as federally threatened or endangered, and this species is represented in the data sets of all five pyrethroids that were examined. There are SMAVs for this species in all five acute data sets ranging from 0.119 to 7 μg/L, which are well above the derived acute criteria for these compounds. The λ-cyhalothrin acute data set also includes *Gasterosteus aculeatus*, of which a subspecies (*G.a. williamsoni*) is endangered. The acute permethrin data set includes seven additional listed species: *Oncorhynchus clarki henshawi*, *Etheostoma fonticola*, *Erimonax monachus*, *Notropis mekistocholas*, *Oncorhynchus apache*, *Salmo salar*, and *Xyrauchen texanus*. All of these acute toxicity values were used in criteria calculation and are well above the derived criteria; hence, there is no evidence that the criteria are underprotective of these

species. The only chronic toxicity value for a listed species was an MATC for *O. mykiss* in the cyfluthrin data set of 0.0133 μg/L, which is much higher than the chronic criterion of 0.00005 μg/L, indicating that the chronic criterion is protective of this species.

All of the acute data sets include species that are not listed but are in the same family or genus as some of those that are. These species were used as surrogates to estimate toxicity values for related TES with the USEPA interspecies correlation estimation software (Web-ICE v. 3.1; Raimondo et al. 2010). Unfortunately, the available bifenthrin and cyfluthrin SMAVs were below the model minimum input values, so toxicity values could not be predicted for bifenthrin or cyfluthrin. *O. mykiss* was used to predict λ-cyahlothrin, cypermethrin, and permethrin acute toxicity values for up to 13 species in the Salmonidae family (Tables S14–S16, Supporting Material http://extras.springer.com/). The predicted acute toxicity values ranged from 0.262 to 0.576 μg/L for cyfluthrin, 0.860 to 1.31 μg/L for cypermethrin, and 3.48 to 11.88 μg/L for permethrin, which are all more than one order of magnitude above their respective acute criteria.

One caveat of this evaluation is that the only TES in the measured or predicted data sets for these pyrethroids were fish, which are relatively insensitive compared to aquatic amphipods and insects. There were no data for TES in these more sensitive taxa, so it is not clear if the derived criteria are protective of these species. No single-species plant studies were found in the literature for use in criteria derivation for any of these pyrethroids, so no estimation could be made for plants on the state or federal endangered, threatened, or rare species lists. Phytoplanktons were unaffected by bifenthrin in a pond study (Sherman 1989); however, bifenthrin seemed to be beneficial in some instances and harmful in others, as reported in a mesocosm study that monitored primary productivity, green algae, chlorophyll, and other end points for photosynthetic organisms (Hoagland et al. 1993). Based on the mode of action, plants should be relatively insensitive to pyrethroids and the calculated criteria should be protective of aquatic plants.

12 Bioaccumulation

Chemicals in surface waters can accumulate in organisms from both the water and food items, which is called bioaccumulation, and eventually the chemicals can move up the food chain from prey to predator. Potential bioaccumulation was assessed to ensure that the derived criteria are set at concentrations that are not likely to cause toxicity due to bioaccumulation. Bifenthrin, cyfluthrin, cypermethrin, λ-cyhalothrin, and permethrin have similar physical–chemical characteristics (Table 1), including molecular weight $<$1,000 and log-normalized octanol–water partition coefficients (log K_{ow}) $>$3.0 L/kg, which indicate that all five compounds have the potential to bioaccumulate.

Low-to-moderate bioaccumulation of pyrethroids has been documented in the literature. For example, wild-caught brown trout (*Salmo trutta*), captured in a

British stream, was found to have accumulated an average 25.4 μg/kg of cyfluthrin and as high as 109 μg/kg in tissues, even though no cyfluthrin could be detected in the water column (Bonwick et al. 1996). Additionally, Surprenant (1986) reported that elimination of bifenthrin from bluegill tissues was very slow, i.e., after 42 days of depuration, fish tissue concentrations of bifenthrin were reduced by about half.

Because the pyrethroids have the potential to bioaccumulate, available data were used to estimate aqueous concentrations not expected to lead to harmful bioaccumulation. Analogous calculations were not done for human consumption of aquatic organisms because there are no tolerance or USFDA action levels for fish tissue (USFDA 2000) for any of these compounds. To calculate an aqueous NOEC, the dietary NOEC of an oral predator (mallard duck; studies listed in Table S17, Supporting Material http://extras.springer.com/) is divided by the bioaccumulation factor (BAF) for a fish. If a BAF is not available for a fish, it can be calculated as the product of the bioconcentration factor (BCF) and a biomagnification factor (BMF) such that BAF = BCF × BMF. BCFs are a measure of the uptake of a chemical by an organism from water alone while BMFs are a measure of the uptake of a chemical by an organism from food sources. BCFs for the pyrethroids of interest varied widely among different species, and were dependent on what portion of an organism was analyzed, with BCFs ranging from 2.6 to 3,280,000 (Table S18, Supporting Material http://extras.springer.com/).

For bifenthrin, one dietary NOEC was available for reproductive effects on mallard duck of 75 mg/kg (Roberts et al. 1986). No BAFs or BMFs were identified for fish, so the BCF of 28,000 L/kg bifenthrin for whole *P. promelas* (McAllister 1988) and a default BMF of 10, based on the log K_{ow} (TenBrook et al. 2010), were used to estimate a BAF as follows:

$$\mathrm{NOEC_{water}} = \frac{\mathrm{NOEC_{oral\_predator}}}{\mathrm{BCF_{food\_item}} \times \mathrm{BMF_{food\_item}}}. \tag{4}$$

The resulting $\mathrm{NOEC_{water}}$ for bifenthrin is 267 ng/L, which is well above the chronic criterion of 0.6 ng/L, indicating that bifenthrin at concentrations equal to or below the chronic criterion will not likely cause harm via bioaccumulation.

This calculation was also performed for the other four pyrethroids. For cyfluthrin, the highest BCF of 854 L/kg for *Lepomis macrochirus* (Carlisle and Roney 1984), a default BMF of 10, and the lowest dietary NOEC for a mallard of 250 mg/kg (Carlisle 1984) were used for a conservative estimation. The $\mathrm{NOEC_{water}}$ estimated for cyfluthrin using this data was 29 μg/L, which is above the aqueous solubility of cyfluthrin (2.3 μg/L; Laskowski 2002). For cypermethrin, the values used in Eq. 4 were the highest fish BCF of 821 L/kg for *O. mykiss*, a default BMF of 10, and a dietary toxicity value for mallard duck of 50 mg/kg, although this dietary NOEC was reported as greater than (>) 50 mg/kg (USEPA 2008). These values resulted in an $\mathrm{NOEC_{water}}$ for mallard of 6.09 μg/L, which is above the aqueous solubility of cypermethrin (4 μg/L; Laskowski 2002). An $\mathrm{NOEC_{water}}$ of 1.34 μg/L was calculated for λ-cyhalothrin with the highest reported BCF of 2,240 L/kg for whole fish *Cyprinus carpio* (Yamauchi et al. 1984), a default BMF of 10, and an oral predator

dietary NOEC of 30 mg/kg for mallard duck (Beavers et al. 1990). This $NOEC_{water}$ is significantly larger than the λ-cyhalothrin chronic criterion of 0.0005 μg/L. Finally, this calculation was completed for permethrin using the highest fish BCF of 2,800 L/kg for *P. promelas*, a default BMF of 10, and the dietary NOEC for mallard duck of 125 mg/kg, giving an $NOEC_{water}$ of 4.46 μg/L, which is nearing the aqueous solubility of 5.7 μg/L. Based on these conservative calculations, these pyrethroids are not likely to cause adverse effects on terrestrial wildlife due to bioaccumulation if their concentrations do not exceed the derived chronic criteria.

13 Assumptions, Limitations, and Uncertainties

Data limitations and important assumptions are reviewed here so that environmental decision makers have information about the accuracy and confidence in the criteria. Assumptions and limitations inherent in the methodology are summarized in the UCDM (TenBrook et al. 2010). The principal limitation for these five pyrethroids was a dearth of chronic data, particularly for the most sensitive species, amphipods and other invertebrates. There were no appropriate paired acute and chronic data for bifenthrin or cypermethrin to calculate ACRs, so the default ACR was used, while measured ACRs were available for cyfluthrin, λ-cyhalothrin, and permethrin. The acute criterion for cyfluthrin calculated with the median fifth percentile was almost identical to the lowest SMAV in the RR data set while the acute criterion for cypermethrin calculated with the median fifth percentile was higher than the lowest SMAV in the RR data set, so these criteria were adjusted downward to be more protective using less robust acute values. There are inherent assumptions in the use of an SSD (TenBrook et al. 2010), and the various distributional estimates can be used to assess uncertainty in the derived criteria for each compound. Of the data that were available for these compounds, not all were from flow-through tests that reported measured concentrations, which can cause overestimation of toxicity values, because pyrethroids are highly sorptive.

Other conspicuous data gaps were regarding temperature effects and mixture toxicity, especially with PBO; additional data on these topics should lead to quantifiable correlations, and these considerations should be added to criteria compliance when available. Also, pyrethroids are known to partition to sediments, and if federal or state sediment quality standards become available for these compounds, partitioning should be predicted based on the derived water quality criteria to ensure that these aqueous concentrations are not leading to potentially harmful sediment concentrations.

14 Comparison to Existing Criteria

To date, the USEPA has not calculated water quality criteria for bifenthrin, cyfluthrin, cypermethrin, λ-cyhalothrin, or permethrin. The CDFG composed a risk assessment report for several pyrethroids, including bifenthrin, cypermethrin,

and permethrin (Siepmann and Holm 2000). CDFG concluded that there were insufficient data to calculate criteria for bifenthrin using the USEPA (1985) method, and instead they reported the lowest acute and chronic toxicity values for guidance. The lowest genus mean acute value (GMAV) for bifenthrin was 3.97 ng/L for *A. bahia*, which is only slightly below the UCDM acute criterion of 4 ng/L; it can be noted that *A. bahia* is a saltwater species, which may be more sensitive than freshwater species. The lowest bifenthrin MATC in the CDFG report was 60 ng/L for *P. promelas*, which would not be protective of *D. magna* with an MATC of 1.9 ng/L (Table 3). The CDFG risk assessment reported interim acute criteria of 2 ng/L for cypermethrin and 30 ng/L for permethrin, which are both higher than the acute criteria calculated using the UCDM by factors of 2 and 3, respectively. Chronic criteria were not calculated for cypermethrin or permethrin because there was insufficient data.

The Netherlands has done generic risk assessment for several pyrethroids and maximum permissible concentrations (MPCs) have been calculated using the Dutch criteria derivation methodology. An MPC is defined as the concentration in the environment above which the risk of adverse effects is considered unacceptable to ecosystems and they are harmonized across media (Crommentuijn et al. 2000). MPCs are analogous to chronic water quality criteria and are used as the basis for setting environmental quality standards in the Netherlands. The Dutch MPCs for bifenthrin, cypermethrin, and permethrin are 1.1, 0.09, and 0.2 ng/L, respectively (Crommentuijn et al. 2000). These values were calculated with a modified EPA method in which an assessment factor ranging from 10 to 1,000 is applied to the lowest available toxicity value. The bifenthrin MPC of 1.1 ng/L is larger than the chronic criterion derived via the UCDM of 0.6 ng/L by a factor of 1.8, but there are no data to indicate that the MPC would be underprotective. The cypermethrin and permethrin MPCs are smaller than the UCDM criteria by a factor of 2.2 and 10, respectively.

In Canada, an interim freshwater quality guideline was derived for permethrin by applying a safety factor of 0.1 to the most sensitive LOEC, which was for *Pteronarcys dorsata* (Anderson 1982). The interim aquatic life guideline for permethrin was derived as 4 ng/L, which is larger than the UCDM chronic criterion of 2 ng/L, but there are no data to indicate that 4 ng/L would be underprotective (CCME 2006). Quebec has also derived its own interim acute criterion for permethrin of 44 ng/L and an interim chronic criterion of 13 ng/L (Guay et al. 2000); the interim acute criterion is larger than the lowest SMAV in the UCD data set and would not be protective of *H. azteca*. In the UK, short-term and long-term predicted NECs (PNECs), analogous to acute and chronic criteria, were recently derived for permethrin using assessment factors (Lepper et al. 2007). The short-term PNEC of 10 ng/L was derived by applying an assessment factor of 10 to the LC_{50} for the mayfly *Hexagenia bilineata*, and the long-term PNEC of 1.5 ng/L was derived by applying an assessment factor of 20 to an LOEC for the caddisfly *B. americanus* (Lepper et al. 2007). The currently adopted long-term (chronic) environmental quality standard (EQS) in the UK for permethrin is 10 ng/L. There are also proposed long-term and short-term EQSs for cypermethrin of 0.1 and 0.4 ng/L, respectively, which are lower than the existing EQSs of 0.2 and 2.0 ng/L, respectively (UKTAG 2008).

15 Comparison to the USEPA (1985) Method

More pyrethroid toxicity data are available now than when CDFG derived criteria for bifenthrin, cypermethrin, and permethrin (Siepmann and Holm 2000) using the USEPA (1985) method. To compare the UCDM criteria to those generated using the USEPA (1985) method, the data sets gathered for this article were used to generate example USEPA criteria for these compounds. The five acute taxa requirements of the SSD procedure in the UCDM were fulfilled for each of these five pyrethroids. There are three additional taxa requirements in the USEPA acute method, as follows:

1. A third family in the phylum Chordata (e.g., fish, amphibian)
2. A family in a phylum other than Arthropoda or Chordata (e.g., Rotifera, Annelida, Mollusca)
3. A family in any order of insect or any phylum not already represented

These three additional requirements were not met for any of these compounds. The bifenthrin, λ-cyhalothrin, and permethrin data sets do not contain any species in a phylum other than Arthropoda or Chordata, but met all of the other taxa requirements. The CDFG has calculated criteria for compounds with incomplete data sets if the missing taxa requirements are known to be relatively insensitive to the compound of interest. The only data available for organisms not in the phyla Arthropoda or Chordata were for saltwater mollusks (*Crassostrea virginica* and *Crassostrea gigas*), which were very insensitive to bifenthrin and λ-cyhalothrin—EC_{50}s could not be calculated for these species because of solubility limits or no responses were observed at the highest concentrations tested (Thompson 1985; Ward 1986a, 1986b, 1987)—so example criteria were calculated for bifenthrin, λ-cyhalothrin, and permethrin. The cyfluthrin and cypermethrin acute data sets were missing two of the additional requirements, so example criteria were not calculated for these compounds.

Acute criteria were calculated by fitting the log-triangular distribution to the acute bifenthrin, λ-cyhalothrin, and permethrin data sets (Tables 2, 8, and 10) and are reported with two significant figures, according to the USEPA (1985) method. The USEPA (1985) method fits the SSD to *genus* mean acute values while the UCDM uses *species* mean acute values, so the UCDM data sets were altered when necessary to calculate genus mean acute values.

Example acute criterion = Final acute value/2

Bifenthrin : Example final acute value (fifth percentile) = 0.0009543 μg/L
Example acute criterion = 0.00048 μg/L

λ-Cyhalothrin: Example final acute value (fifth percentile) = 0.001845 μg/L
Example acute criterion = 0.00092 μg/L

Permethrin : Example final acute value (fifth percentile) = 0.039001 μg/L
Example acute criterion = 0.010 μg/L

The bifenthrin example acute criterion (0.48 ng/L) is almost one order of magnitude lower than the acute criterion calculated by the UCDM (4 ng/L). The λ-cyhalothrin example acute criterion (0.92 ng/L) is almost identical to the acute criterion calculated using the Burr Type III distribution of the UCDM (1 ng/L), and the permethrin example acute criterion (10 ng/L) is identical to the UCDM acute criterion (10 ng/L).

To calculate chronic criteria according to the USEPA (1985) method for compounds with limited chronic data such as the pyrethroids, an ACR procedure is used, which is very similar to the ACR procedure in the UCDM. The ACR procedure cannot be used for cyfluthrin and cypermethrin because acute criteria were not calculated for these compounds. The EPA ACR procedure requires data for three ACRs, which were not available for bifenthrin or permethrin. For λ-cyhalothrin, the same three SMACRs calculated for the UCDM (Table 13) were calculated according to the USEPA (1985) methodology to give a final λ-cyhalothrin ACR of 4.73. The λ-cyhalothrin chronic criterion was calculated by dividing the final acute value by the final ACR:

Example chronic criterion = Final acute value/Final ACR
λ-cyhalothrin example chronic criterion = 0.00039 μg/L

The λ-cyhalothrin example chronic criterion (0.39 ng/L) differs by less than a factor of 2 from the one recommended by the UCDM (0.5 ng/L).

This comparison of criteria calculated using the UCDM and USEPA (1985) method highlights the limitations of the USEPA method. According to the USEPA method, acute criteria could not be calculated for cyfluthrin or cypermethrin, and acute criteria were only calculated for bifenthrin, λ-cyhalothrin, and permethrin by making exceptions for the taxa requirements, and chronic criteria could not be calculated for bifenthrin, cyfluthrin, or cypermethrin. The λ-cyhalothrin acute data set was large and the criteria calculated by the two methods were very similar (1 ng/L vs. 0.92 ng/L). When large data sets are available, criteria calculated using the two methods have been similar, e.g., chlorpyrifos and diazinon (Palumbo et al. (2012)), because the calculation methods in these cases are very similar. When large data sets are not available or data sets are missing a USEPA taxa requirement, the UCDM is able to generate criteria, where the USEPA method gives no results, e.g., malathion (Palumbo et al. (2012)) and cyfluthrin.

16 Final Criteria Statements

The inputs for the final criteria statement are listed in Table 15.

Aquatic life should not be affected unacceptably if the 4-day average concentration of [1] does not exceed [2] μg/L ([3] ng/L) more than once every 3 years on the average and if the 1-h average concentration does not exceed [4] μg/L ([5] ng/L) more than once every 3 years on the average. Mixtures of [1] and other pyrethroids should be considered in an additive manner (see Sect. 7).

Table 15 Final numeric criteria for the five pyrethroids

1 Compound	2 Chronic riterion (μg/L)	3 Chronic criterion (ng/L)	4 Acute criterion (μg/L)	5 Acute criterion (ng/L)
Bifenthrin	0.004	4	0.0006	0.6
Cyfluthrin	0.00005	0.05	0.0003	0.3
Cypermethrin	0.0002	0.2	0.001	1
λ-Cyhalothrin	0.0005	0.5	0.001	1
Permethrin	0.002	2	0.01	10

It is recommended that the freely dissolved pyrethroid concentration is measured for criteria compliance because this appears to be the best predictor of the bioavailable fraction.

17 Summary

Aquatic life water quality criteria were derived for five pyrethroids using a new methodology developed by the University of California, Davis (TenBrook et al. 2010). This methodology was developed to provide an updated, flexible, and robust water quality criteria derivation methodology specifically for pesticides. To derive the acute criteria, log-logistic SSDs were fitted to the medium-sized bifenthrin, cyfluthrin, and cypermethrin acute toxicity data sets while the λ-cyhalothrin and permethrin acute data sets were larger, and Burr Type III SSDs could be fitted to these data sets. A review of the cyfluthrin acute criterion revealed that it was not protective of the most sensitive species in the data set, *H. azteca*, so the acute value was adjusted downward to calculate a more protective criterion. Similarly, the cypermethrin criteria were adjusted downward to be protective of *H. azteca*. Criteria for bifenthrin, λ-cyhalothrin, and permethrin were calculated using the median fifth percentile acute values while the cyfluthrin and cypermethrin criteria were calculated with the next lowest acute value (median first percentile). Chronic data sets were limited in all cases, so ACRs were used for chronic criteria calculations, instead of statistical distributions. Sufficient corresponding acute and chronic data were not available for bifenthrin, cypermethrin, or permethrin, so a default ACR was used to calculate these chronic criteria while measured ACRs were used for cyfluthrin and λ-cyhalothrin. A numeric scoring system was used to sort the acute and chronic data, based on relevance and reliability, and the individual study scores are included in the Supporting Information.

According to the USEPA (1985) method, the data sets gathered for these five pyrethroids would not be sufficient to calculate criteria because they were each missing at least one of the eight taxa required by that method. The USEPA (1985) method generates robust and reliable criteria, and the goal of creating the UCDM was to create a method that also yields statistically robust criteria, but with more

flexible calculation methods to accommodate pesticide data sets of varied sizes and diversities. Using the UCDM, acute and chronic water quality criteria were derived for bifenthrin (4 and 0.6 ng/L, respectively), cyfluthrin (0.3 and 0.05 ng/L, respectively), cypermethrin (1 and 0.2 ng/L, respectively), λ-cyhalothrin (1 and 0.5 ng/L, respectively), and permethrin (10 and 2 ng/L, respectively). Water quality criteria for these five pyrethroids can be used by environmental managers to control the increasing problem of surface water contamination by pesticides.

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