Determination of water quality criteria for three synthetic pyrethroids for the protection of aquatic life

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Abstract: ***Synthetic pyrethroid (SP) pesticides have been widely used in numerous applications, with increasing usage each year. They can be transported from crop fields to adjacent streams and affect aquatic organisms. Public concern over the use of SP pesticides has arisen due to their high toxicity to fish and aquatic invertebrates. The use of water quality criteria (WQC) is aimed at protecting aquatic organisms. However, few WQC values are available for SP pesticides, especially in China. In this study, the acute and chronic WQC of three SP pesticides were derived by the species sensitivity distribution (SSD) method to protect aquatic life. The criterion maximum concentration (CMC) values were 0.0066 μg/L, 0.0037 μg/L and 0.2137 μg/L for bifenthrin, lambda-cyhalothrin and permethrin, respectively. The criterion continuous concentration (CCC) values were 0.0023 μg/L, 0.0029 μg/L and 0.0862 μg/L for bifenthrin, lambda-cyhalothrin and permethrin, respectively. The results provide reference values for water quality management.***

Keywords: pyrethroids; species sensitivity distribution curve methods; water quality criteria

1. Introduction

In the past several decades, pesticides have been increasingly used to improve crop quality, prevent yield losses and enhance market opportunities [1]. In the early 1980s [2], after the ban of some organophosphorus insecticides, the use of SP pesticides increased dramatically because of their high efficacy and generally low mammalian toxicity. Between 1992 and 2002, the total SP pesticide use averaged 125,000 kg/year [3] in the USA. Similarly, China has also seen the extensive application of pesticides, of which approximately 2.8% are SP pesticides [4].
However, regions in which SP pesticides have been used have experienced aquatic toxicity in streams and rivers [5].These pesticides are transported from crop fields to adjacent streams via surface run-off, drains, groundwater and atmospheric deposition and then give rise to transient pulse contamination [6]. In recent years, public concern about SP insecticides has increased due to their high acute toxicity to non-target organisms, such as fish and aquatic invertebrates, at trace levels [2], [7]. Arthropods are vulnerable to the lethal effects of pyrethroid, with acute LC_{50}< 4 ng/L for the most sensitive species [5]. Although SPs are degraded by sunlight and by microorganisms in water, the more recently developed SPs can persist in aquatic environments for a substantial period of time before degradation [7]. The SP concentration in the water samples of California's San Joaquin River and its tributaries ranged from 0.005 to 0.021 μg/L [8], whereas that in the Ebro River Delta (NE Spain) ranged from 0.03 to 35.8 ng/L and 2.6 to 62.4 pg/g for water and sediment [9], respectively. In the Qiantang River, the concentration of bifenthrin in crucian carp muscle ranged from 0.64 to 110.47 μg/kg [10]. In the sediment of urban waterways in California, the concentration of bifenthrin ranged from 2.19 to 219 ng/g dry weight [11]. Permethrin in surface water samples has been detected by the United States Geological Survey (USGS) at concentrations ranging from 0.001 to 0.560 μg/L [12].

Bifenthrin, lambda-cyhalothrin and permethrin represent the most heavily used pyrethroids [8]. In 2002, among the 10 SPs used in California agriculture, permethrin represented45% of the total pesticide application by mass [3]. In 2009, the total amount of SPs used by licensed professional applicators was 138000 kg in California, with bifenthrin and permethrin accounting for 16% and 54% of the total use, respectively [13]. Even more importantly, these three pyrethroids present very significant toxicity to aquatic organisms. For bifenthrin, the median lethal concentration (LC_{50}), median inhibitory concentration (IC_{50}) for days surviving and IC50 for reproduction were 0.86, 0.55 and 0.49 μg/L for daphnia magna [14]. The LC_{50} values of lambda-cyhalothrin for aquatic vertebrate zebrafish were 2.12, 1.11and 0.875 μg/L for 24 h, 48 h and 96 h, respectively [15]. The 96-h LC_{50} of permethrin for short nose sturgeon, razorback sucker, bony tail chub, Cape Fear shiner, Colorado pike minnow and spot fin chub, all of which are endangered and threatened aquatic species, were 1.81, 4.35, 3.49, 4.51, 3.07 and 3.41 mg/L, respectively[16]. D. P. Weston et al. reported that nearly all creek sediment samples collected from Roseville, California, caused toxicity in laboratory exposure to the amphipod Hyalella azteca, and approximately half the samples caused near-complete mortality. Bifenthrin was implicated as the primary cause of this toxicity [17]. In addition, many products containing permethrin are classified as "restricted use pesticides" by the US EPA and Canada because of its toxicity to fish [18]. Nevertheless, their thresholds in aquatic environments are still not clear.

The use of water quality criteria (WQC) is aimed at protecting important commercial and recreational aquatic organisms in the aquatic ecosystem from unacceptable effects of exposure to high concentrations for short periods of time or lower concentrations for long periods of time [19]. Some SPs are highly toxic to aquatic organisms and widely present in the aquatic environment. However, quantitative national water quality criteria for these three SPs are lacking. In the USA, there are currently no quantitative WQCs for bifenthrin, lambda-cyhalothrin and permethrin. The lack of WQCs is a result of many of the available data not being suitable for the criteria derivation methodology recommend by the US EPA. [20].To protect aquatic life from serious harm, it is essential to determine the WQCs for these SPs.

Because different countries are host to different aquatic biota and have different protection targets, they have different national guidelines and recommended methods for deriving the ultimate values of water quality criteria [21].The toxicity percentile rank (TPR) method, which requires the species toxicity data of three phyla and eight taxonomic families, is recommended by the United States in its national WQC guidelines. The assessment factor (AF) method is used by many countries, such as Canada, due to its simplicity and universality, but it is associated with high levels of uncertainty, as it simply multiplies the toxicity data for the most sensitive species by an assessment factor to obtain the final criteria values. The species sensitivity distribution (SSD) method, which is recommended by European Union (EU), Australia and New Zealand, is used to derive the WQC values of target pollutants in this study. The SSD method assumes that all available toxicity data fit a certain distribution. Based on the best-fitting distribution model, the hazard concentrations (HC5) can be obtained, which can then be used to derive the final WQC values. The SSD method can make full use of toxicity data and is less strict than the toxicity percentile rank method in terms of the collection and screening of toxicity data. Compared with the AF method, the SSD method can provide a more reliable criterion value. The use of optimal toxicity data and a model with good fit is very important in the derivation of WQC when using this method. Different toxicity endpoints and models can yield very different criterion values. Based on earlier studies and guidelines, the 96-h LC_{50} for acute toxicity data and NOEC for chronic toxicity data were adopted in this study. After comparing the commonly used models, the logistic model was eventually chosen to fit the data. The aim of our study is to provide a methodological reference and a basis for the derivation of the WQC of SPs. The criteria values obtained can help the government to design an effective water environment management policy, particularly for widely used pesticides.

2. Materials and methods

2.1 Toxicity data collection and screening

All toxicity data for bifenthrin, lambda-cyhalothrin and permethrin were collected from the ECOTOX database of the US Environmental Protection Agency (US EPA, http://cfpub.epa.gov/ecotox/) and China National Knowledge Internet (CNKI, http://www.cnki.net/). The toxicity data cover Chinese native species. The acute toxicity endpoints of lethality were used to derive the acute WQC values, whereas the chronic toxicity endpoints were used to derive the WQC values including lethality and reproduction. The quality assessment and allocation standard of toxicity data during the derivation of criteria values have been comprehensively examined in the literature [19], [22]-[24]. The toxicological experiments should conform to the standard method issued by relevant countries or authoritative international organizations. For screening, acute toxicity data for 96 h of exposure and chronic toxicity data for at least four days of exposure were generally selected. For acute toxicological endpoints, LC_{50} values corresponding to the lethal effect were selected. For chronic toxicological endpoints, the no observed effect concentration (NOEC) was selected. If the acute (or chronic) toxicity data for one species includes values that differ from the other values by a factor of more than 10, the outliers should be rejected.

2.2 Target species

The target species were chosen to represent the actual composition of the aquatic ecosystem. These species must include important commercial and recreational aquatic organisms as well as other important and precious species. According to the species selection requirements of the SSD method, vertebrate animals (especially fish), invertebrate animals and arthropods should be included [25].

2.3 Target compounds

SP pesticides are a class of hydrophobic compounds with low solubility and high adsorption capacities [26]. These pesticides are widely used in production and non-production agriculture. The residues of SP pesticides are widely present in crops, soil and water. Bifenthrin (CAS Registry Number 82657-04-3), lambda-cyhalothrin (CAS Registry Number 91465-08-6) and permethrin (CAS Registry Number 52645-53-1) are the three most commonly used SP pesticides. Permethrin is the SP pesticide used most heavily. In addition, permethrin possesses significant aquatic toxicity. Permethrin is classified as a “restricted use pesticide” by the USA and Canada. Bifenthrin and lambda-cyhalothrin have proven to be extremely toxic to fish and invertebrates. Using these data, acute and chronic WQC values for bifenthrin, lambda-cyhalothrin and permethrin could be obtained.

2.4 Derivation of criteria values

Species sensitivity distribution (SSD) was first proposed by Kooijman and then modified and improved by Aldenberg and Slob [27]. The SSD method requires a large set of measured toxicity data to define a hazard level for the protection of multiple species [28]. This method assumes that the selection of the species is random and representative of the ecosystem. It also assumes that all the available toxicity data fit a certain distribution, which can then be used to estimate the pollutant’s x% harm concentration (HC_x, generally expressed as HC_5,corresponding to the concentration at which 95% species will not be affected by the target pollutants). In general, the larger the toxicity data set, the more reliable the results.

After screening the species toxicity data, they were arranged from largest to smallest. Here, i represents the rank of a species in the data series, and n is the total number of examined species. The cumulative probability (P) for each species is calculated as follows [29]:

P= (i-0.5)/n (1)

SSD assumes that all the data adhere to a certain distribution. The existent guidelines and references recommend many fitting models. The log-normal, log-logistic and Burr Type III fitting models are the most commonly used [30]. However, none of these fitting models have been shown to be suitable for all types of species toxicity data. Therefore, such parameters as the adjusted coefficient of determination (R^2) can be used to compare the suitability of models for a given set of toxicity data and identify the optimum model.

In the SSD method, the acute WQC was defined as

$WQC_{acute}=HC_{5acute}/AF$ (2)

where AF is an assessment factor, which is required by the uncertainty in the HC_5 derivation[31]. At present, no effective method has been developed to accurately quantify this uncertainty. The AF was set at 2 in the current study [21].

There are two approaches to the calculation of the chronic WQC depending on the amount of chronic toxicity data available. The chronic WQC can be calculated in the similar way as the acute WQC if a large set of chronic toxicity data is available. The chronic WQC was defined as:

$WQC_{chronic}=HC_{5chronic}/AF$ (3)

If the chronic toxicity data are insufficient, the chronic WQC can be defined as:

$WQC_{chronic}=HC_{5acute}/(ARC\times AF)$ (4)

where ARC is the acute-chronic ratio. The ARC must be determined using the acute and chronic toxicity data for the same species. If experimental ARC values are not available, the values recommended by national or international organizations can be adopted[32].

3. Results and discussion

3.1 Available toxicity data

In the SSD method, sufficient toxicity data are essential. Numerous species are sensitive to a pollutant. When collecting toxicity data, the data for sensitive species must be taken into consideration. Because some species are not sensitive to the target chemical, the toxicity data will deviate somewhat from a normal distribution. After the collection and screening of the toxicity data, the available acute and chronic toxicity data for lambda-cyhalothrin, bifenthrin and permethrin for different species are listed in Table 1. The species include *Misgurnus anguillicaudatus* [33] and *Monopterus albus* [34], which are unique to China. The outliers present in the toxicity data for some species were eliminated. For toxicity data for the same species with the same exposure time, the geometric mean value was adopted.

Table 1: The available acute and chronic toxicity data of bifenthrin, lambda-cyhalothrin and permethrin

Chemical	Species	Endpoints	Concentration (μg/L)
Bifenthrin	*Cyprinus carpio*	LC_{50}	57.5
	Chironomus tentans	LC_{50}	26.15
	Daphnia magna	LC_{50}	1.4
	Oncorhynchus mykiss	LC_{50}	0.15
	Ceriodaphnia dubia	LC_{50}	0.0975
	Palaemonetes pugio	LC_{50}	0.017
	Cyprinodon variegatus	NOEC	11.7446
	Danio rerio	NOEC	3.2
	Carassius auratus	NOEC	0.8839
	Lepomis macrochirus	NOEC	0.35
	Daphnia magna	NOEC	0.0807
	Palaemonetes pugio	NOEC	0.0099
Lambda-cyhalothrin	*Misgurnus anguillicaudatus*	LC_{50}	21.1714
	Clarias gariepinus	LC_{50}	8
	Channa punctata	LC_{50}	7.92
	Danio rerio	LC_{50}	3.6695
	Tilapia sp.	LC_{50}	2.64
	Oncorhynchus mykiss	LC_{50}	1.1322
	Lepomis macrochirus	LC_{50}	0.7793
	Macropelopia sp.	LC_{50}	0.698
	Erythromma viridulum	LC_{50}	0.493
	Caridina laevis	LC_{50}	0.33
	Procambarus clarkii	LC_{50}	0.16
	Cloeon dipterum	LC_{50}	0.105
	Asellus aquaticus	LC_{50}	0.0752
	Proasellus coxalis	LC_{50}	0.0446
	Caenis horaria	LC_{50}	0.0346

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Table 1 (*continued*)

	Macrobrachium nipponense	LC_{50}	0.0335
	Monopterus albus	LC_{50}	0.0261
	Gammarus pulex	LC_{50}	0.0242
	Chironomus dilutus	LC_{50}	0.0217

	Notonecta glauca	LC_{50}	0.0164
	Americamysis bahia	LC_{50}	0.0041
	Tilapia sp.	NOEC	1
	Channa punctata	NOEC	0.9798
	Clarias batrachus	NOEC	0.7579
	Chironomus tentans	NOEC	0.7266
	Oreochromis mossambicus	NOEC	0.3
	Lithobates pipiens	NOEC	0.1
	Caridina laevis	NOEC	0.1
	Ostracoda	NOEC	0.1
	Armiger crista	NOEC	0.1
	Lymnaea stagnalis	NOEC	0.1
	Mesostoma sp	NOEC	0.1
	Anuraeopsis fissa	NOEC	0.0891
	Asellus aquaticus	NOEC	0.0707
	Cloeon dipterum	NOEC	0.05
	Animalia	NOEC	0.05
	Gammarus pulex	NOEC	0.0416
	Daphnia galeata	NOEC	0.0316
	Caenis horaria	NOEC	0.0316
	Cyclopoida	NOEC	0.025
	Chaoborus obscuripes	NOEC	0.01
	Arthropoda	NOEC	0.01
	Clarias gariepinus	NOEC	0.008
Permethrin	*Xenopus laevis*	LC_{50}	458.2576
	Lymnaea acuminate	LC_{50}	370
	Procambarus blandingii	LC_{50}	210

Table 1 (*continued*)

	Lithobates sphenocephalus	LC_{50}	81.2
	Menidia beryllina	LC_{50}	27.5
	Atherinops affinis	LC_{50}	25.3
	Ptychocheilus lucius	LC_{50}	24.2659
	Cyprinodon variegatus	LC_{50}	21.2804
	Cyprinodon bovinus	LC_{50}	21
	Oncorhynchus kisutch	LC_{50}	17
	Ctenopharyngodon idella	LC_{50}	14.98
	Pimephales promelas	LC_{50}	13.6937
	Gambusia affinis	LC_{50}	11.1686
	Cyprinus carpio	LC_{50}	10.7102
	Micropterus salmoides	LC_{50}	8.5
	Esox lucius	LC_{50}	6.1343
	Lepomis macrochirus	LC_{50}	6.0537
	Xyrauchen texanus	LC_{50}	5.9632
	Mugil cephalus	LC_{50}	5.5
	Oncorhynchus mykiss	LC_{50}	4.947
	Salmo salar	LC_{50}	4.2426
	Notropis mekistocholas	LC_{50}	4.16
	Uca pugilator	LC_{50}	3.6376
	Daphnia magna	LC_{50}	3.6234
	Salvelinus fontinalis	LC_{50}	3.3579
	Etheostoma fonticola	LC_{50}	3.34
	Ictalurus punctatus	LC_{50}	3.1355
	Etheostoma lepidum	LC_{50}	2.71
	Danio rerio	LC_{50}	2.5
	Menidia menidia	LC_{50}	2.2
	Oncorhynchus gilae ssp	LC_{50}	1.7078
	Erimonax monachus	LC_{50}	1.7

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Table 1 (*continued*)

	Oncorhynchus clarkii ssp	LC_{50}	1.5868
	Chironomus dilutus	LC_{50}	0.7602
	Homarus americanus	LC_{50}	0.73
	Ceriodaphnia dubia	LC_{50}	0.6871
	Procambarus clarkii	LC_{50}	0.6438

	Nitocra spinipes	LC_{50}	0.6
	Gammarus pulex	LC_{50}	0.44
	Palaemonetes pugio	LC_{50}	0.4066
	Tropocyclops prasinus	NOEC	34.44
	Algae	NOEC	30.44
	Acroneuria abnormis	NOEC	16
	Salvelinus fontinalis	NOEC	1.6
	Pimephales promelas	NOEC	1.0896
	Planorbella trivolvis	NOEC	0.33
	Danio rerio	NOEC	0.2236
	Palaemonetes pugio	NOEC	0.2

3.2 Species sensitivity distribution

Unlike the toxicity percentile rank method, the species sensitivity distribution method can make full use of the available species toxicity data. Moreover, this method does not lead to overprotection and uncertainty, as in the case of the AF method. In this study, the toxicity data were analyzed using Origin 8.0 software. A variety of distribution models were used to fit the data, and the coefficients of determination (R^2) are listed in Table 2. The best-fitting model, namely, that with the highest R^2 value, was identified. For both the acute and chronic toxicity data, the logistic model provided the best fit for bifenthrin, lambda-cyhalothrin and permethrin. We suspected that the logistic model provides the best fit for pyrethroid pesticides in general. In addition, the results of this study also show that a better model fitting may result from the use of a larger dataset. The largest set of acute toxicity data is available for permethrin. The corresponding R^2 of its logistic model was 0.99141, indicating that it is the best-fitting distribution.

3.3 Water quality criteria for three SPs

Acute water quality criteria for bifenthrin, lambda-cyhalothrin and permethrin were derived by the SSD curve method, and a logistic model was used to fit the data to construct the SSD curve. Fig. 1 shows the fitting distributions of these three pyrethroids. The HC_{5acute} values were determined to be 0.0132 μg/L for bifenthrin, 0.0073 μg/L for lambda-cyhalothrin and 0.4274 μg/L for permethrin. According to equation (2), the final WQC_{acute} values were 0.0066 μg/L, 0.0037 μg/L and 0.2137 μg/L for bifenthrin, lambda-cyhalothrin and permethrin, respectively.

A large set of chronic toxicity data was available for these three chemicals for the derivation of the chronic WQC value using the SSD curve method. Therefore, it is not necessary to calculate the acute-chronic ratio. The logistic fitting model was also used to build the SSD curves of the chronic toxicity data for bifenthrin, lambda-cyhalothrin and permethrin. The SSD curves of these three pyrethroids are presented in Fig. 2. Based on the SSD curve, HC5chronic was determined to be 0.0045 μg/L for bifenthrin, 0.0057 μg/L for lambda-cyhalothrin and 0.1723 μg/L for permethrin. According to equation (3), the final $WQC_{chronic}$ values were 0.0023 μg/L, 0.0029 μg/L and 0.0862 μg/L for bifenthrin, lambda-cyhalothrin and permethrin, respectively.

Table 2: Fitting models of toxicity data

Chemical	Endpoints	Fitting model	R^2
Bifenthrin	LC_{50}	Logistic	0.92998
		Allometric1	0.88815
		ExpDec1	0.82033
		Lorentz	0.76372
		Gumbel	0.64878
	NOEC	Logistic	0.99743
		Lorentz	0.94381
		Allometric1	0.95217
		Gumbel	0.76208
		ExpDec1	0.93407
Lambda-cyhalothrin	LC_{50}	Logistic	0.98331
		Gauss	0.90078
		ExpDec1	0.90977
		Gumbel	0.80963
		Lorentz	0.93054
	NOEC	Logistic	0.95564
		ExpAssoc	0.94817
		ExpDec1	0.95363
		Gauss	0.70916
		Gumbel	0.90849
permethrin	LC_{50}	Logistic	0.99141
		Allometric1	0.72968
		ExpAssoc	0.99135
		ExpDec1	0.97825
		Gumbel	0.83595
	NOEC	Logistic	0.91675
		Gauss	0.86397
		Allometric1	0.87198
		ExpDec1	0.89158
		Gumbel	0.72548

safety factor. The 21-d LOEC is a chronic toxicity datum; therefore, the WQC recommended by Canada and the $WQC_{chronic}$ value in this study are on the same order of magnitude. The recommended WQC value does not consider the acute toxicity data and chronic toxicity data separately. The SSD method can make full use of the toxicity data, and the relevant WQC value can reflect the whole ecosystem better. In addition, the WQC value recommended by Canada may lead to overprotection. In the study conducted at UC Davis [20] the WQC_{acute} values were 0.004 μg/L, 0.001 μg/L and 0.01 μg/L for bifenthrin, lambda-cyhalothrin and permethrin, respectively. The $WQC_{chronic}$ values were 0.6 ng/L, 0.5 ng/L and 2 ng/L for bifenthrin, lambda-cyhalothrin and permethrin, respectively. WQC_{acute} values were also derived by the SSD method in the study conducted at UC Davis. The slight difference in the WQC_{acute} values may be attributed to the collection of the acute toxicity data and the model used to fit the data. For the different protection aims, different toxicological endpoints and exposure times were chosen. Different fitting models can provide very different WQC values. The significant difference in $WQC_{chronic}$ may due to the difference in the derivation method used. In the research conducted at UC Davis, the $WQC_{chronic}$ values were derived by dividing the WQC_{acute} values by ARC. However, in this study, the $WQC_{chronic}$ values were obtained according to the distribution of the chronic toxicity data.

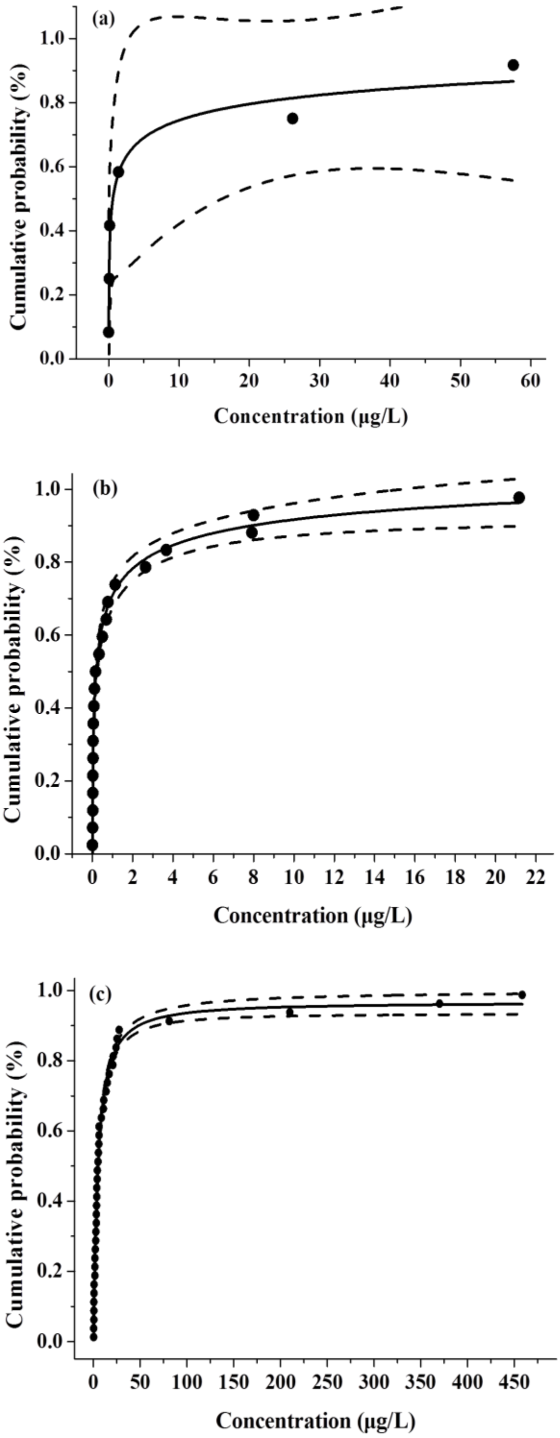

Figure 1: Species sensitivity distribution of acute toxicity data (a, bifenthrin, b, lambda-cyhalothrin, c permenthrin)

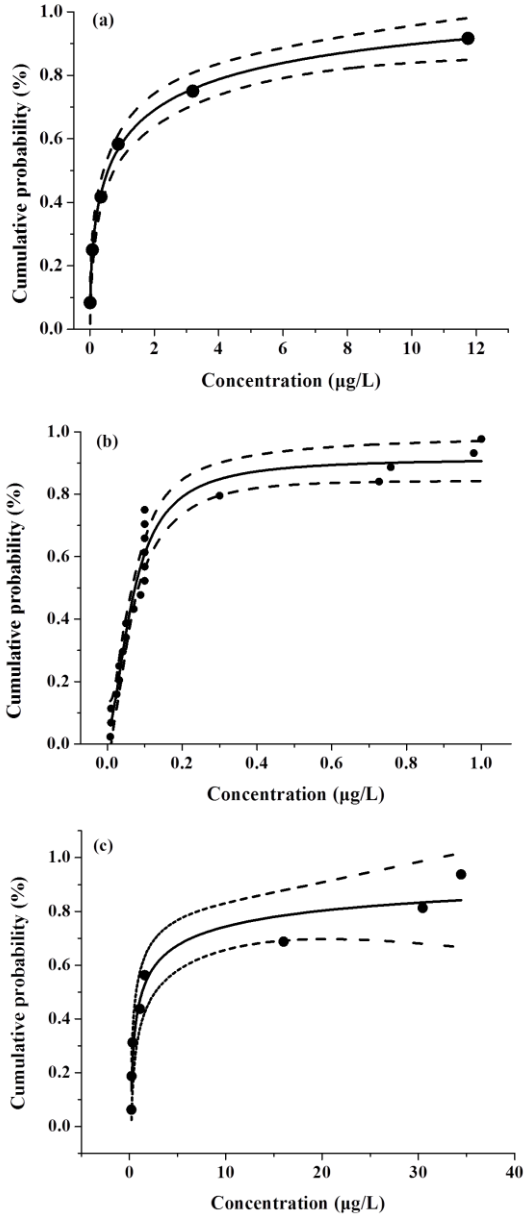

Figure 2: Species sensitivity distribution of chronic toxicity data (a bifenthrin, b lambda-cyhalothrin, c permenthrin)

3.4 Value of the derivation of pyrethroid water quality criteria

Over the past several decades, SP pesticides have been increasingly used in agriculture and many other applications. Bifenthrin, lambda-cyhalothrin and permethrin are the three most representative pyrethroid pesticides. The derivation of the specific WQC for these three SPs is of great valuable and urgent. The WQC values obtained in our results were much lower than those of many other pollutants [21], [29]. This difference was mainly attributed to the extremely high toxicity of SPs to aquatic organisms, such as fish and invertebrates. The use of WQCs is intended to protect the widest range of aquatic organisms. However, due to a lack of toxicity data, national WQCs for pyrethroids have been lacking. China is among the countries without any WQC value for pyrethroids, leading to a lack of water management standards and criteria for SP pesticides. Pyrethroids are ubiquitous in the environment, especially in water. The concentrations of SPs in water samples ranged from 0.005 to 0.021 μg/L [8]. The highest permethrin concentration was 323.9 μg/L in Yiwu, Zhejiang Province, China [35]. The concentrations of these three SPs in environmental media were generally higher than the WQC values obtained in the study. Therefore, the WQC in our study can be applied as a water quality management reference value in China. The government and environmental protection department should attempt to control the concentrations of these pyrethroids in aquatic environments such that they do not exceed the corresponding WQC values obtained in this study.

4. Conclusions

The aim of the use of WQC values is to protect aquatic organisms. The derivation of WQC values for SPs, which are extremely toxic to aquatic organisms, is of environmental significance. The SSD method was adopted to derive WQC values in the study because it can make full use of toxicity data and provide an accurate WQC value. The toxicity data and fitting model are important when using SSD method. Different toxicity endpoints, exposure times and fitting models can yield very different results. Based on the specific protection goals, we should select these factors explicitly and carefully. Despite their difference from previously obtained WQC values, the values obtained in this study can be used as water quality management reference values for China. Future research should conduct broader and more intensive acute and chronic toxicity experiments and obtain more abundant toxicity data to provide the most reasonable and reliable WQC values for pyrethroids, such as the target chemicals of this study.

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