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中国七大水系淡水沉积物中林丹(γ-HCH)的生态风险评估

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摘要: 林丹(γ-HCH)作为曾广泛应用的有机氯农药,自 2000 年在中国停止生产以来,全国范围内环境介质中仍广泛检出,对生物体及自然环境存在潜在危害。在收集 γ-HCH 的沉积物毒性数据基础上,通过物种敏感度分布(Species Sensitivity Distributions, SSD)曲线拟合的方法获得其沉积物质量基准。选取 7 种常用模型进行拟合,通过比较,最终采用 S-Logistic 模型拟合 γ-HCH 急性毒性曲线,得到急性基准值CMC_{sed}=0.00530 μg·g^{-1};采用 S-Gompertz 模型拟合 γ-HCH 慢性毒性曲线,得到慢性基准值CCC_{sed}=0.00106 μg·g^{-1}。我国七大水系 68.2% 的水体沉积物中 γ-HCH 的残留浓度均低于其CCC_{sed},说明其风险较低。但是,在海河和辽河流域某些点位的残留超标,需要引起足够的重视。所获得的沉积物基准值对评估沉积物中 γ-HCH 的生态风险和环境修复具有重要指导意义。

关键词: γ-HCH; 物种敏感度分布; 沉积物质量基准; 相平衡分配法
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Ecological Risk Assessment of γ-HCH for Freshwater Sediment of Seven Major River Systems in China

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Abstract: As one of the widely used organochlorine pesticides, lindane (γ-HCH) has been banned since 2000, but still can be detected in the environment throughout China and may pose potential adverse effects to organisms and the environment. In this study, toxicity data of γ-HCH in sediment were collected and grouped. Then, its sediment quality criterion were derived by fitting the data using Species Sensitivity Distribution (SSD) method. Among the seven commonly used SSD models, S-Logistic and S-Gompertz models fitted the acute and chronic toxicity curves the best, respectively. Developed acute sediment quality criteria was CMC_{sed} = 0.00530 μg·g^{-1}, and chronic sediment quality criteria was CCC_{sed} = 0.00106 μg·g^{-1}. These sediment quality criteria provided scientific guidance for the assessment on ecological risks of γ-HCH and environment remediation.

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Keywords: γ-HCH; SSD; sediment quality criteria; equilibrium partitioning approach

林丹(lindane, >90% 的 γ-HCH)是一种应用非常广泛的有机氯农药,具有一定的疏水性($\log K_{ow}$ = 3.7 ± 0.5)[1],在环境样品,尤其是土壤、沉积物样品中广泛检出[2-5],并可通过食物链富集[6-7],最终对生态环境造成危害。虽然我国于 2000 年停止林丹生产[8],但 γ-HCH 仍在水体环境中频繁检出[9-11]。沉积物作为水生态系统的重要组成部分,是污染物的汇和源,对水环境起着重要的调节作用[12]。

相对于水质基准,世界范围内的沉积物质量基准(Sediment Quality Criteria, SQC)研究起步较晚,于 20 世纪 80 年代初期在北美开始[13-14],随后在欧洲、澳大利亚等国家和地区陆续开展,但目前被广泛认可的沉积物质量基准研究方法主要集中于北美地区[15-16]。我国的沉积物质量基准尚处于起步阶段,且主要集中在重金属基准值研究[17-19],有机污染物基准值十分缺乏。现阶段的沉积物质量基准研究主要是以生物效应数据库和相平衡分配为基础[20],如利用生物效应数据库的效应范围法(Effect Range Approach, ERA)、效应水平法(Effect Level Approach, ELA)等经验型方法,其区别在于生物数据库的适用范围及数理统计方法的不同,再如利用相平衡分配理论的相平衡分配法(Equilibrium Partitioning Approach, EqPA)、组织残留法(Tissue Residue Approach, TRA)等理论型方法[21]。

物种敏感度分布法(Species Sensitivity Distributions, SSD)可以看作是一种基于生物效应数据库的基准研究方法,于 20 世纪 70 年代末被提出,并逐渐应用于推导水质基准[22-23]。相较于其他基准研究方法,SSD 利用不同生物对污染物的敏感性差异建立生物毒性效应曲线,进而得到生态风险阈值 HC_p 来进行基准计算。HC_p 是为保护(100-p)% 的物种所允许的最大环境许可浓度值,p 的取值需考虑环境保护需求、经济发展以及统计科学性等各方面的因素,通常 p 取值为 5[24-25],即能够保护某环境中 95% 的生物。国际上 SSD 通常采用"三门八科"的生物筛选原则,我国提出"三门六科",覆盖生物种类广泛,并且适用于多种类型的环境条件和污染物,随着数据库的不断更新和扩大,基准值可以更新并更加准确[26-27]。目前 SSD 法主要集中在水质基准研究,并且已经得到了普遍认可和比较广泛的应用[28-30],但是尚没有应用在沉积物质量基准推导方面。本文拟将 SSD 应用于 γ-HCH 的沉积物质量基准研究,建立沉积物-物种敏感度分布曲线(SSD_{sed}),最终推导 γ-HCH 沉积物急性基准值 CMC_{sed}、慢性基准值 CCC_{sed},为保护我国淡水生态环境安全提供科学依据。

1 研究方法(Research methods)

1.1 已有毒性数据搜集及筛选

沉积物毒性数据来源于本课题组实测值及目前已发表的相关文献,共搜集直接沉积物毒性数据 5 个。由于沉积物毒性数据量较缺乏,本文搜集了美国国家环保局(US EPA) ECOTOX 水生生物效应数据库(http://cfpub.epa.gov/ecotox/)中的淡水水生生物毒性数据,并通过相平衡分配法将其转化为沉积物毒性数据;经搜集并筛选得到林丹急性生物毒性效应数据 66 个,慢性数据 15 个。

筛选原则为:①选取我国本土及引入我国并稳定繁殖的淡水底栖生物毒性数据,生物区系需最小满足"三门六科"[24],鱼类需满足栖息于水体中下层的条件;②剔除未考虑质量控制、试验条件不合理等有疑点的试验数据,如是否为淡水环境等,优先选取动态试验数据;③选择与生物生长、死亡、繁殖等能反映生物有机体生存状况的毒性效应终点;④急性毒性数据选择 96 h 及 96 h 内标准试验周期数据,若同一生物存在 96 h 内多个试验周期数据,选取试验周期长的数据;⑤慢性毒性数据优先选取 14 d 及以上的 NOEC 或 LOEC 值,若没有 NOEC 或 LOEC 值,也可以用合理的 14 d 以上的 EC_{50} 值和 LC_{50} 值代替;⑥若同一物种存在多个生命阶段毒性数据,在数据条件均合理的情况下,选择物种最敏感生命阶段数据;⑦当某一相同的测试终点有多个合理的毒性数据时,取其几何平均值;⑧数据单位统一换算为 $\mu g \cdot g^{-1}$(干重)。

1.2 毒性数据处理

1.2.1 淡水水生生物毒性数据向沉积物毒性数据转化

当间隙水中污染物浓度达到 C_{water} 时,可对生物产生某种效应,此时对应的沉积物毒性效应浓度 C_{sed} 为[31]:

$$C_{sed}(\mu g \cdot g^{-1}) = \frac{K_{oc} \times f_{oc} \times C_{water}(\mu g \cdot L^{-1})}{1000}$$

式中:C_{sed} 为淡水水体污染物浓度转化后对应的沉积物毒性效应浓度;C_{water} 为淡水水体中污染物的生

物毒性效应浓度;K_{oc}为固相有机碳分配系数,即污染物在沉积物有机碳和水相中的浓度比,采用本课题组实测值 3.04[32];f_{oc}为沉积物中有机碳质量分数,通常采用 1%[33]。

1.2.2　SSD 曲线拟合

将所有搜集到的沉积物生物毒性效应数据进行汇总,并按照浓度由小到大进行排序,对数据进行编号,计算不同生物效应数据所占的累积百分比,即 p =数据编号/(数据总和+1)×100%,得到沉积物毒性效应数据排序表。

设定横坐标为淡水沉积物生物毒性浓度对数值,纵坐标为受影响物种比例;选用 S-logistic(逻辑斯蒂模型)、S-weibull(韦布尔模型)、S-Gompertz(龚珀资模型)、exponential growth(指数增长模型)、Gaussian(高斯模型)、Logarithm(对数模型)、Lorentzian 等 7 种常用模型,采用 origin 8.0 软件进行 SSD 曲线模拟[34-36],并筛选得出拟合最优的 SSD 模型。

1.2.3　基准值计算

观察不同模型的拟合效果,在 P 值相同的情况下,选用 R^2 最高的模型进行 HC_5 计算。急性基准值推导参考美国在短期水质基准制定中采用最终急性值(Final Acute Value, FAV)除以安全系数 2 得到 CMC 的方法,即 $CMC_{sed}=\frac{HC_{5\text{-acute}}}{2}$;由于拟合曲线选用的慢性数据大部分为 NOEC,慢性基准值推导参考最终慢性值(Final Chronic Value, FCV)直接作为 CCC 使用的方法,即 $CCC_{sed}=HC_{5\text{-chronic}}$[37]

2　结果与讨论(Results and discussion)

2.1　SSD 模型拟合结果

应用 origin 9.0 对急慢性毒性数据进行不同模型的拟合,得出最优模型对应的参数如表 1 所示。

S-Logistic 模型对 γ-HCH 沉积物急性毒性数据拟合效果最佳,拟合曲线如图 1a 所示,$HC_{5\text{-acute}}$ = 0.0106 μg·g^{-1},$CMC_{sed}=\frac{HC_{5\text{-acute}}}{2}$=0.00530 μg·g^{-1};S-Gompertz 模型对 γ-HCH 沉积物慢性毒性数据拟合效果最佳,拟合曲线如图 1b 所示,$CCC_{sed}=HC_{5\text{-chronic}}$ =0.00106 μg·g^{-1}。

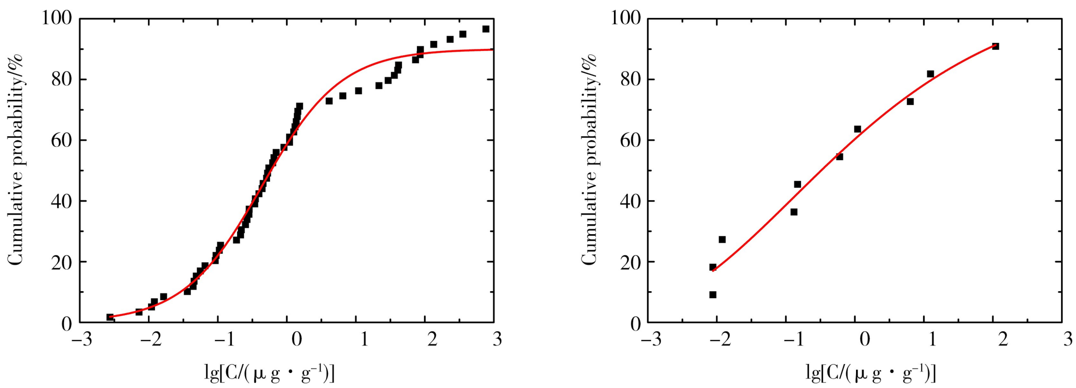

图 1　模型拟合 γ-HCH 急慢性毒性物种敏感度分布(SSD)曲线

注:a.S-Logistic; b.S-Gompertz。

Fig. 1　Models for fitting SSD curve with acute and chronic toxicity data of **γ**-HCH

Note: a.S-Logistic; b.S-Gompertz.

表 1　最优 SSD 模型对 γ-HCH 毒性数据拟合参数

Table 1　Fitting parameters for the optimal SSD models of γ-HCH toxicity data

急性毒性模型 Model for the acute toxicity	拟合公式 Fitting formulas	参数 Parameters	慢性毒性模型 Model for the chronic toxicity	拟合公式 Fitting formulas	参数 Parameters
S-Logistic	$y=\frac{a}{1+e^{-k(x-xc)}}$	a=90.033 xc=-0.357 k=1.752 R^2=0.98615 P<0.0001	S-Gompertz	$y=a\times e^{-e^{-k(x-xc)}}$	a=112.157 xc=-0.882 k=0.542 R^2=0.9666 P<0.0001

2.2 与世界其他地区基准值比较

目前,国际上推导并应用于实践的主要基准值,如表 2 所示。可以看出,本文推导的 γ-HCH 慢性基准值 CCC_{sed} 和急性基准值 CMC_{sed} 均略高于其他国家,但基本都在一个数量级范围之内,是可以比较的。与其他国家发布的基准值产生差距的原因可能有以下几个方面:①推导与计算方法的差异造成基准值不同,如 PEL、ISQG 等值是利用生物效应数据库计算得到的基准值,ESB 是利用相平衡法推导出的理论基准值,而本文推导的 CCC_{sed} 、CMC_{sed} 值则是采用物种敏感度分布曲线对毒性数据进行模型拟合,然后根据推导基准值所需的生物效应累计百分比计算得出;②不同地区间的物种区系差异也是造成基准值差异的主要原因,物种的选择会对 SSD 的拟合结果产生直接影响;③本文推导的基准值也存在一定的不确定性,由于有一部分数据是通过相平衡转化的水体毒性效应数据,该类数据未考虑底栖生物对沉积物的摄食、直接接触等过程,对推导的结果也会存在一定偏差。

2.3 我国七大水系沉积物中林丹浓度与基准值的比较

在中国知网(CNKI)及 web of science 上按照"林丹(lindane)"、"沉积物(sediment)"、"中国(China)"关键词筛选出 2000—2017 年公开发表的关于我国七大水系沉积物中林丹残留浓度的中英文文献 30 篇,涉及长江水系[39-45]、黄河水系[46-52]、珠江水系[53-57]、海河水系[58-60]、淮河水系[61-65]、松花江水系[66]、辽河水系[67],得到沉积物中林丹残留浓度数据 223 个,绘制浓度分布情况如图 2 所示,将所得浓度数据与本文推导基准值进行比较,得到表 3。

总体来说,大部分水体沉积物中 γ-HCH 的浓度

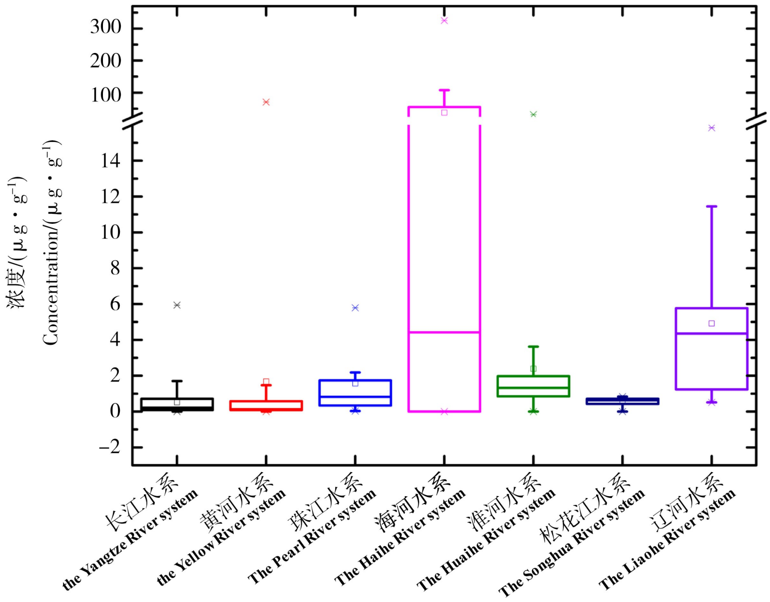

图 2 我国七大水系沉积物中林丹浓度分布情况

Fig. 2 Distributions of lindane in the sediments of seven river systems, China

表 2 γ-HCH 沉积物质量基准推导值比较

Table 2 Comparison of developed sediment quality criteria for **γ-HCH**

基准值推导机构 Offices or departments for SQC development	本文推导基准值 Developed SQC in this study		美国[33] USEPA[33]	加拿大[38] CCME[38]		澳大利亚[26] ANZECC[26]	
基准值/($\mu g \cdot g^{-1}$ dry weight) Sediment Quality Criteria/ ($\mu g \cdot g^{-1}$ dry weight)	CCC_{sed}	CMC_{sed}	ESB	ISQG	PEL	ISQG-Low	ISQG-High
	0.00106	0.00530	0.0037	0.00094	0.00138	0.00032	0.001

注:ESB 代表相平衡沉积物基准;ISQG 代表临时沉积物质量基准值;PEL 代表可能效应水平。

Note: ESB = equilibrium partitioning sediment benchmark; ISQG = interim sediment quality guideline; PEL = probably effect level.

表 3　我国七大水系中林丹浓度与本文推导基准值比较

Table 3　Comparison between the developed sediment quality criteria and the concentrations of lindane in seven river systems

七大水系 Seven river systems	数据点总数 Total number of data	$\leq CCC_{sed}$		$CCC_{sed} \sim CMC_{sed}$		$\geq CMC_{sed}$	
		数据点个数 Number of data	比例/% Ratio/%	数据点个数 Number of data	比例/% Ratio/%	数据点个数 Number of data	比例/% Ratio/%
长江水系 The Yangtze River system	71	58	81.7	12	16.9	1	1,4
黄河水系 The Yellow River system	63	51	81.0	11	17.45	1	1.6
珠江水系 The Pearl River system	13	8	61.5	4	30.8	1	7.7
海河水系 The Haihe River system	19	9	47.4	1	5.3	9	47.4
淮河水系 The Huaihe River system	35	13	37.1	21	60.0	1	2.9
松花江水系 The Songhua River system	10	10	100.0	0	0.0	0	0.0
辽河水系 The Liaohe River system	12	3	25.0	6	50.0	3	25.0
全国范围 Nationwide	223	152	68.2	55	24.7	16	7.2

水平低于本文推导基准值CCC_{sed}，从该角度看，大部分水系生态风险较低。但目前海河水系 47.4% 的采样点 **γ**-HCH 残留浓度高于CMC_{sed}，因此，须加强海河水系治理工作。但由于搜集到的海河水系数据资料采样点大部分集中于污染较重的排污河，导致可能高估海河水系生态风险。

综上所述：本文收集了大量沉积物中 γ-HCH 的急慢性毒性数据，通过沉积物-物种敏感度分布曲线拟合，从而推导 γ-HCH 急慢性基准值 CCC_{sed}、CMC_{sed}基准阈值。针对急性毒性数据，S-Logistic 模型拟合效果佳，得到急性基准值$CMC_{sed}=0.00530$ $\mu g \cdot g^{-1}$；对于慢性毒性数据，S-Gompertz 模型拟合效果佳，得到慢性基准值$CCC_{sed}=0.00106$ $\mu g \cdot g^{-1}$。总体来说，本文推导的基准值和国际上其他国家现行的基准域值相差在一个数量级范围内，相对是可以比较的，因此，SSD 是一个比较可靠的方法。我国大部分水体沉积物中 γ-HCH 的残留浓度均低于其沉积物基准低值CCC_{sed}，说明其风险较低。但是，在个别流域如海河存在一定风险，需要引起足够的重视。

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