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淡水环境中短链氯化石蜡的预测无效应浓度及生态风险评估

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摘要: 短链氯化石蜡(short-chain chlorinated paraffins, SCCPs)是《斯德哥尔摩公约》增列的一类持久性有机污染物。搜集筛选出 SCCPs 对 8 种淡水生物的慢性毒性数据,构建了物种敏感度分布曲线(SSD),推导出 SCCPs 的淡水预测无效应浓度($PNEC_{water}$)为 0.425 μg·L^{-1},淡水沉积物预测无效应浓度($PNEC_{sed}$)为 992.5 μg·kg^{-1}。搜集了国内外部分淡水河流水体及沉积物中 SCCPs 环境暴露数据,运用商值法,评估 SCCPs 的生态风险。结果表明,长江中游和白洋淀水体风险商范围为 2.6 ~ 154.4 和 3.7 ~ 132.5,处于高风险;国外河流 SCCPs 污染水平较低,北美地区与日本淡水河流 SCCPs 风险商小于 1,处于低风险。长江中游沉积物的 SCCPs 的风险商高达 400.6,呈现显著风险,欧洲工业区域淡水沉积物中 SCCPs 存在潜在风险。本研究为 SCCPs 水质标准制定与环境风险管理提供参考依据。
关键词: 短链氯化石蜡;物种敏感度分布;生态风险评估;预测无效应浓度;风险商
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Predicted No Effect Concentration and Ecological Risk Assessment of SCCPs in Freshwater Environments

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Abstract: Short-chain chlorinated paraffins (SCCPs) are a class of persistent organic pollutants newly listed in the Stockholm Convention. In this study, the chronic toxicity data of SCCPs to eight aquatic organisms were collected and screened, and the species sensitivity distribution (SSD) curves were constructed. The predicted no effect concentration (PNEC) of SCCPs for freshwater was derived as 0.425 μg·L^{-1}($PNEC_{water}$) and for freshwater sediment was 992.5 μg·kg^{-1}($PNEC_{sed}$), respectively. The environmental exposure concentrations of SCCPs in freshwater rivers and sediments in China and several other countries were collected, and the ecological risk of SCCPs was evalu-

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ated by quotient method. The results showed that the hazard quotients (HQs) of water bodies in the midstream of the Yangtze River and Baiyangdian Lake were 2.6 ~ 154.4 and 3.7 ~ 132.5, respectively, which were at high risk levels. The HQs of SCCPs in foreign rivers showed that they were at low risk levels. For example, the HQs for rivers in North America and Japan were lower than 1. The HQs of sediments in the midstream of the Yangtze River were up to 400.6, showing significantly high risk in these sediments. However, freshwater sediments in European industrial regions were at low risks. This study could provide information for water quality standard establishment and environmental risk management of SCCPs.

Keywords: SCCPs; SSD; ecological risk assessment; predicted no effect concentration; hazard quotient

氯化石蜡(chlorinated paraffins, CPs),又称多氯代正构烷烃,分子式为 $C_xH_{(2x-y+2)}Cl_y$,按碳链长度可分为 3 类:短链氯化石蜡(C_{10} ~ C_{13}, SCCPs),中链氯化石蜡(C_{14} ~ C_{17}, MCCPs)和长链氯化石蜡(C_{18} ~ C_{30}, LCCPs),氯化程度一般在 30% ~70%之间(按质量计算)[1]。其中短链氯化石蜡(SCCPs)有上千种同族体和异构体,具有良好的热稳定性和化学稳定性,已作为增塑剂和阻燃剂广泛用于金属加工液、涂料、密封剂、粘合剂、皮革处理剂、塑料和橡胶的生产[2]。SCCPs 因其具有环境持久性、生物累积性、长距离迁移性和生物毒性,于 2017 年被列入《关于持久性有机污染物(POPs)的斯德哥尔摩公约》的受控清单[3]。SCCPs 可以干扰内分泌系统[4]和免疫系统[5],影响正常代谢,破坏机体内环境稳定,具有发育毒性[6-7]、致畸性[8]和致癌性[9]。我国是最大的 CPs 生产国和使用国[10-11],在许多河流、湖泊等水体中皆检测出 SCCPs,对水环境有潜在风险。

SCCPs 进入水体后,可以在水生生物体内蓄积[12-13],对水生生物产生毒性。环境剂量的 SCCPs 可以对水生生物的发育、基因表达和激素水平等产生显著影响。Ren 等[14]采用代谢组学方法探讨了 SCCPs 暴露对斑马鱼(*Danio rerio*)胚胎发育和代谢的影响,发现随着 SCCPs 的浓度增加,孵化后幼鱼的存活率显著降低,13 d-LC_{50} 为 34.4 $\mu g \cdot L^{-1}$。另有研究指出,一定剂量的 SCCPs 可通过下调斑马鱼下丘脑-垂体-甲状腺轴相关的 *tyr*、*ttr*、*dio2* 和 *dio3* 的 mRNA 水平影响甲状腺激素水平[15]。

物种敏感度分布法(Species Sensitivity Distribution, SSD)是将不同生物对某种污染物的敏感度通过一定的函数进行拟合[16],采用的拟合模型包括 Logistic、Log-Logistic、Normal、Log-Normal 和 Extreme Value 等,计算求得保护一定百分比生物的污染物浓度。目前 SSD 普遍应用于淡水水生生物水质基准推导[17]。平衡分配法(Equilibrium Partitioning, EP)是美国环境保护局(US EPA)推荐的以污染物在间隙水、沉积物和底栖生物体内的浓度的热力学动态平衡为基础的沉积物基准推导方法,适用于辛醇-水分配系数对数($\lg K_{ow}$)大于 3 的非离子型有机物[18]。

预测无效应浓度(predicted no effect concentration, PNEC)是欧盟风险评价技术导则文件(Technical Guidance Document on Risk Assessment, TGD)[19]中推荐的用于化学物质环境风险评价的毒性安全阈值。本文参考我国最新发布的《淡水水生生物水质基准制定技术指南》(下称“指南”)[20],基于淡水水生生物物种的毒性数据,推导 SCCPs 淡水 $PNEC_{water}$ 与 $PNEC_{sed}$(沉积物以干质量计)。搜集国内外水体及沉积物 SCCPs 暴露数据,利用商值法(HQ)评估国内外水环境 SCCPs 生态风险,为 SCCPs 水质标准制定与环境风险管理提供参考依据。

1　材料与方法(Materials and methods)

1.1　数据获取与筛选

SCCPs 的生态毒性数据来自公开发表的文献及 ECOTOX 毒性数据库(https://cfpub.epa.gov/ecotox/search.cfm)等。参照“指南”,筛选 SCCPs 对水生生物的慢性毒性数据,筛选原则如下:(1)有明确测试终点、暴露时间;(2)优先选择流水式实验及对试验溶液浓度的监控;(3)选择慢性毒性终点包括无可见效应浓度(NOEC)或最低可见效应浓度(LOEC)或 10%效应浓度(EC_{10});(4)弃用离群值(同种生物毒性值相差超过 1 个数量级);(5)对同一物种选择最敏感试验终点的数据;(6)其他弃用毒性数据,包括在实验设计中未设计试验对照组、对照组的试验生物表现异常、稀释用水为去离子水或蒸馏水、暴露时间不适宜、试验所用化合物的理化状态不符合“指南”要求等。

1.2　SSD 构建及 $PNEC_{water}$ 推导

采用 SSD 法获得 SCCPs 水环境预测无效应浓

度($PNEC_{water}$)。大致步骤为如下。对毒性数据进行升序排列,如 1, 2,…, N,计算每个物种毒性数据对应的累计概率。

$$P = R/(N+1)\times 100\%$$

式中:P 为第 R 个物种的累计概率;R 为物种排序等级;N 为物种的总数。选取合适的数学模型构建物种敏感度分布曲线。SSD 曲线上指定百分数对应的浓度即为基准值(HC_X),X 常取 5,表示为 95% 以上的物种受到保护时的浓度。

SSD 曲线的拟合采用"指南"附件 China-WQC 软件,并计算 5% 物种危害浓度(HC_5),单位取 μg·L^{-1}。水体预测无效应浓度计算公式如下。

$$PNEC_{water} = HC_5/AF$$

式中:$PNEC_{water}$ 为水体预测无效应浓度(mg·L^{-1});AF 为评价因子,取值范围为 1~5。本研究取 5[21]。

1.3 SCCPs 淡水 $PNEC_{sed}$ 的推导方法

SCCPs 的推导方法参考 TGD 中推荐的平衡分配法。沉积物 $PNEC_{sed}$ 计算方法如下。

$$PNEC_{sed,wet\ weight} = K_{susp\text{-}water}/RHO_{susp}\times PNEC_{water}\times 1000$$

$$RHO_{susp} = F_{solid\text{-}susp}\times RHO_{solid}+F_{water\text{-}susp}\times RHO_{water}$$

$$K_{susp\text{-}water} = F_{water\text{-}susp}+F_{solid\text{-}susp}\times(K_{p\text{-}susp}/1000)\times RHO_{solid}$$

$$K_{p\text{-}susp} = F_{oc\text{-}susp}\times K_{oc}$$

式中:$PNEC_{sed,wet\ weight}$ 为以湿质量计的沉积物预测无效应浓度(mg·kg^{-1});RHO_{susp} 为悬浮物湿质量,计算得 1 150 kg·m^{-3};$K_{susp\text{-}water}$ 为污染物在悬浮物-水分配系数,计算得 4 028.4 m^3·m^{-3};$F_{solid\text{-}susp}$ 为悬浮物中固体物比例(ϕ_{solid}),默认值为 0.1 m^3·m^{-3};RHO_{solid} 为固相的密度,默认值为 2 500 kg·m^{-3};$F_{water\text{-}susp}$ 为悬浮物中水的比例(ϕ_{water}),默认值为 0.9 m^3·m^{-3};RHO_{water} 为水的密度,默认值为 1 000 kg·m^{-3};$K_{p\text{-}susp}$ 为污染物在悬浮物中的固-水分配系数,计算得 16 110 L·kg^{-1};$F_{oc\text{-}susp}$ 为悬浮物中有机碳比例(w_{OC}),本研究取 0.1 kg·kg^{-1};K_{oc} 为污染物有机碳-水分配系数(L·kg^{-1}),查询 EPI Suite V 4.1 软件数据库 SCCPs 的 K_{oc} 为 161 100 L·kg^{-1}。

根据 TGD 方法得到的 $PNEC_{sed}$ 是以湿质量计的,而沉积物中污染物暴露浓度通常以干质量表示,因此需要进行换算。TGD 默认的湿质量悬浮物含 90% 的水(固相密度为 2 500 kg·m^{-3}),悬浮物的湿质量为 1 150 kg·m^{-3},后者与前者之比为 4.6。由此得出,以干质量计和湿质量计的沉积物 PNEC 之比为 4.6。

1.4 生态风险评估方法

商值法通过污染物的生物毒性数据与自然水体中暴露浓度的比值,评价该污染物在环境中的风险概率和危害程度[22]。风险商值(HQ)的计算公式为:

$$HQ = C/PNEC$$

式中:C 为污染物的水环境暴露浓度。风险程度划分为当 HQ>1 时,为高风险;当 1>HQ>0.1 时,为中风险;当 HQ<0.1 时,为低风险[23]。

2 结果与讨论(Results and discussion)

2.1 水生生物物种和数据

根据数据的筛选原则,搜集整理 SCCPs 慢性毒性数据(表 1),共获得 4 门 7 科 8 种水生生物的 8 个慢性毒性数据,暴露时间为 4~49 d,NOEC 值为 0.005~0.39 mg·L^{-1}。最敏感的水生生物为大型溞(*Daphnia magna*),其次为糠虾(*Mysidopsis bahia*)。中肋骨条藻(*Skeletonema costatum*)的敏感性介于虹鳟(*Oncorhynchus mykiss*)与糠虾之间,最不敏感的是羊角月牙藻(*Selenastrum capricomutum*),表明水生动物对 SCCPs 不一定比水生植物敏感;对比 2 种藻类,中肋骨条藻 4 d-NOEC 为 0.012 mg·L^{-1},而羊角月牙藻 10 d-NOEC 为 0.39 mg·L^{-1},对 SCCPs 的敏感度存在差异。无脊椎动物对污染物比脊椎动物更加敏感,大型溞(*Daphnia magna*)是水生食物链中的初级代谢者,对 SCCPs 最为敏感,这与前人研究一致[24]。但摇蚊(*Chironomus tentans*)作为节肢动物,49 d-NOEC 为 0.061 mg·L^{-1},略大于脊索动物门中的虹鳟和青鳉(*Oryzias latipes*)。

2.2 SCCPs 的淡水 $PNEC_{water}$ 与 $PNEC_{sed}$ 推导

将获得的 4 门 8 种水生生物慢性毒性数据按照"指南"方法构建 SSD 曲线,拟合模型包括 Logistic、Log-Logistic、Normal、Log-Normal 和 Extreme Value,拟合参数如表 2 所示。决定系数(R^2)越接近于 1,均方根误差(RMSE)越接近于 0,残差平方和(SSE)越接近于 0,K-S 检验 P 值>0.05,说明毒性数据拟合最佳。综合 4 项参数,拟合结果优度排序为:Extreme Value > Normal > Log-Normal > Logistic > Log-Logistic。采用拟合较好的极值分布(Extreme Value)模型(图 1),模型 R^2 为 0.9301,用该模型外推,计算 SCCPs 的 HC_5 为 2.1232 μg·L^{-1},推导 SCCPs 的 $PNEC_{water}$ 为 0.425 μg·L^{-1},$PNEC_{sed}$ 推导得 992.5 μg·kg^{-1}(干重)。对比欧盟(EU)先前推导出的 $PNEC_{water}$(0.5 μg·L^{-1})和 $PNEC_{sed}$(1 446.7 μg·kg^{-1})[25]略有不

同,可能是拟合模型的不同导致的部分差异。

2.3　国内外主要淡水水体中 SCCPs 的生态风险分析

搜集国内外水体中 SCCPs 的暴露浓度(表 3)。结果显示,中国流域水体中 SCCPs 的浓度范围为 1 131 ~ 56 305.9 ng·L^{-1},国外部分流域中浓度范围为 1.194 ~ 2 100 ng·L^{-1}。中国淡水水体中 SCCPs 的浓度远高于日本(57.62 ng·L^{-1})和北美地区(37.7 或 1.194 ng·L^{-1}),长江中游 SCCPs 的平均浓度达到 18 989 ng·L^{-1},白洋淀 SCCPs 水体中平均浓度为 7 223 ng·L^{-1}。HQ 值如图 2 所示,长江中游及白洋淀所有样点 HQ 均大于 1,水体环境整体处于高风险,上海淡水水系 HQ 最小值处于低风险,部分水体处于高风险。而日本的 SCCPs 浓度远小于本研究推

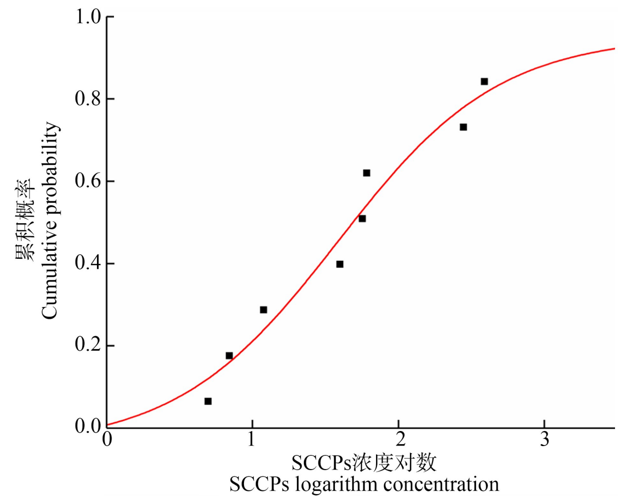


图 1　SCCPs 的物种敏感度分布曲线

Fig. 1　Species sensitivity distribution curve of SCCPs

表 1　短链氯化石蜡(SCCPs)对不同水生物种的最大无效应浓度(NOEC)

Table 1　The no observed effect concentration (NOEC) of short-chain chlorinated paraffins (SCCPs) to different aquatic species

生物分类 Biotaxonomy	中文名 Chinese name	物种拉丁名 Latin name of species	暴露时间/d Exposure time/d	温度/℃ Temperature/℃	NOEC /(mg·L^{-1})	参考文献 References
节肢动物门溞科溞属 Arthropoda Daphniidae	大型溞	*Daphnia magna*	21	20	0.005	[25]
节肢动物门糠虾科 Arthropoda Mysidae	糠虾	*Mysidopsis bahia*	28	25	0.007	[25]
硅藻门圆筛藻科骨条藻属 Bacillariophyta Coscinodiscaceae	中肋骨条藻	*Skeletonema costatum*	4	20	0.012	[25]
脊索动物门鲑科鲑属 Chordates Salmonidae *Salmo*	虹鳟鱼	*Oncorhynchus mykiss*	15 ~ 20	10	0.04	[25]
脊索动物门鳉科青鳉属 Chordates Cyprinodontidae	青鳉	*Oryzias latipes*	20	25	0.057	[25]
节肢动物门摇蚊科摇蚊属 Arthropoda Chironomidae	伸展摇蚊	*Chironomus tentans*	49	21 ~ 23	0.061	[25]
脊索动物门鳉科鳉属 Chordates Cyprinodontidae	对杂色鳉	*Cyprinodon variegatus*	32	25	0.28	[25]
绿藻门小球藻科月牙藻属 Chlorophyta Chlorellaceae	羊角月牙藻	*Selenastrum capricomutum*	10	24	0.39	[25]

表 2　中国 SCCPs 水生生物水质慢性基准推算结果

Table 2　Calculating results of chronic benchmark of SCCPs for the protection of aquatic organisms in China

拟合函数 Fitting function	5%危害浓度(HC_5) 5% Hazardous concentration (HC_5)	决定系数(R^2) Coefficient of determination (R^2)	均方根(RMSE) Root mean square error (RMSE)	残差平方和(SSE) Residual sum of squares (SSE)	K-S 检验 K-S test
逻辑斯蒂分布 Logistic	3.4674	0.8789	0.0886	0.0628	0.9885
对数逻辑斯蒂分布 Log-Logistic	5.0466	0.8421	0.1012	0.0819	0.9796
正态分布 Normal	3.3574	0.9026	0.0795	0.0505	0.9976
对数正态分布 Log-Normal	4.9659	0.8790	0.0886	0.0628	0.9904
极值分布 Extreme Value	2.1232	0.9301	0.0673	0.0362	0.9995

导出的 $PNEC_{water}$(425 ng·L^{-1}),HQ<0.1,表明其水体风险低。这可能与日本对 SCCPs 的环境毒性[5]及生态风险评估较早[26]有关。欧洲地区 Llobregat 河(300 ~ 2 100 ng·L^{-1})和 Da-wen 河(200 ~ 1 700 ng·L^{-1})部分样点 SCCPs 的水体污染浓度也超过了 $PNEC_{water}$,暴露浓度较高样点与 SCCPs 工业分布一致。Zhang 等[27]的研究表明,中国在 2010—2014 年 SCCPs 向水体中的排放量为 2 189.07 t,最大排放源来自金属加工业,并且集中在东部较发达地区。由此可见,中国水体中 SCCPs 的污染现状比较严峻。

2.4 国内外主要淡水沉积物中 SCCPs 的生态风险评估

本研究推算出的淡水 $PNEC_{sed}$(992.5 μg·kg^{-1})与国内外主要淡水沉积物中 SCCPs 暴露浓度比较(表 4)发现,中国长江中游(54 512 ng·g^{-1})与白洋淀(24 454 ng·g^{-1})沉积物中 SCCPs 暴露浓度远远高于 $PNEC_{sed}$(992.5 μg·kg^{-1}),长江中游 SCCPs 浓度最高为 397 600.4 ng·g^{-1},是 SCCPs 淡水 $PNEC_{sed}$ 的 400.6 倍,

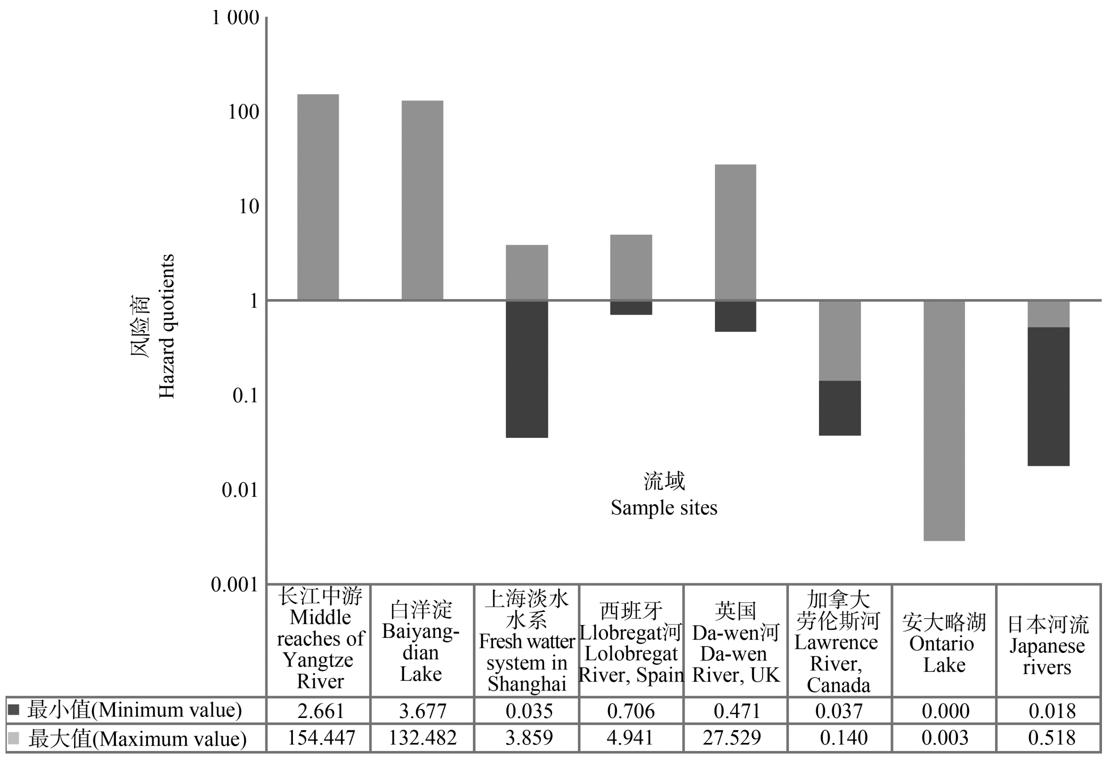


图 2 国内外不同淡水流域水体中 SCCPs 的风险商(HQ)

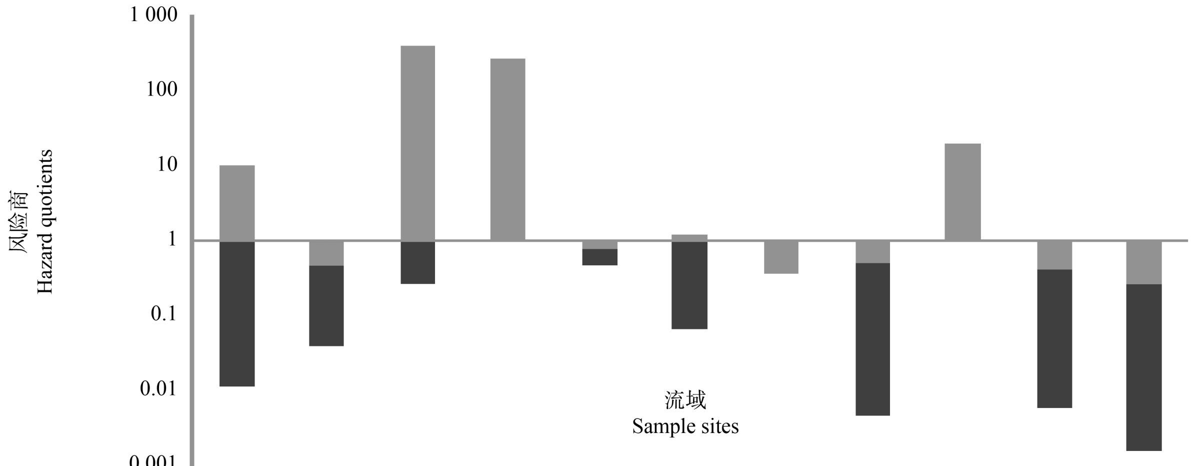
Fig. 2 The hazard quotient (HQ) of SCCPs in different freshwater basins at home and abroad

表 3 国内外部分淡水水体中 SCCPs 暴露数据

Table 3 SCCPs exposure concentration in freshwater environment at home and abroad

(ng·L^{-1})

流域 Basin	范围 Range	均值 Mean	参考文献 References
长江中游 Middle reaches of Yangtze River	1 131 ~ 65 640	18 989	[28]
白洋淀 Baiyangdian Lake	1 562.8 ~ 56 305	7 223	[28]
上海淡水水系 Fresh water system in Shanghai	15.0 ~ 1 640	448	[29]
西班牙 Llobregat 河 Llobregat River, Spain	300 ~ 2 100	—	[30-31]
英国 Da-wen 河 Da-wen River, UK	200 ~ 1 700	—	[32]
加拿大劳伦斯河 Lawrence River, Canada	15.74 ~ 59.57	37.7	[33]
安大略湖 Ontario Lake	0 ~ 1.194	—	[34]
日本河流 Japanese rivers	7.6 ~ 220	57.62	[26,35]



	黄河中游 Middle reaches of the Yellow River	辽河流域 Liaohe River Basin	长江中游 Middle reaches of Yangtze River	白洋淀 Baiyang-dian Lake	珠江北江 Pearl River North River	椒江 Jiaojiang River	捷克河流 Rivers of Czech	日本河流 Japanese rivers	挪威河流 Rivers of Norway	北美五大湖 The Great Lakes of North America	北极地区湖泊 Lakes in the Arctic
■ 最小值(Minimum value)	0.012	0.040	0.254	1.280	0.484	0.068	0.000	0.005	0.000	0.006	0.002
■ 最大值(Maximum value)	9.834	0.484	400.605	256.124	0.816	1.199	0.350	0.488	19.547	0.413	0.259

图 3　国内外不同淡水流域沉积物中 SCCPs 的风险商(HQ)

Fig. 3　The hazard quotient (HQ) of SCCPs in sediments from different freshwater basins at home and abroad

表 4　国内外部分淡水沉积物中 SCCPs 暴露数据

Table 4　Exposure concentration of SCCPs in some freshwater sediments at home and abroad

($ng \cdot g^{-1}$)

流域范围 Basin	范围 Range	均值 Mean	干湿重 Dry and wet weight	参考文献 References
黄河中游　Middle reaches of the Yellow River	11.6 ~ 9 760	903.4	干重 Dry weight	[36]
辽河流域　Liaohe River Basin	39.8 ~ 480.3	74.4	干重 Dry weight	[37]
长江中游　Middle reaches of Yangtze River	251.9 ~ 397 600.4	54 512	干重 Dry weight	[28]
白洋淀　Baiyangdian Lake	1 270 ~ 254 203	24 454	干重 Dry weight	[28]
珠江北江　Pearl River North River	480 ~ 810	610	干重 Dry weight	[38]
椒江　Jiaojiang River	67.4 ~ 1 190	466.3	干重 Dry weight	[39]
捷克河流　Rivers of Czech	0 ~ 347	121.7	干重 Dry weight	[40]
日本河流　Japanese rivers	4.9 ~ 484.4	284.3	湿重 Wet weight	[26]
挪威河流　Rivers of Norway	—	19 400	干重 Dry weight	[41]
北美五大湖　The Great Lakes of North America	5.9 ~ 410	—	干重 Dry weight	[42-44]
北极地区湖泊　Lakes in the Arctic	1.6 ~ 257	77.5	干重 Dry weight	[45]

白洋淀最高浓度样点 254 203 $ng \cdot g^{-1}$ 是 $PNEC_{sed}$ 的 256 倍,HQ 值远>1(图 3),表明 SCCPs 对长江中游和白洋淀水生生物具有高风险。辽河流域(74.4 $ng \cdot g^{-1}$)SCCPs 浓度最低,珠江北江(610 $ng \cdot g^{-1}$)和椒江(466.3 $ng \cdot g^{-1}$)HQ 均低于 1。除挪威外,国外流域沉积物中 SCCPs 的 HQ 低于 1,属于低风险。但在北极地区湖泊中依然有 SCCPs 检出,虽然浓度低于 $PNEC_{sed}$,但北极地区生态稳定与生态恢复能力相对较弱,SCCPs 随空气、洋流不断迁移,给极地地区的生态安全带来潜在风险。

2.5　SCCPs 评价结果不确定性分析

SCCPs 作为新型持久性有机污染物,目前关于其慢性毒性数据较少,对现阶段数据筛选,仅搜集到 4 门 7 科 8 种 8 个慢性毒性数据,毒性数据的丰度

不足导致不能充分反映水生生物的敏感性,随着SCCPs研究的深入,可以获得更多物种的毒性数据,提高推导的SCCPs基准值的确定性。此外SCCPs的种类繁多,分离分析困难,检测没有统一方法,对同一环境样品SCCPs分析结果也不完全相同[46]。

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