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HUANJING KEXUE

经济快速发展区场地污染特征、源-汇关系与管控对策专辑

我国经济快速发展区工业VOCs排放特征及管控对策

孟博文, 李永波, 孟晶, 李倩倩, 史斌, 周喜斌, 李金灵, 苏贵金

VOCs



制药



喷涂



橡胶

京津冀 •

• 长三角

珠三角 •

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鄱阳湖流域水体和水产品中苯酚的暴露特征及人体健康风险评估

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摘要: 苯酚(phenol)被广泛应用于工业生产, 在水环境中大量检出, 具有皮肤灼伤、抑制中枢神经和肝脏损伤等健康毒性。调查了鄱阳湖水体和水产品中苯酚的暴露浓度, 采用商值法和概率风险评估法, 基于本土人群暴露参数评估苯酚对鄱阳湖流域成人的健康风险。分析结果表明, 水体中苯酚浓度范围为 ND ~ 556.26 ng·L⁻¹, 水产品中苯酚含量范围在 11.98 ~ 255.51 μg·kg⁻¹。鄱阳湖湖区成人饮水健康风险值在 3.80 × 10⁻⁷ ~ 8.46 × 10⁻⁵ 范围内, 湖区南部河流聚集处及北部长江交汇处的成人饮水健康风险较高。成人通过食用鄱阳湖不同种类水产品产生的健康风险范围为 2.65 × 10⁻⁵ ~ 1.47 × 10⁻⁴, 食用黄颡鱼和鲢鱼产生的健康风险比其他种类水产品高一个数量级。通过 Monte Carlo 模拟评估苯酚的健康概率风险, 并探讨鄱阳湖流域人群暴露参数敏感性。结果表明鄱阳湖流域成人饮水和水产品食用的 95 百分位健康风险值处于可接受水平, 苯酚环境暴露浓度对健康风险值的影响最大。本研究结果可为鄱阳湖流域水环境中苯酚的风险管控提供参考。

关键词: 鄱阳湖; 苯酚; 水体; 水产品; 人体健康风险

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Human Health Risk Assessment of Phenol in Poyang Lake Basin

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Abstract: Phenol is widely used in the production of insulation and thermal insulation materials, adhesives, perfumes, coatings for food containers, paints, and pharmaceutical production, and is also widely detected in the aquatic environment. Long-term exposure to phenol can elicit adverse effects, such as skin burn, liver and central system damage. Here, phenol concentrations in the water and aquatic products of Poyang Lake were investigated. Human health risks from phenol to adults and adolescents were also assessed based on local population exposure parameters. The exposure concentration range of phenol in the studied water and aquatic products was not detected (ND)-556.26 ng·L⁻¹ and 11.98-255.51 μg·kg⁻¹, respectively. Human health risk based on drinking water in different areas ranged from 3.80 × 10⁻⁷-8.46 × 10⁻⁵. Higher human health risks from drinking water was detected in the southern area of Poyang Lake and at the confluence of the Yangtze River to the north. Health risks caused by different types of aquatic products ranges 2.65 × 10⁻⁵-1.47 × 10⁻⁴. In particular, human health risks from the consumption of yellow catfish and catfish are an order of magnitude higher than for other aquatic products. Probabilistic risk assessment was also conducted through Monte Carlo simulation to analyze the health risk to the population in the Poyang Lake Basin and assess its sensitivity of different exposure parameters. The 95th percentile health risk of drinking water and aquatic product consumption in the Poyang Lake Basin was calculated as being acceptable. Overall, the concentrations of phenol had the greatest impact on the calculated health risk values. This study provides valuable information for phenol risk management in the Poyang Lake basin.

Key words: Poyang Lake; phenol; water; aquatic product; human health risk

苯酚(phenol)被广泛应用于绝缘及隔热材料、粘合剂、香水生产和食品容器的涂料、油漆和制药生产等, 在水环境中被大量检出^[1-3]。我国主要用苯酚生产酚醛树脂、双酚 A 和阿司匹林等^[4,5]。苯酚具有腐蚀性、挥发性和刺激性, 容易对环境和生物造成危害, 被美国环保署(US EPA)列为重点高毒性防护对象, 成为优先控制的 129 种污染物之一^[6,7]。苯酚通过生物链积累对人体和水生生物产生不同程度的急性或者慢性毒性效应。长期饮用含苯酚类有机物污染物的水可能会使人产生头晕、贫血等不良症状, 甚至会造成肝脏和中枢神经系统的损伤^[8-10]。苯酚被

国际癌症研究机构(IARC)列为 3 类致癌物, 现有生物数据无法对其致癌性进行分级评价^[11]。目前国内外对苯酚污染的研究主要集中在苯酚的生态风险, 缺少人体健康风险的研究。

鄱阳湖位于江西省北部, 是我国第一大淡水

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湖^[12,13]. 鄱阳湖是居民饮用水源地,也是江西省乃至全国重要的水产品产地,具有重要的生态和经济地位^[14,15]. 作为江西省重要的淡水渔业基地,鄱阳湖拥有丰富的鱼类资源,其水产品经济占全省水产品经济的 60% 左右^[16]. 作为工业用水和居民饮用水源综合性水域,工业排放的苯酚进入鄱阳湖水体,在水生生物中蓄积^[17]. 苯酚经饮水和食用水产品等途径进入人体,并对健康产生危害,因此有必要开展鄱阳湖流域苯酚污染分布特征及健康风险评估.

本研究分析了鄱阳湖水体中苯酚浓度及分布特征,对鄱阳湖 9 种水产品中苯酚蓄积情况进行了讨论分析. 基于鄱阳湖区域人群暴露参数开展人体健康风险评价,探讨鄱阳湖区域人群饮水风险和不同种类水产品食用风险,以期为我国苯酚污染控制和风险管理工作提供参考.

1 材料与方法

1.1 样品采集

在鄱阳湖设置了 23 个采样点(如图 1),于 2019 年 7 月在每个样点用棕色玻璃瓶采集 2 L 水样,滴加浓盐酸将 pH 调节至 2.0 以抑制微生物活性,4℃ 避光保存. 同时采集鄱阳湖 9 种生物样品,包括:草鱼(*Ctenopharyngodon idellus*)、白鲢(*Hypophthalmichthys molitrix*)、鳙鱼(*Aristichthys nobilis*)、白鲮鱼(*Hemiculter leucisculus*)、黄颡鱼(*Pelteobagrus fulvidraco*)、鲶鱼(*Silurus asotus*)、鳊鱼(*Siniperca chuatsi*)、鲤鱼(*Cyprinus carpio*)和乌鳢(*Ophiocephalus argus* Cantor),每种生物采集 5 个,用铝箔纸包好后于 -20℃ 冷冻保存.

1.2 化学试剂

二氯甲烷(CH_2Cl_2)、甲醇(CH_3OH)、正己烷(C_6H_{14})和乙酸乙酯($\text{CH}_3\text{COOC}_2\text{H}_5$)均为农残级(Mallinckrodt Baker 公司,美国);超纯水(Milli-Q 纯水机,Millipore,美国);HLB 固相萃取柱(500 mg, Waters, 美国);C18 固相萃取柱(500 mg, Supelco, 美国);含 1% 三甲基氯硅烷(TMCS)的 *N*, *O*-双(三甲基硅烷基)乙酰胺(BSTFA)(Supelco, 美国);无水硫酸钠(Na_2SO_4)、浓盐酸(HCl)和氢氧化钠(NaOH)均为优级纯试剂(沪试).

1.3 样品前处理

水样经 0.45 μm 玻璃纤维滤膜过滤后,取 2 L 进行固相萃取. 将 C18 和 HLB 固相萃取柱串联,富集前分别取 5 mL 二氯甲烷、5 mL 甲醇和 5 mL 超纯水淋洗活化固相萃取柱. 萃取时,水样流速保持 5 $\text{mL}\cdot\text{min}^{-1}$ 通过固相萃取柱,抽真空干燥 30 min 后用 10 mL 二氯甲烷进行洗脱,洗脱液流速 2 $\text{mL}\cdot\text{min}^{-1}$.

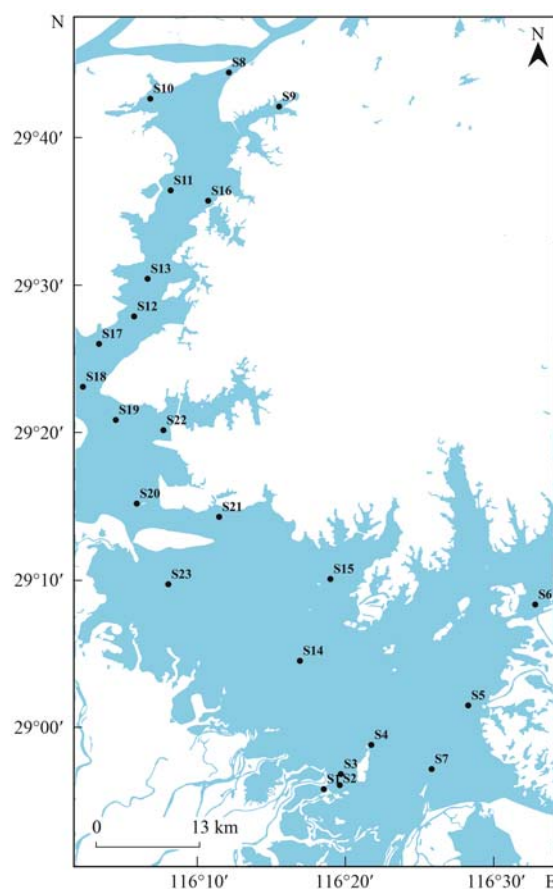


图 1 鄱阳湖采样点分布示意

Fig. 1 Sampling sites in Poyang Lake

收集洗脱液,氮吹至近干,置换溶剂为正己烷,定容至 2 mL,加入 BSTFA 进行衍生化处理,待 GC-MS 分析.

将水产品去鳞、去骨,放入搅拌机中制成匀浆状,冷冻干燥后研磨,准确称取 1 ~ 2 g (精确至 0.000 1 g) 匀质后的冻干鱼样,与硅藻土、石英砂均匀混合后用快速溶剂萃取仪(ASE)进行萃取,萃取溶剂为正己烷与二氯甲烷(1:1, 体积比). 萃取液在 40℃ 下氮吹至 3 mL, 加入到多层硅胶柱中净化,待样品完全流入后加入 10 mL 正己烷与二氯甲烷混合液(9:1, 体积比)洗脱,待其接近流干时再次加入 90 mL 混合液,将洗脱液在 40℃ 和 35 000 ~ 45 000 Pa 条件下旋转浓缩至 3 mL, 转移至 10 mL 刻度管氮吹,置换溶剂为正己烷,定容至 2 mL,加入 BSTFA 进行衍生化处理,待测.

1.4 仪器分析

使用安捷伦公司的 GC6890/MSD5975, DB-5MS 色谱柱(J&W, 美国)规格为 30 m × 0.25 mm × 0.25 μm . 进样口温度:300℃;进样体积:1.0 μL . 柱温箱升温程序:初始温度 40℃ 保持 2.0 min, 以 10℃ · min⁻¹ 的升温速率升至 300℃, 保持 15.0 min, 采用 SIM 模式扫描^[18].

1.5 质量控制

设置全过程空白样品监控分析处理过程中产生的背景值,空白样品的待测物浓度均小于方法检出限.采用外标法定量样品中苯酚,标准曲线设置7个点(0.02、0.05、0.1、0.2、0.5、1.0和2.0 $\text{mg}\cdot\text{L}^{-1}$),目标物的线性回归系数均大于0.995,相对标准偏差(RSD)小于10%.水样中苯酚的回收率为83.5%~92.2%,生物样品中的回收率为75.3%~101.1%.按照样品分析的全部步骤,用超纯水和石英砂重复10次空白平行实验,通过计算平行样品的标准差得到检出限(LOD)为5 $\text{ng}\cdot\text{L}^{-1}$ (水样)和125 $\text{ng}\cdot\text{kg}^{-1}$ (生物样),若浓度低于检出限,为统计分析未检出部分用 $0.5 \times \text{LOD}$ 替代^[19].

1.6 人体健康风险评估商值法

人体健康风险评价,分为致癌风险评价和非致癌风险评价两大类.因苯酚在IARC致癌物分类中为3类致癌物,本研究仅对苯酚对人体健康产生的非致癌性风险进行评估.非致癌风险评价依据US EPA推荐的风险评估方法^[20,21],用某一特定时间下污染物的暴露量与污染物在该暴露途径下的参考剂量之比,对比生物体对非致癌物产生反应的阈值,来评估物质对人体的健康风险HQ,用公式(1)计算^[22~24].人体健康风险评估中,US EPA推荐的非致癌风险阈值为1,当HQ大于1时存在非致癌风险,若HQ小于1时为非致癌风险较小或可忽略不计^[25].

$$\text{HQ} = \frac{\text{ADD}}{\text{RfD}} \quad (1)$$

式中,ADD为污染物的日平均暴露量, $\text{mg}\cdot(\text{kg}\cdot\text{d})^{-1}$; RfD为污染物在某暴露途径下的参考剂量, $\text{mg}\cdot(\text{kg}\cdot\text{d})^{-1}$.

通过监测人体接触的环境介质中污染物浓度,结合不同特征人群在不同环境介质中暴露频率和时间来估算人群摄取污染物的暴露剂量,用公式(2)和(3)计算饮水及水产品摄入途径下苯酚的日平均暴露量(ADD)^[26,27].

$$\text{ADD}_{\text{饮水}} = \frac{c \times \text{IR}_w \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (2)$$

$$\text{ADD}_{\text{食用}} = \frac{c \times \text{IR}_f \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (3)$$

式中,计算饮水途径暴露量时 c 为水体中苯酚的浓度, $\text{mg}\cdot\text{mL}^{-1}$; IR_w 为饮水摄入量, $\text{mL}\cdot\text{d}^{-1}$; 计算食用水产品时, c 为食用水产品中苯酚的含量, $\text{mg}\cdot\text{kg}^{-1}$; IR_f 为水产品摄入量, $\text{kg}\cdot\text{d}^{-1}$; EF为暴露频率, $\text{d}\cdot\text{a}^{-1}$; ED为暴露持续时间, a; BW为平均人体重, kg; AT为平均暴露时间, d.

本研究中,苯酚经口的参考剂量RfD为0.6 $\text{mg}\cdot(\text{kg}\cdot\text{d})^{-1}$,暴露频率(EF)为365 $\text{d}\cdot\text{a}^{-1}$,暴露持续时间(ED)为70 a,平均暴露时间(AT)为25 550 d^[28].

1.7 人体健康风险评估概率分布法

人体健康风险评估中环境暴露参数和人群暴露参数存在不确定性,包括个体的体重、摄入量和实验样品采集等参数在暴露人群中随机变化.为了定量考察不确定性,进行了10 000次模拟抽样,获得饮水和水产品食用的健康风险概率分布.通过Monte Carlo模拟对体重、摄入量、水体及水产品中的暴露浓度等参数的不确定性进行敏感性分析,评估不同参数对健康风险值的影响,确定各参数在健康风险评估中的贡献率. Monte Carlo模拟和敏感性分析均使用Crystal Ball软件完成.

2 结果与讨论

2.1 鄱阳湖流域苯酚污染分布特征

2.1.1 水体中苯酚浓度分布

鄱阳湖区域水体苯酚浓度在ND~556.26 $\text{ng}\cdot\text{L}^{-1}$ 范围内,平均浓度为90.18 $\text{ng}\cdot\text{L}^{-1}$,浓度中位数为19.30 $\text{ng}\cdot\text{L}^{-1}$.鄱阳湖区域所有点位中S8点位的苯酚浓度最高,其次为S2点位,浓度达到422.45 $\text{ng}\cdot\text{L}^{-1}$,S23点位苯酚浓度最低.点位S8所在区域和S1~S7所在区域苯酚浓度较高,S1~S7区域苯酚浓度在67.68~422.45 $\text{ng}\cdot\text{L}^{-1}$ 范围内,基本高于整体浓度均值.由于鄱阳湖是一个季节性和吞吐性湖泊,上承赣江、抚河、信江、饶河和修河五河之水,下接长江^[29],S8点位处于鄱阳湖和长江交汇处,丰水期流速随水位的上升而减小,湖口发生倒灌,长江苯酚浓度偏高^[1]可能是S8点位浓度较高的原因.S1~S7区域汇集赣江、信江和抚河等河流,多条河流中工业废水持续输入在湖区汇聚,可能是导致该区域苯酚浓度偏高的主要原因^[30].

表1为近十年全国不同流域苯酚污染水平,浓度范围在ND~15 500.00 $\text{ng}\cdot\text{L}^{-1}$.大沽和北塘排污河中苯酚浓度最高,是鄱阳湖水体苯酚浓度的近30倍.与其他流域相比,鄱阳湖苯酚污染处于中等水平,与同属长江流域的洞庭湖、黄浦江和苏州河浓度相当.水体中的苯酚污染主要是生活污水和工业废水排放的综合结果,淀山湖周围工厂较少,因此其水环境中苯酚浓度较低,与工业废水排放和人为活动有关.

2.1.2 水产品中苯酚含量

鄱阳湖9种水产品中苯酚含量范围在11.98~255.51 $\mu\text{g}\cdot\text{kg}^{-1}$,均值为94.38 $\mu\text{g}\cdot\text{kg}^{-1}$.图2为鄱

表 1 不同流域苯酚浓度 ¹⁾			
Table 1 Phenol concentrations in different basins			
地区	采样年份	浓度/ng·L ⁻¹	文献
鄱阳湖	2019	ND ~ 556.26	本研究
洞庭湖	2013	ND ~ 859.18	[31]
渭河	2016	ND ~ 3 840.00	[32]
饮马河	2016	56.73 ~ 137.35	[33]
黄埔江	2016	ND ~ 815.00	[34]
苏州河	2016	7.00 ~ 602.00	[34]
淀山湖	2016	ND ~ 91.00	[34]
大沽排污河	2018	ND ~ 15 500.00	[35]
北塘排污河	2018	ND ~ 10 300.00	[35]

1) ND 表示未检出

阳湖 9 种水产品中苯酚含量. 苯酚在水产品中平均含量从高到低为: 鲢鱼 ($187.20\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 黄颡鱼 ($184.56\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 鲤鱼 ($72.56\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 白鲢鱼 ($56.69\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 草鱼 ($51.21\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 鳊鱼 ($49.51\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 鳙鱼 ($44.77\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 乌鳢 ($41.79\text{ }\mu\text{g}\cdot\text{kg}^{-1}$) > 白鲈鱼 ($33.88\text{ }\mu\text{g}\cdot\text{kg}^{-1}$). 黄颡鱼和鲢鱼肌肉组织的苯酚含量远高于其他 7 种水产品, 可能是由于黄颡鱼和鲢鱼同属底栖鱼类, 喜于生活在静水和缓流的浅滩, 容易同时富集来自沉积物中的污染物. 总体上来说, 3 个营养级之间无明显放大现象.

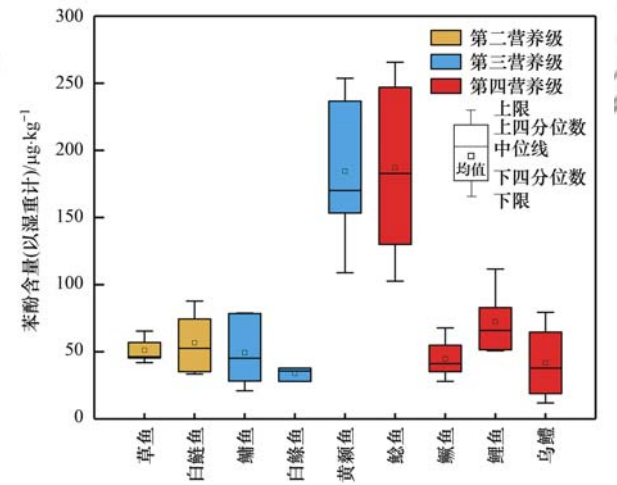


图 2 鄱阳湖水产品中苯酚含量

Fig. 2 Phenol concentrations in aquatic products from Poyang Lake

2.2 商值法健康风险评估

2.2.1 鄱阳湖湖区人群饮水风险

人群环境暴露行为特点是决定环境健康风险评价准确性的关键因子. 在环境介质中对污染物浓度准确定量的情况下, 暴露参数值的选取越接近评价目标人群的实际暴露状况, 则暴露剂量的评价结果越准确, 环境健康风险评价的结果也就越准确. 不同区域人群暴露参数存在差异, 本研究根据鄱阳湖流域人群暴露参数对苯酚的健康风险进行了评估^[36],

鄱阳湖流域成人人体质量为 55.90 kg , 饮水量为 $5.10\text{ L}\cdot\text{d}^{-1}$, 水产品摄入量为 $26.27\text{ g}\cdot\text{d}^{-1}$.

鄱阳湖各样点苯酚的饮水人体健康风险 HQ 值分布如图 3 所示. 鄱阳湖湖区饮水途径的健康风险为 $3.80\times 10^{-7}\sim 8.46\times 10^{-5}$, HQ 最高值与最低值相差 3 个数量级. 所有点位 HQ 值均小于 1, 表明鄱阳湖水体中苯酚的非致癌健康风险处于可接受水平. 鄱阳湖湖区北部和南部的风险值最大, 健康风险自湖区南部和北部向中间逐渐减小. 风险最大的为点位 S8 所处的鄱阳湖湖区北部, 鄱阳湖和长江的交汇处. 其次为湖区南部赣江、信江和抚河等多条河流汇集区域点位 S1 ~ S7. 赣江、信江和抚河等河流沿岸有化工、印染和制药工厂, 工厂废水汇聚导致水体中苯酚暴露浓度较高^[37,38], 这可能是饮水健康风险高于其他点位的原因.

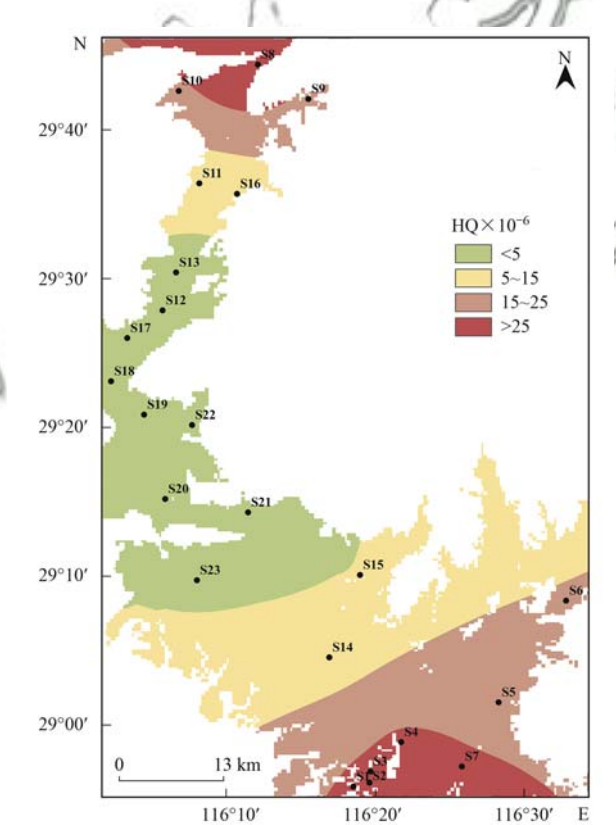


图 3 鄱阳湖不同区域饮水健康风险分布

Fig. 3 Health risk patterns based on drinking water from different areas in Poyang Lake

2.2.2 鄱阳湖不同种类水产品食用风险

根据健康风险评估模型, 计算鄱阳湖不同水产品中苯酚经食用途径产生的健康风险 (图 4). 鄱阳湖 9 种水产品经食用途径产生的健康风险范围为 $2.65\times 10^{-5}\sim 1.47\times 10^{-4}$, 其中鲢鱼导致的水产品食用途径健康风险最高为 1.47×10^{-4} , 其次为黄颡鱼, 健康风险值为 1.45×10^{-4} . 其他 7 种水产品食用途径健康风险范围为 $2.65\times 10^{-5}\sim 5.68\times 10^{-5}$,

相较于这 7 种水产品,鲶鱼和黄颡鱼导致的食用健康风险值高出一个数量级. 第二、三和四营养级之间,由于没有出现生物放大现象,水产品食用风险没有随营养级升高而增大. 因此,鄱阳湖区域人群应减少黄颡鱼和鲶鱼的摄入量,以降低对健康产生的风险.

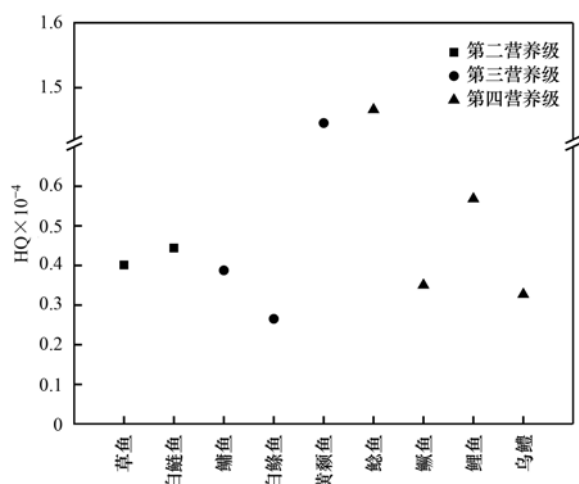


图 4 鄱阳湖不同种类水产品摄入的健康风险

Fig. 4 Health risks based on the consumption of different types of aquatic products

2.3 概率分布法健康风险评估

用 Monte Carlo 模拟做 10 000 次随机抽样,模拟不同环境浓度和暴露参数下,鄱阳湖流域成人饮水和水产品食用造成的健康风险,概率风险分布如图 5 所示. Monte Carlo 模拟结果表明,饮水和水产品食用的健康风险均值分别为 7.20×10^{-6} 和 7.60×10^{-5} ,水产品食用造成的健康风险高出饮水途径的健康一个数量级. 饮水和水产品食用两种暴露途径下 95 百分位点的风险值分别为 2.62×10^{-5} 和 2.11×10^{-4} . 95 百分位点值是合理的最大暴露 (reasonable maximum exposure, RME),若 RME 值超出可接受风险,表明该区域污染严重^[39]. US EPA 推荐的非致癌风险最大可接受水平为 1,鄱阳湖流域成人饮水和水产品食用的 95 百分位点的健康风险值远小于可接受风险,处于可接受水平.

通过敏感性分析(图 6),研究体重、摄入量和苯酚环境暴露浓度等参数变化对健康风险的影响程度. 水产品食用和饮水两种暴露途径下苯酚暴露浓度对健康风险方差的贡献率最大,分别为 73.7% 和 68.5%. 体重对方差的贡献最小且呈负相关性,表明体重较轻的人群面临的健康风险比体重较重的人群更高. 苯酚环境暴露浓度对健康风险值影响更大,为充分保护流域居民人群健康风险,应加强管控苯酚在环境中的暴露水平. 控制环境中苯酚浓度,能更有效地降低人群健康风险.

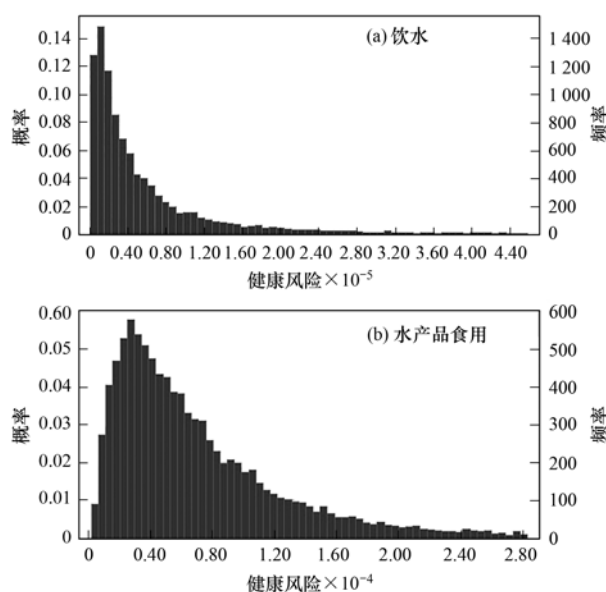


图 5 饮水和水产品食用健康风险概率分布

Fig. 5 Probability distribution of health risks from drinking water and aquatic products

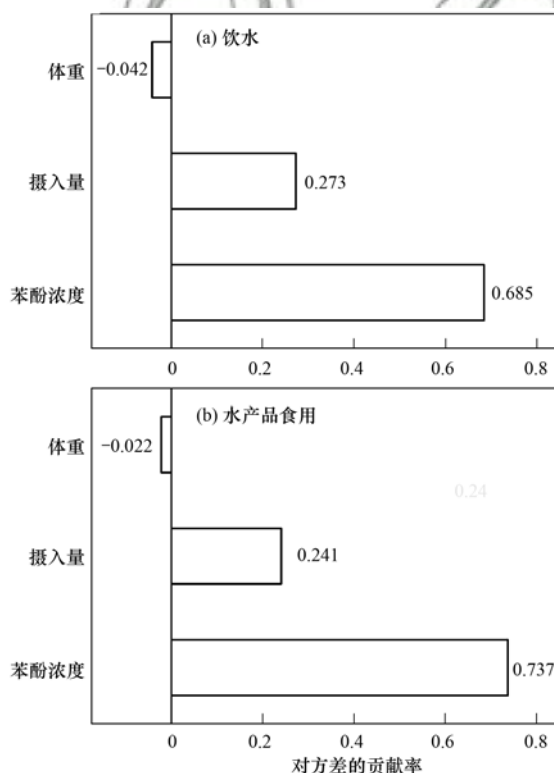


图 6 暴露参数敏感程度比较

Fig. 6 Comparison of exposure parameter sensitivity

3 结论

(1) 鄱阳湖水体中苯酚浓度范围为 ND ~ $556.26 \text{ ng} \cdot \text{L}^{-1}$,与长江流域其他水体浓度相近. 水产品中苯酚含量范围在 $11.98 \sim 255.51 \text{ } \mu\text{g} \cdot \text{kg}^{-1}$,黄颡鱼和鲶鱼苯酚含量较高,是其他种类水产品 2 倍以上,各营养级之间无明显生物放大现象.

(2) 基于鄱阳湖流域成人人群暴露参数,采用

商值法进行人体健康风险评估,饮水途径的健康风险范围为 $3.80 \times 10^{-7} \sim 8.46 \times 10^{-5}$,鄱阳湖湖区南部位点存在相对较高的风险。鄱阳湖 9 种水产品经食用途径产生的健康风险范围为 $2.65 \times 10^{-5} \sim 1.47 \times 10^{-4}$,食用黄颡鱼和鲢鱼会引起较高的健康风险。饮水和水产品摄入途径造成的非致癌风险处于可接受水平,但应加强湖区南部苯酚浓度监控以保障饮水安全。

(3)通过 Monte Carlo 模拟分析鄱阳湖流域人群非致癌健康风险,鄱阳湖流域人群饮水和水产品食用两种暴露途径下 95 百分位的健康风险值分别为 2.62×10^{-5} 和 2.11×10^{-4} ,苯酚环境暴露浓度对健康风险值的影响最大。鄱阳湖流域水体和水产品中苯酚对鄱阳湖流域人群造成的健康风险处于可接受水平。

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