

DOI: 10.7524/AJE.1673-5897.20220116001

孙伟莲, 孙东雷, 黄维, 等. 洗护用品中的抗菌成分对水生态环境的危害作用[J]. 生态毒理学报, 2022, 17(6): 135-143

Sun W L, Sun D L, Huang W, et al. Adverse effects of antibacterial ingredients in sanitary products on aquatic ecological environment [J]. Asian Journal of Ecotoxicology, 2022, 17(6): 135-143 (in Chinese)

洗护用品中的抗菌成分对水生态环境的危害作用

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收稿日期: 2022-01-16 录用日期: 2022-06-08

摘要: 随着抗菌药物在医疗、工农业和日常生活中的广泛应用, 水体抗生素污染已经成为全球公共卫生问题。新型冠状病毒肺炎疫情的暴发促使人们对抗菌卫生用品的使用量进一步增加, 大量杀菌剂随生活废水排入水体, 导致水环境中各种抗菌物质浓度急剧升高, 进一步加重了水体的净化负担。洗护用品中的抗菌成分大多具有广谱杀菌作用, 排入水体后势必对水中的常驻微生物产生危害, 增加水中微生物耐药性, 同时也给水生生物带来威胁。本文系统总结了洗护用品中最常添加的酚类、含氯类和季铵盐类等杀菌剂在国内外水环境中的浓度分布和对微生物、水生生物的毒性作用, 以期为我国科学倡导及管理洗护用品中抗菌成分的添加和使用提供科学依据。

关键词: 抗菌成分; 微生物耐药性; 水生生物毒性; 环境分布; 洗护用品

文章编号: 1673-5897(2022)6-135-09 中图分类号: X171.5 文献标识码: A

Adverse Effects of Antibacterial Ingredients in Sanitary Products on Aquatic Ecological Environment

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Received 16 January 2022 accepted 8 June 2022

Abstract: With the wide application of antibiotics in medical, industrial, agricultural and daily life, antibiotic pollution in water has become a global public health problem. To make matters worse, the COVID-19 pandemic has promoted a further increase in the use of antibacterial hygiene products. As a result, a large number of wastewater containing antibacterial ingredients were discharged, resulting in the increase of the concentration of various antibacterial ingredients in the water environment, and then the burden of water purification increased. Most of the antibacterial ingredients in sanitary products have a broad-spectrum bactericidal effect. When these antibacterial components are discharged into water with wastewater, they are bound to have adverse effects on resident microorganisms in water, increase the drug resistance and threaten the composition of aquatic organisms. In order to provide scientific basis for advocating and managing the addition and use of antibacterial ingredients in sanitary products in China, this review systematically summarized the concentration distribution and adverse effects on microorganisms and aquatic organisms of phenolics, chlorine-containing compounds, quaternary ammonium salts and other fungi-

基金项目: 国家自然科学基金面上项目(82073510)

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cides commonly added in sanitary products in domestic and foreign water environment.

Keywords: antibacterial ingredient; microbial resistance; aquatic toxicity; environmental distribution; sanitary products

近年来,流行性感、新型冠状病毒肺炎和禽流感等突发急性传染病事件频发,促使人们卫生意识大幅增强,推动了消毒杀菌产品的销售和使用。我国国家统计局公布的数据显示,2020 年前三季度我国人均购买洗涤及卫生产品的支出同比增长 13.7%,充分说明新型冠状病毒肺炎疫情极大地刺激了人们对抗菌卫生用品的需求。为了迎合市场和增加销量,生产厂商热衷于在洗护用品中添加杀菌成分,将市售的个人护理产品醒目标注“抗菌”“抑菌”“杀菌”等字样,甚至宣称抗菌产品杀菌率高达 99.999%。但是,由于洗手时间短,抗菌香皂中的杀菌成分在实际应用条件下无法发挥理论的抗菌作用^[1]。另一方面,洗护用品中的抗菌成分随着废水进入水环境,必定给水体微生物和水生生物带来潜在危害^[2]。基于以上 2 点,美国食品药品监督管理局于 2016 年明令禁止销售含有三氯生、三氯卡班和苯酚等 19 种特定活性成分的抗菌洗浴产品^[3],欧盟委员会、东南亚国家联盟和韩国食品药品监督管理局等也相继对抗菌物质的添加做出了更为严格的限制^[4],但中国迄今尚未出台类似政策。事实上,现有研究显示中国此类水环境污染问题逐渐突出,大多数湖泊和河流均能检测到纳克至微克级水平的杀菌剂^[5]。本文检索了国内外洗护用品抗菌成分的最新文献,系统综述了洗护用品中常用的酚类、含氯类、季铵盐类及其他类别杀菌剂在水环境中的分布以及对水体中微生物和水生生物的生物作用,为我国洗护用品中抗菌成分的监督管理提供参考依据。

1 抗菌成分的水环境分布 (Distribution of antibacterial ingredients in water environment)

现有研究表明洗护用品中杀菌成分的排放对微生物耐药性具有重要贡献^[6-7]。随着抗菌洗护用品的广泛应用,酚类、含氯类、季铵盐类、胍类、含氧类和天然杀菌剂等成分被排入水体且在水环境中积累,研究显示这些杀菌剂在国内外地表水、自来水和污水等多种水环境中分布广泛,检出浓度从纳克至微克水平(表 1)。酚类、含氯类和季铵盐类杀菌剂是香皂、洗手液和漱口水等多种洗护用品中最常添加的抗菌成分^[8],受到生产企业的认可,同时添加了这 3 类抗菌成分的产品也受到消费者的青睐。现有的资料提示中国地表水中酚类、含氯类和季铵盐类杀

菌剂浓度普遍高于美国、英国和日本等国家(表 1),表明我国地表水洗护用品中杀菌剂带来的污染不容乐观。此外,目前尚无苯扎溴铵、聚六亚甲基双胍盐酸盐和壳聚糖等多种杀菌剂在我国水环境中浓度分布数据报道,提示这些水环境质量指标的监测亟待加强。

2 抗菌成分对水生态环境的危害作用 (Hazards of antibacterial ingredients on water ecological environment)

2.1 微生物耐药性

众所周知,抗生素滥用会导致环境微生物产生抗生素抗性基因,引起微生物耐药性,给人类健康及环境稳态带来潜在隐患,因此,减缓抗生素耐药性已成为全球重要的公共卫生问题^[22-25]。医疗卫生领域抗生素滥用是引起抗生素耐药的主要原因^[26],针对这一原因,全国已形成规范和共识,严格控制临床抗生素应用。2012 年,我国原卫生部发布的《抗菌药物临床应用管理办法》要求严格控制医疗机构采购抗菌药物的品种与数量,且严格把握使用抗菌药物的指征。然而,遗憾的是,临床用药以外用量很大的洗护用品中抗菌成分应用却被严重忽视。事实上,水环境是耐药菌及耐药基因的主要储存库^[27],日常生活中广泛使用的杀菌成分排放至水体会加剧水环境微生物耐药性。已有研究显示病原微生物暴露于三氯生或三氯卡班后对四环素、环丙沙星和氯霉素的抗性增强,暴露于苯扎氯铵后会增加对氨基青霉素、氯霉素和链霉素等常用抗生素的耐药性^[28-30]。更为重要的是,三氯生、三氯卡班和苯扎氯铵等还可增加病原微生物对多种抗生素产生交叉耐药性^[29-31],表明洗护用品中的杀菌成分随废水排入水体会促进微生物耐药性的发生。此外,酚类、含氯类和季铵盐类等杀菌剂均具有广谱高效杀菌作用,排放至水环境同样会对非目标微生物产生危害,破坏水中微生物正常菌群组成,干扰水的净化和水质调节,同时促使耐药菌株形成,甚至加速超级细菌的产生与传播^[30,32]。更为严重的是,耐药细菌和耐药基因在环境中增殖迁移,造成水源污染且难以被我国现有的自来水处理系统清除^[33];耐药细菌与耐药基因还会在鱼类、虾类和贝类等水产品中富集且可随食物链传播^[34],给人体肠道菌群和免疫功能带来健康威胁。

表 1 洗护用品中抗菌成分的种类、用途和水环境分布

Table 1 Types, uses and distribution in water environment of antibacterial ingredients in washing products

分类 Classification	成分名称 Ingredient name	用途 Application	水环境分布 Distribution in the aquatic environment
酚类杀菌剂 Phenolic bactericides	对氯间二甲苯酚 4-chloro-3,5-dimethylphenol	肥皂、洗手液、消毒剂、香波等 Soaps, hand sanitizers, disinfectants, shampoos, etc.	美国南加州旱季废水 Southern California dry season wastewater, USA; $< 5.59 \mu\text{g}\cdot\text{L}^{-1}$ [9]
			英国塔夫河 River Taff, UK; $< 30 \sim 225 \mu\text{g}\cdot\text{L}^{-1}$ [10]
			英国伊利河 River Ely, UK; $< 30 \sim 301 \text{ ng}\cdot\text{g}^{-1}$ [10]
			日本德岛的河流 Rivers in Tokushima, Japan; $0 \sim 17.8 \text{ ng}\cdot\text{L}^{-1}$ [11]
			中国珠江水系广州段 Guangzhou section of the Pearl River, China; $1.62 \sim 9.57 \mu\text{g}\cdot\text{L}^{-1}$ [8]
含氯类杀菌剂 Chlorine-containing bactericides	三氯生 Triclosan	牙膏、香皂、洗手液、洗面奶、 漱口水、沐浴露等 Toothpaste, soap, hand sanitizer, facial cleanser, mouthwash, shower gel, etc.	中国广州污水处理厂出水 Export water from Guangzhou sewage treatment Plant, China; $2.45 \mu\text{g}\cdot\text{L}^{-1}$ [8]
			中国广州市区家庭自来水 Domestic tap water in Guangzhou, China; $1.60 \text{ ng}\cdot\text{L}^{-1}$ [8]
			中国鄱阳湖流域水体 Poyang Lake Basin, China; $0 \sim 556.26 \text{ ng}\cdot\text{L}^{-1}$ [12]
			—
			—
含氯类杀菌剂 Chlorine-containing bactericides	三氯卡班 Triclocarban	香皂、洗手液、洗面奶、 漱口水、沐浴露等 Soap, hand sanitizer, facial cleanser, mouthwash, shower gel, etc.	加拿大地表水 Canadian surface water; $< 6 \sim 874 \text{ ng}\cdot\text{L}^{-1}$ [13]
			印度托尔萨河 Torsa River, India; $55 \sim 184 \text{ ng}\cdot\text{L}^{-1}$ [14]
			中国广东石井河 Shijing River, Guangdong, China; $35 \sim 1\,023 \text{ ng}\cdot\text{L}^{-1}$ [15]
			中国深圳河 Shenzhen River, China; $90.2 \sim 478 \text{ ng}\cdot\text{L}^{-1}$ [15]
			中国松花江哈尔滨段 Harbin section of Songhua River, China; $0.63 \sim 5.15 \text{ ng}\cdot\text{L}^{-1}$ [16]
含氯类杀菌剂 Chlorine-containing bactericides	次氯酸钠 Sodium hypochlorite	家用消毒剂 Household disinfectant	印度托尔萨河 Torsa River, India; $41 \sim 77 \text{ ng}\cdot\text{L}^{-1}$ [14]
			中国深圳河 Shenzhen River, China; $68.8 \sim 388 \text{ ng}\cdot\text{L}^{-1}$ [15]
			中国松花江哈尔滨段 Harbin section of Songhua River, China; $0.89 \sim 3.41 \text{ ng}\cdot\text{L}^{-1}$ [16]
			中国某扬州游泳池水 Water in a swimming pool in Yangzhou, China; $25 \sim 10\,000 \text{ ng}\cdot\text{L}^{-1}$ [17]
			—

续表1

分类 Classification	成分名称 Ingredient name	用途 Application	水环境分布 Distribution in the aquatic environment
季铵盐类杀菌剂 Quaternary ammonium bactericides	苯扎氯铵(俗称“洁尔灭”) Benzalkonium chloride	皮肤清洁、医务人员手部清洁、洗涤剂 Skin cleaning, medical staff hand cleaning, detergent	中国松花江哈尔滨段 Harbin section of Songhua River, China;0.4 ~ 2.9 ng·L ⁻¹ [16] 中国污水处理厂污泥中 Chinese sewage treatment plant sludge;< 191 μg·g ⁻¹ [18] 中国珠江口沉积物 China Pearl River Estuary sediments;49.3 ~ 1 050 ng·g ⁻¹ [19]
	苯扎溴铵(俗称“新洁尔灭”) Benzalkonium bromide	消毒剂、防腐剂 Disinfectants, preservatives	—
胍类杀菌剂 Guanidine bactericides	聚六亚甲基双胍盐酸盐 Polyhexamethylenebiguanide hydrochloride	洗手液、清洁剂、隐形眼镜护理液 Hand sanitizer, cleaner, contact lens care solution	—
	聚六亚甲基胍盐酸盐 Poly(hexamethylenehydrazine) hydrochloride	万能清洁剂、游泳池水消毒剂 All-purpose cleaner, swimming pool water disinfectant	—
	醋酸氯己定(俗称“洗必定”) Chlorhexidine acetate	牙膏、漱口水 Toothpaste, mouthwash	—
	过氧化氢 Hydrogen peroxide	伤口消毒、下水道去污、 牙膏配制、口腔清洁 Wound disinfection, sewer decontamination, toothpaste preparation, oral cleaning	美国伊利湖 Lake Erie, USA;1.05 ~ 53.92 μg·L ⁻¹ [20] 日本大阪大和川 Osaka Yamato River, Japan;9.38 ~ 22.75 μg·L ⁻¹ [21] 日本国分川 Kokubun River, Japan;8.02 ~ 99.59 μg·L ⁻¹ [21]
含氧类杀菌剂 Oxygenated bactericides	过氧乙酸 Peroxyacetic acid	家用消毒剂 Household disinfectant	—
天然杀菌剂 Natural bactericides	壳聚糖 Chitosan	洗衣液、洗洁精、洗发护发用品、化妆品 Laundry liquid, dish soap, shampoo and conditioner, cosmetics	—
	植物提取物 Plant extracts	洗衣液、洗洁精 Laundry liquid, detergent	—

注:—为未见报道。
Note: — indicates no report.

目前,人们已逐渐意识到洗护用品中抗菌成分对微生物耐药性的重要作用,最近有学者利用宏基因组学技术发现经常使用抗菌牙膏后的牙刷上取样的菌群与正常环境下取样的菌群相比富含种类更多、抗性更强的抗菌剂耐药基因^[35],说明牙刷上抗菌成分残留促进了耐药基因的进化。抗菌牙膏中的三氯生和过氧化物能够在牙刷头积累^[36],长期使用抗菌牙膏会导致牙刷上杀菌剂浓度逐渐增加,在与口腔接触后使人体一直处于杀菌剂暴露,增加杀菌剂引发细菌耐药性的潜在人体健康风险。据统计,目

前全球每年约有 70 万人死于微生物耐药,如果不及时采取控制措施,到 2050 年每年将会有 1 000 万人因微生物耐药而死亡^[37]。随着越来越多的细菌感染难以控制,杀菌剂引发的微生物耐药性已经成为全球性的健康挑战,是人类面临的重大公共卫生威胁^[38-39]。因此,科学合理地管控洗护用品中杀菌剂的使用有助于减少微生物耐药风险。

2.2 水生生物毒性

2.2.1 对氯间二甲苯酚

对氯间二甲苯酚是应用最广泛的酚类杀菌剂,

常被添加到肥皂、洗手液、消毒剂和香波等产品(表1)。现有研究显示当虹鳟鱼暴露于 $(4.2 \pm 0.9) \mu\text{g} \cdot \text{L}^{-1}$ 对氯间二甲苯酚40 d后,鳃、脾、肝和肾等脏器均出现严重损伤,甚至坏死^[40]。事实上,如表1所示,地表水中对氯间二甲苯酚的实际浓度往往高于 $5.1 \mu\text{g} \cdot \text{L}^{-1}$,大于现有实验研究中使用的有害浓度,说明地表水中对氯间二甲苯酚对鱼类的正常生长发育具有危害作用。此外,藻类生长抑制试验、溞类急性活动抑制试验和鱼类急性毒性实验发现,对氯间二甲苯酚对近头状伪蹄形藻、斜生栅藻和稀有鮎鲫具有中等毒性作用,对大型溞、斑马鱼、日本青鳉、虹鳟鱼和蓝鳃鱼具有高毒作用^[41-42],提示对氯间二甲苯酚对水体中鱼类、溞类和藻类等各类生物均可产生危害。

2.2.2 三氯生和三氯卡班

在含氯类杀菌剂中,三氯生和三氯卡班常常添加到牙膏、抗菌皂、漱口水和沐浴露等洗护用品中(表1),然而欧洲化学品管理局尚未批准三氯生作为杀菌物质使用^[43]。研究显示,三氯生和三氯卡班对水体中的藻类、鱼类和低等动物等多种生物都具有明显毒作用,并且能够产生生物富集效应,对低等动物主要表现为致死作用、内分泌干扰效应、生殖毒性及遗传毒性等^[44-45]。急性毒性实验显示三氯生和三氯卡班对黑斑蛙蝌蚪和非洲爪蛙蝌蚪具有高毒作用,且三氯生对日本青鳉鱼、斑马鱼和黑头软口鲮等鱼类也具有高毒效应^[46];残留在给水系统中的三氯生可抑制轮叶黑藻、小球藻和铜绿微囊藻等藻类的生长,且造成鱼类胚胎孵化延迟、发育异常和生殖功能障碍^[47-50]。当虹鳟鱼暴露于 $(0.48 \pm 0.2) \mu\text{g} \cdot \text{L}^{-1}$ 三氯生40 d后,血液细胞DNA明显受损,肌肉组织松弛,肾、肝、脾和腮等重要脏器出现损伤^[40]。值得注意的是,如表1所示,加拿大地表水和我国广东石井河中三氯生浓度的最大值远高于 $0.68 \mu\text{g} \cdot \text{L}^{-1}$,说明地表水中三氯生对水生生物的危害潜力较大。

2.2.3 苯扎氯铵

苯扎氯铵具有强烈的特殊消毒剂刺激性气味,被大量用于皮肤清洁和消毒。2006年,美国环境保护局报道了苯扎氯铵对水环境和水体中的生物(包括鱼、牡蛎和虾等水生生物)具有不利影响,并且明确表示不建议将苯扎氯铵排放到湖泊、海洋等水环境中^[51]。然而,我国目前没有限制排放废水中苯扎氯铵的含量规定。随着人们日常使用,大量苯扎氯铵排入水环境,对鱼类、溞类和藻类的危害作用不容忽视。研究显示淡水甲壳类动物大型溞和网纹蚤暴

露于苯扎氯铵后,可以引发急性毒性、慢性毒性和遗传毒性等多种毒性效应^[52]。当虹鳟鱼暴露于 $180 \mu\text{g} \cdot \text{L}^{-1}$ 苯扎氯铵28 d后,鱼鳃和肝脏中过氧化氢酶活性降低、乙酰胆碱酯酶活性增加^[53],说明苯扎氯铵会干扰虹鳟鱼体内抗氧化作用和代谢过程。此外,苯扎氯铵对水生食物链中斜生栅藻、大型溞和斑马鱼的急性毒性分别属于高毒、剧毒、中毒^[54-55],提示苯扎氯铵的污染会破坏水生生态系统的稳态。

2.2.4 其他杀菌剂

胍类杀菌剂因结构中含有胍基而得名,主要包括氯己定和聚六亚甲基胍,是消毒剂中的新生代产品,早期被认为副作用小安全性高,被当做“万能消毒剂”。2002年,美国环境保护局将胍类杀菌剂中常用的聚六亚甲基双胍盐酸盐列为低环境风险物质^[56],因此胍类杀菌剂在水环境中的分布及毒性作用极少受到关注,对生态环境和人体健康可能造成的危害也被忽视。然而,2020年媒体广泛报道的韩国民众因使用聚六亚甲基胍盐导致的严重健康危害给人们敲响了随意使用消毒剂的警钟^[57]。为了满足谨防细菌滋生的心理,消费者不恰当地在加湿器中加入聚六亚甲基胍盐酸盐,在1994—2011年间对全国60多万人的肺部造成不可逆的肺纤维化,死亡人数高达上万,被认为是韩国历史上最严重的环境灾难之一^[57]。目前对于胍类杀菌剂危害作用的认知还很少,胍类杀菌剂对水环境生物的危害和对人体健康的危害值得研究。

含氧杀菌剂常用的是过氧化氢,俗称双氧水,分解后的最终产物是水和氧气,对环境无污染,被认为是重要的绿色环保产品和化工原料,因此,欧洲化学品管理局于2017年批准过氧化氢用作消毒剂与杀藻剂^[43]。有报道显示,青鳉鱼暴露于高浓度的过氧化氢后会出现抽动、冲撞和惊厥等症状,并且可以与洋地黄皂苷对海洋青鳉鱼产生协同致毒作用^[58]。

除了传统的抗菌抑菌成分外,天然来源的杀菌剂直接提取自天然食物或植物,不属于化学制品,对生态环境和人体健康更为友好,逐渐受到人们的青睐。天然来源抗菌产品的发展可能更有助于开发毒性低、对环境友好的消毒抗菌产品,但天然来源抗菌产品的抗菌机制和实际抗菌效果仍需进一步探讨^[59-60]。

3 总结和展望(Summary and outlook)

综上所述,洗护用品中的抗菌成分在水环境中分布广泛、种类繁多,不仅可以增加水中微生物的耐

药性、促进耐药基因的传播,也对水生生物(鱼类、藻类和溞类等)的生长发育与活动能力产生抑制作用,影响水体自净和生态环境的稳定性。值得注意的是,已有研究表明洗护用品中的杀菌成分在实际使用过程中抗菌效果存疑^[1](时间太短,不能发挥抗菌作用)。因此,美国明令禁止三氯生、三氯卡班和苯酚等杀菌剂在洗护用品中的添加,使废水处理厂进出口水中的三氯生和三氯卡班浓度下降70%以上^[6]。随后,欧盟和韩国也纷纷对洗护用品中抗菌物质的添加做出了更为严格的限制,而我国尚未对洗护用品中抗菌成分添加的政策进行修订,引起人们对水环境中洗护用品杀菌剂带来的潜在风险担忧。当前,全球淡水资源缺乏,水体污染防治迫在眉睫,本文系统总结了洗护用品中主要杀菌成分的水环境分布与效应,为我国科学管控洗护用品中杀菌剂的添加提供了参考依据。未来,需要加强对各种抗菌物质在水环境中浓度分布的监测和毒性效应研究,洗护用品中杀菌剂添加的有效性与必要性有待进一步明确。

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参考文献(References):

- [1] Kim S A, Moon H, Lee K, et al. Bactericidal effects of triclosan in soap both *in vitro* and *in vivo* [J]. The Journal of Antimicrobial Chemotherapy, 2015, 70(12): 3345-3352
- [2] Wu C X, Spongberg A L, Witter J D, et al. Uptake of pharmaceutical and personal care products by soybean plants from soils applied with biosolids and irrigated with contaminated water [J]. Environmental Science & Technology, 2010, 44(16): 6157-6161
- [3] Fischer A. FDA issues final rule on safety and effectiveness of antibacterial soaps [EB/OL]. (2016-09-02) [2022-01-16]. <https://www.fda.gov/news-events/press-announcements/fda-issues-final-rule-safety-and-effectiveness-antibacterial-soaps>
- [4] Merchel Piovesan Pereira B, Tagkopoulos I. Benzalkonium chlorides: Uses, regulatory status, and microbial resistance [J]. Applied and Environmental Microbiology, 2019, 85(13): e00377-e00319
- [5] Su C, Cui Y, Liu D, et al. Endocrine disrupting compounds, pharmaceuticals and personal care products in the aquatic environment of China: Which chemicals are the prioritized ones? [J]. The Science of the Total Environment, 2020, 720: 137652
- [6] Larsson D G J, Flach C F. Antibiotic resistance in the environment [J]. Nature Reviews Microbiology, 2021, 19: 1-13
- [7] Dhama K, Patel S K, Kumar R, et al. The role of disinfectants and sanitizers during COVID-19 pandemic: Advantages and deleterious effects on humans and the environment [J]. Environmental Science and Pollution Research International, 2021, 28(26): 34211-34228
- [8] Tan J H, Kuang H X, Wang C C, et al. Human exposure and health risk assessment of an increasingly used antibacterial alternative in personal care products: Chloroxylenol [J]. The Science of the Total Environment, 2021, 786: 147524
- [9] Loraine G A, Pettigrove M E. Seasonal variations in concentrations of pharmaceuticals and personal care products in drinking water and reclaimed wastewater in southern California [J]. Environmental Science & Technology, 2006, 40(3): 687-695
- [10] Kasprzyk-Hordern B, Dinsdale R M, Guwy A J. The occurrence of pharmaceuticals, personal care products, endocrine disruptors and illicit drugs in surface water in South Wales, UK [J]. Water Research, 2008, 42(13): 3498-3518
- [11] Kimura K, Kameda Y, Yamamoto H, et al. Occurrence of preservatives and antimicrobials in Japanese rivers [J]. Chemosphere, 2014, 107: 393-399
- [12] 徐倩云, 艾舜豪, 高祥云, 等. 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[J]. 环境科学, 2021, 42(3): 1354-1360
Xu Q Y, Ai S H, Gao X Y, et al. Human health risk assessment of phenol in Poyang Lake Basin [J]. Environmental Science, 2021, 42(3): 1354-1360 (in Chinese)
- [13] Lalonde B, Garron C, Dove A, et al. Investigation of spatial distributions and temporal trends of triclosan in Canadian surface waters [J]. Archives of Environmental Contamination and Toxicology, 2019, 76(2): 231-245
- [14] Das Sarkar S, Nag S K, Kumari K, et al. Occurrence and safety evaluation of antimicrobial compounds triclosan and triclocarban in water and fishes of the multitrophic niche of River Torsa, India [J]. Archives of Environmental Contamination and Toxicology, 2020, 79(4): 488-499
- [15] Zhao J L, Ying G G, Liu Y S, et al. Occurrence and risks of triclosan and triclocarban in the Pearl River system, South China: From source to the receiving environment [J]. Journal of Hazardous Materials, 2010, 179(1-3): 215-222
- [16] Li W L, Zhang Z F, Sparham C, et al. Validation of sampling techniques and SPE-UPLC/MS/MS for home and personal care chemicals in the Songhua Catchment,

- Northeast China [J]. *The Science of the Total Environment*, 2020, 707: 136038
- [17] 吴飞, 王冰, 陈晨. 高效液相色谱法测定游泳池水中三氯卡班、三氯生和甲基三氯生的含量[J]. *理化检验-化学分册*, 2020, 56(4): 419-422
- Wu F, Wang B, Chen C. HPLC determination of triclocarban, triclosan and methyltriclosan in swimming pool water [J]. *Physical Testing and Chemical Analysis (Part B: Chemical Analysis)*, 2020, 56(4): 419-422(in Chinese)
- [18] Ruan T, Song S J, Wang T, et al. Identification and composition of emerging quaternary ammonium compounds in municipal sewage sludge in China [J]. *Environmental Science & Technology*, 2014, 48(8): 4289-4297
- [19] Li X L, Luo X J, Mai B X, et al. Occurrence of quaternary ammonium compounds (QACs) and their application as a tracer for sewage derived pollution in urban estuarine sediments [J]. *Environmental Pollution*, 2014, 185: 127-133
- [20] Cory R M, Davis T W, Dick G J, et al. Seasonal dynamics in dissolved organic matter, hydrogen peroxide, and cyanobacterial blooms in Lake Erie [J]. *Frontiers in Marine Science*, 2016, 3: 54
- [21] Sunday M O, Jadoon W A, Ayeni T T, et al. Heterogeneity and potential aquatic toxicity of hydrogen peroxide concentrations in selected rivers across Japan [J]. *The Science of the Total Environment*, 2020, 733: 139349
- [22] von Wintersdorff C J, Penders J, van Niekerk J M, et al. Dissemination of antimicrobial resistance in microbial ecosystems through horizontal gene transfer [J]. *Frontiers in Microbiology*, 2016, 7: 173
- [23] Jian Z H, Zeng L, Xu T J, et al. Antibiotic resistance genes in bacteria: Occurrence, spread, and control [J]. *Journal of Basic Microbiology*, 2021, 61(12): 1049-1070
- [24] Septimus E J. Antimicrobial resistance: An antimicrobial/diagnostic stewardship and infection prevention approach [J]. *The Medical Clinics of North America*, 2018, 102(5): 819-829
- [25] Alanis A J. Resistance to antibiotics: Are we in the post-antibiotic era? [J]. *Archives of Medical Research*, 2005, 36(6): 697-705
- [26] Bojarski B, Kot B, Witeska M. Antibacterials in aquatic environment and their toxicity to fish [J]. *Pharmaceuticals*, 2020, 13(8): 189
- [27] 王丽梅, 罗义, 毛大庆, 等. 抗生素抗性基因在环境中的传播扩散及抗性研究方法[J]. *应用生态学报*, 2010, 21(4): 1063-1069
- Wang L M, Luo Y, Mao D Q, et al. Transport of antibiotic resistance genes in environment and detection methods of antibiotic resistance [J]. *Chinese Journal of Applied Ecology*, 2010, 21(4): 1063-1069 (in Chinese)
- [28] Xiang Y, Wu H, Li L, et al. A review of distribution and risk of pharmaceuticals and personal care products in the aquatic environment in China [J]. *Ecotoxicology and Environmental Safety*, 2021, 213: 112044
- [29] Carey D E. Altered antibiotic tolerance in anaerobic digesters acclimated to triclosan or triclocarban [J]. *Chemosphere*, 2016, 163: 22-26
- [30] Harrison K R, Kappell A D, McNamara P J. Benzalkonium chloride alters phenotypic and genotypic antibiotic resistance profiles in a source water used for drinking water treatment [J]. *Environmental Pollution*, 2020, 257: 113472
- [31] Christensen E G, Gram L, Kastbjerg V G. Sublethal triclosan exposure decreases susceptibility to gentamicin and other aminoglycosides in *Listeria monocytogenes* [J]. *Antimicrobial Agents and Chemotherapy*, 2011, 55(9): 4064-4071
- [32] Lu J, Guo J. Disinfection spreads antimicrobial resistance [J]. *Science*, 2021, 371(6528): 474
- [33] Xu L K, Ouyang W Y, Qian Y Y, et al. High-throughput profiling of antibiotic resistance genes in drinking water treatment plants and distribution systems [J]. *Environmental Pollution*, 2016, 213: 119-126
- [34] 曹弘扬, 汪庆, 赵佳丽, 等. 食源性抗生素耐药菌的污染现状、传播扩散及健康风险研究进展[J]. *中国抗生素杂志*, 2022, 47(10): 1002-1012
- Cao H Y, Wang Q, Zhao J L, et al. Progress on the contamination status, dissemination, and health risks of foodborne antibiotic resistant bacteria [J]. *Chinese Journal of Antibiotics*, 2022, 47(10): 1002-1012 (in Chinese)
- [35] Blaustein R A, Michelitsch L M, Glawe A J, et al. Toothbrush microbiomes feature a meeting ground for human oral and environmental microbiota [J]. *Microbiome*, 2021, 9(1): 32
- [36] Han J, Qiu W, Campbell E C, et al. Nylon bristles and elastomers retain centigram levels of triclosan and other chemicals from toothpastes: Accumulation and uncontrolled release [J]. *Environmental Science & Technology*, 2017, 51(21): 12264-12273
- [37] Stacy A, Andrade-Oliveira V, McCulloch J A, et al. Infection trains the host for microbiota-enhanced resistance to pathogens [J]. *Cell*, 2021, 184(3): 615-627
- [38] Huemer M, Mairpady Shambat S, Brugger S D, et al. Antibiotic resistance and persistence-Implications for human health and treatment perspectives [J]. *EMBO Reports*, 2020, 21(12): e51034
- [39] Nadeem S F, Gohar U F, Tahir S F, et al. Antimicrobial

- resistance: More than 70 years of war between humans and bacteria [J]. *Critical Reviews in Microbiology*, 2020, 46(5): 578-599
- [40] Capkin E, Ozcelep T, Kayis S, et al. Antimicrobial agents, triclosan, chloroxylenol, methylisothiazolinone and borax, used in cleaning had genotoxic and histopathologic effects on rainbow trout [J]. *Chemosphere*, 2017, 182: 720-729
- [41] 朱英. 典型家用消毒剂对稀有鮡鲫毒性及酶活性影响[J]. *环境科学与技术*, 2019, 42(7): 51-56
Zhu Y. Toxicity and enzyme activity effects of typical family-use disinfectants on rare minnow [J]. *Environmental Science & Technology*, 2019, 42(7): 51-56 (in Chinese)
- [42] 朱英, 胡双庆, 沈根祥, 等. 2种典型家用消毒剂对水生生物的急性毒性影响及生态效应阈值研究[J]. *生态毒理学报*, 2018, 13(4): 161-169
Zhu Y, Hu S Q, Shen G X, et al. The study of acute toxicity effects on aquatic organisms and acute ecological threshold (PNECs) of two typical household disinfectants [J]. *Asian Journal of Ecotoxicology*, 2018, 13(4): 161-169 (in Chinese)
- [43] European Chemicals Agency. Information on biocides [EB/OL]. (2022-05-25) [2022-05-30]. <https://echa.europa.eu/en/information-on-chemicals/biocidal-products>
- [44] Brausch J M, Rand G M. A review of personal care products in the aquatic environment: Environmental concentrations and toxicity [J]. *Chemosphere*, 2011, 82(11): 1518-1532
- [45] 陈敏. 三氯生与三氯卡班的生态毒性研究进展[J]. *广东化工*, 2021, 48(1): 62-63
Chen M. Progress in the study of ecotoxicity of triclosan and triclocarban [J]. *Guangdong Chemical Industry*, 2021, 48(1): 62-63 (in Chinese)
- [46] 高昆, 陈娟, 李圆圆, 等. 三氯生和三氯卡班对两栖动物蝌蚪的急性毒性[J]. *生态毒理学报*, 2016, 11(4): 226-231
Gao K, Chen J, Li Y Y, et al. Acute toxicity of triclosan and triclocarban to amphibian tadpoles [J]. *Asian Journal of Ecotoxicology*, 2016, 11(4): 226-231 (in Chinese)
- [47] Ciniglia C, Cascone C, Giudice R L, et al. Application of methods for assessing the geno- and cytotoxicity of triclosan to *C. ehrenbergii* [J]. *Journal of Hazardous Materials*, 2005, 122(3): 227-232
- [48] 张立娜, 王芷茵, 安婧, 等. 水环境中三氯生残留对轮叶黑藻的生态毒性效应[J]. *生态学杂志*, 2019, 38(12): 3754-3761
Zhang L N, Wang Z Y, An J, et al. Ecotoxicological effects of triclosan on *Hydrilla verticillata* in water [J]. *Chinese Journal of Ecology*, 2019, 38(12): 3754-3761 (in Chinese)
- [49] Oliveira R, Domingues I, Grisolia C K, et al. Effects of triclosan on zebrafish early-life stages and adults [J]. *Environmental Science and Pollution Research*, 2009, 16(6): 679-688
- [50] 张立娜, 王芷茵, 安婧. 底泥中三氯生残留对轮叶黑藻的生态毒性效应[J]. *农业环境科学学报*, 2019, 38(9): 2057-2065
Zhang L N, Wang Z Y, An J. Ecotoxicological effects of triclosan on *Hydrilla verticillata* in sediments [J]. *Journal of Agro-Environment Science*, 2019, 38(9): 2057-2065 (in Chinese)
- [51] United States Environmental Protection Agency (US EPA). Reregistration eligibility decision for alkyl dimethyl benzyl ammonium chloride (ADBAC) [R]. Washington DC: US EPA, 2006
- [52] Lavorgna M, Russo C, D'Abrosca B, et al. Toxicity and genotoxicity of the quaternary ammonium compound benzalkonium chloride (BAC) using *Daphnia magna* and *Ceriodaphnia dubia* as model systems [J]. *Environmental Pollution*, 2016, 210: 34-39
- [53] Antunes S C, Nunes B, Rodrigues S, et al. Effects of chronic exposure to benzalkonium chloride in *Oncorhynchus mykiss*: Cholinergic neurotoxicity, oxidative stress, peroxidative damage and genotoxicity [J]. *Environmental Toxicology and Pharmacology*, 2016, 45: 115-122
- [54] 李祥英, 杨法辉, 李秀环, 等. 两种季铵盐阳离子表面活性剂对水生生物的毒性效应[J]. *农业环境科学学报*, 2012, 31(4): 673-678
Li X Y, Yang F H, Li X H, et al. Toxicity of two quaternary ammonium cationic surfactants to aquatic organisms [J]. *Journal of Agro-Environment Science*, 2012, 31(4): 673-678 (in Chinese)
- [55] Chen Y, Geurts M, Sjollem S, et al. Acute toxicity of the cationic surfactant C12-benzalkonium in different bioassays: How test design affects bioavailability and effect concentrations [J]. *Environmental Toxicology and Chemistry*, 2014, 33(3): 606-615
- [56] de Oliveira Melo A, Santos D B D, Silva L D, et al. Molluscicidal activity of polyhexamethylene biguanide hydrochloride on the early-life stages and adults of the *Biomphalaria glabrata* (Say, 1818) [J]. *Chemosphere*, 2019, 216: 365-371
- [57] Yoon J, Kang M S, Jung J, et al. Humidifier disinfectant consumption and humidifier disinfectant-associated lung injury in South Korea: A nationwide population-based study [J]. *International Journal of Environmental Research and Public Health*, 2021, 18(11): 6136

- [58] 高东辉, 李昌伟, 吴霓, 等. 过氧化氢、洋地黄皂苷对海洋青鳉鱼的致毒作用[J]. 海洋环境科学, 2017, 36(6): 801-805, 819
- Gao D H, Li C W, Wu N, et al. The toxicity of hydrogen peroxide and digitonin on *Oryzias melastigma* [J]. Marine Environmental Science, 2017, 36 (6): 801-805, 819 (in Chinese)
- [59] 刘治平, 杨华明. 植物源性消毒与抗菌制剂发展现状[J]. 中国消毒学杂志, 2019, 36(4): 302-303
- [60] 刘光荣, 翟春涛, 金敏蓉, 等. 天然抗菌剂对5种致病菌的抑制作用研究[J]. 中国真菌学杂志, 2020, 15(2): 88-92
- Liu G R, Zhai C T, Jin M R, et al. The inhibitory effects of natural antibacterial agent on 5 pathogenic microorganisms [J]. Chinese Journal of Mycology, 2020, 15(2): 88-92 (in Chinese)
- [61] Brose D A, Kumar K, Liao A, et al. A reduction in triclosan and triclocarban in water resource recovery facilities' influent, effluent, and biosolids following the US Food and Drug Administration's 2013 proposed rulemaking on antibacterial products [J]. Water Environment Research, 2019, 91(8): 715-721 ◆