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季铵盐抗菌剂在环境中的迁移转化行为及其毒性效应

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摘要: 季铵盐(QACs)是一类广泛使用的阳离子杀菌剂,流感和新冠肺炎大流行导致其使用量剧增. 在其使用或使用后处理处置过程中,QACs可通过各种途径释放到环境中,在水体、沉积物和土壤等多种介质中频繁检出. QACs有较强的表面活性和非专一性的生物毒性,对生态系统构成潜在威胁. 围绕QACs在环境介质中的迁移转化、生物毒性效应和细菌出现QACs抗性的主要机制等方面,系统梳理了QACs在环境中的迁移转化行为及其潜在的毒性效应. 结果发现好氧生物降解是QACs在环境中的主要衰减途径,降解反应以QACs不同位置C的羟基化来起始,后经过脱羧、脱甲基和β-氧化反应,最终矿化为CO_2和H_2O. 环境浓度的QACs不会对生物产生致死效应,但会显著影响*Daphnia magna*等水生生物生长繁殖,毒性效应主要受自身结构、受试生物种类和暴露时长等因素影响. 探究了QACs对*Microcystis aeruginosa*急性毒性的作用机制,发现QACs主要通过破坏光合系统,导致电子传递受限,构成氧化胁迫,破坏细胞膜来抑制*Microcystis aeruginosa*的生长. QACs在环境中的浓度低于其杀菌浓度,且其生物降解易形成浓度梯度,利于诱导细菌出现QACs抗性. 归纳出细菌对QACs抗性机制主要有改变细胞膜结构和组成、形成生物膜、外排泵基因的过度表达以及通过水平转移获取抗性基因. 由于作用对象和机制的相似性,QACs也会诱导细菌产生抗生素抗性,主要通过协同抗性和交叉抗性来实现. 根据目前的研究现状,提出了未来应重点围绕QACs在实际环境介质中的毒性效应以及对环境微生物抗性的诱导机制展开研究.

关键词: 季铵盐(QACs); 迁移转化; 毒性效应; 抗性机制; 协同抗性

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Migration, Transformation, and Toxicity of Quaternary Ammonium Antimicrobial Agents in the Environment

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Abstract: Quaternary ammonium compounds (QACs) are one type of widely used cationic biocide, and their usage amount is growing rapidly due to the flu and COVID-19 pandemic. Many QACs were released into the environment in or after the course of their use, and thus they were widely detected in water, sediment, soil, and other environmental media. QACs have stronger surface activity and non-specific biotoxicity, which poses a potential threat to the ecosystem. In this study, the environmental fate and potential toxicity of QACs were documented in terms of their migration and transformation process, biological toxicity effects, and the main mechanisms of bacterial resistance to QACs. Aerobic biodegradation was the main natural way of eliminating QACs in the environment, and the reaction was mainly initiated by the hydroxylation of C atoms at different positions of QACs and finally mineralized to CO_2 and H_2O through decarboxylation, demethylation, and β-oxidation reaction. Toxicological studies showed that QACs at environmental concentrations could not pose acute toxicity to the selected biotas but threatened the growth and reproduction of aquatic organisms like *Daphnia magna*. Their toxicity effects depended on their molecular structure, the tested species, and the exposed durations. Additionally, our team first investigated the toxicity effects and mechanisms of QACs toward *Microcystis aeruginosa*, which showed that QACs depressed the algae growth through the denaturation of photosynthetic organelles, suppression of electron transport, and then induction of cell membrane damage. In the environment, the concentrations of QACs were always lower than their bactericidal concentrations, and their degradation could induce the formation of a concentration gradient, which facilitated microbes resistant to QACs. The known resistance mechanisms of bacteria to QACs mainly included the change in cell membrane structure and composition, formation of biofilm, overexpression of the efflux pump gene, and acquisition of resistance genes. Due to the similar targets and mechanisms, QACs could also induce the occurrence of antibiotic resistance, mainly through co-resistance and cross-resistance. Based on the existing data, future research should emphasize the toxicity effect and the potential QACs resistance mechanism of microorganisms in real environmental conditions.

Key words: quaternary ammonium compounds (QACs); migration and transformation; toxic effects; mechanism of resistance; co-resistance

季铵盐(quaternary ammonium compounds, QACs)是一类由一个带正电的中心N^+和其连接的有机基团以及带负电的卤素原子构成的阳离子杀菌剂,主要通过烷基链改变细胞膜的磷脂双分子层,破坏细胞膜,使细胞膜内的物质流失导致细菌死亡[1,2]. 根据其化学结构式,QACs主要分为三大类(如图1): ①直链烷基铵类、②咪唑类和③吡啶类[3]. 一般地,氯化QACs杀菌效果强于其他QACs[4],含苯环QACs强于不含苯环类[5]. 其中,直链烷基铵类的烷基三甲基铵盐(alkyltrimethylammonium compounds, ATMACs)、二烷基二甲基铵盐(dialkyldimethylammonium

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compounds, DADMACs) 和苄基烷基二甲基铵盐 (benzylammonium compounds, BACs) 是最常用的 QACs 类杀菌剂.

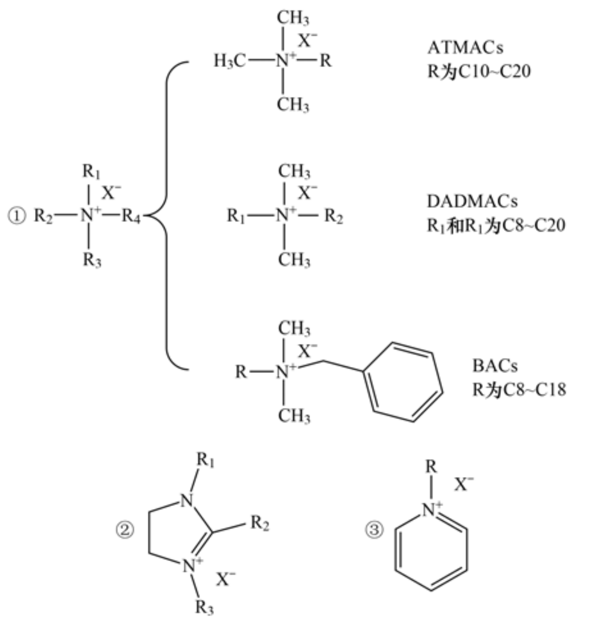

图 1　典型 QACs 结构式

Fig. 1　Structural formulas of typical QACs

由于其良好的杀菌性能和双亲性, QACs 被广泛应用在工业、农业和医护行业[6~8]. 美国环保署在 2015 年公布的报告中指出 ATMACs、DADMACs 和 BACs 在美国 5 年内总产量约4 536 ~ 22 680 t (1 000万 ~ 5 000万磅)[9]. 近年来, 流感和新冠肺炎大流行, 更高的消毒要求导致了更多杀菌剂生产和使用[10,11]. 有数据表明, 用于对抗新冠病毒的杀菌剂中一半以上是以 QACs 作为活性成分[12]. QACs 在使用过程中会有意或无意释放到环境中, 据测算, 约 75% 的 QACs 被排放到污水处理系统, 其余则直接释放到环境中[13]. 由于传统污水处理技术的局限性, QACs 在污水处理厂不能被完全去除, 如澳大利亚某市政污水处理厂进水中 ATMAC(C16) 浓度范围为 7.7 ~ 27 $\mu g \cdot L^{-1}$, 出水浓度约为 1.1 $\mu g \cdot L^{-1}$[14]. 同时, QACs 的正电性和疏水性导致其易吸附在活性污泥中, 最终随污水处理厂出水或活性污泥农用释放到环境中. 如墨西哥一处利用污水处理厂出水灌溉的农田土壤中 BAC(C12) 浓度为 81 $\mu g \cdot kg^{-1}$[15], 显著高于其他未灌溉区土壤. 目前 QACs 在世界范围内的水体、沉积物和土壤中均有检出, 在水中浓度范围为几 ~ 几十 $\mu g \cdot L^{-1}$[16~18], 在沉积物和土壤中浓度范围为几十 ~ 几千 $\mu g \cdot kg^{-1}$[15,19~21]. QACs 在环境中的浓度低于其杀菌浓度 (10 ~ 50 $mg \cdot L^{-1}$)[22], 且极易吸附在微生物体、土壤和沉积物等介质表面, 生物可利用性降低, 导致 QACs 在环境中的赋存时间延长. 长期低剂量的 QACs 暴露易诱导微生物对其产生抗性, 导致 QACs 杀菌失效. 据统计, 已有 16 起流行疾病的暴发是由于微生物出现抗性导致 QACs 杀菌剂失效引起的[23].

由于大量生产和使用, QACs 广泛存在于多种环境介质中, 对生态系统和人体健康构成潜在威胁, 了解其环境行为及效应是实现有效管控的基础. 本文结合国内外相关文献, 综述了 QACs 在环境中的迁移转化行为、毒性效应、微生物出现 QACs 抗性及诱导抗生素抗性的主要机制, 以期为充分了解 QACs 的环境风险提供重要参考.

1　QACs 在环境中的主要迁移转化过程

QACs 在环境中的迁移转化方式主要包括吸附和降解, 其在活性污泥、土壤和沉积物等固相介质中的吸附分为可逆吸附和不可逆吸附, 可逆吸附受降解过程影响. QACs 在环境中的降解主要包括光解和生物降解. 目前仅有一篇关于 QACs 在水体中光解的研究, 发现 QACs 能够吸收的太阳光谱有限, 自然光解缓慢, 半衰期为 12 ~ 94 d, 相比非生物降解, 生物降解是 QACs 在环境中的主要消除方式[24]. 在厌氧条件下, QACs 很难被生物降解利用, 而在好氧条件下可以作为碳源被生物降解利用, 半衰期为数小时到十几天[25,26].

1.1　QACs 的主要吸附特征

QACs 具有带正电的头部 (N^+) 和长的疏水性尾部 (碳氢链), 进入环境后容易通过静电作用和疏水作用吸附在微生物、活性污泥、土壤和沉积物等带负电的介质上[27], 目前的研究主要聚焦于 QACs 在活性污泥和土壤中的吸附机制.

活性污泥对 QACs 的吸附主要是通过有机质的疏水作用实现的, 吸附强弱与 QACs 结构有关, 一般 QACs 烷基链越长越易被吸附[28~31]. 有研究发现活性污泥对 4 种 QACs 的吸附能力依次为: ATMAC (C16) > BAC (C16) > BAC (C12) > ATMAC (C12), 吸附亲和力与烷基链长呈正相关; QACs 烷基链较短时, 苄基能增加吸附亲和力[29]. 此外, pH 也是重要的影响因素, 随着 pH 值的升高, 活性污泥对 QACs 的吸附量降低, 主要是由于高 pH 会导致活性污泥中的有机质被大量溶解, 产生大量带负电的酸性基团, 这些酸性基团与 QACs 发生静电作用, 从而抑制了活性污泥对 QACs 的吸附[29,32].

土壤是一个由微生物、黏土矿物、腐殖质及其他固体和可溶性物质组成的复杂有机-无机复合体. 土壤中的有机质、黏土矿物、氧化物和微生物细胞壁均可以作为 QACs 的吸附剂. Sarkar 等[33] 研究了

QACs 在 3 种土壤中的吸附行为,发现土壤矿质黏土成分是影响 QACs 在土壤中吸附强度的主要因素,蒙脱石对 QACs 的吸附强度大于高岭土. 进一步,土壤矿质胶体性质也会对 QACs 吸附产生影响,土壤矿质胶体吸附的阳离子可与带正电的 QACs 进行离子交换,快速达到吸附平衡,与 QACs 浓度无关,但当 QACs 浓度高于土壤阳离子交换容量(cation exchange capacity,CEC)的 80% 时,由于空间位阻效应,土壤吸附 QACs 速率降低[34]. Ilari 等[35]考察了不同离子浓度(0.002、0.02 和 0.1 $mol \cdot L^{-1}$ NaCl)对蒙脱土吸附 BAC(C12)的影响特征,发现最大吸附容量与离子浓度呈正相关. Ndabambi 等[36]发现土壤对不同链长的 BAC(C8 ~ C14)的吸附能力与土壤 CEC 含量和 BAC 烷基碳链长度呈正相关,表明土壤对 BAC 的吸附过程主要受离子交换和疏水作用影响. 土壤的铁铝等氧化物会改变黏土矿物的表面积和净电荷零点,影响土壤团聚体的稳定,改变土壤对 QACs 的吸附,氧化铁 Zata 电位的改变也会加强或抑制吸附,关于土壤中氧化物对 QACs 吸附机制的研究有待深入. 土壤微生物是土壤最活跃的组分之一且带负电荷,也会影响土壤对 QACs 的吸附性,同时,吸附态的 QACs 也会影响其在土壤中的生物有效性[33].

综上,QACs 在环境中的吸附过程主要受 QACs 自身理化性质和环境介质理化性质影响,吸附机制主要涉及离子交换、静电作用及疏水作用等.

1.2 QACs 的生物降解特征

虽然环境介质对 QACs 有较强的吸附性,但在目前报道的机制中,生物降解才是其在环境中自然衰减的主要方式. 有研究模拟了 10 种 QACs 在自然水环境中的生物转化,发现其均能被生物降解,半衰期为 0.5 ~ 1.6 d[37]. 本课题组研究了 BAC(C12)在两种不同类型土壤(碱性耕地土壤和酸性森林土壤)中的好氧生物降解,降解半衰期分别为 4.66 d 和 17.33 d,土壤有机质含量和微生物群落结构对其降解速率存在显著影响[38].

QACs 的好氧生物降解主要由于 *Xanthomonas*、*Aeromonas* 和 *Pseudomonas* 等功能菌的生长利用. Oh 等[26]将 BAC(C12 和 C14)作为好氧间歇式反应器的唯一碳源,发现 12 h 内大部分的 BAC(C12 和 C14)被降解,*Pseudomonas* 丰度显著上升. 目前,研究者在好氧条件下分离出一些有效降解 QACs 的微生物,如 *Pseudomonas fluorescens* TN4、*Pseudomonas putida* A(ATCC 12633)和 *Aeromonas hydrophila* sp. K 等[39~41].

QACs 的好氧生物降解反应主要由不同位置碳的羟基化来起始,目前报道的 QACs 好氧生物降解主要有以下 3 条途径(如图 2):①烷基末端 C 原子的 ω-羟基化起始反应,后经脱乙酸作用和末端羟基氧化形成羧酸-QACs,后经脱羧反应及多次脱甲基

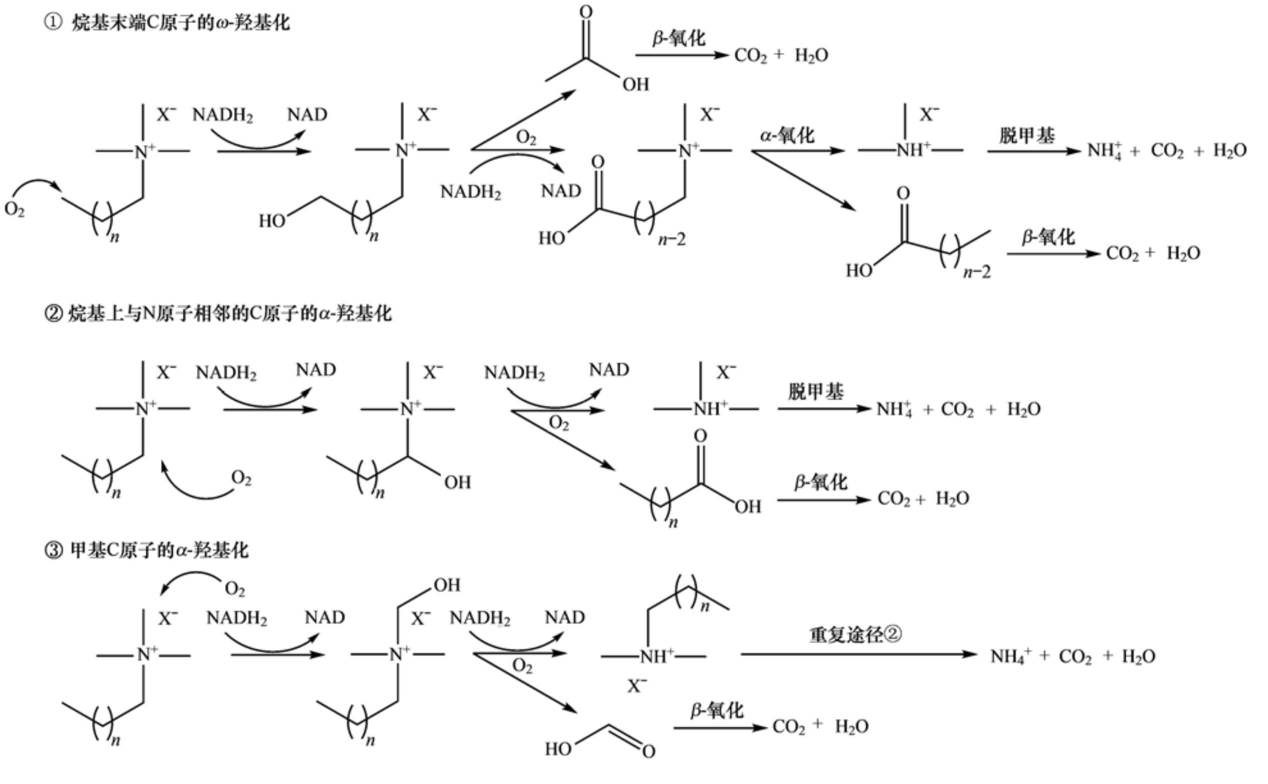

图 2 QACs 的好氧生物降解途径

Fig. 2 Aerobic biodegradation pathway of QACs

反应降解为 NH_4^+、CO_2 和 H_2O,乙酸和羧酸基团则通过β-氧化被降解为 CO_2 和 H_2O;②烷基上与 N 原子相邻的 C 原子的 α-羟基化起始反应,后经脱烷基作用生成叔铵化合物和长链羧酸,叔铵化合物经过多次脱甲基后产生 NH_4^+、CO_2 和 H_2O,长链羧酸则通过β-氧化被降解为 CO_2 和 H_2O;③甲基 C 原子的α-羟基化起始反应,然后脱甲酸,之后按途径②被降解为 CO_2 和 H_2O[13,42,43]. 3 种途径的起始能量负荷是相同的,但途径②产生的中间产物(叔铵化合物)链长更短,比途径①和③的产物毒性小,因此,途径②是主要的生物转化机制[13]. 在降解过程中微生物的单加氧酶、胺氧化酶和环羟基化加氧酶发挥关键作用[26,44,45]. 上述途径均是纯菌株降解利用 QACs 的主要方式,在多种微生物共存、多种电子受体共存的实际环境介质中,QACs 的主要降解途径是否会发生变化,环境中的多种理化因子是否对其产生影响均有待进一步研究.

2 QACs 的生物毒性效应

作为一种非专一性杀菌剂,环境中残留的 QACs 会对鱼类、藻类和微生物等多种生物产生毒性效应,尤其是会诱发细菌出现抗性,导致杀菌失效,有引发传染病流行的潜在风险. 本部分主要对目前关于 QACs 生物毒性的研究进行归纳总结,便于研究者深入理解其可能带来的潜在生态风险.

2.1 急性毒性

QACs 对于土壤微生物的急性毒性浓度往往高达数百$mg\cdot kg^{-1}$. 对土壤微生物硝化作用的 48 h 急性抑制试验发现,BAC 半数效应浓度(EC_{50})值为 221 $mg\cdot kg^{-1}$[46];对土壤中 *Bacillus cereus* 纯菌株生长抑制的 EC_{50}(48 h)为 500 $mg\cdot kg^{-1}$[47].

表 1 总结了目前报道的关于 QACs 对水生生物的急性毒性效应浓度. 环境浓度下 QACs 不会对水生生物构成急性毒性,其急性毒性效应浓度与其自身结构、受试生物类型及受试时间紧密相关. 在所报道的受试生物中, *Daphnia magna* 对 QACs 最敏感[48]. 如 Kreuzinger 等[17]对比了 48 h 内 BAC(C12、C14 和 C16)对几种水生生物生长繁殖影响的 EC_{50}值,发现 BAC(C12、C14 和 C16)对 *Daphnia magna* 的 EC_{50} 值为 0.041 $mg\cdot L^{-1}$,显著低于 *Brachionus calyciflorus* (0.125 $mg\cdot L^{-1}$) 和 *Tetrahymena thermophila*(2.941 $mg\cdot L^{-1}$). 一般地,QACs 的急性毒性强度与其碳链长度成正比,可能是因为长链 QACs 更容易被吸附到生物膜上,破坏细胞膜结构. 如 96 h 内,BAC(C12)、BAC(C14)和 BAC(C16)对 *Chlorella vulgaris* 的生长抑制 EC_{50}值分别为 0.203、0.174 和 0.161 $mg\cdot L^{-1}$[49]. 此外,其毒性效应还受暴露时间影响,Li[50]将 *Dugesia japonica* 暴露在同一浓度 BAC(C12)下,发现半数致死浓度(LC_{50})与暴露时间呈反比.

表 1 QACs 对水生生物的急性毒性

Table 1 Acute toxicity of QACs to aquatic organisms

QACs 类别	受试生物	测试时长/终点	浓度/$mg\cdot L^{-1}$	文献
BAC(C12)	*Dugesia japonica*	24 h/死亡	LC_{50} = 4.02	[50]
	Dugesia japonica	48 h/死亡	LC_{50} = 2.27	[50]
	Dugesia japonica	72 h/死亡	LC_{50} = 0.43	[50]
	Dugesia japonica	96 h/死亡	LC_{50} = 0.21	[50]
	Chlorella vulgaris	96 h/生长抑制	EC_{50} = 0.203	[48]
	Microcystis aeruginosa	96 h/生长抑制	EC_{50} = 3.614	[51]
BAC(C14)	*Chlorella vulgaris*	96 h/生长抑制	EC_{50} = 0.174	[49]
BAC(C16)	*Chlorella vulgaris*	96 h/生长抑制	EC_{50} = 0.161	[49]
BAC(C18)	*Oryzias latipes*	96 h/死亡	LC_{50} = 2.12	[52]
CTAB/ATMAC(C16)	*Rainbow trout*	24 h/死亡	LC_{50} = 0.6	[53]
	Daphnia magna	24 h/活动抑制	EC_{50} = 0.058	[53]
	Chlorella vulgaris	96 h/生长抑制	EC_{50} = 0.156	[49]
CTAC/ATMAC(C16)	*Chlorella vulgaris*	96 h/生长抑制	EC_{50} = 0.137	[49]
	Chlorella vulgaris	96 h/生长抑制	EC_{50} = 0.15	[54]
BAC(C8 ~ C18)	*Ceriodaphnia dubia*	24 h/死亡	LC_{50} = 0.403 7	[55]
	Daphnia magna	48 h/活动抑制	EC_{50} = 0.038 2	[55]
BAC(C12 和 C14)	*Oryzias latipes*	96 h/死亡	LC_{50} = 0.246	[56]
	Daphnia magna	48 h/活动抑制	EC_{50} = 0.04111	[56]
BAC(C12、C14 和 C16)	*Daphnia magna*	48 h/抑制生长繁殖	EC_{50} = 0.041	[17]
	Brachionus calyciflorus	48 h/抑制生长繁殖	EC_{50} = 0.125	[17]
	Tetrahymena thermophila	24 h/抑制生长繁殖	EC_{50} = 2.941	[17]
BEC	*Cyprinus carpi*	96 h/死亡	LC_{50} = 4.57	[57]

为了进一步明确 QACs 对水生生物急性毒性机制，本课题组利用差异分析蛋白组学探究了 BAC(C12)对铜绿微囊藻的急性毒性效应及其机制，发现 96 h 内 BAC(C12)对 *Microcystis aeruginosa* 生长抑制的 EC_{50} 为 3.61 $mg\cdot L^{-1}$[51]. 在此剂量下，藻的光合活性降低了 36%，但内源藻毒素和乳酸脱氢酶的胞外释放量显著增高，藻细胞出现质壁分离、类囊体模糊及类囊体膜堆积等现象. 基于差异分析蛋白组学分析，表明 BAC(C12)主要通过破坏光合系统，导致电子传递受限，构成氧化胁迫，破坏细胞膜来抑制 *Microcystis aeruginosa* 的生长，同时内源微囊藻毒素合成量上升，并通过破损的细胞膜释放到环境中，从而增加了 BAC(C12)对水生生物的危害.

2.2　慢性毒性

低于作用浓度的 QACs 不会直接杀死细菌，但会抑制细菌活性. 当进水 BACs 浓度达到 5 $mg\cdot L^{-1}$ 时，氨氧化细菌的活性受到抑制[58]. Yang 等[59]发现 BAC(C12)会导致湖泊水体微生物的 *amoA* 和 *nifH* 基因的丰度下降，表明 BAC(C12)可能会影响微生物驱动的氮循环过程. 除了影响细菌的氮循环功能外，QACs 还会对微生物构成氧化胁迫，有研究发现将好氧膜生物反应器暴露于十六烷基三甲基溴化铵(CTAB)后，反应器中细菌细胞色素 C 氧化酶含量减少，呼吸电子传递体系功能受阻，胞内活性氧自由基含量上升[60,61]，本课题组的研究也发现了 BAC(C12)会对 *Microcystis aeruginosa* 构成氧化胁迫[51].

QACs 对于 *Daphnia magna*、*Ceriodaphnia dubia* 和 *Rainbow trout* 等水生生物的慢性毒性效应浓度普遍在 $\mu g\cdot L^{-1}$ 级. Lavorgna 等[55]探究了 BAC(C18)对 *Daphnia magna* 和 *Ceriodaphnia dubia* 生殖的影响，发现 21 d 内，0.06 $\mu g\cdot L^{-1}$ 的 BAC(C18)抑制了 *Daphnia magna* 的生殖，7 d 内 3.39 $\mu g\cdot L^{-1}$ 的 BAC(C18)抑制了 *Ceriodaphnia dubia* 的生殖，此两种受试生物常为鱼类等水生生物的食物，在食物链中发挥关键作用，低浓度 QACs 的长期慢性毒性效应需引起重视.

哺乳动物对于 QACs 的敏感性较低，效应浓度普遍在数百 $mg\cdot kg^{-1}$[62]. 有研究发现老鼠摄入 120 $mg\cdot kg^{-1}$ QACs 后，其后代数量减少并且怀孕间隔时间增加[63]. 最近有学者在人体血液样本中检测到了不同链长的 QACs，并发现 QACs 会增加血液中炎症细胞因子水平，抑制线粒体功能，破坏胆固醇稳态，效应与 QACs 浓度显著相关[64].

3　QACs 诱导细菌出现 QACs 抗性的主要机制

低于作用浓度的 QACs 会诱导细菌出现抗性，如长期暴露在 BACs 环境中的 *Staphylococcus aureus* 会对 BACs 产生抗性，最低抑菌浓度(minimum inhibitory concentration, MIC)从 5 $mg\cdot L^{-1}$ 上升到 10 $mg\cdot L^{-1}$[65]. 对单菌株的研究发现，微生物主要通过修饰细胞膜结构、抑制膜孔蛋白对 QACs 的运输、药物外排泵蛋白高表达及捕获水平移动元件等机制获得 QACs 抗性.

细胞膜是 QACs 杀菌的主要靶位点，细菌常通过改变细胞膜的结构和组成，抑制 QACs 的进入，降低对 QACs 的敏感性. 例如，*Pseudomonas aeruginosa* BACs 抗性株细胞膜中的磷脂和脂肪酸的含量显著高于敏感株[66]，在 BACs 诱导下，*Bacillus cereus* 细胞膜中短链脂肪酸含量显著上升[67]；*Pseudomonas aeruginosa* 通过增加亚精胺(一种聚阳离子)的合成来稳定膜电荷，以减少 QACs 的进入[66]. 细菌还可以调控细胞膜运输蛋白的含量来降低对 QACs 的敏感性. 有研究发现膜孔蛋白基因的下调能够降低 *Pseudomonas*[68,69] 和 *Escherichia coli*[70] 对 BACs 的敏感性.

形成生物膜是细菌保护自身免受环境压力的常见抗性机制，可以提高对 QACs 的抗性. 例如，*Staphylococcus epidermidis* CIP53124 形成生物膜后降低了对 BACs 的敏感性[71]. 从乳制品行业中分离出的 BACs 抗性 *Escherichia coli* 形成生物膜的能力强于敏感株，敏感株暴露于 BACs 一段时间后其形成生物膜的能力也显著提升[72]. 此外，多菌种生物膜对 QACs 的抗性强于单菌种生物膜[73,74]. Giaouris 等[74]发现多菌种生物膜的形成能够显著降低 *Pseudomonas putida* 对 BACs 的敏感性.

外排泵蛋白的过度表达也可以使细菌产生 QACs 抗性. 外排泵是包含跨膜区的膜蛋白，其形成的通道能够主动将物质从细胞质或者细胞膜中去除[75~77]. 有研究表明 QACs 抗性往往与 Qac 外排泵有关，Qac 外排泵蛋白家族主要包括 QacA、QacB、QacC、Qac EΔ1、QacG、QacH 和 QacJ，其中 QacA、QacB 是主促进者(major facilitator, MF)家族，而其余的则是小多药抗性(small multidrug resistance, SMR)家族[13,78]. 有研究发现，*Listeria monocytogenes* 通过 QacH 外排泵的高表达，降低了对 BACs 敏感性，而在外排泵抑制剂存在的条件下，恢复了对 BACs 的敏感性[79,80]. 此外，细菌还可以借助抗生素类外排泵实现对 QACs 的外排. 例如，氟喹诺酮类抗生素外排泵 CemABC、NorA 和氨基糖苷类抗生素外排泵 EmrE 可协同将 QACs 外排[80,81]，此类外排泵的过度表达，会使细菌对 QACs 和相应的抗生素的耐受性提升 2~8 倍[82~85].

抗性基因的水平转移是细菌获得 QACs 抗性的另一重要途径,许多 QACs 抗性基因通常位于质粒、整合子和转座子等可移动遗传元件(mobile genetic elements,MGEs)上. 例如,编码 Qac 外排泵的多种 *qac* 基因(*qacA*、*qacB*、*qacC*、*qacEΔ1*、*qacG*、*qacH*、*qacJ*)均位于质粒上[78];携带 QACs 抗性基因 *bcrABC* 的不相容质粒(IncP)广泛分布在 QACs 亚抑制浓度环境中,且可以在所有的革兰氏阴性菌之间转移[86,87];转座子 *Tn6188* 上也发现携带 *qacH* 和 *emrE* 基因[80]. 这些 MGEs 可以通过水平转移在不同细菌之间传播,加剧了 QACs 抗性的扩散[88,89].

在环境中,QACs 的浓度远低于其 MIC,且 QACs 在环境中可以被好氧降解,形成浓度梯度,为细菌产生 QACs 抗性创造了良好的条件. Yang 等[59]发现 10 $\mu g \cdot L^{-1}$ QACs 会诱导水体中微生物群落中 *qacA/B* 丰度急速上升. 但实际环境中微生物应对 QACs 的主要抗性机制是什么、不同类型微生物的抗性机制是否存在差异、诱导环境微生物出现 QACs 抗性的最低浓度以及诱导 QACs 抗性基因水平转移的最低浓度是多少,以上问题均有待揭示.

4 QACs 对细菌抗生素抗性的协同选择机制

由于作用机制和作用对象的相似性,QACs 大量使用还可能诱导细菌出现抗生素抗性. 细菌出现抗生素抗性将会导致更长的住院时间,更高的死亡率,以及更大的经济负担,抗生素抗性菌的出现和传播已经成为一个日益严重的全球性问题[90~92]. 由于其广泛存在性、高传播性以及危害性,抗生素抗性基因(antibiotic resistance genes,ARGs)已经被列为一类新型污染物. 越来越多的证据表明,QACs 会诱导细菌产生抗生素抗性. Han 等[93]将从天然水体中分离出的 *Pseudomonas aeruginosa* 在含 3 种 QACs 的筛选培养基上培养 30 d 后,发现其对四环素和环丙沙星的敏感性均显著降低. 将湖泊水体微生物暴露在不同浓度的 QACs 下,发现磺胺类抗性基因(*sulⅠ*)、四环素类抗性基因(*tetA*、*tetM*)和喹诺酮类抗性基因(*qnrD*)的丰度均有不同程度的上升[57]. 目前认为关于 QACs 对细菌抗生素抗性的选择机制主要包括协同抗性和交叉抗性.

协同抗性机制是指 QACs 抗性基因与 ARGs 位于同一 MGEs 上. 多种 QACs 抗性基因和 ARGs 均位于Ⅰ类整合子(*IntⅠ*,位于质粒或转座子上的 DNA 整合元件)上[94~96]. Heuer 等[97]对 16 种耕地土壤进行分析,发现*sulⅠ* 和 *qacEΔ1* 与 *IntⅠ* 成显著正相关. 对 *Escherichia coli* 和 *Klebsiella pneumoniae* 进行基因组分析,证实*IntⅠ* 上携带 β-内酰胺类抗生素抗性基因(bla_{IMP-11})、氨基糖苷类抗生素抗性基因(*aacA1*)、*sulⅠ* 和 *qacEΔ1*[98]. Han 等[93]发现在 48 个水体样本中,7 种 ARGs 与*IntⅠ* 和 *qacEΔ1* 显著正相关,5 种环境浓度下的 QACs 均显著促进了 ARGs 抗性质粒(RP4 质粒)在大肠杆菌间的接合转移. 有研究通过对2 666条公开可用的细菌染色体和1 926个质粒进行分析,发现 QACs 抗性基因和 ARGs 之间存在正相关关系, *qacEΔ1* 在质粒上被频繁检出[99].

交叉抗性是指细菌通过外排泵、形成生物膜和改变细胞膜组分等通用抗性机制获得 QACs 抗性的同时也获得抗生素抗性. 如上文所述,一些外排泵本身就是多药外排泵,如 NorA、CemABC 和 QacC 外排泵也可同时将某些 β-内酰胺类抗生素排出体外[100]. 生物膜是细菌保护自身免受环境压力的一种通用机制,McBain 等[101]发现 QACs 能够促进细菌形成生物膜,降低了细菌对 QACs 和抗生素的敏感性. 同时,细菌也可以通过改变细胞膜脂肪酸和磷脂的含量来降低细胞膜的通透性,减少 QACs 和抗生素的摄入,从而降低细菌的敏感性[102].

QACs 导致微生物出现应激效应也会诱导 ARGs 的产生. Luo 等[60]发现膜生物反应器中添加 CTAB 后,多种 ARGs 的表达量升高,其中四环素类抗性基因(*tetO* 和 *tetW*)表达量增加得最多. 证实是由于 CTAB 损害了细胞膜和呼吸磷酸化,干扰电子传递体系,使得 O_2 传递过程受阻,细胞出现应激反应,导致外排泵机制的 ARGs 过度表达[60,103].

5 展望

(1)关于土壤和水等实际环境介质中 QACs 降解过程及微生物响应机制研究. 实际环境中存在除分子氧外的其他多种电子受体,QACs 的起始反应和降解机制有可能发生变化,产生毒性更强的中间产物. 进一步,作为非专一性的杀菌剂,QACs 及其降解产物会对微生物群落结构和相互作用关系产生怎样的影响? 明确这些问题有助于明确 QACs 的环境行为和效应.

(2)关于 QACs 对更多生物的长期慢性毒性效应研究,尤其是在生态循环中发挥重要作用的生物. 现有数据表明 QACs 在环境中的浓度不会对受试生物构成致死威胁,但对 *Daphnia magna* 等水生物有显著慢性毒性效应,且毒性效应浓度受生物种类影响显著. 为了全面了解 QACs 的环境风险,亟待分析其对更多生物的长期慢性毒性效应及其机制. 进一步,QACs 在环境中常与其他污染物共存,其复合毒性效应研究也应给予充分关注.

(3)关于环境微生物群落对 QACs 胁迫的主要抗性机制研究.单菌株的抗性机制研究发现,由于细胞外膜结构的差异,革兰氏阳性菌和革兰氏阴性菌对 QACs 抗性机制存在显著差异.在实际环境中,多种微生物共存,其应对 QACs 胁迫的主要抗性机制是什么,不同类型微生物的应对策略是否存在差异以及它们之间的互作关系均不清晰.更重要的是,QACs 是否会诱导环境微生物出现 QACs 抗性和促进抗性基因在不同种属间水平转移,最低的作用浓度是多少？明确这些问题有助于制定有效的 QACs 管控措施.

6　结论

(1)QACs 在水体、土壤和沉积物中被广泛检出,在水体中浓度范围为几～几十$\mu g \cdot L^{-1}$,在沉积物和土壤中浓度范围为几十～几千$\mu g \cdot kg^{-1}$.吸附和好氧生物降解是其在环境中的主要迁移转化过程,吸附机制主要涉及离子交换、静电作用和疏水作用.关于纯菌株对 QACs 降解的研究发现,在好氧条件下, QACs 可被 *Pseudomona*、*Aeromonas*、*Xanthomonas* 和 *Bacillus* 等种属细菌作为碳源或能源利用,降解速度与微生物种属、共存离子显著相关.在分子氧和还原辅酶Ⅱ(NADPH)的参与下,单加氧酶催化的羟基化反应为 QACs 生物降解的起始步骤,差别仅在于发生羟基化的碳位点不同.

(2)目前尚未发现环境浓度下 QACs 会对受试生物构成急性致死效应,但环境浓度下 QACs 的长期暴露会抑制 *Daphnia magna* 的生长繁殖.QACs 的毒性效应主要受自身结构、暴露时间和受试生物种类的显著影响.一般地,烷基链越长毒性越强,暴露时间越长效应浓度越低,在已有关于水生生物研究中, *Daphnia magna* 对 QACs 最敏感.

(3)有机质和矿物质对 QACs 的可逆性吸附延长了其在环境中的赋存时间,长期、低剂量 QACs 胁迫会诱导微生物出现 QACs 抗性.此外,QACs 的生物转化导致其在环境中形成浓度梯度,也利于微生物适应胁迫而出现抗性.除了降解利用,微生物主要通过修饰细胞膜结构、抑制膜孔蛋白对 QACs 的输送、药物外排泵蛋白高表达、捕获 MGEs 获得 QACs 抗性基因以及形成生物膜等机制提高对 QACs 的抗性.此外,QACs 还可以通过协同抗性和交叉抗性机制诱导微生物出现抗生素抗性,加大环境中 ARGs 的传播风险.

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