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近 15 年我国土壤抗生素污染特征与生物修复研究进展

赵晓东^{1,2}, 乔青青¹, 秦宵睿¹, 李晓晶^{2*}, 李永涛³

(1. 太原师范学院生物科学与技术学院, 榆次 030619; 2. 农业农村部环境保护科研监测所, 农业农村部产地环境污染防治重点实验室, 天津市农业环境与农产品安全重点实验室, 天津 300191; 3. 华南农业大学资源环境学院, 广州 510642)

摘要: 近年来, 抗生素及其抗性基因 (ARGs) 污染问题受到国内外学者的广泛关注, 土壤作为抗生素和 ARGs 迁移转化重要的汇给土壤生物和人类健康带来了威胁. 从近 15 年的相关调查研究结果来看, 全国各地土壤遭受了不同程度的抗生素污染. 生物修复是一种绿色且环保的修复技术, 在治理抗生素污染土壤方面有着巨大的潜力. 通过综述近 15 年我国土壤抗生素污染的时空变化特征, 总结植物、动物和微生物在抗生素污染土壤修复方面的应用, 特别介绍了微生物电化学系统在消减土壤中抗生素和 ARGs 的研究进展, 并指出了相关研究存在的不足和未来发展的方向, 以期能为土壤污染修复提供科学依据.

关键词: 抗生素; 抗生素抗性基因 (ARGs); 土壤; 生物修复; 微生物电化学系统

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Characteristics of Antibiotic Contamination of Soil in China in Past Fifteen Years and the Bioremediation Technology: A Review

ZHAO Xiao-dong^{1,2}, QIAO Qing-qing¹, QIN Xiao-rui¹, LI Xiao-jing^{2*}, LI Yong-tao³

(1. College of Biological Sciences and Technology, Taiyuan Normal University, Yuci 030619, China; 2. Tianjin Key Laboratory of Agro-Environment and Agro-Product Safety, Key Laboratory of Original Agro-Environmental Pollution Prevention and Control, Agro-Environmental Protection Institute, Ministry of Agriculture and Rural Affairs, Tianjin 300191, China; 3. College of Natural Resources and Environment, South China Agricultural University, Guangzhou 510642, China)

Abstract: In recent years, the contamination of antibiotics and their resistance genes (ARGs) has attracted the extensive attention of researchers at home and abroad. Soil is an important sink for the migration and transformation of antibiotics and ARGs, which pose a threat to soil organisms and human health. According to the relevant investigations in the past 15 years, the soil has been polluted by antibiotics to varying degrees in China. Bioremediation is a green and environment-friendly remediation technology, which has great potential in the remediation of antibiotic-contaminated soil. This review summarized the spatial and temporal characteristics of antibiotic pollution of soils in China in the past 15 years and the application of plants, animals, and microorganisms in the remediation of antibiotic-contaminated soil. In particular, the recent research advances of microbial electrochemical systems in removing antibiotics and ARGs in soil were reviewed, and the unaddressed issues of relevant research and the direction of future development were proposed, in order to provide a scientific basis for soil pollution remediation.

Key words: antibiotics; antibiotic resistance gene (ARGs); soil; bioremediation; microbial electrochemical system

抗生素是由微生物、高等动物和植物在其生命活动过程中所产生或通过其它途径获取的, 具有抗病原体或其它活性的次级代谢产物, 并能在低微浓度下干扰其它生活细胞发育功能的有机物质^[1,2]. 1928 年 9 月英国细菌学家亚历山大·弗莱明首次发现了青霉素, 这标志着抗生素在人类世界的正式诞生. 20 世纪 50 年代初, 在美国畜禽养殖业为提高饲料效率、预防疾病和促进畜禽生长, 首次在饲料中人工添加抗生素等添加剂^[3,4], 从此开启了抗生素在畜禽领域的应用.

近年来, 人类对肉蛋奶等畜产品日益增长的需求促进了我国畜禽养殖业的飞速发展, 导致抗生素在畜禽养殖业的投入迅猛增加甚至滥用^[5]. Zhang 等^[6]对中国抗生素投入量的调查研究发现, 2013 年中国抗生素的总使用量达近 92 700 t, 兽用消耗占抗生素总使用量的 52%. 抗生素进入动物体内很难被吸收, 最后通过粪便和尿液的排泄率达 28% ~ 88%^[7], 这造成了抗生素在动物粪便中较高的残留量, 以四环素类抗生素为例, 其在猪粪固相部分中的

残留含量高达 0.14 ~ 183.45 mg·kg⁻¹^[7~9].

畜禽粪尿等动物废弃物可作为肥料施入土壤补充土壤有机物质, 供给作物所需的必需营养元素, 这是防治土壤退化、恢复土壤生态系统的重要方式^[10]. 据估计, 我国每年畜禽粪肥的生产量超过了 30 亿 t^[11], 但是随着动物废弃物在农业土壤的大量使用, 畜禽粪肥中的抗生素也随之侵入土壤^[12]. 对我国抗生素向环境中排放量的调查表明, 近 84% 的抗生素排放量 (54 000 t) 来自畜禽养殖场^[6]. 抗生素入侵土壤后不仅会引发细菌耐药性, 而且抗生素和耐药菌会通过地表径流和随水下渗等方式迁移至地表水、地下水以及其它环境介质, 甚至通过农作物对人类健康造成潜在的威胁^[13,14]. 有研究表明, 不仅在儿童和孕妇群体中检测到抗生素的存

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作者简介: 赵晓东 (1990 ~), 男, 博士, 副教授, 主要研究方向为土壤有机污染修复, E-mail: zhaoxiaodong@tynu.edu.cn

* 通信作者, E-mail: lixiaojing@caas.cn

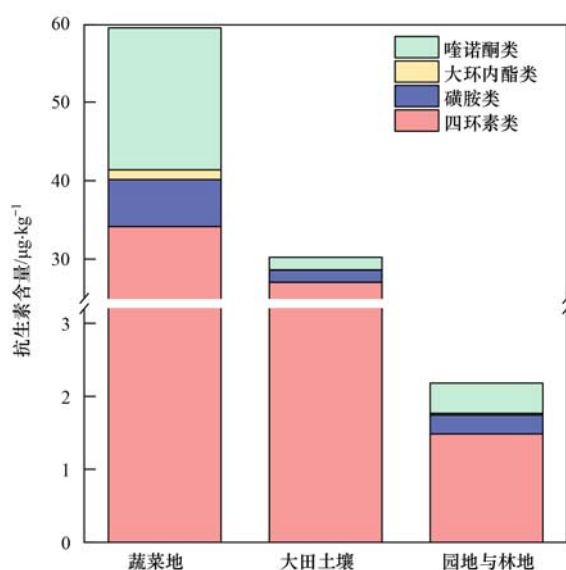
在^[15,16],而且 93% 的老年人也受到抗生素的危害^[17],甚至 12% 的新生儿存在对抗生素的耐药性^[18].

1 土壤中抗生素的污染特征

1.1 土壤中抗生素的空间分布特征

作为抗生素进入农田土壤的主要源头,畜禽养殖场附近的农田土壤所遭受抗生素的污染达到了很高的程度^[13],例如在上海某养殖场附近农田土壤中的四环素类、磺胺类抗生素和氯霉素的含量分别高达 5~25、6~33 和 3~18 $\text{mg}\cdot\text{kg}^{-1}$ ^[19].但也有研究表明,养殖场对周边土壤中抗生素分布的影响不大,并未引起较高的生态风险,这可能与养殖场的运作模式以及所在地区的气候特征有关^[20,21].较大田土壤而言,蔬菜地因其较高的土地利用强度和畜禽粪便投入等因素导致了较严重的抗生素污染(图 1),而农田土壤中的抗生素残余含量总体显著高于园地和林地等非农田土壤^[22~24].尽管抗生素在农田土壤的残留量主要取决于畜禽粪肥投入、污水灌溉等农作措施,但也有研究表明在粪肥投入量相等的情况下,水稻田因具有较高的微生物量,其土壤中抗生素残余量明显高于花生地^[25].此外,农作物作为人类生存的重要物质基础也在不同程度上受到了抗生素的污染.研究发现,玉米可食用部分富集了相对较高的抗生素,蔬菜次之,其余 3 种作物由高到低顺序依次为水稻、白菜和萝卜,且四环素类和喹诺酮类是主要的抗生素类型^[26,27].

四环素类、磺胺类、喹诺酮类和大环内酯类抗



整理自文献^[20,23,24,28~41]

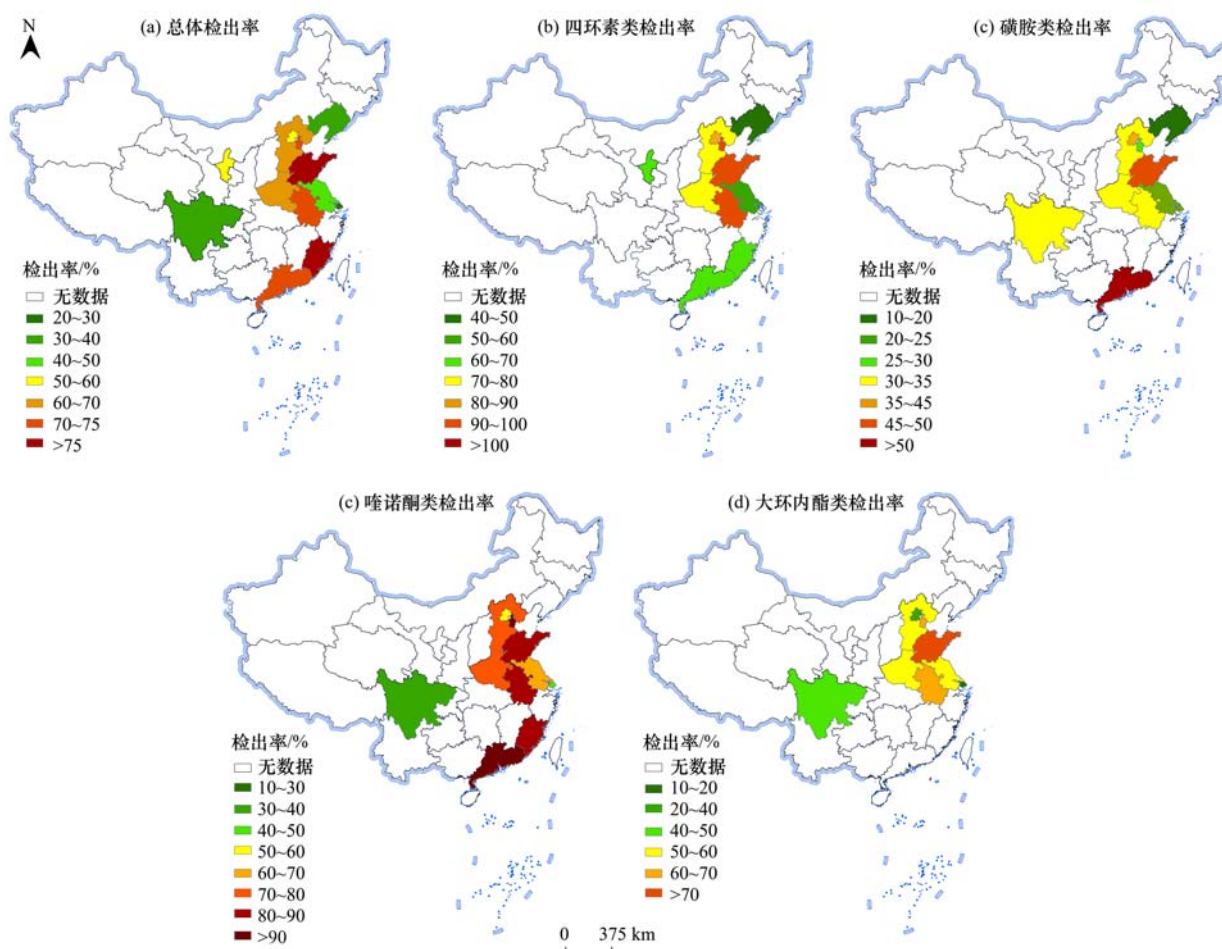
图 1 不同土地利用方式下抗生素含量的变化

Fig. 1 Changes in antibiotic content under different land use patterns

生素是畜禽养殖场中最常使用的 4 大类抗生素,通过对近 15 年的相关调查进行总结发现,我国表层土壤(0~20 cm)中抗生素的平均检出率高达 58% (图 2),检出率较高的地区主要包括山东(76%)、福建(76%)、广东(75%)、天津(71%)和安徽(71%)^[20,23,24,28~41].四环素类抗生素在表层土壤中被检出的频率最高(45%~99%,平均 76%),喹诺酮类次之(16%~99%,平均 73%),而磺胺类和大环内酯类的检出率相对较低.同时,辽宁、宁夏、福建和广东等地区的表层土壤中抗生素的含量明显高于其他省,达 15~116 $\mu\text{g}\cdot\text{kg}^{-1}$,而新疆地区最低,仅为 0.044 $\mu\text{g}\cdot\text{kg}^{-1}$ (图 3).四大类抗生素在表层土壤中的含量由高到低依次为四环素类、喹诺酮类、磺胺类和大环内酯类. Pan 等^[42]选取四环素、磺胺甲噁唑、诺氟沙星和红霉素作为典型抗生素对其环境行为进行研究,发现抗生素在土壤中的吸附能力从大到小排序为:四环素>诺氟沙星>红霉素>磺胺甲噁唑,而抗生素吸附能力的强弱直接影响其在土壤中的空间分布.

基于前人研究结果可知,四环素类和喹诺酮类抗生素主要分布于表层土壤,并随土层深度加深呈下降趋势,而磺胺类抗生素呈相反趋势(图 4)^[35,43].按照抗生素在土壤中的迁移强弱将其大致分为两类:第一类是在土壤中迁移能力强的抗生素,包括磺胺类和大环内酯类抗生素^[44].有调查研究均表明,磺胺类抗生素因其强烈的迁移能力很容易进入水生环境(4~291 $\text{ng}\cdot\text{L}^{-1}$),其在水环境中的残余含量较其它类型抗生素高出 6~25 倍,对水生生态系统造成极高的潜在风险^[45~49]. Zhao 等^[22]调查研究表明,大环内酯类抗生素因其在土壤中较弱的吸附能力,其残余含量随着土层的增加而明显增加.这也是磺胺类和大环内酯类抗生素在作物中积累较少的主要原因之一.第二类是在土壤中迁移能力弱的抗生素,如四环素类和喹诺酮类抗生素^[22,50],因其特殊的结构致使其强烈的吸附于土壤胶体^[51],这使得这两类抗生素在表层土中会残留很长时间.此外,四环素类和喹诺酮类抗生素易被微塑料通过氢键等方式吸附,从而在土壤中发生共迁移,最终可能会加剧其在环境中的生态风险^[52,53].

除抗生素自身的特性外,抗生素在土壤中迁移转化明显受到土壤理化性质的制约,这也是影响土壤环境中抗生素分布特征的重要因素.土壤 pH、阳离子交换量、有机质、铁氧化物和层状硅酸盐黏土矿物均强烈影响着抗生素在土壤的吸附能力^[54~57].抗生素在土壤中主要通过范德华力、色散力及氢键等分子间作用力(物理吸附)和络合或螯合作用(化



数据整理自文献[20,23,24,28~41]; 基于自然资源部标准地图服务网站审图号 GS(2020)4615 号标准地图制作, 底图无修改, 香港、澳门和中国台湾地区资料暂缺, 下同

图2 我国表层土壤(0~20 cm)中抗生素的检出率

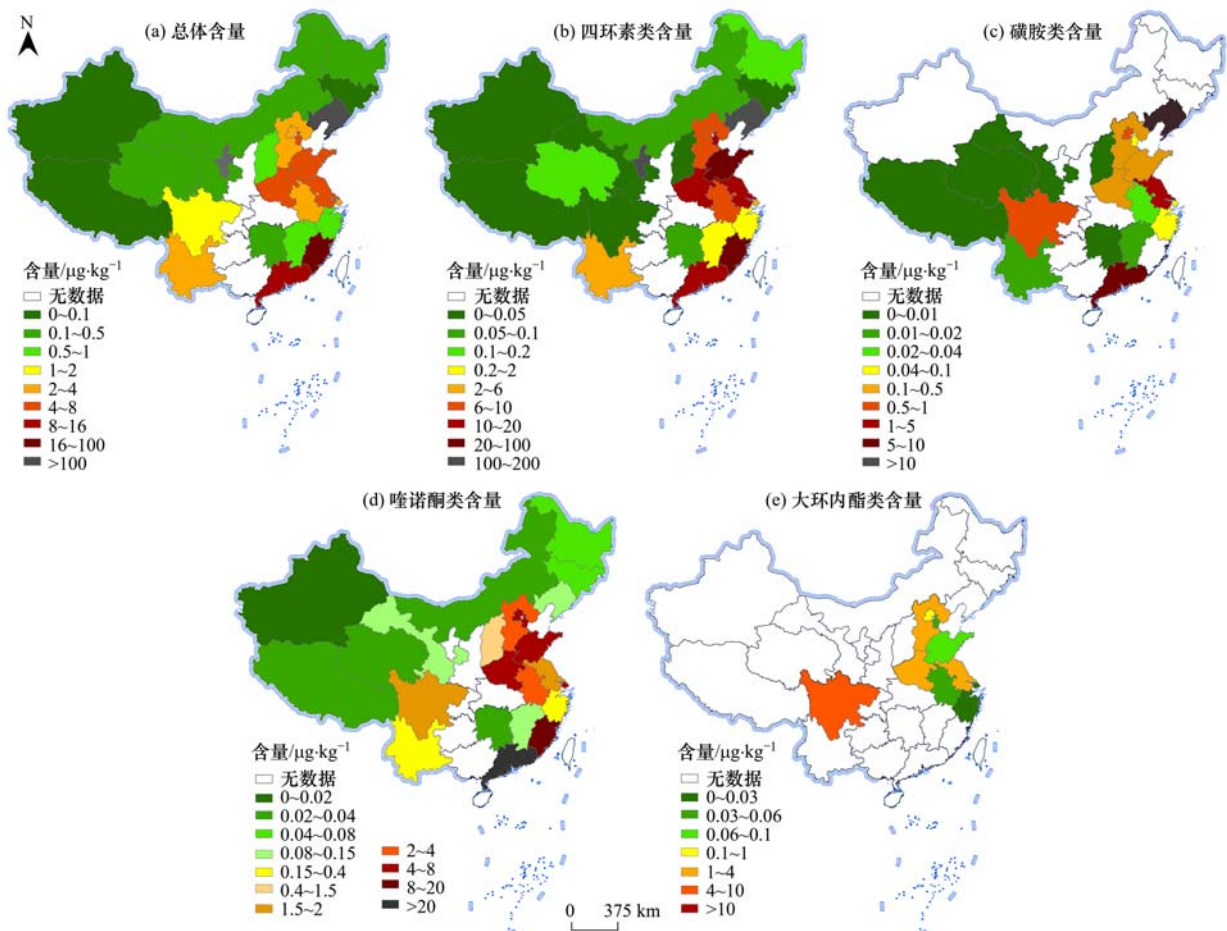
Fig. 2 Detection rate of antibiotic in topsoil (0-20 cm) of China

学吸附)与土壤胶体进行吸附. Bao 等^[58]研究发现, 红壤中富含的高铁铝氧化物是造成四环素在红壤中强烈吸附的主要原因. 然而去除红壤中的铁铝氧化物并不一定会降低四环素的吸附能力, 究其原因是由于铁铝氧化物通常在有机质表面形成包膜, 一旦铁铝氧化物被去除, 有机质反而会被暴露于土壤中吸附四环素^[59]. 密度泛函是揭示抗生素吸附机制的重要工具^[60], Zhao 等^[61]基于密度泛函理论研究了 22 种磺胺类抗生素在介孔碳上的吸附, 发现磺胺类抗生素吸附的主要驱动力为 π - π 相互作用、疏水效应和氢键.

1.2 土壤中抗生素的时间分布特征

相比于抗生素在土壤中的空间分布, 目前对我国土壤中抗生素的时间分布特征的研究相对较少, 且主要集中于季节变化规律^[62]. 水分和温度是决定抗生素随时间变化的重要因素, 一方面适宜的水热条件有利于加快土壤微生物对抗生素的降解, 另一方面低温可增强土壤对抗生素的吸附^[63]. 此外, 强降雨也会造成抗生素在土壤中的淋溶与迁移^[64]. 有

研究指出在中国东部长三角城市周边地区, 由于冬季低温低湿条件降低了抗生素的降解和迁移, 从而导致冬季土壤中抗生素的含量高于夏季^[22]. 同时, 秋冬季较为频繁的畜禽粪肥施用也会造成冬季土壤中抗生素的含量高于夏季, 尤其是喹诺酮类抗生素^[65]. Pan 等^[28]调查了抗生素在不同种植年限大棚土壤中的分布规律, 发现抗生素在新棚(1~6 a)土壤中的残留量高于老棚(7~30 a). Zhang 等^[66]研究也表明, 棚龄在 6 a 以上的土壤中抗生素含量远高于 5 a 以内棚龄的土壤. 究其原因, 一方面可能是长期的粪肥投入导致老棚土壤已被驯化较为丰富的抗生素高效降解菌, 另一方面这也可能与粪肥的投入量有关. 通过总结先前文献发现, 尽管土壤中抗生素含量随时间的推移呈现出一定的波动, 但结合拟合分析结果发现抗生素含量整体随时间呈下降趋势(图 5), 这不仅在侧面暗示了日趋完善的国家相关政策法规对抗生素使用的规范性, 也在一定程度上反映了当前修复技术对抗生素污染土壤的有效治理.



整理自文献[20,23,24,28~41]

图3 我国表层土壤中抗生素的含量变化

Fig. 3 Changes in antibiotic content in topsoil of China

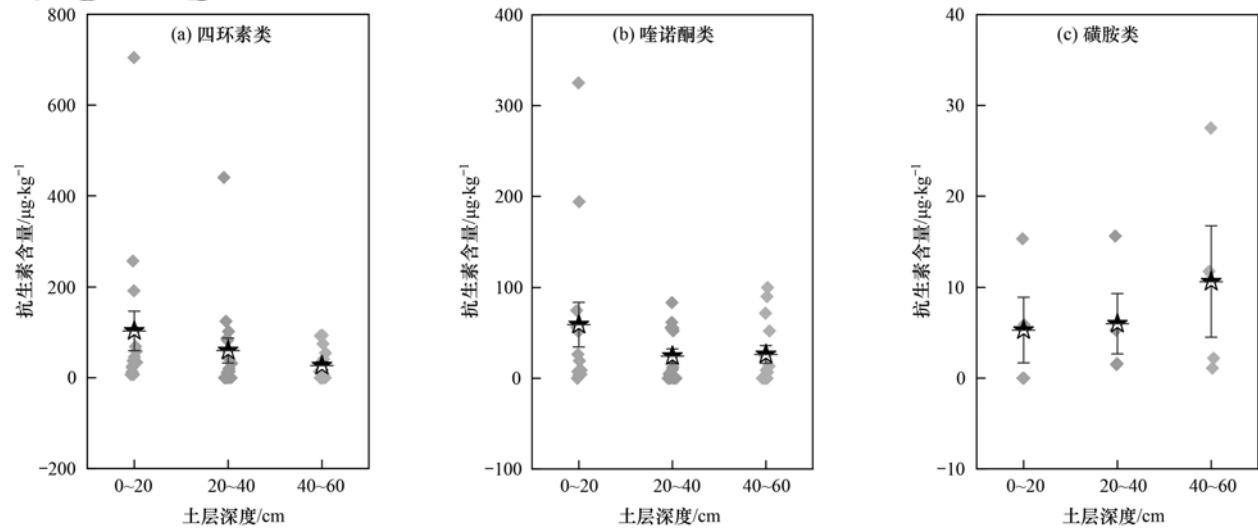


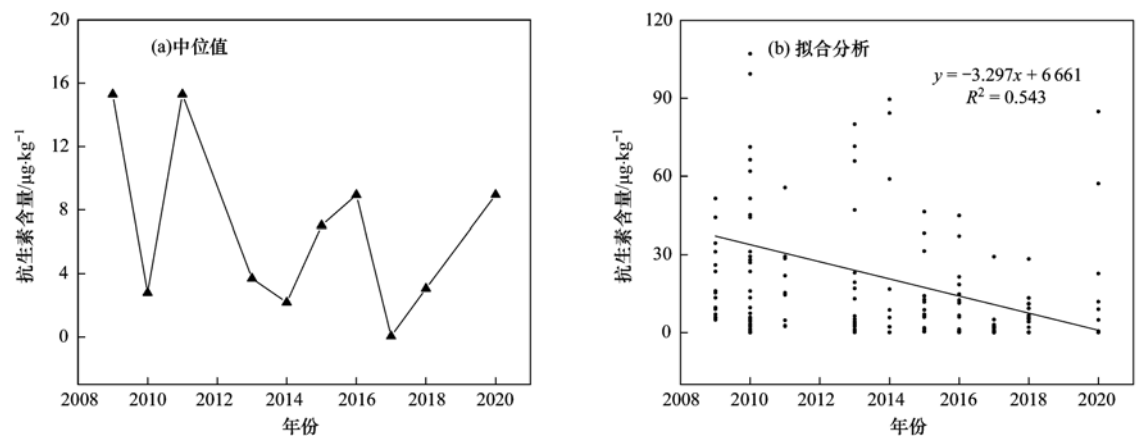
图4 不同土层中抗生素含量的变化

Fig. 4 Changes in antibiotic content in different soil layers

2 土壤中抗生素抗性基因的污染特征

微生物耐药性包括固有耐药性和获得耐药性,除部分微生物具有天然耐药性之外,其余主要通过自身基因突变和抗性基因水平转移获得^[67,68]. 抗生

素抗性基因(ARGs)的水平转移一方面会通过移动遗传元件(MGEs,包括质粒、整合子和转座子等)进行,另一方面噬菌体(细菌病毒)也在此转移过程中起着不可忽视的作用^[69~71]. Chen等^[72]研究发现,有机肥的长期施用不仅造成了携带ARGs的土壤噬

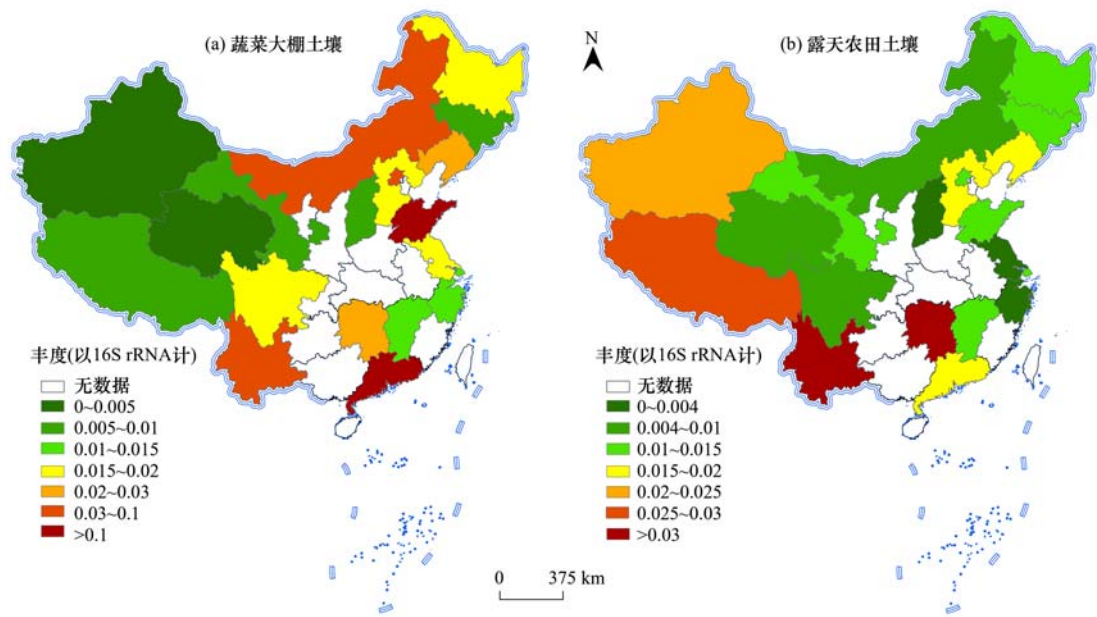


整理自文献[20,23,24,28~41]
图5 土壤中抗生素随时间的变化及拟合分析
Fig. 5 Changes in antibiotics in soil with time and fitting analysis

菌体种群的显著改变,而且加强了噬菌体与细菌的关联度,从而扩大了 ARGs 的宿主范围,最终增加了 ARGs 在土壤环境中的传播风险.此外,即使耐药菌株已经死亡,其携带 ARGs 的裸露 DNA 也有可能吸附在土壤颗粒,并在适当条件下转移至其它微生物体内^[73].

近年来,抗生素的滥用进一步加剧了 ARGs 的扩增与传播,全球每年因细菌耐药性而死亡的人数高达 70 万,因此 ARGs 已经被世界卫生组织列为危害全球健康的三大威胁之一,对人类感染类疾病的预防与治疗造成了严峻的考验^[74~76]. Fang 等^[77]调查了 ARGs 及人类致病菌在鸡粪肥和设施菜地土壤中的分布情况,发现四环素抗性基因是粪肥中抗性基因的主要类型,而在设施菜地土壤中多重耐药基

因占主要成分,同时抗生素、ARGs 和人类致病菌展现出显著的正相关关系.黄福义等^[78]对种植不同作物农田土壤 ARGs 的污染特征进行了研究,发现 ARGs 在土壤中的丰度按由大到小顺序依次为:香蕉土>水稻土>花生土>甘蔗土>柑橘土,同时在种植香蕉和水稻土壤中检测出丰度较高的移动遗传原件基因,这进一步增加了 ARGs 的传播风险. Zeng 等^[31]对我国蔬菜大棚和露天土壤中 ARGs 的分布特征进行了系统的调查,结果表明,广东和山东两地蔬菜大棚土壤中的 ARGs 含量明显高于其他地区,其丰度高达 0.11~0.12 (以 16S rRNA 计)[图 6 (a)],而露天土壤中 ARGs 的丰度显著低于蔬菜大棚,且云南和湖南等地存在着较高的污染风险[图 6 (b)]. *tet*、*sul* 和 *qnr* 基因是农业土壤中分布最广泛



改自文献[31]
图6 我国土壤中 ARGs 丰度的变化
Fig. 6 Changes in ARGs abundance in soil of China

的 ARGs,其中 *sul* 基因丰度明显高于 *tet* 基因和 *qnr* 基因. 有研究也表明, ARGs 广泛分布于我国的农田土壤中,主要受到了施肥、灌溉、作物产量、地膜覆盖和农药等因素的影响,其中畜禽粪肥是其主要的影响因素^[79]. 在粪肥改良的土壤中, ARGs 会通过弹尾虫 (*Folsomia candida*)-捕食性螨 (*Hypoaspis aculeifer*) 食物链传播,从而加剧其传播风险^[80].

然而,在实际土壤中并非只存在抗生素一种污染物,复合污染对 ARGs 的胁迫现象十分普遍. 重金属作为抗生素的同源污染物(同为饲料添加剂),其含量与 ARGs 丰度呈正相关关系^[13,19,68]. 在我国,尽管近年来动物饲料中已被禁止添加抗生素,但是在用于治疗动物疾病的抗生素和重金属的协同选择作用下也会造成 ARGs 的扩增^[81]. 微塑料是指直径小于 5 mm 的塑料颗粒,作为一种新型污染物其粒径小、比表面积大、分布广且在环境中残留时间长,成为了其它污染物的重要载体,引起了国际社会的广泛关注^[82]. 对我国 19 个省的调查结果显示,土壤样品中尺寸较大的塑料含量在 0.1 ~ 324.5 kg·hm⁻²之间,可能有数万亿基于地膜的微塑料颗粒被排放到土壤中^[83]. 尽管目前缺乏对全国尺度的微塑料污染土壤调查数据,但根据青藏高原、云南、渤海和黄海沿岸等局部区域土壤的微塑料污染分布特征,可以预测我国土壤环境中的微塑料污染状况不容乐观^[84]. 有研究表明,微塑料通过改变土壤性质提高了土壤中铜和四环素的生物有效含量,增加了对微生物的选择压力,导致了耐药菌在微塑料表面及周围的富集,最终加剧了 ARGs 的扩增与传播风险^[85]. 微塑料粒径越大和风化程度越强,越会促进其对周围环境中抗生素和重金属的吸附,从而增加微塑料表面 ARGs 的丰度和数量^[86]. 此外,残留在土壤中的杀虫剂也可能通过共选择机制加剧 ARGs 的污染^[87].

3 抗生素污染土壤生物修复技术

农业农村部农业生态与资源保护总站于 2019 年 8 月发布的《受污染耕地治理与修复导则(NY/T 3499-2019)》中指出污染耕地的修复技术应具有环境友好性,防止修复过程中对周边环境产生二次污染. 因此,生物修复作为一种绿色、环境友好型的修复技术在土壤污染治理过程中起着关键的作用. 生物修复技术主要包括植物修复、动物修复和微生物修复(图 7),本文主要从这三大方面对目前的国内外最新动态进行总结.

3.1 植物修复

植物修复技术以其投入成本低、环境干扰小且

修复效果佳等特点广泛应用于环境中各种污染物的修复^[88],它通过利用植物根系对污染物的积累、固定、挥发、降解和矿化,从而达到去除环境介质中污染物的目的^[89,90]. 植物积累是植物根系直接将污染物从环境转移至地上部组织中的过程^[91],主要通过筛选超积累植物如蜈蚣草(*Pteris vittata*)、东南景天(*Sedum alfredii* Hance)和伴矿景天(*Sedum plumbizincicola*)等,待污染物转移至植物之后将其收获进行集中处理. 相比重金属的植物提取,有机物的直接吸收取决于目标污染物的理化性质如辛醇-水分配系数^[92]. 植物固定是利用某些植物对污染物的机械固定作用(如根际的吸附与沉淀),以阻止污染物的迁移,减少其向生态系统和食物链的转移^[93]. 植物降解是指在植物酶的驱动下,污染物在根际或者植物组织中的完全或部分降解,其降解机制主要是植物酶将目标污染物完全矿化为无机化合物如 CO₂ 和水,或转化为稳定的化合物贮藏在植物组织中^[90]. 植物代谢活动及其相关微生物将目标污染物转化为挥发性化合物并释放到大气过程称为植物挥发,该过程还伴随着对污染物的稀释和分散,这有助于植物及微生物对污染物的降解^[94].

运用黑麦草(*Lolium perenne*)和伴矿景天对土壤中抗生素(初始含量分别为 15 ~ 903 μg·kg⁻¹和 0.6 ~ 12.7 μg·kg⁻¹)的去除率分别达 3.4% ~ 28.8% 和 36% ~ 76%^[95,96]. 相比于单作多花黑麦草(*Lolium multiflorum*)和印度芥菜(*Brassica juncea*),轮作后对抗生素的去除率增加了 46% ~ 54%,且 ARGs 丰度被进一步降低^[97]. 虽然植物修复有着得天独厚的优势,但其缺点也不容忽视,比如许多植物对污染物敏感,导致其生长缓慢,难以产生足够的生物量进行植物修复^[98],再如受体内酶活性和数量的限制,植物本身对有机污染物的降解能力较弱等^[99],因此植物-微生物联合修复技术成为了近年来的研究热点.

植物-微生物联合修复技术分为植物与专性菌株的联合修复和植物与菌根的联合修复,前者是利用植物根系与污染土壤中专性降解菌的协同作用,后者是利用菌根根际特殊的微生态区域所维持的较高的微生物种群密度和生理活性强化微生物对污染物的降解能力^[100]. 豆科植物能在根部形成根瘤,通过利用植物与根瘤菌的天然共生系统,不仅增强植物固氮能力,更重要的是增强植物和微生物对土壤中污染物的去除能力. 植物根系通过分泌营养物质有助于增加根际与专性降解菌等功能微生物的群落数量,改变种群结构和丰度并促进共代谢作用,有利于对污染物的降解^[101]. 湿地植物红树(*Rhizophora*

apiculata) 和 卤 蕨 (*Acrostichum aureum*) 具有较好的根系供氧能力和根系分泌物, 有利于微生物群落的生长, 从而有效刺激微生物去除喹诺酮类抗生素 (初始含量为 $10 \text{ mg} \cdot \text{kg}^{-1}$ 时, 半衰期为 10 d)^[102]. 利用植物紫茉莉 (*Mirabilis jalapa*) 和孔雀草 (*Tagetes patula*) 与土霉素降解菌 *Phyllobacterium myrsinacearum* 和 *Rhodotorula mucilaginosa* 修复土霉素污染土壤, 对高 ($30 \text{ mg} \cdot \text{kg}^{-1}$)、低含量 ($5 \text{ mg} \cdot \text{kg}^{-1}$) 土霉素的去除率分别达 17% ~ 59% 和 31% ~ 71%, 且孔雀草的总体去除效果优于紫茉莉^[103].

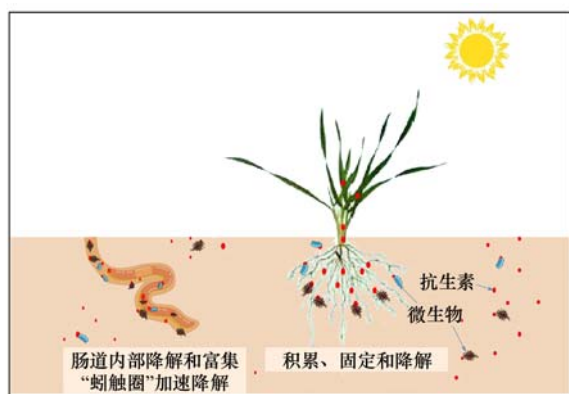


图 7 土壤生物修复技术原理

Fig. 7 Principles of soil bioremediation technology

3.2 动物修复

土壤动物是土壤生境中不可或缺的组成部分, 主要包括蚯蚓、蚂蚁、鼯鼠、变形虫、线虫、蜘蛛和千足虫等, 而蚯蚓是土壤动物中重要的物种, 也是目前为止土壤动物中研究最多的一类, 占土壤生物量的 60% ~ 80%^[104]. 蚯蚓按生态类型可划分为表层种、深层种和内层种这 3 种, 常见的蚯蚓有赤子爱胜蚓 (*Eisenia fetida*)、白颈环毛蚓 (*Pheretima californica*)、威廉环毛蚓 (*Metaphire guillelmi*) 和壮伟环毛蚓 (*Amyntas robustus*) 等^[105]. 由于蚯蚓的挖掘、消化和排泄活动具有很强的改造土壤结构、改善土壤理化性质、刺激微生物活动和促进反应的能力, 这会直接和间接地促进有机污染物的生物降解^[106].

目前为止, 该技术已经成功地实现对土壤中重金属^[107,108]、多环芳烃^[109]、农药^[110~112]、多氯联苯^[113,114]和抗生素^[115~119]等多种污染物的治理. 例如, Yang 等^[116]发现蚯蚓在显著提高土壤中金霉素及其代谢产物的去除的同时也降低了 *tetR*、*tetD*、*tetPB*、*tetG*、*tetA*、*sulI*、*TnpA*、*ttgB* 和 *intI1* 等基因的丰度, 在培养第 28 d, ARGs 的总丰度降低了 35% ~ 44%. 由于“蚓触圈”自身特殊的环境条件, 导致四环素在蚯蚓肠道、粪便和非“蚓触圈”土壤中的降解

菌存在差异, *Ralstonia* sp. 和 *Sphingomonas* sp. 是土壤中潜在的降解菌, 蚯蚓肠道 (*Pseudomonas* sp. 和 *Arthrobacter* sp.) 和 蚓 粪 (*Comamonas* sp.、*Acinetobacter* sp. 和 *Stenotrophomonas* sp.) 中的降解菌将 4-差向四环素脱水成 4-差向脱水四环素^[117]. 利用蚯蚓去除土壤中有机污染物的机制主要包括以下两方面: 第一, 蚯蚓通过表皮接触或进食吸收有机污染物, 肠道独特的厌氧状态为厌氧和/或兼性厌氧菌提供了有利的微环境, 使需氧和厌氧细菌的数量为土壤中的 12 ~ 20 倍和 10 ~ 4 000 倍^[120], 污染物一方面会在肠道和消化系统中加速降解和代谢^[121], 另一方面可能在蚯蚓体内富集^[122]. 第二, 蚯蚓在土壤中的移动和掘穴活动增强了土壤的通气性, 增加了微生物与污染物的接触几率^[123], 并通过蚯蚓的粘液分泌物以及对有机物质的分解以提供充足的碳源, 刺激降解菌在土壤中快速生长, 强化微生物对有机污染物的降解能力^[124]. 值得一提的是, 尽管蚯蚓的存在可以有效修复抗生素污染土壤, 但高含量抗生素也会诱导蚯蚓 DNA 链断裂和遗传毒性, 并造成 ARGs 在蚯蚓肠道的传播, 从而对蚯蚓生长造成威胁^[125,126].

3.3 微生物修复

土壤微生物修复是利用土著微生物或人工驯化功能菌基于其自身的代谢作用将目标污染物作为能源物质或者以共代谢的方式去除, 通过分泌胞外降解酶或者将污染物吸收至细胞内由胞内酶降解, 从而实现土壤污染治理的目的^[127,128]. 抗生素作为一种广谱性的杀菌物质对细菌和放线菌活性具有抑制作用, 会影响土壤呼吸、氮/铁循环等过程, 但对真菌活性影响不大, 甚至可能增加真菌生物量^[129,130]. 尽管抗生素的存在对古菌总量影响微弱, 但却会改变其群落结构, 尤其是产甲烷古菌, 对抗生素尤为敏感^[131,132]. 微型藻类、纤毛虫等原生生物也会受到抗生素的抑制, 例如土霉素的存在会使游仆虫 (*Euplotes crassus*) 基因组内 rDNA 序列多样性及其碱基组成发生改变, 从而显著抑制游仆虫的个体生存能力、细胞分裂效率和种群自然增长率, 这也表明原生生物可通过基因组高度的可塑性以适应抗生素污染胁迫^[133]. 抗生素对微生物的选择性压力会引发特异性微生物群落的形成, 诱导耐药菌株的产生, 同时也能促进对抗生素的生物降解^[134]. 虽然抗生素降解菌主要以耐药细菌为主 (它们通过取代基氧化与裂解、羟基化/去羟基化等过程降解抗生素), 但某些真菌 (如白腐真菌、镰刀菌、聚多曲霉、微紫青霉等) 也被发现具有降解四环素类、磺胺类和喹诺酮类抗生素的能力^[135,136]. 此外, 微生物还能

协同降解抗生素,例如类节杆菌(*Paenarthrobacter* sp.)可将磺胺甲噁唑降解为3-氨基-5-甲基异噁唑,为类诺卡氏菌(*Nocardioide* sp.)生长提供碳源,从而实现对磺胺甲噁唑的协同降解^[137]。

微生物修复主要分为原位修复和异位修复,原位修复主要是通过向土壤中微生物提供生长所需能源,促进土壤中微生物对污染物的降解,同时也可向环境中添加高效降解菌剂进行污染物原位修复。而异位修复是将污染土壤转移至特定环境中,利用高效降解菌对其进行集中修复。微生物原位修复法虽然成本低,但是易受场地环境的影响,修复难度相对较大。而异位修复法的修复效果较原位修复好,但是修复成本高,且不适合大面积污染场地的修复。无论原位修复还是异位修复都有各自的适用范围,其难易程度主要取决于目标污染物的化学结构和土壤中微生物的数量和活性,因此高效降解菌的筛选一直是研究的热点。史艳财^[138]筛选出8种四环素类抗生素的降解菌,其中2株四环素降解菌(*Citrobacter werkmanii*、*Providencia rettgeri* strain)、4株土霉素降解菌(*Citrobacter werkmanii*、*Bacillus megaterium*、*Arthrobacter* sp.、*Bacillus altitudinis*)、1株金霉素降解菌(*Pseudomonas putida*)以及1株四环素和金霉素的降解菌(*Comamonas testosteroni*),均为革兰氏阴性菌。解开治等^[139]研发了高效降解四环素类抗生素的菌剂,其对土壤中四环素、土霉素、多西环素的去除效率均高于95%。红霉素酯酶源于大肠杆菌质粒或绿脓杆菌假单胞菌,具有降解红霉素的潜力,将红霉素酯酶(*ereA*)表达于大肠杆菌表面,构建出的基因工程菌*E. coli ereA*可有效提高对红霉素的降解效率^[140]。然而,由于土壤环境条件较差、菌剂缺乏营养来源,以及土著微生物对入侵的高效降解菌的“排斥效应”,使得向污染土壤中施入菌剂的实际效果很难达到室内控制实验的预期效果^[141],而将降解菌株固定于载体可提高细胞渗透性、增加与污染物的接触面积、增强微生物稳定性,最终有利于降解菌在污染土壤中的定殖与修复^[142~144]。微生物固定化载体需要具有低成本、环保、机械和化学稳定性高、固定化细胞有足够空间、防止不必要的蛋白质变性、增强底物与固定化细胞之间的相互作用等特点,常用的载体包括天然类、合成聚合物凝胶和无机材料等^[145]。运用多孔蔗渣颗粒作为降解菌的固定化载体基质可以给菌株提供必要的有机物和营养物质,提高其对土壤重金属离子的耐受性,从而增强菌株对土壤环境的定殖和适应能力^[143]。生物炭具有高碳含量、高阳离子交换容量、高孔隙率、高稳定性和丰富的表面官能团等优点,被广泛

报道为潜在的固定化基质,它通过吸附、包封、共价键和交联等固定方式不仅为微生物的生长繁殖提供了空间,还可以提供少量的营养,抵御外界环境因素对微生物生长的影响^[146]。将具有降解四环素能力的贝莱斯芽孢杆菌(*Bacillus velezensis*)固定化,制备磁性固定化微生物复合材料,可有效增强该菌的降解效果,其降解率较仅添加该菌株的对照处理提高了20%^[147]。尽管对降解菌剂的研制与应用备受关注,但如何进一步增强高效降解菌在污染土壤中的定殖能力和实际修复效果目前仍旧是一个巨大的挑战^[148]。此外,尽管高效降解菌的添加有助于污染土壤的治理,但此过程可能引发的生物入侵与生物安全问题同样需要高度警惕。

3.4 土壤微生物电化学修复

随着有机污染物的氧化降解,污染土壤中氧化剂(电子受体)逐渐消耗以致缺乏限制了微生物的修复效率,而微生物电化学系统(MES)能够提供固体阳极作为永不枯竭的电子受体,在将土壤中有有机污染物降解的同时还能将此过程中的化学能直接转化为生物电能,既解决了微生物修复效率提升的技术瓶颈,又实现了污染治理与固废资源化利用的双功效,且该过程无二次污染^[149]。MES是基于产电呼吸的一种电化学装置,起初为双室构型,由电极材料、阳极室、阴极室和隔膜组成^[150]。其工作原理如图8(a)所示,主要分为底物生物氧化、阳极还原、外电路电子传输、质子迁移和阴极反应共5个步骤^[151]。双室微生物燃料电池的缺点在于阴极需要曝气,为此出现了单室微生物燃料电池[图8(b)],与双室的主要区别是阴极直接暴露于空气中,这样氧气会直接自发地扩散至阴极的亲水层从而避免了曝气所需的能量消耗。

3.4.1 电子传递机制

在MES中,电子主要通过直接和间接传递两种方式进行转移,直接传递包含纳米导线和膜蛋白两种途径,间接传递主要利用电子中介体(如蒽醌类物质)进行^[152,153]。产电微生物主要富集于MES阳极表面及附近,典型的产电菌包括:*Geobacter lovleyi*、*Geobacter sulfurreducens*、*Shewanella putrefaciens*、*Shewanella oneidensis*和*Thermococcus carboxydophilus*等,它们通过在阳极表面形成生物膜或者分泌氧化还原性电子传递介质完成向阳极的电子传输^[154~156]。然而,在正常环境中产电微生物向阳极传递电子的过程会受到诸多因素的限制,例如细胞外膜的c型细胞色素在向胞外传递电子的过程中会受到其辅助因子含量和固态电子受体极性等因素的影响^[157]。鉴于此,国内外学者基于产电微生物与

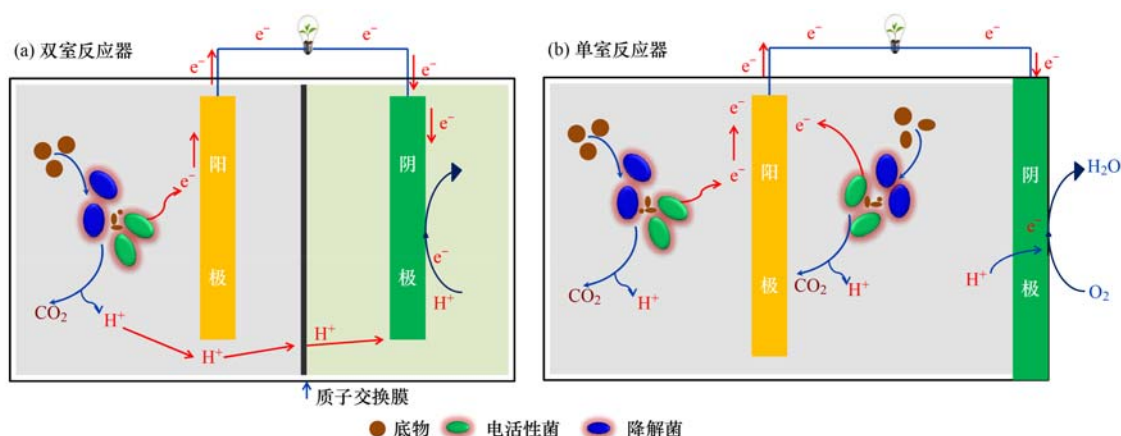


图 8 微生物电化学系统构型

Fig. 8 Schematics of microbial electrochemical systems

阳极间的作用机制,通过合成生物学手段改造细胞、导电纳米材料修饰细胞、在阳极表面固定化细胞等方式,对产电微生物向阳极的电子传递过程进行强化^[158]. Cao 等^[159]通过在外膜和跨膜区引入银纳米颗粒,极大地提高了希瓦氏菌的产电效率,其最大电流密度和功率密度分别达 $3.9 \text{ mA} \cdot \text{cm}^{-2}$ 和 $0.7 \text{ mW} \cdot \text{cm}^{-2}$,库仑效率高达 81%,是截至当前产电效率最高的 MES.

与生物阳极不同的是,生物阴极的电活性微生物通常大部分是自养型亲电微生物,且生物阴极的胞外电子传递机制目前尚不清楚^[160]. 胞外电子传递过程是双向的,生物阴极的电子传递通常为向内型传递过程,即阴极作为固体电子供体,电子穿过微生物的细胞膜进入细胞内部^[161]. 参照生物阳极的胞外电子传递机制,生物阴极的胞外电子传递机制可能分为两种:直接电子传递和中介体电子传递机制^[162]. 直接电子传递即通过微生物细胞膜蛋白或者纳米导线与阴极或者其它细胞直接接触进行电子传递,而中介体电子传递则是利用内生或外生的氧化还原化合物在阴极与微生物之间穿梭从而进行电子传递. 除 *Shewanella oneidensis* MR-1、*Geobacter sulfurreducens* 和 *Alcaligenes faecalis* 等具有双向胞外电子传递功能的微生物外,大部分电活性微生物只拥有单向胞外电子传递的能力,即它们只能作为电能的生产者或者消耗者而富集于电极附近^[163]. 同时,同一双向传递型电活性菌株在阳极和阴极部位的胞外电子传递机制也不尽相同,例如 *G. sulfurreducens* 的 PccP 蛋白在向内型电子传递过程是必需的,但在向外型电子传递过程中并不需要^[164].

3.4.2 在抗生素污染土壤中的应用

目前为止,MES 已经广泛应用于水体、沉积物和土壤中多种有机、无机污染物的修复研

究^[165~169]. 关于抗生素污染修复最早的研究出现于 2011 年,Wen 等^[170]运用一个单室空气阴极 MES 去除废水中的头孢曲松钠,研究发现头孢曲松钠 ($50 \text{ mg} \cdot \text{L}^{-1}$) 能够在 24 h 内降解 91%,其降解率较对照提高了 78%,在葡萄糖存在的条件下添加头孢曲松钠后系统输出最大功率密度增加了近 5 倍. 此后相关的研究逐渐受到国内外关注,但主要集中于废水处理. 研究使用的反应器构型多种多样,按其操作原理主要分为以下三大类:第一类主要结合厌氧微生物降解和功能微生物的电化学刺激作为其关键降解机制,即通过提供电子刺激微生物的代谢;第二类的研究重点在于生物阴极,其降解机制主要涉及直接的电化学还原和生物降解反应;第三类主要运用阴极产生的自由基攻击并降解目标抗生素^[171].

由于土壤自身的复杂性、异质性以及不同地域土壤类型的差异,导致 MES 对抗生素污染土壤的修复难度高于水环境,尤其是土壤巨大的内阻对电子传递效率的制约所造成的修复效率低下问题. 通过向土壤中添加二氧化硅胶体^[172]、砂粒^[173]、导电碳纤维^[174]和生物质炭^[175]等均可有效提高土壤的电子传递和修复效率. 例如,添加生物质炭可迅速提高土壤 MES 的产电效率,特别是添加木屑处理,其累积电荷输出较对照处理高出 36%,这也为土壤碳封存提供了新思路^[175].

Zhao 等^[176]采集了黑土、潮土、黄棕壤和红壤这 4 种类型土壤进行研究,发现 MES 更适用于高电导率的土壤,这可能是由于高电导率降低了土壤 MES 的内阻^[177],即高离子强度引起的高电导率更有利于 MES 的电子传递(图 9),从而加快了 MES 中的氧化还原反应,进而提高了对抗生素的降解效能. 然而,以四环素为代表的抗生素易被土壤吸附,这很大程度地限制了它们在土壤 MES 中的降解速率^[176]. 因此,如何降低四环素在土壤中的吸附非常

关键,可以适当添加一些无害的物质与四环素竞争吸附位点.但需要注意的是,一旦四环素被解吸出来,很可能会增加它向其它环境介质的迁移风险,因此考虑这条建议的前提是原位修复.磷是植物和微生物生长发育必需的营养元素,在正常土壤 pH 条件下,无机磷在土壤中主要以磷酸根离子或磷酸氢根离子存在.有研究发现,磷酸根离子的存在会与草甘膦竞争吸附位点,从而抑制草甘膦在土壤中的吸附^[178].在土壤 MES 运行过程中加入磷可与四环素竞争吸附,从而提高四环素在土壤中的生物可利用性(图 10),最终有利于 MES 对四环素的降解(去除率较对照提高了 25%)^[51,179].同时土壤 MES 具有快速降解抗生素的能力,在实验第 7 d,四环素在闭路处理中的降解率已经达到 42%~50%,较相应的开路处理显著提高 42%~67% ($P < 0.05$),而在无电极处理中的降解效率仅为 6%^[179].不仅如此,即使针对混合抗生素污染的土壤,该系统依旧具有很好的修复效果^[180].此外,相比于研究较为成熟的阳极氧化降解, MES 阴极近年来也受到了关注,且对抗生素的去除效率与阳极相近^[179,181],这可能是阴极的供电子系统促进了功能菌的还原降解^[182].

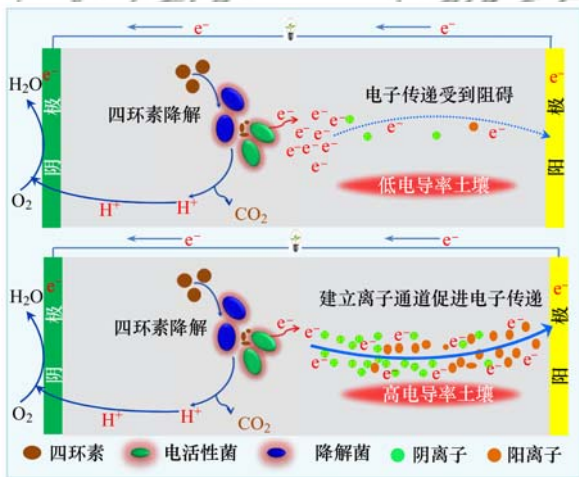


图 9 电子在高、低电导率土壤 MES 中的模拟传递过程

Fig. 9 Simulated electron transfer process in MES filled with high or low conductivity soils

微生物是 MES 运行过程的反应主体,深入研究 MES 中参与抗生素降解的功能菌群是阐明其生物降解机制和 ARGs 扩增与传播机制的重要基础.在 MES 运行过程中,功能微生物之间的协作、竞争和拮抗等作用构建了一个密切的代谢关系网络.例如,在 MES 的产电过程中,发酵细菌 *Anaerolineaceae* sp. 可将小分子糖类分解为短链脂肪酸和 H_2 ,电活性微生物 *Geobacter* sp. 则可以利用短链脂肪酸如乙酸盐或丙酸盐等作为电子供体完成其生长代谢过程^[183,184].对于抗生素的生物降解,需要行使不同功

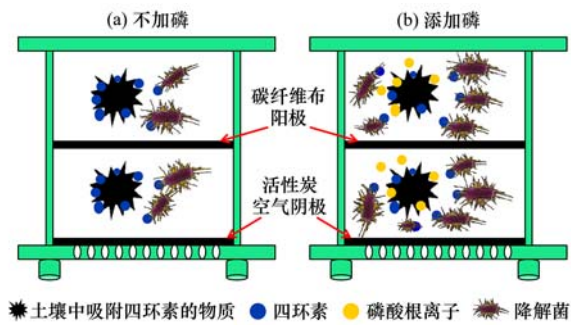


图 10 土壤 MES 中添加磷对四环素降解的影响

Fig. 10 Effect of phosphorus addition in soil MES on tetracycline degradation

能微生物之间的相互协作完成降解过程.通过 Network 关联分析发现土壤 MES 可以提供生物电流刺激系统中的微生物代谢活性,强化细菌、真菌和古菌之间的协作关系,建立一个涉及产电、降解和氮转化在内的功能微生物代谢网络(图 11),从而促进对抗生素的降解效率^[181].土壤 MES 在降解抗生素的过程中,由某些功能菌分泌的酶起着重要的作用^[185].借助宏基因组技术在土壤 MES 阴极部位分辨出 4 个潜在的功能酶基因,分别为电子传递黄素蛋白 2,3-氧化还原酶(EC 1.3.8.7)、富马酸还原酶(EC 1.3.5.4)、多酚氧化酶(EC 1.10.3.-)和醌氧化酶(EC 1.10.3.14)^[179].进一步发现, *Gemmatimonadetes bacterium* SCN 70-22、*Azohydromonas australica* 和 *Steroidobacter denitrificans* 很可能分泌 EC1.3.8.7 编码的电子转移黄素蛋白 2,3-氧化还原酶,以提高电子从阴极到亲电菌的转移效率. *Gemmatirosa kalamazoonesis* 可能分泌 EC 1.10.3.14 编码的醌氧化酶,以加速土壤 MES 阴极的胞外电子传递过程,从而提高了四环素的生物降解速率^[179].此外,由 EC 1.10.3-编码的多酚氧化酶可能与四环素在 MES 中的降解有关.

尽管 Song 等^[186]研究发现 MES 在降解抗生素的过程中会加剧 ARGs 对生态环境的潜在威胁,但多数研究均表明 MES 能够有效控制 ARGs 的扩增与传播^[187~189].Yan 等^[171]结合交叉领域的相关研究基于以下两方面原因肯定了该系统在消减 ARGs 方面的应用前景:第一,该系统是在厌氧生物技术的基础上构造的,而先前研究证实了厌氧处理工艺可以有效去除 ARGs.第二,该系统较传统厌氧处理方法产生极低的剩余生物量,这在一定程度上降低了 ARGs 扩增和传播的载体.通过构建土壤 MES 进一步明确了该系统对 ARGs 的消减机制:一方面, MES 给予微生物电流刺激调控并重塑了土壤微生物的种间关系,建立了一个不同于对照处理的微生物代谢网络,降低了耐药菌的数量^[181];另一方面, MES 具

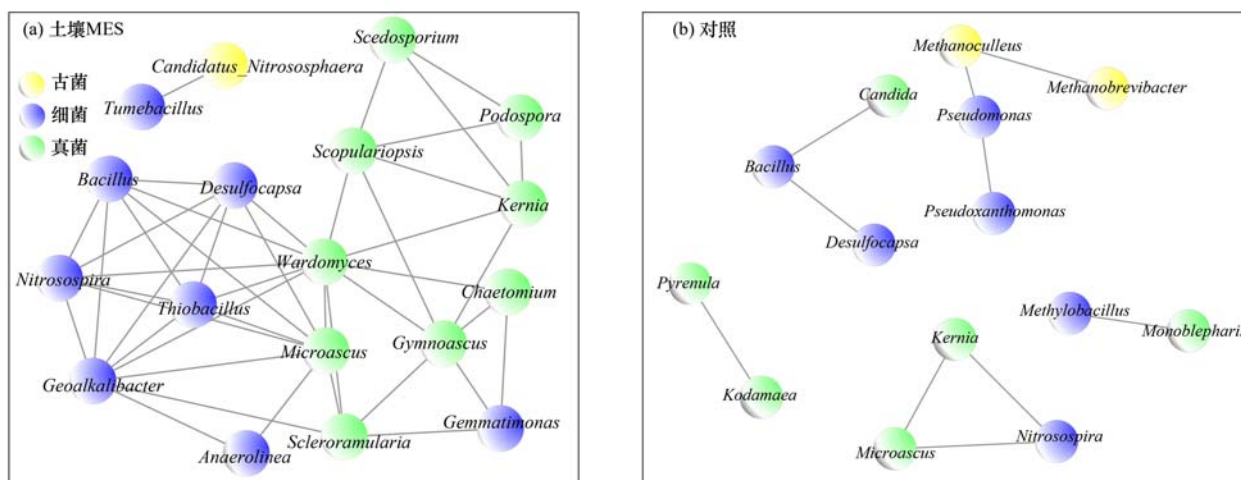


图 11 土壤 MES 对微生物种间关系的影响

Fig. 11 Effect of soil MES on interspecific relationship of microorganisms

有快速降解土壤中抗生素的能力,从而减缓了抗生素对微生物的选择性压力,进而降低了 ARGs 的扩增与传播风险^[179]. Chen 等^[190]通过综述前人研究提出在 MES 中加入某些添加剂,包括有机质、亚硝酸盐、过氧化钙、铜、表面活性剂等,可增强对 ARGs 的消减.例如,将过氧化钙添加至 MES 中不仅可加速胞外电子转移过程从而提高对抗生素的降解效率,而且过氧化钙会破坏 DNA,并减少一些酶和蛋白质,最终促进对 ARGs 的消减^[191].

4 展望

(1) 存在于土壤中的抗生素和 ARGs 不仅会给土壤环境带来威胁,也会通过迁移扩散等途径增加其对其他环境介质的污染风险.目前尚缺乏土壤中抗生素和 ARGs 在全国尺度上连续多年的追踪调查,增加相关研究将为揭示抗生素污染特征和制定相关政策法规等提供更多的科学依据.尽管经过对现有文献数据的统计分析发现,土壤中抗生素含量随时间推移呈现一定的下降趋势,但依旧不能放松对抗生素使用的管控和污染修复技术的研发.

(2) 生物修复是基于自然环境本身存在的生物资源来实现污染治理,针对抗生素污染土壤的生物修复技术目前还不成熟,尚有一些不足之处需要完善:第一,植物和动物可通过积累或富集等方式吸收土壤中的抗生素,对于吸收在生物体内的抗生素如何处理需进一步完善.第二,考虑到高含量抗生素的生物体自身的毒害,对于养殖场附近污染土壤的修复目前适用性低.第三,尽管高效降解菌在人工控制实验中对抗生素的降解效果较好,但实际环境的复杂性限制了其原有的修复效率,另外如何在保证生物安全的同时提高降解菌在污染土壤中去除效率值

得深思.

(3) 虽然 MES 在废水处理领域已经初步实现从实验模拟到实际应用的跨越,但在污染土壤修复方面距离实际应用仍存在一定的距离.尽管如此,土壤 MES 依旧具有巨大的研究潜力,未来主要从以下几方面着手:第一, MES 的产电特性可为全球能源转型提供新思路,然而其产电性能目前受到诸多因素的限制,在我国实现“碳达峰、碳中和”目标的背景下,急需研发新能源技术来改变由传统化石能源发电的现状,因此提升 MES 的产电性能是当前亟待解决的问题之一.第二,迄今为止,多数相关研究主要通过统计学手段结合现有文献对降解功能菌与 ARGs 宿主菌进行推测,然而这些降解菌是否真正起作用,究竟是哪些功能基因起到关键作用,如何通过调控关键功能基因进一步促进抗生素在 MES 中的降解有待深入研究.第三,污染代谢产物在不同降解菌间的跨膜转运是耗能过程,这将在一定程度上制约微生物对有机污染物的降解效能.重组不同菌体中的降解功能基因,构建具有高效降解能力的基因工程菌,采用微生物电化学技术对其降解效能作进一步强化,将有助于定向增强污染物的生物降解效率.第四,由于土壤 MES 需要淹水条件,且土壤巨大的内阻降低了电子传递效率,从而限制了对抗生素等污染物的修复效果,因此土壤 MES 目前较难应用于原位土壤修复,未来应努力打破淹水条件对土壤 MES 的限制,着重探索提高土壤电子传递效率的方法,如添加生物质炭,或者在不影响土壤质量的前提下适当提高土壤电导率等.第五, MES 对 ARGs 的去除机制尚无确切定论,仍需进行深入研究.

5 结论

(1) 我国关于土壤中抗生素污染状况调查的相

关研究最初出现于2008年左右.从近15年的调查结果来看,全国各地土壤均不同程度地受到了抗生素及其抗性基因的污染,尤其是广东、福建和山东地区.

(2) 抗生素对蔬菜地的污染通常较大田等其它生产类型的土壤更为严重,且四环素类和喹诺酮类抗生素主要残留于表层土壤,而磺胺类和大环内酯类抗生素因其自身较强的迁移能力常见于深层土壤甚至水环境中.由于不同季节的气候条件存在差异,导致土壤中抗生素的残余量在秋冬季高于夏季.另外,在关注土壤中抗生素的同时,重金属、微塑料及杀虫剂等对ARGs在土壤中扩增与传播的影响也不容忽视.

(2) 植物通过积累、固定、挥发、降解和矿化等方式或者联合微生物修复抗生素污染土壤.蚯蚓常被用于动物修复,通过将抗生素在肠道内部降解、富集及强化“蚓触圈”土壤降解菌活性以实现其对污染土壤的修复.耐药细菌是抗生素的主要降解菌,将降解菌株固定于载体有利于其在污染土壤中的定殖与修复.

(3) 土壤MES是一种绿色高效的修复技术,该系统可以提供生物电流刺激土壤微生物的代谢活性、重构种间关系、调控功能酶的分泌,不仅增强了微生物对抗生素的降解效率,而且也有效控制了ARGs的扩增传播风险.

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