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卤代有机污染物对剩余污泥厌氧消化产能的影响及机制

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摘要: 卤代有机化合物(HOPs)作为重要的工业化学品,在加工、运输和使用过程中被大量释放到环境中,最终富集在污水处理厂的剩余污泥中.厌氧消化是剩余污泥资源化处置的重要手段,可转化有机物为挥发性脂肪酸和甲烷等高价产品.然而, HOPs 对剩余污泥厌氧消化产能的影响及其作用机制尚未得到系统阐述,因此,采用系统文献综述的方法,通过系统文献综述,分析了 HOPs 对污泥厌氧消化产甲烷效能、关键过程及微生物群落的影响规律.结果表明,多数 HOPs 因具有较高毒性而抑制厌氧消化关键阶段,导致甲烷产量降低;部分低毒性 HOPs 则表现出低浓度促进、高浓度抑制的"激素效应".HOPs 主要通过影响增溶、水解、产酸和产甲烷四个关键阶段来调控厌氧消化效能,其中对产酸和产甲烷阶段的影响最为显著.HOPs 可通过改变微生物群落结构、抑制关键酶活性和干扰代谢途径等方式影响厌氧微生物的功能.本研究揭示了 HOPs 对污泥厌氧消化的影响机制,并针对当前研究不足提出了未来研究方向,为剩余污泥的资源化利用和安全处置提供理论基础.

关键词: 卤代有机污染物; 剩余污泥; 厌氧消化; 微生物

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Effect of halogenated organic pollutants on anaerobic digestion capacity of waste activated sludge and its mechanisms. ZHAO Zheng-zheng¹, WU Yang¹, ZHENG Xiong^{1,2,3,*}, LONG Min¹, CHEN Yin-guang^{1,3} (1.State Key Laboratory of Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Tongji University, Shanghai 200092, China; 2.Key Laboratory of Yangtze River Water Environment, College of Environmental Science and Engineering, Tongji University, Shanghai 200092, China; 3.Shanghai Institute of Pollution Control and Ecological Security, Shanghai 200092, China). *China Environmental Science*, 2025,45(2): 892~901

Abstract: Halogenated organic compounds (HOPs), as important industrial chemicals, are extensively released into the environment during their production, transportation, and usage, ultimately accumulating in waste activated sludge (WAS) from wastewater treatment plants. Anaerobic digestion is a crucial approach for resource recovery from WAS, converting organic matter into valuable products such as volatile fatty acids and methane. However, the effects of HOPs on the anaerobic digestion capacity of WAS and their underlying mechanisms have not been systematically elucidated. Through a comprehensive literature review, this study analyzed the impacts of HOPs on methane production efficiency, key processes, and microbial communities during sludge anaerobic digestion. The results revealed that most HOPs inhibit key stages of anaerobic digestion due to their high toxicity, leading to reduced methane yield, while some low-toxicity HOPs exhibit a "hormesis effect" with promotion at low concentrations and inhibition at high concentrations. HOPs primarily regulate anaerobic digestion efficiency by affecting four critical stages: solubilization, hydrolysis, acidogenesis, and methanogenesis, with the most significant impacts on acidogenesis and methanogenesis. HOPs can influence the function of anaerobic microorganisms by altering microbial community structure, inhibiting key enzyme activities, and interfering with metabolic pathways. This study unveils the mechanisms of HOPs' effects on sludge anaerobic digestion and proposes future research directions addressing current knowledge gaps, providing a theoretical foundation for resource recovery and safe disposal of WAS.

Key words: halogenated organic pollutants; waste activated sludge; anaerobic digestion; microorganism

卤代有机化合物是指一个或多个卤素原子替代了氢原子的有机化合物,在工业生产中常被用作润滑剂、增塑剂和添加剂^[1-2].在生产、运输和使用过程中,大量卤代有机污染物(HOPs)被释放到环境

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中^[3-4],由于其种类多、性质多样、基质复杂和潜在危害大等特点^[5-6],引起了广泛关注^[7-8].例如,在某些地区,废水中对氯酚(4-CP)的浓度已达到 200.0mg/L^[9],其可通过食物链在人体内积累,对生态环境产生严重风险^[10-11].作为污染物重要的“汇”,常规污水处理厂对 HOPs 处理效果有限,导致超过 50%的 HOPs 通过吸附、降解等作用转移至剩余污泥中^[12-13],对污泥的资源化利用构成潜在威胁.

剩余污泥除了“污染”属性,同时兼具“资源”属性^[14-15].截至 2022 年,我国剩余污泥年产量超 1300 万 t(含水率按 80%计).剩余污泥中含有大量有机物(如蛋白质、多糖、脂类),其含量占 50%~70%^[16],具有较高的生物能源回收潜力.厌氧消化是剩余污泥资源化处置的重要手段^[17-18],可以合成 VFAs 和甲烷等 high 价值产品^[19-20],具有较高的经济可行性和处理有效性^[21-22].厌氧消化是一个微生物主导的过程,其水解、产酸、产甲烷阶段依赖于各类微生物的协同作用^[23-25].作为外源污染物,HOPs 可能影响厌氧消化过程中功能微生物的活性及功能表达,从而影响厌氧消化的效能^[26].然而,目前针对 HOPs 对剩余污泥厌氧消化产能的影响及其作用机制研究尚不明晰.

本文首先探究 HOPs 对污泥厌氧消化产甲烷效能的影响,其次探讨了 HOPs 对污泥厌氧消化关键过程(增溶、水解、产酸和产甲烷)的影响,随后研究了 HOPs 对厌氧消化微生物群落及功能的影响,最后揭示了 HOPs 对污泥厌氧消化效能的影响及作用机制,以期为剩余污泥高值化利用提供理论支撑和技术指导.

1 HOPs 对剩余污泥厌氧消化效能影响

HOPs 主要分为氟代、氯代和溴代有机污染物.不同种类的 HOPs 对剩余污泥厌氧消化产甲烷的影响各不相同.研究表明,多数 HOPs 对厌氧消化产甲烷具有抑制作用;但由于微生物对不同污染物的耐受性不同,部分 HOPs 表现出类似于低浓度促进、高浓度抑制的“激素效应”.代表性 HOPs 对污泥厌氧消化产甲烷效能的影响如表 1 所示.

氟代有机污染物对污泥厌氧消化产甲烷效能影响的研究主要集中在全氟和多氟烷基物质(PFAS)及部分含氟药物.PFAS 是一种非聚合含氟碳链的合成化学品,广泛应用于冰箱、消防泡沫、纺织品和食

品包装等各个行业^[27-28].其中,全氟辛酸(PFOA)和全氟辛烷磺酸(PFOS)是最具代表性的两种,其在固体和水生基质中的浓度可达 mg/kg 和 mg/L 级别^[29].研究表明,PFOA 和 PFOS 均对污泥厌氧消化产甲烷有抑制作用,且 PFOA 的抑制作用更加显著^[30],这与它们对污泥厌氧消化过程的毒性效应机制有关:PFOA 主要通过其毒性抑制微生物,影响细菌和古菌群落结构,而 PFOS 则通过其毒性和官能团(如磺酸)引起细菌和古菌群落结构的变化^[31-32].例如,在 Choi 等^[31]的研究中,当浓度达到 100mg/L 时,PFOS 对产甲烷的抑制率为 7.4%,而 PFOA 的抑制率高达 34.2%.此外,氟代有机污染物中还包含环丙沙星(CIP)、诺氟沙星(NOR)和氟尿嘧啶(FLU)等药物.这些含氟药物对于污泥厌氧消化产甲烷效能的影响存在“激素效应”^[33].例如,Tang 等^[34]研究发现,随着 CIP 浓度从 0.5mg/L 提升至 2mg/L 时,其对产甲烷效能的影响从提高 22.9%转为抑制 34.3%.

氯代有机污染物对产甲烷效能影响的研究主要集中于含氯内分泌干扰物、氯化脂肪烃和氯酚.三氯生(TCS)是一种典型的内分泌干扰物,广泛应用于药物和个人护理用品的生产^[35].研究表明,TCS 对污泥厌氧消化产甲烷效能的影响存在“激素效应”.当污泥中 TCS 浓度为 200mg/kg 时,厌氧体系甲烷产率提高 17.3%;而当浓度增加为 1500mg/kg 时,甲烷积累量降低 13.2%^[36-37].氯化脂肪烃是另一类重要的氯化有机污染物,常见于含水层、土壤和废水中,通常在污泥厌氧消化过程中会抑制甲烷的生成.Yu 等^[38]研究表明,4 种常见的多氯脂肪烃(二氯甲烷、氯仿、三氯乙烯和过氯乙烯)对甲烷生成有不同程度的抑制作用.氯仿对甲烷生成的抑制作用最强,0.09mg/L 的氯仿溶液完全抑制了甲烷的生成;而 3.9mg/L 的二氯甲烷和 18mg/L 的三氯乙烯也表现出对一定的抑制作用,14.5mg/L 的全氯乙烯则无抑制作用.进一步研究发现,多氯脂肪烃的抑制作用与其氯化程度和分子结构相关^[39].除此之外,氯酚也是一类重要含氯化合物,常用于木材防腐和杀虫剂^[40].氯酚类物质对产甲烷效能有显著的抑制作用,尤其是分子中氯原子位于芳香环上的位置更容易与微生物发生相互作用.Puyol 等^[41]研究表明,相比于 2,4-二氯苯酚(2,4-DCP)、2,4,6-三氯苯酚(2,4,6-TCP)、五氯苯酚(PCP)对污泥厌氧消化产甲烷的抑

制作用最强,8mg/L 的 PCP 可抑制 50%的甲烷生成.

溴代有机污染物对污泥厌氧消化产甲烷效能影响的研究主要集中在溴代阻燃剂和溴代杀菌剂.多溴联苯醚(PBDE)和四溴双酚 A(TBBPA)是两种广泛使用的阻燃剂.研究表明,8mg/kg PBDE 可抑制 25%甲烷产量^[42];而 0.1mg/L TBBPA 则可提高 8.79%的甲烷产率;但当浓度升高至 4.0mg/L 时,甲烷产率下降 28.9%^[43].苯扎溴铵(BK)作为一种典型的表面活性剂类杀菌剂,在浓度达到 8.69mg/kg VSS 时,抑

制 17.2%的甲烷生成^[44].除此之外,十溴联苯醚、二溴二苯醚、六溴环十二烷等溴代有机污染物也对厌氧消化产甲烷效能产生抑制^[45-46].

综上所述,不同种类 HOPs 对污泥厌氧消化产能的影响各异,多数 HOPs 对产甲烷效能有显著抑制作用,部分低毒性 HOPs 对呈现低浓度促进、高浓度机制的“激素效应”.因此,进一步探究不同污染物对产甲烷效能的影响机制,对于深入理解 HOPs 对厌氧消化产甲烷效能的差异具有重要意义.

表 1 HOPs 对污泥厌氧消化产甲烷效能的影响

Table 1 Effects of HOPs on methane production efficiency during sludge anaerobic digestion

有机物类别	名称	浓度	温度(°C)	培养天数(d)	甲烷积累量(mL)	促进/抑制率(%)	参考文献
氟代有机物	PFOA	0.1mg/L	37	28	314.7	-3.85	[31]
	PFOA	100mg/L	37	28	217.8	-34.2	[31]
	PFOS	0.1mg/L	37	28	317.2	-3.91	[31]
	PFOS	100mg/L	37	28	305.7	-7.40	[31]
	PFOA	60mg/g	37±1	15	104.2	-19.2	[32]
	PFOA	3.0mg/L	-	8	-	-19.7	[33]
	PFOA	170mg/kg	37±1	60	159.9	-18.9	[47]
	CIP	0.5mg/L	36±1	40	407.44	+23.0	[34]
	CIP	2mg/L	36±1	40	217.64	-34.3	[34]
	CIP	50mg/L	35±1	45	66.7	-42.2	[48]
	NOR	1.7mg/L	35±1	37	6.7	+26.4	[49]
	FLU	5mg/L	37	20	-	无影响	[50]
FLU	50mg/L	37	20	-	-80.0	[50]	
氯代有机物	TCS	200mg/kg	35±1	-	74.7	+17.3	[37]
	TCS	1500mg/kg	35±2	60	94.1	-13.2	[36]
	BmimCl	20mg/L	35±1	24	288.95	-7.00	[51]
	PCP	8mg/L	30±1	25	-	-50.0	[41]
溴代有机物	PBDE	8mg/kg	37±1	-	161.5	-25.00	[52]
	TBBPA	0.1mg/L	35±1	60	259.0	+8.79	[43]
	TBBPA	4.0mg/L	35±1	60	171.5	-29.0	[43]
	BK	8.69mg/g	33±2	33	615.3	-17.2	[44]

2 HOPs 对剩余污泥厌氧消化关键过程的影响机制

2.1 HOPs 对增溶阶段的影响

剩余污泥是一个复杂的生物系统,含有丰富的可降解有机物,其中大部分以非溶解/颗粒形式存在^[42].厌氧消化的第一步是增溶阶段,将颗粒有机化合物转化为可溶性底物^[25].蛋白质和多糖是污泥的重要组成部分,约 61.5%的 COD 来源于这两类化合物^[53].因此 HOPs 对增溶作用的影响可以通过可溶性有机底物(即 SCOD、可溶性蛋白和可溶性多糖)的浓度来评估.

总体来说,绝大部分 HOPs 对于增溶作用的影响并不明显,这与增溶作用是非微生物过程有关^[54].研究表明,PFOA、NOR、5-FLU 等氟代有机污染物对增溶阶段没有显著影响,可能相较其他 HOPs,这些污染物的化学结构较为简单,不会对微生物产生吸附或毒性^[32,49-50].然而,部分研究发现,某些 HOPs 对轻微抑制增溶作用.这类 HOPs 具有较强毒性,会刺激微生物分泌胞外多聚物质(EPS),增加传质阻力^[47,55],从而抑制有机物的释放^[56].例如,当得克隆(DPs)浓度从 30.3mg/kg TSS 增加到 3034.1mg/kg TSS 时,可溶性蛋白浓度从 1129.8 降低到 870.1mg/

L^[57].高浓度的 DP_s 减少了可溶性有机底物含量,抑制了污泥厌氧消化的增溶阶段.一旦增溶作用停滞,可用于微生物合成甲烷的有机底物就会减少,这是 HOP_s 降低污泥厌氧消化产甲烷潜力的原因之一^[33].此外,部分 HOP_s 对增溶过程具有促进作用.这类 HOP_s 由于具有较强活性,能够破坏 EPS 的结构,促进有机底物的释放^[58].例如,Zou 等^[37]研究表明,低浓度的 TCS 促进了污泥中可溶性有机物的释放.Wang 等^[59]发现投加 TCC 有利于污泥的增溶,随着 TCC 浓度从 0 增加至 1403mg/kg TSS,厌氧体系中 SCOD 的浓度从 7689mg/L 提高 9250mg/L,表明更多的颗粒有机物转化为可溶性底物.不同 HOP_s 对增溶阶段的影响不同,这主要与不同物质的特性有关.

2.2 HOP_s 对水解阶段的影响

厌氧消化的水解过程是指在缺氧条件下,有机物质被厌氧微生物分解为简单的有机化合物^[60].在这一过程中,可溶性蛋白和多糖被进一步分解^[61],转化为小分子有机物,如氨基酸和葡萄糖^[62].厌氧消化是由多种酶驱动的综合微生物生化过程,蛋白酶和 α -葡萄糖苷酶是水解阶段的关键酶,HOP_s 对水解阶段的影响与其对酶活性的影响有关^[42].

总体来说,HOP_s 对污泥厌氧消化水解阶段影响较小.Xie 等^[33]和 Jiao 等^[47]的研究均证明了 PFOA 对污泥厌氧消化水解阶段无显著影响.Wang 等^[59]的研究中,蛋白酶在对照组和添加 TCC 反应器之间的活性无明显变化,即 TCC 对牛血清白蛋白的降解几乎没有影响,这与实验结果中牛血清蛋白浓度未变化相一致.同样,Jiao 等^[47]的研究也发现 PFOA 对蛋白酶和 α -葡萄糖苷酶活性无显著影响.然而,部分研究发现 HOP_s 对水解阶段有抑制作用.这类 HOP_s 对参与水解过程的微生物产生毒性作用,抑制酶的活性,从而减缓或阻碍水解过程.Wang 等^[36]发现高浓度的 TCS 暴露(超过 550mg TCS/kg TSS)对水解有一定的毒性.Zhao 等^[52]研究 PBDE 对污泥厌氧消化过程的影响时发现,PBDE 降低了牛血清蛋白和葡聚糖的降解率,分别为对照的 75.8%和 84.4%.

综上所述,HOP_s 对污泥厌氧消化水解阶段的影响总体较小.毒性较高的 HOP_s 通过影响参与水解过程的微生物酶活性,抑制水解过程.而低毒性 HOP_s 在低浓度时可增强微生物功能来抵御外界危害,但当污染物浓度过高时,其功能则会受到抑制.

2.3 HOP_s 对产酸阶段的影响

厌氧消化的产酸阶段是决定生物质转化代谢途径的关键阶段^[63].在该阶段中酸的种类和浓度的变化与微生物群落的种群结构及代谢途径的改变有关^[64].这些产物是产甲烷阶段的主要底物,因此产酸阶段的效率直接影响到甲烷的产量和速率^[65,66].

总体来说,大部分 HOP_s 会抑制污泥厌氧消化产酸阶段.Zhang 等^[57]的研究表明,DP_s 虽然没有影响挥发性脂肪酸(VFAs)的组成,但是 VFAs 的最大积累量仅为 27.8mg COD/g VSS,为空白组最大产量的 0.13 倍.Wang 等^[36]发现,TCS 暴露水平越高,其对酸化过程的抑制作用越大.当 TCS 暴露水平增加至 1200mg/kg TSS 时,产酸相关微生物的相对活性下降了 21.5%.该现象主要由于 HOP_s 的存在会干扰微生物的正常代谢途径,导致产酸过程中所需酶活性降低或代谢中间产物积累,从而抑制酸的产生.乙酸激酶(AK)和丁酸激酶(BK)是酸化过程中的两种主要酶,它们负责水解产物生成乙酸和丁酸.Jiao 等^[47]研究发现,PFOA 的存在显著降低了 AK 和 BK 的活性,PFOA 占据酶的活性位点,诱导氧化应激,导致蛋白酶合成障碍,抑制了产酸酶活性.Liang 等^[63]研究也表明,在 100mg/L PFOA 条件下,葡萄糖激酶和丙酮酸激酶的丰度分别降低 8%和 28.1%,与 PFOA 对产酸阶段抑制的实验结果一致.

此外,部分 HOP_s 在低浓度或特定类型下可以促进产酸阶段.微生物通过基因表达调整或代谢途径的改变来适应这些化合物的存在,从而更有效地利用底物进行产酸.Zou 等^[37]发现 200mg/kg TSS 的 TCS 作用下,AK 的活性是对照反应器的 2.66 倍,显著增加了污泥厌氧消化期间 VFAs 的积累,其中乙酸是增加总 VFAs 产量的主要贡献者.TCS 对酸化的促进作用强于产甲烷作用,使 VFA 的生成速率超过消耗速率,导致 VFAs 的积累.此外,Wang 等^[59,67]研究发现,TCC 存在下,AK 和 BK 活性分别增强 34.7%和 10.8%,说明 TCC 暴露可显著促进酸化相关微生物的相对活性.对于一些毒性较低的 HOP_s,产酸菌群能够适应其存在,产生解毒酶或抵抗机制(如泵出系统),从而维持代谢功能.例如,Hoshiko 等^[50]研究发现,FLU 不会对污泥的酸化过程产生影响.

综上所述,HOP_s 对污泥厌氧消化产酸阶段的影响取决于其类型和浓度.高浓度的 HOP_s 通常抑制产

酸过程,干扰微生物的代谢途径和酶活性,从而降低产酸效率.然而,低浓度或特定类型的 HOPs 可能通过微生物的适应性改变和代谢途径调整来提高产酸效率.

2.4 HOPs 对产甲烷阶段的影响

甲烷生成是污泥厌氧消化的最后一步,不同的 HOPs 对该阶段的影响虽有所不同,但机制相似.总体来说,HOPs 会对甲烷菌产生毒性,抑制其功能,导致产甲烷相关酶(如 F420)活性下降.Wang 等^[32]的研究表明,PFOA 浓度升高(3~60 $\mu\text{g/g}$ TS)导致甲烷产量较对照下降 11.1~19.2%,辅酶 F420 的活性也有所下降.同时,HOPs 可能通过影响污泥厌氧消化过程的产物组成,进而影响甲烷的生成^[66].Tang 等^[34]研究发

现,低剂量的 CIP 有利于产酸菌的生长,为产甲烷菌的生长提供了更多的碳源,促进了甲烷的产生.而在 Zou 等^[37]的研究中,在 200mg/kg TSS TCS 的作用下,AK 的活性是对照组的 2.66 倍,辅酶 F420 的活性比对照组提高了 1.17 倍.TCS 对 AK 活性的促进作用大于 F420,导致 TCS 对酸化的促进作用强于产甲烷作用.

综上所述,由于 HOPs 自身的毒性,大多数 HOPs 对产甲烷阶段产生抑制作用.但是产甲烷阶段的最终影响取决于污泥厌氧消化四阶段强弱的权衡.某些低毒性或低浓度 HOPs 对前三阶段的促进作用强于对产甲烷阶段的抑制作用,也可能导致最终的甲烷产量的提升.

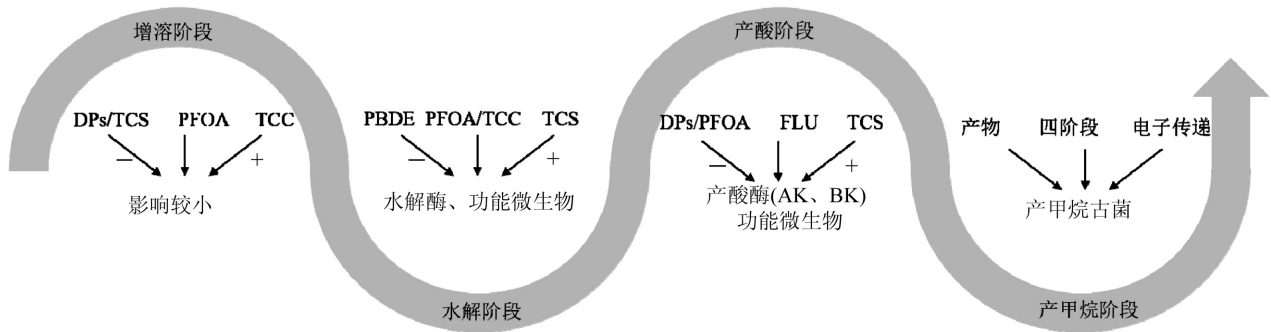


图 1 HOPs 对污泥厌氧消化关键过程影响

Fig.1 Effect of HOPs impacts on the crucial stages during sludge anaerobic digestion

3 HOPs 对厌氧消化微生物群落及功能的影响机制

厌氧消化是一种复杂的生物转化过程,涉及多种微生物,包括细菌和古菌,这些微生物在不同的厌氧消化阶段发挥关键作用^[68].表 2 展示了参与污泥厌氧消化过程各个阶段的微生物.研究表明,HOPs 对厌氧消化相关微生物的影响,主要表现在其对微生物功能、活性、结构及相对丰度的影响^[69].这些影响贯穿于厌氧消化的各个生物过程.由于 HOPs 具有较高极性和易发生亲核取代反应的特点,它们可以直接影响污泥中参与厌氧消化过程的微生物活性.一些对 HOPs 较为敏感的微生物可能会被抑制,而一些能够耐受或部分代谢这些污染物的微生物则可能成为优势种.这种微生物群落结构的改变可能会减缓或促进厌氧消化的不同阶段,从而影响整个厌氧消化过程的效率和稳定性.

3.1 HOPs 对水解阶段微生物的影响

HOPs 对水解阶段相关微生物的丰度和结构产生显著影响,这些变化会进一步影响微生物的功能^[51].多数 HOPs 会降低微生物的丰度,抑制水解酶的活性,进而减缓复杂有机物向简单小分子的转化,影响整个厌氧消化过程的效率和稳定性.

水解过程中,Proteobacteria 和 Bacteroidetes 在门水平上占主导作用^[71-72].Wang 等^[32]研究发现,在 60 $\mu\text{g/g}$ TSS PFOA 暴露下,Proteobacteria 和 Bacteroidetes 丰度分别显著降低了 9.2%和 37.9%,这与 PFOA 对水解阶段产生抑制的结果一致.Wang 等^[36]还提出,TCS 显著降低了 Firmicutes 的相对丰度,但对其他厌氧微生物的毒性较小,有利于其他种群的生长,增加了微生物群落的多样性,例如 TCS 增加了负责有机物分解的放线菌的相对丰度.此外,一些毒性较低的 HOPs 会提升水解微生物的丰度.某些微生物可能对这类 HOPs 具有耐受性或降解能力,从而在这种环境下占据竞争优势,导致这些耐受性或降

解能力强的水解相关微生物丰度增加^[73]。张静^[74]提出, DPs 的存在增加了 *Proteobacteria* 的相对丰度,当 DPs 含量为(3034.1±101.7) mg/kg TSS 时, *Proteobacteria* 的相对丰度较空白组提高了 15.3%,进一步证明了 DPs 促进有机物分解的效果。此外, *Burkholderiales* 和 *Rhodobacterales* 能够刺激溶解性有机物的释放,在

0.5mg/L 的 CIP 条件更有利于这些功能菌的富集^[22], 从而促进水解过程。

综上所述,多数 HOPs 对水解阶段相关微生物的功能产生抑制作用,但由于某些微生物对外源污染物具有耐受性,这些微生物的丰度可能会增加,从而在特定条件下促进水解功能。

表 2 参与污泥厌氧消化各过程的代表性微生物^[70]

Table 2 Representative microorganisms involved in the critical stages during sludge anaerobic digestion^[70]

过程	微生物域	微生物属	微生物物种
水解	细菌	<i>Acetivibrio, Aminobacterium, Aminomonas, Anaeromusa, Anaerospaera</i>	<i>Pseudomonas</i>
		<i>Bacillus, Bacteroides, Bifidobacterium, Butyrivibrio</i>	<i>mendocina</i>
酸化	细菌	<i>Caldanaerobacter, Caldicellulosiruptor, Campylobacter, Cellulomonas, Clostridium, Devosia, Spiroquetas, Eubacterium,</i>	<i>Bacillus halodurans</i>
		<i>Fervidobacterium, Fibrobacter, Fusobacterium,</i>	<i>Clostridium</i>
		<i>Gelria, Gracilibacter, Halocella, Lactobacillus</i>	<i>hastiforme</i>
		<i>Paludibacter, Peptococcus, Peptoniphilus, Proteiniborus, Pseudomonas, Psychrobacter,</i>	<i>Gracilibacter</i>
		<i>Ralstonia, Ruminoclostridium, Ruminococcus</i>	<i>thermotolerans</i>
		<i>Selenomonas, Shewanella, Sporanaerobacter, Streptococcus, Streptomyces</i>	<i>Thermomonas</i>
		<i>Thermanaerovibrio, Thermomonas, Thermomonospora, Thermotoga, Treponema, Trichococcus</i>	<i>haemolytica</i>
			<i>Moorella</i>
			<i>thermoacetica</i>
			<i>Desulfotignum</i>
产甲烷	古菌	<i>Acetobacterium, Clostridium, Desulfotignum, Eubacterium, Holophaga, Moorella, Ruminococcus, Sporomusa, Thermoanaerobacter, Treponema</i>	<i>phosphitoxidans</i>
			<i>Holophaga foetida</i>
		<i>Methanobrevibacter smithii</i>	
		<i>Methanobacterium, Methanobrevibacter, Methanococcus, Methanoculleus, Methanosaeta, Methanomicrobium, Methanosarcina, Methanospirillum, Methanothermobacter</i>	<i>Methanobrevibacter arboriphilus</i>
			<i>Methanococcus vannielii</i>

3.2 HOPs 对产酸阶段微生物的影响

大多数 HOPs 对厌氧消化产酸微生物具有抑制作用。例如,当 TCS 暴露水平增加到 1200mg/kg TSS 时,产酸微生物的相对活性下降了 21.5%^[36]。同时 CIP 通过与脱氧核糖核酸旋转酶结合抑制产酸微生物的活性^[34],从而减少产酸过程中的产物,影响产甲烷阶段的效能。此外,HOPs 可能通过与微生物的代谢途径中间体反应,形成毒性更强的代谢产物,从而抑制微生物的代谢活性。这类代谢产物可能进一步干扰微生物的能量代谢途径(如甲烷生成路径或糖酵解途径),降低产酸效率。例如,在 CIP 暴露下,羟化、胺化、脱氟、脱羧和哌嗪断环过程不仅能降解 CIP,还能降低其毒性。然而,随着 CIP 用量的增加,微生物降解中间体的能力下降。此外,Zhang 等^[57]的研究发现,DPs 抑制了污泥厌氧消化过程中脂肪酸的生物

合成。微生物的活性和功能决定了其对环境变化的适应能力。在 HOPs 的影响下,部分产酸微生物能够快速抵抗有害化学物质,增加其丰度;而其他微生物由于其活性和功能受到影响,丰度下降。在产酸过程中, *Firmicutes* 和 *Actinobacteria* 在门水平上占主导作用。当 DPs 含量为 3034.1mg/kg TSS 时, *Firmicutes* 和 *Actinobacteria* 的相对丰度分别降至空白组的 72.6% 和 64.1%^[74]。在属水平上, PFOA 暴露下, VFAs 的生成者 *Chloroflexi* 的相对丰度由 18.2% 降至 10.8%^[47]。同时,与产 VFAs 相关的 *Longilinea sp.* 和 *Candidatus_Cometibacter sp.* 的相对丰度分别比对照降低了 20.5% 和 9.6%。与酸化相关的 *Clostridium* 和 *Acidibacter* 的丰度也呈现下降趋势,这与产酸量降低的实验结果相一致。在张静^[64]的研究中, DPs 浓度从 (30.3±1.2) mg/kg TSS 增加到 (3034.1±101.7)

mg/kg TSS 时,与合成 VFAs 相关的 *Mycobacterium* 相对丰度降至空白组的 46.5%。*Clostridiales*, *Bacteroidales*, *Aminicenantales* 和 *Cloacimonadales* 能够将葡萄糖转化为乙酸和 H_2/CO_2 作为中间体。随着 CIP 浓度升高,这些细菌的活性也逐渐降低。当 CIP 浓度达到 2.0mg/L 时, *Cloacimonadales* 和 *Aminicenantales* 相对丰度也有所下降^[34]。

综上所述,大多数 HOPs 对产酸微生物产生抑制作用。HOPs 通过影响产酸菌的活性、干扰代谢途径和微生物群落组成来进一步影响其产酸功能。需要进一步研究微生物和产酸过程的关系,以全面理解 HOPs 对污泥厌氧消化过程的影响

3.3 HOPs 对产甲烷阶段微生物的影响

在产甲烷过程中,产甲烷古菌占主导作用。由于其群落结构较为单一^[73],对环境条件非常敏感,其活性更容易受到外源污染物的影响^[75-76]。Wang 等^[36]研究表明,TCS 抑制了产甲烷菌的活性,进而抑制了乙酸的降解,导致甲烷产量下降,这与 Reyes-Contreras 等^[77]的结论一致。Silva 等^[28]发现,当 PFOA 和 PFOS 浓度为 0.1mg/L 时,产甲烷古菌的活性显著降低至 7.9%~14.8% 和 4.4%~6.6%;当 PFAS 浓度高于 0.1mg/L 时,这一抑制作用更为明显;而氢营养产甲烷菌不受此影响。Wang 等^[32]发现,暴露于 PFOA 中 Euryarchaeota 的相对丰度显著下降,与甲烷产量减少的实验结果一致,进一步证实了产甲烷古菌在产甲烷阶段的重要作用^[78]。*Methanosaeta sp.* 是厌氧消化过程中的优势菌,可以在厌氧消化过程中利用乙酸产生甲烷;暴露于 PFOA 后,其相对丰度减少了 20.4%。进一步研究发现,PFOA 可引起生物体氧化应激和蛋白质功能异常^[79]。PFOS 引起的氧化应激导致脂质过氧化,阻碍膜合成过程,破坏细胞膜完整性^[80]。过度的氧化应激会引发细胞失活或死亡,是 PFAS 抑制甲烷产生的主要原因之一。

综上所述,产甲烷古菌相较于其他微生物来说更容易受到 HOPs 的胁迫,导致产甲烷阶段的效率降低。

4 结语

本研究系统考察了 HOPs 对剩余污泥厌氧消化产甲烷效能、关键过程及微生物群落和功能的影响。结果表明,HOPs 对不同阶段的综合影响导致了污泥

厌氧消化产甲烷效能的变化。多数 HOPs 对厌氧消化的关键阶段产生抑制作用,这些 HOPs 通常具有较高的毒性,显著抑制微生物活性,从而降低了甲烷产量。部分毒性较低的 HOPs 因微生物对其具有耐受性,导致微生物丰度增加,活性提高,进而促进甲烷生成。然而,当污染物浓度超过微生物的耐受性时,其功能也会受到负面影响。然而,目前研究仍显不足,未来研究可集中于以下方向:

首先,作用机制仍需进一步探究。目前 HOPs 对厌氧消化微生物的毒性作用机制了解有限。特别在实际厌氧消化过程中,多种污染物通常同时存在,需研究不同污染物之间的相互作用和协同效应对产能的影响机制。因此,未来可结合分子信息学等先进技术,进一步从基因层面深入探究复合污染物对厌氧消化过程的影响机制。其次,检测和治理策略有待开发。鉴于 HOPs 对污泥厌氧消化产能的不利影响,有必要监测污水处理厂各处理工段中 HOPs 的浓度变化趋势,识别其在污水/污泥复杂介质中的迁移转化规律。如通过液相色谱-串联质谱法、酶联免疫法、毛细管电泳法、免疫分析法等先进技术进行 HOPs 检测。同时,需开发高效绿色处理技术,以减少 HOPs 在污水/污泥处理过程中的负面影响。这不仅包括改进现有处理工艺,还涉及开发新型技术和材料,以提高对 HOPs 的去除效率。主要包括热处理^[81]、光催化^[82]、高级氧化等(包括羟基自由基氧化和硫酸盐自由基氧化)^[83]。因此,应进一步改进 WAS 处理工艺,防止卤代有机物进入厌氧消化池。

最后,消化污泥后续处置值得关注。一般来说,消化污泥可用于土地利用,回收其氮、磷、钾等营养元素,从而缓解日益严重的全球营养储备减少问题。然而,由于消化污泥富含 HOPs 等新污染物,其后续土地利用可能对生态环境造成潜在威胁。因此,未来需要高度重视消化污泥的后续处置,通过识别 HOPs 等新污染物生物降解规律,研发新型安全化无害化污泥处置技术,实现消化污泥的资源化利用,如利用污泥制备生物炭及高级氧化的催化剂^[84]等。

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