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## 典型新污染物在我国垃圾填埋渗滤液中的赋存、迁移与去除\*

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**摘要** 新污染物由于具有生物毒性、环境持久性、生物累积性等特征, 对生态环境和人体健康造成潜在风险, 其环境安全问题日益受到重视. 近年来, 垃圾填埋渗滤液陆续检出各种新污染物, 使其逐渐成为新污染物不容忽视的源. 本文在梳理文献的基础上, 系统总结了药物及个人护理品、抗生素抗性基因、内分泌干扰物、全氟化合物、微塑料为主的典型新污染物在我国垃圾填埋渗滤液中的赋存特征, 探讨了影响其赋存水平的因素, 分析了其在地表水、土壤、地下水等周边环境介质的迁移行为, 比较了渗滤液处理工艺对新污染物的去除效果. 总体上, 我国垃圾填埋渗滤液中新污染物赋存浓度范围跨度较大, 且主要研究集中在东部发达区域, 东北和西北欠发达地区研究较少. 填埋场年龄、渗滤液理化性质、自然条件等因素均会影响其赋存特征, 但是具体影响机制缺乏深入阐释. 垃圾填埋渗滤液中新污染物通过排放、渗透等方式可以迁移到周边土壤与地表水和地下水中, 但是大气迁移鲜有研究. 现有渗滤液处理工艺(生物+膜处理)可有效去除多数新污染物, 但是转化规律及去除机制仍不明晰. 最后, 本文对垃圾填埋场渗滤液中新污染物未来研究重点进行了展望, 以期为填埋场新污染物的环境管理提供科技支持.

**关键词** 垃圾填埋渗滤液, 新污染物, 赋存, 迁移, 去除.

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## The occurrence, migration, and removal of typical emerging contaminants in landfill leachate in China

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**Abstract** The environmental safety of emerging contaminants (ECs) is receiving increasing attention because of their biotoxicity, environmental persistence, bioaccumulation and other

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characteristics, which pose potential risks to the ecological environment and human health. In recent years, various ECs have been detected in landfill leachate, making it a “source” of ECs that cannot be ignored. Based on the literatures, this review systematically summarized the distribution characteristics of typical ECs in China's landfill leachate, including pharmaceuticals and personal care products, antibiotic resistance genes, endocrine disrupting compounds, perfluorochemicals and microplastics, and discussed the factors affecting their distribution levels. Their migration behavior in surface water, soil, groundwater and other surrounding environmental media was analyzed, and the removal effect of ECs in different leachate treatment processes was compared. In general, the concentration range of ECs in landfill leachate in China is large, and the studies are main concentrated in the eastern developed regions, while less studies are conducted in the northeastern and northwestern less developed regions. The landfill age, physicochemical properties of leachate and natural conditions can affect the fate characteristics of ECs, but the specific influence mechanisms are not well understood. The ECs in landfill leachate can migrate to surrounding soil, surface water and groundwater through discharge and infiltration, but atmospheric transport has rarely been studied. The existing leachate treatment processes (biological and membrane treatment) can effectively remove most of the ECs, but the transformation patterns and removal mechanisms are still unclear. Finally, this review provides an outlook on the future research priorities of ECs in landfill leachate and provides scientific and technical support for the environmental management of ECs in landfills.

**Keywords** refuse landfill leachate, emerging contaminants, occurrence, migration, removal.

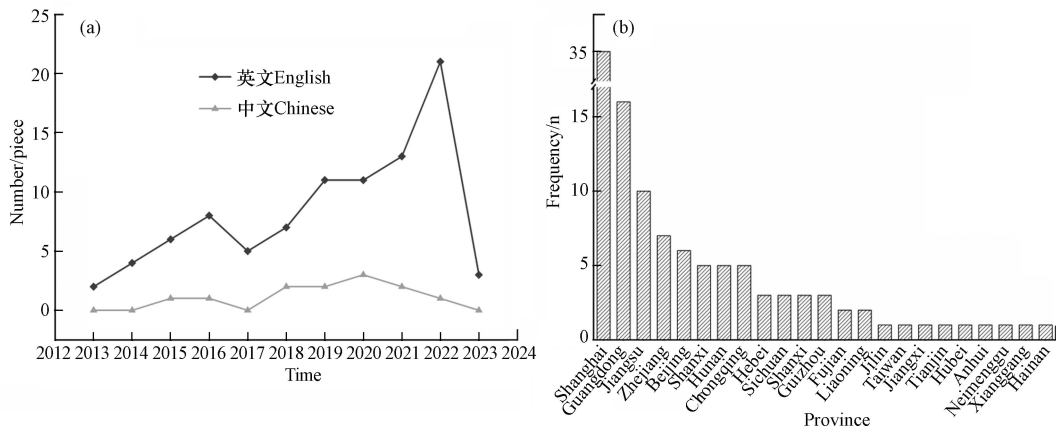
近年来,随着我国社会经济的高速发展和城市化进程的加速,城市生活垃圾产生量迅速增加,根据《2020年全国大、中城市固体废物污染环境防治年报》统计数据,2019年全国196个大、中城市生活垃圾产生量23560.2万t,预计2030年将达到3.5亿t<sup>[1]</sup>.相对焚烧与堆肥等处理方式,卫生填埋由于相对廉价仍是我国处理处置城市垃圾的主要手段.垃圾填埋渗滤液作为填埋过程中持续产生的二次污染物,不仅含有较高的有机质以及营养物质(氮磷化合物),而且越来越多的研究显示药物及个人护理品、抗生素抗性基因、全氟化合物等新污染物在渗滤液中检出<sup>[2-5]</sup>.同时,渗滤液可通过渗透、扩散等作用进入地下水<sup>[6]</sup>、地表水<sup>[7]</sup>、土壤<sup>[8]</sup>等周边环境.因此,垃圾渗滤液已经成为新污染物影响周边环境不可忽视的源.

由于新污染物具有生物毒性、环境持久性、生物累积性等特征,对生态环境和人体健康具有较大的潜在风险,其日益受到各国政府和民众的广泛关注,我国也已将新污染物治理列入“十四五”规划和2035年远景目标.目前我国对于新污染物的研究主要集中于地表水<sup>[9]</sup>、沉积物<sup>[10]</sup>、土壤<sup>[11]</sup>以及市政污水<sup>[12]</sup>等,而对垃圾填埋渗滤液中的新污染物研究较少.本文总结了药物及个人护理品(pharmaceuticals and personal care products, PPCPs)、抗生素抗性基因(antibiotic resistance genes, ARGs)、内分泌干扰物(endocrine disrupting compounds, EDCs)、全氟化合物(perfluorochemicals, PFCs)、微塑料(microplastics, MPs)为主的新污染物在我国垃圾填埋渗滤液中的赋存特征、影响因素、迁移行为和去除技术,并提出未来亟待发展的研究方向,旨在为我国新污染物的治理和风险管控提供理论支撑.

## 1 垃圾填埋渗滤液中新污染物研究现状(Research status of ECs in landfill leachate)

通过 Web of Science (WoS) 以及中国知网(CNKI)数据库检索我国垃圾填埋场渗滤液中新污染物(PPCPs、ARGs、MPs、EDCs、PFCs)的研究文献,截止到2023年1月共有103篇,统计数据见图1(a).由图1(a)可知,2016年之后相关研究逐渐增多,约占统计数据的90%,且整体呈上升趋势.此外,全国有23个省份的垃圾填埋场渗滤液中至少检测到1种新污染物,见图1(b).由图1(b)可知,相关研究主要集中在东部沿海发达地区,上海是研究涉及最多的城市,高达35篇,其次是广东和江苏,分别为16和10篇,西部和北部地区相关研究较少.虽然相较其它环境介质中新污染物的研究,垃圾填埋渗滤

液中新污染物研究起步较晚,但是通过文献分析发现其作为新污染物重要的“源”逐步引起国内学者的关注.目前,由于经济发展水平、消费模式、管理需求等因素的限制,垃圾填埋渗滤液中新污染物主要集中在东部发达区域,未来需要全面研究我国不同地域垃圾填埋渗滤液中新污染物的赋存特征.



$\beta$ -内酰胺和质子泵抑制剂、解离性麻醉剂和拟交感神经化合物等,其中抗生素检出种类约占 40%,检出频率较高的抗生素类别为磺胺类(磺胺嘧啶和磺胺甲恶唑)、四环素类(四环素和土霉素)和大环内酯类(红霉素和罗红霉素);非抗生素类检出频率较高的药物为吉非罗齐、卡马西平、咖啡因、美托洛尔和舒必利;检出浓度范围跨度较大,从  $0.6 \text{ ng}\cdot\text{L}^{-1}$  到  $38.59 \text{ mg}\cdot\text{L}^{-1}$ (以各种药物的最大浓度统计).由统计结果发现,我国垃圾填埋渗滤液中检出的 PPCPs 主要是药物,个人护理品在渗滤液中很少检出.此外,对检出浓度较高的前 25 种 PPCPs 进行分析(图 3),结果显示浓度最高的药物为磷酸三(2-氯乙基)酯,同时对这 25 种 PPCPs 进行分类统计,结果发现抗生素类药物仅占 32%,非抗生素类药物的检出比例较高.根据现有文献报道,渗滤液中部分非抗生素类药物(苯扎贝特和卡马西平)对藻类的急性毒性高于抗生素类药物<sup>[20]</sup>,同时 Yin 等<sup>[21]</sup>研究表明在饮用水、地表水和污水处理厂中检出的 PPCPs 的代谢产物或中间产物比母体具有相同甚至更高的毒性,这在一定程度上反映出非抗生素类药物更值得关注.目前,我国学者对渗滤液中 PPCPs 的研究更多集中在抗生素类药物,非抗生素类药物的研究报道相对较少,而对于 PPCPs 代谢物及中间产物的研究鲜有报道.

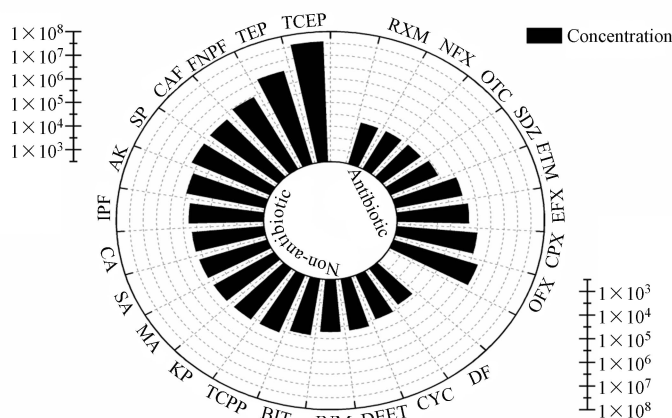


图 3 前 25 种 PPCPs 在垃圾填埋场渗滤液中的检出浓度( $\text{ng}\cdot\text{L}^{-1}$ )

(TCEP: 磷酸三(2-氯乙基)酯, TEP: 磷酸三乙酯, FNPF: 非诺洛芬, OFX: 氧氟沙星, CAF: 咖啡因, SP: 舒必利, CPX: 环丙沙星, AK: 安赛蜜, IPF: 布洛芬, CA: 氯贝酸, EFX: 恩氟沙星, SA: 水杨酸, MA: 甲芬那酸, KP: 酮洛芬, TCPP: 磷酸(1-氯丙-2-基)酯, ETM: 红霉素, BIT: 苯并异噻唑啉酮, INM: 吡啶美辛, DEET: 避蚊胺, CYC: 甜蜜素, SDZ: 磺胺嘧啶, DF: 双氯芬酸, OTC: 土霉素, NFX: 诺氟沙星, RXM: 罗红霉素)

Fig.3 Concentrations of the top 25 PPCPs in landfill leachate ( $\text{ng}\cdot\text{L}^{-1}$ )

(TCEP: tris (2-chloroethyl) phosphate, TEP: triethyl phosphate, FNPF: fenopfen, OFX: ofloxacin, CAF: caffeine, SP: sulphiride, CPX: ciprofloxacin, AK: acesulfame, IPF: ibuprofen, CA: chlorbesic acid, EFX: enrofloxacin, SA: salicylic acid, MA: mefenamic acid, KP: ketoprofen, TCPP: tris (1-chloropropyl-2-yl) phosphate, ETM: erythromycin, DEET: DEET, BIT: benzoisothiazolinone, INM: indomethacin, CYC: cyclamate, SDZ: sulfadiazine, DF: diclofenac, OTC: oxytetracycline, NFX: norfloxacin, RXM: roxithromycin)

### 2.1.2 影响因素

目前研究发现,不同填埋过程产生的渗滤液中 PPCPs 检出浓度有较大差异,从  $\text{ng}\cdot\text{L}^{-1}$ — $\mu\text{g}\cdot\text{L}^{-1}$  不等,影响渗滤液中 PPCPs 浓度差异的因素主要包括垃圾填埋龄<sup>[2,22-25]</sup>、季节与降雨<sup>[24,26-27]</sup>、重金属<sup>[28-29]</sup>、渗滤液理化性质<sup>[27-28,30]</sup> 以及当地人口规模与消费水平<sup>[31]</sup> 等.根据现有研究报道,垃圾填埋龄与渗滤液中大多数抗生素的浓度呈负相关( $r=-0.10$ — $-0.42$ ),较高填埋龄渗滤液中抗生素浓度相对较低<sup>[25]</sup>.目前,降雨对垃圾填埋场渗滤液中 PPCPs 浓度的影响还不明确, Lu 等<sup>[26]</sup> 研究发现,由于雨水的稀释作用,旱季垃圾渗滤液中 PPCPs 浓度要高于雨季,而 Yu 等<sup>[32]</sup> 研究表明,降雨的淋洗作用对填埋场具有更显著的影响,导致雨季垃圾渗滤液中 PPCPs 浓度较高.季节对渗滤液中 PPCPs 的浓度存在不同的研究结论, Sui 等<sup>[33]</sup> 研究表明大多数 PPCPs 浓度没有发生明显的季节性变化( $P<0.05$ ),而 Yu 等<sup>[27]</sup> 研究表明 PPCPs 浓度,特别是咖啡因,与温度存在正相关.重金属与抗生素有较强的相关性, He 等<sup>[29]</sup> 研究表明磺胺类和喹诺酮类抗生素与 Co、Cu 和 Ni 等重金属呈显著正相关( $P<0.01$ ),同时, PPCP 的赋存水平与渗滤液 pH( $P<0.01$ )和氮水平有较强相关性,而氮水平与人口规模( $r=0.733$ ,  $P<0.01$ )和人均 GDP( $r=0.485$ ,  $P<0.05$ )显著相关<sup>[31]</sup>.2020 年在上海老港垃圾填埋场渗滤液中 PPCPs 检出浓度较高的为咖啡因和双氯

酚酸<sup>[7]</sup>,而在 2021 年 PPCPs 检出浓度较高的为阿苯达唑、避蚊胺、舒必利和咖啡因,这表明 PPCPs 在渗滤液中的种类和浓度会随着消费模式及经济情况发生变化<sup>[5,27]</sup>。此外,目前的研究具有较大的地域局限性,由于研究地区主要集中在我国中东部和西南区域,并不能获取全国范围内垃圾填埋场渗滤液中 PPCPs 的时空分布规律,鉴于我国地域环境、经济水平、消费方式等差异,有必要开展区域性和连续性的研究。

## 2.2 ARGs

### 2.2.1 赋存特征

近些年来,抗生素药物滥用致使细菌耐药性增强,带有抗生素抗性基因的细菌进入环境后,通过转化与转导等水平转移至人类病原菌,使得致感染性疾病的病原菌日益复杂、种类增多,若多种 ARGs 同时转移到致病菌中,就会导致超级细菌的产生,极大地增加环境健康风险,这也使 ARGs 成为人们广泛关注的一类新污染物<sup>[34]</sup>。目前,在我国垃圾填埋场渗滤液中已广泛检出 ARGs,见表 1 所示,在 13 个省份 18 个城市垃圾渗滤液中共检测到 21 种 ARGs 类型和 540 个 ARGs 亚型,其中上海检出的 ARGs 种类最多,为 18 种 ARGs 类型和 369 个 ARGs 亚型,同时也是文献报道次数最多的城市,高达 31 次,而在陕西铜川检出的 ARGs 种类最少,为 2 种 ARGs 类型和 4 个 ARGs 亚型。由此可见,城市化与经济程度将影响垃圾渗滤液中 ARGs 的浓度和种类。此外,根据现有研究报道,我国垃圾渗滤液中主要的 ARGs 种类为磺胺类、大环内酯类、 $\beta$ -内酰胺类、四环素类及氨基糖苷类。

表 1 我国部分垃圾填埋场渗滤液 ARGs 检出丰度

Table 1 The abundance of ARGs detected in landfill leachate of some cities in China

物质 Chemical	地区 Region	丰度 Abundance	参考文献 References
磺胺类抗性基因 <i>sul 1</i>	各地	$(5.51 \times 10^{-6} - 4.97 \times 10^{-1})$ ARG copies/16S rRNA copies	[41]
	陕西	$(4.45 \pm 1.02 - 6.31 \pm 0.49)$ lg copies/ng DNA	[42]
	上海	$(5.6 \pm 0.9)$ lg copies/ng DNA	[23]
	贵州	7.58 lg copies/ng DNA	[25]
	浙江	7.97 lg copies/ng DNA	[25]
	江苏	8.81 lg copies/ng DNA	[25]
磺胺类抗性基因 <i>sul 2</i>	陕西	$(1.98 \pm 0.62 - 2.70 \pm 0.51)$ lg copies/ng DNA	[42]
	贵州	5.89 lg copies/ng DNA	[25]
	浙江	6.88 lg copies/ng DNA	[25]
	江苏	6.62 lg copies/ng DNA	[25]
四环素类抗性基因 <i>tet Q</i>	浙江	$(9.57 \pm 1.32)$ lg gene copies/L	[42]
	陕西	$(3.24 \pm 0.24 - 3.64 \pm 0.90)$ lg copies/ng DNA	[42]
	贵州	6.29 lg copies/ng DNA	[25]
	浙江	5.16 lg copies/ng DNA	[25]
	江苏	5.91 lg copies/ng DNA	[25]
四环素类抗性基因 <i>tet M</i>	陕西	$(2.48 \pm 0.04 - 3.01 \pm 1.38)$ lg copies/ng DNA	[42]
	贵州	5.45 lg copies/ng DNA	[25]
	浙江	4.66 lg copies/ng DNA	[25]
	江苏	4.59 lg copies/ng DNA	[25]
氟喹诺酮类抗性基因 <i>mexF</i>	浙江	$(11.92 \pm 0.22)$ lg gene copies/L	[24]
	贵州	4.33 lg copies/ng DNA	[25]
	江苏	3.81 lg copies/ng DNA	[25]
$\beta$ -内酰胺抗性基因 <i>bla CTX-M</i>	上海	$(4.1 \pm 0.7)$ lg copies/ng DNA	[23]
	贵州	$(1.75 \pm 0.30)$ lg copies/ng DNA	[25]
	浙江	5.26 lg copies/ng DNA	[25]
	江苏	4.38 lg copies/ng DNA	[25]

续表 1

物质 Chemical	地区 Region	丰度 Abundance	参考文献 References
四环素类抗性基因 <i>tet O</i>	各地	$(2.03 \times 10^{-7}—4.26 \times 10^{-2})$ ARG copies/16S rRNA copies	[41]
四环素类抗性基因 <i>tet W</i>	各地	$(2.03 \times 10^{-7}—4.06 \times 10^{-3})$ ARG copies/16S rRNA copies	[41]
氨基糖苷类抗性基因 <i>aad A1</i>	上海	$(5.5 \pm 0.8)$ lg copies/ng DNA	[23]

注: 各地指调查的成都、重庆、上海、深圳、长沙、唐山和广东等地垃圾渗滤液。

All localities refer to Chengdu, Chongqing, Shanghai, Shenzhen, Changsha, Tangshan, Guangdong, etc.

### 2.2.2 影响因素

已有研究在报道垃圾填埋场渗滤液 ARGs 赋存特征与组成的同时, 对影响其赋存的自然因素以及渗滤液特征理化性质之间的关联进行了研究探讨. 根据现有研究报道, 垃圾渗滤液中 ARGs 种类及丰度受到多种因素的影响, 主要包括抗生素、重金属、可移动基因元件、微生物群落、填埋场年龄、渗滤液理化性质、自然条件等. 磺胺类 ARGs 丰度与磺胺类抗生素(磺胺嘧啶、磺胺吡啶、磺胺甲恶唑、磺胺甲基嘧啶、磺胺二甲嘧啶)的增加存在显著相关性( $P < 0.05$ )<sup>[22]</sup>, 但是多数研究结果表明抗生素与 ARGs 相关性较弱, 如 Wu 等<sup>[35]</sup>研究发现除四环素类抗生素(TCs)与 *tetW* 和 *tetQ* 具有相关性( $r$  分别为 0.88 和 0.81), ARGs 丰度与检测抗生素浓度没有关联性, Zhao 等<sup>[36]</sup>研究结论也表明二者无关联性. 重金属与 ARGs 丰度具有较强的相关性, 如 Wu 等<sup>[35]</sup>研究结果表明大多数 ARGs 丰度与检测重金属浓度显著相关( $P < 0.05$ ), 尤其是 Cd 和 Cr, Yu 等<sup>[24]</sup>研究发现 ARGs 丰度与重金属(Zn、Cu 和 Co)具有显著关联性( $P < 0.05$ ), Zhang 等<sup>[8]</sup>研究结果显示 ARGs(*tetC*、*tetW*、*sul1*、*sul2*、*int11* 和 *FOX*)与重金属(Cu、Zn、Ni 和 Cr)显著相关( $P < 0.05$ ). ARGs 丰度与可移动基因元件、微生物群落结构也具有紧密关联性, 如 Wu 等<sup>[23]</sup>和 Su 等<sup>[37]</sup>, 且随着填埋场年龄的增加逐渐升高. 另外, 由于渗滤液组分较复杂, ARGs 丰度与渗滤液理化性质存在不同的研究结论, Su 等<sup>[38]</sup>研究表明 ARGs 丰度与渗滤液理化参数(如 COD、总氮、氨氮、硝酸盐及亚硝酸盐)没有关联性, 而 Zhao 等<sup>[36]</sup>研究发现 ARGs 丰度与渗滤液理化参数(COD、TOC、总氮、总磷、氨氮、电导率)具有负相关性( $P < 0.05$ ), Wang 等<sup>[25]</sup>研究显示 COD 与 *tetM* ( $P < 0.05$ )、 $\text{NH}_4^+\text{-N}$  与 *ermB*、*blaCTX-M* 和 *int11* ( $P < 0.01$ )、 $\text{NO}_3\text{-N}$  与 *tetQ* 和 *mexF* ( $P < 0.05$ )以及 pH 与 *ermB*、*mefA*、*blaCTX-M* 和 *int11* ( $P < 0.01$ )均呈正相关, 因而需要进一步深入研究不同类别的 ARGs 与渗滤液组分的特异性关联. 除此之外, 渗滤液中其他新污染物也会影响 ARGs 丰度与传播, Shi 等<sup>[39]</sup>研究发现微塑料选择性的富集渗滤液中 ARGs, 其中 *strB* 和 *blaTEM* 富集量最大, 而 *mefA*、*ermB*、*tetM* 和 *tetQ* 轻微富集在微塑料, Su 等<sup>[40]</sup>进一步研究发现微塑料老化过程将增强对 ARGs 的富集, 机理分析表明在微塑料表面存在非随机、更紧密和更稳定的 ARGs 细菌关系影响了 ARGs 的传播. 然而, 值得关注的是, 这些影响因素均为单因子研究, 影响因素之间的相互作用并不明晰, 这对探究垃圾填埋渗滤液中 ARGs 丰度与多种因素联合影响机制明晰了研究方向.

### 2.3 MPs

1950 年到 2015 年, 世界范围内塑料的生产量从约 50 万 t 大幅度增加至 49 亿 t, 有研究预测表明, 2050 年将会有 120 亿 t 塑料进入垃圾填埋场或自然环境<sup>[43-44]</sup>. 2004 年 Thompson 首次提出 MPs 的概念<sup>[45]</sup>, 通常粒径  $< 5$  mm 的塑料碎片被称为微塑料<sup>[46]</sup>. MPs 稳定性极强, 在环境中很难降解, 已在海洋、湖泊、空气、沉积物及垃圾填埋场固体垃圾中广泛检出<sup>[47]</sup>. 垃圾填埋场固体垃圾中的 MPs 浓度为 20000—91000 items·kg<sup>-1</sup><sup>[47]</sup>, 远高于沉积物(260 items·kg<sup>-1</sup>)<sup>[48]</sup> 和地表水(3.4—25.8 items·L<sup>-1</sup>)<sup>[49]</sup>.

目前, 由于渗滤液成分较为复杂, 受限于 MPs 的测定分析技术, 其污染现状调查非常有限, 现有研究主要集中在我国上海和江苏部分城市, 见表 2. 如表 2 所示, 共有 36 种 MPs 在渗滤液中检出, 浓度范围在 0.42—382 items·L<sup>-1</sup>, 检出浓度较高的 MPs 为聚乙烯、聚丙烯、聚酰胺和聚对苯二甲酸乙二醇酯. 此外, 不同地区垃圾填埋场渗滤液中 MPs 检出浓度存在差异, 但对于影响因素的研究还十分有限, 更多的研究集中在渗滤液中 MPs 检测方法的优化<sup>[50]</sup>、去除方法<sup>[51-54]</sup>及其作为载体对其他污染物的影响<sup>[53,55]</sup>等. 现有研究发现 MPs 检出浓度与填埋场年龄以及环境因素有关, Su 等<sup>[3]</sup>研究了垃圾填埋场年龄对 MPs 浓度的影响, 结果发现随着填埋场年龄的增加, 聚丙烯微塑料可以被氧化降解, 另外, 环境因素(水分、盐度、高温、厌氧环境及微生物)也会促进塑料垃圾分解为 MPs, 进而使垃圾填埋场渗滤液中 MPs 浓度增加<sup>[51,56]</sup>.

表 2 MPs 在垃圾填埋场渗滤液中检出情况

Table 2 MPs detected in landfill leachate

地区 Region	物质 Chemical	浓度/(items·L <sup>-1</sup> ) Concentration		参考文献 References
		最小值 Minimum	最大值 Maximum	
		上海、无锡 苏州、常州	PP、PVC、PS、ABS、PMMA、PET、PMDS、PTFE、PU、 EVA、PES、EP、PF、PPC、ALK、PE	
广东	PU、PAT、PA、PEC、PP、PE、PS、PET	3	25	[58]
上海	PE、PP、PS、PET、EPM、PVC、玻璃纸	4	13	[37,59]
上海	PP、PA、PE、PES、人造丝	200	382	[50]
上海	EP、PET、PVAC、CN、PS、PP、EPM、PA、PES、PE、Acrylic	0.63	1.77	[60]
苏州	PE、PP、PA、PVA、PVB、PMP、PDO、PSB、PAA、PBMA、PVP、RUBBER	218.3	252.5	[54]

注: PP:聚丙烯, PVC:聚氯乙烯, PS:聚苯乙烯, ABS:聚丙烯腈、丁二烯和苯乙烯聚合物, PMMA:聚甲基丙烯酸甲酯, PET: 聚对苯二甲酸乙二醇酯, PMDS:聚二甲基硅氧烷, PTFE:聚四氟乙烯, PU:聚氨酯, EVA: 乙烯-醋酸乙烯共聚物, PES: 聚酯纤维, EP: 环氧树脂, PF: 酚醛树脂, PPC: 聚碳酸亚丙酯, ALK: 醇酸树脂, PE: 聚乙烯, PAT: 聚芳酯, PA: 聚酰胺, PEC: 氯化聚乙烯, EPM: 乙丙橡胶, PVAC: 聚乙烯醇, CN: 涂层尼龙, PVA: 聚乙烯醇, PVB: 聚乙烯醇缩丁醛, PMP: 聚4-甲基戊烯-1, PDO: 聚2,5-二苯基-1,3,4-噁二唑, PSB: 聚苯乙烯-丁二烯, PAA: 聚丙烯酸, PBMA: 聚甲基丙烯酸正丁酯, PVP: 聚乙烯吡咯烷酮, RUBBER: 顺式聚异戊二烯。

Note: PP: polypropylene, PVC: polyvinyl chloride, PS: polystyrene, ABS: acrylonitrile-butadiene-styrene, PMMA: poly(methylmethacrylate), PET: polyethylene terephthalate, PMDS: polydimethylsiloxane, PTFE: poly tetra fluoroethylene, PU: polyurethane, EVA: ethylene vinyl acetate copolymer, PES: polyester, EP: epoxy resin, PF: phenolic resin, PPC: polypropylene carbonate, ALK: alkyd resin, PE: polyethylene, PAT: polyaryl ester, PA: polyamide, PEC: polyethylene chlorinated, EPM: ethylene propylene rubber, PVAC: polyvinyl acetate, CN: coated nylon, PVA: polyvinyl alcohol, PVB: polyvinyl butyral, PMP: polymethylpentene, PDO: Poly(2,5-diphenyl-1,3,4-oxadiazole), PSB: Poly(styrene-co-butadiene), PAA: polyacrylic acid, PBMA: poly(*n*-Butylamethacrylate), PVP: poly(*n*-vinyl pyrrolidone), RUBBER: poly-(*cis*1,4-Isoprene).

## 2.4 EDCs

内分泌干扰物被定义为“干扰体内负责平衡、繁殖、发育和行为的激素合成、分泌、运输、结合、作用或消除的外源性物质”, 主要包括双酚类、烷基酚类、邻苯二甲酸酯类、激素类、有机氯杀虫剂、除草剂和某些重金属(铅、镍、汞)等<sup>[61]</sup>, 广泛添加于家居产品、生活用品(杀虫剂、洗涤剂和防晒霜等)、工业产品(增塑剂)等消费品中。与其他环境污染物相比, EDCs 在低剂量环境浓度下就可产生慢性毒性效应, 并对人体生长、发育、生殖产生影响。目前在我国垃圾填埋场渗滤液中均有检出, Gong 等<sup>[62]</sup>在垃圾渗滤液中检测到双酚 A(18.11  $\mu\text{g}\cdot\text{L}^{-1}$ )、苯酚(2.76  $\mu\text{g}\cdot\text{L}^{-1}$ )、邻苯二甲酸二正丁酯(4.86  $\mu\text{g}\cdot\text{L}^{-1}$ )、邻苯二甲酸二正辛酯(0.21  $\mu\text{g}\cdot\text{L}^{-1}$ )和邻苯二甲酸二乙基己酯(9.16  $\mu\text{g}\cdot\text{L}^{-1}$ ); Huang 等<sup>[63]</sup>在广东省垃圾填埋场渗滤液中检测到 11 种双酚类化合物, 总浓度为 32130  $\text{ng}\cdot\text{L}^{-1}$ , 双酚 A 是主要污染物。现有数据分析结果表明在我国垃圾渗滤液中共检出 42 种 EDCs, 浓度范围为 0.96—178000  $\text{ng}\cdot\text{L}^{-1}$ , 并对检出浓度较高的前 25 种 EDCs 进行分类, 结果显示邻苯二甲酸酯类和雌激素类为主要污染物(图 4)。然而, 目前关于影响 EDCs 赋存水平及组成的文献数据还未见报道, 因此尚难以给出对 EDCs 赋存水平及组成影响的清晰认识。

## 2.5 PFCs

全氟化合物是以烷基碳链结构为骨架、氢原子被氟原子全部或部分取代的一类有机化合物<sup>[64-65]</sup>, 因其良好的表面活性、热/化学稳定性以及疏水和疏油的特性, 被广泛应用于食品包装、纺织品、涂料、化妆品等工业和商业产品<sup>[66]</sup>。鉴于其可能引起生殖发育、免疫毒性和内分泌干扰等多种潜在危害效应, 人们对 PFCs 污染的关注度越来越高。目前, PFCs 在我国湖泊、河流、沉积物、土壤、大气等环境介质中已广泛检出, 近年来才逐渐关注垃圾填埋场渗滤液中 PFCs 的污染水平, 而且研究主要集中在北京、上海等经济发达地区。

在我国垃圾填埋渗滤液中共检出 20 种 PFCs, 其浓度水平跨越 6 个数量级, 浓度范围为 nd—214000  $\text{ng}\cdot\text{L}^{-1}$ , 并对检出的 PFCs 进行分类(图 5), 结果显示 PFOA(长链全氟化合物)浓度最高, 但是短链 PFCs 在渗滤液中所占比例较高, 由于短链 PFCs 在水相中的高溶解度及低辛醇/水分配系数, 推测淋洗后更容易分配于渗滤液液相。根据现有的研究报道, 在上海未处理渗滤液中检出的全氟烷基酸类化合物浓度最高, 赋存总浓度高达 292000  $\text{ng}\cdot\text{L}^{-1}$ , 其中 PFOA 浓度为 214000  $\text{ng}\cdot\text{L}^{-1}$ , 其检出浓度高于其他地区 1—3 个数量级<sup>[4]</sup>, 而西南地区研究数据表明垃圾渗滤液中 PFCs 浓度相对较低, 且主要以全氟丁

烷羧酸和全氟丁烷磺酸等短链为主<sup>[67]</sup>,这可能与城市的经济水平和工业发展水平相关.此外,Buck等<sup>[68]</sup>研究表明长链全氟化合物比短链全氟化合物具有更强的生物累积性,因此,长链全氟化合物应得到更多的关注.此外,国外已开展影响渗滤液中PFCs赋存特征和组成的因素及机制研究,如降雨、填埋废物类型与时间、渗滤液理化性质,但是我国鲜有相关研究报道,亟需系统性研究以获得进一步认识.

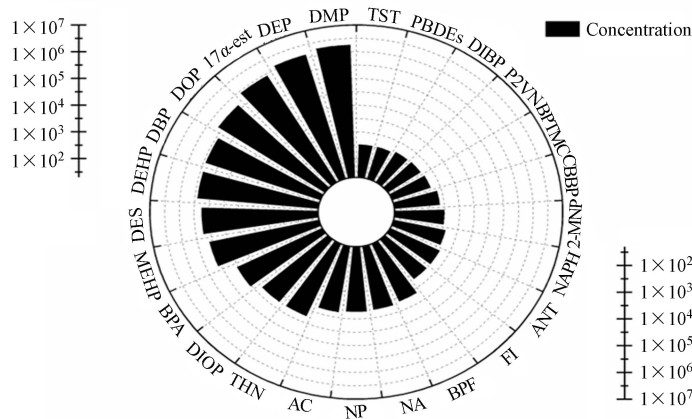


图4 前25种EDCs在垃圾填埋场渗滤液中的检出浓度( $\text{ng}\cdot\text{L}^{-1}$ )

(DMP: 邻苯二甲酸二甲酯, DEP: 邻苯二甲酸二乙酯, 17 $\alpha$ -est: 17 $\alpha$ -乙炔雌二醇, DOP: 邻苯二甲酸二正辛酯, DBP: 邻苯二甲酸二丁酯, DEHP: 2-苯二甲酸单(2-乙基己基)酯, DES: 乙烯雌酚, MEHP: 2-苯二甲酸双(2-甲基丙基)酯, BPA: 双酚 A, DIOP: 邻苯二甲酸二异辛酯, THN: 1,2,3,4-四氢萘, AC: 苊, NP: 壬基酚, NA: 芴, BPF: 双酚 F, EI: 雌酮, ANT: 蒽, NAPH: 萘, 2-MNP: 2-甲基萘, BBP: 邻苯二甲酸丁苄酯, BPTMC: 双酚 TMC, P2VN: 2-乙烯基萘, DIBP: 邻苯二甲酸二异丁酯, PBDEs: 多溴二苯醚, TST: 睾酮)

Fig.4 The top 25 EDCs in landfill leachate( $\text{ng}\cdot\text{L}^{-1}$ )

(DMP: dimethyl phthalate, DEP: diethyl phthalate, 17 $\alpha$ -est: 17 $\alpha$ -acetylene estradiol, DOP: di-n-octyl phthalate, DBP: dibutyl phthalate, DEHP: mono (2-ethylhexyl) 1,2-benzoate, DES: diethylstilbestrol, MEHP: bis (2-methylpropyl) 1,2-benzoate, BPA: bisphenol A, DIOP: diisooctyl phthalate, THN: 1,2,3,4-tetraphthalene, AC: acenaphthene, NP: nonyl phenol, NA: fluorene, BPF: bisphenol F, EI: oestrone, ANT: anthracene, NAPH: naphthalene, 2-MNP: 2-methylnaphthalene, BBP: butyl benzyl phthalate, BPTMC: bisphenol TMC, P2VN: 2-vinylnaphthalene, DIBP: diisobutyl 2-vinylnaphthalene phthalate, PBDEs: polybrominated diphenyl ethers, TST: testosterone)

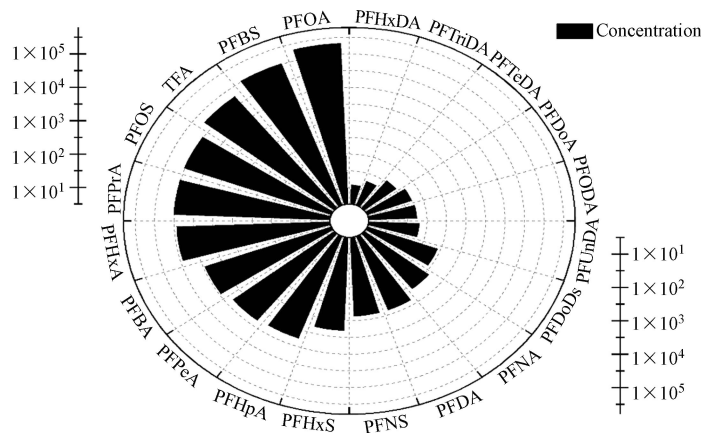


图5 PFCs在垃圾填埋场渗滤液中的检出浓度( $\text{ng}\cdot\text{L}^{-1}$ )

(PFOA: 全氟烷基辛酸, PFBS: 全氟丁烷磺酸, TFA: 三氟乙酸, PFOS: 全氟辛烷磺酸, PFPrA: 全氟烷基丙酸, PFHxA: 全氟烷基己酸, PFBA: 全氟烷基丁酸, PFPeA: 全氟烷基戊酸, PFHpA: 全氟烷基庚酸, PFHxS: 全氟己烷磺酸, PFNS: 全氟烷基壬酸, PFDA: 全氟烷基癸酸, PFNA: 全氟烷基壬酸, PFDoDs: 全氟十一烷磺酸钠, PFUnDA: 全氟烷基十一酸, PFODA: 全氟烷基十八酸, PFDoA: 全氟烷基十二酸, PFTeDA: 全氟烷基十四酸, PFTrDA: 全氟烷基十三酸, PFHxDA: 全氟烷基十六酸)

Fig.5 The PFCs in landfill leachate( $\text{ng}\cdot\text{L}^{-1}$ )

(PFOA: perfluoroalkyl octanoic acid, PFBS: perfluorobutane sulfonic acid, TFA: trifluoroacetic acid, PFOS: perfluorooctane sulfonic acid, PFPrA: perfluoropropionic acid, PFHxA: perfluoroalkyl hexanoic acid, PFBA: perfluoroalkyl butyric acid, PFPeA: perfluoropentanoic acid, PFHpA: perfluoroalkyl heptanoic acid, PFHxS: perfluorohexane sulfonic acid, PFNS: perfluorononic acid, PFDA: perfluoroalkyl decanoic acid, PFNA: perfluoroalkyl nonanoic acid, PFDoDs: Sodium perfluoroctane sulfonate, PFUnDA: Perfluoroalkyl undecanoic acid, PFODA: Perfluoroalkyl octadecanoic acid, PFDoA: Perfluoroalkyl dodecanoic acid, PFTeDA: Perfluoroalkyl tetradecanoic acid, PFTrDA: Perfluoroalkyl tridecanoic acid, PFHxDA: Perfluoroalkyl hexadecyl acid)



### 3 垃圾填埋渗滤液中新污染物环境迁移(Environmental migration of ECs in landfill leachate)

垃圾渗滤液是新污染物重要的储库,同时也是环境中新污染物的重要污染源。目前,渗滤液处理设施可有效去除渗滤液中的常规指标,如 COD、BOD、总氮、氨氮、总磷等,但是对于新污染物的去除效果有限<sup>[26]</sup>,排放将对受纳环境构成潜在威胁,但是目前我国有关新污染物通过垃圾填埋场渗滤液排放或渗透等途径向周边环境迁移行为的研究文献较少。Zhang 等<sup>[8]</sup>研究表明受纳地表水中 *sul1*、*sul2* 和 *int1* 的绝对浓度范围为 107.76—108.26 copies·mL<sup>-1</sup>,比河流中的浓度高约 1 个或 2 个数量级,出水会导致地表水中 ARGs 的富集。同时,黄等<sup>[69]</sup>研究表明在垃圾填埋场背景点河水中检测到 57 种 ARGs,下游河水中检测到 158 种 ARGs,下游河流 150 种抗性基因显著富集( $P<0.05$ ),生活垃圾填埋场可能对河水中的抗性基因的多样性和丰度产生影响。然而,Yu 等<sup>[7]</sup>比较了垃圾填埋场附近地表水与同一流域远离垃圾填埋场地地表水中 PPCPs 的浓度,研究表明垃圾填埋场渗滤液对周边地表水中 PPCPs 的贡献可以忽略不计。与地表水相比,由于氧化还原相对减少、缺少光降解和微生物降解效率低,地下水中的污染物更持久且更难消除<sup>[70]</sup>。因此,垃圾渗滤液对地下水的影响关注度更高。Han 等<sup>[6]</sup>研究表明渗滤液对地下水的污染主要出现在 1000 m 内,对地下水造成严重污染发生在 200 m 内。Han<sup>[71]</sup>等比较了原始渗滤液与处理过的渗滤液和地下水之间相关性,结果显示两组之间呈显著正相关( $P<0.05$ ),表明垃圾渗滤液对附近地下水有负面影响。但 Peng 等<sup>[70]</sup>研究表明垃圾填埋场附近地下水中水杨酸与渗滤液的相关性很差( $r=-0.06$ ),抗生素和个人护理产品浓度与垃圾填埋场的距离呈负相关, $r$  分别为 -0.17 和 -0.30,同时发现垃圾填埋场附近的地下水与不受垃圾填埋场影响的地下水中 PPCPs 没有显著性差异,推测与目标物性质以及复杂地下水水文地质情况差异有关,需要进一步深入研究。Zhang 等<sup>[8]</sup>研究发现受纳表层土壤中 ARGs 的浓度比对照土壤高 1.59—4.98 个数量级,表明渗滤液灌溉会导致 ARGs 向垃圾填埋场附近土壤迁移并发生富集。然而,Liu 等<sup>[72]</sup>研究显示填埋场表层土壤中邻苯二甲酸酯的含量与汉江平原相比无显著差异,甚至还显著低于城郊和城市,表明填埋场对附近表层土壤邻苯二甲酸酯污染没有明显影响,但是其研究还发现在更深的土壤中邻苯二甲酸酯的含量会变高,土壤剖面显示邻苯二甲酸酯的含量从土壤表层到深层呈现“高-低-高-低”的变化趋势,说明地下土壤中的邻苯二甲酸酯不仅来自垂直渗透,还来自地下,可能是地下水的水文状况对深层土壤的影响造成的。

此外,新污染物不仅可以向地表水、地下水和土壤中迁移,部分物质通过挥发作用还会扩散到大气中(图 6)。目前,对垃圾填埋场中新污染物的大气环境行为已有研究,主要集中在固体垃圾对大气新污染物的扩散影响,如 Tao 等<sup>[73]</sup>研究表明安徽亳州垃圾填埋场气溶胶中的 ARGs 对作业区域的环境有明显影响,并可能通过扩散影响周围环境;Tian 等<sup>[74]</sup>研究了天津两个不同的垃圾填埋场周围大气中全氟化合物的浓度,结果表明垃圾填埋场是环境中全氟化合物的重要来源。目前渗滤液对于大气中新污染物的扩散还鲜有研究。对于挥发性或半挥发性新污染物可以通过挥发性释放从渗滤液排放至大气环境中<sup>[75]</sup>,并因其具有持久性与远距离迁移潜能对大气环境及周边植物体等造成污染或损害。因此,关于渗滤液中新污染物的逸散研究值得重视,可以全面阐释垃圾填埋场对周围大气中新污染物的贡献。

### 4 垃圾填埋渗滤液中新污染物的去除(Removal of ECs from landfill leachate)

目前,我国垃圾渗滤液处理厂对渗滤液的处理主要包括“预处理+生化处理+深度处理”3 个处理单元,其主流的处理工艺为“预处理(格栅+隔油+调节池)+膜生物反应器(MBR+UF)+纳滤(NF)/反渗透(RO)”<sup>[76]</sup>。如表 3 所示,相较于渗滤液处理厂常规工艺,该工艺在实际运用中可有效去除垃圾渗滤液的 ECs。Yang 等<sup>[77]</sup>研究表明外置 MBR 与集成膜工艺结合在去除 ARGs 方面表现出色,其绝对丰度降低 3—6 个数量级;Yan 等<sup>[4]</sup>研究表明 MBR-NF/RO 系统可有效去除渗滤液中的全氟烷基酸,去除效率为 88.2%—99.4%;Zhang 等<sup>[60]</sup>研究表明 MBR+UF+NF/RO 系统对垃圾渗滤液中 MPs 去除效率为 75%;Sui 等<sup>[33]</sup>研究表明相较于原始渗滤液 PPCPs 浓度(0.39—349  $\mu\text{g}\cdot\text{L}^{-1}$ ),MBR+UF 工艺处理后的渗滤液 PPCPs 浓度( $<\text{LOQ}$ —10.6  $\mu\text{g}\cdot\text{L}^{-1}$ )明显降低。但该工艺在运行时仍存在诸多问题,如预处理过程不能有效地去除难生化降解的有机污染物<sup>[78-79]</sup>,生化处理过程对老龄渗滤液可生化性不高,运行过程中需加入大量碳源<sup>[80]</sup>,膜处理过程易堵塞且会对膜造成污染,同时会产生大量的膜浓缩液<sup>[81]</sup>等。因此,有必要探究新的渗滤液 ECs 去除技术以克服上述工艺存在的问题。

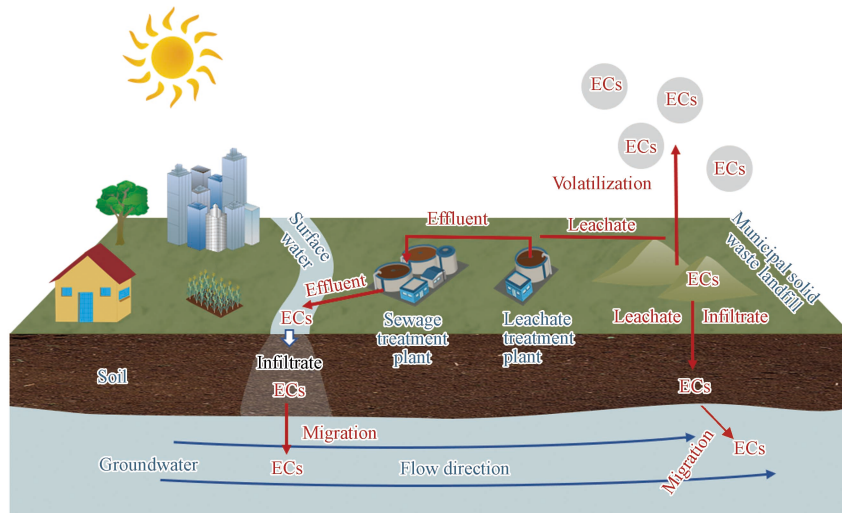


图 6 垃圾渗滤液中新污染物迁移示意图

Fig.6 Migration of ECs in landfill leachate

表 3 渗滤液处理厂 ECs 处理工艺

Table 3 The treatment process of ECs in leachate treatment plant

处理工艺 Treatment process	化合物 Compound	去除效率/% Removal efficiency	参考文献 References
A/O/O+MBR+NF	PFCs	74.71—100	[4]
UASB+A/O+MBR+RO	PFCs	89.07—100	[4]
两段式A/O+MBR+NF	PFCs	95.49—100	[4]
A/O/O+MBR+RO	PFCs	98.51—100	[4]
两段式A/O+MBR+RO	PFCs	99.35—100	[4]
MBR+DTRO	EDCs	61.2—100	[62]
两段式A/O+UF+RO	ARGs	97.74—99.74	[8]
氧化沟+颗粒活性炭	PPCPs	0—80	[26]
硝化+厌氧+臭氧化工艺	PPCPs	0—85	[26]
厌氧+硝化工艺	PPCPs	0—43	[26]
MBR+DTRO	PPCPs	98—100	[82]
塔式生物反应器	抗生素	76.75	[38]
卧式生物反应器	抗生素	48.36	[38]
MBR+UF	PPCPs	-1.5—100	[33]
MBR+CMF+RO	有机磷阻燃剂	98	[83]
生化处理	PFCs	43.3—93.2	[84]
生化处理+RO	PFCs	91.7—99.4	[84]
生化处理+MF+AC	PFCs	53.7—94.1	[84]
A/O+二沉池	有机磷酸酯	4.71—97.6	[85]
厌氧+缺氧+好氧	甲基硅氧烷	50—100	[86]
A/O+UF+RO	EDCs	97.4—98.7	[87]
MBR+NF+RO	有机磷阻燃剂	55.7—99.8	[88]
UASB+反硝化+硝化+UF+RO	PFCs	97.4—99.5	[67]
O/A/O+A/O生物盘+曝气池+生物沉淀池厌氧池+硝化+反硝化	PFCs	52—96	[89]
砂滤+DTRO+紫外消毒	PPCPs	87.2—100	[90]
A/O+MBR+UF+RO	抗生素	96—100	[28]
硝化+反硝化+UF+RO	EDCs	97.8—100	[63]

续表 3

处理工艺 Treatment process	化合物 Compound	去除效率/% Removal efficiency	参考文献 References
AO+AO/A <sup>2</sup> O	ARGs	32.81—99.99	[91]
MBR+A/O+UF+NF+RO	MPs	58.33	[60]
MBR+NF+RO	MPs	98.4	[54]
UASB+MBR+UF	ARGs	10.88—89.03	[92]

基于上述工艺的缺点,我国众多学者也再不断探索其他的垃圾渗滤液 ECs 去除工艺(表 4),如光化学催化、电化学催化、高级氧化和人工湿地等.利用低成本矿物填充臭氧鼓泡塔臭氧氧化垃圾渗滤液中阿特拉津,相较于传统的臭氧氧化工艺,其去除效率提高 27%<sup>[93]</sup>.如采用 Fenton 氧化去除垃圾渗滤液中 ARGs,其去除效率高达 99%<sup>[94]</sup>.通过处理垃圾渗滤液的污水再循环人工湿地系统去除有机污染物,可将多环芳烃浓度从 0.91—98.04 ng·L<sup>-1</sup> 降低到 0.65—38.72 ng·L<sup>-1</sup>,去除效率高达 85%<sup>[95]</sup>.采用 Fenton 氧化膜法浓缩垃圾渗滤液中的有机污染物,可将邻苯二甲酸二乙酯浓度从(1.37 ± 0.11) mg·L<sup>-1</sup> 降低到(0.28 ± 0.02) mg·L<sup>-1</sup>.此外,耦合工艺的关注点也越来越高,如使用颗粒活性炭催化臭氧系统同时降解垃圾渗滤液反渗透浓缩液中的多种 ECs,其去除了约 80%—100% 的抗生素和 ARGs<sup>[96]</sup>.采用 Fenton 氧化絮凝与光 Fenton 联用工艺去除纳滤浓缩液中的难降解有机物,对邻苯二甲酸酯和多环芳烃的去除率为 80%—90%<sup>[97]</sup>.目前,上述方法在去除 ECs 方面展现出巨大的潜力,但大部分处于室内模拟阶段,其实际意义还需进一步研究.

表 4 实验室规模 ECs 处理工艺

Table 4 The treatment process of ECs in laboratory scale

处理工艺 Treatment process	物质 Chemical	去除效率/% Removal efficiency	参考文献 References
电凝法	PFCs	33.8—75.2	[98]
芬顿氧化	EDCs	26.3—79.4	[99]
芬顿氧化+混凝+光芬顿	多环芳烃	80—90	[97]
硫酸盐煅烧蛋壳	ARGs	94.48—99.92	[100]
煤基磁性活性炭	PFCs	72.8—89.6	[101]
紫外+芬顿氧化	EDCs	100	[102]
GAC/O <sub>3</sub>	抗生素/ARGs	80—100	[96]
氯化	ARGs	54.3—77.6	[94]
芬顿氧化	ARGs	99.9	[94]
人工湿地	EDCs/多环芳烃	85	[95]

注: GAC, 颗粒活性炭. Note: GAC, Granular activated carbon.

## 5 展望(Prospects)

随着我国新污染物治理行动方案的发布实施,新污染物日益受到人们的广泛关注,但是总体的研究仍处于起步阶段,作为重要污染源和汇的垃圾填埋渗滤液中新污染物研究数据还十分有限,存在定性定量数据不全面、影响作用机制不明晰、环境迁移规律不明确、处理技术不完善等问题,因此,后续研究可从以下方面开展工作:

(1) 新污染物种类和数量众多,目前缺少高通量的非靶向筛查方法全面识别新污染物类别,同时现有文献建立的定量监测方法缺乏标准化的方法体系,对于基质复杂的渗滤液组分无法准确验证检测数据的有效性,特别是对于 MPs 与 ARGs,建立新污染物标准监测方法已经成为亟待解决的首要任务.

(2) 由于我国各地经济发展水平、产业结构、消费模式、人口密度等社会因素,以及气候、降雨等自然环境因素的差异,尚需全面开展不同区域垃圾填埋渗滤液中新污染物的赋存特征研究,同时系统性研究阐明这些因素的影响程度及其作用机制,以期全面评估垃圾填埋体系新污染物污染水平提供

基础数据。

(3) 新污染物通过垃圾渗滤液排放或渗透等方式进入周边环境, 迁移扩散机制缺少系统性和全面性研究, 尤其对于地下水和大气迁移, 存在地下水水文地质环境复杂以及大气样品收集困难等问题, 开展填埋场周边环境介质中新污染物全方位监测研究, 对于加强其环境安全至关重要。

(4) 现有生物处理结合膜处理技术的渗滤液处理工艺, 可以有效处理多数新污染物, 但是去除机制及沿程演变规律尚未明晰, 此外, 仍需研发经济性高、循环利用性强、可操作性的新技术和新工艺, 高效低耗能的控制渗滤液中新污染物。

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