採 佐 神 草 (HUANJING KEXUE)

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目 次

持久性、迁移性和潜在毒性化学品环境健康风险与控制研究现状及趋势分析 张少轩,陈安娜,陈成康,景侨楠,刘建国(3017) 我国厨余垃圾资源化技术的多维绩效评价 杨光,史波芬,周传斌(3024) 基于 MSPA 和电路理论的京津冀城市群热环境空间网络	
基于 MSPA 和电路理论的京津冀城市群热环境空间网络	
乔治,陈嘉悦,王楠,卢应爽,贺疃,孙宗耀,徐新良,杨浩,李莹,王方(3034)城市空间格局与热环境响应关系:以合肥市区为例	
天津市"十三五"期间 PM _{2.5} 减排效果评估 肖致美,徐虹,蔡子颖,张裕芬,刘茂辉,孙猛,李鹏,杨宁,戢运峰(3054)	
一清洁取暖对保定而采暖期 PM,中磋店与浴胶的影响 岁字基 张凯 赵妤希 任家豪 段菁素 李欢欢 美健 乳志區 李博文(3063)	
大气环流型对珠三角 2015~2020 年臭氧变化的影响	
南京地区细颗粒物污染输送影响及潜在源区 谢放尖,郑新梅,窦焘焘,杨峰,刘春蕾,李洁,谢轶嵩,王艳,胡建林,陈长虹(3071)大气环流型对珠三角 2015~2020 年臭氧变化的影响	
$2010 \sim 2020$ 年成都市在制 $1M_{2.5}$ 相 $0_{3.8h}$ 行来的健康双盘开刊 108 分 深圳市 2022 年春季新冠疫情管控期间空气质量分析 108 分	
西安市大气降水的主要化学组分及其来源 周东,黄智浦,李思敏,王森,牛振川,熊晓虎,冯雪(3142)	
世界的 2022年至于的 2022年至于	
无定河流域地表水硝酸盐浓度的时空分布特征及来源解析	
太福河水体与机快物中重壶属的学节变化付证与75条件价 … 夕鹛在,赤难仁,称婷婷,对生辉,尚佳欣,戏佳怡,顾心龙,权艳干(5184) 北京市北运河水体中抗生素污染特征及风险评估 …	
淮河下游湖泊表层水和沉积物中 PPCPs 分布特征及风险评估	
四宁市浅层地下水化学特征及形成机制····································	
密云水库细菌群落组成结构及影响因素	
- 纳米季价铁改性生物灰对水甲氨氮的吸附特性及机制 除文静,石峻岭,李雪婷,张李金,对富强,除止祝,龙维海,杨殿海(3270)	
喜经酚钾改性椰壳生物岩对水中Cd(Ⅱ)和N;(Ⅱ)的丰除性能及机制 张凤恕 王翦珑 曹昆注 刘桥京 兵进进 刘立恒(3278)	
網改性净水污泥水热炭对水体中磷的吸附特性及底泥内源磷的固定 ············何李文泽,陈钰,孙飞,李艳君,杨顺生,张志鹏(3288)城镇生活污水处理厂出水硝酸盐浓度及同位素组成的影响因素	
我国自然生态系统氮沉降临界负荷评估 黄静文, 刘磊, 颜晓元, 遗超普 (3321)	
气恢变化和人尖活动对东部沿海地区 NDV1 变化的影响分析 金岩松,金凯,土飞,对春葭,秦鹏,宗全利,对佩如,陈明利(3329) 基于 InVEST 模型和 PLUS 模型的环杭州湾生态系统碳储量 丁岳,王柳柱,桂峰,赵晟,朱望远(3343)	
基于 Meta 分析的污水处理工艺对微塑料去除效果影响 符立松,侯磊,王艳霞,李晓珠,王万宾,梁启斌(3309)我国自然生态系统氮沉降临界负荷评估 符立松,侯磊,王艳霞,李晓珠,王万宾,梁启斌(3309)我国自然生态系统氮沉降临界负荷评估 卷岩松,金凯,王飞,刘春霞,秦鹏,宗全利,刘佩茹,陈明利(3329)基于 InVEST 模型和 PLUS 模型的环杭州湾生态系统碳储量 丁岳,王柳柱,桂峰,赵晟,朱望远(3343)河西走廊中段荒漠绿洲土壤生态化学计量特征 孙雪,龙永丽,刘乐,刘继亮,金丽琼,杜海峰,陈凌云(3353)乌梁素海东部流域非生长季草地土壤细菌群落结构的垂向差异 李文宝,张博尧,史玉娇,郭鑫,李兴月(3364)芦芽山华北落叶松林土壤剖面细菌群落分布格局 老晓雅,刘晋仙,贾彤,吴铁航,柴宝峰(3376)植被类型对黄土高原露采矿山复垦土壤碳循环功能基因的影响 赵姣,马静,朱燕峰,于昊辰,张琦,陈浮(3386)施用生物炭对麦田土壤细菌群落,对麦生长的影响	
与采系海尔部流域非任大学早地工集细图群洛结构的垂间差异····································	
植被类型对黄土高原露采矿山复垦土壤碳循环功能基因的影响 赵姣,马静,朱燕峰,于昊辰,张琦,陈浮(3386)	
一 明光日中国年度列上发展图针役印刷 —————本卫红,胜初风,处为对,内阁观,瓜门田,内云云,瓜入石,不几刀,尽刀刀(J 1 00)	
生物炭对热带地区辣椒种植土壤N ₂ O排放及其功能基因的影响 ····································	
覆膜和有机无机配施对夏玉米农田温室气体排放及水氮利用的影响 蒋洪丽、雷琪、张彪、吴淑芳(3426)	
不同类型地膜覆盖对土壤质量、根系生长和产量的影响 ············穆晓国,高虎,李梅花,赵欣茹,郭宁,靳磊,李建设,叶林(3439)基于 PMF 模型的某铅锌冶炼城市降尘重金属污染评价及来源解析 ····································	
云南 5 城市道路扬尘 PM _{2.5} 中重金属含量表征及健康风险 ····································	
PMF 和 RF 模型联用的土壤重金属污染来源解析与污染评价:以西北某典型工业园区为例	
基于 APCS-MLR 受体模型和地统计法的矿区周边农用地土壤重金属来源解析 张传华,王钟书,刘力,刘燕(3500)	
PCA-APCS-MLR 和地统计学的典型农田土壤重金属来源解析	
皖江经济带耕地重金属健康风险评价及环境基准	
PCA-APCS-MLR 和地坑订字的典型农田土壤重金属来源解析	
矿业废弃地重金属形态分布特征与迁移转化影响机制分析····································	
- 个回种尖疏采里金属晶集特值及健康风险 ····································	
山东省典型污灌区土壤-小麦重金属健康风险评估 ··········王菲,费敏,韩冬锐,李春芳,曹文涛,姚磊,曹见飞,吴泉源(3609)基于机器学习方法的小麦镉富集因子预测 ··················	
《环境科学》征订启事(3062) 《环境科学》征稿简则(3116) 信息(3164, 3259, 3572)	



基于 Meta 分析的污水处理工艺对微塑料去除效果影响

符立松1,侯磊1,王艳霞1,李晓琳1,王万宾2,梁启斌1*

(1. 西南林业大学生态与环境学院, 昆明 650224: 2. 云南省生态环境科学研究院, 昆明 650034)

摘要:污水处理厂(WWTPs)是微塑料"源-汇"过程的重要场所,污水处理工艺和微塑料特性对微塑料去除效果的影响有待深入探讨. 综述了 57 篇文献中 78 个 WWTPs 的微塑料赋存特征,基于 Meta 分析探讨了不同污水处理工艺和微塑料赋存特征对其去除效果的影响. 结果表明:①进水和出水中微塑料的丰度范围分别为 $1.56 \times 10^{-2} \sim 3.14 \times 10^4$ n·L⁻¹和 $1.70 \times 10^{-3} \sim 3.09 \times 10^2$ n·L⁻¹,污泥中的微塑料赋存丰度范围为 $1.80 \times 10^{-1} \sim 9.38 \times 10^3$ n·g⁻¹;②以氧化沟、生物膜和传统活性污泥为核心工艺的 WWTPs 对微塑料的总去除率(>90%)优于序批式活性污泥、厌氧-缺氧-好氧和缺氧-好氧工艺;③一级处理对微塑料的去除率(62.87%)略高于二级处理(55.78%)和三级处理(58.45%),"格栅+沉砂池+初沉池"是一级处理中去除率最高的工艺组合,膜生物反应器在二级处理中去除率最高,过滤为三级处理最佳技术;④WWTPs 对薄膜、泡沫和碎片微塑料的去除率(>90%),高于纤维(88.36%)和球状(87.23%),粒径>0.5 mm的微塑料比<0.5 mm的更容易去除,WWTPs 对聚乙烯(PE)、聚对苯二甲酸乙二醇酯(PET)和聚丙烯(PP)的去除效果较好(>80%).

关键词:微塑料: Meta 分析: 污水处理工艺; 赋存特征; 去除效果

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Effects of Wastewater Treatment Processes on the Removal Efficiency of Microplastics Based on Meta-analysis

FU Li-song¹, HOU Lei¹, WANG Yan-xia¹, LI Xiao-lin¹, WANG Wan-bin², LIANG Qi-bin¹*

(1. College of Ecology and Environment, Southwest Forestry University, Kunming 650224, China; 2. Yunnan Research Academy of Eco-environmental Sciences, Kunming 650034, China)

Abstract: Microplastics (MPs) are ubiquitous emerging pollutants that have been found in the marine, freshwater, air, and soil environments. Wastewater treatment plants (WWTPs) play an important role in releasing MPs to the environment. Therefore, understanding the occurrence, fate, and removal mechanism of MPs in WWTPs is of great importance towards microplastic control. In this review, the occurrence characteristics and removal rates of MPs in 78 WWTPs from 57 studies were discussed based on Metanalysis. Specifically, the key aspects regarding MPs removal in WWTPs, such as wastewater treatment processes and MPs shapes, sizes, and polymer compositions were analyzed and compared. The results showed that: ① the abundances of MPs in the influent and effluent were $1.56 \times 10^{-2} - 3.14 \times 10^4 \,\mathrm{n \cdot L^{-1}}$ and $1.70 \times 10^{-3} - 3.09 \times 10^2 \,\mathrm{n \cdot L^{-1}}$, respectively. The abundance of MPs in the sludge ranged from 1.80×10^{-1} to $9.38 \times 10^3 \,\mathrm{n \cdot g^{-1}}$. ② The total removal rate (>90%) of MPs by WWTPs using oxidation ditch, biofilm, and conventional activated sludge treatment processes was higher than that using sequencing batch activated sludge, anaerobic-anoxic-aerobic, and anoxic-aerobic processes. ③ The removal rate of MPs in primary, secondary, and tertiary treatment process were 62.87%, 55.78%, and 58.45%, respectively. The combination process of "grid + sedimentation tank + primary sedimentation tank" had the highest removal rate towards MPs in primary treatment processes, and the membrane bioreactor had the highest one beyond other secondary treatment processes. Filtration was the best process in tertiary treatment. ④ The film, foam, and fragment MPs were easier to remove (>90%) than fiber and spherical (<90%) MPs by WWTPs. The MPs with particle size larger than 0.5 mm were easier to remove than those with particle size smaller than 0.5 mm. The removal efficiencies of polyethylene (PE), polyethylene terephthalate (PET), and polypropylene (PP) MPs were higher than 80%. **Key words**: micr

微塑料广泛存在于水体^[1,2]、土壤^[3]、大气^[4]和沉积物^[5,6]等环境介质中,甚至在人体粪便^[7]、肺部组织^[8]和血液^[9]中均检出微塑料颗粒,可能导致潜在的健康风险而备受关注.目前,针对微塑料污染特征、输移过程、生态效应和复合污染等方面开展了广泛研究,污水处理厂(WWTPs)是微塑料的"汇",同时也是自然环境中微塑料重要的"源",是影响微塑料输移过程的重要环节^[10~12].因此,充分了解微塑料在污水处理厂中的赋存特征及其去除状况,对于微塑料的污染管控至关重要.

国内外学者对单个污水处理厂开展大量研究, 但不同区域的污水处理厂中微塑料赋存特征和去除 率差异较大,如韩国某污水处理厂进水中微塑料丰 度高达 $3.14 \times 10^4 \text{n·L}^{-1[13]}$,中国珠江口的污水处理厂进水丰度仅为 $1.56 \times 10^{-2} \text{n·L}^{-1[14]}$;在 Ziajahromi等[15]的研究中,澳大利亚的 3 个污水处理厂总去除率均超过 98%,而在 Do 等[16]的研究结果中,越南的 3 个污水处理厂分别为 25.5%、21.8%和25.3%.因此,单一污水处理厂研究结果很难准确反映不同污水工艺对微塑料的去除效果. Meta 分析是基于文献资料定量化综合评价多个同

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作者简介: 符立松(1996~),男,硕士研究生,主要研究方向为微塑 料环境行为,E-mail; fls1348623800@126.com

* 通信作者,E-mail:qbliang@swfu.edu.cn

类独立研究结果的统计学方法,广泛应用于生态环境领域,Liu等^[17]将风险比(risk ratio,RR)作为Meta分析的效应量,探讨了污水处理厂分级处理对微塑料的去除效果,并深入分析去除机制,但未开展污水处理厂中微塑料的总去除率和微塑料聚合物组成对去除效果的影响研究.

本研究采用单组率的 Meta 分析方法探讨污水处理厂对微塑料的去除效果,将微塑料去除率作为 Meta 分析的效应量,可更加直观地反映污水处理工艺、微塑料粒径、形状和聚合物类型等因素对微塑料去除效果的影响. 本文基于 57 篇文献中 78 个污水处理厂的微塑料赋存特征,借助 Meta 分析探究污水处理厂总体、分级处理和工艺单元对微塑料的去除效果,讨论了污水处理厂对形状、粒径和聚合物类型的去除效果. 本研究结果可为深入理解微塑料在污水处理厂中的迁移过程和截留机制奠定基础,以期为高效去除微塑料的污水处理工艺选择及微塑料管控提供参考.

1 材料与方法

1.1 数据收集

基于中国知网(CNKI)和 Web of Science(WOS)数据库,以"微塑料(microplastics, MPs)"、"污水处理厂(wastewater treatment plants, WWTP)"和"污水处理厂(sewage treatment plants, STP)"等为主题词检索公开发表文献,文献数据截至 2022 年 4 月 1 日,并对检索结果进行筛选.文献保留需具备以下信息:①处理工艺名称和进出水微塑料丰度;②微塑料聚合物类型、形状和粒径占比数据;③污水处理厂基本信息(区位和日处理能力等).基于以上原则并剔除重复研究,共获得 57 篇文献,其中 22 篇文献涉及我国污水处理厂.

1.2 数据分类

本文将污水和最终排泥(按干重计)中的微塑料丰度单位统一为:n·L⁻¹和n·g⁻¹,统计污水中检出频次超过7次的特定形状、颜色和聚合物类型的微塑料丰度;污泥中统计检出频次3次及以上的.现有文献对微塑料形状的命名和粒径范围的界定不统一,为便于统计分析,将线状和纤维状统称为纤维,片状、块状和碎片状统称为碎片,薄片和薄膜统称为薄膜,颗粒、微珠和球形统称为球状,泡沫和海绵状统称为泡沫;将微塑料粒径范围统一为4个区间:<0.1、0.1~0.5、0.5~1和1~5 mm. 污水处理厂对微塑料的总去除率按工艺类型分组进行Meta分析,某工艺出现频次超过3篇文献的单列分组,而将小于3篇文献的工艺归于对应的工艺分组,

具体如下:氧化沟(oxidation ditch,OD)、生物膜法(biofilm)、传统活性污泥法(conventional activated sludge,CAS)、序批式活性污泥法(sequencing batch reactor,SBR)、厌氧-缺氧-好氧工艺(anaerobicanoxic-oxic,A²O)和缺氧-好氧工艺(anoxic-oxic,AO);分级去除率按一级处理、二级处理和三级处理进行分组;各工艺单元(构筑物和构筑物组合)的去除率根据单个构筑物或构筑物组合进行分组.

1.3 Meta 分析

将污水处理厂对微塑料的去除率 (R_i) 指定为 Meta 分析的效应量,参照 Erni-Cassola 等 $^{[18]}$ 和 Lim 等 $^{[19]}$ 的计算方法,将进水的微塑料丰度作为总体样本数量 (A_{in}) ,合并效应量 (R) 为单个污水处理厂去除率的加权平均值,见式 (1) 和式 (2). 经过"Shapiro-Wilk test"正态检验后,选择接近正态分布的样本估计方法即反正弦转换 (PAS) 进行 Meta 分析. 通过 I^2 统计量检验方法对效应量数据进行异质性检验 $^{[20]}$,计算方法见式 (3) 和式 (4). 除泡沫微塑料分组的去除率数据外,本研究所有分组的效应量数据的异质性检验结果均满足 $I^2 \geq 50\%$,表明各研究结果间存在异质性,采用随机效应模型进行合并及亚组分析以进一步检测异质性来源 $^{[21]}$.

$$R_i = (A_{\rm in} - A_{\rm out})/A_{\rm in} \times 100\%$$
 (1)

$$R = \sum_{i=1}^{n} W_i R_i \tag{2}$$

$$Q = \sum_{i=1}^{n} W_{i} (R_{i} - \overline{R}_{i})^{2}$$
 (3)

$$I^2 = \lceil Q - (n-1) \rceil / Q \tag{4}$$

式中, R_i 表示第 i 个污水处理厂对微塑料去除率,%,即效应量; A_{in} 和 A_{out} 分别表示第 i 个污水处理厂或工艺单元进水和出水的微塑料丰度, $n \cdot L^{-1}$;R 表示合并效应量; W_i 表示第 i 个污水处理厂的权重;Q 表示检验统计量; \overline{R}_i 表示所有污水处理厂效应量的均值;n 表示效应量样本数.

1.4 数据处理

使用 GetData Graph Digitizer 2.25 提取文献中图的进、出水微塑料污染特征丰度和去除率数据,并用 Microsoft Excel 2019 软件建立数据库.在 SPSS 25.0 中采用最小显著差异法(LSD)完成数据差异性分析(P < 0.05). Meta 分析基于 R 语言的 Meta 包完成,并运用 Origin 2020 软件绘图.采用漏斗图和 Egger's 法对文献的发表偏倚性进行定性和定量分析[22],检验结果显示,漏斗图呈现对称且检验值 P > 0.05,表明所收集的文献和数据不存在发表偏倚.

2 结果与分析

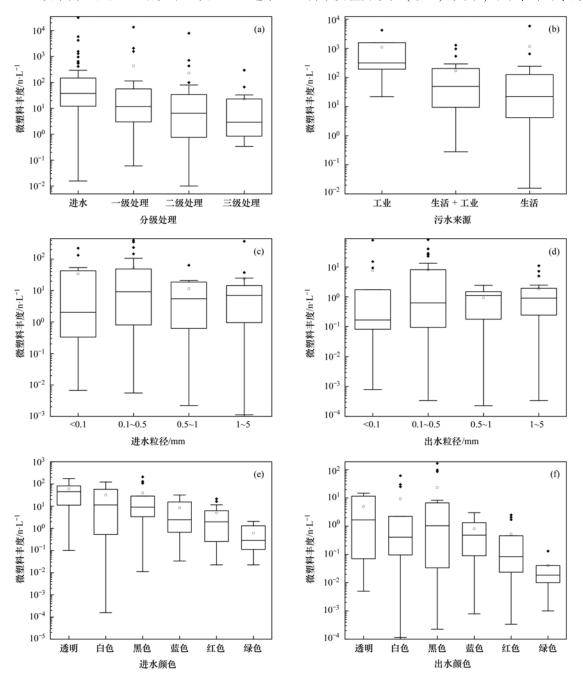
2.1 微塑料在污水处理厂的赋存特征

2.1.1 污水中微塑料的赋存特征

文献综述结果显示,国内外污水处理厂进水的 微塑料丰度差异较大,在 $1.56 \times 10^{-2} \sim 3.14 \times 10^4$ $n \cdot L^{-1}$ 之间,均值为 636.30 $n \cdot L^{-1}$,中值为 37.59 $n \cdot L^{-1}$ [图 1(a)].以生活污水为主要处理对象的污水处理厂,其进水微塑料丰度中值为 22.15 $n \cdot L^{-1}$,低于以工业废水为主要处理对象的污水处理厂进水

丰度中值(317.50 $n \cdot L^{-1}$),生活污水和工业废水混合的进水丰度中值为 49.24 $n \cdot L^{-1}$,介于前两者之间[图 1(b)].从一级处理到三级处理,微塑料丰度逐渐降低[图 1(a)],污水处理厂最终出水微塑料丰度在 $1.70 \times 10^{-3} \sim 3.09 \times 10^2 n \cdot L^{-1}$ 之间,均值为22.94 $n \cdot L^{-1}$,中值为 $1.97 n \cdot L^{-1}$; 进水微塑料丰度均值为出水的 27.74 倍,表明污水处理厂具有较高的微塑料去除能力.

污水处理厂的进、出水中检出不同形状的微塑料丰度差异较大(表1).其中,纤维、碎片、薄膜和



- (a)进水和不同处理阶段出水中微塑料的丰度;(b)不同污水来源进水中的微塑料丰度;
- (c)和(d)进、出水中不同粒径区间的丰度;(e)和(f)进、出水中微塑料不同颜色的丰度

图 1 污水处理厂进、出水中微塑料的赋存特征

Fig. 1 Occurrence characteristics of microplastics in the influent and effluent of wastewater treatment plants

球状是进水中赋存较多的微塑料形状,其占比最高值分别为 $91.97\%^{[23]}$ 、 $93.73\%^{[24]}$ 、 $73\%^{[25]}$ 和

70. $40\%^{[13]}$,纤维和碎片的丰度均显著高于薄膜、球状和泡沫(P < 0.05).

表 1 污水处理厂中不同形状微塑料的丰度1)

Table 1 Abundance of microplastics with different shapes in wastewater treatment plants

形状	进水丰度/n·L ⁻¹	进水占比/%	出水丰度/n·L-1	出水占比/%	检出频次
纤维	3. 36 ~4 600	6. 27 ~ 91. 90	2. 28 × 10 ⁻⁴ ~ 107. 66	5. 60 ~ 100	62
碎片	0. 11 ~ 3 400	3. 62 ~ 93. 70	$1.36 \times 10^{-3} \sim 101.01$	0. 67 ~ 80	54
薄膜	0. 35 ~ 1 300	2 ~ 73	0. 01 ~ 29. 32	0. 67 ~ 72. 46	31
球状	0. 03 ~22 100	0. 43 ~ 70. 40	$2.23 \times 10^{-4} \sim 277$	1. 16 ~93. 20	25
泡沫	0.06 ~21.56	1 ~ 21. 40	ND ~ 15. 67	ND ~ 5. 07	7

1) ND 表示未检出;进水丰度和出水丰度分别表示进、出水中某形状微塑料的丰度;进水占比和出水占比分别表示进、出水中某形状的占比; 检出频次表示某形状在进水中检出的次数

污水处理厂进、出水中不同粒径微塑料丰度如图 1(c) 和图 1(d) 所示, $0.1 \sim 0.5$ mm 为赋存最多的微塑料粒径区间,其次为 < 0.1 mm,进水的丰度均值分别为 52.15 n·L⁻¹和 33.55 n·L⁻¹,而出水的丰度均值则分别为 8.03 n·L⁻¹和 7.73 n·L⁻¹. 污水处理厂中检出各种颜色的微塑料丰度如图 1(e) 和图 1(f) 所示,在 WWTP 中检出的微塑料颜色主要有 6 种,进水中透明的丰度均值最高,为 59.61

 $n \cdot L^{-1}$. 污水处理厂中赋存的微塑料聚合物类型众多,检出频次从高至低依次为聚乙烯(PE)、聚丙烯(PP)、聚酯(PES)、聚对苯二甲酸乙二醇酯(PET)、聚酰胺(PA)、聚苯乙烯(PS)和聚氯乙烯(PVC)是污水处理厂进水中赋存最广泛的聚合物(表 2),其最高占比分别为 57.10% [14]、35.91% [26]、89.66% [23]、83.33% [15]、71.43% [27]、37.5% [28]和52.5% [29].

表 2 污水处理厂主要检出的聚合物类型1)

Table 2 Mainly detected polymer types in wastewater treatment plants

聚合物名称	密度 ^[30] /g·cm ⁻³	进水丰度 /n·L ⁻¹	进水占比/%	出水丰度 /n·L ⁻¹	出水占比/%	检出频次
PE	0.89 ~ 0.98	0. 09 ~ 541	4. 08 ~ 57. 10	$1.25 \times 10^{-3} \sim 59.95$	3. 19 ~ 73. 30	28
PB/a	0.92	0. 10 ~ 97. 30	2. 50 ~ 35. 91	$2.23 \times 10^{-4} \sim 33.06$	2. 90 ~ 29. 60	23
PET	0.96 ~ 1.45	0.02 ~ 123.64	1. 82 ~ 83. 30	4. $47 \times 10^{-4} \sim 98.88$	4 ~ 88. 54	17
PES	1. 24 ~ 2. 30	0.02 ~618.21	1. 60 ~ 89. 66	$2.23 \times 10^{-4} \sim 18.72$	2. 90 ~ 100	17
PA	1. 02 ~ 1. 16	0. 05 ~43. 37	2 ~71.43	0.01 ~35.84	1. 23 ~40	15
PS	1.05	0. 01 ~91. 50	1. 90 ~ 37. 50	0. 01 ~ 12. 36	4 ~45.94	14
PVC	1. 16 ~ 1. 58	0. 07 ~ 712. 42	1. 30 ~ 52. 50	6. 56 ~ 42. 96	9. 30 ~ 40	13

1)进水丰度和出水丰度分别表示进、出水中单种聚合物的丰度;进水占比和出水占比分别表示进、出水中单种聚合物的占比;检出频次表示单种聚合物在进水中检出的次数

2.1.2 污泥中微塑料的赋存特征

污水处理厂最终排泥中的微塑料污染特征如图 2 所示. 丰度在 $1.80 \times 10^{-1} \sim 9.38 \times 10^{3} \, \text{n·g}^{-1}$ 之间; CAS 工艺污泥中的微塑料丰度均值最高 (1384.91 n·g^{-1}),是丰度最低的 SBR 工艺的 184.65 倍,其余工艺的污泥微塑料丰度数量级相当. 污泥中赋存最多的形状为纤维 (219.01 n·g^{-1}),其次为球状 (19.32 n·g^{-1}) 和碎片 (13.69 n·g^{-1}); $0.1 \sim 0.5$ mm 在污泥中的丰度均值最高 (20.45 n·g^{-1}),其次是 <0.1 mm (6.48 n·g^{-1})即粒径 <0.5 mm 的微塑料占主要地位. 污泥中检出的主要颜色则为透明 (20.24 n·g^{-1}),其次是黑色 (14.59 n·g^{-1}),PET 在污泥中检出的丰度最高 (7.23 n·g^{-1}),其次为 PA (5.17 n·g^{-1})和 PES (3.97 n·g^{-1}),最低为 PP (1.47 n·g^{-1}).根据污泥中微塑料丰度和排泥量的统计结果,得到污水处理厂通过污泥外排微塑料数量在

4. 23×10⁷~1. 51×10¹¹n·d⁻¹之间.

2.2 不同工艺污水处理厂对微塑料的去除效果

不同工艺的污水处理厂总去除率 Meta 分析结果如图 3(a) 所示,总去除率大小依次为: OD (93.40%) > biofilm (92.28%) > CAS (90.84%) > SBR (84.91%) > $A^2O(81.41\%)$ > AO (71.31%).各级处理中工艺单元对微塑料去除率如图 3(b) 所示,一级处理对微塑料的去除率最高(62.87%),其次为三级处理(58.45%)和二级处理(55.78%).

在一级处理中,格栅、沉砂池和初沉池的工艺组合对微塑料的去除效果最好,去除率为78.47%,其次为沉砂池与初沉池的工艺组合(65.65%)和沉砂池与格栅组合(45.30%),单独使用沉砂池的去除率仅为13.88%,初沉池对微塑料的去除率比沉砂池的高,为43.82%.在二级处理中,MBR工艺

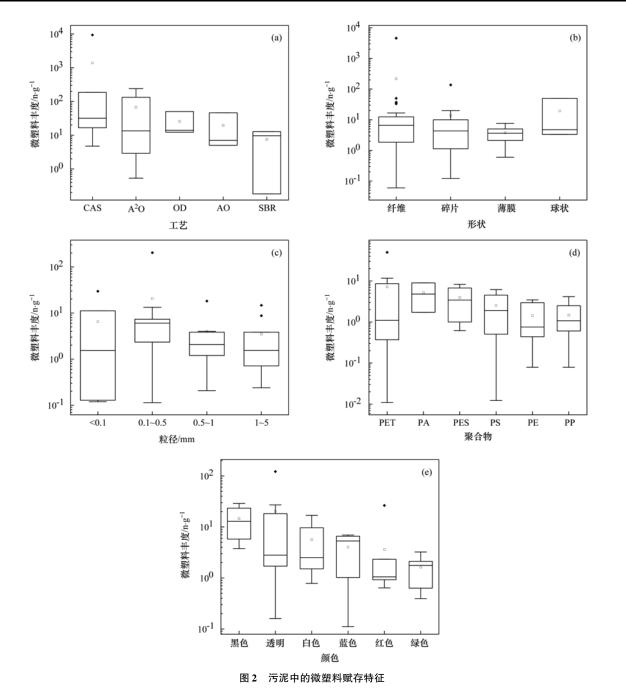


Fig. 2 Occurrence characteristics of microplastics in sludge

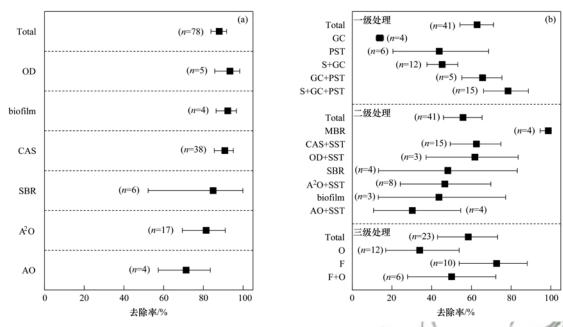
(membrane bioreactor)对微塑料的去除效果最优,去除效率高达98.74%. CAS(62.52%)和 OD(61.77%)两个工艺单元去除率较为相近,其余工艺单元去除率分别为 SBR(48.14%)、A²O(46.61%)、biofilm(43.68%)和 AO(30.27%).在三级处理中,过滤技术(72.71%)对微塑料的去除率最高,深度氧化工艺的最低(33.99%),而两者组合的去除率居中(50.04%).

2.3 污水处理厂对不同形状、粒径和聚合物类型 微塑料的去除效果

如图 4(a) 所示, 污水处理厂对不同形状的去除率均高于 80%, 其中对薄膜微塑料的去除率最高

(94.59%), 其次是泡沫(91.18%)和碎片(90.89%),而纤维(88.36%)和球状(87.23%)的去除率相对较低.在分级处理中,一级处理(60.91%)和二级处理(62.70%)对纤维的去除率稍高于三级处理(57.39%),二级处理对碎片的去除率最高(81.30%),其次为三级处理(71.66%),一级处理(60.67%)相对较低.薄膜和球状随处理分级的增加去除率逐渐升高,三级处理去除率分别为90.39%和78.47%.

污水处理厂对不同粒径的去除效果如图 4(b) 所示, $0.5 \sim 1 \text{ mm}(90.63\%)$ 和 $1 \sim 5 \text{ mm}(89.87\%)$ 的总去除率最高, $0.1 \sim 0.5 \text{ mm}(83.84\%)$ 和 < 0.1



(a)不同工艺污水处理厂对微塑料的总去除率,(b)分级处理和工艺单元对微塑料的去除率; Total 表示全组,GC 表示沉砂池, PST 表示初沉池,S 表示格栅,SST 表示二沉池,O 表示深度氧化,F 表示过滤技术,n 表示样本数

图 3 不同处理工艺去除微塑料的 Meta 分析结果

Fig. 3 Meta-analysis results of microplastic removal efficiencies by different treatment processes

mm(81.63%)的相对较低. 在分级处理中,一级处理和二级处理对 < 0.1 mm 的去除率相当,三级处理最高(70.74%). 0.1 ~ 0.5 mm 的去除率随分级呈升高趋势,三级处理去除率为 85.56%. 一级处理和三级处理对 0.5 ~ 1 mm 的去除率相近,二级处理最低(52.05%).1~5 mm 则与 0.1~0.5 mm 呈相反趋势,最高为一级处理(88%). 污水处理厂对不同聚合物的去除效果如图 4(c)所示,PE、PET、PP、PVC和 PES 的总去除率均高于 79%,PA 和 PS 的相对较低,去除率分别为 62.27% 和 55.57%.

3 讨论

3.1 污水处理厂中微塑料的污染特征

不同污水处理厂的微塑料丰度差异受服务人口、采样季节、污水来源(生活或工业)、社会经济和生活方式等因素的影响. Zhang 等^[31]分析结果显示,污水处理厂进水微塑料丰度与服务人口、服务面积和第三产业(游客数量)呈显著正相关(P < 0.01). Yuan 等^[32]发现污泥中的微塑料丰度与工业废水比例、服务人口数量和区域经济发展水平呈正比. 泰国某污水处理厂在旱季和雨季污水样品中检出的微塑料丰度范围分别为 76~192 n·L⁻¹和 36~68 n·L⁻¹,并且在雨季的最终出水中未检测到微塑料^[33]. Long 等^[34]分析了不同进水来源的微塑料丰度,结果表明工业废水中微塑料丰度(2.56 n·L⁻¹)是生活污水(1.44 n·L⁻¹)的 1.8 倍. 此外,样品采集、预处理和表征分析等方法也会影响污水处理厂

进水微塑料丰度的统计. 尽管出水中微塑料丰度相对较低,但污水处理量大,出水中微塑料的排放总量仍然很高,排放到水体环境的微塑料在 8.48×10^4 ~ 1.46×10^{11} n·d ⁻¹之间,均值(3.79×10^9 n·d ⁻¹)约为 Liu 等 ^[17] 研究结果(7.2×10^9 n·d ⁻¹)的 50%,为 Cheng 等 ^[35]的(1.6×10^9 n·d ⁻¹) 两倍有余,而中值(2.72×10^7 n·d ⁻¹)比 Sun 等 ^[10]的研究结果(2×10^6 n·d ⁻¹)高一个数量级. 因此,污水处理厂尾水的排放被认为是自然水体中微塑料的重要来源.

污水中的微塑料大部分被截留于污泥中,且与 处理工艺的去除率有关[10,32,36]. 本文统计最终排泥 中的微塑料丰度(1.80×10⁻¹~9.38×10³n·g⁻¹)数 值范围比 Cheng 等[35] 估计的 (9.70 ~ 186.70 n·g⁻¹)更宽泛,不同污水处理厂对微塑料的去除效 果差异较大. 纤维、碎片和球状为污泥中赋存较多 的微塑料形状(见图2),其中纤维和碎片易在沉降 过程被截留至污泥中[37-39],而球状易被吸附至污泥 中[40]. 粒径 < 0.5 mm 的微塑料因其比表面积相对 较大,易被污泥吸附并迁移至其中[37,40,41]. PET 在污 泥中赋存最多,是因为密度较高的微塑料(见表2) 更容易从污水沉降至污泥中[40,42]. 尽管污水中大部 分的微塑料被去除至污泥中,但污泥的资源化利用 和不当处置仍会导致大量的微塑料以多种方式排放 到环境中. 据报道,全球不同国家的污水处理厂每年 有 1.0×10⁶~1.0×10¹⁴n·a⁻¹微塑料通过污泥排放 到环境中[43]. 污泥中的微塑料可通过填埋、焚烧和 农田回用等方式进入自然环境,其中,农田回用为主

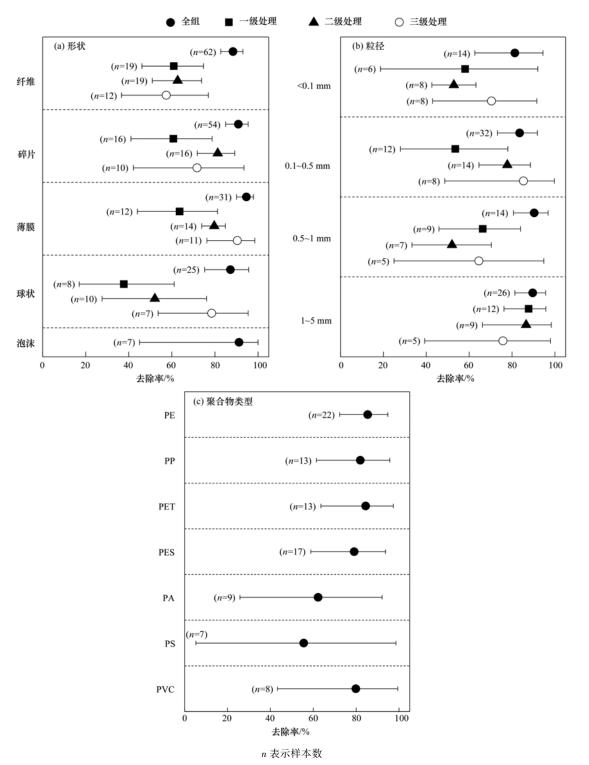


图 4 微塑料形状、粒径和聚合物类型对污水处理厂去除率影响的 Meta 分析结果

Fig. 4 Meta-analysis results of effects of microplastics shape, particle size, and polymer composition on their removal efficiencies by wastewater treatment plants

要利用方式. Koutnik 等^[44]研究表明,美国全国有51%的污泥用于农业回用,22%为填埋场填埋,16%污泥焚烧,11%为其他处置方式;农业回用释放微塑料为 $7.85 \times 10^{14} \sim 1.08 \times 10^{15} \,\mathrm{n\cdot a^{-1}}$. 西班牙的马德里地区,因污泥农用导致微塑料进入土壤环境的数量为 $1.0 \times 10^{13} \,\mathrm{n\cdot a^{-1[45]}}$. 因此,污泥的农业回用被认为是农田土壤微塑料的主要来源之—^[46].

3.2 污水处理厂对微塑料的去除机制

一级处理中,微塑料的去除机制主要通过格栅的截留、沉砂池和初沉池的沉降来实现.格栅能够截留大尺寸的微塑料,如细格栅的栅条间距为 2.5 ~10 mm,可去除粒径 > 2.5 mm 的微塑料^[47],而很难截留小粒径的.沉砂池对微塑料去除效果较差,仅对高密度微塑料有一定去除效果,原因可能是沉砂

池的水力停留时间(HRT)较短(约4 min),且多为湍流,微塑料不易沉降去除^[41].同时,曝气沉砂池中附着在泥沙上的微塑料可能会因曝气而脱落,反而增加水中微塑料的丰度^[36],对旋流沉砂池而言,流体剪切力会导致微塑料进一步破碎,据 Lv 等^[48]报道,在经旋流沉砂池处理后,污水中 0.25 ~ 0.5 mm的微塑料增加了 27.27%,甚至新增了 < 0.25 mm的微塑料增加了 27.27%,甚至新增了 < 0.25 mm的微塑料,而初沉池的 HRT 较长(0.5 ~ 2 h),水流速度缓慢,粒径相对较大的微塑料易在初沉池中沉降^[10,12,17,41].因此,初沉池对微塑料的去除率高于格栅和沉砂池.同时,初沉池和格栅、沉砂池的任意组合均可提升微塑料的去除效果,特别是格栅、沉砂池和初沉池的工艺组合对微塑料的去除效果最好,与 Liu 等^[17]的研究结果类似.

二级处理中,主要通过微塑料和微生物之间的 相互作用(如生物膜)和二沉池的沉降作用来去除 污水中的微塑料. MBR 工艺在二级处理中表现出了 优异的微塑料去除性能,Lares 等[23]研究发现,MBR 工艺出水中的微塑料丰度(0.4 n·L⁻¹)低于 CAS 工 艺的最终出水(1.0 n·L⁻¹),去除率为99.40%. MBR 的滤膜孔径通常为 0.1 μm, 可高效去除小粒 径微塑料^[36],Li 等^[49]还发现大部分微塑料被截留 于 MBR 系统的生物膜载体侧,表明生物膜的吸附效 应可能是微塑料去除的原因之一,但 MBR 工艺同时 也面临着维护成本高和膜污染易产生等问题. CAS 和 OD 工艺对微塑料的平均去除率分别为 62.52% 和 61.77%,其 HRT 通常约为 25 h 和 > 16 h [50],有 利于在生化池中形成大量的活性污泥,微生物分泌 的胞外聚合物(EPS)会吸附微塑料形成大颗粒絮 体,增加絮体的相对密度和沉降速度,易于微塑料在 二沉池中沉降去除[36,51];另一方面,微塑料可作为 微生物生长的载体,其表面形成生物膜,增强对微塑 料的吸附而沉降至污泥中[35,36,51]. A2O 工艺虽然与 CAS 和 OD 工艺的去除机制类似,但其 HRT(约7~ 14 h)较短^[17,36],不利于微塑料表面生物膜的形成, 不能有效地去除微塑料,与 Liu 等[17]研究的结果一 致. 此外, A²O 工艺的污泥回流量较大, 部分沉降至 污泥的微塑料会回流至生化池中[48],从而导致去除 率不高,如贾其隆等[52]研究结果显示,作为二级处 理的 A2O 工艺对微塑料的去除率仅为 12.92%. AO 工艺与 A2O 工艺相近, HRT 较短, 污泥回流量较大, Jiang 等[38] 研究表明, AO 生化池仅可去除约 16.90%的微塑料. 相对地, SBR 工艺无污泥回流系 统,且 HRT 长于 A2O 工艺,因此对微塑料的去除率 相对较好. 较长的 HRT 和污泥龄(SRT)能够确保微 塑料与污水、污泥之间的充分接触,从而促进微塑

料在生化池的污泥絮体和 EPS 中聚集,并最终在污泥中沉降[32,53]. Yuan 等[32]分析得出, CAST 工艺对微塑料的去除率优于 A^2O 工艺的原因与 HRT 和 SRT 有关, CAST 反应池的设计 HRT 和 SRT 分别为 20 h 和 16 d, 而 A^2O 的分别为 12 h 和 14 d.

三级处理中,微塑料的去除效果因工艺而异,尤 其是过滤技术,取决于过滤介质的孔径和堵塞频率. 孔径较小的过滤技术可去除的微塑料粒径范围更 广,特别是膜过滤技术,如拥有纳米级孔径的超滤 (ultrafiltration, UF) 和反渗透(reverse osmosis, RO) 可去除粒径更小的微塑料. Pramanik 等[54]研究结果 显示,超滤对粒径为 1.88 µm 的微塑料去除率高达 96%. Ziajahromi 等^[55]研究表明,超滤和反渗透组合 可完全去除 > 0.19 mm的微塑料.类似地,孔径为10 μm 的盘式过滤(disc filter, DF) 对微塑料去除率可 达 79. 40% [13], 但在 Talvitie 等 [56] 的研究中, DF 的 去除率仅为 40.0%, 而使用孔径为 20 μm 的 DF, 去 除率却高达98.50%.可能是因为过滤孔径越小,微 塑料堵塞滤膜的速率越快,从而导致反冲洗次数增 多,造成去除率降低.相比膜过滤技术,砂滤工艺在 污水处理中更为常用,在 Hidayaturrahman 等[13]的 研究中,砂石直径约为 0.80~1.20 mm 的快速砂滤 (rapid sand filter, RSF) 去除了 73.80% 的微塑料. Magni 等[25]报道称, RSF 可去除 50% 以上二级处理 出水的微塑料. 而在 Talvitie 等[56]的研究中,由粒径 分别为3~5 mm 与 0.1~0.5 mm 的砾石和石英组 成的 RSF 可高效去除微塑料,去除率高达 97%,但 也会将微塑料破碎成更小的颗粒[57]. 其次,微塑料 与滤料或滤膜之间存在架桥效应、静电吸附和自由 扩散等作用,可提升去除率,Enfrin等[58]研究表明, 由于微塑料和滤膜表面之间存在静电作用,微塑料 可被吸附至滤膜表面上. Xu 等[51] 认为 RSF 滤料可 通过与微塑料表面羟基的亲水作用吸附微塑料,也 可能与 EPS 缠绕在一起形成聚集体,被滤料截留. 但随着时间的推移,滤膜孔隙易被微塑料堵塞而形 成滤饼层,进而降低过滤性能.作为三级处理的另一 类工艺,深度氧化对微塑料的去除效果较差,与Liu 等[17]的研究结果一致,原因是深度氧化过程可改变 微塑料的物理和化学性质[17,36], Kelkar 等[59]研究表 明氯化消毒可能会破坏高密度聚乙烯(HDPE)微塑 料原有的化学键,并形成 C-C-C 不对称链、C-C-C 对称链、CH, 扭曲和 CH, 弯曲等新的化学结 构. Lin 等[60]研究发现在紫外线照射下,微塑料的表 面形态(裂纹、褶皱、隆起)、化学特征(化学键断 裂)和疏水性(疏水性降低)可发生显著变化.同样, 臭氧处理也可引起微塑料在物理和化学性质上的变

化^[61]. 因此,深度氧化过程可能会使部分微塑料进一步破碎开裂成次级微塑料,导致微塑料丰度增加和去除率降低. 但 Hidayaturrahman 等^[13]报道,通过臭氧处理去除微塑料的效率(89.90%)高于盘式过滤(79.40%)和快速砂滤(73.80%). 因此,深度氧化对微塑料的去除效果尚未形成共识,其去除机制需要进一步深入研究.

3.3 微塑料形状、粒径和聚合物类型对去除效率 的影响

不同形状微塑料的物理化学特性各异,是影响 其去除效果的重要因子. 纤维多由 PET 和 PES 制 成[42,62],密度相对较大,且其表面易形成生物膜或 因其较为柔软容易变形,在一级处理和二级处理过 程中更容易与絮体聚合并通过沉降去除[10,36,41,63]. Cai 等[64]研究发现,由于纤维表面相对光滑,横截面 尺寸较小,容易纵向通过三级处理中过滤介质,粒径 较小的纤维(<0.2 mm)甚至可穿透孔径为 1×10^{-7} mm 的反渗透处理系统,使得在三级处理出水中纤 维的占比高于二级处理出水. 陈瑀等[27] 研究结果显 示,经过三级处理后,纤维占比由二级处理出水的 69%提高到了100%.碎片在二级处理中的去除效 率(81.30%)高于一级处理和三级处理[图4(a)], 与 Liu 等[17] 研究的结果一致,可能是因其层状结构 的特点而易被活性污泥吸附沉降去除[17];其次,碎 片通常具有棱角、扭曲、分叉、弯曲和粗糙的表面。 使其易于被微生物定殖以增加相对密度,从而利于 其从污水中去除[34]. Bilgin 等[41]推测,相比纤维、 球状和薄膜,碎片的沉降速度更快.球状微塑料主要 去除机制包括:一方面由于其自身粒径小、比表面 积大的特点,更有利于絮体的吸附和生物膜的形成 而沉降至污泥^[40,65],如 Liu 等^[40]在污泥中检测到球 状微塑料的比例(17.10%)比污水中的高(3.70%~ 5.80%),Yuan 等[32]的结果也显示,相对于曝气沉 砂池,生化池中污泥的球状微塑料占比更高,表明生 化池中活性污泥大量吸附球状微塑料而实现更好去 除:另一方面,球状微塑料大多由 PE 和 PP 制 成[53,66],密度较低而易浮于表水,可在一级处理中 随浮渣被去除. 薄膜和泡沫状的微塑料密度相对较 低,且表面积较大,易漂浮于水面而随浮渣被去 除[37]. 在 Blair 等[67]的研究中,薄膜大部分在一级 处理过程中被去除. 同时, Ali 等[36]认为粒径与污水 悬浮固体(<20 μm)相似的薄膜和纤维易被 EPS 吸 附而产生沉降. 但本文研究结果显示,三级处理对于 薄膜和球状的去除效果更好,原因是过滤技术可有 效截留一级处理和二级处理中未能去除的小粒径球 状和薄膜.

粒径亦为影响微塑料去除的重要因素,污水处 理厂总体对 0.5~1 mm 和 1~5 mm 的微塑料去除 率均比 0.1~0.5 mm 和 < 0.1 mm 的高. 较大粒径 即 0.5~5 mm 可在一级处理中得到较好地去除,原 因是其易被格栅截留或在沉砂池和初沉池通过重力 沉降被去除[36,40],如 Dris 等[68]研究结果显示,在经 过一级处理后 1~5 mm 的占比从 45% 下降到了 7%. 在 Liu 等[40]的研究中,污水处理厂进水中的微 塑料平均粒径为 0.52 mm, 在经一级处理后平均粒 径减少至 0.24 mm. 而 < 0.5 mm 的微塑料更容易在 二级处理中被生物絮凝体吸附或生物膜覆盖,从而 沉降至污泥中^[35], Magni 等^[25]研究结果显示, 0.1~ 0.5 mm 的微塑料颗粒在回流污泥中占比最高 (54%). < 0.5 mm 的微塑料也可被三级处理中孔 径较小的过滤技术截留. Mintenig 等[69] 研究发现过 滤技术对 < 0.5 mm 的微塑料去除率高达 93%,在 Cai 等[64]的研究中,经过 RO 处理后,0.05~0.1 mm 的微塑料丰度由 2.40 n·L-1降至 0.06 n·L-1.但如 前所述,微塑料在污水处理厂的去除过程中,受机械 力、流体剪切力和氧化等作用影响,微塑料易被破 碎成更小粒径的微塑料[51],从而造成小粒径微塑料 丰度的增加,去除率降低. 因此,可以认为污水处理 厂总体对 0.5~5 mm 的微塑料去除效果比 < 0.5 mm 的好,与 Liu 等[17]的研究结果一致.

聚合物类型对去除率的影响主要是密度差异(见表 2),据 Long 等^[34]报道,微塑料去除率随聚合物密度的升高而增加;高密度聚合物可通过重力沉降在沉砂池和初沉池中被去除,如 PET 和 PES 因其高密度的性质而具有较高的去除率,最高达97.70%和 97.79%^[42];低密度聚合物(如 PE 和 PP)自身易漂浮于表水^[70]或者与中等密度(如 PA 和 PS)被气浮工艺产生的气泡带到表水^[65],随浮渣被去除.因此,若沉砂池、初沉池和气浮工艺相结合,可以实现微塑料高效去除.此外,具有特定表面电荷的聚合物易被污水处理厂去除,如带正电荷的PE 和 PS 对带负电荷的活性污泥具有高亲和力^[71].

4 展望

(1)作为一种新污染物,水体中微塑料的样品采集、预处理和表征分析等方法仍缺乏统一标准,对于形状、粒径的分类和描述也未达成共识,导致不同研究结果存在差异,进而影响 Meta 分析结果及结论,同时还阻碍了基于赋存特征的微塑料污染风险评价等研究的开展和微塑料污染管控标准的制定.

(2)今后可重点针对氧化沟、生物膜和活性污

泥等工艺开展研究,研发高效去除微塑料的生活污水处理工艺组合及运行参数.

- (3)污水处理厂三级处理中的紫外和臭氧消毒,可能导致污水中微塑料的氧化和破碎,产生粒径更小的微、纳塑料,随出水排放而进入环境,增加污染风险.因此,需针对污水深度处理工艺中微、纳塑料的产生机制及影响因素开展研究,明晰其形成的关键过程和因子,为控制出水中微、纳塑料提供科学依据.
- (4)微塑料形状、粒径和聚合物类型等因素对污水处理工艺性能和微塑料去除率的影响研究还很少,具体的影响机制仍不明晰,有待深入研究.

5 结论

- (1)污水处理厂进水和出水的微塑料丰度分别在 $1.56 \times 10^{-2} \sim 3.14 \times 10^{4} \text{ n·L}^{-1}$ 和 $1.70 \times 10^{-3} \sim 3.09 \times 10^{2} \text{ n·L}^{-1}$ 之间,进水中微塑料形状以纤维和碎片为主,颜色主要以透明为主,粒径以 < 0.5 mm为主,PE 和 PP 是主要的检出聚合物类型. 最终排泥的微塑料丰度在 $1.8 \times 10^{-1} \sim 9.38 \times 10^{3} \text{ n·g}^{-1}$ 之间,纤维的丰度均值最大, < 0.5 mm为主要赋存粒径,PET 在污泥中检出的丰度均值最高,颜色以透明和黑色为主.
- (2)以 OD、CAS 和 biofilm 为核心工艺的污水处理厂对微塑料的去除效果较好,总去除率均超过90%,其次为 SBR 和 A²O 工艺,最差为 AO 工艺(71.31%).一级处理(62.87%)对微塑料的去除效果优于二级处理(55.78%)和三级处理(58.45%),"格栅+沉砂池+初沉池"是一级处理中去除率最高的工艺组合(78.47%),而沉砂池的去除效果最差(13.88%),MBR 工艺为二级处理最佳工艺单元(98.74%),过滤技术在三级处理中去除率最高(72.71%).
- (3)污水处理厂可有效去除薄膜、泡沫和碎片, 粒径>0.5 mm 的微塑料,对 PE、PET 和 PP 的去除 率在 80%以上.纤维易被一级处理和二级处理去 除,而碎片在二级处理中去除率较高,三级处理则是 对薄膜和球状的去除效果较好.一级处理易去除 >0.5 mm的微塑料,而三级处理对 < 0.5 mm 的微 塑料去除效果最佳.

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HUANJING KEXUE

Environmental Science (monthly)

Vol. 44 No. 6 Jun. 15, 2023

CONTENTS

Research Status and Trend Analysis of Environmental and Health Risk and Control of Persistent, Mobile, and Toxic Chemicals	
	·· ZHANG Shao-xuan, CHEN An-na, CHEN Cheng-kang, et al. (301
Assessment of the Multidimensional Performances of Food Waste Utilization Technologies in China	_
Spatial Network of Urban Heat Environment in Beijing-Tianjin-Hebei Urban Agglomeration Based on MSPA and Circuit Theory	
Relationship Between Urban Spatial Pattern and Thermal Environment Response in Summer: A Case Study of Hefei City	
Assessment of Emission Reduction Effect of Major Air Pollution Control Measures on PM _{2,5} Concentrations During 13th Five-Y	ear Period in Tianjin
Do col II de Col de la la la marca de la colonidada de la	XIAO Zhi-mei, XU Hong, CAI Zi-ying, et al. (305
Effect of Clean Heating on Carbonaceous Aerosols in PM _{2,5} During the Heating Period in Baoding	VIE F. " ZHENG V. DOLLT
Transport Influence and Potential Sources of PM ₂₅ Pollution for Nanjing Impact of Atmospheric Circulation Patterns on Ozone Changes in the Pearl River Delta from 2015 to 2020	WANG V. LILLD. VIN F. (200
Impact of Atmospheric Circulation Patterns on Ozone Changes in the Pearl River Delta from 2015 to 2020 Effects of Tropical Cyclones on Ozone Pollution in Hainan Island	
Analysis of Causes and Sources of Summer Ozone Pollution in Rizhao Based on CMAQ and HYSPLIT Models	
Health Benefit Evaluation for PM _{2.5} as Well as O _{3.8h} Pollution Control in Chengdu, China from 2016 to 2020	ZHANG Ying, HAN Qi-qi, WEI Xiao-yu, et al. (310
Emission Inventory of Airborne Pollutants from Biomass Combustion in Guizhou Province Main Chemical Components in Atmospheric Precipitation and Their Sources in Xi'an	
Main Chemical Components in Atmospheric Precipitation and Their Sources in Ai an Distribution, Respiratory Exposure, and Traceability of Atmospheric Microplastics in Yichang City	
Hydrochemical Evolution in the Yarlung Zangbo River Basin	
Temporal and Spatial Distribution Characteristics and Source Analysis of Nitrate in Surface Water of Wuding River Basin	
Seasonal Variation Characteristics and Pollution Assessment of Heavy Metals in Water and Sediment of Taipu River	
Pollution Characteristics and Risk Assessment of Antibiotics in Beiyun River Basin in Beijing	
Tracking Riverine Nitrate Sources and Transformations in the Yiluo River Basin by Nitrogen and Oxygen Isotopes	
Distribution Characteristics and Risk Assessment of PPCPs in Surface Water and Sediments of Lakes in the Lower Reaches of the	he Huaihe Kiver
	WU Yu-sheng, HUANG Tian-yin, ZHANG Jia-gen, et al. (32)
Characteristics and Driving Mechanisms of Shallow Groundwater Chemistry in Xining City	
Groundwater Pollution Risk Assessment in Plain Area of the Yarkant River Basin	
Composition Structure and Influence Factors of Bacterial Communities in the Miyun Reservoir	
Photo-Degradation Mechanism and Pathway for Tetracycline in Simulated Seawater Under Irradiation of Visible Light	
Adsorption Characteristics and Mechanism of Ammonia Nitrogen in Water by Nano Zero-valent Iron-modified Biochar	
Removal Performance and Mechanism of Potassium Permanganate Modified Coconut Shell Biochar for Cd(II) and Ni(II) in A	Aquatic Environment
Phosphorus Adsorption in Water and Immobilization in Sediments by Lanthanum-modified Water Treatment Sludge Hydrochar	
Factors Affecting Nitrate Concentrations and Nitrogen and Oxygen Isotope Values of Effluents from Waste Water Treatment Plan	
Effects of Wastewater Treatment Processes on the Removal Efficiency of Microplastics Based on Meta-analysis	
Assessment of Critical Loads of Nitrogen Deposition in Natural Ecosystems of China	
Impacts of Climate Change and Human Activities on NDVI Change in Eastern Coastal Areas of China	
Ecosystem Carbon Storage in Hangzhou Bay Area Based on InVEST and PLUS Models	
Soil Stoichiometry Characterization in the Oasis-desert Transition Zone of Linze, Zhangye	
Vertical Differences in Grassland Bacterial Community Structure During Non- Growing Season in Eastern Ulansuhai Basin	
Distribution Pattern of Bacterial Community in Soil Profile of Larix principis-rupprechtii Forest in Luya Mountain	
Effects of Vegetation Types on Carbon Cycle Functional Genes in Reclaimed Soil from Open Pit Mines in the Loess Plateau	
Effects of Biochar Application on Soil Bacterial Community Diversity and Winter Wheat Growth in Wheat Fields	
Effects of Different Planting Years of Dendrocalamus brandisii on Soil Fungal Community	
Effects of Biochar Amendment on N ₂ O Emission and Its Functional Genes in Pepper Growing Soil in Tropical Areas	
Effects of Mulching and Application of Organic and Chemical Fertilizer on Greenhouse Gas Emission and Water and Nitrogen U	Jse in Summer Maize Farmland
Effects of Different Types of Plastic Film Mulching on Soil Quality, Root Growth, and Yield	
Pollution Assessment and Source Analysis of Heavy Metals in Atmospheric Deposition in a Lead-zinc Smelting City Based on Pl	
Characterization and Health Risk of Heavy Metals in $PM_{2.5}$ from Road Fugitive Dust in Five Cities of Yunnan Province \cdots	
Pollution Characteristics and Risk Assessment of Heavy Metals in Surface Dusts and Surrounding Green Land Soils from Yellow	
	River Custom Tourist Line in Lanzhou
Source Apportionment and Pollution Assessment of Soil Heavy Metal Pollution Using PMF and RF Model: A Case Study of a T-	William River Custom Tourist Line in Lanzhou
Typotholimon and Foreign to continue of co	LI Jun, LI Kai-ming, WANG Xiao-huai, et al. (347) Sypical Industrial Park in Northwest China
	ypical Industrial Park in Northwest China
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and G	
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and	ypical Industrial Park in Northwest China
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and	ypical Industrial Park in Northwest China
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Source Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics Characteristics and Risk Evaluation of Heavy Metal Contamination in Paddy Soils in the Three Gorges Reservoir Area	UJ Jun, IJ Kai-ming, WANG Xiao-huai, et al. (34) ypical Industrial Park in Northwest China
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Source Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics Characteristics and Risk Evaluation of Heavy Metal Contamination in Paddy Soils in the Three Gorges Reservoir Area	UJ Jun, IJ Kai-ming, WANG Xiao-huai, et al. (34) ypical Industrial Park in Northwest China
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Source Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics	
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Source Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics	
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and of Source Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics	UI Jun, II Kai-ming, WANG Xiao-huai, et al. (347) Sypical Industrial Park in Northwest China GAO Yue, LÜ Tong, ZHANG Yun-kai, et al. (348) Geostatistical Method ZHANG Chuan-hua, WANG Zhong-shu, LIU Li, et al. (350) WANG Mei-hua (350) LIU Ya-jun, LI Cai-xia, MEI Nan, et al. (352) LIU Hai, WEI Wei, SONG Yang, et al. (352) AN Yong-long, YIN Xiu-lan, LI Wen-juan, et al. (352) TANG Jin-lai, ZHAO Kuan, HU Rui-xin, et al. (356)
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Cource Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics Characteristics and Risk Evaluation of Heavy Metal Contamination in Paddy Soils in the Three Gorges Reservoir Area Health Risk Assessment and Environmental Benchmark of Heavy Metals in Cultivated Land in Wanjiang Economic Zone Evaluation and Source Analysis of Soil Heavy Metal Pollution in a Planting Area in Wanquan District, Zhangjiakou City Heavy Metal Concentration, Source, and Pollution Assessment in Topsoil of Chuzhou City	UI Jun, II Kai-ming, WANG Xiao-huai, et al. (347) Sypical Industrial Park in Northwest China GAO Yue, LÜ Tong, ZHANG Yun-kai, et al. (348) Geostatistical Method ZHANG Chuan-hua, WANG Zhong-shu, LIU Li, et al. (350) WANG Mei-hua (350) LIU Ya-jun, LI Cai-xia, MEI Nan, et al. (352) LIU Hai, WEI Wei, SONG Yang, et al. (352) AN Yong-long, YIN Xiu-lan, LI Wen-juan, et al. (352) TANG Jin-lai, ZHAO Kuan, HU Rui-xin, et al. (356)
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Geostatistics of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics Characteristics and Risk Evaluation of Heavy Metal Contamination in Paddy Soils in the Three Gorges Reservoir Area Health Risk Assessment and Environmental Benchmark of Heavy Metals in Cultivated Land in Wanjiang Economic Zone Evaluation and Source Analysis of Soil Heavy Metal Pollution in a Planting Area in Wanquan District, Zhangjiakou City Heavy Metal Concentration, Source, and Pollution Assessment in Topsoil of Chuzhou City Analysis on the Distribution Characteristics and Influence Mechanism of Migration and Transformation of Heavy Metals in Minin Ecological Risk Assessment and Source Apportionment of Heavy Metals in Mineral Resource Base Based on Soil Parent Materia	
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and Geostatistics of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics Characteristics and Risk Evaluation of Heavy Metal Contamination in Paddy Soils in the Three Gorges Reservoir Area Health Risk Assessment and Environmental Benchmark of Heavy Metals in Cultivated Land in Wanjiang Economic Zone Evaluation and Source Analysis of Soil Heavy Metal Pollution in a Planting Area in Wanquan District, Zhangjiakou City Heavy Metal Concentration, Source, and Pollution Assessment in Topsoil of Chuzhou City Analysis on the Distribution Characteristics and Influence Mechanism of Migration and Transformation of Heavy Metals in Minin Ecological Risk Assessment and Source Apportionment of Heavy Metals in Mineral Resource Base Based on Soil Parent Materia	
Source Analysis of Soil Heavy Metals in Agricultural Land Around the Mining Area Based on APCS-MLR Receptor Model and of Source Analysis of Heavy Metals in Typical Farmland Soils Based on PCA-APCS-MLR and Geostatistics	