

目 次

中国城市碳排放强度的时空演变、动态跃迁及收敛趋势	杨清可, 王磊, 朱高立, 李颖, 范业婷, 王雅竹 (1869)
交通运输业碳排放效率时空演变及趋势预测	郑琰, 蒋雪梅, 肖玉杰 (1879)
基于扩展 STIRPAT 模型 LMDI 分解的碳排放脱钩因素	张江艳 (1888)
基于 LEAP 模型的工业园区碳达峰路径: 以南京某国家级开发区为例	李慧鹏, 李荔, 殷茵, 何文大, 宿杰, 赵秋月 (1898)
高校碳排放核算与分析: 以北京 A 高校为例	曹睿, 封莉, 张立秋 (1907)
北京市制造业减排降碳协同效应分析和驱动因素	俞珊, 韩玉花, 牟洁, 张双, 张增杰 (1917)
碳排放权交易价格与全要素生产率: 来自中国的证据	吴雪萍 (1926)
中国城市 PM _{2.5} 和 PM ₁₀ 时空分布特征和影响因素分析	李江苏, 段良苏, 张天娇 (1938)
2017~2021 年苏皖鲁豫交界区域 PM _{2.5} 和 O ₃ 时空变化特征及影响因素	陈伟, 徐学哲, 刘文清 (1950)
疫情管控期西安 PM _{2.5} 和 O ₃ 污染特征及成因分析	原晓红, 张强, 李琦, 谢文豪, 刘跃廷, 樊亭亭, 姜旭朋 (1963)
苏南五市秋冬季 PM _{2.5} 化学组成特征和空间差异	冯蔚, 丁峰, 尚玥, 谢鸣捷 (1975)
湖北咸宁细颗粒物 PM _{2.5} 来源	罗怡, 朱宽广, 陈璞琰, 田军, 谢旻, 战杨志豪, 赵润琪 (1983)
邢台氨减排对京津冀 PM _{2.5} 改善的溢出效益	边泽君, 闻超玉, 郎建奎, 范晓茜, 夏祥晨, 周颖 (1994)
聊城市冬季 PM _{2.5} 载带金属元素污染特征、风险评价及来源分析	张敬巧, 朱瑶, 曹婷, 燕丽, 王淑兰, 刘铮 (2003)
高原城市拉萨典型 VOCs 排放源成分谱特征	郭淑政, 叶春翔, 林伟立, 陈熠, 曾立民, 尹晴晴, 刘雪莉 (2011)
北京市工业园区 VOCs 污染特征及健康风险评估案例: 高新技术产业的环境影响	王洁, 姚震, 王敏燕, 陈速敏, 龙腾, 王海林, 李红, 郭秀锐, 郝江虹, 聂磊 (2019)
高温极端天气影响下的成都平原一次典型臭氧污染过程分析	雷丽娟, 张懿, 罗伊娜, 张潇, 冯森 (2028)
水环境中抗病毒药物的存在、行为与风险	葛林科, 李璇艳, 曹胜凯, 郑金帅, 张蓬, 朱超, 马宏瑞 (2039)
黄河小浪底水库地表水中重金属的时空变化与概率健康风险	王亮, 邓雪娇, 王潇磊, 李明, 刘奕尧, 姜亚敏, 涂响, 张坤峰 (2054)
藏东多曲河流域锑富集水化学特征及控制因素	李敬杰, 连晟, 王明国, 张智印, 张涛 (2067)
西北内陆区降水稳定同位素时空分布特征及其水汽来源	张炎炎, 辛存林, 郭小燕, 张博, 陈宁, 史延飞 (2080)
基于水化学和氢氧同位素的泰安城区岩溶地下水补给来源及演化过程	孟令华 (2096)
丹江流域山区地表水-地下水水化学特征及其影响因素	张子燕, 伏永朋, 王宁涛, 谭建民, 刘亚磊 (2107)
金塔盆地鸳鸯池灌区地下水水化学特征及控制因素	王晓燕, 韩双宝, 张梦南, 尹德超, 吴玺, 安永会 (2118)
德阳市平原区浅层地下水水化学特征与健康风险评价	刘楠, 陈盟, 高东东, 吴勇, 王橹橹 (2129)
典型城市河网沉积物微塑料时空分布特征	许万璐, 范一凡, 钱新 (2142)
抚仙湖流域尺度氮排放清单构建及关键源解析	王延杰, 梁启斌, 王艳霞, 侯磊, 陈奇伯, 王伟, 李晓琳, 高俊淑 (2150)
过氧化钙/海泡石海藻酸钠缓释凝胶复合材料的制备及其对内源磷的控制性能	曲思彤, 单苏洁, 王崇铭, 吴玲予, 李大鹏, 黄勇 (2160)
矿物超细颗粒的形成机制、结构特征及其环境行为和效应	刘振海, 张展华, 袁语欣, 朱盼盼, 陈威, 张勇 (2171)
生物炭固定化菌复合材料在环境修复中的应用研究进展	孙淑玉, 黄梦鑫, 孔强, 张焕新, 刘继伟 (2185)
微塑料对沸石吸附水体氨氮的影响及其机制	练建军, 谢诗婷, 吴培, 孟冠华, 陈波 (2195)
紫外老化作用对纳米生物炭吸附环丙沙星的影响机制	马锋锋, 薛之一, 赵保卫 (2203)
土地利用影响下永定河流域浮游植物群落与环境因子响应	郭善嵩, 胡恩, 丁一桐, 张嘉渭, 孙长顺, 卢悦, 潘保柱 (2211)
鄱阳湖湿地细菌群落多样性和可培养细菌功能基因丰度	喻江, 王淳, 龙永, 刘贵花, 李春杰, 范国权, 于镇华 (2223)
典农河沉积物细菌群落结构特征及其与重金属的关系	刘双羽, 蒙俊杰, 邱小琮, 周瑞娟, 李霖 (2233)
污水处理厂尾水排放对受纳河流细菌和真菌微生物群落的影响	郭有顺, 余仲, 郝文彬, 孟凡刚 (2246)
宏基因组学分析深度处理阶段污水中细菌的赋存特征及其功能	胡健双, 王燕, 周政, 汪雅琴, 王秉政, 李激 (2259)
大别山区生境质量时空特征及自然-人为因素驱动机制	郑亚平, 张俊华, 田惠文, 朱航成, 刘舒, 丁亚鹏 (2268)
基于连续小波变换、SHAP 和 XGBoost 的土壤有机质含量高光谱反演	叶森, 朱琳, 刘旭东, 黄勇, 陈蓓蓓, 李欢 (2280)
秸秆还田、覆膜和施氮对旱地麦田土壤质量的影响	叶子壮, 王松燕, 陆潇, 史多鹏, 吕慎强, 李嘉, 杨泽宇, 王林科 (2292)
长期施用有机肥对土壤微塑料赋存及迁移特征的影响	王长远, 马啸驰, 郭德杰, 刘新红, 马艳, 罗佳 (2304)
土壤盐分变化对 N ₂ O 排放影响: 基于 Meta 分析	黄艺华, 余冬立, 史祺琦, 胡磊, 潘永春 (2313)
基于土地利用/覆被动态变化的粤港澳大湾区碳储量评价与预测	郑慧玲, 郑辉峰 (2321)
基于 InVEST 模型的伏牛山地区生态系统碳储量时空变化模拟	张哲, 时振钦, 朱文博, 孙梓欣, 赵体侠, 邓文萍, 刘志强 (2332)
防护林建设过程中土壤微生物养分限制与有机碳组分之间的关系	徐凤璟, 黄懿梅, 黄倩, 申继凯 (2342)
长期秸秆还田褐土有机碳矿化特征及其驱动力	赵宇航, 殷浩凯, 胡雪纯, 解文艳, 刘志平, 周怀平, 杨振兴 (2353)
冬绿肥覆盖对土壤团聚体及有机碳和 AMF 多样性的影响	鲁泽让, 陈佳钰, 李智贤, 李永梅, 罗志章, 杨锐, 田明洋, 赵吉霞, 范茂攀 (2363)
辽河流域氮素时空分布及其对土地利用和降雨的响应	周波, 李晓光, 童思陈, 吕旭波, 郭朝臣, 雷坤 (2373)
高强度农业种植区不同景观池塘氧化亚氮排放特征	张欣悦, 肖启涛, 谢晖, 刘臻婧, 邱银国, 罗菊花, 徐向华, 段洪涛 (2385)
生物炭与不同类型氮肥配施对菜地土壤反硝化细菌群落的影响	柳晓婉, 刘杏认, 高尚洁, 李贵春 (2394)
太岳山不同林龄人工油松林土壤微生物特征	马义淑, 曹亚鑫, 牛敏, 张明昱, 程曼, 文永莉 (2406)
基于稀疏样点的土壤重金属含量模拟方法	张佳琦, 潘瑜春, 高世臣, 赵亚楠, 景胜强, 周艳兵, 邹允兵 (2417)
基于特定源-风险评估模型的兰州黄河风情线绿地土壤重金属污染优先控制源分析	李军, 李旭, 李开明, 焦亮, 台喜生, 臧飞, 陈伟, 脱新颖 (2428)
西南不同类型紫色土 pH 变化、重金属累积与潜在生态风险评估	张海琳, 张雨, 王顶, 谢军, 张跃强, 张宇亭, 王洁, 石孝均 (2440)
重庆某铁矿区周边耕地土壤重金属污染评价及来源解析	廖泽源, 李杰芹, 沈智杰, 李彩霞, 罗程钟, 梅楠, 张成, 王定勇 (2450)
金属矿区周边农田土壤与农作物重金属健康风险评估	魏洪斌, 罗明, 向奎, 查理思 (2461)
赤泥基纳米零价铁对多金属污染土壤修复效果	刘龙宇, 杨世利, 赵黄诗雨, 常凯威, 余江 (2473)
施锌对碱性土壤-小麦幼苗体系累积镉的影响	张瑶, 王天齐, 牛硕, 杨阳, 陈卫平 (2479)

长期施用有机肥对土壤微塑料赋存及迁移特征的影响

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摘要: 为探明设施土壤中微塑料的赋存特征与施肥模式、种植年限及土壤深度之间的响应关系, 通过现场采样、显微镜观察、傅里叶变换红外光谱(FTIR)和扫描电子显微镜(SEM)等方法, 对长期施用有机肥土壤中微塑料丰度、形状、粒径、颜色、种类和表面形貌特征及有机肥中微塑料的丰度、粒径和种类进行分析. 结果表明, 设施土壤中不同种植年限下(2016~2021年)表层土壤中微塑料的平均丰度随着种植年限的升高而升高, 化肥(CF)、有机肥(OF)和生物炭+有机肥(BOF)处理中微塑料丰度不同, 其中每年OF处理微塑料的丰度最高, 2021年长期施用OF的表层土壤微塑料丰度最高, 为 $543.33 \text{ n} \cdot \text{kg}^{-1}$. 3种施肥处理土壤中微塑料平均年增长速率分别为11.16%、12.61%和9.17%. 微塑料丰度在不同深度土层中表现为随土层深度的增加而显著降低, 且BOF处理中微塑料向深层土壤迁移效率最高. 微塑料形状主要为纤维状、碎片状、颗粒状和薄膜状, 在0~40 cm深度土壤中, 纤维状微塑料在3种施肥类型土壤中均占主导地位. 在40~60 cm土壤中CF处理无纤维状和薄膜状微塑料, 颗粒状微塑料占比最高(57.14%); OF处理无薄膜状微塑料, 颗粒状微塑料和纤维状微塑料占比最高(36.00%); BOF处理中占比最高的为颗粒状微塑料(41.18%), 占比最少的为薄膜状微塑料(5.88%). 3种施肥处理土壤中小尺寸(<0.5 mm)微塑料占比最高, 在CF、OF和BOF这3种处理土壤中PE为主要微塑料种类, 且在微塑料表面均呈现多孔、撕裂、裂痕和磨损等特点. 有机肥中微塑料平均丰度高达 $577.78 \text{ n} \cdot \text{kg}^{-1}$. 长期施用有机肥的设施土壤中微塑料丰度与有机肥密切相关, 且微塑料迁移也与施用肥料种类密切相关. 研究对于明晰不同长期施肥模式下设施土壤中微塑料的分布特征和迁移具有重要意义.

关键词: 微塑料; 有机肥; 土壤深度; 设施农田; 迁移

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Effects of Long-term Application of Organic Fertilizer on the Occurrence and Migration Characteristics of Soil Microplastics

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Abstract: To explore the response relationship between the occurrence characteristics of microplastics in the facility soil (fertilization mode, planting years, and soil depth), the abundance, shape, particle size, color, type, and surface morphology characteristics of microplastics in the soil with long-term application of organic fertilizer, as well as the abundance, particle size, and type of microplastics in organic fertilizer, were analyzed through on-site sampling, microscopic observation, Fourier transform infrared spectroscopy (FTIR), and scanning electron microscopy (SEM). The results showed that the average abundance of microplastics in the surface soil under different planting years (2016–2021) increased with the increase in planting years, and the abundance of microplastics in the chemical fertilizer (CF), organic fertilizer (OF), and biochar organic fertilizer (BOF) treatments differed, among which the abundance of microplastics in the OF treatment was the highest per year, and the highest abundance of microplastics in the topsoil with long-term application of OF in 2021 was $543.33 \text{ n} \cdot \text{kg}^{-1}$. The average annual growth rates of microplastics in the soil treated by the three fertilization treatments were 11.16%, 12.61%, and 9.17%, respectively. The abundance of microplastics decreased significantly with the increase in soil depth in different soil layers, and the migration efficiency of microplastics to deep soils was the highest in the BOF treatment. The shapes of the microplastics were mainly fibrous, fragmentary, particle, and film, and in the soil at a depth of 0–40 cm, fibrous microplastics were dominant in the soil of all three fertilization types. In 40–60 cm soil, CF-treated fibrous and thin-film microplastics were not detected, and particle microplastics accounted for the highest proportion (57.14%). The OF treatment did not have the presence of film microplastics, and particle microplastics and fibrous microplastics accounted for the highest proportion (36.00%). The highest proportion in the BOF treatment was particle microplastics (41.18%), and the lowest proportion was that of film microplastics (5.88%). The proportion of small-size (<0.5 mm) microplastics in the three fertilization-treated soils was the highest, and PE was the main microplastic species in the three treated soils of CF, OF, and BOF; additionally, the characteristics of porous, torn, cracked, and worn on the surface of microplastics were all presented. The average abundance of microplastics in organic fertilizer was as high as $577.78 \text{ n} \cdot \text{kg}^{-1}$. The abundance of microplastics in the soil of facilities for the long-term application of organic fertilizers was closely related to organic fertilizers, and the migration of microplastics was also closely related to the types of fertilizers applied. It is of great significance to clarify the distribution characteristics and migration of microplastics in facility soils under different long-term fertilization modes.

Key words: microplastics; organic fertilizer; soil depth; facility farmlands; migrate

随着塑料产业的不断发展和塑料产品的大量使用, 环境中使用的塑料产品经过光照、氧化、物理

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磨损和生物降解等作用破碎和风化成粒径小于 5 mm 的颗粒, 即形成了微塑料^[1]. 微塑料具有比表面积大^[2]、体积小^[3]、吸附污染物能力强^[4]、环境危害性大且不易降解^[5]等特点. 这些特点使得微塑料更易在水体、沉积物和土壤等环境介质中赋存, 并通过生物摄食在食物链中迁移, 进而危害人类健康^[6].

目前微塑料环境污染研究主要集中在水体环境中, 受土壤环境的复杂性和微塑料分析方法的局限性的影响, 土壤中微塑料富集、迁移及分布研究相对较少^[7]. 微塑料主要通过农用地膜覆盖^[8]、污水灌溉^[9]、有机肥施用^[10]、污泥的土地利用^[11]及大气沉降^[12]等途径进入农田土壤, 逐渐改变土壤结构和质地, 对土壤理化和生物性状产生影响, 使土壤生态功能被破坏, 并且可能在植物中富集, 对食品安全产生影响^[13]. 微塑料还会通过灌溉、径流、翻耕及生物活动等作用向深层土壤和地下水中迁移^[14]. 有研究表明, 覆膜土壤中微塑料丰度显著高于未覆膜土壤^[15,16], Zhang 等^[17]同样报道我国农用地膜会在土壤中残留, 且会使作物产量显著下降^[18]. 农业灌溉水源以及污泥中同样也检测出大量的微塑料存在^[19,20], 这就导致了农田土壤中微塑料的积累. Yang 等^[21]报道施肥显著增加农田土壤中的微塑料富集量. 但我国肥料施用对农田土壤微塑料影响的研究鲜见报道, 因此探明不同肥料施用的影响对避免土壤微塑料污染具有重要意义.

设施农业因其种植产量大、经济效益高, 在我国乡村振兴建设中有着重要地位^[22]. 但设施农业生产中对地膜的需求量大、化学品投入多且肥料施用量大, 都会导致微塑料富集的程度增加^[23]. 微塑料的富集也会对作物营养元素吸收产生影响, 进而影响植物生长^[24]. 设施栽培过程中塑料制品的大量应用, 以及有机肥的长期施加, 极易在土壤中富集, 增加微塑料环境污染的风险. 由于有机肥在设施生产中占据重要地位, 且有机肥的长期施加对微塑料赋存及迁移影响尚不明确, 因此分析设施土壤中长期施用有机肥对土壤微塑料赋存及迁移特征的影响, 对了解及控制设施农业微塑料污染具有深远意义.

综上, 本文以 3 种长期施肥(化肥, chemical fertilizer, CF; 有机肥, organic fertilizer, OF; 生物炭+有机肥, biochar organic fertilizer, BOF)的设施土壤为研究对象, 探索微塑料在不同长期施肥土壤中的分布及迁移特征, 并探究有机肥中微塑料赋存情况, 以期对微塑料的减排处理和农业生产管理管控提供科学依据, 并为设施栽培土壤防止微塑料污染提供基础数据.

1 材料与方法

1.1 研究区域概况

本试验地点位于江苏省南京市六合区竹镇镇江苏省农业科学院六合动物科学基地辣椒试验大棚. 该地区气候为温带季风气候, 平均年降水量为 914.6 mm, 平均气温为 15.6℃. 试验土壤类型为马肝土(黄棕壤). 试验地块中使用的农业覆盖膜是由 PE 制成的黑色膜. 辣椒在每年的 3 月和 8 月种植, 所有处理均覆盖地膜, 持续覆盖完整的生长期, 收获后进行人工清理, 并在下次种植重新铺设新地膜, 同时在生长季进行田间常规化管理.

本试验时间自 2016 ~ 2021 年, 共设 3 个处理. 处理 1: 化肥处理(CF), 每 hm^2 施用 N、 P_2O_5 和 K_2O 的量分别为 300、200 和 350 kg; 处理 2: 有机肥处理(OF)每 hm^2 施用 15 t 有机肥(猪粪秸秆有机肥)并施用化肥; 处理 3: 生物炭+有机肥处理(BOF), 每 hm^2 施用 9 t 生物炭(水稻秸秆)和 6 t 有机肥并施用化肥. 处理 2 和处理 3 化肥施用量一致, 除肥料施用外, 各处理田间管理一致, 每个处理设 3 个重复小区, 每小区排列随机, 各个小区长和宽分别为 6.3 m 和 3.3 m, 有效面积约为 21 m^2 .

1.2 样品采集与预处理

本研究采集 2016 ~ 2021 年秋季种植并收获作物后的 0 ~ 20 cm 深度土壤样品以及 2016 年、2019 年和 2021 年的 20 ~ 40 cm 和 40 ~ 60 cm 的土壤样品. 采集过程中挑出植物根和砾石. 从每个随机处理小区[长期施用化肥(CF)改良土壤、长期施用有机肥(OF)改良土壤和长期施用生物炭+有机肥(BOF)改良土壤]中分别采集 5 个不同位置的样品并混匀, 送至实验室. 将样品在 45℃ 的烘箱中烘干, 破碎, 过 5 mm 筛, 并用铝箔包好, 储存在干燥处, 用于微塑料的测定. 在整个过程中避免使用塑料制品, 以避免样品污染.

选取 2016 年、2019 年和 2021 年施用的有机肥, 烘干, 过 5 mm 筛, 用铝箔包好, 保存在干燥处, 用于后续微塑料的测定.

1.3 微塑料提取

1.3.1 土壤微塑料的提取

采用密度浮选分离法进行土壤中微塑料的分离^[25]. 首先将 100 g 土壤样品(干重)转移到一个干净的锥形瓶中, 加入 500 mL 饱和氯化钠溶液, 磁力搅拌 30 min, 瓶口用铝箔密封, 以防止样品污染, 并静置过夜, 等待固体和液体分离完全, 确保微塑料完全悬浮在上清液中. 随后, 将上清液转移至另一个干净的锥形瓶中, 并在剩余的沉积物中加入饱和

碘化钠溶液, 进行二次浮选. 重复上述相同的步骤来分离固体和液体. 为避免有机质的干扰, 在提取的上清液中加入 30% 的过氧化氢溶液, 然后用铝箔将瓶口密封, 在 45℃ 的水浴中加热 48 h. 待有机质消解后, 使用 $\Phi 50 \text{ mm} \times 0.45 \mu\text{m}$ 醋酸纤维素滤膜对上清液进行真空抽滤, 将抽滤好的滤膜置于干净的玻璃培养皿中保存, 以备后续对微塑料的观察.

1.3.2 有机肥微塑料的提取

称取 15 g(干重)有机肥置于 100 mL 烧杯中, 称量后加入氯化锌溶液 ($1.7 \sim 1.8 \text{ kg} \cdot \text{L}^{-1}$) 60 mL, 充分搅拌(2 min)并静置过夜(12 h). 静置完成后, 将悬浮液转移到另一个烧杯中并加入 60 mL 30% 的 H_2O_2 以去除有机物, 充分搅拌后静置 24 h, 使过氧化氢与有机物充分反应. 将过氧化氢处理后的溶液进行真空抽滤, 将得到的滤膜浸入乙醇溶液中进行超声处理, 使得滤膜上的物质分散在乙醇溶液中. 将乙醇溶液中的滤膜取出, 并用乙醇多次清洗滤膜. 将乙醇溶液浓缩, 继而将其滴加在高反玻璃上, 待乙醇完全挥发后进行激光红外成像光谱仪(laser direct infrared, LDIR)的测试. LDIR 的测试: 选择颗粒分析模式, 选择微塑料谱库建立方法, 设置自动测试方法(匹配度 > 0.65), 进行测试. 有机肥中微塑料测定由上海微谱检测科技集团股份有限公司完成.

1.4 微塑料观察与鉴定

使用金相显微镜(CX40M, Ningbo Sunny, China)在放大 50 ~ 100 倍下通过软件程序 IMSA-2000 对干燥滤膜上的微塑料进行观察、拍照和计数. 土壤样品中微塑料的丰度用 $\text{n} \cdot \text{kg}^{-1}$ 表示, 即微塑料的数量与土壤干重之比.

傅里叶变换红外(Fourier transform infrared spectroscopy, FTIR)光谱仪(Nicolet iS50, Thermo, USA)用于验证大于 1 mm 的可疑微塑料. 光谱在反射模式下采集, 采集范围为 $4000 \sim 400 \text{ cm}^{-1}$, 扫描 32 次. 每个样品在不同的点采集两次, 以获得更高的

光谱和吸收峰匹配结果. 与已知的聚合物光谱相比, 确定微塑料的聚合物类型, 并确定微塑料的类型^[26].

利用扫描电子显微镜(scanning electron microscope, SEM)对土壤微塑料样品表面进行观察, 以了解土壤中微塑料的表面形貌特征. 用 FTIR 分析对微塑料进行鉴定后, 样品涂上铂薄膜, 用 10 kV 可变真空钨丝扫描电子显微镜(EVO-LS10, 蔡司, 德国)观察. 由于微塑料表面的侵蚀程度不同, 在不同的表面位置和粗糙度下至少进行 3 次可视化分析.

1.5 数据计算与分析

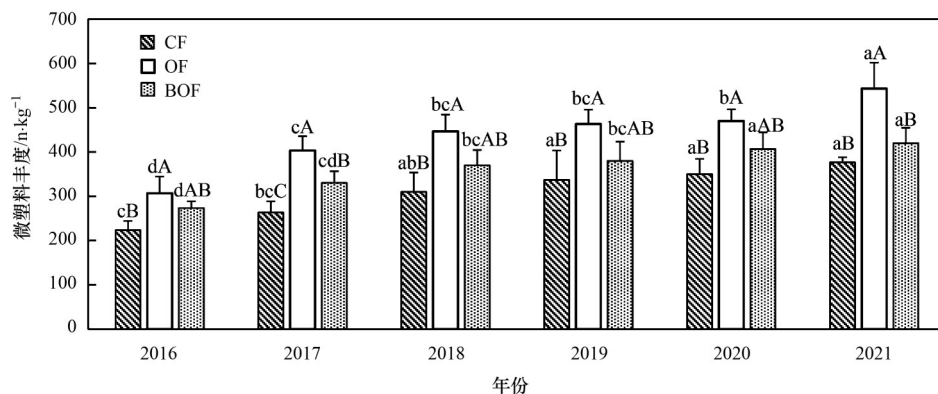
使用 Origin 2022b 和 WPS 对图表进行绘制, 使用 SPSS v. 27(IBM 公司, Armonk, NY, USA)进行双因素方差分析检验, 来确定 $P < 0.05$ 时种植年限和施肥类型、土壤深度和施肥类型是否具有统计学意义, 使用单因素方差分析用于评估不同施肥类型之间的显著性差异.

2 结果与分析

2.1 不同种植年限及施肥模式对土壤中微塑料的影响

通过检测, 所有土壤样品中均检测出微塑料存在, 表层土壤微塑料丰度见图 1. 随着种植年限的增加微塑料的丰度呈上升趋势, 其中所有土壤样品在 2016 ~ 2021 年的微塑料平均丰度分别为: 267.78、332.22、375.56、393.33、408.89 和 $446.67 \text{ n} \cdot \text{kg}^{-1}$, 其中 2021 年长期施用 OF 的土壤微塑料丰度最大, 丰度为 $543.33 \text{ n} \cdot \text{kg}^{-1}$.

不同施肥处理不同土层中微塑料丰度见图 2, 不同土层微塑料丰度的方差分析结果显示, 3 种施肥处理在不同土层的微塑料丰度存在显著差异 ($P < 0.001$). 3 种处理中 0 ~ 20、20 ~ 40 和 40 ~ 60 cm 土层中微塑料丰度均呈现递减的趋势, 且 CF 处理中 0 ~ 20 cm 土层微塑料向 20 ~ 40 cm 土层的



不同大写字母表示同年份不同施肥间土壤微塑料显著性差异, 不同小写字母表示同施肥不同年份间土壤微塑料显著性差异

图 1 不同种植年限表层土壤中微塑料丰度

Fig. 1 Abundance of microplastics in surface soil with different planting years

迁移效率为 17.70%, 向 40 ~ 60 cm 土层的迁移效率为 2.45%; OF 处理中 0 ~ 20 cm 土层微塑料向 20 ~ 40 cm 土层的迁移效率为 19.75%, 向 40 ~ 60 cm 土层的迁移效率为 6.02%; BOF 处理中 0 ~ 20 cm 土层微塑料向 20 ~ 40 cm 土层的迁移效率为 26.71%, 向 40 ~ 60 cm 土层的迁移效率为 13.71%,

BOF 处理迁移效率最高。

3 种施肥处理微塑料丰度除不同土层差异, 在微塑料增长程度上也存在差异. 在 0 ~ 20 cm 土层下微塑料的年平均增长速率存在差异, 但差异不显著. CF、OF 和 BOF 处理中微塑料的平均年增长速率分别为 11.16%、12.61% 和 9.17%.

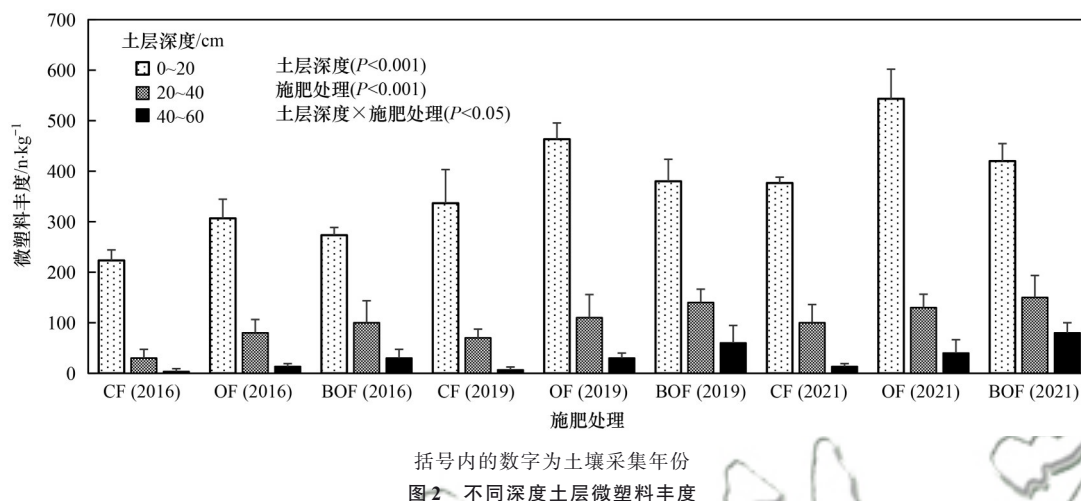


Fig. 2 Abundance of microplastics in soil layers at different depths

2.2 土壤中微塑料的形状及粒径分布特征

图 3 结果表明, 在土壤样品中发现了 4 种主要形状的微塑料(纤维、碎片、颗粒和薄膜), 微塑料的形状占比如图 4(a) 所示. 在 0 ~ 20 cm 范围内, CF 处理中 4 种类型微塑料占比分别为 38.33%、21.60%、16.30% 和 23.77%, OF 处理中 4 种类型微塑料占比分别为 40.86%、22.84%、21.57% 和 14.72%, BOF 处理中 4 种类型微塑料占比分别为 40.37%、15.84%、19.57% 和 24.22%. 20 ~ 40 cm 范围内, CF 处理中纤维状微塑料占比为 60.00%, 其次是颗粒状微塑料(25.00%), 碎片状(10.00%)和薄膜状(5.00%)占比较少, OF 处理中纤维状微塑料占比为 59.38%, 其次是碎片状微塑料(21.88%), 颗粒状(15.63%)和薄膜状(3.13%)占比较少, BOF 处理中纤维状微塑料占比为 66.67%, 其次是颗粒状微塑料(12.82%), 碎片状(10.26%)和薄膜状(10.26%)占比较少. 在 40 ~ 60 cm 深度土层中, CF 处理中仅存在颗粒状微塑料(57.14%)和碎片状微塑料(42.86%), OF 处理中纤维状微塑料和颗粒状微塑料占比均为 36.00%, 碎片状微塑料占比为 28.00%, BOF 处理中颗粒状微塑料占比为 41.18%, 其次是碎片状微塑料(35.29%), 纤维状微塑料(17.65%)和薄膜状微塑料(5.88%)占比较少.

通过显微镜观察微塑料粒径并计数, 将粒径分为 <0.5、0.5 ~ 1、1 ~ 3 和 3 ~ 5 mm 共 4 个范围, 如图 4(b) 所示, 土壤中微塑料丰度均随粒径增大而减

小. 表层土壤中, CF、OF 和 BOF 这 3 个处理中微塑料的粒径主要分布在 < 0.5 mm 范围, 其中粒径 < 0.5 mm 的微塑料占比分别为 55.52%、53.55% 和 61.18%, 0.5 ~ 1 mm 范围的微塑料占比分别为 27.05%、27.41% 和 26.40%, 1 ~ 3 mm 范围的微塑料占比分别为 9.96%、10.91% 和 6.52%, 3 ~ 5 mm 范围的微塑料占比分别为 7.47%、8.12% 和 5.90%. 随着土壤深度的增加, CF 和 OF 处理大尺寸微塑料占比逐渐减少, 而 BOF 处理中 < 0.5 mm 微塑料占比为 33.33%, 0.5 ~ 1 mm 微塑料占比为 25.64%, 1 ~ 3 mm 微塑料占比为 23.08%, 3 ~ 5 mm 微塑料占比为 17.95%, 大尺寸微塑料占比增多. 在 40 ~ 60 cm 土壤深度时 CF 和 OF 处理中微塑料粒径均 < 1 mm, BOF 处理中 < 1 mm 微塑料占比为 94.12%, 1 ~ 3 mm 微塑料占比为 5.88%.

2.3 微塑料的聚合物类型及表面形貌特征

微塑料聚合物类型是确定其来源的主要方法. 通过傅里叶红外光谱, 对微塑料聚合物的红外吸收特征进行分析, 发现研究区域主要有 5 种微塑料类型(图 5), 分别为: 聚乙烯(polyethylene, PE)、聚氯乙烯(polyvinyl chloride, PVC)、聚丙烯(polypropylene, PP)、聚对苯二甲酸乙二醇酯(polyethylene terephthalate, PET) 和 聚 苯 乙 烯(polystyrene, PS). 在 CF、OF 和 BOF 这 3 个处理中 PE(52.14%、49.24% 和 55.96%) 均占主要地位, 其次是 PVC(16.04%、14.50%、11.93%)、PP(12.83%、

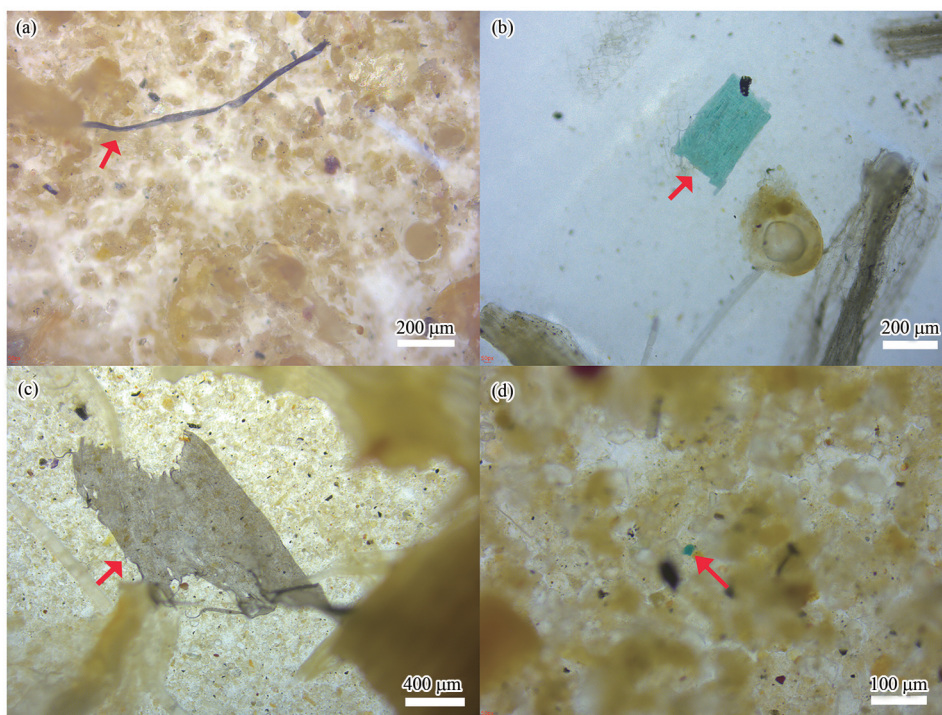


图3 不同形状微塑料显微照片

Fig. 3 Micrographs of microplastics of different shapes

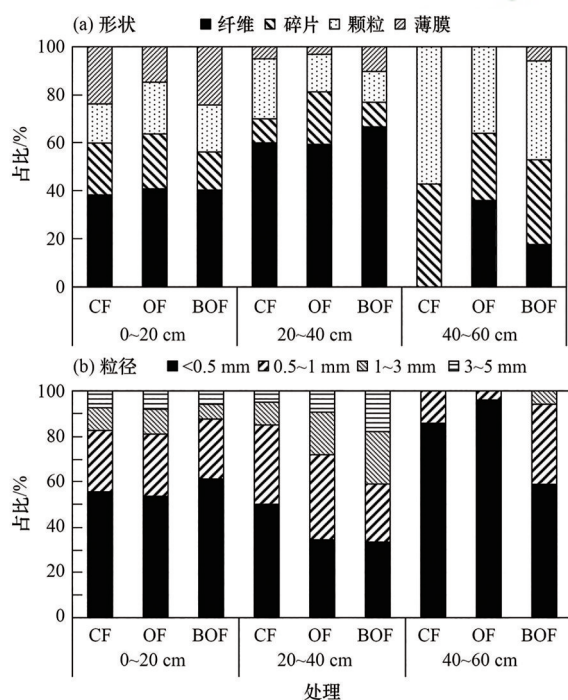


图4 不同深度及不同施肥土壤微塑料占比

Fig. 4 Proportions of microplastics in the soil with different depths and different fertilization treatments

13.74%、11.01%) 和 PET (13.37%、10.31%、11.93%), 占比最少的为 PS (5.35%、12.21% 和 9.17%)。

通过扫描电子显微镜(SEM)图像显示(图6), 因农事操作及时间因素, 微塑料经历了不同程度的侵蚀和风化, 其表面特征表现为多孔、撕裂、裂缝和磨损等特点。

2.4 有机肥中微塑料种类及丰度

进一步探讨施用有机肥对土壤中微塑料的贡献, 所有检测的有机肥样品中均发现微塑料(表1). 在有机肥中检测出4种类型微塑料: PE、PET、PP 和 PVC, 与土壤中微塑料类型相同. 有机肥中微塑料平均丰度为 $577.78 \text{ n} \cdot \text{kg}^{-1}$. 2016年施用的有机肥中微塑料平均丰度为 $333.33 \text{ n} \cdot \text{kg}^{-1}$, 2019年施用的有机肥中微塑料的平均丰度为 $733.33 \text{ n} \cdot \text{kg}^{-1}$, 2021年施用的有机肥中微塑料的平均丰度为 $666.67 \text{ n} \cdot \text{kg}^{-1}$. 其中2016年和2019年有机肥中PET占比最高, 分别为60.00%和72.73%, 在2016年有机肥中, 其次是PP(20.00%)和PE(20.00%); 在2019年有机肥中, 位居第二的是PVC(18.18%)和PP(9.09%). 2021年有机肥中PVC占比最高(50.00%), 位居第二的是PP(20.00%)、PET(20.00%)和PE(10.00%). 有机肥中微塑料存在两种粒径, 2016年微塑料粒径 $<0.1 \text{ mm}$ 的占比为80.00%, $0.1 \sim 0.5 \text{ mm}$ 微塑料占比为20.00%; 在2019年和2021年施用的有机肥中仅存在 $<0.1 \text{ mm}$ 的微塑料。

3 讨论

3.1 长期施用有机肥对土壤微塑料丰度影响

虽然微塑料在农田土壤中有一定的积累和降解趋势, 但由于其持久性强, 导致它们在环境中持续积累^[24,27], 使得土壤中微塑料随着种植年限的增加其丰度不断上升. 且通过对比其他农业土壤微塑料

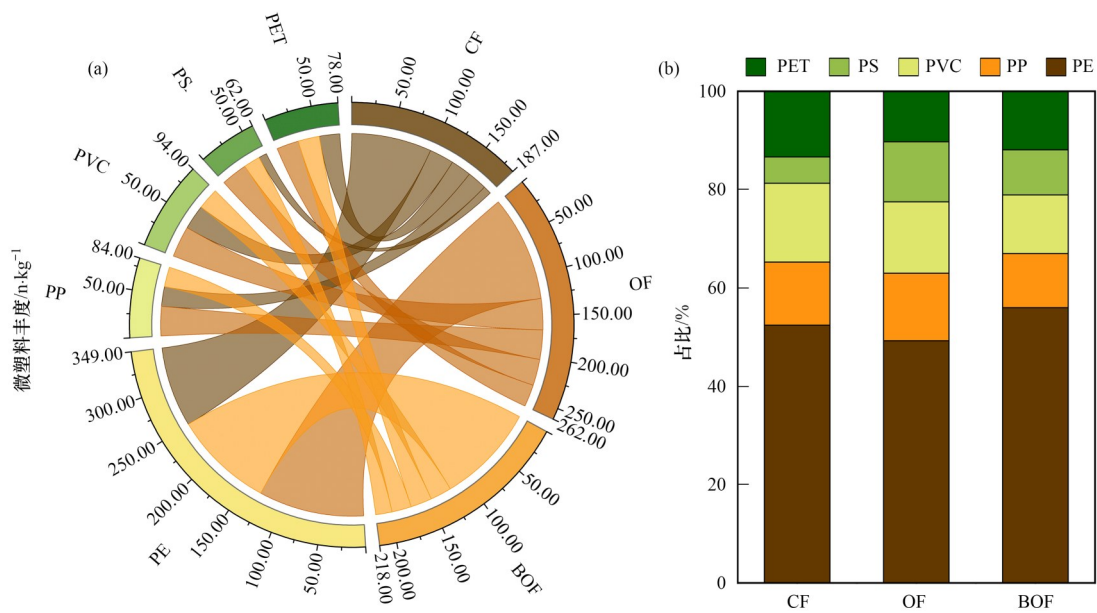


图 5 表层土壤微塑料聚合物类型

Fig. 5 Types of microplastic polymers in surface soil

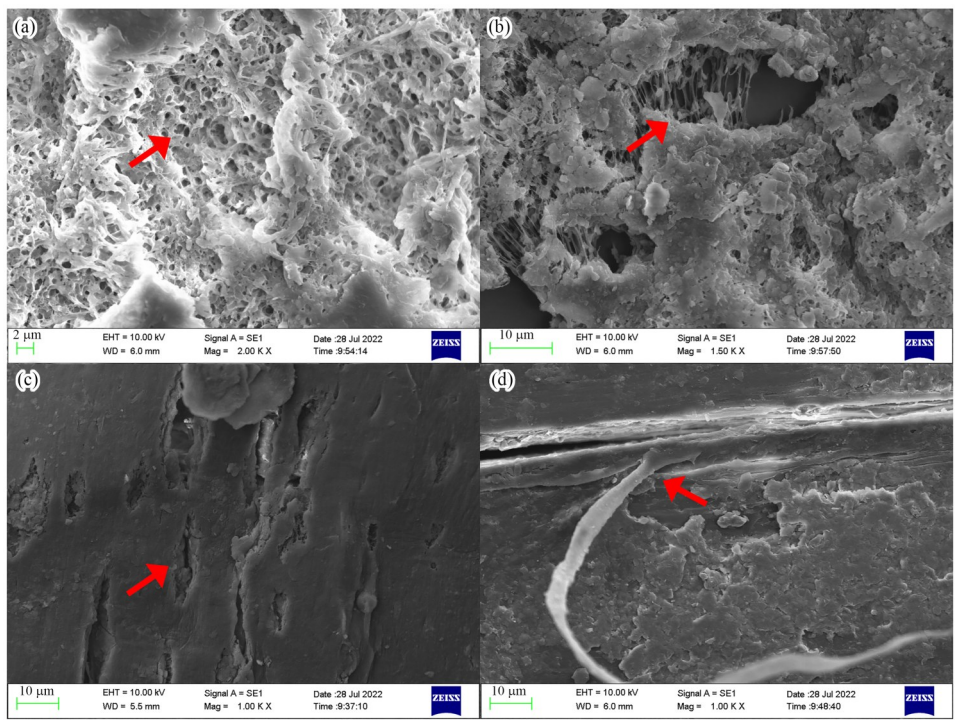


图 6 微塑料表面形貌特征

Fig. 6 Surface morphological characteristics of microplastics

表 1 不同年份有机肥中微塑料种类和粒径丰度/ $n \cdot kg^{-1}$

Table 1 Species and particle size abundance of microplastics in organic fertilizers in different years/ $n \cdot kg^{-1}$

有机肥年份	微塑料种类					微塑料粒径/mm	
	PS	PE	PET	PP	PVC	<0.1	0.1 ~ 0.5
2016	0	66.67	200	66.67	0	266.67	66.67
2019	0	0	533.33	66.67	133.33	733.33	0
2021	0	66.67	133.33	133.33	333.33	666.67	0

丰度发现,本研究结果远低于陕西省农业土壤中微塑料的丰度($1\,430 \sim 3\,410 \text{ n} \cdot \text{kg}^{-1}$)^[28]和中国西南部土壤微塑料丰度($7\,100 \sim 42\,960 \text{ n} \cdot \text{kg}^{-1}$)^[29];高于青藏高原东北设施农业土壤中微塑料丰度($20 \sim 110 \text{ n} \cdot \text{kg}^{-1}$)^[30];本研究农业土壤微塑料丰度与杭州湾沿海平原农业土壤中微塑料丰度($503.3 \text{ n} \cdot \text{kg}^{-1}$)^[31]相近。

长期施用3种肥料土壤中微塑料向下迁移效率不同,这可能是因为微塑料主要在土壤表层产生,仅有少部分可以通过土壤生物、土壤水分迁移和翻耕等过程迁移至深层土壤^[32],微塑料的大量残留可能会影响植物种子和根系的生长发育^[33]。并且在3种处理中,施加生物炭的土壤微塑料向下迁移效率最高,可能由于生物炭的施加使土壤容重降低,含水率增加^[34],使微塑料更易向下迁移。

先前研究表明,微塑料广泛存在于农业土壤中^[35]。以上研究的异质性较高的可能原因是取样方法、施肥类型和施肥方法、农业土壤用途和栽培过程中的人类活动等均存在差异^[36]。在本试验于2016年、2019年和2021年施用的有机肥中检测出微塑料平均丰度分别为333.33、733.33和666.67 $\text{n} \cdot \text{kg}^{-1}$,且在有机废弃物的消化过程中,有机肥中微塑料的丰度为 $14 \sim 895 \text{ n} \cdot \text{kg}^{-1}$ ^[10],在猪粪中也检测出微塑料^[37]。由此发现,微塑料不仅会在堆肥过程中进入有机肥,养殖生产过程中也会存在一定的微塑料迁移至猪粪中。所以随着肥料的长期施用,种植年限的升高,覆膜时间的增加,导致大量地膜风化残留、降解后变为微塑料,逐渐积累于农田土壤中,且因长期得不到治理,导致种植的时间越久,微塑料的积累程度越高^[38]。

3.2 长期施用有机肥对土壤微塑料形状和粒径的影响

目前,相关研究中不同土层微塑料丰度的垂直分布并不一致^[39],研究结果的不一致受多种因素影响,如生物干扰、机械干扰、径流、渗透、作物轮作和施肥等。在 $0 \sim 20 \text{ cm}$ 耕作层的CF和BOF处理中纤维状微塑料和薄膜状微塑料占比较多,可能主要来源于农用地膜的破碎^[40],且CF和BOF处理土壤的理化性质可能易使农用地膜的破碎。而OF处理中薄膜状微塑料占比较少,可能因有机肥中带入部分微塑料造成。且由于地膜的不易降解性,使得农田土壤中纤维状微塑料和薄膜状微塑料占比较大。在 $20 \sim 40 \text{ cm}$ 土层中3种处理中均为纤维状微塑料占比最高,这可能由于种植时土壤翻动、耕作等原因使得薄膜状微塑料碎裂为纤维状微塑料,向下层土壤迁移。在 $40 \sim 60 \text{ cm}$ 土层中CF处理仅存在碎片状微塑料和颗粒状微塑料,OF处理中无薄膜状微塑料,

只有BOF处理中存在少量的薄膜状微塑料,可能因为薄膜状微塑料不易向下层土壤迁移,但是BOF处理中生物炭对土壤孔隙度的增大作用^[41]使得薄膜状微塑料向下迁移。同时,生物炭也会影响土壤的物理、化学和生物性质,进而影响土壤水分和养分保持、微生物活性和作物生长,从而也会影响土壤中微塑料的丰度^[42,43]。

以往的研究证实,云南高原耕地和瑞士泛滥平原等土壤中也以小尺寸微塑料为主^[44,45]。微塑料主要通过机械、化学和生物手段形成小颗粒,微塑料的小颗粒容易被生物吸收^[46,47]。因此,小体积的微塑料对生态系统和人类健康的危害更大。并且通过分析 $40 \sim 60 \text{ cm}$ 土壤深度微塑料的形状和粒径占比,发现CF和OF处理中不存在薄膜状微塑料和大尺寸微塑料,BOF处理由于生物炭会增大土壤孔隙度,土壤更松散,使得更丰富的微塑料向下迁移,长期施用会对环境带来更大的风险。

本研究中有有机肥微塑料粒径也存在差异,且每年施用的有机肥中微塑料以小尺寸为主。Yang等^[48]调查了猪粪中微塑料,发现猪粪中微塑料的年平均丰度是 $(1\,250 \pm 640) \text{ n} \cdot \text{kg}^{-1}$,在施用猪粪土壤中微塑料明显高于不施用猪粪的土壤^[7]。刘淑丽等^[49]在鸟类粪便中也检测到微塑料 $[(4.93 \pm 4.25) \text{ n} \cdot \text{g}^{-1}]$ 。另一项研究发现,鸡粪中存在高达 $(129.8 \pm 82.3) \text{ n} \cdot \text{g}^{-1}$ 的微塑料颗粒^[38]。粪便中微塑料的存在表明,它们已经进入了动物和人类,可能是来自受到污染的水、空气和食物中^[50,51]。因此,不同施肥处理中微塑料粒径的差异可能是肥料来源不同造成的,有机肥成分相对复杂造成有机肥中小尺寸微塑料的丰度增加,使得土壤中微塑料的丰度增加,且这些小尺寸微塑料还会使土壤风险升高。

3.3 长期施用有机肥对土壤微塑料聚合物类型及表面形态的影响

对研究区域中土壤微塑料污染现状调查,发现研究区的主要塑料制品有农用地膜(PP和PVC)、塑料捆扎绳和编织袋(PP)、塑料袋(PE)、塑料包装瓶(PET)和泡沫包装(PS)等,与检测出微塑料类型相同。相关研究表明PE(20.90%)和PA(0.30%)是中国5省农田中含量最高的微塑料^[52],在特定的区域中微塑料类型与微塑料的来源密切相关^[53]。不同施肥处理影响微塑料类型,本研究结果表明有机肥处理土壤中微塑料各聚合物类型更为丰富,且在本研究中的有机肥中也检测出多种微塑料类型,除PE和PVC类型微塑料外,其他类型微塑料占比较高,这可能与有机肥的来源复杂有关。Zhang等^[54]发现施用有机肥的土壤比施用其他肥料的土壤中微塑料更多样且

更丰富, 这表明有机肥向土壤中输入了额外的微塑料, 这与本研究的结果一致. 因此, 肥料、农药和农膜等投入品及包装物不能有效处理或作为废物丢弃可能会增加土壤中微塑料的丰度^[31]. Corcoran 等^[55]认为微塑料表面的纹理特征与线性裂纹平行的边缘具有易被氧化的特点, 故微塑料被氧化后的表面特征使得微塑料比表面积增大, 增加了对污染物的吸附, 使得微塑料对环境的风险增大.

4 结论

(1) 通过对长期定位试验小区土壤检测发现, 2021 年长期施用 OF 的土壤微塑料的丰度最高 ($543.33 \text{ n} \cdot \text{kg}^{-1}$), 且设施土壤中微塑料丰度随种植年限的上升而升高, CF、OF 和 BOF 处理中微塑料的年平均增长速率分别为 11.16%、12.61% 和 9.17%.

(2) 不同施肥处理对微塑料丰度存在影响, 其中 OF 微塑料丰度最高, 且有机肥会向土壤中带入大量微塑料.

(3) 微塑料在不同深度土层中表现为随土层深度的增加, 微塑料丰度逐渐减少, 且 BOF 处理中微塑料向深层土壤迁移效率最高.

(4) 长期施肥设施土壤中发现纤维、碎片、颗粒和薄膜微塑料这 4 种形态微塑料. 纤维状微塑料在 0~40 cm 土壤中占比最高, 在 40~60 cm 土壤中 CF 处理颗粒状微塑料占比最高 (57.14%), OF 处理颗粒状微塑料和纤维状微塑料占比最高 (36.00%), BOF 处理中颗粒状微塑料占比最高 (41.18%).

(5) 在各土层的 3 种施肥处理土壤中微塑料均以小尺寸 (<0.5 mm) 为主, 且在 CF、OF 和 BOF 3 种施肥类型土壤中 PE 微塑料占主导地位, 占比分别为 52.14%、49.24% 和 55.96%.

(6) 长期施肥的设施土壤中检测出的微塑料表面均呈现多孔、撕裂、裂痕和磨损等特点.

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CONTENTS

Spatio-temporal Evolution, Dynamic Transition, and Convergence Trend of Urban Carbon Emission Intensity in China	YANG Qing-ke, WANG Lei, ZHU Gao-li, <i>et al.</i> (1869)
Spatio-temporal Evolution and Trend Prediction of Transport Carbon Emission Efficiency	ZHENG Yan, JIANG Xue-mei, XIAO Yu-jie (1879)
Research on Carbon Emission Decoupling Factors Based on STIRPAT Model and LMDI Decomposition	ZHANG Jiang-yan (1888)
Carbon Peak Pathways of Industrial Parks Based on the LEAP Model: A Case Study of a National Development Zone in Nanjing	LI Hui-peng, LI Li, YIN Yin, <i>et al.</i> (1898)
Accounting and Analysis of Carbon Emissions in Universities: A Case Study of Beijing A University	CAO Rui, FENG Li, ZHANG Li-qiu (1907)
Synergy Effects and Driving Factors of Pollution and Carbon Emission Reduction in Manufacturing Industry in Beijing	YU Shan, HAN Yu-hua, MU Jie, <i>et al.</i> (1917)
Carbon Emissions Trading Prices and Total Factor Productivity: Evidence from China	WU Xue-ping (1926)
Analysis of Spatio-temporal Distribution Characteristics and Influencing Factors of PM _{2.5} and PM ₁₀ in Chinese Cities	LI Jiang-su, DUAN Liang-rong, ZHANG Tian-jiao (1938)
Spatial-temporal Characteristics and Influencing Factors of PM _{2.5} and Ozone in the Border Area of Jiangsu, Anhui, Shangdong, and Henan from 2017 to 2021	CHEN Wei, XU Xue-zhe, LIU Wen-qing (1950)
Analysis of the Characteristics and Causes of PM _{2.5} and O ₃ Pollution in Xi'an During the Epidemic Lockdown Period	YUAN Xiao-hong, ZHANG Qiang, LI Qi, <i>et al.</i> (1963)
PM _{2.5} Chemical Composition and Spatial Variability in the Five Cities of Southern Jiangsu During Fall and Winter	FENG Wei, DING Feng, SHANG Yue, <i>et al.</i> (1975)
Source Apportionment of Fine Particles in Xianning, Hubei	LUO Yi, ZHU Kuan-guang, CHEN Pu-long, <i>et al.</i> (1983)
Spillover Benefits of Ammonia Emission Reduction in Xingtai Area to the Improvement of Atmospheric PM _{2.5} Concentration in the Beijing-Tianjin-Hebei Region	BIAN Ze-jun, WEN Chao-yu, LANG Jian-lei, <i>et al.</i> (1994)
Characteristics, Risk Assessment, and Sources of the Polluted Metallic Elements in PM _{2.5} During Winter in Liaocheng City	ZHANG Jing-qiao, ZHU Yao, CAO Ting, <i>et al.</i> (2003)
Composition Characteristics of Typical VOCs Sources in the Highland City of Lhasa	GUO Shu-zheng, YE Chun-xiang, LIN Wei-li, <i>et al.</i> (2011)
VOCs Pollution Characteristics and Health Risk Assessment in Typical Industrial Parks in Beijing: Environmental Impact of High and New Technology Industries	WANG Jie, YAO Zhen, WANG Min-yan, <i>et al.</i> (2019)
Analysis of a Typical Ozone Pollution Process in the Chengdu Plain Under the Influence of High Temperature Extremes	LEI Li-juan, ZHANG Yi, LUO Yi-na, <i>et al.</i> (2028)
Presence, Behavior, and Risk of Antiviral Drugs in the Aqueous Environment	GE Lin-ke, LI Xuan-yan, CAO Sheng-kai, <i>et al.</i> (2039)
Spatio-temporal Variation and Probability Health Risk of Heavy Metals in Surface Water of Xiaolangdi Reservoir in the Yellow River	WANG Liang, DENG Xue-jiao, WANG Xiao-lei, <i>et al.</i> (2054)
Hydrochemical Characteristics and Control Factors of Strontium Enrichment of the Duoqu River Basin in Eastern Xizang	LI Jing-jie, LIAN Sheng, WANG Ming-guo, <i>et al.</i> (2067)
Characteristics of Stable Isotopes in Precipitation and Its Moisture Sources in the Inland Regions of Northwest China	ZHANG Yan-yan, XIN Cun-lin, GUO Xiao-yan, <i>et al.</i> (2080)
Recharge Source and Evolution Process of Karst Groundwater in Tai'an Urban Area Based on Hydrochemistry and Hydrogen and Oxygen Isotopes	MENG Ling-hua (2096)
Hydrochemical Characteristics and Influencing Factors of Surface Water and Groundwater in the Mountainous Area of Danjiang River Basin	ZHANG Zi-yan, FU Yong-peng, WANG Ning-tao, <i>et al.</i> (2107)
Hydrochemical Characteristics and Control Factors of Groundwater in Yuanyangchi Irrigation Area, Jinta Basin	WANG Xiao-yan, HAN Shuang-bao, ZHANG Meng-nan, <i>et al.</i> (2118)
Hydrochemical Characteristics and Health Risk Assessment of Shallow Groundwater in the Plain Zone of Deyang City	LIU Nan, CHEN Meng, GAO Dong-dong, <i>et al.</i> (2129)
Spatial and Temporal Distribution of Microplastics in the Sediments of Typical Urban River Network	XU Wan-lu, FAN Yi-fan, QIAN Xin (2142)
Construction of Nitrogen Emission Inventory at Sub-basin Scale and Analysis of Key Sources in Fuxian Lake Watershed	WANG Yan-jie, LIANG Qi-bin, WANG Yan-xia, <i>et al.</i> (2150)
Preparation of Lanthanum Crosslinked Calcium Peroxide/Sepiolite/Sodium Alginate Composite Hydrogels and Their Elimination Performance for Endogenous Phosphorus	QU Si-tong, SHAN Su-jie, WANG Chong-ming, <i>et al.</i> (2160)
Formation Mechanism, Structural Characteristics of Ultrafine Mineral Particles, and Their Environmental Effects	LIU Zhen-hai, ZHANG Zhan-hua, YUAN Yu-xin, <i>et al.</i> (2171)
Research Progress in Application of Biochar-immobilized Bacteria Composites in Environmental Remediation	SUN Shu-yu, HUANG Meng-xin, KONG Qiang, <i>et al.</i> (2185)
Effect of Microplastics on Ammonia Nitrogen Adsorption by Zeolite in Water and Its Mechanism	LIAN Jian-jun, XIE Shi-ting, WU Pei, <i>et al.</i> (2195)
Mechanism of Ultraviolet Aging Effect on the Adsorption of Ciprofloxacin by Nano-biochar	MA Feng-feng, XUE Zhi-yi, ZHAO Bao-wei (2203)
Response of Phytoplankton Communities and Environmental Factors Under the Influence of Land Use in the Wuding River Basin	GUO Shan-song, HU En, DING Yi-tong, <i>et al.</i> (2211)
Bacterial Community Diversity and Functional Gene Abundance of Culturable Bacteria in the Wetland of Poyang Lake	YU Jiang, WANG Chun, LONG Yong, <i>et al.</i> (2223)
Bacterial Community Structure and Its Relationship with Heavy Metals in Sediments of Diannong River	LIU Shuang-yu, MENG Jun-jie, QIU Xiao-cong, <i>et al.</i> (2233)
Impacts of Treated Wastewater on Bacterial and Fungal Microbial Communities in Receiving Rivers	GUO You-shun, YU Zhong, HAO Wen-bin, <i>et al.</i> (2246)
Metagenomics Reveals the Characteristics and Functions of Bacterial Community in the Advanced Wastewater Treatment Process	HU Jian-shuang, WANG Yan, ZHOU Zheng, <i>et al.</i> (2259)
Spatio-temporal Characteristics of Habitat Quality and Natural-human Driven Mechanism in Dabie Mountain Area	ZHENG Ya-ping, ZHANG Jun-hua, TIAN Hui-wen, <i>et al.</i> (2268)
Hyperspectral Inversion of Soil Organic Matter Content Based on Continuous Wavelet Transform, SHAP, and XGBoost	YE Miao, ZHU Lin, LIU Xu-dong, <i>et al.</i> (2280)
Effects of Straw Retention, Film Mulching, and Nitrogen Input on Soil Quality in Dryland Wheat Field	YE Zi-zhuang, WANG Song-yan, LU Xiong, <i>et al.</i> (2292)
Effects of Long-term Application of Organic Fertilizer on the Occurrence and Migration Characteristics of Soil Microplastics	WANG Chang-yuan, MA Xiao-chi, GUO De-jie, <i>et al.</i> (2304)
Effect of Different Soil Salinities on N ₂ O Emission: A Meta-analysis	HUANG Yi-hua, SHE Dong-li, SHI Zhen-qi, <i>et al.</i> (2313)
Assessment and Prediction of Carbon Storage Based on Land Use/Land Cover Dynamics in the Guangdong-Hong Kong-Macao Greater Bay Area	ZHENG Hui-ling, ZHENG Hui-feng (2321)
Simulation of Temporal and Spatial Changes in Ecosystem Carbon Storage in Funiu Mountains Based on InVEST Model	ZHANG Zhe, SHI Zhen-qin, ZHU Wen-bo, <i>et al.</i> (2332)
Relationship Between Microbial Nutrient Limitation and Soil Organic Carbon Fraction During Shelterbelts Construction	XU Feng-jing, HUANG Yi-mei, HUANG Qian, <i>et al.</i> (2342)
Characteristics and Driving Forces of Organic Carbon Mineralization in Brown Soil with Long-term Straw Returning	ZHAO Yu-hang, YIN Hao-kai, HU Xue-chun, <i>et al.</i> (2353)
Effects of Winter Green Manure Mulching on Soil Aggregates, Organic Carbon, and AMF Diversity	LU Ze-rang, CHEN Jia-yu, LI Zhi-xian, <i>et al.</i> (2363)
Spatial and Temporal Distribution of Nitrogen in the Liaohe River Basin and Its Responses to Land Use and Rainfall	ZHOU Bo, LI Xiao-guang, TONG Si-chen, <i>et al.</i> (2373)
Nitrous Oxide Emissions from Ponds in Typical Agricultural Catchment with Intensive Agricultural Activity	ZHANG Xin-yue, XIAO Qi-tao, XIE Hui, <i>et al.</i> (2385)
Effects of Biochar Combined with Different Types of Nitrogen Fertilizers on Denitrification Bacteria Community in Vegetable Soil	LIU Xiao-wan, LIU Xing-ren, GAO Shang-jie, <i>et al.</i> (2394)
Investigation of Soil Microbial Characteristics During Stand Development in <i>Pinus tabulaeformis</i> Forest in Taiyue Mountain	MA Yi-shu, CAO Ya-xin, NIU Min, <i>et al.</i> (2406)
Simulation of Heavy Metal Content in Soil Based on Sparse Sample Sites	ZHANG Jia-qi, PAN Yu-chun, GAO Shi-chen, <i>et al.</i> (2417)
Identification Priority Source of Heavy Metal Pollution in Greenspace Soils Based on Source-specific Ecological and Human Health Risk Analysis in the Yellow River Custom Tourist Line of Lanzhou	LI Jun, LI Xu, LI Kai-ming, <i>et al.</i> (2428)
Heavy Metal Accumulation and Assessment of Potential Ecological Risk Caused by Soil pH Changes in Different Types of Purple Soils in Southwest China	ZHANG Hai-lin, ZHANG Yu, WANG Ding, <i>et al.</i> (2440)
Assessment and Source Analysis of Heavy Metal Pollution in Arable Land Around an Iron Ore Mining Area in Chongqing	LIAO Ze-yuan, LI Jie-qin, SHEN Zhi-jie, <i>et al.</i> (2450)
Health Risk Assessment of Heavy Metals in Farmland Soils and Crops Around Metal Mines	WEI Hong-bin, LUO Ming, XIANG Lei, <i>et al.</i> (2461)
Effect of Red Mud-based Nano Zero-valent Iron on Remediation of Polymetallic Contaminated Soil	LIU Long-yu, YANG Shi-li, ZHAO Huang-shi-yu, <i>et al.</i> (2474)
Effects of Zinc Application on Cadmium Accumulation in Alkaline Soil-Wheat Seeding Systems	ZHANG Yao, WANG Tian-qi, NIU Shuo, <i>et al.</i> (2480)