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引用本文:

张海红,石岩松,崔文杰,王予頓,秦莉,端正花. 微塑料对农田污染物迁移的促进作用[J]. *农业资源与环境学报*, 2023, 40(3): 619–625.

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微塑料对农田污染物迁移的促进作用

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摘要:为探讨微塑料(MPs)在农田系统的污染情况及可能的作用途径,以湖南省湘潭市3个镇的水稻田为研究对象,测定分析了该地区水稻田56个点位的表层土壤中MPs的分布特征,并在水稻成熟后随机选取14个点位收集稻谷,分析了MPs对土壤中双酚A(BPA)与镉(Cd)向稻谷中迁移的影响。结果表明,该地区水稻田中MPs以老化的薄膜、颗粒、碎片、纤维4种形态存在。3个镇的水稻田土壤中MPs的丰度相似,丰度范围为48~1 771 个·kg⁻¹,主要成分为聚乙烯(PE)和聚氯乙烯(PVC),占比分别为46.77%和24.19%,且聚碳酸酯(PC)含量均显著低于苯二甲酸乙二醇酯(PET)含量,二者平均含量分别为2.51 μg·kg⁻¹和113.35 μg·kg⁻¹。相对于非解聚土壤,解聚土壤中BPA含量与稻谷中的BPA含量呈现出显著的正相关关系($P<0.01$),表明随着环境中MPs老化程度的加重,BPA有二次释放的风险。另外,土壤中Cd的含量也与MPs丰度表现出显著的正相关关系($P<0.001$),表明MPs促进了Cd向稻谷中的转移。本研究对全面、客观评估农田系统中MPs的污染水平和健康风险具有参考意义。

关键词:微塑料;稻田;双酚A;镉;迁移

中图分类号:X56;X505;X71 文献标志码:A 文章编号:2095-6819(2023)03-0619-07 doi: 10.13254/j.jare.2022.0289

Promoting effect of microplastics on the migration of pollutants in farmland

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Abstract: To investigate the pollution caused by microplastics (MPs) in agricultural systems and their possible mechanisms of action, the distribution characteristics of MPs in the surface soils of 56 sites in paddy fields of three towns in Xiangtan City, Hunan Province, China, were determined. Fourteen sites were randomly selected to collect rice grains after harvest. The effect of MPs on the migration of bisphenol A (BPA) and cadmium (Cd) from soil to rice grains were analyzed. Four forms of aging MPs were detected in soils: films, particles, fragments, and fibers. The abundance of MPs in the three towns was similar, ranging from 48 items · kg⁻¹ to 1 771 items · kg⁻¹; the main components were polyethylene (PE) and polyvinyl chloride (PVC), accounting for 46.77% and 24.19%, respectively, and the content of polycarbonate (PC) was significantly lower than that of ethylene terephthalate (PET) (2.51 μg · kg⁻¹ and 113.35 μg · kg⁻¹, respectively). BPA in depolymerized soils showed a more significant positive correlation with the content of BPA in rice grains than in un-depolymerized soils ($P<0.01$), indicating a potential risk of secondary release of BPA during the aging of MPs in the environment. In addition, the Cd content in soils also exhibited a significant positive correlation ($P<0.001$) with the abundance of MPs, promoting the transformation of Cd from soils to rice grains. This study contributes to the overall and objective evaluation of the pollution and health risks posed by MPs in agricultural systems.

Keywords: microplastics; paddy; bisphenol A; cadmium; transformation

收稿日期:2022-05-14 录用日期:2022-07-25

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基金项目:国家自然科学基金项目(41807487);国家级大学生创新创业训练项目(202010060009)

Project supported: The National Natural Science Foundation of China (41807487); National College Students Innovation and Entrepreneurship Training Program of China (202010060009)

塑料制品具有化学稳定和抗腐蚀等优良特性,因而生产量和使用量逐年稳步增长,但是目前只有少部分被回收利用,大部分被废弃在环境中^[1]。塑料在生产、使用以及环境的老化过程中会降解形成微塑料(Microplastics, MPs)^[2]。研究表明,每年排放到陆地中的MPs的数量是排放到海洋中的4~23倍^[3]。农田中MPs的主要来源包括农用地膜残留、使用未经处理的污水灌溉和使用污水处理厂排放的污泥作为土壤改良剂等^[4~5]。塑料地膜使用后若得不到有效回收,会长期存在于土壤中^[6],对土壤密度、孔径分布等物理性质产生影响,同时可能影响土壤中生物的活性和多样性,以及植物健康^[7]。

除了MPs本身对环境的影响外,塑料产品在制造过程中也会加入很多添加剂以获得更好的性能。然而这些添加剂通常不会与塑料聚合物本身结合成为聚合物大分子的一部分^[8],导致添加剂会在使用过程中逐渐释放出来^[9]。随着塑料在环境中逐渐分解变成小塑料颗粒,其表面积增加,扩散到环境界面的路径缩短,添加剂以及未反应单体的释放量呈指数增长,形成“毒性债务”(Toxicity debt)^[10]。另外,MPs的添加剂可能会产生比MPs本身更强的毒性效应^[11]。

塑料产品的很多添加剂是内分泌干扰物,如广泛使用的双酚A(BPA)^[12]。BPA是聚碳酸酯(PC)的合成原料以及重要的增塑剂,已有研究证明其会对生物产生毒性作用^[13]。此外,MPs对重金属表现出很强的吸附能力^[14~15]。重金属镉(Cd)在土壤母质中含量较低,主要通过农药、化肥和工业污染进入农田^[16]。湖南地区由于采矿区与水稻种植区较近,土壤中Cd含量显著高于其他地区^[17]。随着季节交替和土壤环境变化,MPs也会释放其吸附的重金属^[18]。

因此,本研究选取湖南省湘潭市的水稻田作为研究区域,调查该地区MPs的丰度及其污染特征,并分析MPs对稻田土壤中BPA和Cd向稻谷中迁移的贡献,以评估其对人类健康的潜在风险。

1 材料与方法

1.1 样品采集

研究区域位于湖南省湘潭市的水稻生产基地。利用大样本统计法,选取杨嘉桥镇(Y1~Y16,共16个)、河口镇(H1~H20,共20个)和谭家山镇(T1~T20,共20个)三个镇共计56个点位作为研究对象,采样点位置如图1所示,研究区位于北纬27°36'~27°54',东

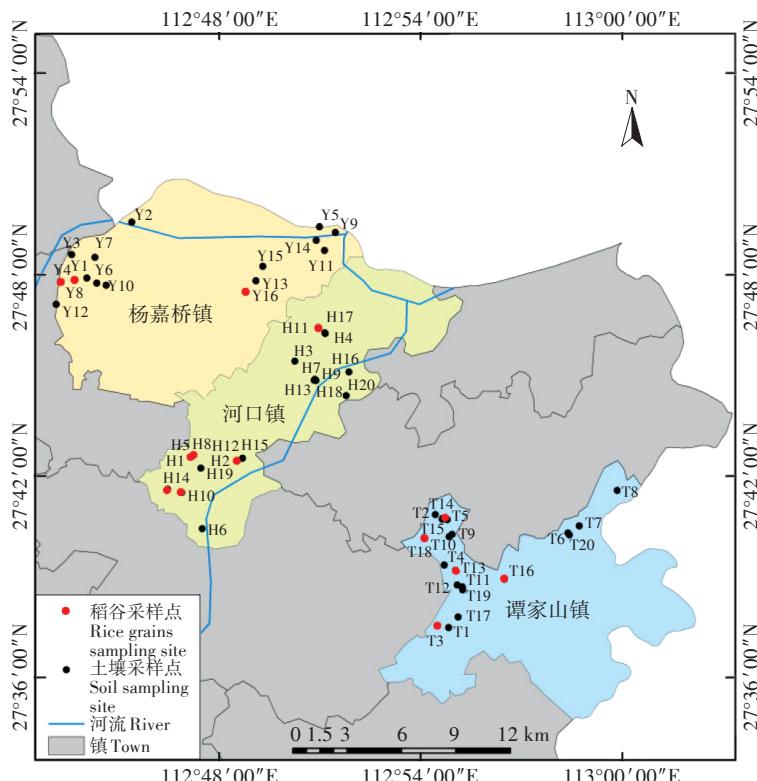


图1 研究区域位置及采样点分布图

Figure 1 The distribution of sampling sites in the study area

经 $112^{\circ}40' \sim 113^{\circ}00'$,海拔0~89 m。土样采集时间为2020年4月20日。用不锈钢铲采集土壤表层10 cm土样,并在水稻成熟后随机收集该地区14个采样点位的稻谷。将采集的土壤和稻谷样品装入牛皮纸袋并运回实验室,放置于-20 ℃冰柜内保存备用。

1.2 MPs的筛选、鉴定与表征

参照Duan等^[19]和岳俊杰等^[20]的研究方法,将土样于70 ℃下干燥24 h,期间用铝箔覆盖防止受到空气沉降物的污染。土样捣碎后用2 mm筛网过滤,称取1 kg(干质量)土壤作为1份样品。使用浓度为1.6 g·mL⁻¹的氯化锌溶液进行浓度分选,搅拌30 min后静置6 h,收集上层包含悬浮物的液体,使用0.45 μm尼龙滤膜(Whatman,美国)抽滤分离悬浮物,上述浓度分选重复3次。收集的悬浮物加入30% (V/V)过氧化氢溶液,在70 ℃下水浴加热12 h以消解天然有机物,再次使用0.45 μm尼龙滤膜抽滤,干燥后采用体视显微镜(SZX10,Olympus Corp.,日本)对滤膜上的微塑料从左到右Z字形进行识别、拍照和计数。采用显微傅立叶变换红外光谱仪(FT-IR,Thermo Nicolet iN10,美国)对微塑料进行鉴定。土壤中MPs丰度单位为个·kg⁻¹。选取其中典型的MPs用扫描电子显微镜(SEM,Hitachi SU3500,日本)进行表征。

1.3 有机物测定

1.3.1 解聚土壤样品中有机物的测定

环境中的MPs难以直接分离测定,高分子PC与苯二甲酸乙二醇酯(PET)可以解聚成小分子单体BPA和对苯二甲酸(PTA),用解聚后的单体含量通过公式换算得出实际PC与PET的含量。使用Wang等^[21]的检测方法,土样研磨后干燥并用2 mm筛网过滤,称取1 g土样并加入250 ng D₄-TPA和50 ng ¹³C₁₂-BPA作为内标,再加入0.1 g氢氧化钾和20 mL正戊醇,135 ℃加热30 min,冷却后用超纯水萃取两次解聚物。振荡离心后再用超纯水定容至50 mL,按照上述过程做一个正戊醇加氢氧化钾的空白对照。用移液枪吸取5 mL离心管中的反萃取水溶液,将其转移至15 mL离心管中,pH调节至2~3,用HLB固相萃取柱(Poly-106 Sery HLB 3 cc/60 mg;CNW Technologies,中国)纯化,压缩氮气浓缩溶液并用甲醇溶液定容至1.5 mL。离心后取上清液,使用液相二级质谱(LC-MS/MS,Agilent Technologies,美国)测定BPA和PTA含量。BPA和PTA加标回收率分别为92.6%±0.8%和96.3%±1.4%,最低检测限分别为0.10 μg·kg⁻¹和4.60 μg·kg⁻¹,经此换算的PET和PC检测限分别为5.3 μg·

kg⁻¹和0.1 μg·kg⁻¹^[21]。

1.3.2 非解聚土壤和稻谷样品中BPA的测定

土壤和稻谷样品研磨干燥并用2 mm筛网过滤,取1 g土样并加入50 ng ¹³C₁₂-BPA作为内标,再加入10 mL 2:8 (V/V)的甲醇-0.001 mol·L⁻¹乙酸钠混合液,混合均匀后加入4 mL 7:3 (V/V)的正己烷-乙醚混合萃取液,萃取3次,离心后取上清液,用LC-MS/MS测定BPA含量。

1.4 重金属Cd的测定

土壤样品采用四分法缩分至约150 g。经过干燥和研磨后,取0.5 g土壤样品和稻谷样品分别放入石墨消解仪(GS-20,南京滨正红仪器有限公司),加入7 mL硝酸(65%)和3 mL氢氟酸(40%)于180 ℃消解2 h,待消解液冷却至室温后用滤膜过滤,使用电感耦合等离子体质谱仪(ICP-MS,Agilent 7500a,美国)测定其中Cd的含量。标准溶液和样品溶液测定时均加入5 μg·L⁻¹ Re内标进行质量控制,加标回收率为98.9%±2.6%。

1.5 质量控制

为防止实验室潜在的背景塑料污染,在整个暴露期间,用超纯水冲洗所有实验器皿3次,并用铝箔覆盖。研究人员穿着棉质实验室工作服和丁腈手套,用75%乙醇清洗显微镜和工作台,然后对样品进行鉴定和量化^[22]。所有样品预处理均在通风柜中进行。分别设置3个空白对照样品,以评估实验期间的背景MPs以及BPA和Cd污染,操作同上。

1.6 统计分析

使用ArcGIS 10.2绘制采样点分布图。使用Origin Pro 9.0进行相关性分析和图表绘制,通过单因素方差分析(ANOVA)检测差异显著性($P<0.05$)。

2 结果与讨论

2.1 湘潭地区水稻田土壤样品中MPs的分布特征

MPs污染已被视为农业生态系统中一个全球性的新兴问题^[23]。湘潭地区三个镇的水稻田土壤中MPs的丰度相似,丰度范围为48~1 771个·kg⁻¹,其中杨嘉桥镇的MPs平均污染程度较高(图2)。PC和PET含量也表现出类似的空间分布,即杨嘉桥镇的PC和PET含量略高于其他两个镇。但是三个镇的PC含量均显著低于PET含量,PC和PET平均含量分别为2.51 μg·kg⁻¹和113.35 μg·kg⁻¹。因此,该地区受到MPs污染的程度和种类相似。通过傅立叶红外检测及OMNIC图库比对发现,该地区的塑料成分主要

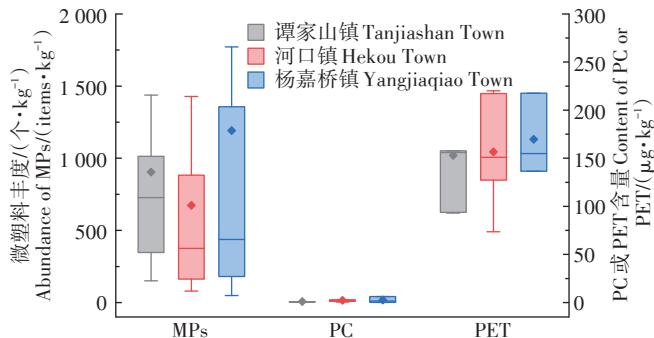


图2 土壤中微塑料丰度及PC和PET的含量

Figure 2 Abundance of MPs and contents of PC and PET in soils

为聚乙烯(PE)和聚氯乙烯(PVC),占比分别为46.77%和24.19%,还包括聚苯乙烯(PS)、PET橡胶(RSB)、尼龙(PA)等(图3)。

有研究表明,上海某农田浅层土壤中MPs的丰度为78个·kg⁻¹,深层土壤中MPs的丰度为62.50个·kg⁻¹,MPs的主要成分为聚丙烯(PP)和PE^[24]。武汉城郊某蔬菜种植地中MPs的丰度为320~12 560个·kg⁻¹,PP和PA是其主要成分^[25]。而本研究区域土壤中MPs丰度跨度也较大,并且主要成分为PE和PVC,表明不同类型土壤中MPs丰度和种类具有较大差异,这可能是由研究区域差异及土壤中MPs的量化无统一标准方法导致的^[26]。MPs输入农业生态系统土壤的途径有多种^[27],其中,农业活动(包括塑料地膜的使用、污泥的肥料利用和污水灌溉)为主要方式^[28]。地膜覆盖可以通过改善作物生长条件来提高作物产量,近年来我国耕地地膜的覆盖面积达到 1.84×10^7 hm²^[29]。但是,它们大多有意或无意地被留在农业土壤中,并在自然条件下逐步分解成微型薄膜^[30-31]。PVC是我国农田种植中使用最广泛的地膜材质^[32]。本实验研究对象为农田土壤,该地区水稻育苗和培植过程中大量使用地膜,使得土壤中MPs含量相对较高,并且成分以PVC为主。农田中残留的大量塑料对土壤的性质造成了负面影响^[33],其对农业土壤中微塑料污染的贡献率需进一步探讨。

土样经过浮选后,用体视显微镜对MPs进行观察,可观察到薄膜、颗粒、碎片、纤维4种形态(图4)。SEM扫描结果显示,所有类型的MPs表面粗糙,有细小的裂纹和碎片。研究表明,老化后的MPs会吸附更多的有机物和重金属^[34-35]。因此,本研究进一步探讨了微塑料对土壤有机物和重金属迁移的影响。

2.2 MP_s对稻谷中BPA污染水平的贡献

BPA是PC的合成原料,研究表明PC会向环境中

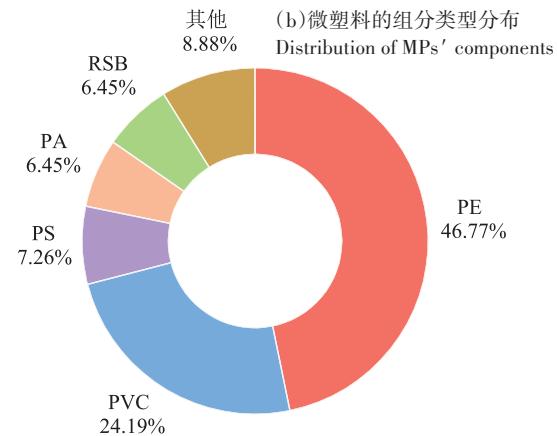
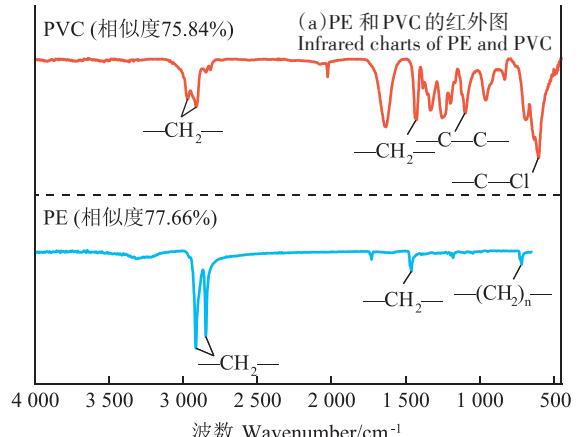


图3 土壤中微塑料的组分类型

Figure 3 The components of MPs in soils

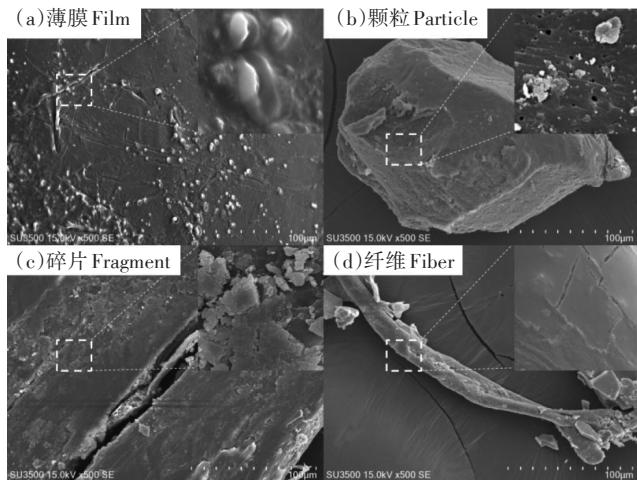


图4 微塑料形态的显微镜及SEM表征

Figure 4 Morphology of MPs characterized by microscopy and SEM

释放BPA^[36]。BPA虽然不是PET的合成材料,但是在加工过程中会使用BPA作为添加剂,因此PET产品使用过程中也会释放BPA^[37]。本研究区域内PC的含

量很低,远小于PET的含量,但二者均可向土壤中释放BPA。目前我国尚未针对食品中BPA含量发布相关规定,《土壤环境质量农用地土壤污染风险管控标准(试行)》(GB 15618—2018)规定,食品接触材料中BPA迁移限量为 $0.6 \text{ mg} \cdot \text{kg}^{-1}$ 。本研究中稻谷样品的BPA含量较低,为 $1.85\text{--}3.34 \mu\text{g} \cdot \text{kg}^{-1}$ (图5),均未超过相关BPA含量标准。相较于未解聚土壤,解聚土壤中BPA与稻谷中的BPA含量呈现出显著的正相关关系($P=0.005$)。解聚土壤中的BPA含量明显提高,表明随着MPs老化程度加重以及环境变化,BPA具有二次释放的风险。因此,MPs土壤污染长期存在,其释放的BPA长期积累有可能通过食物链的传递对人体健康造成潜在威胁。

2.3 MPs对Cd污染水平的贡献

MPs在环境的物理或化学作用下会发生老化,除了表面产生细小裂缝外,其表面官能团也发生改变,这些因素共同导致了老化MPs对重金属吸附量的增加^[38]。研究表明,土壤中MPs与重金属含量高度相关,如Zhou^[39]等通过MPs去除前后对比研究发现,土壤中存在MPs时Cd的含量高于MPs去除后土壤中Cd的含量^[39]。吸附在MPs上的重金属(如Cd)主要以离子形式存在,而该形态具有很高的生物可利用性^[40]。

MPs对金属离子的吸附受pH的影响,在偏酸性环境中吸附量会减少^[41],而水稻根际土壤的pH比其他区域低^[42],这将会导致随着土壤环境的动态变化,非根际土壤中MPs对Cd的富集。当MPs迁移到水稻根部附近时可能释放其吸附的重金属,使水稻暴露在高于生物有效浓度的重金属中,导致更多的Cd被水稻吸收并蓄积在稻谷中。如图6所示,土壤中Cd的

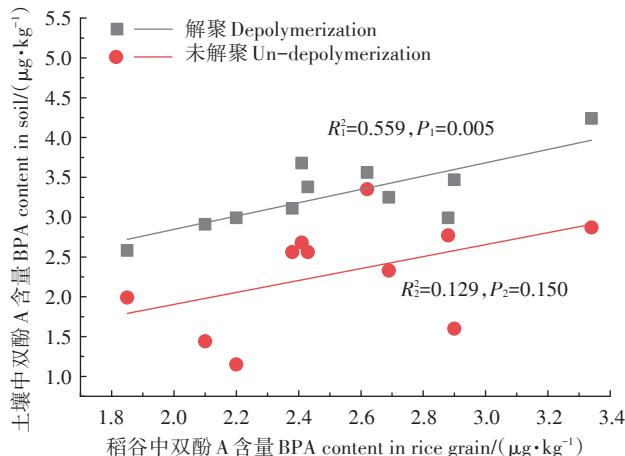


图5 稻谷与土壤中BPA含量的相关性

Figure 5 Correlation between BPA content in rice grains and soils

含量与MPs丰度表现出显著的正相关关系($P<0.001$),稻谷中Cd的含量与土壤中Cd的含量也呈线性正相关($P<0.001$),即随着土壤中MPs的增加,土壤和稻谷中的重金属Cd含量均增加。另外,尺寸较小的MPs可以进入作物中^[43],为重金属在作物中的蓄积起到载体作用,MPs与重金属同时存在条件下还可能会产生机理更复杂的联合毒性效应^[44-45]。因此,微塑料污染长期存在,且持续老化,将促进重金属对农作物的毒性效应,从而可能通过食物链的传递对人体健康造成潜在威胁。

另外,在研究土壤中微塑料与其他环境污染物的相关关系时,目前采用的方法均为直接测定土壤中其他污染物的含量,而不是直接测定微塑料上吸附的污染物含量^[39],这在一定程度上降低了该相关性结果的可靠性。究其原因,目前土壤微塑料的筛分主要以浮选消解分离方式为主^[20],反应比较剧烈,有可能使微塑料上吸附的污染物脱落。在微塑料提取的过程中,其他污染物也可能与土壤中的有机质结合进而从溶液中过滤出去。土壤中微塑料的筛分提取方法还需要进一步优化,不同污染物在微塑料-土壤界面的分配问题也值得进一步探讨。

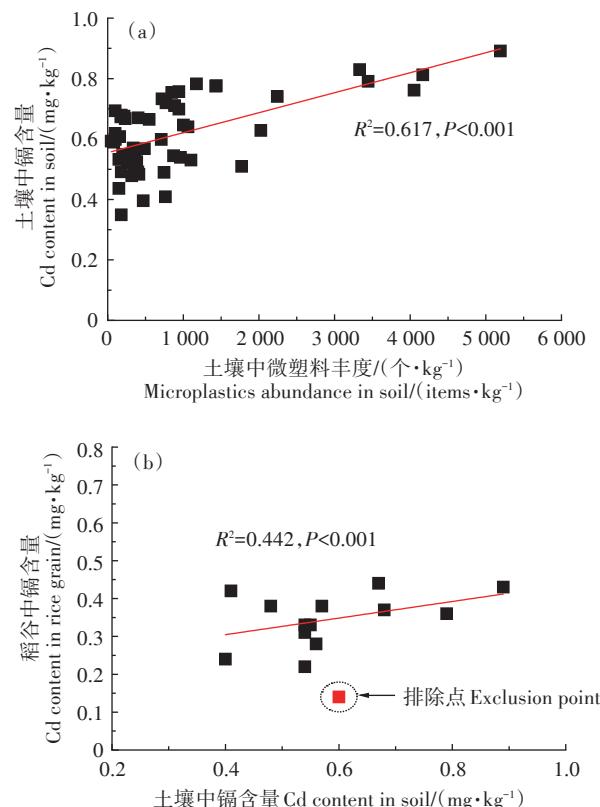


图6 微塑料丰度与镉含量相关性分析

Figure 6 Correlation between MP abundance and Cd content

3 结论

(1)湘潭地区水稻田土壤中微塑料丰度范围为48~1 771个·kg⁻¹,成分以PE和PVC为主。

(2)稻谷中的BPA含量与解聚土壤中BPA含量呈现出显著的正相关关系,微塑料的存在也促进了土壤中的Cd向稻谷中迁移。

(3)土壤中微塑料具有促进污染物向食物链迁移进而对人体健康造成威胁的潜在风险。

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