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土壤微塑料与重金属、持久性有机污染物和抗生素作用影响因素综述

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摘　要：微塑料是难以降解的污染物，可作为众多污染物的载体，在土壤中极易与其他污染物发生复合效应，对土壤产生不利影响。本文系统分析了聚乙烯(PE)、聚氯乙烯(PVC)、聚对苯二甲酸乙二醇酯(PET)、聚丙烯(PP)、聚苯乙烯(PS)和聚酰胺(PA)六种微塑料与重金属、持久性有机污染物和抗生素在土壤中的相互作用及其影响因素。在土壤介质中，微塑料与重金属的作用受微塑料比表面积、老化程度、极性以及土壤中 H^+、低分子酸的影响；微塑料与持久性污染物的作用受微塑料疏水性、橡胶域丰度、污染物极性以及土壤有机质的影响；微塑料与抗生素的作用受微塑料比表面积、极性以及土壤中的腐殖质等物质的影响。研究结果将为阐明微塑料在土壤环境中与其他污染物的相互作用影响因素提供理论支撑。

关键词：土壤；微塑料；重金属；持久性有机污染物；抗生素

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Key influencing factors for interactions between microplastics and heavy metals, persistent organic pollutants, and antibiotics in soil

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Abstract: Microplastics are difficult to degrade and could carry many kinds of pollutants. Furthermore, microplastics could compound with other pollutants in soil and exert adverse effects on the soil environment. Here, we systematically study the interaction of microplastics, including polyethylene (PE), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), and polyamide (PA), with heavy metals, persistent organic pollutants (POPs), and antibiotics in soil and its influencing factors. In the soil environment, the interaction mechanism between microplastics and heavy metals is mainly affected by the specific surface area, aging degree, and polarity of microplastics. The H^+ and low molecular acid in the soil would also affect the interaction process. The interaction mechanism between microplastics and POPs is mainly influenced by the hydrophobicity of microplastics, abundance of rubber domain, and polarity of pollutants or soil organic matter. The interaction between microplastics and antibiotics is mainly affected by specific surface area, polarity of microplastics, and humus in the soil environment. The results of this study will provide theoretical support for clarifying the influencing factors of the interaction mechanism between microplastics and other pollutants in the soil environment.

Keywords: soil; microplastics; heavy metals; persistent organic pollutants; antibiotics

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塑料制品在人们生活中无处不在。据报道2019年全球的塑料产量达3.68亿t[1],预计至2050年将会达到330亿t[2]。塑料制品通过自然降解过程会形成微塑料(Microplastics),其一般定义为直径<5 mm的塑料颗粒[3]。除了塑料制品的降解,微塑料也被广泛应用在个人护理用品中,目前在大气、淡水、海洋和土壤中均发现了微塑料的存在[4-5]。2018年的一项研究表明,陆地上的微塑料可能是海洋中的4~23倍[6]。微塑料进入土壤的途径主要有施用有机肥、使用地膜、大气沉降、污水灌溉等[7-9](图1)。

由于土壤中光线较差,氧气含量较少,微塑料在土壤中的降解速度较为缓慢,HORTON等[2]预计微塑料会在土壤中存在超过100年。微塑料会通过创造水分的运输通道破坏土壤结构的完整性、改变土壤的理化性质,如降低土壤的持水能力[10-11],进而影响土壤容重和肥力,对土壤中的生物产生不利影响[6,8,12-13]。

以"微塑料"为关键词对中国知网和Web of Science的数据进行分析,得到共现网络分析图(图2)。目前关于微塑料的研究更多集中在水环境,土壤环境的研究热度相对较低[5,14-15]。由于土壤有机质、土壤质地和团聚体结构相互影响,土壤环境中微塑料分离和检测难度较大[9,16]。

重金属、持久性有机污染物(POPs)和抗生素类污染物是目前土壤中较为常见的污染物[17-20]。相关研究表明,微塑料作为这些污染物在环境中迁移的载

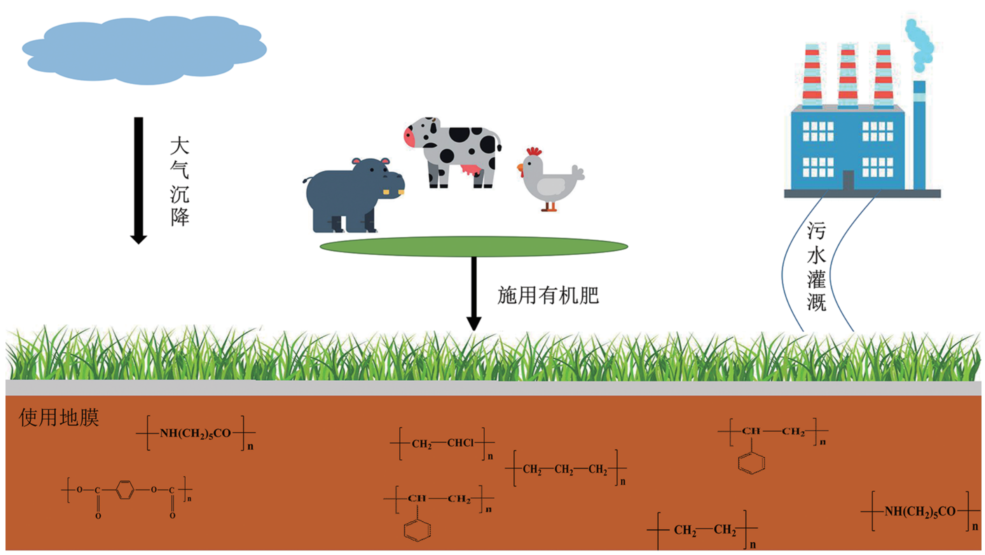

图1 土壤中微塑料来源

Figure 1 Source of microplastics in soil

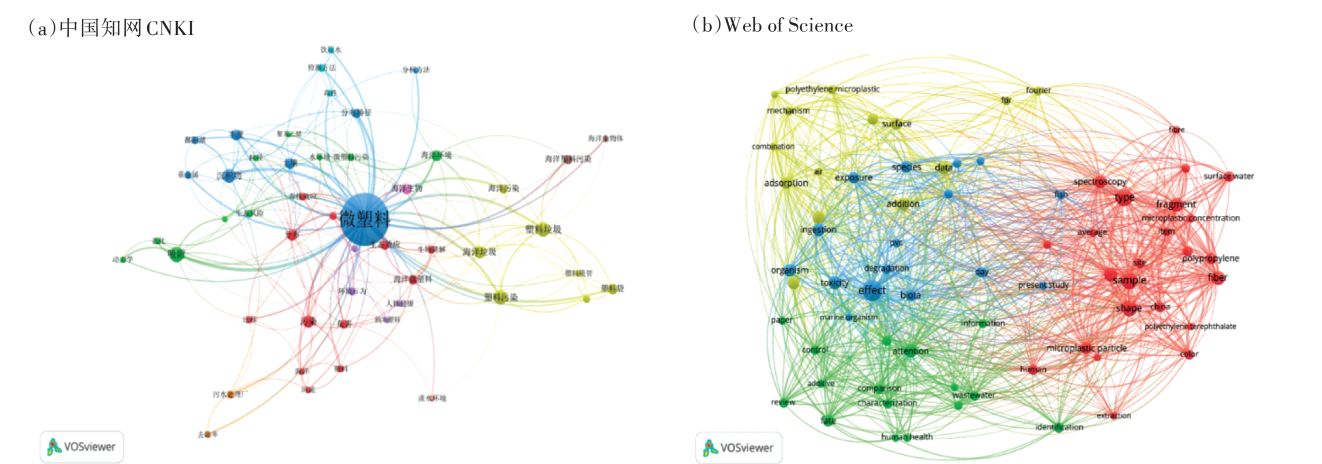

图2 以微塑料为关键词的共现网络分析图

Figure 2 Co-occurrence network analysis diagrams with microplastic as the keyword

体，能够富集疏水性有机物，如多环芳烃（PAHs）、重金属以及多种抗生素[12，21-23]。被微塑料吸附的污染物在微塑料降解过程中会再次释放到土壤环境[24]，对土壤造成持续危害。土壤对人类生存和发展至关重要，探究微塑料与污染物的界面吸附作用不仅可为土壤微塑料污染的风险评估提供理论依据，而且为后续关于土壤中微塑料与其他污染物相互作用的研究提供重要参考。若要揭示微塑料与其他污染物在土壤中的作用机理，首先需要分析和研究微塑料的结构及特性。

1 不同微塑料结构

环境中较为常见的微塑料主要有聚乙烯（PE）、聚氯乙烯（PVC）、聚对苯二甲酸乙二醇酯（PET）、聚丙烯（PP）、聚苯乙烯（PS）和聚酰胺（PA）等，它们通常具有不同的颜色、质地和尺寸（图3），见表1。

微塑料与污染物的分子间存在多种相互作用（图4）。PE、PP和PET为非极性塑料，其中PE和PP表面仅由C—C键和C—H键组成；PVC和PS因含有氯离子或苯环而发生极性的改变，PA具有酰胺基，为极性聚合物[25]。PS微塑料可通过π-π键与其他芳香族有机化合物发生相互作用[28]。PVC表面存在的卤素原子易与苯环的π电子（作为电子给体）产生卤素键[29]。PA存在由C—O键和N—H键组成的酰胺基，易通过氢键的形式与污染物形成吸附，使得PA的吸附能力远大于其他材质微塑料[30]；PA更具亲水性，因此对亲水性有机化合物（如抗生素）具有更高的吸附亲和力[28]。

图中标尺均为1 mm

All scale bars are 1 mm

图3 从加拿大安大略湖支流、海滩和海滩沉积物样品中鉴定出的微塑料实物图[25]

Figure 3 Samples example of microplastics identified in lake bottom sediment from tributaries, beaches and the nearshore of Lake Ontario[25]

2 土壤中微塑料与重金属相互作用影响因素

土壤中重金属主要包括汞、铬、铅、锌、铜、钴和镍

表1 常见的微塑料及其特性[11，26-27]

Table 1 Characteristics of the common microplastics[11，26-27]

微塑料种类 Microplastic type	结构式 Structural formula	密度 Density/（g·cm⁻³）	结晶度 Crystallinity	玻璃化温度 Glass transition temperature/℃
高密度PE	$[CH_2—CH_2]_n$	0.93~0.97	70%~95%	-110
PP	$[CH_2—CH_2—CH_2]_n$	0.85~0.95	50%~80%	-49~-20
PS	$[CH(C_6H_5)—CH_2]_n$	1.04~1.11	低	90
PET	$[O—C(=O)—C_6H_4—C(=O)—O]_n$	1.37~1.45	70%~80%	73~78
PVC	$[CH_2—CHCl]_n$	1.16~1.58	高	60~100
PA	$[NH(CH_2)_5CO]_n$	1.08	35%~45%	-60

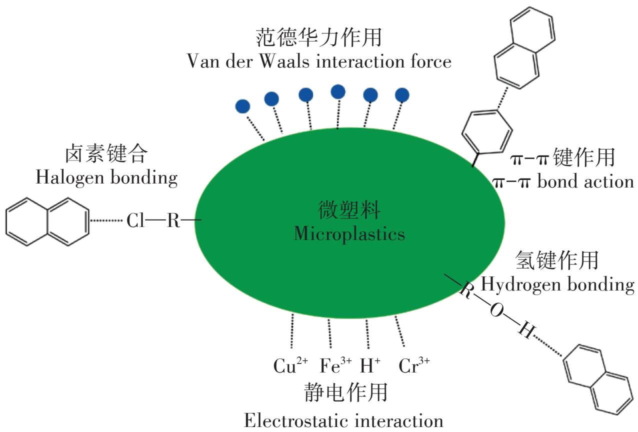

图4 微塑料与污染物之间相互作用的主要机理

Figure 4 Main mechanism of interaction between microplastics and pollutants

等，主要来自岩石、矿物质和人类活动等[31]。重金属在城市和农村中普遍存在[32]，极有可能与微塑料同时出现在土壤中[33]。相关研究表明，长期种植蔬菜的土壤中存在微塑料富集重金属的现象[34]。部分微塑料在热力学上呈亚稳态，其降解时可能会加速微塑料所吸附重金属的迁移过程。土壤中微塑料对重金属的吸附作用主要受两方面影响。

（1）微塑料的影响。微塑料自身的比表面积和相关基团会对吸附产生明显影响，YANG等[35]的研究表明，PA所特有的酰胺基易受环境因素（如紫外线、温度）的影响，且PA本身具有较大的比表面积，其对Cu^{2+}的吸附明显高于其他微塑料（PE、PS、PET、PVC）。塑料自身的老化也会影响土壤中微塑料对重金属的吸附，如紫外照射可使微塑料表面更易负载负电荷[36]，进而吸附更多重金属。与原始微塑料相比，污泥中的老化微塑料对重金属具有更强的吸附能力，因为老化后微塑料更为粗糙和多孔[12]（图5），微塑料的老化也会导致其变成更小颗粒，比表面积增大。老化后的微塑料结构会发生系列变化，包括聚合物分子链断裂、歧化以及表面含氧官能团（如酯基团、酮基团等）增加[37-40]。

微塑料的极性也会影响其对重金属的吸附过程，极性较小的微塑料更易带负电，会与带正电的重金属离子通过静电作用相互结合[41]。相关研究也表明，微塑料在接触水的过程中会在表面产生生物膜并吸附重金属[42]。

（2）土壤的影响。在吸附实验中，PE微塑料表现出与土壤相似的吸附特性，其吸附容量与土壤颗粒相近，但在两者混合后，土壤与微塑料会对重金属发生竞争吸附，土壤所吸附的重金属量是微塑料吸附量的5~10倍[43]。相较之下，微塑料在土壤中对重金属的吸附水平则较低，推测因为H^+会与重金属离子发生竞争吸附，进而占据微塑料表面的吸附位点[44-45]（图6）。

土壤中的低分子有机酸（如苹果酸、柠檬酸）会与重金属络合[31,35,46]，柠檬酸还会影响微塑料吸附位点的数量，高浓度柠檬酸会增加微塑料的吸附位点，进而影响微塑料对重金属的吸附。同时，土壤中的金属离子会对微塑料产生竞争吸附，如土壤中Ca^{2+}和Mg^{2+}的存在会抑制微塑料对Cu^{2+}的吸附[35]。

3 土壤中微塑料与持久性有机污染物相互作用影响因素

持久性有机污染物具有难降解、可长距离迁移等特点，易在环境中持久存在，主要包含杀虫剂、二噁英（PCDDs）和呋喃（PCDFs）[47]。土壤中较为典型的POPs包括多环芳烃（PAHs）、多氯联苯（PCBs）、农药

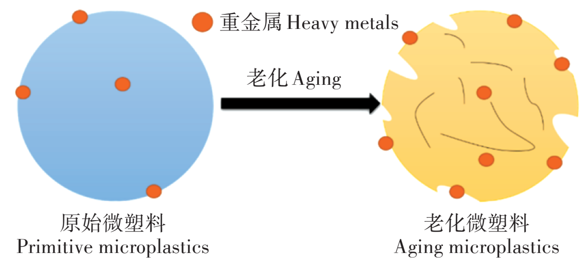

图5 微塑料老化前后对重金属的吸附变化示意图

Figure 5 Adsorption changes of heavy metals on microplastics before and after aging

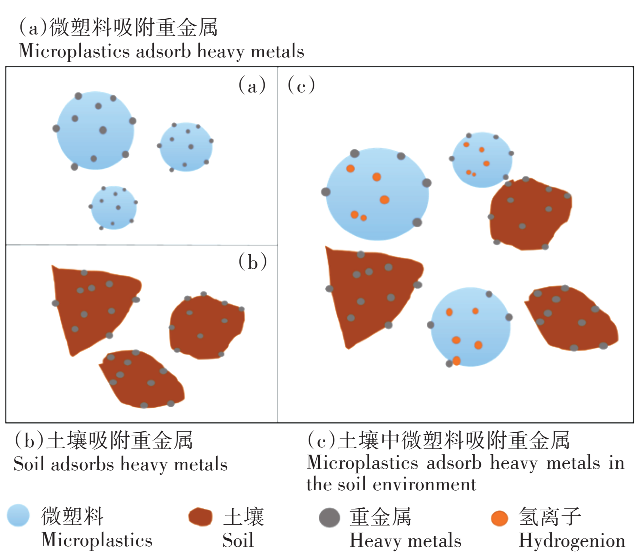

图6 吸附过程示意图

Figure 6 Adsorption process diagram

和硝基芳香化合物等[48]。在土壤中，微塑料对POPs的吸附主要受以下两个方面影响。

（1）微塑料的影响。微塑料自身具有疏水性，易吸附疏水性有机污染物（如PAHs、PCBs）[49-50]。微塑料橡胶域的丰度也会影响微塑料的吸附行为[44]。当温度高于其玻璃化转变温度（Tg）时，微塑料聚合物会从玻璃态变为橡胶态。玻璃态聚合物结构致密，分子间几乎没有空隙，而橡胶态聚合物分子间空隙较大[51]。PE微塑料的Tg值为−110 ℃，PET和PVC微塑料的Tg值较高，分别为73~78 ℃和81 ℃；在实验室（25 ℃）条件下，PE微塑料具有最丰富的橡胶域[52]。相较于玻璃结构的微塑料PP、PS、PVC，橡胶态的PE微塑料能吸附更多的有机污染物[26]。SEIDENSTICKER等[53]的研究表明，微塑料对有机污染物的吸附与微塑料的极性相关，PE微塑料对非极性疏水性有机物菲的吸附明显强于极性物质丙酸。相关研究表明，PS微塑料对非极性有机污染物芘以及弱极性污染物2，2′，4，4′-四溴二苯醚（BDE47）在土柱中的迁移有明显的促进作用，但对极性有机污染物双酚A（BPA）、双酚F（BPF）、4-壬基酚（4-NP）的迁移几乎没有影响，这与非极性污染物在PS微塑料上的解吸滞后有关[54]。

（2）土壤的影响。土壤中微塑料对持续性有机污染物的吸附量取决于土壤环境的温度、pH值和盐度[55]。例如，Na^+的存在会降低农药在土壤中的溶解度，从而促进农药在微塑料上的吸附[55]。土壤中黏土矿物、金属氧化物及氢氧化物、腐殖质、微生物等小颗粒物质会通过范德华力、氢键、离子交换、电荷转移、配体交换和阳离子桥联等作用不同程度地结合有机污染物，与微塑料对有机污染物产生竞争吸附[56-58]。HÜFFER等[59]的研究表明，PE微塑料与土壤有机质相比，对有机污染物的吸附较差，但PE微塑料会对土壤产生稀释效应，导致土壤对有机污染物阿特拉津吸附量减少，加速阿特拉津在土壤中的迁移。由于土壤环境的复杂性，微塑料与持久性有机污染物的相互作用影响因素还需进一步探索。

4 土壤中微塑料与抗生素相互作用影响因素

抗生素目前被广泛应用于养殖业，其主要包含磺胺类、四环素类、氟喹诺酮类、大环内酯类、β-内酰胺类等[60]。农田中可同时检测出多种抗生素，随着土层深度的增加，部分抗生素（如四环素和氯四环素）的平均含量随之增加[61]。关于微塑料与抗生素在土壤中相互作用的研究较少，相关研究表明，微塑料会一定程度抑制抗生素的降解，加速部分抗生素（如土霉素）的迁移[62]（图7），进而对土壤环境造成严重危害[24]。在土壤中，微塑料对抗生素的吸附主要受以下两个方面影响。

（1）微塑料的影响。微塑料为抗生素提供吸附位点，随着微塑料颗粒的增大，比表面积减小，其对抗生素的吸附量逐渐降低[63]。极性微塑料对极性抗生素的吸附性更强[64]，非极性微塑料PE对非极性抗生素的吸附能力强于极性微塑料PS[65]。SHEN等[63]的研究表明，微塑料老化程度对微塑料吸附抗生素的影响并不大，推测微塑料与抗生素之间氢键为主要作用力。LI等[21]的研究也表明，相较于其他四种微塑料（PS、PP、PE、PVC），PA对抗生素的吸附更强，这可能与PA表面发达的孔隙结构有关。

（2）土壤的影响。土壤中有机质和无机颗粒物表面主要通过氢键、范德华力、色散力和诱导力等相互作用，也可以通过阳离子交换、静电、键桥、配位或络合等多种作用与微塑料竞争吸附抗生素[60]。如土壤黏粒、有机质和氧化铁都会通过阳离子交换作用吸附抗生素[60]。土壤中的腐殖质不仅会占据微塑料的吸附位点，还会与抗生素（如四环素）产生静电排斥，影响微塑料对抗生素的吸附[60,63,66]。

5 结论与展望

在土壤介质中，微塑料与重金属、持久性有机污染物、抗生素的相互作用受多种因素的影响。微塑料的结构性质，如比表面积、极性以及表面官能团等都会对微塑料与这几类污染物的作用产生影响。比表面积越大的微塑料能吸附更多的污染物；相较于非极

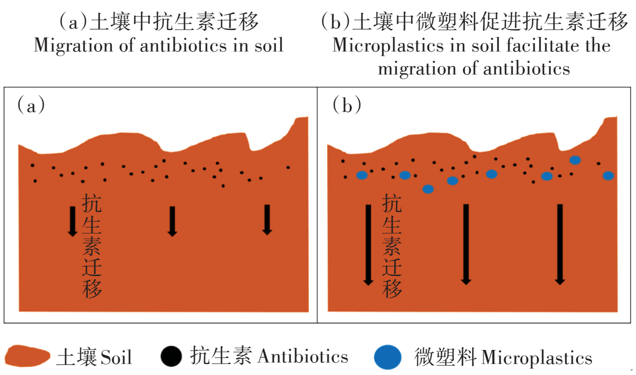

图7 微塑料加速抗生素在土壤中迁移模型

Figure 7 Model of microplastics accelerating antibiotic migration in soil

性微塑料,极性微塑料可以吸附更多的重金属、极性有机污染物;老化的微塑料结构会发生改变,可以吸附更多的重金属,但对微塑料与抗生素之间的相互作用影响不大;微塑料的疏水性有利于微塑料吸附更多的疏水性有机污染物,橡胶域丰度越大的微塑料可以吸附更多的有机污染物。土壤中的物质也会对污染物相互作用产生影响,如土壤中的H^+会与重金属在微塑料表面发生竞争吸附,小分子有机酸会与重金属形成络合物;土壤中的有机质会与持久性有机污染物发生竞争吸附;土壤黏粒、有机质、腐殖质会与抗生素在微塑料表面发生竞争吸附。

微塑料给土壤系统带来的负面影响已逐渐引起关注,但对微塑料在土壤环境中与其他污染物的相互作用及影响机制的研究较少。未来关于微塑料与其他污染物相互作用的研究可以从以下几个方面着手:

(1)制定土壤中微塑料的分离标准和检测方法。微塑料广泛存在于土壤中,土壤环境微塑料总量已超过水体环境微塑料。由于土壤环境较为复杂,目前分离和检测土壤中微塑料的方法有限,导致研究进展缓慢。学者们在研究时会采用不同的方法,不利于后续研究。

(2)探究微塑料对污染物在土壤中迁移的影响。土壤中的有机质及无机颗粒物会对污染物产生吸附,微塑料对土壤产生的稀释作用在一定程度上减少了土壤对部分持续性有机污染物及抗生素的吸附,进而加速污染物在土壤中的迁移,对土壤生态系统造成严重危害,但目前国内外相关研究较少。

(3)探究微塑料吸附污染物在土壤中的复合效应。微塑料对土壤生态系统的影响已经成为研究热点。微塑料不仅对土壤的理化性质造成影响,而且危害土壤中的动植物健康。微塑料在吸附重金属、抗生素、持久性污染物等物质后还会产生复合效应,如抑制污染物的降解,降低土壤中生物多样性,但对这方面的研究甚少。

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