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生物滞留池中多环芳烃去除研究进展

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摘要:对多环芳烃主要去除机理及相关影响因素进行详尽综述。介绍多环芳烃来源、转移与源强,综述生物滞留池中土壤介质吸附、生物降解、植物吸收以及根际协助去除多环芳烃的机制以及影响去除效果的主要因素,包括含碳量、有机物组成、环境因子等的研究进展,指出了有待进一步探讨的问题。

关键词:生物滞留池;雨水径流;多环芳烃;去除机理;影响因素;研究进展
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Advance on the removal efficiency of polycyclic aromatic hydrocarbons in bioretention cells

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Abstract: There are a large number of persistent organic pollutants (polycyclic aromatic hydrocarbons) in the stormwater runoff, and many researchers have studied the removal of polycyclic aromatic hydrocarbons in bioretention cells. However, as the construction of the China's sponge city has just started, the relevant research has not yet been reported. Herein, the authors present a review of the main removal mechanism of polycyclic aromatic hydrocarbons and the influencing factors in bioretention cells. Firstly, the origin, transfer and source of polycyclic aromatic hydrocarbons are briefly introduced. Secondly, main removal mechanisms in the rhizosphere are elaborated including the soil media adsorption, degradation, plant uptake and stimulation of microbiological activity. Besides, main factors affecting the removal of the polycyclic aromatic hydrocarbons, such as the content of carbon, environmental factors and the composition of organic matter are concluded as well. Finally, current gaps for future research are discussed. In this paper, the research progress on the removal of polycyclic aromatic hydrocarbons in bioretention cells abroad can provide scientific support for the optimization of bioretention cells and the efficiency improvement of persistent organic pollutants in our country.

Key words: bioretention cell; stormwater runoff; polycyclic aromatic hydrocarbons; removal mechanism; influencing factors; research progress

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生物滞留池也叫雨水花园,是利用植物、土壤以及微生物共同作用达到削减洪峰流量以及净化雨水目标的浅凹绿地。生物滞留池具有占地面积小,安装、维护成本低,美化环境等优点[1-3],是一种被广泛采用的低影响开发设计(LID)暴雨控制措施[4]。常见的生物滞留系统类型有雨水花园、高位花坛、绿色屋顶、树池等。大量研究表明生物滞留池在处理雨水径流污染物方面效果显著[5-13]。多环芳烃(PAHs)是城市地表雨水径流中常见的污染物之一,其作为一种持久性有机污染物,具有高毒、潜在致癌等特点[14]。近 20 年来,国外在生物滞留池对 PAHs 的去除效率、去除机理、影响因素等方面取得了较为丰硕的研究成果[15-18],相比之下,由于我国海绵城市建设刚刚起步,相关的低影响开发设计研究较少,针对有毒有机污染物 PAHs 的研究更是少之又少,而我国城市道路和土壤受 PAHs 污染又很严重[16-19],因此对国外有关生物滞留池中 PAHs 的去除以及相关影响因素的研究成果进行归纳总结非常必要,可为我国海绵城市中相关设施的设计提供理论支撑和科学依据。

1　PAHs 来源、转移与源强研究

1.1　来源、转移

PAHs 主要来源于有机物不完全燃烧、交通排放和石油泄漏、道路封闭涂层等方面[20-22]。一年四季道路灰尘中 PAHs 总量有所差异,冬季最高,春季最低[23]。PAHs 的来源可以通过检验进水水样中低分子质量(2 ~3 个环)PAHs 和高分子质量(4 ~6 个环)PAHs 的质量浓度来确定,低分子质量 PAHs 质量浓度高说明 PAHs 主要来源于原油和精炼油的释放[24],而高分子质量 PAHs 占主导地位则表明 PAHs 主要来源于有机物燃烧过程[24-25],比如 Stein 等[26]提出当荧蒽/芘的比值在 0.9 ~1.2,且菲/蒽比值低于 21 时,PAHs 主要来源于燃烧过程,与 Maher 等[27]、Gschwend 等[28]得出的结论大致相同。这种比值方法最初是建立在沉积物基础上的,但是现已应用于雨水径流研究中[29]。另外一种初步判断 PAHs 来源的方法是通过 PAHs 的同分异构体即蒽和菲、芴和芘、苯并蒽和的浓度比值来确定,张录龙等[30]运用同分异构浓度比值法,初步判断 4 环多环芳烃、5 环多环芳烃来源于燃烧源,萘来源于燃油泄漏。

随着城市化进程的不断提高,不透水区域面积逐渐增大,透水区域面积逐渐减少。降雨期间,径流将不透水表面(比如屋顶、街道等)大量的 PAHs 带入水体,破坏水生生态系统,危害人类健康[31-32];还有部分 PAHs 则下渗进入透水表面(土壤),造成非点源污染[33]。

1.2　源强

目前,对于雨水径流中多环芳烃的监测主要是现场调查和实验室模拟[7-8,29,34-36],谢继锋等[37]监测比较了 4 种下垫面中 PAHs 质量浓度,其中草地的 PAHs 浓度最低,大约是 5 μg/L;Bardin 等[38]、Lundy 等[39]监测商业区和工业区 PAHs 质量浓度范围是 1.7×10^{-3} ~1.5 mg/L;而道路中的多环芳烃浓度相对低一点,范围是 0.03 ~6 μg/L[39-40];James 等[41]监测停车场的 PAHs 浓度范围是 11 ~191 μg/L,而煤焦油密封停车场的 PAHs 浓度较高,可高达 5 890 μg/L[21,42]。除此以外,也有不少学者采用模型估算雨水径流中 PAHs 的含量。Murakami 等[43]建立了冲刷模型来模拟屋顶和道路径流的颗粒态 PAHs,但是这个模型需要将初始颗粒上的 PAHs 量作为已知输入,这也大大限制了模型的应用;Zhang 等[44-45]利用北京城市道路中现场数据,建立了城市道路累积径流中悬浮物和 PAHs 的相关关系方程,并进一步建立分析模型,同时考虑道路径流中悬浮物堆积和冲刷过程来估算 PAHs 负荷,但是模型颗粒相中 PAHs 的平衡浓度需要是已知的,这在实践中很少有案例。上述提到的都是单场降雨模拟,而 Zheng 等[19]利用变化的时间步长建立了持续性的模型来模拟悬浮物和 PAHs 在不透水的城市表面的堆积-冲刷循环。

2　生物滞留池中 PAHs 去除机理研究

生物滞留池可有效捕获雨水径流中烃类污染物,PAHs 去除率一般比较高,有的甚至可以超过 85%[7-8,29,35-36]。烃的去除机理主要包括土壤介质吸附、生物降解、植物吸收以及植物根际协助去除等方面[7-8,29,34-36],挥发可能也是 PAHs 从暴雨中损失的一种机理,但是通过挥发损失的 PAHs 似乎只是很少的一部分[35-36]。下面详细介绍 PAHs 去除的主要机理。

2.1　介质吸附

PAHs 土壤和沉积物等介质吸附是 PAHs 去除的主要过程[34-36,46-47]。正辛醇-水分配系数(K_{ow})是判断

有机污染物吸附能力的指示性参数,这个系数越高,表明 PAHs 越易被吸附。研究发现高分子质量的 PAHs 比低分子质量的 PAHs 的正辛醇-水分配系数大,溶解度低,更易被吸附[48-49]。谢继锋等[37]指出径流中 58% ~69% PAHs 是颗粒态的,其中以 5 ~6 环 PAHs 为主,大多数高分子质量的 PAHs 浓度与悬浮物浓度正相关[19,22,29,50-52]。因此,去除进水悬浮物是一种去除进水中颗粒相关污染物很好的方法。PAHs 在介质中的垂向分布存在差异,主要被吸附在表层土,下层土中 PAHs 的量很少[29,33,35,49]。吸附到介质中的这个过程是可逆的,土壤会慢慢解吸,具有再生能力[1-2,29,34-35,53]。

2.2 生物降解

石油烃一般在有氧条件下容易发生降解,尤其是在 pH、温度和营养物等条件适宜,不限制微生物生长的情况下更是如此[54-56]。尽管某些 PAHs 在无氧条件下也会发生生物降解,但是在这种条件下的生物降解速率明显小于有氧条件[55-58]。

生物降解速率快慢主要由两方面因素决定,分别是总细菌数(16SrRNA)和生物降解功能基因(酚氧化酶和萘双加氧酶)。生物滞留介质中总细菌数越大,萘降解速率越快[34-35];生物降解功能基因的量也与污染物降解是正相关的[59]。但是污染物进行生物降解所需的酶并不是特定不变的,比如,萘双加氧酶仅催化多种 PAHs 降解的初始步骤[60]。另外,生物降解速率还与 PAHs 的分子质量相关,Leroy 等[22]对比分析受 3 种 PAHs(菲、芘和苯并芘)为期 2 a 污染的滞留池中土壤 PAHs 残留量,菲、芘和苯并芘分别代表的是 3 环、4 环和 5 环 PAHs,结果表明菲比芘降解更快,芘比苯并芘降解更快。

2.3 植物吸收及根际协助作用

大多数生物滞留池表层都有植物生长[2,22],植物的存在对于去除 PAHs 是有利的[34-35]。Lefevre 等[35]指出种有植物的生物滞留柱中^{14}C-萘去除效率比无植物的生物滞留柱高的多;Siciliano 等[61]发现在种有植物的系统中 PAHs 生物降解的量是无植物的系统中 PAHs 的 2 倍。

PAHs 被植物直接吸收的量较少,在实验室规模的生物滞留池中添加^{14}C 标记的萘,仅有 2% ~23% 的萘被植物吸收[35]。植物除了直接吸收以外,溶解性有机物可以直接通过植物蒸腾在植物的气孔上蒸发去除[62-64]。污染土壤中植物最重要的作用是刺激根际微生物活性[65-67],Jouanneau 等[68]、Sun 等[69]指出菲、芘、苯并芘主要由植物根际固定,很少会被植物吸收转移到地上植物部分。Lefevre 等[35]研究表明种有植物的介质中细菌生物降解萘的速率比无植物介质显著高的多。植物根部分泌氧可以提高土壤中的氧气含量[70],从而促进有氧生物降解。植物释放的分泌物可以作为污染物类似物诱导污染物生物降解,刺激整个细菌群[61,71-72],或提高疏水性有机污染物——萘的生物利用度[73]。

3 PAHs 去除效率研究

尽管关于生物滞留池 PAHs 来源、去除机理方面国内外研究很多,但是相关的去除效果研究不多,表 1 中列举了不同研究背景下基于质量的 PAHs 去除效率,Hong 等[36]在实验室条件下观察到雨水径流中萘的去除率大约是 80% ~95%;Leroy 等[22]添加已知量的 PAHs 到滞留池中,并检测渗出水中的 PAHs 的量,结果显示去除率高达 99%;Lefevre 等[35]在实验室柱装置中进行试验,比较分析有无植物对萘去除效果,结果表明无植物的情况下对萘的去除率为 78%,有植物的情况下去除率是 93%。由表 1 可见 PAHs 在大多数情况下去除效果不错,但是如果是基于浓度的去除率,就没有这么稳定,Diblasi 等[29]发现生物滞留池基于质量的去除率是 87%,而基于浓度的去除率是 31% ~99%,基于浓度的去除没有考虑入渗损失量,且每场降雨的浓度变化大,因此很难精确计算基于浓度的去除效率[75-76]。

表 1 生物滞留池中 PAHs 去除效率

Table 1 Removal efficiency of PAHs in bioretention cells

研究背景	污染物类型	去除率/%	参考文献
实验室	萘(有植物)	93	Lefevre 等[35]
	萘(无植物)	78	Lefevre 等[35]
	萘	大于 90	Hong 等[36]
现场	PAHs	87	Diblasi 等[29]
	PAHs	97	David 等[74]
	PAHs	大于 99	Leroy 等[22]

4 影响生物滞留池中 PAHs 去除的因素

4.1 含碳物质和有机物组成

尽管土壤和沉积物上的矿物颗粒可以将 PAHs 滞留,但是土壤有机物(SOM)似乎才是 PAHs 的主要直

接载体[17],SOM 是由许多含碳物质组成,比如煤和黑炭(BC)。BC 是不同含碳颗粒(烟灰、碳、木炭、焦炭)的集合项,土壤和沉积物中含碳物质是很好的 PAHs 吸附剂[17,77-78],黎舒雯[52]分析径流中 PAHs 与总溶解性碳(TOC)的相关性,R^2 普遍高于 0.7。针对煤和 BC 对 PAHs 的转移以及 PAHs 吸附特性的影响研究较多,Yang 等[47]发现土壤中分子质量小的 PAHs(2 个环)与煤颗粒的相关系数 R 高达 96%,显著水平 $p<0.0001$,而分子质量大一些的 PAHs 更易受 BC 控制,与 BC 相关($R=0.79$,显著水平 $p<0.0001$)。Liu 等[79]调查北京不同地区(城市、偏远平原和山区)PAHs 土壤污染与 BC 之间的关系,结果表明分子质量大 PAHs(4 ~ 6 环)与 BC 的相关性明显强于分子质量小的 PAHs(2 ~ 3 环),但是并未调查土壤中不同含碳物质(CMs)在雨水径流过程中如何影响 PAHs 转移,而 Zheng 等[18]比较 4 种类型 CMs 对 10 种 PAHs 的吸附,基于试验数据利用分配系数的对数值来进行比较,碳对应对数值的中值是 7.2,至少是其他 3 种 CMs 对应对数中值的 1.3 倍,结果表明炭对 PAHs 的吸附能力更强。Zheng 等[17]也指出 PAHs 的富集比 ER(沉积物中 PAHs 浓度与土壤中 PAHs 浓度的比值)与有机碳含量正相关(R^2 可高达 0.9899),采集的 2 种土壤的有机物组成不同,而这 2 种土壤对应的 ER 也不同,设想 ER 也与土壤有机物组成有一定关系,Zheng 等[18]进而分析了不同 CMs 的富集,验证了之前的假设。

4.2　植被类型

植被类型是影响污染物去除能力的重要因素之一。Lefevre 等[35]比较分别种有草原草和三叶草的生物滞留池中 2 种植物对萘的吸收,结果发现草原草对 PAHs 的吸收是三叶草的 3 倍多;Leroy 等[22]比较分析了大型植物(灯芯草、黄菖蒲、蠡草)和普通小草这 4 种植物对受 PAHs 污染土壤的修复作用,结果表明种植有黄菖蒲的试验装置中 PAHs 降解速率最快,去除 50% PAHs 大致需要 90 d。

此外,研究发现种有深根植物的生物降解功能性基因和总细菌数比未种植物或者非常浅根植物(草坪)要多[34],萘矿化速率大。Aprill 等[80]也指出有深根的当地草原上的草比无植被的情况对土壤中 4 种 PAHs 的生物降解速率显著提高,因为植物根系可以为微生物附着提供载体,旺盛的微生物活动提高对 PAHs 降解速率。

4.3　环境因子

环境因子可在一定程度上影响微生物的新陈代谢从而影响对 PAHs 的降解效果,主要的环境因子有温度、土壤 pH、氧气含量、无机盐含量、降水等。

吸附反应通常是放热的,因此吸附的程度随着温度的下降而升高,Hiller 等[81]试验表明温度从 4 ℃升高至 27 ℃,萘、菲、芘吸附量分别下降了 27.3% 、17.0% 和 27.4% ;Margesin 等[82]指出 25 ~ 30℃是土壤中细菌、真菌最适合的生存温度,温度过高或者过低会降低酶活性导致 PAHs 降解能力下降;另外,土壤 pH 会影响代谢酶活性,改变代谢过程中酶促反应速率。

生物滞留系统中 PAHs 的生物降解主要是好氧过程,PAHs 的有氧降解速率比无氧降解速率快[83],氧气的供给为有氧代谢提供了物质条件;PAHs 在沉积物上吸附量随着盐度的增加而增大,El-Sheikh 等[84]证明当 NaCl 浓度升高至 150 mg/L,菲在土壤表面吸附显著升高;Diblasi 等[29]研究分析了城市雨水径流中 PAHs 的去除效率,发现降雨量小,进水 PAHs 浓度高,对 PAHs 去除效果好;Zheng 等[17]对于 3 个降雨强度对 PAHs 富集特性的影响进行了研究,结果表明降雨强度越小,PAHs 富集比越高,有利于 PAHs 去除。

4.4　降解菌种类

微生物降解被认为是 PAHs 在自然界中降解的方式,要提高微生物的降解效果,就要对降解菌进行驯化、培养、富集、分离、鉴定。常见的 PAHs 降解菌包括假单胞菌属、黄杆菌属、诺卡氏菌属、分枝杆菌属等,不同 PAHs 对应的降解菌不同,真菌是 4 环以下 PAHs 的降解菌,研究较多的白腐真菌是常见真菌之一,可以产生诸如木素过氧化物酶等胞外酶和胞内酶,起到降解低环 PAHs 的效果[85],而 5 环及以上的 PAHs,它的降解可通过细菌和真菌的联合作用,比单一菌作用的效果显著,降解速率更快[86]。

4.5　其他

除了上述影响因子之外,初始 PAHs 浓度、土壤颗粒尺寸、土壤年限、吸附质溶解度及接触时间等在不同程度上会影响生物滞留系统中 PAHs 的去除效果。

吸附量随着初始 PAHs 浓度的升高而增大。Ping 等[87]发现初始菲浓度越高,菲的吸附量也相应增加;Awoyemi[88]用不同初始浓度的萘(1.6 mg/L、4.2 mg/L 和 8.2 mg/L)进行试验并证明吸附速率与初始浓度成

正比,相似地,Hall 等[89]发现芘初始浓度从 0.376 mg/L 增加到 1.795 mg/L,它在硅胶上的吸附能力从 0.178 mg/L 增加到 1.296 mg/g。

Wang 等[90]对比了不同粒径介质对 PAHs 的吸附能力,结果表明较小粒径的介质具有更大的表面积,吸附能力更强。相似地,疏水性有机污染物,比如菲,被发现在小颗粒沉积物上累积量大于大颗粒沉积物[91]。

Leroy 等[92]对种有植物的洼地进行了为期 2 a 的 12 场暴雨的进、出水 PAHs 浓度监测,研究表明两种洼地出水的 PAHs 浓度都显著高于进水浓度(种有大型植被洼地的 PAHs 去除率-106%,种有草的洼地 PAHs 去除率-236%),可能是因为土壤受污染年数长(6 a),PAHs 慢慢渗出。因此,表层土壤需要在有规律的时间间隔去除。

吸附质的溶解度显著影响吸附剂的吸附能力,不同种类多环芳烃在水溶液中溶解度不同,水中溶解度低的种类,沉积物中的吸附量会增加。PAHs 的溶解度和分子量之间存在反比关系,Balati 等[93]的试验指出 PAHs 中的萘,芘和菲的溶解度随分子量的增加而呈递减趋势。

当疏水性有机化合物与土壤接触时,一部分通过物理吸附快速吸附到土壤组分(如有机物质,黏土矿物质)上,而剩余部分则需要较长时间达到吸附平衡,Zeledón-Toruño 等[94]使用初始 PAHs 浓度为 100 mg/L 的芘,苯并(k)荧蒽,苯并(a)芘,苯并(g,h,i)苝和芴,在接触时间 2 h 内达到 70% 的去除率,在 24 h 内逐渐达到平衡,芘,苯并(k)荧蒽,苯并(a)芘、苯并(g,h,i)苝的最大去除效率都高于 82%,芴的去除率是 78.4%。

5 有待进一步研究的问题

虽然国外关于生物滞留池中去除多环芳烃的研究相对成熟,但是目前还是存在一些问题有待解决。(a)Leroy 等[92]提出定期更换滞留池表层土壤,这个措施的最佳时间是在冬季,且每 10 a 去除一次,但是更换表层土的实际频率应该依具体情况确定,确定的依据有待深入研究;(b)进行表层土壤置换处理时使用机器将对植物造成伤害,因此需要进一步探究此措施进行之后植物的生长发育情况以及滞留池对污染物滞留能力的改变;(c)低分子质量 PAHs 在土壤中受许多微生物比如假单胞菌[95]或白腐真菌[96]控制,可以自然降解,但是高分子质量的 PAHs 是更具持久性的,不易降解,因此很有必要确定 PAHs 在生物滞留池中最终存在形式,判断是否会造成土壤污染问题;(d)设计参数比如植被密度,对污染物去除影响的相关信息很少,需进一步研究确定;(e)为了更好地模拟 PAHs 的非点源污染,评估环境风险,土壤中主要含碳物质的含量、迁移和吸附能力需进一步探索,因 SOM 组分在降雨初期径流中变化最大,而且典型含碳物质,尤其是黑炭组分在雨水径流的情况下的转移机制需进一步研究。

6 结 论

PAHs 主要来源于有机物不完全燃烧、交通排放和石油泄漏、道路封闭涂层等。在生物滞留池中,PAHs 通过土壤介质吸附、生物降解和植物吸收、挥发等几种方式去除,其中挥发是最不显著的,吸附作用和降解是主要的去除机理。由于雨水径流中悬浮物浓度与 PAHs 正相关,土壤介质通过将悬浮物滞留于沉积物或土壤表层,同时达到将 PAHs 吸附去除的效果,另外,生物滞留池中植物的存在提高了 PAHs 的去除效果,尤其是深根植物。虽然植物直接吸收的量较少,但是植物根际为微生物提供了适宜的生境条件,加速了 PAHs 的生物降解。除此之外,含碳物质的含量、有机物组成以及环境因子也影响生物滞留池中 PAHs 的去除。因此,进行生物滞留池设计时,综合考虑这些影响因素,优化设计生物滞留池,将为有效、稳定去除 PAHs 提供理论指导和科学依据。

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