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铁炭内电解垂直流人工湿地对污水厂尾水深度脱氮效果

郑晓英¹, 朱星¹, 周翔¹, 徐亚东², 王菊², 韦诚², 高雅洁¹, 周橄¹

(1. 河海大学环境学院, 浅水湖泊综合治理与资源开发教育部重点实验室, 南京 210098; 2. 江苏省水资源服务中心, 南京 210029)

摘要: 针对污水厂尾水总氮(TN)含量偏高、微生物可利用碳源低的问题, 构建铁炭内电解垂直流人工湿地(ICIE-VFCW)装置, 研究了ICIE-VFCW对尾水的处理效果, 并采用紫外-可见光光谱(UV-VIS)、凝胶过滤色谱(GFC), 进一步探讨了ICIE-VFCW强化脱氮机制. 结果表明, ICIE-VFCW可提高系统对尾水中COD的去除, 出水COD可稳定在 $30\text{ mg}\cdot\text{L}^{-1}$ 以下, 全年、暖季、寒季COD平均去除率较普通垂直流人工湿地分别可提高10.16%、9.81%、11.22%. 系统出水TN可维持在 $10\text{ mg}\cdot\text{L}^{-1}$ 以下, 全年、暖季、寒季TN平均去除率较普通垂直流人工湿地分别提高13.72%、12.90%、16.17%. 经过人工湿地处理后, 污水中有机物的腐殖度、芳香度及相对分子质量(M_r)均有所下降, 且ICIE-VFCW中 M_r 下降更为明显. 湿地基质掺杂铁炭可促进尾水中大分子有机物转化为小分子, 为微生物提供更多可利用碳源, 从而提高脱氮效率.

关键词: 污水厂尾水; 垂直流人工湿地; 铁炭内电解; 深度脱氮; 相对分子质量分布

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Removal of Nitrogen in Municipal Secondary Effluent by a Vertical Flow Constructed Wetland Associated with Iron-carbon Internal Electrolysis

ZHENG Xiao-ying¹, ZHU Xing¹, ZHOU Xiang¹, XU Ya-dong², WANG Ju², WEI Cheng², GAO Ya-jie¹, ZHOU Gan¹

(1. Key laboratory of Integrated Regulation and Resources Development on Shallow Lakes, Ministry of Education, College of Environment, Hohai University, Nanjing 210098 China; 2. Water Resources Service Center of Jiangsu Province, Nanjing 210029, China)

Abstract: Aiming at the problem of high concentration of total nitrogen (TN) and low available carbon source for microorganisms in municipal secondary effluent, the vertical flow constructed wetland associated with iron-carbon internal electrolysis (ICIE-VFCW) was applied to investigate the removal efficiencies of pollutants in municipal secondary effluent. Moreover, the mechanism for enhanced nitrogen removal was primarily discussed by the applications of UV visible spectrum (UV-VIS) and gel filtration chromatography (GFC). The results showed that the ICIE-VFCW could improve the COD removal efficiencies and the effluent COD of less than $30\text{ mg}\cdot\text{L}^{-1}$ could be stably obtained. The average COD removal efficiencies of the whole year, warm months, and cold months could be increased by 10.16%, 9.81%, 11.22%, respectively, compared to the control group. The effluent TN of the ICIE-VFCW could be maintained below $10\text{ mg}\cdot\text{L}^{-1}$, and the average TN removal efficiencies of the whole year, warm months and cold months could be increased by 13.72%, 12.90%, 16.17%, respectively. Besides, compared to the influent, the humification, aromaticity and average relative molecular weight (M_r) in the effluent obviously decreased, and the M_r decreased more significantly in the ICIE-VFCW. The ICIE-VFCW could promote the conversion of refractory organics in municipal secondary effluent to the small and readily biodegradable molecules, which could enhance the utilization of organic compounds by microorganisms, thus improving the removal efficiency of nitrogen.

Key words: municipal secondary effluent; vertical flow constructed wetland; iron-carbon internal electrolysis; deep denitrification; distribution of molecular weight

目前,我国大多数污水处理厂以二级处理为主,即使污水厂尾水达到《城镇污水处理厂污染物排放标准》(GB 18918-2002)一级A/B标准,仍属于劣V类水,对于受纳水体而言还是重要的污染源^[1].同时考虑到国际公认的水体发生富营养化的临界值 $[\text{TN } 0.2\text{ mg}\cdot\text{L}^{-1}, \text{TP } 0.2\text{ mg}\cdot\text{L}^{-1}]$ ^[2],二级出水依旧是造成受纳水体富营养化的主要原因之一^[3].污水处理厂提标改造,尾水进行深度处理,是未来的发展趋势,北京市、天津市已先后发布了更为严格的城

镇污水处理厂污染物排放标准.

人工湿地是近年来发展起来的一种新型生态处理技术,具有污染物去除效果稳定、投资省、运行成本低等显著优点,是削减二级出水中氮磷、有机

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作者简介: 郑晓英(1976~),女,博士,副教授,主要研究方向为污水处理及其资源化, E-mail: zhxyqq1118@163.com

物等污染物的有效工艺之一^[4~7],为污水处理厂尾水的深度处理开辟了一条新途径.然而,经过污水厂生化处理单元处理后,尾水的碳源低、可生化性差(BOD_5/COD 为 $0.2\sim0.35$, BOD_5/TN 约为 1).尾水中有机碳源主要有富里酸、腐殖酸以及亲水性有机酸、核酸、氨基酸和表面活性剂等有机物^[8],难以被微生物降解、利用.同时,硝酸盐氮是尾水中氮素的主要成分,占 TN 的 80% 左右^[9,10].反硝化脱氮是人工湿地脱氮的主要方式,尾水中可利用碳源不足限制了反硝化作用的进行,影响湿地系统的脱氮效率^[11~13].且冬季低温污染物去除效率低^[14~16],这也是限制人工湿地进一步推广应用的重要原因之一.

铁炭内电解法作为目前国内外研究较为成熟的电化学工艺,已经在制药废水、印染废水、焦化废水等难降解废水的处理上得到了广泛应用.铁炭内电解与其他生化处理工艺相结合,可以改善废水性质、提高废水可生化性、提高废水的处理效果^[17].为此,本研究构建新型铁炭内电解强化脱氮人工湿地(vertical flow constructed wetland associated with iron-carbon internal electrolysis, ICIE-VFCW),通过在湿地基质中掺杂铁炭,形成铁炭内电解与人工湿地耦合工艺,利用铁炭内电解改善尾水可生化性,为湿地微生物提供更多可利用碳源以强化湿地对氮素等污染物的去除,并采用紫外-可见光光谱(UV-VIS)、

凝胶过滤色谱(GFC)等深入分析铁炭内电解人工湿地的强化脱氮机制.本研究通过开发一种高效脱氮的人工湿地处理工艺,以期为人工湿地的进一步推广应用提供技术支持.

1 材料与方法

1.1 人工湿地系统的构建

垂直流人工湿地能够形成良好的好氧/缺氧环境,有利于尾水的深度脱氮,因此选择垂直潜流湿地作为实验装置^[18].实验采用自行设计的下向垂直流人工湿地装置,共2套(分别为对照组和实验组),材料选用有机玻璃,装置采用圆柱体,尺寸为 $\Phi\times H=20\text{ cm}\times65\text{ cm}$,以粗砂和天然砾石作为人工湿地主要基质,种植芦苇作为湿地植物.装置底部设集水区,通过穿孔有机玻璃板集水,集水区高 5 cm .为更加真实地模拟人工湿地,装置四周采用遮光布包裹.人工湿地构造如表1所示.为方便取样分析,沿程设置5个取样口,分别采集粗砂层、第一砾石基质层中部、第一砾石基质层、第二砾石基质层中部、第二砾石基质层的出水.人工湿地装置如图1所示.污水经蠕动泵打入湿地,通过底部的出水口、流量计控制出水,并维持湿地水位恒定,湿地孔隙率为 35% ,设计水力停留时间为 2 d .实验装置于2015年6月构建,经近半个月后,系统生物膜生长成熟,出水稳定.

表1 垂直流人工湿地构造¹⁾
Table 1 Structure of vertical flow constructed wetland

项目	对照组	实验组
湿地植物	芦苇,种植密度为 $20\text{ 株}\cdot\text{m}^{-2}$	芦苇,种植密度为 $20\text{ 株}\cdot\text{m}^{-2}$
粗砂层	粒径 $2\sim4\text{ mm}$,厚 5 cm	粒径 $2\sim4\text{ mm}$,厚 5 cm
第一砾石基质层	粒径 $4\sim8\text{ mm}$,厚 20 cm	粒径 $4\sim8\text{ mm}$,厚 20 cm ,掺杂砾石基质层质量 1% 的铁炭
第二砾石基质层	粒径 $4\sim8\text{ mm}$,厚 20 cm	粒径 $4\sim8\text{ mm}$,厚 20 cm
砾石承托层	粒径 $8\sim16\text{ mm}$,厚 5 cm	粒径 $8\sim16\text{ mm}$,厚 5 cm

1) 铁炭颗粒为 $10\sim30\text{ mm}$ 的块状,由铁和炭高温煅烧而成,铁炭质量比为 $m_{Fe}:m_C=5:1$

1.2 研究方法

实验前通过调研污水厂尾水以确定尾水的水质特征,采用实验室自配水模拟污水厂尾水,具体成分如表2所示.实验期间(2015年7月~2016年6月)进水主要污染物平均浓度为: $COD\ 53.16\text{ mg}\cdot\text{L}^{-1}$ 、 $BOD_5\ 6.73\text{ mg}\cdot\text{L}^{-1}$ 、 $TN\ 16.68\text{ mg}\cdot\text{L}^{-1}$ 、 $NH_4^+-N\ 2.02\text{ mg}\cdot\text{L}^{-1}$ 、 $NO_3^--N\ 12.58\text{ mg}\cdot\text{L}^{-1}$ 、 $TP\ 0.23\text{ mg}\cdot\text{L}^{-1}$.

经过1个月的稳定运行后,采集进出水及沿程各取样口水样,每月4次,采样时间为上午09:00~

表2 模拟污水厂尾水的水质

Table 2 Water quality of the simulated tail water of wastewater treatment plants

成分	浓度 / $\text{mg}\cdot\text{L}^{-1}$	组分	浓度 / $\text{mg}\cdot\text{L}^{-1}$
$C_6H_{12}O_6$	22	KH_2PO_4	1
腐殖酸钠	10	$MgSO_4\cdot7H_2O$	33
海藻酸钠	4	$CaCl_2$	3
牛血清白蛋白	2	$MnSO_4$	3
NH_4Cl	15	$FeSO_4\cdot7H_2O$	2
$NaNO_3$	97	$MnSO_4\cdot4H_2O$	0.4

11:00,并于当日送至实验室进行水质分析.常规水

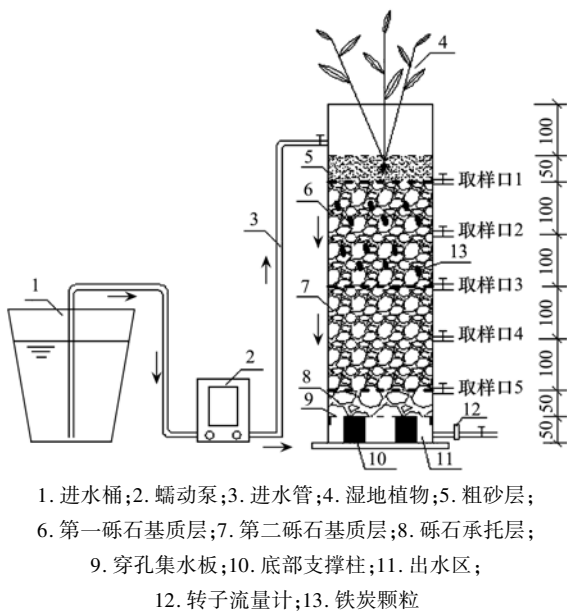


图1 垂直流人工湿地装置及取样点分布
Fig. 1 Vertical flow constructed wetland system and sampling locations

质指标分析方法参考文献[19]. 沿程水样吸光度采用紫外-可见光光谱仪(普析通用T6新世纪)进行测定,扫描波长范围为200~700 nm,扫描步长为2 nm. 以超纯水作基线,去离子水为空白. 各水样有机物相对分子质量分布采样 GFC(凝胶过滤色谱, LC-10ADVP)测定,测定前用0.45 μm 微孔滤膜过滤,进样量为20 μL . 测定条件:TSK4000 型色谱柱,超纯水为流动相,液体动力注入,视差检测器型号 RID-10A,控制器型号 SCL-10AVP,柱温箱型号 CTO-10SVP.

2 结果与讨论

2.1 垂直流人工湿地对尾水中有机物的去除

两组人工湿地对尾水中 COD 的去除效果如图2所示. 从中可知,实验组全年对 COD 的去除效果优于对照组. 采用 SPSS 软件进行统计学分析,两组湿地 COD 去除率存在显著性差异($P < 0.05$). 湿地对 COD 的去除,呈现出季节性变化,随气温的升高而提高. 实验组全年出水 COD 稳定在30 $\text{mg} \cdot \text{L}^{-1}$ 以下,满足地表水Ⅳ类标准(30 $\text{mg} \cdot \text{L}^{-1}$),而对照组在寒季(江苏地区12月到次年2月,平均气温14.7℃)出水 COD 超过30 $\text{mg} \cdot \text{L}^{-1}$. 对照组和实验组 COD 全年平均去除率分别为47.48%、57.64%,暖季(江苏地区3~11月,平均气温26.3℃)平均去除率分别为50.10%、59.91%,寒季平均去除率分别为39.61%、50.83%,实验组全年、暖季、寒季

COD 平均去除率分别高出对照组10.16%、9.81%、11.22%. 可见,基质掺杂铁炭能够提高湿地对 COD 的去除效果,且冬季的提升效果更为明显. 这可能主要是在污水浸渍条件下,掺杂在湿地基质中的铁炭形成原电池,阳极发生电极反应 $\text{Fe} - 2\text{e} = \text{Fe}^{2+}$ ($E_0 = -0.44 \text{ V}$),阴极发生电极反应 $\text{O}_2 + 2\text{H}_2\text{O} + 4\text{e} = 4\text{OH}^-$ ($E_0 = +0.40 \text{ V}$)^[20]. 铁炭内电解过程产生大量活性的 $[\text{H}]$ 和 Fe^{2+} ,使尾水中的复杂有机物发生开环、断链等作用,另外,在活性 Fe^{2+} 被 O_2 氧化成 Fe^{3+} 的过程中,会产生具有强氧化性的 $\cdot\text{OH}$ 、 $\text{O} \cdot$,破坏—CN和C=O键^[21]. 原电池反应过程中产生的活性基团促进了难降解有机物分解为小分子,强化了微生物对尾水中有机物的利用,促进了湿地对 COD 的去除.

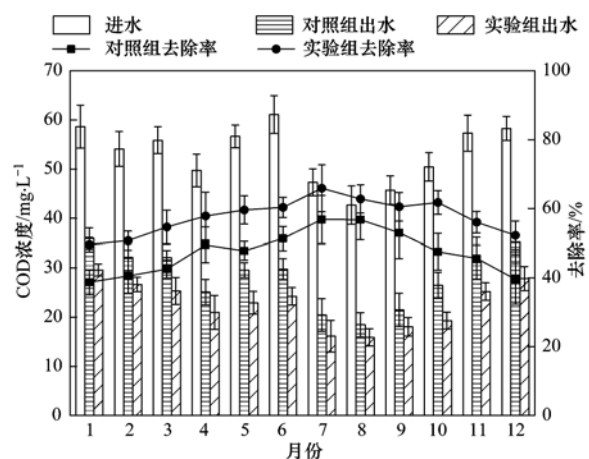


图2 垂直流湿地进出水 COD 浓度及平均去除率

Fig. 2 Concentrations of COD in influent and effluent of vertical flow constructed wetland and average removal efficiencies

两组湿地沿程溶解性有机物(DOC)及其光谱特征如表3所示(10月12日测定结果). 总体来看,DOC及各波长的吸光度沿程下降,且对照组各采样点高于实验组. 实验组采样点3的DOC明显升高,可能是经过第一砾石基质层(铁炭层)后,尾水中部分不溶性有机物转变成了可溶性有机物. a_{300}/a_{400} 可表征污水腐殖度和芳香度,该比值越大,说明尾水中腐殖度和芳香度越小,其难降解污染物含量越少^[22, 23]. 两组湿地出水中 a_{300}/a_{400} 均大于进水,表明经过人工湿地的处理,尾水的腐殖度和芳香度均有所下降. 湿地中腐殖度和芳香度高的物质难以被微生物降解,实验组各采样点 a_{300}/a_{400} 大于对照组,表明铁炭内电解可降低尾水的腐殖度和芳香度. a_{250}/a_{365} 可作为水中溶解性有机物分子质量大小的评价指标,该比值越大,尾水中有机物平均分子质量越小^[24]. 表

3 中 a_{250}/a_{365} 总体上沿程降低,表明污水中有机物分子质量逐渐下降,且实验组 a_{250}/a_{365} 高于对照组,说明实验组污水中大分子有机物向小分子转化更为彻底. 对照组 a_{250}/a_{365} 沿程均匀下降,而实验组经过铁炭层(取样点 1~3)后, a_{250}/a_{365} 显著升高,表明铁炭层是大分子有机物向小分子转化的主要单元.

表 3 湿地各采样点水样 DOC 质量浓度及光谱特征

Table 3 DOC and spectral characteristics of influent, effluent and sampling points along the path of vertical flow constructed wetland								
湿地名称	水样	DOC $\text{mg}\cdot\text{L}^{-1}$	a_{250}	a_{300}	a_{365}	a_{400}	a_{300}/a_{400}	a_{250}/a_{365}
对照组	进水	12.62	0.136	0.069	0.028	0.014	4.93	4.86
	1	11.17	0.131	0.066	0.026	0.012	5.50	5.04
	2	10.03	0.127	0.062	0.024	0.011	5.64	5.29
	3	8.55	0.107	0.061	0.020	0.010	6.10	5.35
	4	8.03	0.097	0.061	0.018	0.009	6.78	5.39
	5	7.67	0.095	0.059	0.017	0.009	6.56	5.59
	出水	7.31	0.091	0.053	0.016	0.008	6.63	5.69
实验组	1	11.32	0.131	0.067	0.027	0.013	5.15	4.85
	2	9.75	0.121	0.063	0.022	0.011	5.73	5.50
	3	10.84	0.132	0.066	0.023	0.010	6.60	5.74
	4	7.42	0.091	0.055	0.015	0.009	6.11	6.07
	5	5.14	0.073	0.047	0.011	0.007	6.71	6.64
	出水	4.06	0.064	0.042	0.009	0.006	7.00	7.11

2.2 垂直流人工湿地对尾水中氮素的去除

两组湿地对尾水中 NH_4^+-N 、TN 的去除效果如图 3 所示. 两组人工湿地对 NH_4^+-N 均有良好的去除

效果,出水浓度达到地表水 IV 类标准 ($1.5\text{ mg}\cdot\text{L}^{-1}$),接近于 II 类标准($0.5\text{ mg}\cdot\text{L}^{-1}$),全年去除率相对稳定.

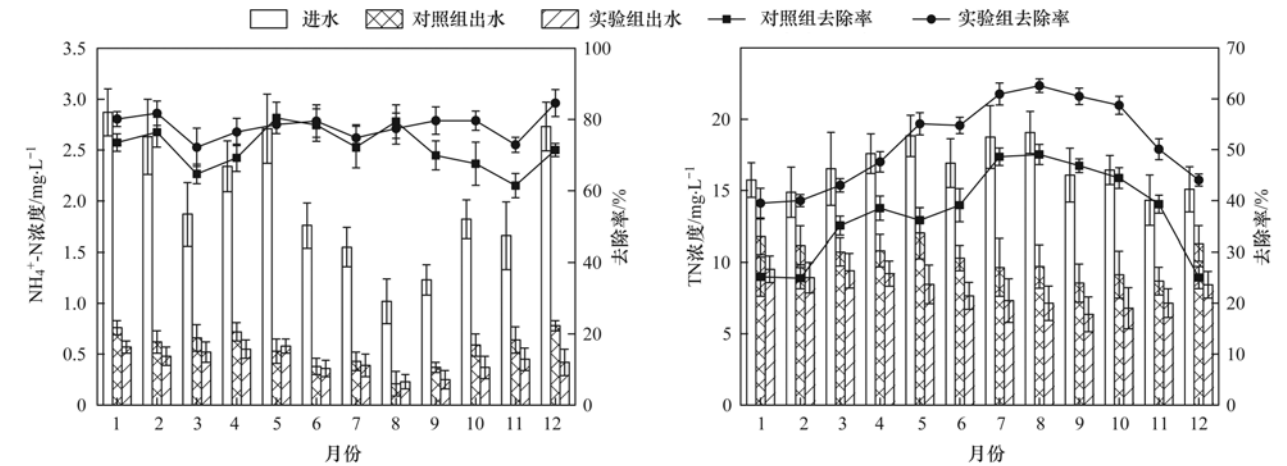


图 3 垂直流湿地进出水氨氮、TN 浓度及平均去除率

Fig. 3 Concentrations of NH_4^+-N and TN in influent and effluent of vertical flow constructed wetland and average removal efficiencies

两组湿地对 TN 的去除率表现出明显的季节性,去除率随温度升高而升高. 采用 SPSS 软件进行统计学分析表明,两组湿地 TN 去除率存在显著性差异 ($P<0.05$),实验组全年 TN 平均去除率均高于对照组,且出水浓度在 $10\text{ mg}\cdot\text{L}^{-1}$ 以下,满足现阶段北京(2012 年)、天津(2015 年)更为严格的污水排放标准. 对照组和实验组全年 TN 平均去除率分别为 37.69%、51.41%,暖季为 41.91%、54.81%,寒季为 25.03%、41.20%,实验组全年、暖季、寒季 TN 平均去除率分别高出对照组 13.72%、12.90%、16.17%.

由此可见,湿地基质掺杂铁炭有利于系统脱氮,尤其是提高冬季低温的脱氮效率. 这一方面是因为湿地基质掺杂的铁炭可改善尾水可生化性,为后续微生物脱氮提供更多的可利用碳源,从而提高脱氮效率. 另一方面, Fe^{2+} 和 Fe^{3+} 是微生物生命活动中重要的电子传递体系,铁炭内电解过程中产生的 Fe^{2+} 和 Fe^{3+} 可以参与这种电子传递^[25, 26],从而加速细胞中的电子传递效率,提高微生物活性,改善系统脱氮效率,特别是冬季低温脱氮效率.

两组人工湿地沿程氮素形态分布如图 4 所示

(10月12日测定结果). 从中可知进出水及沿程污水中的氮素主要以 $\text{NO}_3^- \text{-N}$ 为主,占TN的70%以上,而 $\text{NH}_4^+ \text{-N}$ 在湿地系统中得到了较好的去除,占TN比例始终在8%以下. 实验组有机氮[Org-N, $\text{Org-N} = \text{TN} - (\text{NH}_4^+ \text{-N}) - (\text{NO}_2^- \text{-N}) - (\text{NO}_3^- \text{-N})$]基本沿

程下降,占TN比例由16.79%下降到6.48%,而对照组Org-N占TN比例均在10%以上,这主要是由于铁炭层中铁炭内电解可促进含氮难降解有机物的分解,从而使Org-N占比显著下降. 由于缺乏可利用碳源,对照组还出现了 $\text{NO}_2^- \text{-N}$ 积累的现象.

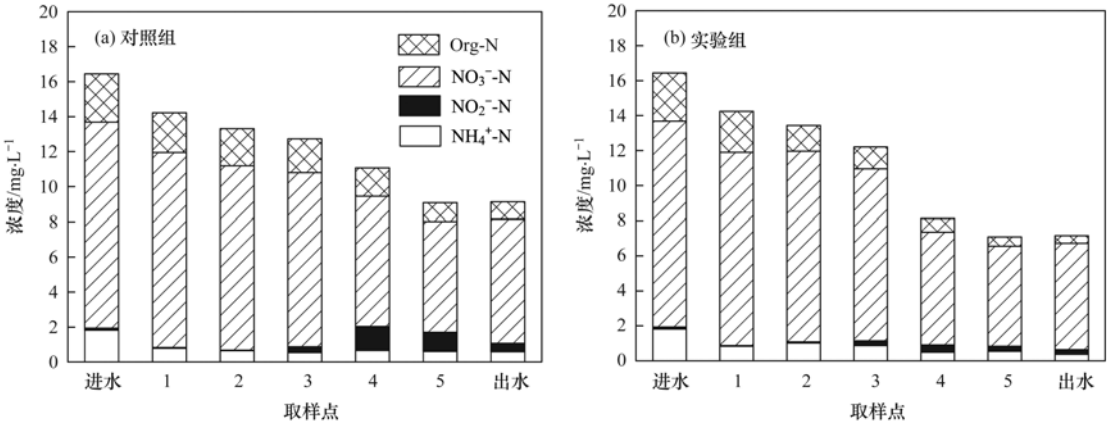


图4 垂直流湿地沿程氮形态分布

Fig. 4 Distribution of nitrogen forms along the path of vertical flow constructed wetland

2.3 垂直流人工湿地中有机物相对分子质量沿程分布特征

表4为两组湿地进出水及各采样点溶解性有机物的 M_r . 从中可知,湿地处理前后,尾水中有机物的相对分子质量均有所下降,对照组和实验组分别降低了25.82%、39.56%;且实验组经过铁炭层(取样点1

~3)后, M_r 下降幅度较大,分别为13.73%、24.15%,这与DOC质量浓度及光谱特征变化相一致.

两组人工湿地有机物沿程分子质量分布如图5所示. 湿地污水中有机物相对分子质量主要集中在 $50 \times 10^3 \sim 500 \times 10^3$ 之间,且有机物向小分子转变. 经过湿地处理后,对照组和实验组 $<10 \times 10^3$ 的有

表4 湿地进出水及各采样点水样溶解性有机物的相对分子质量 $\times 10^3$

Table 4 Molecular weight values in influent, effluent and sampling points along the path of vertical flow constructed wetland $\times 10^3$

湿地名称	水样						
	进水	1	2	3	4	5	出水
对照组	364.8	350.6	329.6	321.5	301.2	279.5	270.6
实验组	364.8	353.3	314.7	276.7	244.8	227.7	220.5

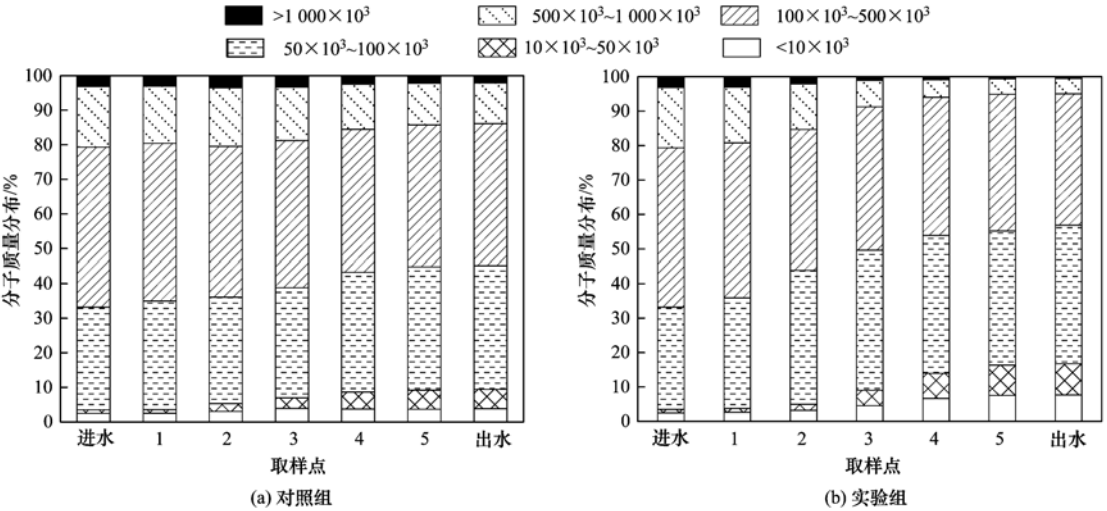


图5 垂直流湿地有机物相对分子质量分布

Fig. 5 Distribution of molecular weight of organics in influent, effluent and sampling points along the path of vertical flow constructed wetland

机物质量分数分别增加了 1.31% 和 5.15%, $10 \times 10^3 \sim 50 \times 10^3$ 的有机物质量分数分别增加了 4.54% 和 8.11%。而 $500 \times 10^3 \sim 1\,000 \times 10^3$ 的有机物质量分数分别减少了 4.80% 和 13.19%, $>1\,000 \times 10^3$ 的有机物质量分数分别减少了 0.94% 和 2.56%。此外,经过铁炭层(取样点 1~3)后, $500 \times 10^3 \sim 1\,000 \times 10^3$ 和 $>1\,000 \times 10^3$ 的有机物质量分数显著下降,分别下降了 8.52% 和 1.97%。可见,湿地基质掺杂铁炭能有效促进大分子有机物向小分子物的转变^[27]。

3 结论

(1) ICIE-VFCW 可强化系统对污水厂尾水中 COD 的去除,全年出水 COD 可稳定在 $30 \text{ mg} \cdot \text{L}^{-1}$ 以下。ICIE-VFCW 较不掺杂铁炭普通湿地全年、暖季、寒季 COD 平均去除率分别可提高 10.16%、9.81%、11.22%。

(2) 无论湿地基质是否掺杂铁炭,垂直流湿地对尾水中 $\text{NH}_4^+ - \text{N}$ 去除效果良好,出水稳定在 $1.5 \text{ mg} \cdot \text{L}^{-1}$ 以下,接近 $0.5 \text{ mg} \cdot \text{L}^{-1}$,掺杂铁炭对 $\text{NH}_4^+ - \text{N}$ 去除影响不大。

(3) ICIE-VFCW 可提高湿地的脱氮效率,湿地出水 TN 可维持在 $10 \text{ mg} \cdot \text{L}^{-1}$ 以下。ICIE-VFCW 较不掺杂铁炭普通湿地全年、暖季、寒季 TN 平均去除率分别提高 13.72%、12.90%、16.17%。

(4) ICIE-VFCW 可促进尾水中难降解大分子有机物转变为小分子易降解有机物,减小污水中有机物的腐殖度和芳香度,改善尾水可生化性,为湿地微生物提供更多可利用碳源,促进反硝化脱氮,提高湿地系统的脱氮效率。

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