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EDDS 对土壤铜镉有效性及蓖麻吸收转运的影响

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摘要: 为探究螯合剂对植物吸收重金属的影响, 以蓖麻(*Ricinus communis* L.) 为供试植物, 通过土培和盆栽试验, 研究不同含量乙二胺二琥珀酸(EDDS)对土壤中铜镉形态和植物吸收、转运的影响。结果表明, EDDS显著增加了土壤有效态铜和镉含量, 培养 15 d 时, 增幅分别为 43.01%~103.55% 和 51.78%~69.43%, 同时促进了可还原态铜向弱酸提取态转化, 增加了土壤铜的移动性。EDDS 促进了蓖麻对铜的吸收、转运与富集。EDDS 2.5 和 EDDS 5.0 处理时, 地上部铜含量是对照的 4.88 倍和 16.65 倍($P < 0.05$), 根部是对照的 2.89 倍和 3.60 倍($P < 0.05$), 铜转运系数显著提高了 72.73% 和 381.82%。EDDS 5.0 处理时, 蓖麻地上部和根部的铜提取量分别是对照处理的 14.08 倍和 2.16 倍, 总铜提取量是对照处理的 4.70 倍($P < 0.05$)。此外, EDDS 显著增加了蓖麻镉含量, EDDS 2.5 处理时, 地上部和根部分别增加了 15.15% 和 57.42%, 蓖麻总镉提取量显著提高了 13.44%。综上可知, EDDS 能增加土壤铜镉的有效性, 促进蓖麻对铜镉的吸收, 提高蓖麻的修复效率, 其中 5.0 mmol·kg⁻¹ EDDS 更有利于蓖麻对铜的提取, 而 2.5 mmol·kg⁻¹ EDDS 处理对镉的提取有较高的增加效果。

关键词: 铜(Cu); 镉(Cd); 乙二胺二琥珀酸(EDDS); 蓖麻; 植物修复; 重金属形态

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Effect of EDDS Application on Soil Cu/Cd Availability and Uptake/transport by Castor

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Abstract: To investigate the effect of chelating agents on plant uptake of heavy metals, castor (*Ricinus communis* L.) was used as the test plant. Soil culture and pot experiments were conducted to study the effects of different concentrations of ethylenediamine disuccinic acid (EDDS) on the forms of Cu and Cd in soil and their absorption and transport by castor. The results showed that the application of EDDS significantly increased the content of available Cu and Cd. After 15 days of cultivation, the available Cu and Cd concentrations in the soil increased by 43.01%-103.55% and 51.78%-69.43%, respectively. EDDS promoted the conversion of reducible Cu to weak acid extractable and increased the mobility of Cu. Meanwhile, the application of EDDS promoted the absorption, transport, and enrichment of Cu in castor. Under the application of 2.5 mmol·kg⁻¹ EDDS and 5.0 mmol·kg⁻¹ EDDS, the Cu concentrations in the shoots were 4.88 times and 16.65 times higher than that of the control ($P < 0.05$), and the Cu concentrations in the roots were 2.89 times and 3.60 times higher than that of the control ($P < 0.05$), respectively. The Cu transport coefficient significantly increased by 72.73% and 381.82% when treated with EDDS 2.5 and EDDS 5.0. Simultaneously, the phytoextraction of Cu in shoots, roots, and their sum were 14.08, 2.16, and 4.70 times higher than that of the control ($P < 0.05$), respectively, when treated with EDDS 5.0. Furthermore, EDDS significantly increased the Cd concentrations in castor. When treated with EDDS 2.5 the shoots and roots increased by 15.15% and 57.42%, respectively, and the phytoextraction of total Cd significantly increased by 13.44%. Generally, the EDDS treatment could increase the available Cu and Cd in soil, promote the uptake of Cu and Cd, and improve the phytoremediation efficiency of castor. Among them, the addition of 5.0 mmol·kg⁻¹ EDDS had the best effect for Cu, whereas the addition of 2.5 mmol·kg⁻¹ EDDS had a higher increase in the phytoextraction of Cd.

Key words: Cu; Cd; N,N'-ethylenediamine disuccinic acid (EDDS); castor; phytoremediation; heavy metal forms

全球土壤重金属污染日趋严重, 已成为当前主要环境挑战之一^[1]。重金属进入土壤后, 不仅会降低粮食产量, 破坏生态环境, 还会通过食物链进入人体, 威胁人类健康^[2,3]。有研究表明, 矿区周边农田土壤以多种重金属复合污染为主, 其中铜镉复合污染最为常见^[4-6]。据《全国土壤污染状况调查公报》^[7]显示, 我国土壤铜和镉的点位超标率分别为 2.1% 和 7.0%, 严重影响粮食生产。因此, 开展矿区农田土壤铜镉污染修复具有重要意义。

植物修复是目前运用较多的绿色修复技术, 具有成本低和不引起二次污染等优势^[8]。其中植物提取是指利用超富集植物从土壤中吸取重金属并转移到地上部, 经收获而去除土壤重金属的方法。植物对重金属的耐性、地上部生物量和重金属含量是影响植

物提取效率的主要因素^[9]。但是, 重金属超富集植物往往存在生物量小、修复周期长和修复效率较低等缺陷, 限制了植物修复技术的大规模应用^[10]。为了进一步弥补植物修复技术的不足, 螯合剂强化植物提取土壤重金属是目前较为有前景的措施^[11]。

螯合剂可与许多种重金属离子形成稳定的水溶性络合物, 使重金属由不溶态转为可溶态而从土壤表面解吸出来^[12]。近年来, N,N'-乙二胺二琥珀酸(EDDS)和 N,N'-二乙酸(GLDA)因其在环境中具有较好的生物可降解性而被认为是具有良好应用前景的

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重金属淋洗剂和活化剂^[13-15]. EDDS是EDTA的结构异构体,具有生物降解的优势,对植物和土壤微生物群落的毒性小,在土壤中残留时间短,其环境风险低于EDTA,在某些方面,EDDS是EDTA的可行替代品^[16,17]. EDDS对重金属的螯合作用在施用7 d后达到峰值,随着EDDS的降解逐渐降低^[18];同时,EDDS分解能释放氮,促进植物生长,提高修复效率^[19]. 陈银萍等^[20]研究表明,适宜浓度的EDDS有利于紫苜蓿幼苗的生长和对镉的吸收积累,有助于细胞壁中镉向可溶部分转移及降低细胞器和线粒体中镉含量. 添加5.0 mmol·kg⁻¹ EDDS可促进向日葵对镉的吸收,土壤镉去除率比对照增加了181.51%^[21];EDDS能与铜形成高稳定性的Cu-EDDS络合物,促进黑麦草对铜的吸收^[22]. 也有研究表明,施用EDDS显著提高了植物地上部铜含量,虽增加了土壤氮含量,但降低了植物生物量^[23].

目前的研究多集中于生物量小的超富集植物,即使植物体内重金属含量增加,但提取总量仍然较低,难以满足实际需要;且多数研究集中于单一的重金属污染修复,缺乏多金属复合污染土壤中,螯合剂对不同重金属的修复效果对比. 所以,在植物修复过程中需从生物量小的超富集植物转向生物量大且对多种重金属有较强富集能力的植物. 蓖麻是一种油料作物,环境适应能力强,具有生物量大、根系发达、经济价值较高等特点,对多种重金属均具有较高的耐性和富集能力^[24]. 因此,本文以蓖麻为供试植物,结合土培和盆栽试验,研究EDDS对蓖麻植株铜镉分布、吸收、转运及富集的强化效果,分析不同时长下EDDS对土壤铜镉有效性、形态、pH、可溶性有机碳(DOC)的影响及相关性,探讨EDDS对土壤铜镉的作用机制和蓖麻提取铜镉的差异,以期对铜镉复合污染农田土壤修复提供科学依据.

1 材料与方 法

1.1 供试材料

供试土壤为水稻土,采自湖北省黄石市阳新县富池镇(N 29°48'40";E 115°25'53")耕层0~20 cm,自然风干后去除植物残体和石砾,磨细过10目和100目筛备用. 其基本理化性质为:pH 7.79,阳离子交换量20.30 cmol·kg⁻¹, ω (有机质)19.72 g·kg⁻¹, ω (全磷)0.70 g·kg⁻¹, ω (全氮)1.63 g·kg⁻¹, ω (总铜)352.25 mg·kg⁻¹, ω (有效态铜)50.48 mg·kg⁻¹, ω (总镉)1.05 mg·kg⁻¹, ω (有效态镉)0.35 mg·kg⁻¹. 供试植物为蓖麻,种子采自于湖北省黄石市铜绿山矿区,自然风干,去除瘪种,装袋保存备用. 供试试剂EDDS

购买于合肥博美生物科技有限公司,为分析纯试剂.

1.2 试验方法

土培试验:称150 g过10目筛的风干土壤于塑料杯中,以溶液形式加入EDDS,使土壤 m_B (EDDS)分别为0、2.5和5.0 mmol·kg⁻¹,分别用EDDS 0、EDDS 2.5和EDDS 5.0表示. 用去离子水维持土壤含水量(质量分数)为25%,室温下培养45 d,3次重复,分别在第1、7、15、25和45 d取样.

盆栽试验:称取过10目筛的风干土壤1.50 kg于塑料盆中(直径15 cm,高12 cm),按每kg土施入基肥100 mg N(尿素);100 mg K₂O(氯化钾);100 mg P₂O₅(过磷酸钙). 盆底部放置塑料袋,防止土粒和养分随水流失. 充分混匀土壤和肥料,加入去离子水,维持土壤含水量为25%左右. 14 d后,播入经75%酒精消毒的蓖麻种子,每盆6颗,维持含水量在25%左右,温度25℃,每天8 h光照. 待蓖麻长出两片叶后间苗,每盆留3株,30 d后,向盆中加入不同质量摩尔浓度的EDDS溶液(pH 7.7),土壤 m_B (EDDS)分别为0、2.5和5.0 mmol·kg⁻¹,种植30 d后收获植物样. 每个试验重复3次.

1.3 测定项目及方法

土壤基本理化性质参照《土壤农化分析》^[25]. 采用pH计(Mettler-Toledo FE20)在水土比为2.5:1下测定土壤pH. 土壤可溶性有机碳(DOC)含量采用1:5土水比提取,振荡离心,0.45 μm滤膜过滤,TOC分析仪测定(德国Vario). 土壤有效铜和镉含量采用DTPA法提取^[26],待测液中铜采用原子吸收分光光度计测定(Varina, AAS240FS),镉采用石墨炉原子吸收光谱仪测定(Agilent, 240Z). 土壤不同形态铜和镉含量采用改进的BCR连续提取法提取^[27]. 蓖麻铜和镉含量采用HNO₃-HClO₄(体积比4:1)消煮,定容过滤,原子吸收分光光度计测定.

1.4 数据处理

采用Excel 2007处理和分析数据,用SPSS 22.0进行单因素方差分析,用Duncan法在P<0.05水平上进行显著性比较,用Origin 8.5作图.

用以下公式计算蓖麻对铜和镉的转运系数(TF)、富集系数(BCF)和提取量:

$$\text{转运系数(TF)} = \frac{\text{植物地上部重金属含量}}{\text{根部重金属含量}}$$

$$\text{富集系数(BCF)} = \frac{\text{植物体内重金属含量}}{\text{土壤重金属含量}}$$

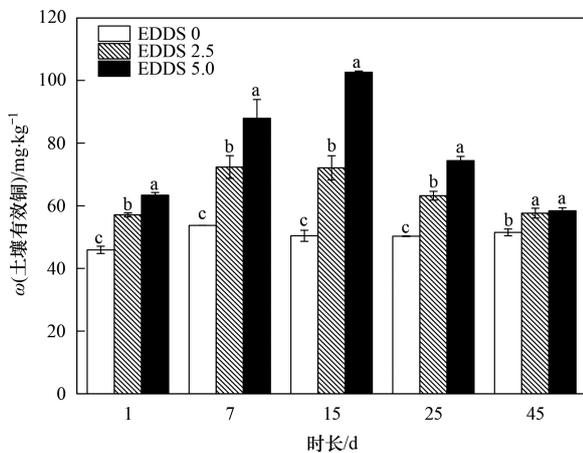
$$\text{提取量} = \text{植物体内重金属含量} \times \text{植物生物量}$$

2 结果与分析

2.1 EDDS对土壤铜和镉有效性的影响

2.1.1 DTPA提取态铜镉含量

不同培养时长下,EDDS对土壤铜有效性的影响如图1. 添加EDDS后均显著增加了土壤有效铜含量,并随着EDDS含量升高而增加,随着培养时长先增加后降低,在培养1 d后,EDDS 2.5和EDDS 5.0处理中土壤有效铜含量与对照相比分别增加了24.32%和38.02%. 培养15 d后,EDDS 5.0处理中土壤有效铜含量最高,为102.61 mg·kg⁻¹,比对照增加了103.55%. 培养45 d后,不同含量EDDS对有效铜含量的影响不显著,但活化效果均显著高于对照处理.



不同小写字母表示同一培养时长下处理间的显著性差异($P < 0.05$)

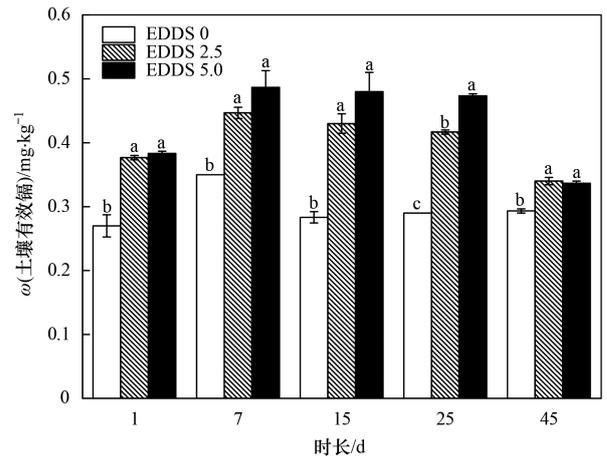
图1 EDDS不同培养时长下对土壤有效态铜含量的影响

Fig. 1 Effect of incubation time after EDDS addition on available Cu concentration in soil

EDDS对土壤镉有效性的影响如图2. 土壤有效镉含量的变化趋势与有效铜大致相同,但EDDS 2.5和EDDS 5.0处理对土壤镉的活化效果差异不显著. 培养1 d后,添加EDDS处理下,土壤有效镉含量与对照相比增加了39.52%和41.96%;培养到第7、15和25 d时,EDDS对土壤镉均有较好的效果,EDDS 5.0处理中土壤有效镉含量与相应培养时长的对照相比,分别增加了39.06%、69.43%和63.21%;培养45 d时,EDDS对镉的活化效果最低. 因此,EDDS对土壤铜镉的活化受培养时长和EDDS含量的影响.

2.1.2 土壤铜和镉形态的变化

随着培养时长的变化,EDDS对土壤铜形态的影响如图3. 培养1~15 d时,添加EDDS可显著增加弱酸提取态铜含量. 第1 d时,EDDS 5.0处理中弱酸提取态铜含量占9.82%,比对照增加了3.43%,可还原态铜含量降低了2.68%,而可氧化态和残渣态铜含量与对照无明显差异. 第15 d时,EDDS 2.5和EDDS 5.0



不同小写字母表示同一培养时长下处理间的显著性差异($P < 0.05$)

图2 EDDS不同培养时长下对土壤有效态镉含量的影响

Fig. 2 Effect of incubation time after EDDS addition on available Cd concentration in soil

处理中,弱酸提取态铜含量占比分别增加了1.42%和2.68%,可还原态和可氧化态含量与对照相比虽有下降,但差异不明显. 同一EDDS水平处理下,弱酸提取态铜含量随着培养时长增加而降低,EDDS 5.0处理中,弱酸提取态铜含量占比从第1 d的9.82%降低到45 d的5.96%,可还原态铜从35.33%增加到39.39%,可氧化态和残渣态铜含量无显著变化. 因此,EDDS处理下弱酸提取态铜含量主要是通过可还原态转化而来,随着培养时长增加,EDDS分解,导致弱酸提取态铜向可还原态转化.

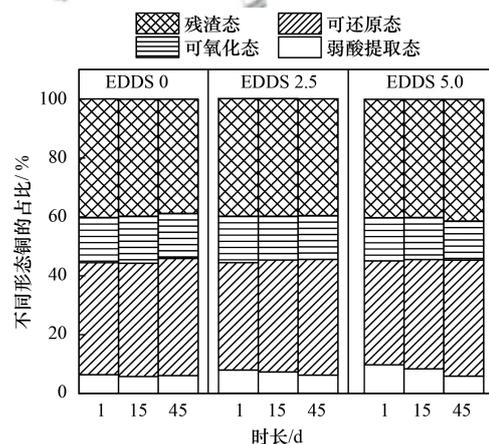


图3 EDDS不同培养时长下土壤铜形态的变化

Fig. 3 Effect of incubation time after EDDS addition on Cu fractions in soil

EDDS对土壤镉形态的影响如图4. 培养1 d后,EDDS对弱酸提取态镉含量的影响不大,但随着培养时长增加,添加EDDS显著增加了土壤弱酸提取态镉含量占比,培养15 d后,EDDS 2.5和EDDS 5.0处理弱酸提取态镉含量分别占53.97%和56.04%,显著高于对照处理,残渣态含量占比从18.80%下降到

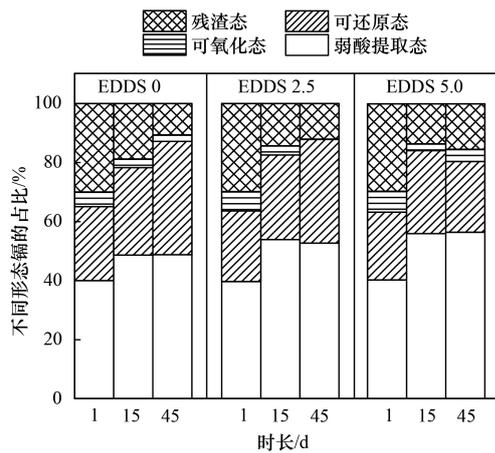


图4 EDDS不同培养时长下土壤镉形态的变化
Fig. 4 Effect of incubation time after EDSS addition on Cd fractions in soil

14.24%和13.46%,而可还原态和可氧化态镉与对照相比差异不显著. 培养45 d后, EDDS 2.5和EDDS 5.0处理的弱酸提取态镉含量占比与同时长对照相比,分别增加了3.98%和7.73%.

同一EDDS水平下,延长培养时长显著增加了弱酸提取态镉含量,EDDS 2.5处理中,培养45 d后,弱酸提取态镉占比从39.73%增加到52.75%,与第1 d同处理下相比,可还原态镉含量占比增加11.21%,可

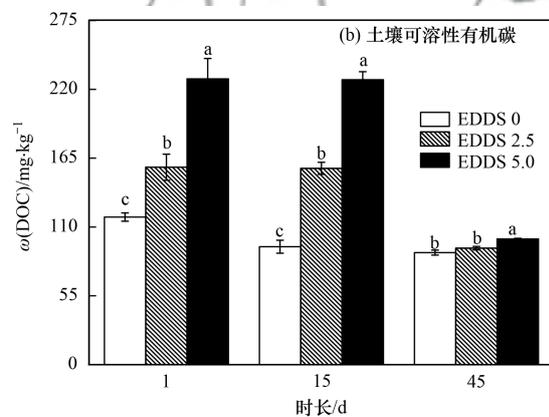
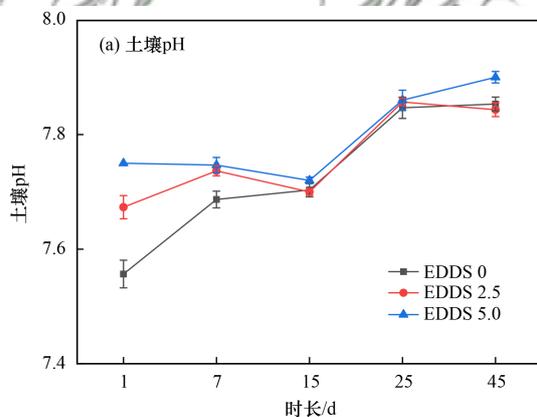
氧化态镉降低6.45%,残渣态镉降低17.79%. 而EDDS 5.0处理中,培养45 d与1 d相比,土壤弱酸提取态镉含量占比增加16.11%,残渣态镉降低14.27%.

可知,随着培养时长增加,EDDS可促进残渣态和可氧化态镉向弱酸提取态和可还原态镉转化.

2.2 土壤pH和可溶性有机碳的变化

EDDS对土壤pH和可溶性有机碳(DOC)含量的影响如图5. 添加EDDS能提高土壤pH,但增幅不大,培养1 d时,EDDS 2.5和EDDS 5.0处理中,土壤pH分别增加了0.10和0.20个单位,这可能与EDDS本身呈弱碱性有关,EDDS含量越高,pH增幅越大,但差异不显著. 随着培养时长增加,各处理下的土壤pH均有一定程度升高,培养到45 d,EDDS 2.5和EDDS 5.0处理与培养1 d相比,升高0.20个单位.

添加EDDS显著增加了土壤DOC含量,培养1 d时,EDDS 2.5和EDDS 5.0处理中,DOC含量分别比对照增加了33.74%和93.62%. 随着培养时长增加,EDDS含量逐渐降低,培养到45 d,EDDS 5.0处理的 $\omega(\text{DOC})$ 为 $100.42 \text{ mg}\cdot\text{kg}^{-1}$,与培养1 d相比下降了56.03%. 因此,EDDS能有效增加土壤DOC含量,但DOC可随着培养时长增加而分解.



不同小写字母表示同一培养时长下处理间的显著性差异($P < 0.05$)

图5 EDDS不同培养时长下土壤pH和可溶性有机碳的变化

Fig. 5 Effect of incubation time after EDSS addition on pH and soluble organic carbon in soil

培养1、15和45 d,土壤有效态铜和镉含量与土壤pH、DOC的相关性分析如表1. 可以看出,土壤有效态铜和镉含量与土壤DOC含量呈极显著正相关,

相关系数分别为0.701和0.745. 土壤pH与有效态铜镉含量的相关性不明显,可能是因为添加EDDS后,土壤本身具有一定的缓冲能力,因此整合剂溶液对

表1 土壤pH、DOC和有效态铜镉的皮尔森相关系数¹⁾

Table 1 Pearson correlation coefficient of soil pH, DOC, and available Cu and Cd

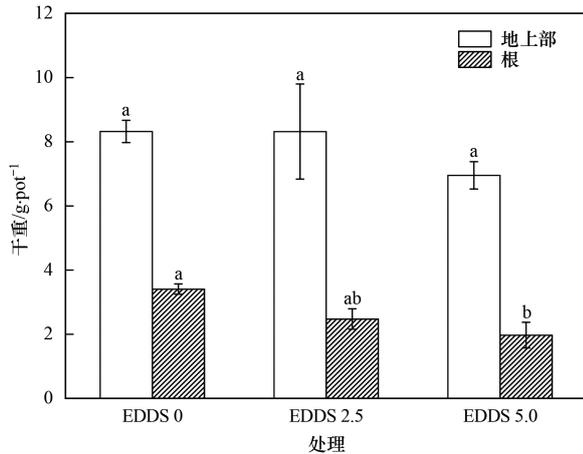
	DTPA-Cu	DTPA-Cd	土壤 pH	土壤 DOC
DTPA-Cu	1			
DTPA-Cd	0.896**	1		
土壤 pH	0.013	-0.017	1	
土壤 DOC	0.701**	0.745**	-0.285	1

1)**为 $P < 0.01$

土壤pH的影响不大^[28].

2.3 EDDS对蓖麻生长和吸收铜镉的影响

EDDS对蓖麻生物量的影响如图6. 添加EDDS降低了蓖麻地上部和根部生物量,未添加EDDS的处理中,每盆蓖麻地上部和根部干重分别为8.32 g和3.41 g,EDDS 5.0处理时,蓖麻地上部生物量与对照相比降低了16.47%,但差异不显著,而每盆根部生物量为1.97 g,与对照相比显著降低了42.23%. 可知,EDDS在一定程度上降低蓖麻生物量,抑制植物的生长.



不同小写字母表示处理间的显著性差异 ($P < 0.05$),下同

图6 EDDS对蓖麻生长的影响

Fig. 6 Effect of EDDS on dry weight of castor

EDDS促进了蓖麻对土壤铜的吸收(图7). EDDS 2.5和EDDS 5.0处理中,蓖麻地上部 $\omega(\text{Cu})$ 分别为 $30.91 \text{ mg}\cdot\text{kg}^{-1}$ 和 $105.44 \text{ mg}\cdot\text{kg}^{-1}$,是对照处理的4.88倍和16.65倍($P < 0.05$);添加EDDS后,蓖麻根部

$\omega(\text{Cu})$ 从 $57.39 \text{ mg}\cdot\text{kg}^{-1}$ 增加到 $166.02 \text{ mg}\cdot\text{kg}^{-1}$ 和 $206.73 \text{ mg}\cdot\text{kg}^{-1}$,且蓖麻根部铜含量显著高于地上部.

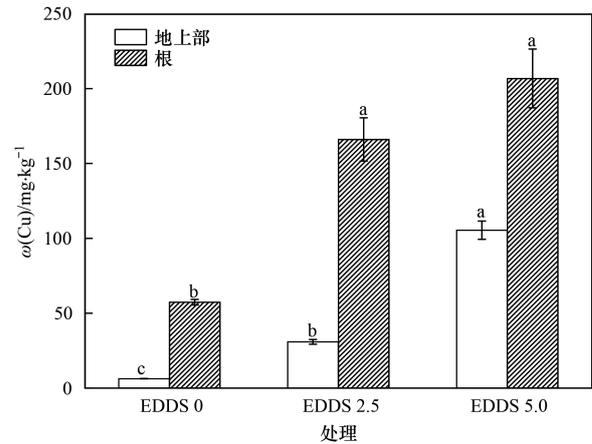


图7 EDDS对蓖麻铜含量的影响

Fig. 7 Effect of EDDS on Cu concentration in castor

EDDS显著增加了蓖麻地上部对铜的提取(表2). 添加EDDS后,地上部铜提取量比对照处理增加了3.84倍和13.08倍($P < 0.05$);根部铜提取量从每盆 $194.52 \mu\text{g}$ 增加到 $412.30 \mu\text{g}$ 和 $419.64 \mu\text{g}$,但不同EDDS水平间根部铜提取量差异不显著. EDDS 5.0处理中,每盆蓖麻总铜提取量达 $1160.15 \mu\text{g}$,是对照处理的4.70倍. EDDS促进了铜向蓖麻地上部的转运,EDDS 2.5和EDDS 5.0处理后,蓖麻的铜转运系数显著增加了72.73%和381.82%. 可知,EDDS能促进蓖麻对铜的吸收,有利于促进铜从根部向地上部转运. 添加EDDS促进了蓖麻根系和茎对铜的富集,EDDS 5.0处理后, $\text{BCF}_{\text{根}}$ 和 $\text{BCF}_{\text{茎}}$ 分别比对照增加了2.60倍和13.00倍.

表2 EDDS对蓖麻富集和转移铜的影响¹⁾

Table 2 Effect of EDDS on the accumulation and transportation of Cu in castor

处理	地上部铜量 / $\mu\text{g}\cdot\text{pot}^{-1}$	根部铜量 / $\mu\text{g}\cdot\text{pot}^{-1}$	总铜量 / $\mu\text{g}\cdot\text{pot}^{-1}$	转运系数 (TF)	根富集系数 ($\text{BCF}_{\text{根}}$)	茎富集系数 ($\text{BCF}_{\text{茎}}$)
EDDS 0	$52.59 \pm 0.83\text{c}$	$194.52 \pm 2.08\text{b}$	$247.10 \pm 2.65\text{c}$	$0.11 \pm 0.00\text{c}$	$0.15 \pm 0.00\text{c}$	$0.02 \pm 0.00\text{c}$
EDDS 2.5	$254.36 \pm 24.81\text{b}$	$412.30 \pm 47.62\text{a}$	$666.66 \pm 71.21\text{b}$	$0.19 \pm 0.02\text{b}$	$0.44 \pm 0.04\text{b}$	$0.08 \pm 0.00\text{b}$
EDDS 5.0	$740.52 \pm 70.10\text{a}$	$419.64 \pm 87.13\text{a}$	$1160.15 \pm 153.00\text{a}$	$0.53 \pm 0.05\text{a}$	$0.54 \pm 0.05\text{ab}$	$0.28 \pm 0.02\text{a}$

1)不同小写字母表示处理间存在显著性差异,下同

EDDS处理显著促进了蓖麻对镉的吸收,蓖麻地上部镉含量受EDDS施用量影响不大,EDDS 2.5和EDDS 5.0处理后,蓖麻地上部 $\omega(\text{Cd})$ 分别为 $1.23 \text{ mg}\cdot\text{kg}^{-1}$ 和 $1.34 \text{ mg}\cdot\text{kg}^{-1}$ (图8),与不添加EDDS处理相比增加了15.15%和25.46%;根部 $\omega(\text{Cd})$ 分别为 $5.25 \text{ mg}\cdot\text{kg}^{-1}$ 和 $5.51 \text{ mg}\cdot\text{kg}^{-1}$,比对照分别增加了57.42%和65.17%($P < 0.05$).

EDDS对蓖麻提取镉的影响如表3. 添加EDDS对蓖麻地上部和根部镉提取量均影响不显著,但总

镉提取量在EDDS 2.5处理下达最大值每盆 $22.96 \mu\text{g}$,显著高于CK和EDDS 5.0处理. EDDS降低了镉从蓖麻根部向地上部的转运,EDDS 2.5和EDDS 5.0处理后,镉转运系数分别降低了27.27%和24.24%,但添加EDDS的处理与对照间差异不显著;EDDS显著促进了蓖麻根对镉的富集,EDDS 2.5和EDDS 5.0处理后,蓖麻根系镉富集系数从2.81增加到5.14和5.39. 可知,EDDS促进了蓖麻根对镉的吸收,并使镉主要富集在根部.

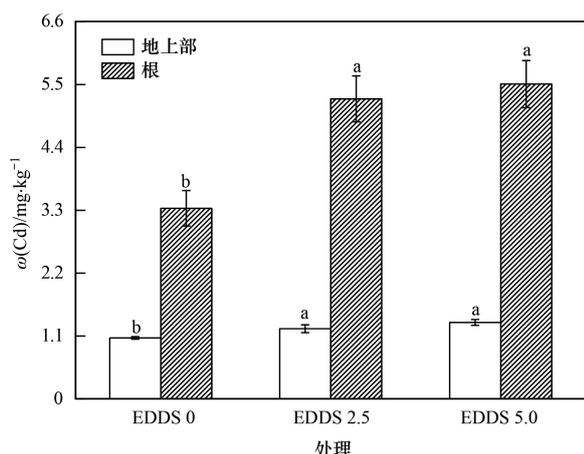


图8 EDDS对蓖麻镉含量的影响

Fig. 8 Effect of EDDS on Cd concentration in castor

3 讨论

3.1 EDDS对土壤重金属有效性的影响

螯合剂可促进土壤重金属的溶解,并形成稳定的络合物,增加重金属的活性和生物有效性^[29]. 罗洋等^[30]研究表明,EDDS可以与土壤中的镉离子结合成配位基,增加镉的移动性,进而提高土壤镉的有效性. Yang等^[31]研究表明,在不同pH下铜均能与EDDS形成稳定的Cu-EDDS络合物,增加其有效性. 本研究(图1和图2)表明,添加EDDS并培养45 d,对土壤铜和镉均有较好的活化效果,但在培养期间,活化效果先增加后降低,在培养7 d和15 d时,EDDS对土壤铜和镉的活化效果最佳. EDDS 5.0处理时,培养15 d后土壤有效铜和镉的含量分别提高了103.55%和69.43%,25 d后活化效果减弱,土壤有效态重金属含量下降,其原因是DOC与土壤DTPA-Cu和DTPA-Cd呈显著的正相关关系(表1),DOC含量是影响DTPA提取态重金属的主要因素^[33],培养后期EDDS逐渐分解,DOC含量降低,螯合能力减弱. 文献[18]和[32]均得到类似结果,EDDS对重金属的螯合作用在施用7 d达到高峰,7~11 d EDDS开始降解,57 d后完全分解.

另外,螯合剂可改变重金属形态,增加重金属在土壤中的迁移性,使土壤重金属由较稳定的形态向较活跃的形态转化^[34]. 本研究中添加EDDS,增加了弱酸提取态铜含量,促进了可还原态铜向弱酸提取态的转化,这与Zhao等^[35]研究的结果一致,EDDS能

直接与弱结合态金属组分络合,促进金属从土壤固相释放到溶液中,并以较慢的速度促进强结合态金属组分的释放,形成金属螯合物,增加土壤重金属的活性. 但在不同培养时长,EDDS虽增加了弱酸提取态镉含量,但其他形态的镉变化规律存在差异,具体原因还有待进一步探究. 螯合剂对重金属的影响因重金属类型而异^[36],镉的移动性弱,与EDDS结合时需要更长时间,同时镉和铜与EDDS的稳定常数存在差异,溶液中Cu²⁺和Cd²⁺与EDDS的稳定常数分别为18.4和10.8,铜能与EDDS形成具较高热力学稳定性的Cu-EDDS络合物,因此EDDS更有利于与铜结合,增加土壤铜的移动性.

3.2 EDDS对蓖麻吸收铜镉的影响

3.2.1 不同含量EDDS对蓖麻生物量的影响

土壤重金属的有效性和植物生物量是决定植物修复效率的关键因素^[37],选用生物量大的植物和增加土壤有效态重金属含量是提取更多重金属的有效措施. 不同量EDDS对植物生长的影响不同,高施入量会对植物产生毒害,如导致植株矮小、叶片发黄且根系短小^[38]. 本研究中,与对照相比,添加EDDS并未对蓖麻地上部生长产生明显的影响,可能是因为加入EDDS含量较低,对蓖麻地上部毒害小,以及地上部吸收的铜镉对蓖麻的胁迫作用不明显^[30]. 而添加EDDS后,根部生物量从每盆3.41 g下降到1.97 g,EDDS抑制了蓖麻根系的生长;当螯合剂作用于土壤时,会与金属离子形成络合物,然后,金属离子或者螯合剂-金属离子络合物进入植物根部,并通过内皮层快速运输到芽,会对根通道细胞产生破坏^[22],同时螯合物可以从根细胞的质膜中提取阳离子,对根细胞造成损伤. 另外,EDDS能增加土壤重金属的有效性,活化的重金属会对植物产生氧化损伤和代谢紊乱. 当EDDS的毒性超过植物的自我防御能力时,植物的生物量就会下降. Hai等^[39]研究表明,施用螯合剂EDDS、EDTA和NTA均降低了黑麦草生物量. Wang等^[40]研究发现,在 $\omega(\text{Cd})$ 为 $2.12 \text{ mg} \cdot \text{kg}^{-1}$ 的土壤中,添加 $3 \text{ mmol} \cdot \text{L}^{-1}$ EDDS使苋菜的根和茎干重分别降低了3.69%和17.57%. Xu等^[41]研究发现, $5.0 \text{ mmol} \cdot \text{kg}^{-1}$ EDDS抑制了向日葵的生长,破坏了细胞的完整性和改变了叶绿体结构.

表3 EDDS对蓖麻富集和转移镉的影响

Table 3 Effect of EDDS addition on the accumulation and transportation of Cd in castor

处理	地上部镉量 / $\mu\text{g} \cdot \text{pot}^{-1}$	根部镉量 / $\mu\text{g} \cdot \text{pot}^{-1}$	总镉量 / $\mu\text{g} \cdot \text{pot}^{-1}$	转运系数 (TF)	根富集系数 (BCF _根)	茎富集系数 (BCF _茎)
EDDS 0	8.90±0.39ab	11.34±1.10a	20.24±1.30b	0.33±0.03a	2.81±0.11b	1.04±0.02c
EDDS 2.5	9.94±0.69a	13.03±1.41a	22.96±1.97a	0.24±0.03a	5.14±0.39a	1.20±0.07ab
EDDS 5.0	9.29±0.48a	11.08±1.76a	20.38±2.09b	0.25±0.02a	5.39±0.40a	1.31±0.05a

3.2.2 EDDS对铜与镉在蓖麻中转运效果的影响机制分析

植物修复是一种绿色可持续的重金属修复技术,但对重金属有效性低的土壤,比如碱性土壤,如何提高土壤重金属的移动性是增强植物修复效果的关键.在植物修复中,螯合物一方面增加金属的流动性,促进根系吸收和转运重金属;另一方面改变重金属在植物体内的储存形式,降低重金属对植物的毒害.有研究表明,施加适量EDDS能显著增加土壤有效态重金属含量,促进植物吸收^[30,34,42],本研究也得到类似结果.Zhao等^[22]研究表明,EDDS与铜形成Cu-EDDS络合物,提高了铜在植物体内的迁移能力,促进铜向植物地上部运输.Wang等^[43]研究表明,在铜污染的土壤中添加5 mmol·kg⁻¹EDDS,鸭跖草茎中铜含量增加15%~47%.本研究中,添加EDDS提高了蓖麻根和茎的铜生物富集系数与转运系数,促进了铜从土壤向植物地上部的转运.而镉主要富集在蓖麻的根部,EDDS增加了镉从土壤向蓖麻根部的转移.镉与铜在蓖麻中的转运存在差异的原因可能是,植物体内并没有专一性的镉离子通道和转运载体^[44],同时,镉的运输和转运还受植物蒸腾速率和气孔导度的影响,蒸腾速率增大更有利于促进Cd²⁺通过载体蛋白和离子通道向地上部运输^[45].

本研究结果表明,添加EDDS后,地上部铜提取量比对照增加了3.84倍和13.08倍($P<0.05$);根部铜提取量增加1.12倍和1.16倍.EDDS 5.0处理时,蓖麻总铜提取量达每盆1 160.15 μg,是对照处理的4.70倍.而EDDS 5.0处理后蓖麻的总镉提取量低于EDDS 2.5处理.Wang等^[46]也得到类似结果,即低剂量的EDDS更能促进孔雀草和美洲商陆对镉的提取,这可能与EDDS对植物的毒性有关,由于生物量降低而引起提取量降低.由此说明,EDDS更能促进蓖麻对铜的提取,提高对土壤铜的修复效率.

4 结论

(1)EDDS可活化土壤中铜镉,增加铜镉的生物有效性,促进可还原态铜向有效态的转化.培养15 d时,土壤有效铜和镉含量分别增加了43.01%~103.55%和51.78%~69.43%,25 d后活化效果逐渐降低.

(2)添加EDDS促进了蓖麻根和地上部对铜镉的吸收,其中添加5.0 mmol·kg⁻¹EDDS时,蓖麻地上部和根部铜含量是对照处理的16.65倍和3.60倍,蓖麻地上部和根部镉含量分别增加了25.46%和65.17%,但该含量EDDS会对根系产生一定胁迫.

(3)添加EDDS显著增加蓖麻地上部铜提取量,

促进铜向地上部转移,强化蓖麻对铜的去除效率,当添加5.0 mmol·kg⁻¹EDDS时,蓖麻地上部铜提取量比对照增加了13.08倍,铜转运系数比对照增加了381.82%.而EDDS使镉主要富集在蓖麻根部,添加2.5 mmol·kg⁻¹EDDS和5.0 mmol·kg⁻¹EDDS,蓖麻根富集系数分别增加了82.92%和91.81%.

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