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钙镁磷肥对石灰、海泡石组配修复镉污染稻田土壤的影响

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摘要: 改良剂组配(例如羟基磷灰石和沸石)可有效降低土壤中重金属的生物有效性,抑制水稻对重金属的积累,从而提高农产品的安全水平. 本文通过田间试验,研究钙镁磷肥施用对石灰、海泡石组配降低稻田土壤有效态Cd含量、水稻各部分中Cd含量的影响. 结果表明,施用钙镁磷肥可显著促进石灰、海泡石组配的钝化修复效果. 与石灰、海泡石组配处理相比,施用钙镁磷肥,土壤中有有效态Cd含量和糙米中Cd含量显著降低,在施用 $2\,250\text{ kg}\cdot\text{hm}^{-2}$ 钙镁磷肥的条件下,土壤中 Cd^{2+} 发生沉淀或络合反应生成难溶性磷酸镉而降低土壤酸可提取态、可还原态和可氧化态Cd的占比,土壤中有有效态Cd含量显著($P < 0.05$)降低46.97%,糙米中Cd含量降低至 $0.04\text{ mg}\cdot\text{kg}^{-1}$,低于《食品安全国家标准》(GB 2762-2017)中糙米镉限量($0.2\text{ mg}\cdot\text{kg}^{-1}$)标准,且糙米产量增加28.34%. 相关性分析结果表明,水稻根、茎叶和糙米中Cd含量与土壤中有有效态Cd含量,水稻糙米中Cd含量与根和茎叶中Cd含量呈极显著正相关关系($P < 0.01$),土壤有效态Cd含量是影响水稻糙米中Cd含量的关键性因素,钙镁磷肥可提高石灰与海泡石对土壤中有有效态Cd降低效果来减少水稻根和茎叶对Cd的吸收,进而降低糙米中Cd含量. 上述结果表明,施用钙镁磷肥提高了石灰、海泡石组配降低糙米中Cd含量的效果,可实现Cd污染稻田水稻安全生产和增产.

关键词: 污染稻田土壤; 镉; 钙镁磷肥; 石灰; 海泡石; 组配修复

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Effect of Calcium Magnesium Phosphate on Remediation Paddy Soil Contaminated with Cadmium Using Lime and Sepiolite

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Abstract: The combined application of amendments, such as hydroxyapatite and zeolite, can effectively reduce both the bioavailability of heavy metals in soil and the bioaccumulation of heavy metals by rice, thus improving the safety of agricultural products. In this work, the effect of calcium magnesium phosphorus fertilizer on the content of available Cd in paddy soil and Cd accumulation in various parts of rice by lime combined with sepiolite was studied using a plot experiment. The results show that calcium magnesium phosphorus fertilizer can significantly promote the remediation effect of lime combined with sepiolite. Compared with the treatment with lime combined with sepiolite, the application of calcium magnesium phosphorus fertilizer significantly decreases the available Cd content in the soil and the Cd accumulation in the brown rice. With calcium magnesium phosphorus fertilizer of $2\,250\text{ kg}\cdot\text{hm}^{-2}$, Cd^{2+} in soil formed less soluble cadmium phosphate, the content of extractable, reducible and oxidizable Cd was reduced, and the percentage of available Cd content in the soil was significantly ($P < 0.05$) reduced by 46.97%. Meanwhile, the Cd content in brown rice was decreased to $0.04\text{ mg}\cdot\text{kg}^{-1}$, which is far lower than the level required by the National Standard for Food Safety (GB 2762-2017) ($0.2\text{ mg}\cdot\text{kg}^{-1}$). In addition, the production of brown rice was increased by 28.34%. Correlation analysis shows that the Cd content in rice roots, straw, and brown rice was positively correlated with the available Cd content in soil ($P < 0.01$), and between the Cd content in brown rice and in roots and straw ($P < 0.01$). In general, the results indicate that the available Cd content in soil was the key factor affecting the Cd content in brown rice. The application of calcium magnesium phosphorus fertilizer can improve the amendment of lime combined with sepiolite for available Cd in soil and reduce the Cd absorption of rice roots and straw so that the Cd content in brown rice is reduced. The results show that the application of calcium magnesium phosphorus fertilizer enhances the effect of reducing the Cd content in brown rice by lime combined with sepiolite. Finally, it can both meet the safety requirements of brown rice quality and increase the yield of brown rice in Cd-contaminated paddy soil.

Key words: contaminated paddy soil; cadmium; calcium magnesium phosphorus fertilizer; lime; sepiolite; combination remediation

重金属具有隐蔽性、能通过食物链的放大作用在人体富集等特点而备受关注^[1-3]. 据调查,我国有19.4%的农田重金属含量超标,主要重金属污染元素为Cd、Hg、As、Cu和Pb^[4]. 其中,Cd因在人体积累会引发肾功能失调、破坏骨骼,导致骨质疏松、骨痛等而受到重视^[5,6]. 水稻吸收污染农田土壤中

Cd,导致糙米中Cd含量超过《食品安全国家标准》

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(GB 2762-2017) 中糙米镉限量 ($0.2\text{ mg}\cdot\text{kg}^{-1}$) 标准,因此,对一些中轻度重金属污染农田的边修复边生产显得尤为重要。

众多重金属污染农田土壤修复技术中,钝化修复效果好、操作简单和经济,适用于大面积污染土壤修复且不影响农作物的生产。一般说来,多种钝化剂联合使用可达到更好的修复效果,石灰组配黏土矿物钝化修复重金属污染土壤受到广泛关注^[7~9]。然而,长期施用石灰和黏土矿物(如海泡石)会破坏土壤团粒结构,造成土壤板结,进而影响土壤中微生物活性,最终导致粮食减产^[10],因此需要减少石灰和黏土矿物(如海泡石)的用量,寻找一种既能促进石灰组配黏土矿物(如海泡石)对重金属的钝化修复效果,又不影响甚至增加粮食产量的修复材料。有研究表明,钙镁磷肥不仅可以提高作物产量,还能降低 Cd 的迁移性、植物可利用性,减少水稻对 Cd 的吸收^[11],但使用钙镁磷肥辅助石灰组配黏土矿物

(如海泡石)对重金属污染农田进行修复暂未见报道。然而,生产钙镁磷肥的原料磷矿中含有伴生元素 Cd,导致钙镁磷肥本身含有一定的 Cd^[12,13],在使用过程中,若添加量不当,不仅不能降低土壤中 Cd 的毒性,还会造成新的 Cd 污染,因此科学施用非常重要。

本文通过大田试验研究施用钙镁磷肥对石灰和海泡石组配钝化修复 Cd 污染农田效果的影响,以期 Cd 污染农田组配钝化修复过程中合理施加钙镁磷肥提供依据。

1 材料与方法

1.1 供试材料

供试田间试验点位于湖南省某退役工业区附近农田。供试改良剂石灰、海泡石和钙镁磷肥均为市售产品,供试农田土壤和改良剂基本理化性质见表 1。供试水稻品种为晚稻天优华占,籼型三系杂交水稻。

表 1 供试土壤和改良剂基本理化性质¹⁾

Table 1 Basic physicochemical properties of tested soil and amendments

项目	pH 值	碱解氮 / $\text{mg}\cdot\text{kg}^{-1}$	有效磷 / $\text{mg}\cdot\text{kg}^{-1}$	速效钾 / $\text{mg}\cdot\text{kg}^{-1}$	有机质 / $\text{g}\cdot\text{kg}^{-1}$	Cd/ $\text{mg}\cdot\text{kg}^{-1}$	
						全量	有效态
土壤	6.62	201.72	21.01	65.47	25.12	4.98	1.33
石灰	—	—	—	—	—	0.46	—
海泡石	—	—	—	—	—	0.21	—
钙镁磷肥	—	—	—	—	—	0.61	—

1) “—”表示未检验

1.2 试验设计

本试验共设置 5 个处理,分别为对照(CK)、石灰+海泡石(LS)、石灰+海泡石+钙镁磷肥1 $125\text{ kg}\cdot\text{hm}^{-2}$ (LSP1)、石灰+海泡石+钙镁磷肥2 $250\text{ kg}\cdot\text{hm}^{-2}$ (LSP2)和石灰+海泡石+钙镁磷肥3 $375\text{ kg}\cdot\text{hm}^{-2}$ (LSP3),其中,石灰用量为 $1\,575\text{ kg}\cdot\text{hm}^{-2}$,海泡石用量为 $1\,125\text{ kg}\cdot\text{hm}^{-2}$ 。每个处理重复 3 次,共 15 个处理小区。每个处理小区面积为 30 m^2 ,随机排列。基肥的施加、田间水分管理和病虫害防治等措施按照传统农业生产模式进行,种植 70 d 水稻成熟后进行样品采集。

1.3 样品收集与分析

在田间试验小区采集水稻及其根际土壤,水稻带回实验室先用自来水冲洗干净,再用去离子水冲洗 3 次后,将水稻鲜样根、茎叶和米分开装入纸袋,于 105°C 下杀青 30 min 后, 60°C 下烘至恒重,称量根、茎叶和糙米重量,粉碎后装袋备用。土壤样品在自然条件下风干,剔除杂物分别过 10、20 和 100 目筛备用。

土壤样品中全量 Cd 采用 $\text{HNO}_3\text{-HClO}_4\text{-HF}$ 法消

解,水稻样品采用 $\text{HNO}_3\text{-HClO}_4$ 法消解^[14],采用改进 BCR 法连续提取土壤中 Cd 的赋存形态^[15,16],用第一步提取的酸可提取态 Cd 代表土壤中 Cd 的有效态含量,用 ICP-MS 测定处理后溶液中 Cd 含量,以国家标准物质土壤 GBW07405(GSS-5)和国家标准物质柑橘叶 GSB-11 进行质量控制,同时做空白试验,土壤样品全量消解 Cd 的回收率可达 98.04%,植物样品消解 Cd 的回收率可达 105.26%,对 4 种 Cd 形态进行质量平衡计算,消解了 BCR 残渣态和原始土壤中 Cd 含量,其中残渣态 Cd 的回收率为 95.15%~101.94%;土壤 pH 值采用水土比 2.5:1 浸提、pH 计(雷磁,PHS-3C)测定,土壤有机质含量采用低温外热重铬酸钾氧化-比色法测定,土壤碱解氮含量采用碱解扩散法测定,有效磷含量采用碳酸氢钠浸提-钼锑抗比色法测定,速效钾采用乙酸铵提取法测定^[17]。

1.4 数据分析

所有数据均采用 Excel 2013 分析,所有图由 OringinPro 8.5 制作,采用 SPSS 16 进行单因素方差分析(One-way ANOVA)不同处理结果间的显著性差异($P<0.05$)。

2 结果与讨论

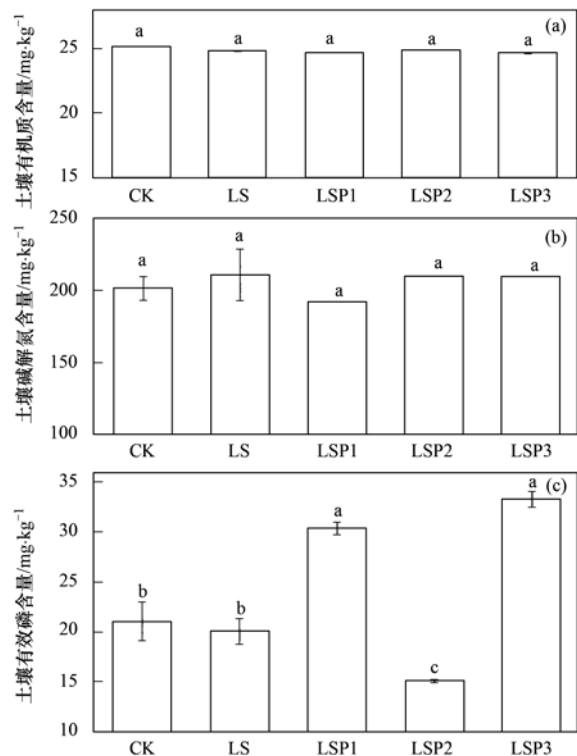
2.1 施用钙镁磷肥对石灰、海泡石组配修复下土壤理化性质的影响

石灰、海泡石和钙镁磷肥均是碱性物质,可提高酸性土壤的 pH 值,改善土壤性质. LS、LSP1、LSP2 和 LSP3 处理下土壤 pH 值相对于 CK 处理(6.6)均显著提高 1 个 pH 值单位以上($P < 0.05$). 施用钙镁磷肥,土壤 pH 值和有机质含量呈缓慢升高趋势,但对石灰、海泡石组配改变土壤 pH 值无显著性影响,土壤中有机质含量无显著性变化.

土壤碱解氮含量和有效磷含量是反映土壤肥力的一个重要指标^[18,19],图 1(b)表示不同处理下土壤碱解氮含量的变化情况. 除 LSP1 处理外,加入改良剂的 LS、LSP2 和 LSP3 处理相对于 CK 处理土壤碱解氮含量均有提高,分别提高 4.45%、4.10% 和 4.10%,而 LSP1 相对于 CK 处理土壤碱解氮含量减少 4.57%,LS、LSP2 和 LSP3 这 3 个处理之间基本无差异. 由图 1(c)可知,石灰、海泡石组配(LS)对土壤有效磷含量影响不大,相比于 CK 处理,有效磷含量略微降低了 4.63%. 钙镁磷肥的加入,LSP1 和 LSP3 处理相比于 CK 和 LS 均显著提高($P < 0.05$)了土壤有效磷含量,与 LS 处理相比,LSP1 和 LSP3 土壤有效磷含量分别显著($P < 0.05$)提高 51.46% 和 65.96%,但 LSP2 处理下,土壤有效磷含量却显著($P < 0.05$)下降了 24.79%,这可能是在 LSP2 的钙镁磷肥用量下水溶性磷酸根与土壤中 Cd^{2+} 通过沉淀或络合反应生成磷酸镉等难溶性磷酸盐;或在这一浓度下可能更有利于水稻对磷的吸收;也可能是土壤中可溶性磷酸盐通过吸附作用与有机质结合,导致土壤中有效磷含量降低.

2.2 施用钙镁磷肥对石灰、海泡石组配修复下土壤 Cd 赋存形态及其有效性的影响

Cd 对生物毒害作用的大小主要与酸可提取态含量有关^[20],采用 BCR 法连续提取土壤中 Cd 的赋存形态. 从图 2 可以看出,CK 处理土壤中 Cd 主要以酸可提取态和可还原态形式存在,分别占比 32.47% 和 59.70%,而可氧化态与残渣态占比较低,一共仅 7.83%,表明 CK 处理土壤中 Cd 的生物毒性较大,这可能与该处理下土壤呈酸性有关. 与 CK 处理相比,石灰、海泡石组配(LS)显著降低($P < 0.05$)了土壤中酸可提取态、可还原态和可氧化态 Cd 占比,分别降低 82.13%、81.45% 和 65.55%,残渣态 Cd 占比显著($P < 0.05$)提高 1 507.59%,与 Hamid 等^[21]的研究结果一致,这可能是因为石灰、海泡石施用提高了土壤 pH 值,促进土壤中 Cd^{2+} 与



不同字母表示具有显著性差异($P < 0.05$),下同
图 1 不同处理下土壤有机质含量、碱解氮含量以及有效磷含量变化情况

Fig. 1 Content changes of soil organic matter, alkaline nitrogen and available phosphorus under different treatments

硅酸盐和氢氧根等形成沉淀从而增加土壤对 Cd 的吸附^[22],也可能是海泡石通过离子交换或络合的方式与 Cd^{2+} 结合,以及石灰、海泡石中含有的 Ca^{2+} 与 Cd^{2+} 发生共沉淀从而增加土壤中残渣态 Cd 的占比,减小 Cd 的生物可利用性^[23]. 施加钙镁磷肥,土壤中 Cd 主要存在形式为残渣态,其次为酸可提取态和可还原态,可氧化态最低,与 LS 处理相比,施用 $1125 \text{ kg} \cdot \text{hm}^{-2}$ 钙镁磷肥(LSP1),土壤中酸可提取态 Cd 占比提高 21.03%,可还原态和可氧化态 Cd 占比分别显著($P < 0.05$)提高 112.28% 和 20.43%,而残渣态显著($P < 0.05$)降低 16.82%;施用 $2250 \text{ kg} \cdot \text{hm}^{-2}$ 钙镁磷肥(LSP2),土壤中酸可提取态、可还原态和可氧化态 Cd 占比分别显著($P < 0.05$)降低 26.72%、47.15% 和 18.27%,而土壤残渣态 Cd 占比显著($P < 0.05$)升高 8.44%;施用 $3375 \text{ kg} \cdot \text{hm}^{-2}$ 钙镁磷肥(LSP3),土壤中酸可提取态和可还原态 Cd 占比分别降低 6.37% 和 12.37%,可氧化态 Cd 占比显著($P < 0.05$)降低 16.12%,而土壤残渣态 Cd 占比升高 2.29%. 总的来说,除了 LSP1 处理,钙镁磷肥的施用均能减少酸可提取态、可还原态和可氧化态 Cd 的占比,降低 Cd 的生物毒性,与吴文成等^[24]的研究结果相一致.

有研究表明,土壤中有效态 Cd 含量与土壤中

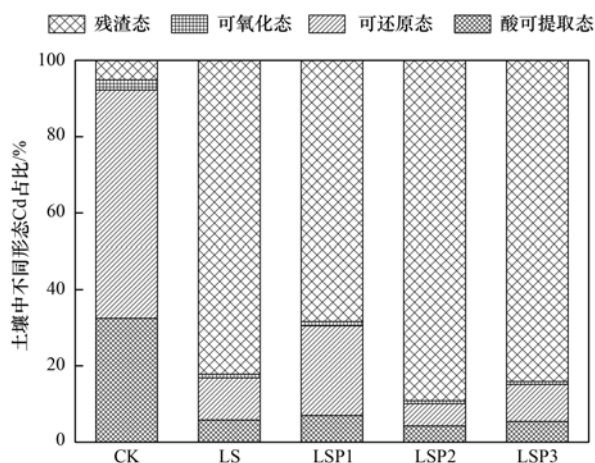


图2 土壤中Cd形态的占比情况

Fig. 2 Percentages of Cd forms in soil

磷酸根含量以及其它阳离子的存在密切相关^[21]. 由图3可知,CK处理土壤中有有效态Cd含量与其余加入改良剂的4个处理相比均有显著性差异($P < 0.05$). 与CK处理相比,石灰、海泡石组配(LS)土壤中有有效态Cd含量显著($P < 0.05$)降低43.79%,这与前文石灰、海泡石组配(LS)显著降低土壤中酸可提取态、可还原态和可氧化态Cd占比的原因一致. 施加不同浓度的钙镁磷肥,对石灰、海泡石组配降低土壤中有有效态Cd含量影响效果不同,但均低于CK处理土壤中有有效态Cd含量. 与LS处理相比,施用1125 kg·hm⁻²钙镁磷肥(LSP1),土壤中有有效态Cd含量显著($P < 0.05$)增加50.36%,这可能是钙镁磷肥在这一用量下,其本身Cd含量大于土壤中磷酸根固定的Cd含量,导致土壤有效态Cd含量升高;施用2250 kg·hm⁻²钙镁磷肥(LSP2),土壤中有有效态Cd含量显著($P < 0.05$)降低46.97%,这可能是因为在该处理下,钙镁磷肥中水溶性磷酸根离子(PO_4^{3-} 、 HPO_4^{2-} 、 H_2PO_4^-)与土壤中 Cd^{2+} 通过沉淀或络合作用生成磷酸镉而减少土壤中有有效态Cd含量^[25];施用3375 kg·hm⁻²钙镁磷肥(LSP3),土壤中有有效态Cd含量降低9.79%,表明该处理下,磷酸根通过沉淀或络合作用减少的有效态Cd含量与钙镁磷肥本身带入的Cd含量可基本抵消,因此,其土壤中有有效态Cd降低效果反而不如LSP2处理. 施用钙镁磷肥可改变土壤中有有效态Cd含量,2250 kg·hm⁻²钙镁磷肥(LSP2)可显著($P < 0.05$)促进石灰、海泡石组配对土壤中Cd的钝化效果.

2.3 施用钙镁磷肥对石灰、海泡石组配修复下水稻各部位镉分布的影响

不同处理下水稻各部位Cd含量情况见图4. CK处理水稻各部位Cd含量均显著($P < 0.05$)高出除LSP1处理外的其它处理25.80%~875%,LSP1中

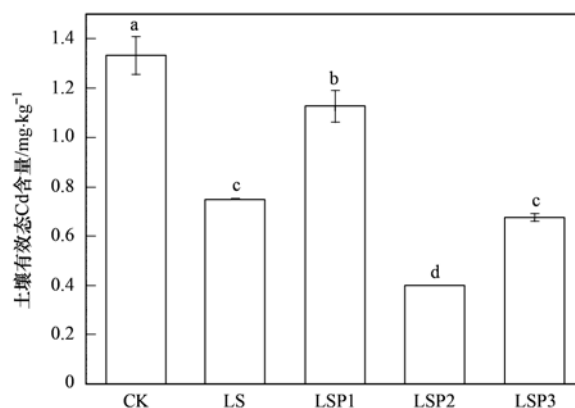


图3 不同处理下土壤中有有效态Cd含量的变化情况

Fig. 3 Content changes of available Cd in soil under different treatments

Cd含量高于CK处理0%~30.76%. 石灰、海泡石组配(LS)相对于CK处理,水稻根、茎叶和糙米中Cd含量均显著($P < 0.05$)降低58.96%、54.28%和20.51%,但糙米中Cd含量仍高于《食品安全国家标准》(GB 2762-2017)中糙米镉限量(0.2 mg·kg⁻¹)标准. 施用钙镁磷肥后,石灰、海泡石组配对水稻根、茎叶和糙米中Cd含量的影响随钙镁磷肥用量的不同而不同,LSP1处理水稻根、茎叶和糙米中Cd含量显著高于LS处理($P < 0.05$),分别高出165.51%、118.75%和65.24%;LSP2处理水稻根和茎叶中Cd含量低于LS处理,分别降低11.95%和47.50%,糙米中Cd含量相对于LS处理,显著降低87.09%($P < 0.05$);LSP3处理相对于LS处理,糙米中Cd含量显著($P < 0.05$)降低22.58%. 加入钙镁磷肥的3个处理中,仅LSP2处理糙米中Cd含量0.04 mg·kg⁻¹低于《食品安全国家标准》(GB 2762-2017)中糙米镉限量(0.2 mg·kg⁻¹)标准,结果表明,在中等浓度(2250 kg·hm⁻²)钙镁磷肥辅助下,石灰、海泡石组配可确保Cd污染农田边修复边安全生产的要求.

图4(d)表示不同处理下水稻糙米干重变化情况,石灰和海泡石组配(LS)使水稻糙米干重相对于CK处理增加15.01%,这可能是因为石灰、海泡石的施用提高了土壤pH值,使土壤有效态Cd含量降低,减小了Cd对水稻的毒害作用. 与LS处理相比,施用1125 kg·hm⁻²钙镁磷肥(LSP1),水稻糙米干重减少4.67%,这可能是因为相对于LS处理其土壤中含有更多的有效态Cd,对水稻产生毒害作用,使水稻减产;施用2250 kg·hm⁻²钙镁磷肥(LSP2),水稻糙米干重增加28.34%,这可能是因为该浓度下的钙镁磷肥使土壤中有有效态Cd含量相对于LS处理显著($P < 0.05$)降低46.97%(图3),显著减小了Cd对水稻的毒害作用,使糙米产量增加;施用3375

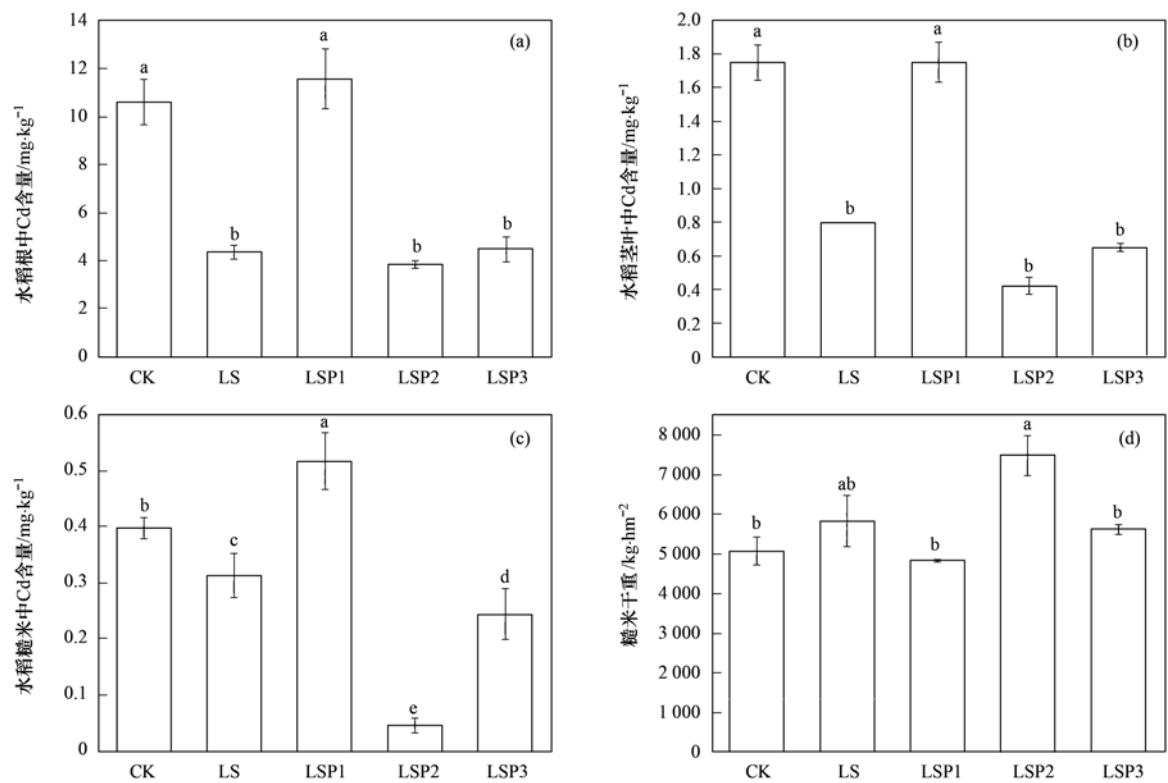


图4 不同处理下水稻根、茎叶和糙米中Cd含量情况以及糙米干重变化情况

Fig. 4 Cd content in rice roots, straw, and brown rice and dry weight of brown rice under different treatments

kg·hm⁻²钙镁磷肥(LSP3),水稻糙米干重减少3.74%,这可能与该条件下土壤中有效态Cd含量密切相关.总的来说,中等浓度(2 250 kg·hm⁻²)钙镁磷肥的施用不仅能够很好地促进石灰、海泡石组配对土壤Cd的钝化,保证糙米中Cd含量低于《食品安全国家标准》(GB 2762-2017)中糙米镉限量(0.2 mg·kg⁻¹)标准,还能保证水稻产量,很好地实现Cd污染稻田的安全生产和增产.

表2为水稻各部位Cd含量与土壤pH值、有效磷和有效态Cd含量的相关性分析.分析结果表明,土壤有效态Cd含量和土壤pH值呈极显著负相关关系($P < 0.01$),水稻根、茎叶中Cd含量和土壤

pH值呈显著负相关关系($P < 0.05$),水稻根、茎叶和糙米中Cd含量与土壤有效态Cd含量均呈极显著正相关关系($P < 0.01$),土壤中有有效态Cd含量对抑制水稻吸收Cd非常关键.此外,水稻糙米中Cd含量与根、茎叶中Cd含量呈极显著正相关关系($P < 0.01$),说明糙米中Cd含量会随着水稻根、茎叶中Cd含量的降低而降低.水稻根、茎叶和糙米中Cd含量以及土壤有效态Cd含量与土壤有效磷含量均呈正相关关系,但均无显著性差异,该结果可能是由于水溶性磷酸根与土壤中有有效态Cd反应生成难溶性磷酸镉,使土壤中有有效磷和有效态Cd含量同时降低.

表2 水稻各部位Cd含量与土壤pH值、有效磷含量和有效态Cd含量的相关性¹⁾

Table 2 Correlation between Cd content in various parts of rice and soil pH, available phosphorus content and available Cd content

	pH 值	土壤有效磷	土壤有效态 Cd	根中 Cd	茎叶中 Cd	糙米中 Cd
pH 值	1					
土壤有效磷	0.224	1				
土壤有效态 Cd	-0.736 **	0.254	1			
根中 Cd	-0.567 *	0.298	0.876 **	1		
茎叶中 Cd	-0.541 *	0.313	0.913 **	0.945 **	1	
糙米中 Cd	-0.349	0.510	0.846 **	0.841 **	0.838 **	1

1) * 表示 $P < 0.05$; ** 表示 $P < 0.01$

3 结论

(1)施用钙镁磷肥改善了土壤理化性质,与石灰、海泡石组配处理相比,LSP1 和 LSP3 处理显著

提高($P < 0.05$)了土壤有效磷含量,分别提高51.46%和65.96%.在施用2 250 kg·hm⁻²钙镁磷肥的条件下,大部分水溶性磷酸根通过与土壤中Cd²⁺发生沉淀或络合反应生成难溶性磷酸镉降低土壤

Cd 的有效性.

(2) 与石灰、海泡石组配处理相比,施用2 250 kg·hm⁻²钙镁磷肥,可促进土壤酸可提取态、可还原态和可氧化态 Cd 转化为残渣态 Cd,使土壤残渣态 Cd 占比显著($P < 0.05$) 升高 8.44%,而土壤酸可提取态、可还原态和可氧化态 Cd 占比分别显著($P < 0.05$) 降低 26.72%、47.15% 和 18.27%;土壤中有有效态 Cd 含量显著($P < 0.05$) 降低 46.97%.

(3) 在镉浓度为 4.98 mg·kg⁻¹ 的稻田土壤上,施用2 250 kg·hm⁻²钙镁磷肥,协助石灰、海泡石组配使糙米中 Cd 含量降低至 0.04 mg·kg⁻¹,低于《食品安全国家标准》(GB 2762-2017) 中糙米镉限量(0.2 mg·kg⁻¹) 标准,此外,还增加了糙米产量.水稻糙米中 Cd 含量与土壤中有有效态 Cd 含量呈极显著正相关关系($P < 0.01$),土壤中有有效态 Cd 含量是影响水稻糙米中 Cd 含量的关键因素之一.

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