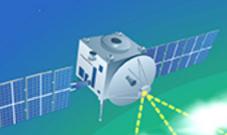


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ENVIRONMENTAL SCIENCE

ISSN 0250-3301 CODEN HCKHDV HUANJING KEXUE

PM_{2.5}和O₃污染协同防控区的遥感精细划定与分析 李沈鑫,邹滨,张凤英,刘宁,薛琛昊,刘婧



O₃ PM_{2.5}

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PM_{2.5}



- 主办 中国科学院生态环境研究中心
- ■出版斜学出版社





2022年10月

第43卷 第10期 Vol.43 No.10

ENVIRONMENTAL SCIENCE

第43卷 第10期 2022年10月15日

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无机钝化剂对镉污染酸性水稻土的修复效果及其机制

张剑1,孔繁艺2,卢升高2*

(1. 浙江省温州市植物保护与土壤肥料管理站,温州 325000; 2. 浙江大学环境与资源学院,杭州 310058)

摘要:以浙江南部重金属镉(Cd)轻度污染酸性稻田为对象,以当地应用最广泛的 3 种无机钝化剂(硅钙镁钾肥、钙镁磷肥和石灰)为材料,通过田间试验研究了不同用量(750、1500和2250 kg·hm $^{-2}$)钝化剂阻控土壤酸化与稻米 Cd 积累的效果与化学机制.结果表明,3 种钝化剂可有效地改良土壤酸化和降低水稻稻米 Cd 积累,施用2250 kg·hm $^{-2}$ 硅钙镁钾肥、钙镁磷肥和石灰分别增加土壤 pH 值 0.62、0.65 和 0.86 单位,减少交换性酸总量 67%、69% 和 78%,降低糙米镉含量 73%、68% 和 77%.施用2250 kg·hm $^{-2}$ 钝化剂可使 Cd 轻度污染稻田上种植水稻糙米中 ω (Cd)低于 0.2 mg·kg $^{-1}$,达到国家食品安全标准;与对照比较,3 种钝化剂均显著降低土壤中 DTPA 提取有效态镉含量,降低弱酸提取态(F1)和可还原态(F2)Cd含量,增加残渣态(F4)Cd含量;相关分析表明糙米 Cd含量与土壤 pH 与交换性阳离子含量呈显著负相关,与 DTPA-Cd、F1-Cd、F2-Cd 和交换铝含量呈显著正相关.应用最小二乘路径模型分析了糙米 Cd含量、Cd有效性和化学形态与土壤性质的关系,土壤交换性阳离子对糙米 Cd含量、有效镉和水稻产量直接影响的路径系数分别为-0.566、-0.866 和 0.873,土壤 pH 主要通过直接影响有效镉而间接影响糙米镉含量.田间试验证明,这 3 种钝化剂是实现镉污染酸性水稻土水稻安全生产的有效技术,它们主要通过影响土壤交换性阳离子而直接影响土壤镉生物有效性,进而影响糙米中镉的积累,研究结果可为受污染耕地水稻安全生产和酸化土壤改良提供科学依据.

关键词:钝化剂; pH; 有效镉; 交换性阳离子; 偏最小二乘路径模型 (PLS-PM)

中图分类号: X53 文献标识码: A 文章编号: 0250-3301(2022)10-4679-08 DOI: 10.13227/j. hjkx. 202112273

Remediation Effect and Mechanism of Inorganic Passivators on Cadmium Contaminated Acidic Paddy Soil

ZHANG Jian¹, KONG Fan-yi², LU Sheng-gao²*

(1. Wenzhou Plant Protection and Soil Fertilizer Management Station, Zhejiang Province, Wenzhou 325000, China; 2. College of Environmental and Resource Sciences, Zhejiang University, Hangzhou 310058, China)

Abstract: Cadmium (Cd) is one of the main pollutants in acidic paddy fields, and its accumulation in rice (Oryza sativa L.) and subsequent transfer to the food chain is an important environmental issue in China. In our field study, three types of inorganic passivators (silicon-calcium-magnesium-potassium fertilizer (SCMK), calcium magnesium phosphate fertilizer (CMP), and lime (L) at the rate of 750, 1500, and 2 250 kg·hm⁻², respectively) were applied to acidic paddy soils polluted by the heavy metal Cd in southern Zhejiang province. The objective of this study was to reveal the effects and chemical mechanisms of passivators on soil acidification and Cd accumulation in rice. The field experimental results showed that the three passivators could effectively improve soil acidification and reduce Cd accumulation in rice grains. The application of 2 250 kg·hm⁻² SCMK, CMP, and L increased soil pH by 0.62, 0.65, and 0.86 units; decreased exchangeable acidity by 67%, 69%, and 78%; and reduced the content of Cd in brown rice by 73%, 68%, and 77%, respectively. The application of 2 250 kg·hm⁻² SCMK, CMP, and L reduced the content of Cd in brown rice planted on polluted paddy rice fields to lower than 0.2 mg·kg⁻¹, which reached the national food safety standard. Compared with the control, the application of SCMK, CMP, and L significantly (P<0.05) decreased the content of available Cd extracted by DTPA; decreased the contents of weak acid-extractable (F1) and reducible (F2) Cd; and increased the content of residual (F4) Cd. Correlation analyses indicated that Cd content in brown rice was significantly negatively correlated with soil pH and exchangeable cation content and significantly positively correlated with DTPA-Cd, weak acid-extractable (F1) and reducible (F2) Cd, and exchangeable Al contents. The partial least squares path model (PLS-PM) was used to analyze the relationship between the Cd content of brown rice, DTPA-Cd, and various chemical forms of Cd and soil properties. The direct path coefficients of soil exchangeable cations on Cd content in brown rice, available cadmium, and rice yield were -0.566, -0.866, and 0.873, respectively. Soil pH indirectly affected Cd content of brown rice mainly by directly affecting available Cd in soil. Field experiments demonstrated that the three passivators SCMK, CMP, and L were effective technologies for the safe production of rice in acidic paddy soils polluted by Cd. The possible mechanism for passivators reducing the bioavailability of Cd in soil and its accumulation in brown rice contributed to increased exchangeable cations in the soils. These findings could provide a scientific basis for the safe production of rice in acidic paddy soil polluted by heavy metals.

Key words: passivator; pH; bioavailable Cd; exchangeable cations; partial least squares path model (PLS-PM)

水稻(*Oryza sativa* L.)是我国最主要的粮食作物,它对土壤中的镉(Cd)有较强的富集能力,导致稻米容易发生 Cd 污染.由于南方地区稻田土壤普遍酸化加剧了稻米 Cd 污染,导致"镉米"问题频发^[1].如何治理 Cd 污染农田,保障稻米食品安全是我国农产品安全的重要内容.目前,Cd 污染稻田水稻安全生产的主要措施是采用低 Cd 积累水稻品

种、原位钝化、水分管理和优化施肥等技术^[2]. 其中低 Cd 积累水稻品种和原位钝化技术结合是目前

收稿日期: 2021-12-27; 修订日期: 2022-01-25

基金项目: 浙江省受污染耕地安全利用试点项目; 国家重点研发计划项目(2016YFD0800401)

作者简介: 张剑(1975~),男,硕士,高级农艺师,主要研究方向为受污染耕地治理与安全利用,E-mail;wznyz@126. com

* 通信作者,E-mail:lusg@zju.edu.cn

广泛推荐的技术,石灰等无机钝化剂以成本低、效果好和二次污染少的优势在污染土壤修复实践中得到广泛应用.

有田间和盆栽试验研究证明^[3,4],石灰等无机钝化剂可以显著提高土壤 pH,并降低土壤有效态 Cd 含量和作物各部位 Cd 含量,一定程度上增加作物产量; 硅钙镁钾肥作为近年来新兴的一种富含 Si、Ca、Mg和 K 的碱性矿质肥料,其施用可以显著增加土壤 pH和交换性阳离子,并将镉转变为更稳定的形态,减少镉的生物富集系数,并满足作物对钙、镁、硅元素的吸收利用^[5,6];钙镁磷肥由于强碱性和富含 Ca 和 Mg等元素广泛应用于重金属污染土壤修复^[7,8],它们是浙江南部受污染耕地安全利用中的主要推荐产品.由于土壤酸化往往提高土壤中镉的生物有效性,促进水稻对 Cd 的吸收^[9].有研究表明^[9,10],pH 越低土壤中重金属越活跃,农作物 Cd 越易超标.因此,酸化土壤中 Cd 污染修复必须考虑治理酸化,从根本上控制土壤中 Cd 的生物有效性.

根据文献[11],我国已经全面开展了受污染耕地安全利用工作,农业部门也针对污染稻田安全利用推荐了数种钝化剂,但存在钝化机制不明确且缺乏合理的钝化剂施用量等问题.本研究以浙江地区广泛应用的3种无机钝化剂为材料,通过大田试验探讨了不同钝化剂用量对 Cd 污染酸化稻田的修复效果,分析钝化剂对水稻产量、土壤酸度、交换性阳离子、土壤镉有效性与形态和稻米镉含量的影响,通过相关分析和偏最小二乘路径模型(PLS-PM)明确影响 Cd 生物有效性及其在水稻中积累的土壤性质,通过探明无机钝化剂改良土壤酸化和治理重金属污染的机制,以期为稻田 Cd 污染耕地安全利用与酸化治理提供科学依据.

1 材料与方法

1.1 试验材料

田间试验在浙江省南部某污染酸性水稻田,试验田表层土壤(0~20 cm) pH 为 5. 19, ω (全氮)为 2. 1 g·kg⁻¹, ω (速效磷)为 10. 51 mg·kg⁻¹, ω (黏粒)为 234. 3 g·kg⁻¹, ω (有机碳)为 19. 33 g·kg⁻¹, ω (土壤总 Cd)为 0. 61 mg·kg⁻¹. 试验土壤总 Cd 含量高于生态环境部规定的风险筛选值^[12],农产品监测表明稻米 Cd 超标,存在农产品污染风险. 根据国家土壤环境质量类别划分,该田块为安全利用类耕地. 本试验所用 3 种纯化剂为当地农业部门受污染耕地安全利用主推的土壤调理剂,分别是硅钙镁钾肥,钙镁磷肥和石灰. 本试验用材料均为市场成品,其中硅钙镁钾肥主要成分为 CaSiO₃、Ca₃(PO₄)₂和 MgSiO₃.

1.2 试验设计与样品采集

本文采用田间小区试验,共设30个小区,每个 小区面积为30 m²,小区之间用田埂分隔,田埂用两 层黑色塑料薄膜扎住,防止小区之间水分和肥料交 换. 每种钝化剂采用 3 个施用量(750、1500和2250 kg·hm⁻²),分别用Ⅰ、Ⅱ和Ⅲ表示,每个处理设3个 重复,随机区组设计. 钝化剂在水稻播种前 15 d 施 入耕层翻耕后充分混匀,按照常规水稻种植季节播 种、插秧、施肥和收割.水稻灌浆-蜡熟期保持田面 水位3~5 cm, 蜡熟期后排干水分. 其他田间管理和 防病虫害措施相同. 试验水稻品种为前期筛选出的 低积累水稻品种——甬优 15. 水稻成熟后每个小区 单独测产,测定每个小区的实际产量,并同步采集土 壤和水稻样品. 土壤样品按小区内多点随机取样法 采集,经自然风干磨细过筛备用.水稻样品用超纯水 清洗后,在烘箱于105℃下杀青30 min,籽粒部分在 70℃烘干,脱粒去壳后制得糙米样品,研磨粉碎过 100 目筛保存备用.

1.3 土壤性质分析方法

土壤基本理化性质采用常规分析方法^[13]. 土壤 pH(土水比为1:2.5)采用 pH 计(PB-21, Sartorius) 测定,交换性酸采用氯化钾交换-中和滴定法,有机质的测定采用重铬酸钾外加热法,全氮采用半微量开氏法,速效磷为碳酸氢钠浸提-钼锑抗比色法(Olsen 法),土壤颗粒分析采用吸管法. 土壤交换性阳离子采用乙酸铵(pH 7.0)浸提-等离子体原子发射光谱测定(ICP-AES).

1.4 土壤重金属分析方法

土壤总 Cd 采用 HNO,-HF-H,O, (6:2:2, 体积 比)-微波消解法(MASTER40 SINEO,中国),土壤有 效态 Cd 采用二乙烯三胺五乙酸-氯化钙-三乙醇胺 (DTPA-CaCl,-TEA)缓冲溶提取. 糙米采用 HNO,-H₂O₂微波消解法. 土壤中重金属形态分析参照欧盟 标准物质局 BCR 连续提取法[14],具体为:弱酸提取 态或可交换态及碳酸盐结合态(F1)采用 0.11 mol·L⁻¹ CH₃COOH 提取,可还原态或铁锰氧化物结 合态(F2)为 0.5 mol·L⁻¹ NH₂OH·HCl提取,可氧化 态或有机物及硫化物结合态(F3)为 0.02 mol·L-1 HNO_3 (30% H_2O_2) + 3.2 $mol \cdot L^{-1} NH_4 OAc$ (20% HNO₃)(pH 2.0)提取,残渣态(F4)为 HNO₃-HF-HCl溶解. 提取液中 Cd浓度采用石墨炉原子吸收光 谱仪(PerkinElmer AAnalyst 800)测定. 土壤和糙米 分析过程采用标准物质 GBW07405 (GSS-5)和 GBW10044 (GSB-22)作质量控制.

1.5 数据处理与分析

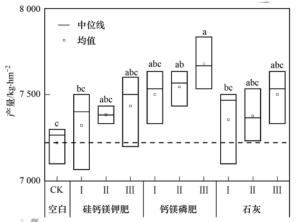
数据整理和统计分析分别采用 Excel 2020 和

SPSS 26.0,采用单因素方差分析(ANOVA),处理间的差异用 Duncan 法作显著性检验(P < 0.05 和 P < 0.01). 可视化分析作图采用 origin 2022 和 R (4.1.2),其中偏最小二乘路径模型分析用"plspm"包分析,相关性分析用"corrplot"包分析,方法采用皮尔逊相关性分析.

2 结果与分析

2.1 无机钝化剂对水稻产量的影响

无机钝化剂对水稻产量的影响见图 1. 结果表明施用钝化剂对水稻产量具有一定的增产效应,水稻产量随着钝化剂用量增加而增加,但与对照比较,钝化剂硅钙镁钾肥和石灰对水稻产量的影响未达显著水平,钙镁磷肥 II 和 III 处理组水稻产量较对照组分别提高 4.5% 和 6.3%,钙镁磷肥用量达1500kg·hm⁻²以上时,水稻产量显著增加.



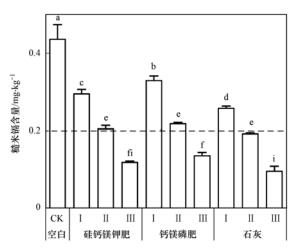
不同小写字母表示处理间差异显著(P < 0.05); I 表示施用量为 750 kg·hm⁻², II 表示施用量为 1500 kg·hm⁻², III 表示施用量为 250 kg·hm⁻²; 虚线为空白组产量的平均值

图 1 无机钝化剂对水稻产量的影响

Fig. 1 Effect of inorganic passivators on rice yield

2.2 无机钝化剂对水稻籽粒 Cd 含量的影响

不同钝化剂对水稻糙米镉含量影响见图 2. 对照处理的糙米 ω (Cd) 为 0. 44 mg·kg⁻¹,超过《食品安全国家标准食品中污染物限量》(GB 2762-2017)中对糙米中 Cd 污染物的限量标准(0.2 mg·kg⁻¹),稻米 Cd 严重超标.施用钝化剂后稻米 Cd 含量显著降低,钝化剂用量达2 250 kg·hm⁻²时,糙米 Cd 含量降至 0.2 mg·kg⁻¹以下,符合国家食品安全标准.与对照比较,钝化剂用量为2 250 kg·hm⁻²时,3 种钝化剂降低糙米镉含量 68%~70%.本试验的3种纯化剂降低糙米 Cd 的效果以石灰较好,硅钙镁钾肥次之.田间小区试验表明,研究区轻度 Cd 污染酸性水稻田要实现水稻安全生产的钝化剂用量需在2 250 kg·hm⁻²以上.



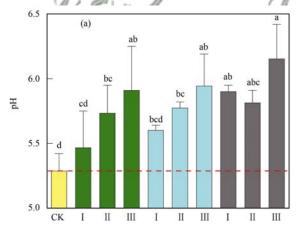
不同小写字母表示处理间差异显著(P<0.05);虚线为《食品安全国家标准食品中污染物限量》(GB 2762-2017)中对糙米中Cd污染物的限量标准($0.2~\text{mg·kg}^{-1}$)

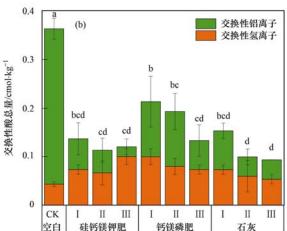
图 2 无机钝化剂对糙米中 Cd 含量的影响

Fig. 2 Effect of inorganic passivators on the content of Cd in brown rice

2.3 无机钝化剂对土壤 pH 和酸度的影响

不同钝化剂处理对土壤 pH 值和交换性酸的影响见图 3. 结果表明, 3 种无机钝化剂施用明显提高





不同小写字母表示处理间差异显著(P<0.05); 红色虚线表示空白组中 pH 的均值

图 3 不同无机钝化剂对土壤酸度的影响

Fig. 3 Effect of inorganic passivators on soil acidity

土壤 pH,但750 kg·hm⁻²施用量的钙镁磷肥和硅钙镁钾肥对提高土壤 pH 的效果未达显著差异.与对照比较,2250 kg·hm⁻²硅钙镁钾肥、钙镁磷肥和石灰分别提高土壤 pH值0.62、0.65和0.86个单位.石灰提高土壤 pH值的效果优于钙镁磷肥和硅钙镁钾肥[图3(a)].不同钝化剂处理对土壤交换性酸和交换性铝离子的影响表明[图3(b)],钝化剂显著地降低土壤的交换性酸(EA)和交换性铝离子(EAl)含量.与对照比较,钝化剂用量为2250 kg·hm⁻²时,硅钙镁钾肥、钙镁磷肥和石灰分别降低交换性酸67%、69%和78%,交换性铝含量94%、81%和88%.然而,钝化剂增加土壤交换性氢离子(EH)含量.

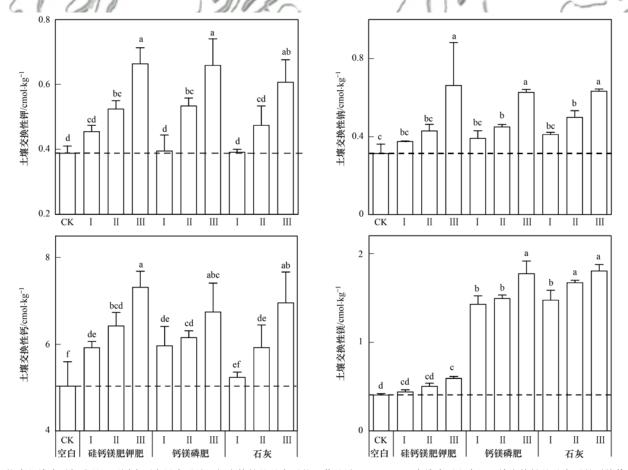
2.4 无机钝化剂对土壤交换性离子组成的影响 不同无机钝化剂处理对土壤交换性盐基离子的 影响见图 4,与对照比较,3种钝化剂施用都不同程度地增加了各盐基离子含量,且随着钝化剂用量的增加,交换性盐基离子含量增多.3种钝化剂用量为750 kg·hm⁻²时,土壤中交换性钾含量没有显著增加,当用量为1500 kg·hm⁻²和2250 kg·hm⁻²时,钙镁磷肥和硅钙镁钾肥处理显著增加交换性钾含量,而石灰只在2250 kg·hm⁻²时显著增加;当钝化剂用量达2250 kg·hm⁻²时,3种钝化剂均显著增加交换性钠、交换性钙和镁离子含量,750 kg·hm⁻²和1500 kg·hm⁻²硅钙镁钾肥处理未能显著增加土壤交换性镁离子.不同无机钝化剂对土壤 pH、交换性酸和交换性阳离子影响的 F 检验表明(表1),钝化剂对 pH、交换性酸总量(EA)、EAl、交换性 K、Na和 Ca有极显著影响.

表 1 不同无机钝化剂对土壤 pH、交换性酸和交换性阳离子影响的 F 检验 $^{1)}$

Table 1 The F-test on the effects of inorganic passivators on soil pH, exchangeable acid, and exchangeable cations in soils

项目	рН	EH	EAl	EA	K	Na 🦰	Ca Mg	Total
硅钙镁钾肥	6 **	3.8*	47. 9 **	23. 5 **	16 **	14. 4 ** 16.	5 ** 2. 8	130. 9
钙镁磷肥	6. 1 **	3.8*	32. 6 **	15. 5 **	19. 1 **	11 ** 9.	1 ** 145. 2 **	242, 6
石灰	10. 4 **	1.1	44. 7 **	26 **	12. 1 **	11. 4 ** 13.	7 ** 164 **	283. 4

1) F 值是显著性差异的水平,值越大,处理的效果越显著; *表示P < 0.05; **表示P < 0.01,下同



柱中竖线表示标准差;不同小写字母表示处理间交换性盐基离子的显著差异(P < 0.05);虚线表示空白组土壤交换性盐基离子的平均值

图 4 不同无机钝化剂对土壤交换性盐基离子的影响

Fig. 4 Effect of inorganic passivators on exchangeable base in soils

2.5 无机钝化剂对土壤 Cd 有效性与化学形态的 影响

图 5 是不同无机钝化剂处理对土壤 DTPA 提取 态镉含量的影响,与 CK 比较,无机钝化剂的施用显 著降低土壤中有效态镉含量,并且随着钝化剂施用 量增加,土壤中 DTPA 提取的有效态镉含量显著减 少. 当钝化剂用量达2 250 kg·hm⁻²时,硅钙镁钾肥、 钙镁磷肥和石灰处理分别降低 DTPA-Cd 含量 28%、 28%和31%.

不同钝化剂处理对土壤重金属 Cd 形态的影响 见图 6. 结果表明, 钝化剂显著降低了土壤中弱酸提 取态(F1)和可还原态(F2)Cd含量,显著增加了土 壤中残渣态(F4)Cd含量,表明3种钝化剂明显改 变了土壤中 Cd 的化学形态,将易溶性的 F1 和 F2 形态 Cd 转变为难溶性的 F4 形态. 例如,施用石灰 降低 F1 含量 20.8%~34.6%,降低 F2 含量 16.5% ~20.7%,增加 F4 含量 12.4%~14.2%.结果表明 (表2),无机钝化剂可以显著改变土壤中不同化学 形态镉的分布,降低可交换态镉含量,形成更稳定的 残渣态.

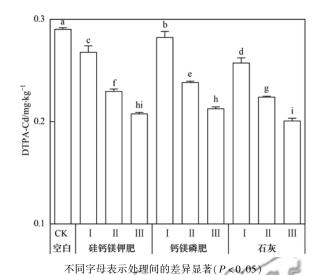


图 5 不同无机钝化剂处理对 DTPA-Cd 含量的影响

Effect of inorganic passivators on the content of DTPA-extractable Cd in soils

表 2 不同无机改良剂对根际土壤镉形态和稻谷镉含量 F 检验的统计量

Table 2 The F-test on the effects of inorganic modifiers on cadmium forms in rhizosphere soil and cadmium content in rice

项目 DTPA-Cd Grain-Cd F1 F2 F3 F4 Total 硅钙镁钾肥 201.528 ** 148.971 ** 20.281 ** 9.714 ** 20.434 ** 7.412 ** 408.34	
	1
钙镁磷肥 205 ** 139. 249 ** 21. 277 ** 5. 293 ** 6. 483 ** 8. 213 ** 385. 515	15 //
石灰 233.75 ** 165.608 ** 34.193 ** 23.583 ** 3.175 * 54.743 ** 515.052	52

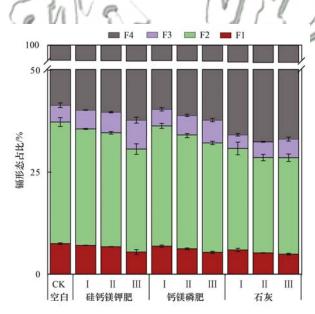


图 6 不同无机钝化剂对土壤中 Cd 化学形态的影响

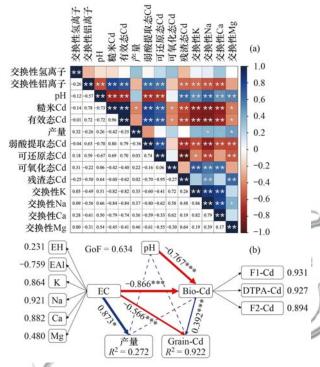
Fig. 6 Effect of inorganic passivators on chemical forms of Cd in soils

2.6 糙米 Cd 含量与土壤 Cd 有效性、化学形态和 土壤性质的相关性分析

水稻糙米 Cd 含量与土壤中 Cd 有效态、化学形 态和土壤性质的相关性分析见图 7(a). 糙米 Cd 含 量(Grain-Cd)与 DTPA-Cd、F1-Cd 和 F2-Cd 呈显著

正相关,而 DTPA-Cd 与 F1-Cd、F2-Cd 呈显著正相 关,与F3-Cd、F4-Cd、交换性K、Na、Ca和Mg呈显 著负相关. 土壤 pH 与 Grain-Cd、DTPA-Cd、F1-Cd 和 F2-Cd 呈显著负相关,与 F4-Cd、交换性 K、Na、 Ca和 Mg 呈显著正相关. 土壤交换性铝离子(EAI) 与 pH、F4-Cd、土壤交换性 K、Na 和 Ca 呈显著负相 关,与 Grain-Cd、DTPA-Cd、F1-Cd、F2-Cd 呈显著正 相关. F1-Cd 和 F2-Cd 与 F4-Cd、交换性 K、Na 和 Mg 呈显著负相关. 相关性分析表明,土壤 DTPA-Cd 与土壤 pH 呈显著负相关(r = -0.72**),与糙米 Cd 含量呈显著正相关(r=0.96**),表明稻米镉含量 受土壤中镉的形态及土壤 pH 的强烈影响. 在此基 础上,选出与糙米 Cd 含量和水稻产量相关性高的 指标,通过偏最小二乘路径模型分析各项指标之间 的逻辑关系,结果见图 7(b). 该模型拟合优度 (GoF)为0.634,其中潜变量交换性阳离子(EC)用 交换性氢离子(EH)、交换性铝(EAI)、交换性 K、 Na、Ca和Mg这6项可测量变量来表征;潜变量生 物有效镉(Bio-Cd)用 F1-Cd、F2-Cd 和 DTPA-Cd 这 3 项可测量变量表征. 土壤 pH 对 Bio-Cd 的路径系 数为 - 0.767,达到显著水平.EC 对水稻产量、糙米 Cd 和 Bio-Cd 的路径系数分别为 0.873、-0.566 和

-0.866,达到显著水平. 潜变量 Bio-Cd 对糙米 Cd 的路径系数为 0.392,达到显著水平. 土壤 pH 对糙米 Cd 和水稻产量的路径系数分别为 -0.045 和 0.069,未达到显著水平,pH 对糙米 Cd 的影响主要是通过影响 Bio-Cd 而间接影响糙米 Cd. 而 EC 对产量和糙米 Cd 的间接效应系数分别为 -0.414, Bio-Cd 对糙米 Cd 的间接效应系数为 0.034,可见交换性阳离子和生物有效镉对水稻产量和镉含量的影响既有直接影响也有间接影响,且直接影响更明显.



(b) 中虚线表示路径未达到显著水平,蓝色线表示路径系数为正,红色表示路径系数为负,线条粗细表示路径系数大小;可测量变量框外数值表示该观察变量对潜变量的贡献权重;*表示P < 0.05; **表示P < 0.01, ***表示P < 0.001

图 7 各项环境因子关联分析

Fig. 7 Correlation analysis of various environmental factors

3 讨论

田间试验结果表明,硅钙镁钾肥、钙镁磷肥和石灰这3种无机钝化剂具有修复水稻土酸化和 Cd污染的联合修复效应,且钝化剂的修复效果随着钝化剂用量的增加而增强. 当3种钝化剂的施用量达2250 kg·hm⁻²时,可使中轻度 Cd污染酸性稻田生产的糙米安全达标,其中石灰在减少土壤镉移动性,降低糙米镉含量上效果较好;钙镁磷肥在施用量超过1500 kg·hm⁻²时,水稻产量较其它处理组有显著增加. 3种钝化剂显著地提高土壤 pH 可能是由于试验材料本身 pH 值很高,且含有 Ca²⁺和 Mg²⁺等盐基离子,能与土壤胶体中 H⁺和 Al³⁺离子发生交换反应^[5,7],从而提高土壤 pH. 施用钝化剂能显著减

少土壤中交换性酸总量,但从土壤交换性 H+和 Al3+的变化看, 钝化剂显著降低交换性 Al3+含量, 而 增加交换性 H+含量,这可能与土壤酸化过程中交 换性 H+与 Al3+离子的消长关系有关,强酸性土壤 中的酸度主要由交换性 Al3+贡献, 钝化剂中盐基离 子替换 Al3+后, Al3+的水解产生 H+, 可能影响土壤 中的交换性 H+含量. 钝化剂降低土壤中有效 Cd 和 糙米中镉含量的化学机制可能是: 钝化剂增加了土 壤 pH 值和交换性盐基离子含量,增强了镉的沉淀 或吸附,降低了镉的移动性,进而降低了水稻对镉的 吸收积累量. 夏文建等[15]的研究表明 CEC 和 pH 对 土壤重金属有效态的影响起到了关键的调控作用. 土壤中发生的溶解反应、沉淀反应、吸附解析等动 态过程,它们的反应平衡很大程度上受 pH 值变化 的影响[16],pH 升高可以增加铁锰氧化物等变价胶 体的负电荷,增强土壤胶体对 Cd2+的吸附能力[17]. 理论模型表明,控制镉迁移率最可能的沉淀物就是 形成 Cd(OH)2, 土壤 pH 值升高可导致土壤 OHT的 溶出,进而和镉形成氢氧化物沉淀,降低土壤 Cd 的 有效性[18]. 而毒性浸出程序(TCLP)结果证明:在 pH < 7 时, 随着 pH 升高, 土壤浸出液中可检测到的 镉含量减少[19]

本试验的钝化剂中较高的钙含量也可能是影响水稻吸收累积 Cd 的重要因素. 含钙钝化材料中 Ca²⁺和 Cd²⁺具有相似的化学性质,钝化剂中所含的盐基离子(Ca²⁺和 Mg²⁺)能与水稻根表面的镉离子竞争吸收,从而抑制水稻对镉的吸收^[7,8,20]. 水培试验监测了在一定的镉胁迫下不同钙浓度溶液中水稻的生长,结果表明钙的应用除了竞争吸附位点,降低水稻对镉的吸收积累,还可以间接保护细胞壁和质膜^[21]. 钝化剂中钾元素的存在可以提高作物的抗逆性,有效降低水稻各营养器官重金属的积累^[22,23]. 元素镉对水稻是非必需元素,但 Cd 很容易被吸收到水稻各部位^[24],碱性肥料的施用可以有效缓解水稻镉的积累^[25~28].

石灰类钝化剂材料自身水解产生 OH⁻和CO₃²⁻,在土壤亲和力强的吸附点位上与 Cd²⁺形成氢氧化物沉淀、碳酸盐沉淀、金属-碳酸盐共沉淀物或金属氧化物等溶解度较低的化合物,从而降低 Cd活性^[29].关于石灰修复土壤 Cd 污染的化学机制已有系统研究^[30],主要原因是石灰可以有效增加土壤pH值和交换性钙离子量,减少稻米对重金属的吸收,并增加重金属的稳定性等机制. 硅钙镁钾肥是富含 Si、Ca、Mg 和 K 的一种碱性肥料,合理施用可以显著增加土壤 pH 值和交换性盐基离子^[5,6,31]. 硅钙镁钾肥降低糙米 Cd 含量的机制除了提高土壤 pH

值,也与多种元素共同作用降低 Cd 生物有效性有关.据报道,硅可与重金属形成复合物沉淀,抑制重金属在作物体内转运^[32].有研究表明,硅钙镁钾肥可以有效改善土壤酸度,增加土壤的阳离子交换能力^[33].因此,施用硅钙镁钾肥可能通过提升土壤 pH 值降低土壤 Cd 活性,硅钙等元素抑制水稻植株 Cd 吸收和积累量,最终降低糙米 Cd 含量. 钙镁磷肥主要是富含 Ca、Mg 和 P 的一种碱性肥料^[34],其施用对作物重金属吸收和积累的影响与硅钙镁钾肥类似.此外,钙镁磷肥中 P 元素的溶出进一步促进作物的生长^[35,36],这与本研究的结果一致.

偏最小二乘路径模型分析表明[图 7(b)], 钝 化剂施用后改变土壤 pH 值而直接影响 Cd 的生物 有效性, 通过降低 Cd 的生物有效性间接影响糙米中 Cd 的含量. 同样, 土壤交换性酸和阳离子组成也直接影响 Cd 的生物有效性, 进一步影响水稻中 Cd 的积累. 土壤 pH 对水稻 Cd 积累的影响机制已有许多证明^[37,38], 本研究进一步证明了土壤交换性阳离子在控制水稻 Cd 积累的重要性, 为土壤钝化剂筛选提供了科学依据.

4 结论

- (1)无机钝化剂硅钙镁钾肥、钙镁磷肥和石灰显著提高土壤 pH,降低土壤交换性酸总量和交换性铝离子含量,增加土壤交换性盐基离子含量,有效地阻控土壤酸化.
- (2) 无机钝化剂显著降低土壤有效态 Cd 浓度,降低水稻对土壤中 Cd 的吸收和积累,降低水稻可食部分的 Cd 风险. 对于 Cd 轻度污染酸性水稻土,钝化剂施用量达2 250 kg·hm⁻²可使糙米 ω(Cd) 低于 0.2 mg·kg⁻¹,达到国家食品安全标准,实现污染耕地水稻安全生产. 无机钝化剂能有效促进土壤中Cd 由弱酸态和可还原态向残渣态转化,降低 Cd 的生物有效性. 钝化剂中含有的 Si、Ca²⁺和 Mg²⁺等盐基离子可与土壤中 Cd²⁺发生竞争和拮抗作用,从而抑制水稻对 Cd 的吸收.
- (3)PLS-PM 分析表明,EC 和 pH 对土壤中生物有效镉有显著的直接影响,它们通过影响 Cd 的生物有效性进而影响水稻 Cd 的积累.而土壤交换性阳离子也对水稻产量和水稻 Cd 的积累产生显著的直接影响,表明土壤交换性阳离子在污染土壤水稻安全生产中起到重要作用. 3 种钝化剂具有土壤酸化与重金属污染联合修复作用,研究结果阐明了钝化剂调控土壤中 Cd 生物有效性的化学机制,为污染耕地水稻安全生产和酸化土壤改良提供科学依据.

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