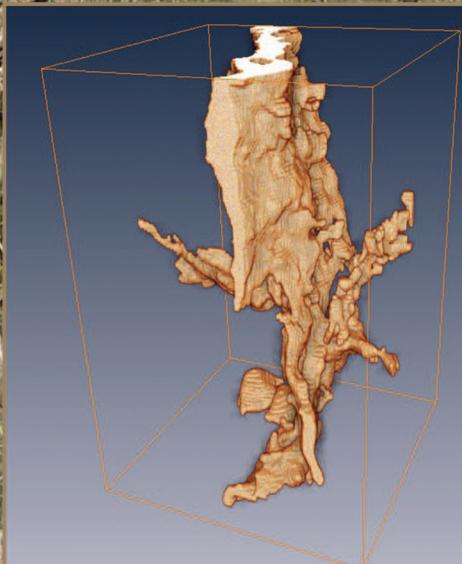


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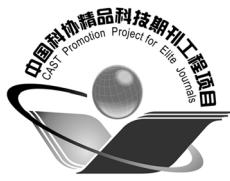
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钝化剂-锌肥降低烟草镉含量长期效果研究^{*}

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摘要 采用田间小区试验, 通过外源添加 1.5 mg kg^{-1} 镉(Cd)模拟轻度Cd污染土壤, 研究赤泥、油菜秸秆、玉米秸秆及其组合并配施硫酸锌降低土壤Cd有效性和烟草各部位Cd含量的长期效果。结果表明, 施用钝化剂后, 烟叶产量增加了7.0%~32.1%, 中上等烟叶比例提高了2.9%~20.2%, 烟草经济效益提高了6.6%~31.3%。钝化剂处理土壤有效态Cd含量降低显著, 2012年和2013年降幅分别为18.6%~47.4%、16.0%~27.0%。烟草各部位Cd含量排序为下部叶>中部叶、上部叶>茎秆, 且均与土壤有效态Cd含量呈极显著正相关关系。2011—2013年钝化剂处理烟草叶片Cd平均含量较对照分别降低了28.3%、28.1%、15.2%, 烟草各部位Cd含量年变化幅度下部叶最大, 茎秆最小, 叶位间Cd含量差异随钝化时间延长逐渐缩小。钝化剂降低土壤-烟草系统Cd生物有效性效果稳定持久, 其中油菜秸秆优于玉米秸秆, 有机无机复合钝化剂效果最好。配施锌肥后, 2011—2013年烟草叶片Cd含量较对照分别降低了19.1%、23.3%、17.9%, 且下部和中部叶片降低效果达到显著水平; 中部和上部叶Cd、Zn含量呈显著负相关关系。因此, 在轻度Cd污染土壤上, 赤泥油菜秸秆复合钝化剂配施锌肥是降低烟叶镉含量兼顾高产的最佳生产措施。

关键词 烟草; 镉; 锌肥; 钝化剂; 有效性; 稳定性

中图分类号 S154.4, X131.3 **文献标识码** A

我国农田土壤镉(Cd)超过《土壤环境质量标准(GB15618-1995)》^[1]Ⅱ级的比例高达11.9%~21.1%^[2], 较其他重金属超标严重, 并以每年 0.004 mg kg^{-1} 速度增加, 为农田环境质量首要监控元素^[3]。土壤中Cd易被植物吸收, 进入食物链危害人体健康。烟草是Cd高富集植物, 其中烟叶Cd积累量占吸收总量的50%~80%^[4-5]。香烟中Cd燃烧点低于其他重金属元素, 易迁移至烟气粒相中被人体吸收^[6]。镉在人体中半衰期为13.6~23.5 a^[7], 有报道指出终身吸烟者Cd积累量是不吸烟者(15 mg)的2倍, 而血Cd浓度高达4倍~5倍^[8]。长期血Cd浓度偏高可引起人体某些器官病变甚至癌变^[9-10]。因此, 降低烟草Cd含量成为烟草和健康等领域关注焦点。

土壤中Cd有效态含量直接影响植物吸收。目前, 以工农业废弃物为钝化剂的原位钝化技术,

因资源丰富、成本低廉、容易操作等优点, 成为治理Cd污染土壤的研究热点。其中, 治铝工业副产物赤泥由于其呈碱性且比表面积大, 被广泛用于修复重金属污染的水和土壤^[11-12]。秸秆中含有丰富的有机官能团, 如-COOH、-OH、C=O、-SH, 可与Cd结合形成稳定化合物, 降低土壤Cd生物有效性^[13-15]。此外, 利用Zn-Cd拮抗作用, 在Cd污染土壤中施锌肥亦可有效地降低植物Cd吸收^[16-18]。但关于钝化剂-锌肥联合技术在降低轻度Cd污染土壤Cd有效性及钝化时效性评价尚少有研究报道。本试验通过模拟轻度Cd污染土壤, 研究赤泥、玉米秸秆和油菜秸秆钝化剂配施锌肥降低土壤Cd有效性和烟草Cd含量长期效果, 从中筛选出最优钝化组合, 为轻度Cd污染土壤上低Cd含量烟草生产提供理论依据和技术指导。

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1 材料与方法

1.1 供试材料

田间试验在中国农业科学院德州实验站进行。供试烟草品种为NC55, 由中国农业科学院烟草研究所提供。供试土壤为潮土(Fluvaquic soil), pH为8.90(水土比5:1^[19]), 有机质含量为12.0 g kg⁻¹(重铬酸钾外加热法测定^[20]), 阳离子交换量为8.3 cmol kg⁻¹(氯化钡-硫酸法测定^[20]), 黏粒含量为17.6% (激光粒度分析仪测定^[21]), 土壤背景镉含量为0.152 mg kg⁻¹(王水-氢氟酸消解法测定^[1])。

试验所采用的钝化剂材料为赤泥、油菜秸秆、玉米秸秆(由王立群^[22]盆栽试验确定)。其中, 赤泥(pH=11.1)取自中国铝业山东分公司, X射线衍射分析其矿物组成为: Fe₂O₃(28%)、Al₂O₃(21%)、SiO₂(20%)、Na₂O(11%)、CaO(6.2%)、TiO₂(3.3%)、MgO(1.3%)、K₂O(0.26%)。105℃下烘干至恒重, 磨碎后过1 mm筛备用。玉米秸秆和油菜秸秆于70℃烘干至恒重, 磨碎后过1 mm筛备用。赤泥、油菜秸秆、玉米秸秆3种钝化剂背景镉含量分别为0.26、0.34、0.22 mg kg⁻¹, EDTA提取态含量均小于0.01 mg kg⁻¹, 因此可以忽略钝化剂背景Cd含量对试验的影响。

1.2 试验设计

试验采用双因素裂区设计, 小区面积4 m²(2 m×2 m), 外源添加1.5 mg kg⁻¹Cd, 以CdSO₄·8H₂O形式加入, 平衡2个月后, 设置试验处理(镉盐、钝化剂、锌肥于2010年豇豆试验^[23]中施入)。主处理为不同钝化剂处理, 分别为: (1)对照(CK)、(2)0.5%赤泥(w/w)(RM)、(3)0.1%油菜秸秆(w/w)(RS)、(4)0.1%玉米秸秆(w/w)(CS)、(5)0.5%赤泥+0.1%油菜秸秆(RMRS)、(6)0.5%赤泥+0.1%玉米秸秆(RMCS)。每个处理重复5次, 其中2次重复为添加锌肥处理(Zn含量4.3 mg kg⁻¹, 以ZnSO₄·7H₂O形式加入, 因试验场地有限玉米秸秆不设锌肥处理)。2011—2013年种植烟草, 每年选取长势均匀的烟苗于5月初移栽, 每小区10株。种植前施入纯氮60 kg hm⁻², 并按照N:P₂O₅:K₂O=1:1:2比例施加磷肥、钾肥。烟田日常管理同常规生产田。

1.3 样品采集与分析

2012—2013年两季烟草收获后, 各小区五点法采集0~20 cm土壤, 风干, 过0.15 mm尼龙筛。土壤有效态Cd含量采用EDTA浸提法测定: 称取2.000 g土壤样品于50 ml离心管中, 加入0.02 mol L⁻¹ EDTA-Na₂溶液25 ml, 室温下200 r min⁻¹往复振荡2 h, 离心机4 000 r min⁻¹离心0.5 h, 过0.45 μm滤膜, 滤液利用电感耦合等离子体质谱仪(ICP-MS)测定Cd含量。

烟草收获茎秆及上、中、下部叶(2011年除外, 未考虑叶位间Cd含量差异), 带回实验室清洗、杀青、烘至恒重、称量后粉碎备用。称取0.300 0 g样品于聚四氟乙烯消解罐中, 加入6 ml浓硝酸和2 ml双氧水微波消解, 消解液经赶酸、过滤、定容至50 ml待测。利用ICP-MS测定烟草Cd、Zn含量。

1.4 数据处理

利用Microsoft Excel 2007进行均值、标准差计算和作图, 利用SAS 9.0的ANOVA模块(LSD法, α=0.05)对试验处理进行方差分析和均值比较。

2 结果

2.1 不同钝化剂处理下烟叶产量及经济效益

2013年烟叶生物量(表1)数据显示, 添加不同钝化剂后, 各部位烟叶干重较对照有不同程度的提高, 上、中、下三部位叶片提高幅度分别为2.4%~28.3%、2.3%~20.2%、15.5%~34.7%; 进而也提高了单株叶片产量, 增产幅度为7.0%~32.1%, 增产效果排序为: RMRS, RMCS>RS, RM>CS, 其中RMRS处理增产效果最明显, 单株产量达161.8 g。施用钝化剂后品质较高的中、上部烟叶干重增加, 也提高了中上等烟叶比例, 增幅为2.9%~20.2%, 进而显著提高了烟草经济效益, 其中RMRS处理产值最高, 较对照增加了31.3%。

2.2 不同钝化剂处理下土壤有效态Cd含量变化

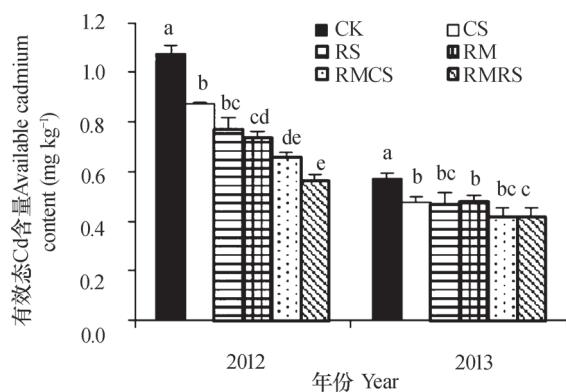
由图1可知, 2012—2013两季烟草土壤有效态Cd含量不施钝化剂处理均最高, 分别为1.07、0.57 mg kg⁻¹。施用钝化剂后, 两季烟草土壤有效态Cd含量分别降低了18.6%~47.4%(2012年), 16.0%~27.0%(2013年), 钝化剂钝化效果排序为: RMRS>RMCS>RS, RM>CS, 其中油菜秸秆(RS)

表1 不同钝化剂处理下烟草生物量及经济效益

Table 1 Tobacco biomass and economic benefit relative to treatment

处理 Treatment	上部叶 Upper leaf	中部叶 Middle leaf	下部叶 Bottom leaf	单株产量 Yield (g plant ⁻¹)	中上等 烟叶比例 Ratio of middle and high grade leaves (%)	产值 Production value (yuan hm ⁻²)
	(g)	(g)	(g)	(g plant ⁻¹)		
CK ± Zn	4.49 ± 0.24c	6.09 ± 0.30d	5.30 ± 0.26d	122.5 ± 5.7d	61.9 ± 2.6c	26 413 ± 967e
CS ± Zn	4.60 ± 0.38c	6.23 ± 0.23ed	6.12 ± 0.20c	131.1 ± 4.5c	63.7 ± 0.3bc	28 150 ± 1 027de
RS ± Zn	4.76 ± 0.54bc	6.71 ± 0.32bc	6.41 ± 0.38bc	138.3 ± 3.3c	66.9 ± 3.5b	30 642 ± 1 373c
RM ± Zn	4.79 ± 0.11bc	6.40 ± 0.44bcd	6.57 ± 0.10b	135.4 ± 1.5c	65.8 ± 2.5bc	30 013 ± 1 328cd
RMCS ± Zn	5.25 ± 0.18ab	6.89 ± 0.30ab	6.26 ± 0.05bc	149.6 ± 6.2b	71.9 ± 1.6a	34 672 ± 1 505b
RMRS ± Zn	5.76 ± 0.18a	7.32 ± 0.23a	7.14 ± 0.24a	161.8 ± 2.6a	74.4 ± 4.0a	38 805 ± 1 281a

注：同一列中无相同字母表示处理间在 $p < 0.05$ 水平上差异显著 Note: Different letters in the same column mean significant differences at 0.05 level between treatments



注：CK为对照；CS为玉米秸秆；RS为油菜秸秆；RM为赤泥；RMCS赤泥+玉米秸秆；RMRS赤泥+油菜秸秆；图中同一指标不同字母表示 $p < 0.05$ 水平上的显著性。下同 Note: CK stands for control; CS for corn straw; RS for rape straw; RM for red mud; RMCS for red mud plus corn straw; RMRS for red mud plus rape straw; Different letters in the same index show significant differences with $p < 0.05$. The same below

图1 钝化剂处理下土壤有效态Cd含量年变化

Fig.1 Annual variation of available cadmium content in the soil relative to treatment

钝化效果优于玉米秸秆(CS)，有机无机钝化剂组合较单施钝化剂效果更为显著，赤泥油菜秸秆复合钝化剂最大程度地降低了土壤有效态 Cd 含量。

2.3 不同钝化剂处理下烟草各部位Cd吸收积累

图2显示烟草不同部位Cd含量分布规律为：下部叶 > 中部叶 > 上部叶 > 茎秆，添加钝化剂后

显著降低了2011—2013三季烟草叶片Cd平均含量，降幅分别为28.3%、28.1%、15.2%，此外上、中、下三部位叶Cd含量随钝化时间延长差异逐渐缩小，比例由1:1.81:2.00(2012年)降至1:0.97:1.05(2013年)，其中附加值较高的中、上部叶片Cd含量的降低对提高烟叶品质尤为重要。烟草各部位Cd含量年度变化茎秆较小，下部叶最大2013年较2012年降低了52.7%。三季烟草试验结果一致表明钝化剂降低烟草Cd含量效果长期稳定，钝化剂降低效果排序为：RMRS, RMCS>RS, RM>CS；该顺序与钝化剂降低土壤有效态Cd含量排序基本一致，说明土壤有效态Cd含量直接影响烟草Cd吸收。2012年烟草试验土壤有效态Cd含量与茎秆和各部位叶片Cd含量分别呈极显著($p < 0.01$)正相关关系(图3)；由于2013年烟草试验叶位间Cd含量差异减小，土壤有效态Cd含量直接与烟草叶片Cd含量呈极显著的正相关关系。钝化剂通过降低土壤有效态Cd含量，降低烟草根部Cd吸收，且土壤有效态Cd含量越低，烟草叶位间Cd含量差异越小。

2.4 施锌肥对各部位叶Cd含量影响及烟叶Cd、Zn间关系

图4结果表明，2011—2013三季施锌肥后烟草叶片Cd平均含量分别降低了19.1%、23.3%、17.9%，尤其是Cd积累较多的中部、下部叶，施锌

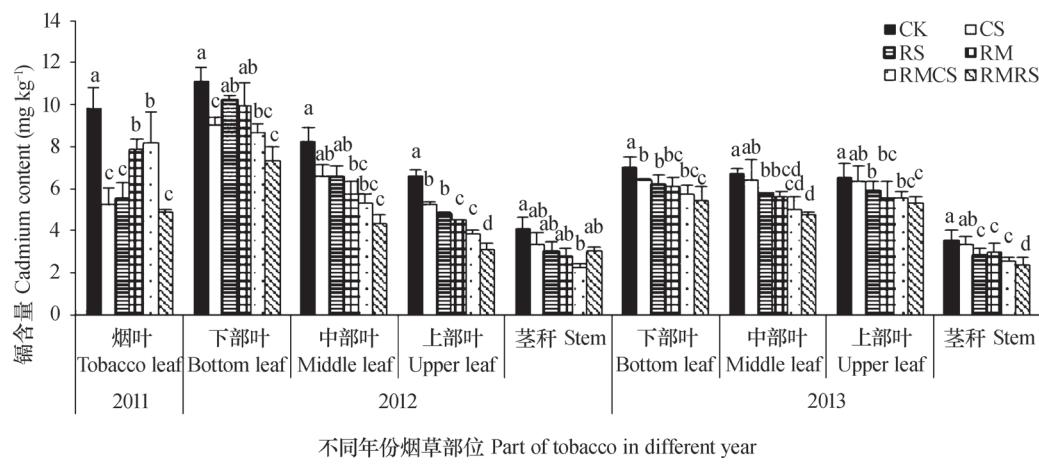


Fig.2 Cadmium content in different parts of tobacco grown in soils with different treatments

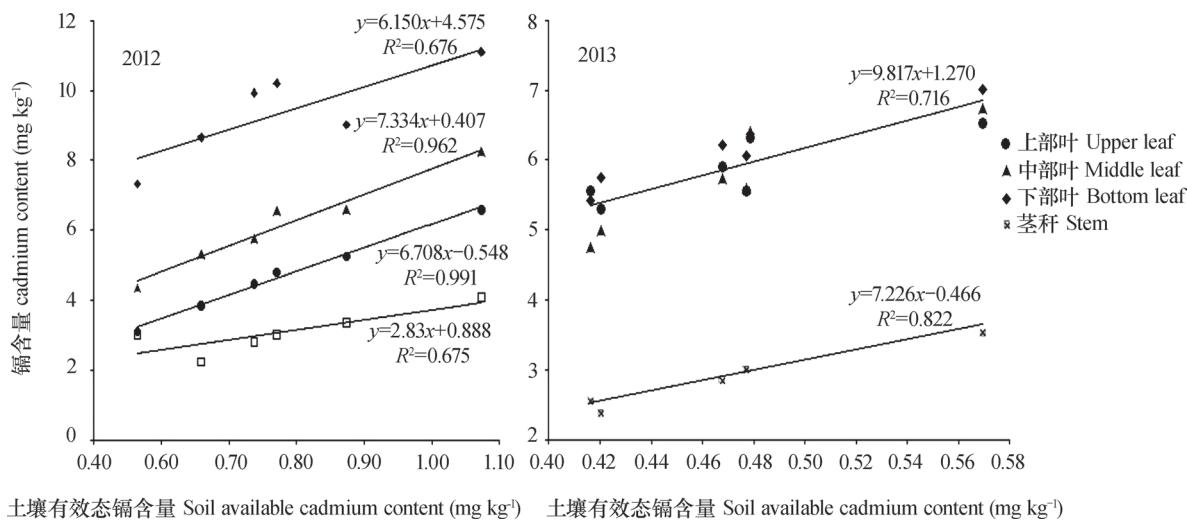


图3 土壤有效态Cd含量与烟草各部位Cd含量关系

Fig.3 Relationship between soil available cadmium content and cadmium contents in different parts of tobacco

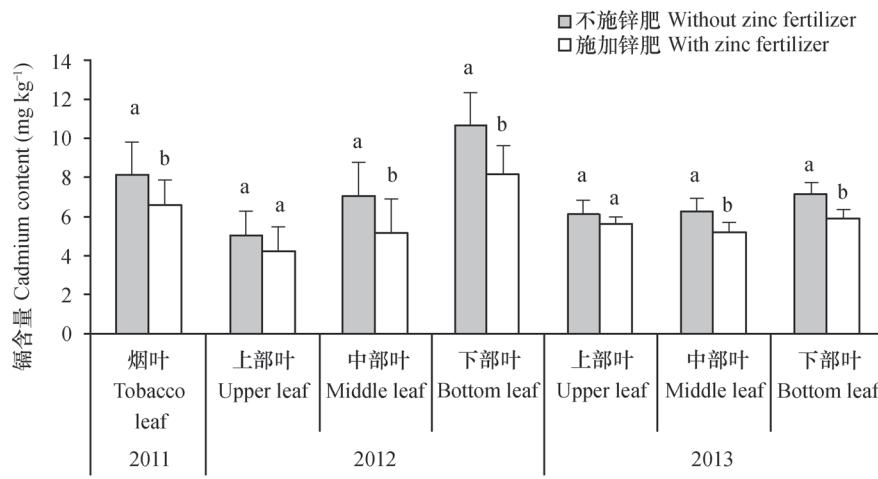


图4 锌肥处理下烟草不同叶位镉含量

Fig.4 Cadmium contents in leaves of different position of tobacco grown in soils with or without zinc fertilizer

肥后Cd含量降低达显著水平。在时间尺度上，锌肥降低烟草Cd含量效果有下降趋势，这可能与土壤有效态Zn含量降低有关。2013年烟草试验叶片Cd、Zn含量可以看出，中、上部叶片Cd、Zn含量

呈显著负相关关系，而下部叶呈显著正相关关系（图5），两者关系在叶位间表现不一致可能与植物体内Zn的转运再分配有关。

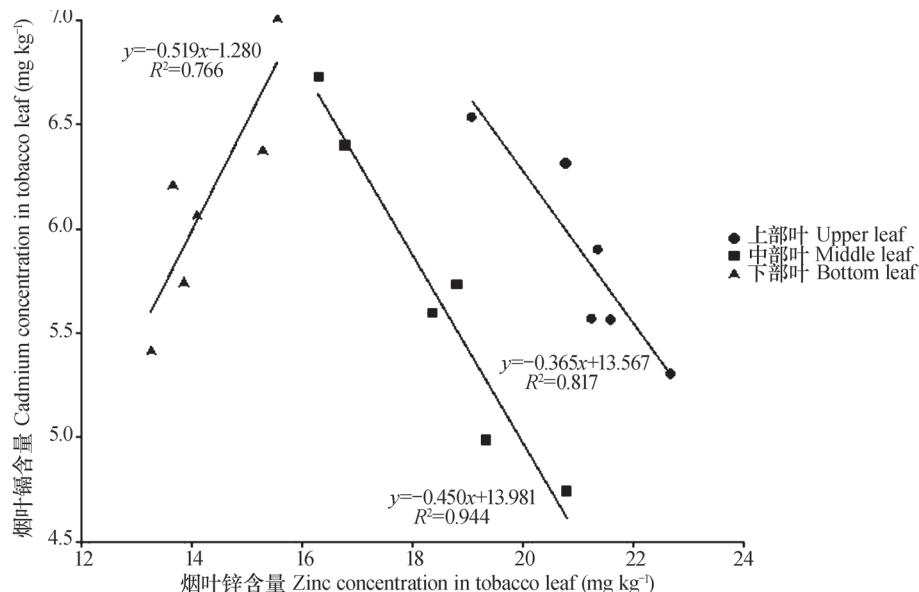


图5 烟叶Cd、Zn含量关系

Fig.5 Relationships between cadmium and zinc contents in tobacco leaves

3 讨 论

在轻度Cd污染土壤中施加钝化剂可以提高整株烟叶产量（表1），尤其是高品质的中部和上部叶干重，直接提高了中上等烟叶比例，最终增加了烟农经济效益。有研究指出在重金属污染土壤上添加钝化剂，可显著降低重金属植物毒害，提高生物量^[24]；胡钟胜等^[25]研究发现在重金属污染的植烟土壤中添加钝化剂后，烟叶产量提高了1倍~2倍。钝化剂施用增加烟草生物量可能原因有：（1）赤泥^[24, 26]或有机物料^[27-28]施入污染土壤可显著提高微生物活性；而土壤微生物参与有机物分解和矿质元素循环，直接为植物生长提供必需的营养元素。（2）赤泥中含有植物生长所必需的K、Ca、Mg、Fe等营养元素^[29]，可促进植物生长发育；有机物料不仅提供矿质养分，还可以改善土壤团粒结构，提高土壤肥力，进而提高植物产量^[28]。

（3）施用钝化剂不同程度地降低了烟草各部位Cd吸收积累，可能减轻了Cd对烟草生理生化过程的毒害，尤其是光合磷酸化过程，增加了烟草光合产

物积累。

有机或无机钝化剂施用均能促进土壤Cd从植物易吸收形态向难溶态转化，降低植物Cd吸收积累。有报道指出赤泥可促进Cd由水溶态和交换态向铁锰氧化态转化^[30-31]，因为赤泥中铁铝氧化物可对重金属产生专性吸附并将其固定到氧化物晶格层间^[31]，Luo等^[30]进一步利用X射线吸收近边结构技术发现Cd与赤泥内部基团形成XCdOH稳定化合物。秸秆因富含有机官能团可促进Cd向有机结合态和残渣态转变^[15]；此外巯基的Cd亲和力强，与Cd可形成稳定化合物^[32]，因此富含巯基的油菜秸秆更利于降低土壤Cd有效性。有机-无机复合钝化剂因吸附位点多，故固定Cd能力优于单一钝化剂^[13, 33]。三季烟草试验及丁琼等^[23]豇豆试验结果一致表明，钝化剂降低土壤Cd有效性效果稳定持久，其中富含巯基的油菜秸秆好于玉米秸秆，有机无机复合钝化剂优于单一钝化剂。

本试验结果表明施锌肥可降低烟草各部位叶Cd含量，且Cd积累量越多的部位，降低效果越显著。Yang等^[17]盆栽试验发现施锌肥可大幅度

提高黄瓜幼苗Zn含量, 显著降低地上部Cd含量。McKenna等^[18]研究表明Zn不仅抑制莴苣和菠菜根系Cd吸收, 还阻止Cd经木质部向地上部运输。Cd-Zn拮抗机制可能是Cd、Zn在根细胞及木质部薄壁细胞中存在共同的吸收转运蛋白, 如铁锌转运蛋白ZIP和重金属转运蛋白HMA4^[34-35]。烟草中、上部叶Cd、Zn含量呈显著负相关关系, 与丁琼等^[23]豇豆试验结果一致; 而下部叶呈正相关关系, 与Keller等^[36]报道第2层烟叶(共4层) Cd-Zn关系一致。Erenogiu等^[37]指出锌在植物体内是易移动元素, 老叶中14%~35%锌可通过韧皮部转移至新叶和根中。考虑造成烟草各部位叶片Cd、Zn含量关系不一致原因, 可能与烟草生长中后期生长点转移至中、上部叶, 下部叶Zn转移再分配有关。

4 结 论

本试验研究表明, 在轻度Cd污染土壤上, 添加赤泥、油菜秸秆、玉米秸秆及有机无机钝化剂组合, 可显著提高烟草单株产量尤其是高品质的中、上部叶干重, 优质烟叶比例提高可以增加烟农经济效益, 钝化剂降低土壤-烟草系统Cd有效性效果稳定持久, 油菜秸秆因具有特殊的巯基基团固定土壤Cd能力较玉米秸秆强; 有机无机复合钝化剂因Cd吸附位点增加, 使土壤Cd形态更多地向植物难吸收的铁锰氧化态、有机结合态转化, 降低土壤Cd植物有效性效果更好。利用Cd-Zn拮抗作用钝化剂配施锌肥可进一步降低烟草Cd含量。但工农业废弃物成分复杂, 重金属含量超标的工农废弃物应用于农业种植可能会引起土壤重金属累积, 具有一定风险性, 因此, 我国急需制订完整的工农业废弃物农用污染物限量标准, 以引导我国农田土壤钝化修复技术健康发展。本试验证实清洁的赤泥油菜秸秆复合钝化剂配合锌肥是降低轻度Cd污染土壤上土壤-植物系统Cd有效性的有效措施。

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EFFECTS OF LONG-TERM AMENDMENT WITH PASSIVANT AND ZINC FERTILIZER ON CADMIUM REDUCTION IN TOBACCO GROWING IN A Cd CONTAMINATED FIELD

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Abstract With rapid industrialization and urbanization in progress over the recent decade in China, the issue of Cd contamination of agricultural soils has become more and more serious. According to the surveys and statistics available that show about 7% of the agricultural soils have already exceeded the National Standard for Environmental Quality in Cd contamination and relative to other heavy metals and metalloids, cadmium (Cd) should be deemed as an element of the first priority for control. Besides, tobacco is a high Cd accumulator, storing Cd mainly in its leaves, which, if consumed by chronic smokers, would leave Cd accumulated in their bodies. Therefore how to decrease Cd content in tobacco plant, especially in its leaves, has been a hot topic in the fields of tobacco industry and public health. In a three-year field plot experiment, red mud and/or plant straw was amended in addition, or not, to Zn fertilization to explore its long-term effect on Cd *in-situ* immobilization in the soil and Cd accumulation in tobacco plant, so as to find a best way to reduce cadmium content in tobacco growing in contaminated soils. The experiment was designed to have five treatments in terms of amendment (red mud, rape straw, corn straw, red mud plus rape straw, and red mud plus corn straw) and two levels of Zn fertilization. The soil in the experiment was artificially contaminated with 1.5 mg kg⁻¹ cadmium added to the soil. Results show that compared with control, the yield of tobacco leaves and the ratio of mid-high grade leaves in amendment treatments were significantly increased by 2.6%~32.3% and 2.9%~20.2%, respectively, which brought about an increase in economic benefit by 6.6%~31.3%. Among the treatments, the amendment of red mud plus rape straw was found to be the highest in both yield and economic benefit. The amendments stably decreased soil available Cd contents by 18.6%~47.4% in 2012, and 16.0%~27.0% in 2013. The plants from the treatment were found to follow an order, i.e. bottom leaf > middle leaf and upper leaf > stem in terms of Cd content in plant. The contents of Cd in tobacco were most positively related to soil available Cd content. Therefore, the Cd content in tobacco leaves in 2011, 2012 and 2013 decreased by 28.3%, 28.1% and 15.2% on average, respectively in all the amendment treatments. The effect was the most apparent in bottom leaves and the least in stem. Rape straw, full of thiols, was higher in

Cd immobilization capacity than corn straw. Application of zinc fertilizer in addition to the amendments further significantly lowered Cd content in different parts of the tobacco plant at different seasons. In conclusion, the treatment of red mud, rape straw plus zinc fertilizer is the most effective one in this experiment to decrease Cd phytoavailability in soil-tobacco system. Therefore it can be extrapolated as a high cost-efficient measure to decrease Cd content in tobacco plants and increase yield of tobacco leaves in lightly Cd contaminated soils.

Key words Tobacco; Cadmium; Zinc fertilizer; Amendment; Phytoavailability; Immobilization

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