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典型黑色岩系地质高背景区土壤和农产品重金属富集 特征与污染风险

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摘要:土壤重金属污染是我国的重要环境问题,而区域地质高背景是土壤重金属富集的重要影响因素.黑色岩系风化土壤普遍存在重金属富集的现象,潜在生态环境风险较高,但目前农产品重金属富集的基础数据较缺乏,限制了黑色岩系地质高背景区的土地安全利用与农产品安全生产.对重庆典型黑色岩系地质高背景区 313 件土壤和 289 件主要农产品重金属的富集特征、污染风险和土壤重金属赋存形态进行研究.结果表明,相对于我国土壤环境背景值和重庆市表层土壤,研究区土壤 Cd、Cr、Cu、Zn和 Se 富集,Pb 不富集;其中98.7%的土壤 Cd 含量超过风险筛选值,47.3%的土壤 Cd 含量超过风险管制值,Cd 的污染水平和潜在生态风险最高,是研究区土壤的特征污染物.土壤 Cd 主要以离子交换态存在(40.6%),其次为残渣态(19.1%)和弱有机结合态(16.6%); Cr、Cu、Pb、Se 和 Zn 主要以残渣态存在,Se 和 Cu 的有机结合态、Pb 的铁锰结合态也占较高比例;说明 Cd 的迁移能力和活性高于其它重金属.研究区主要农产品富集重金属的能力较弱,约18.7%的农产品 Cd 超标,超标倍数相对较低,重金属污染风险较低.研究可为黑色岩系地质高背景区土地安全利用和农产品安全生产提供科学依据.

关键词:地质高背景区; 土壤; 农作物; 镉(Cd); 硒(Se); 赋存形态

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Accumulation and Pollution Risks of Heavy Metals in Soils and Agricultural Products from a Typical Black Shale Region with High Geological Background

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Abstract: Soil polluted by heavy metals (HMs) is an important environmental issue in China, and regional geological background is a vital factor that influences the enrichment of HMs in soils. Previous studies have shown that soils derived from black shales are commonly enriched in HMs and present high potential eco-environmental risks. However, few studies have investigated the HMs in different agricultural products, which inhibit the safe use of land and safe production of food crops in black shale regions. This study investigated the concentrations, pollution risks, and speciation of HMs in soils and agricultural products from a typical black shale region in Chongqing. The results showed that the study soils were enriched in Cd, Cr, Cu, Zn, and Se but not in Pb. Approximately 98.7% of total soils exceeded the risk screening values, and 47.3% of total soils exceeded the risk intervention values. Cd had the highest pollution level and potential ecological risks and was the primary pollutant in soils of the study area. Most of the Cd resided in ion-exchangeable fractions (40.6%), followed by residual fractions (19.1%) and weak organic matter combined fractions (16.6%), whereas Cr, Cu, Pb, Se, and Zn were dominated by residual fractions. Additionally, organic combined fractions contributed to Se and Cu, and Fe-Mn oxide combined fractions contributed to Pb. These results indicated that Cd had higher mobility and availability than those of other metals. The agricultural products presented a weak ability to accumulate HMs. Approximately 18.7% of the collected samples with Cd exceeded the safety limit, but the enrichment factor was relatively low, indicating low pollution risks of the heavy metals. The findings of this study could provide guidelines for safe use of land and safe production of food crops in black shale regions with high geological background.

Key words: high geological background; soil; food crops; cadmium (Cd); selenium (Se); speciation

土壤重金属污染是我国目前的主要环境问题之一,除矿冶、工业和农业等典型人为活动造成土壤重金属污染外,区域地质高背景也是土壤重金属富集的重要因素[1~3]. 我国浙江部分地区、珠江三角洲和西南碳酸盐岩地区土壤重金属的富集均不同程度受到地质高背景的影响[4~6],其中,西南地区大面积土壤镉等重金属自然富集,土壤重金属富集特征与环境效应被广泛关注[7~10]. 有研究表明,碳酸盐岩地质高背景区土壤重金属的生物活性较低[11],农产品污染风险低,例如广西岩溶区稻田土壤 Cd 含

量超过风险筛选值的点位占比超过 50%,但在安全利用区和严格管控区,水稻籽实 Cd 的超标率分别为 11.5% 和 16.4% [12]. 而受硫化物风化产酸等因素影响,黑色岩系地质高背景区土壤重金属的生物活性相对较高,以 0.01 mol·L⁻¹ CaCl,溶液对重金

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属的提取率为例,重庆典型黑色岩系地区土壤 Cd 的平均提取率为6.14% [13],浙江黑色岩系地区土壤 Cd 的平均提取率为15.9% [14],均显著高于碳酸盐岩风化土壤平均值1.9% [11].然而,与西南碳酸盐岩地质高背景区相比,目前关于黑色岩系地区农产品重金属富集特征与污染风险的研究相对较少,有研究表明,即使土壤 Cd 含量和生物有效性较高,但黑色岩系地区玉米的超标率仍较低 [13,15].然而,其它农产品中重金属的污染风险依然未知,相关基础数据的缺乏制约了黑色岩系地质高背景区土地安全利用和农产品安全生产.因此,本文选择重庆市城口县的典型黑色岩系地质高背景区,系统采集区内土壤和多种主要农产品,研究样品重金属总量及典型土壤样品中重金属赋存形态特征,通过揭示黑色岩系地区土壤与农产品的重金属富集特征与污染风

险,以期为区域重金属污染风险防控和农产品安全 生产提供科学依据.

1 材料与方法

1.1 研究区域概况

研究区位于重庆市城口县(图1),属亚热带山地气候,具有气候温和、雨量充沛、日照较足、四季分明和冬长夏短等特征.地形地貌以低山河谷、中山和峰丛台地为主.研究区位于城巴断裂带以北,区内地层主要为寒武系下统鲁家坪组,岩性主要以含炭质板岩和泥质板岩为主,中上部夹少量薄层灰岩,部分板岩含黄铁矿星点和结核.成土母质主要为第四系新冲积黄色冲积物、碳酸盐岩风化物、沙泥岩和碳酸盐风化物.土壤类型为黄壤和黄棕壤.研究区耕地以旱地为主,主要种植玉米、红薯、土豆和萝卜等农作物.



Fig. 1 Location of the study area and sampling sites

1.2 样品采集与分析

本文采集了研究区内主要地块的土壤和对应农产品样本,表层土壤样品(0~20 cm)根据地块形状选择"X"型或"S"型由主样点向周边辐射 30~50 m确定 4~6个分样点,等份组成一个混合样,共采集土壤样品 313 件.农产品采集于收获盛期,在采样点地块内视不同情况采用棋盘法、梅花点法、对角线法和蛇形法等进行多点取样,然后等量混匀组成一个混合样品,共采集农产品样品 313 件,其中土豆48 件,草莓 27 件,玉米 80 件,萝卜 47 件,红薯 42件,板栗 45 件;另有 24 件为中药材样品,未纳入本文讨论.

土壤样品自然风干后过10目孔径尼龙筛,剔除粗粒级石块和植物根系,使用玛瑙球磨机破碎成200目粒径.采用重铬酸钾-硫酸220℃加热消煮,氧化还原容量法测定土壤有机碳(TOC).采用离子选择性电极(ISE,雷磁 PHSJ-3F)以土水比1:2.5测定土壤pH.采用X射线荧光光谱仪(XRF,荷兰帕纳科PW4400)测定土壤Cr、Cu和Zn含量;采用混合酸

(HF、HNO₃、HClO₄和 H₂SO₄)消解,逆王水溶解残渣,消解液转移至25 mL 比色管中定容,使用电感耦合等离子体质谱(ICP-MS)测定 Cd 和 Pb 含量.采用HNO₃ + HF + HClO₄ 消解,再用 HCl 溶解残渣,消解液转移至25 mL 比色管中定容,采用 KBH₄ 还原,原子荧光光谱仪(AFS,北京海光 AFS-9760)测定 Se含量.每批次样品中插入4个不同含量的国家一级土壤标准物质(GSS-5、GSS-19、GSS-21和GSS-26为一组),各项技术指标的原始一次性合格率均为100%.

农作物样品洗净晾干后采用切碎机切碎,再使用无污染破碎机粉碎后用于分析测试. 农作物重金属含量分析按照《生态地球化学评价样品分析技术要求(试行)》(DD 2005-03)[16]执行,采用 HNO₃ 和 H_2O_2 在微波消解仪消解,ICP-MS 测定 Cd、Cr、Cu、Pb 和 Zn 含量;在 HCl 介质(6 mol·L⁻¹)中用 KBH₄将六价 Se 还原成四价 Se,采用 AFS 测定 Se 含量.目标元素检出限均满足 DD 2005-3^[16]的检出限要求.每批次按要求插入准确度控制的国家一级标准

物质2件,同时选取加标回收样品2件.国家一级标 准物质测定值准确度合格率均为100%,重复性检验 合格率 100%, 加标回收率均处于 90%~110% 之间.

1.3 重金属赋存形态分析

本文选择30件典型土壤样品,按照文献[16] 的方法进行连续提取,将重金属形态划分为水溶态、 离子交换态、碳酸盐态、弱有机结合态、铁锰结合 态、强有机结合态和残渣态,采用 ICP-MS 测试提取 液中 Cd、Cr、Cu、Pb 和 Zn 含量,采用原子荧光光谱 法测试 Se 含量,该方法对 Cd、Cr、Cu、Pb、Zn 和 Se 的回收率分别为82%~104%、84%~102%、88%~ 104%、80%~104%、87%~104%和87%~103%.

1.4 数据处理与分析

本研究数据的整理采用 Excel 软件完成,数据 的多元相关分析采用 IBM SPSS 软件完成,图表的绘 制采用 Origin 软件完成,采样点位图使用 ArcMap 绘制.

本文采用 Müller $^{[17]}$ 建立的地累积指数 (I_{geo}) 法

对研究区土壤重金属污染水平进行评价,计算公式 如下:

$$I_{\text{geo}} = \log_2 C/1.5B$$

式中, C 为目标重金属的实测含量; B 为目标重金 属的地球化学背景值,采用重庆市表层土壤重金属 含量平均值^[18]:ω(Cd)、ω(Cr)、ω(Cu)、ω(Pb)和 ω(Zn)分别为 0.27、74、24、27 和 79 mg·kg⁻¹; 1.5 为校正系数,用于校正人为和自然影响.污染等级划 分如表1所示.

本文采用 Håkanson^[19] 提出的生态风险因子 (E.) 法对研究区土壤各重金属的潜在风险进行评 估,计算公式如下:

$$E_r = (C/B)T$$

式中,C为目标重金属的实测含量;B为目标重金 属的地球化学背景值,采用重庆市表层土壤重金属 含量平均值[18]. T为毒性因子, Cd 为 30, Cr 为 2, Cu、Ni 和 Pb 为 5, Zn 为 1^[19]. 风险生态等级划分如 表 2 所示.

表 1 土壤污染等级划分

		Tab	le 1 Classification	n of pollution lev	el of soils	100	JA CAS
污染等级	未污染	未污染至中度污染	中度污染	度至重度污染	重度污染	重度至极度污染	极度污染
$I_{ m geo}$	€0	0~1	1 ~2	2 ~ 3	3 ~ 4	4 ~ 5	(≥5
1	65	11	表 2 土壤潜在	生态风险等级均	100 10	24)	1

表2 土壌潜在生态风险等级划分

0 11 3	1 - 4	table 2 Classification of po	tentiai ecologicai risks ol	SOIIS	
污染等级	低潜在风险	中等潜在生态风险	较高潜在生态风险	高潜在风险	极高潜在风险
参数	$E_{\rm r} < 40$	$40 \le E_{\rm r} < 80$	$80 \le E_{\rm r} < 160$	$160 \le E_r < 320$	$E_{\rm r} \ge 320$

2 结果与讨论

2.1 研究区土壤重金属富集特征与污染风险

研究区土壤 pH 变化范围为 4.94~8.14(平均 值为 6.24),其中 pH ≤ 5.5 的样本占比为 16.9%, 5.5 < pH ≤ 6.5 的样本占比为 50.2%, 酸性土壤(pH ≤6.5)共计占比约为 67.1%; 中性土壤(6.5 < pH ≤7.5)占比约为27.1%;碱性土壤(pH>7.5)占比 仅为5.8%.研究区土壤整体以酸性为主,可能与黑 色岩系中硫化物风化产酸导致土壤不同程度酸化有 关^[20]. 从不同种植类型来看,土壤 pH 的变化为:草 莓地(6.78) > 板栗地(6.36) > 玉米地(6.34) > 萝 卜地(6.16) > 红薯地(6.11) > 土豆地(5.92),差异 小于1个 pH 单位. 研究区土壤 $\omega(TOC)$ 变化范围为 0.29%~4.67%(平均值为1.93%),从不同种植类 型来看,板栗地(2.08%)>红薯地(2.00%)>萝卜 地(1.97%)> 玉米地(1.92%)> 草莓地(1.83%) >土豆地(1.72%),由于板栗地属林地,其 $\omega(TOC)$ 高于其它种植类型的土壤.

有研究表明,研究区属于典型的黑色岩系地质 高背景区,土壤重金属源于富重金属黑色岩系风化 成土过程[21,22]. 本文对研究区 313 件土壤样品的分 析结果表明: 土壤 ω (Cd)、 ω (Cr)、 ω (Cu)、 ω(Pb)、ω(Se) 和 ω(Zn) 的平均值分别为 3.29、 145、54.3、24.7、1.68 和 292 mg·kg⁻¹(表 3). 除 Pb 外,其它重金属含量平均值均高于我国土壤环境 背景值[23]和重庆市表层土壤均值[18].以重庆市表 层土壤[18]为参比,研究区土壤重金属的平均富集系 数由大到小依次为: Cd(12.2) > Se(8.40) > Zn (3.69) > Cu(2.26) > Cr(1.96) > Pb(0.91). 以富 集系数最高的 Cd 为例,不同种植类型土壤 ω (Cd) 平均值变化趋势为:板栗地(5.32 mg·kg⁻¹)>玉米 地(3.54 mg·kg⁻¹) > 土豆地(2.83 mg·kg⁻¹) > 红薯 地(2.47 mg·kg⁻¹) = 草莓地(2.47 mg·kg⁻¹)≈萝卜 地 $(2.45 \text{ mg} \cdot \text{kg}^{-1})$,属林地的板栗种植土壤 $\omega(\text{Cd})$ 平均值最高,而其它类型土壤 $\omega(Cd)$ 差异较小. 陈 梓杰等^[13]对该区域农田土壤的研究表明, ω (Cd)、 $\omega(Cr)$ 、 $\omega(Cu)$ 、 $\omega(Pb)$ 和 $\omega(Zn)$ 平均值分别为:

5.11、157、88.2、20.6 和 488 mg·kg⁻¹, ω (Cd)、 ω (Cu)和 ω (Zn)明显高于本研究,这可能与母岩重金属含量差异造成的区域土壤重金属空间分布不均有关.以 Cd 为例,研究区内母岩 ω (Cd)范围为 $0.44\sim64.5$ mg·kg^{-1[21,24]},本文土壤 ω (Cd)范围为 $0.23\sim29.1$ mg·kg⁻¹,标准差达 3.81 mg·kg⁻¹,而前人研究发现该区域土壤 ω (Cd)可高达 64.5 mg·kg⁻¹,标准差达 11.3 mg·kg^{-1[21]},Zn 和 Cr 等

其它重金属元素也呈类似区域分布特征. 与研究区类似的土壤重金属分布空间不均现象普遍存在于地质高背景区,如广西岩溶地质高背景区土壤 $\omega(Cd)$ 为 0. 068 ~ 16. 905 mg·kg^{-1[12]},浙江黑色岩 系 地 区 土 壤 $\omega(Cd)$ 为 0. 07 ~ 13. 03 mg·kg^{-1[14]},云南典型地质高背景区 $\omega(Cd)$ 为 0. 085 ~ 6. 25 mg·kg^{-1[25]},说明区域土壤重金属的富集与母岩的地球化学继承性有关^[26].

表 3 研究区土壤重金属元素含量统计

Table 3 Statistics of heavy metal concentrations in the study soils

指标	最小值 /mg·kg ⁻¹	最大值 /mg·kg ⁻¹	中位值 /mg·kg ⁻¹	平均值 /mg·kg ⁻¹	标准差 /mg·kg ⁻¹	> 风险筛选值 占比/%	>风险管制值 占比/%	重庆市表层土壤 均值 ^[18] /mg·kg ⁻¹	中国土壤背景值 ^[23] /mg·kg ⁻¹
Cd	0. 23	29. 1	1. 98	3. 29	3. 81	98. 7	47. 3	0. 27	0. 10
Cr	55.8	631	121	145	77. 3	24. 0	0	74	61.0
Cu	16. 9	347	42. 2	54. 3	39. 7	25. 2	_	24	22. 6
Pb	14.8	72. 5	23.7	24. 7	5. 01	0	0	27	26. 0
Se	0. 24	17.4	1. 19	1.68	1. 79	1)	_	0. 20	0. 29
Zn	60. 9	2 840	210	292	285	48. 6	_	79	74.2

1) — 表示无风险筛选值或风险管制值

研究区土壤各重金属间均存在显著正相关(*P* <0.01,表4),说明该区域土壤重金属具有同源性. Pb 与其它重金属的相关系数相对较小,且 Pb 在土壤中的富集程度最低,可能与母岩中 Pb 含量较低有关^[21,24]. Cd、Cr 和 Cu 与土壤 pH 呈显著正相关,说明高 pH 条件有利于重金属的富集,其中 Cd 含量与pH 相关性最高(*R* = 0.223, *P* < 0.01,图 2),这与前

人在碳酸盐岩地区的研究结果类似^[12],但本文的相关系数较低.各重金属含量均与 TOC 呈显著正相关(P<0.01),与碳酸盐岩地区的结果类似^[12],说明高有机质含量有利于土壤重金属的富集,土壤腐殖质丰富的官能团可为重金属提供大量的吸附点位^[27,28],这也一定程度上解释了板栗地土壤 Cd 含量高于其它类型土壤的现象.

表 4 研究区土壤重金属含量与主要理化参数间的 Pearson 相关系数¹⁾

47 4	101	Table 4 Com	relation coefficie	nts of heavy metals	and soil propertie	s in the study soil	s	
(0)	рH	TOC	Cd	Cr	Cu	Pb	Se	Zn
pН	1							
TOC	0. 112 *	1						
Cd	0. 223 **	0. 283 **	1					
\mathbf{Cr}	0. 122 *	0. 263 **	0. 841 **	1				
Cu	0. 126 *	0. 255 **	0. 856 **	0. 885 **	1			
Pb	0.088	0. 285 **	0. 430 **	0. 454 **	0. 537 **	1		
Se	0.062	0. 327 **	0. 696 **	0. 857 **	0. 826 **	0. 413 **	1	
Zn	0. 109	0. 269 **	0. 905 **	0. 808 **	0. 879 **	0. 395 **	0.717 **	1

1)*表示在 0.05 级别相关性显著; **表示在 0.01 级别相关性显著

与《土壤环境质量 农用地土壤污染风险管控标准(试行)》(GB 15618-2018)^[29]相比,研究区约98.7%的土壤样本 Cd 含量超过风险筛选值,47.3%的样本 Cd 含量超过风险管制值,其它重金属超过风险筛选值的占比由大到小依次为:Zn(48.6%)>Cu(25.2%)>Cr(24.0%).可见 Cd 是研究区土壤污染风险最高的特征污染物.若按照标准对研究区土地环境质量进行评价,优先保护类、安全利用类和严格管控类的占比分别为 1.3%、51.4%和47.3%,近50%的土地原则上应采取严格管控措施,且主要为酸性土壤(图2).

值得关注的是,除 Cd 等有害元素超标外,研究 区土壤也呈现 Se 富集的特征,土壤 ω (Se)平均值

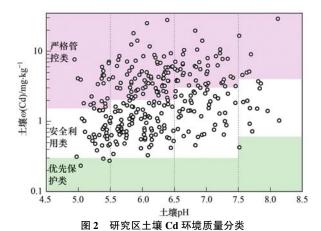
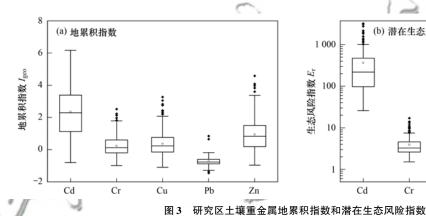


Fig. 2 Classification of the environmental quality of Cd in the study soils

 $(1.68 \text{ mg·kg}^{-1})$ 远高于王惠艳等 $^{[30]}$ 提出的我国早地土壤的富 Se 阈值 $(0.30 \text{ mg·kg}^{-1})$. 根据自然资源部发布的《天然富硒土地划定与标识》 $(DZ/T 0380-2021)^{[31]}$,研究区中酸性土壤 $(pH \le 7.5)$ 中约有93%的土壤 $\omega(Se) \ge 0.4 \text{ mg·kg}^{-1}$ (范围为 $0.40 \sim 17.4 \text{ mg·kg}^{-1}$, 平均值为 1.78 mg·kg^{-1} , N = 294);而所有碱性土壤 $(pH > 7.5)\omega(Se) \ge 0.3 \text{ mg·kg}^{-1}$ (范围为 $0.42 \sim 7.04 \text{ mg·kg}^{-1}$, 平均值为 1.75 mg·kg^{-1} , N = 19).

研究区土壤地累积指数平均值的变化规律为: Cd(2.34) > Zn(0.94) > Cu(0.37) > Cr(0) > Pb(-0.74), Cd的污染水平最高, Zn次之[图3(a)]. 根据污染等级划分, 对 Cd而言, 未污染土壤占比为2.20%, 未污染至中度污染占比为19.8%, 中度污染占比为20.8%, 中度至重度污染占比为21.1%, 重度污染占比为24.0%, 重度至极度污染占比为9.60%,

极度污染占比为 2.50%, 研究区土壤 Cd 污染水平 总体属于中度和重度污染. 对 Zn 而言,未污染土壤 占比为13.7%,未污染至中度污染占比为40.9%,中 度污染占比为 31.0%,中度至重度污染占比为 11.8%, 重度污染及以上占比为 2.6%, 整体属于中 度污染水平. Cr 和 Cu 污染水平低, Pb 无污染. 生态 风险指数平均值变化规律为:Cd(365) > Cu(11.3) > Pb(4.57) > Cr(3.91) > Zn(3.69) [图 3(b)], 由于 Cd 的毒性因子远高于其它重金属,因此其 E_r 值 与其它重金属具有数量级的差异. 对 Cd 而言, 研究 区具有低潜在风险的土壤占比仅为1.90%,中等潜 在风险占比为 15.70%, 较高潜在风险占比为 21.7%,高潜在风险占比为20.4%,极高潜在风险占 比为40.3%.其它重金属元素属于低潜在风险,这 与研究区土壤重金属富集程度和污染水平的结果一 致,说明 Cd 是研究区土壤的特征污染物.



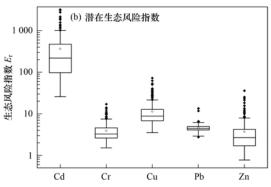


Fig. 3 Geo-accumulation index and ecological risk index of heavy metals in study soils

2.2 研究区土壤重金属赋存形态特征

对研究区30件典型土壤样品的化学连续提取 结果表明: 土壤 Cd 主要以离子交换态存在 (40.6%),其次为残渣态(19.1%)和弱有机结合态 (16.6%),而铁锰结合态(10.9%)、碳酸盐结合态 (7.10%)和强有机结合态(4.40%)占比相对较低, 水溶态仅占 1.30%; Cr 主要以残渣态存在 (89.8%),其余各形态占比极低(图 4). Cu 主要以 残渣态(61.5%)存在,弱有机结合态(22.7%)次 之,铁锰结合态(10.4%)再次之. Pb 主要赋存于残 渣态(56.5%),铁锰结合态(30.1%)次之. Zn 主要 为残渣态(79.9%),强有机结合态(5.88%)、铁锰 结合态(6.71%)和弱有机结合态(5.01%)占比相 近,其他形态占比极低. Se 主要赋存于残渣态 (54.6%),强有机结合态(19.2%)与弱有机结合态 (23.0%)相近,其它形态占比极低. 总体来看,有机 结合态中 Se 的占比最高:铁锰结合态中 Pb 的占比 最高,Cd、Cu 和 Zn 依次降低;离子交换态和碳酸

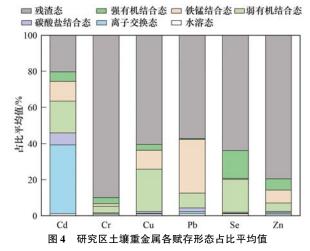


Fig. 4 Speciation distribution of heavy metals in study soils

盐结合态中 Cd 的占比最高. 前人对韩国黑色页岩风化土壤重金属的研究表明, 残渣态 Zn 和 Cu 的占比在 80%以上, 残渣态 Pb 的占比约为 60%, 而残渣态 Cd 的占比基本小于40%, 可交换态 Cd 的占比约为 20% [32]. Zhang 等[33] 对浙江黑色页岩风化土壤的研

究表明,可交换态与碳酸盐结合态 Cd 的占比合计为 38. 22%,而两种结合态 Pb 的占比仅为 3. 70%,与本研究区土壤重金属的形态分布特征结果类似. Cd 的活性态(水溶态、离子交换态和碳酸盐结合态)占比最高,说明其具有较高的迁移能力和环境活性,而 Cr、Cu、Pb、Se 和 Zn 以残渣态为主,迁移能力和环境活性较低. 研究区不同土壤重金属的赋存状态存在明显差异,以 Cd 为例,离子交换态的占比为 21. 9%~57. 7%,碳酸盐结合态的占比为 1.01%~17. 4%,弱有机结合态的占比为 10. 4%~24. 4%,说明区域土壤地球化学组成的异质性较高.

2.3 研究区农产品重金属富集特征与污染风险

尽管研究区土壤 Cd 等重金属富集,且 Cd 的活性较高,但对主要农产品中重金属含量(以鲜重计)的研究结果表明,研究区农产品中重金属含量整体较低(表5),其中 118 件农产品(占比约为 40.8%,主要为玉米、萝卜、红薯和草莓)的 Cd 低于检测限 $(0.03~\text{mg}\cdot\text{kg}^{-1})$; 276 件农产品(占比约为 95.5%)的 Pb 低于检测限 $(0.1~\text{mg}\cdot\text{kg}^{-1})$.从农产品种类来看,板栗 ω (Cd)和 ω (Cu)平均值最高,分别为 0.10 ω (Cu)平均值最高,分别为 0.10 ω (Se)平均值在各农作物中差异较小.与《食品安全

国家标准 食品中污染物限量》(GB 2762-2017)[34] 相比,9件玉米样本 Cd 超过污染物限值(0.1 mg·kg⁻¹),占比为11.3%,富集倍数为1.10~8.34 (平均值为4.28);17件蔬菜类农产品(土豆6件、 萝卜9件和红薯2件)Cd超过污染物限值(0.1 mg·kg⁻¹),占比为12.4%,富集倍数为1.01~2.80 (平均值为 1.64); 8 件草莓样本 Cd 超过污染物限 值(0.05 mg·kg⁻¹),占比为 29.6%,富集倍数为 1.20~3.80(平均值为2.09); 若按0.1 mg·kg⁻¹限 值来计算,20 件板栗样本 Cd 超标,占比为 44.4%, 富集倍数为1.06~2.74(平均值为1.64).所有农产 品样本 Cr 和 Pb 均不超标. 前人对该区域 50 件玉米 样本的研究表明, ω (Cd) (以干重计) 为 0.001 ~ 1.36 mg·kg⁻¹,整体超标率为14%,籽粒 Zn 含量平 均值最高[13],与本研究的结果相似.总体来看,研究 区农产品基本处于安全水平,部分农产品表现为 Cd 超标,超标率约为18.7%,草莓和板栗超标率较高, 但超标倍数相对较低. 与本研究区类似的土壤 Cd 富集且超标率高而农产品 Cd 不富集且超标率较低 的现象普遍存在于我国西南地质高背景 区[9~12,35,36],尽管目前的土壤环境质量标准考虑了 pH 等因素差异,但其在应用于地质高背景区土壤 Cd 污染风险管控时显得过于保守.

表 5 研究区主要农产品重金属鲜重含量统计值¹⁾/mg·kg⁻¹

V6 1/1	Table 5	Statistics of heavy	metal concentration	ns in collected ag	ricultural products/	mg•kg ⁻¹	
项目	类型	Cd	Cr	Cu	Pb	Se	Zn
	最小值	< 0.03	0. 03	0.004	< 0. 1	< 0. 001 5	2. 44
土豆 (N=48)	最大值	0. 15	0. 10	2. 22	< 0. 1	0. 12	8. 43
(11 = 40)	平均值	0.07	0.06	1. 27	2)	0. 01	4. 15
草莓	最小值	< 0.03	0.06	0. 15	< 0. 1	0.002	0.49
早母 (N=27)	最大值	0. 19	0. 19	0. 58	< 0.1	0. 01	1. 37
(17 = 27)	平均值	0.04	0. 11	0. 34	_	0. 01	0. 95
工业	最小值	< 0.03	0.09	0.49	< 0.1	0. 01	5. 57
玉米 (N=80)	最大值	0.83	0. 94	2. 95	0. 14	0. 10	30. 3
(17 = 00)	平均值	0.07	0. 19	1. 25	0.05	0. 03	13. 9
帯し	最小值	< 0.03	0.03	0. 15	< 0.1	0. 01	1. 14
萝卜 (N=47)	最大值	0. 28	0. 21	0.42	< 0.1	0. 18	9. 10
(11 = 47)	平均值	0.07	0.07	0. 26	_	0. 03	2. 64
红曲	最小值	< 0.03	0.04	0.50	< 0.1	0. 01	1.05
红薯 (N=42)	最大值	0. 24	0. 24	1. 35	< 0.1	0. 22	9. 09
(11 = 42)	平均值	0.04	0.08	0. 93	_	0. 03	1.84
+c mi	最小值	< 0.03	0.05	3. 12	< 0. 1	0. 004	4. 84
板栗 (N=45)	最大值	0. 27	0.30	7. 60	0.71	0. 16	18. 4
(17 = 43)	平均值	0. 10	0. 11	4. 93	0. 11	0. 03	7. 50

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1) 低于检测限的样品以检测限的 1/2 赋值计算平均值; 2) —表示无法计算

2.4 研究区土壤-农产品重金属迁移累积规律与影响因素

土壤是农产品中重金属的主要来源,鉴于 Cd 的高毒性和研究区土壤 Cd 的富集程度与活性,本 文重点探讨 Cd 在土壤-农产品系统的迁移累积规律 与影响因素. 从高于检测限的农产品对 Cd 的生物 富集系数(BCF:农产品 Cd 含量与土壤 Cd 含量之比)来看,不同农产品可食用部分对 Cd 的富集系数为:玉米(0.128)>土豆(0.064)>萝卜(0.045)>板栗(0.038)>草莓(0.033)>红薯(0.029),整体

属于微弱摄取^[37]. 玉米 Cd 的 BCF 高于前人所报导的 0.045^[13], 土豆和萝卜等根类作物对 Cd 的富集能力也较强. 筛选高于检测限的农产品样本与对应土壤 Cd 含量数据进行分析,结果表明: 板栗 Cd 含量与土壤 Cd 含量相关性较高(r=0.619); 其次为块根类农作物(萝卜、土豆和红薯),均与土壤 Cd含量呈一定正相关,其中红薯的相关性较高(r=0.507); 玉米和草莓 Cd含量与土壤 Cd含量无相关性(图5). 说明土壤 Cd含量高低可较好地指示块根类农产品中 Cd 的富集情况,这与前人在其它黑色岩系地质高背景区的研究结果相似^[20].

尽管农产品 Cd 含量与土壤 Cd 含量呈不同程 度的正相关,且Cd的活性态占比高,但农产品中Cd 的富集程度较低,可能受到其它因素影响.首先,1.0 mol·L-1MgCl, 提取的离子交换态并不等同于生物 有效态,前者常用于反映重金属的迁移和释放等环 境活性^[8,22], 而 0.01 mol·L⁻¹CaCl₂, 提取态是反映生 物有效性的常用指标[38~41],由表 4 和图 2 可知,研 究区土壤 ω (Cd)与 pH 呈显著正相关(r=0.223). 而 Cd 的生物有效性随 pH 的升高而降低, 当土壤 pH > 6 时, Cd 的生物有效性显著降低[13,38,39], 研究 区约60%的土壤样本 pH > 6,较高的 pH 导致土壤 Cd 生物有效性低,可能是农产品 Cd 富集程度低的 重要原因之一,类似现象也存在于岩溶地区土壤-水 稻系统[11]. 此外,值得关注的是,研究区还存在明显 的 Se 富集现象(表 3),土壤中一定含量范围的 Se 可降低 Cd 的生物有效性和根对 Cd 的吸收能 力[42,43],例如,盆栽试验研究结果表明,土壤基施硒 肥用量的增加可有效降低 Cd 的生物有效性和稻米 Cd 含量[44]; 野外调查结果表明,偏碱性的土壤条件 有利于水稻吸收 Se 而不利于吸收 Cd. 且在土壤 ω (Se)高于0.4 mg·kg⁻¹, ω (TOC)高于1.5%时,稻

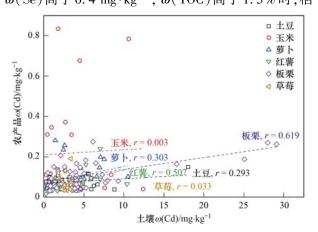


图 5 研究区农产品 Cd 含量与土壤 Cd 含量的关系

Fig. 5 Relationship between Cd concentrations in agricultural products and soils

米 Cd 含量与土壤 Se 含量存在显著负相关^[45]. 研究 区土壤基本符合该环境条件, 但农产品 Cd 含量与土壤 Se 含量和农产品 Se 含量之间并无明显相关性, 土壤 Se 富集对农作物吸收 Cd 的影响机制还有待于进一步研究.

3 结论

- (1)研究区土壤 Cd、Cr、Cu、Se 和 Zn 富集,Pb 不富集; Cd 的富集程度最高,98.7%的土壤样本 Cd 含量超过我国农用地土壤污染风险筛选值,47.3%的样本 Cd 含量超过风险管制值,近 50%的土地原则上应采取严格管控措施. Cd 的污染水平为中度和重度污染,潜在生态风险最高; Cr、Cu 和 Zn 以中度污染为主,Pb 无污染.
- (2)研究区土壤 Cd 主要以离子交换态存在(40.6%),其次为残渣态(19.1%)和弱有机结合态(16.6%),铁锰结合态(10.9%)、碳酸盐结合态(7.10%)和强有机结合态(4.40%)占比相对较低;Cr、Cu、Pb、Se 和 Zn 主要以残渣态存在,此外,有机结合态 Se 和 Cu 的占比也较高,铁锰结合态 Pb 的占比较高(30.1%).
- (3)研究区主要农产品对重金属的富集能力较弱,农产品 Cd 超标率约为 18.7%,其中草莓和板栗的超标率较高,但超标倍数相对较低;农产品重金属污染风险较低.

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