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重庆市煤矸山周边农产品镉健康风险评价及土壤环境 基准值推导

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摘要:以重庆市煤矸山周边农用地土壤和农产品(玉米和水稻)为研究对象,测定土壤和农产品(玉米和水稻)中 Cd 含量,评估摄入农产品(玉米和水稻)对人体的潜在健康风险,并基于物种敏感性分布法(SSD)推导土壤环境基准值.结果表明,重庆煤矸山周边旱地土壤 Cd 含量超风险筛选值的点位占 55.8%,水田土壤 Cd 含量超风险筛选值的点位占 31.6%,土壤 Cd 以较高生态危害和高生态危害为主,分别占 47.4% 和 36.8%.玉米 Cd 含量超标点位占 4.4%,水稻 Cd 含量均未超标.健康风险评价表明因食用玉米和水稻摄入 Cd 的非致癌健康风险可忽略,食用玉米摄入 Cd 存在可耐受致癌健康风险,食用水稻摄入 Cd 存在不可耐受致癌健康风险,且玉米和水稻 Cd 含量敏感度最高. SSD 推导出煤矸山周边旱地土壤在 $pH \le 5.5$ 、 $5.5 < pH \le 6.5$ 、 $6.5 < pH \le 7.5$ 和 pH > 7.5 时 Cd 的环境基准值分别为 0.491、0.382、0.376 和 0.588 mg·kg $^{-1}$,水田土壤 Cd 的环境基准值为 0.807 mg·kg $^{-1}$.水田土壤和旱地土壤 $pH \le 7.5$ 时,现行土壤标准(GB 15618-2018)相对偏宽松.应加强煤矸山周边土壤 Cd 污染防治和农产品安全利用研究,并对土壤 Cd 环境基准值进行适当调整.

关键词:煤矸山;土壤;镉(Cd);物种敏感性分布(SSD);玉米;水稻中图分类号: X820.4 文献标识码: A 文章编号: 0250-3301(2023)09-5264-11 **DOI**: 10.13227/j. hjkx. 202210159

Health Risk Assessment and Environmental Benchmark of Cadmium in Farmland Soils around the Gangue Heap of Coal Mine, Chongqing

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Abstract: To analyze the health risk assessment and environmental benchmark of cadmium in farmland soils surrounding the gangue heap of a coal mine in Chongqing, Hakanson, the ecological risk index and health risk assessment were used. Meanwhile, the soil environmental reference value of the regional cultivated land was inverted based on the species sensitive distribution model (SSD). The results showed that the dryland soil was polluted by Cd, with an over-standard rate of 55. 8%, and the paddy field soil was polluted by Cd, with an over-standard rate of 4.4%, and the rice was not polluted by Cd. The Hakanson ecological risk index showed that Cd was mainly characterized in soils by high ecological risk and considerable ecological risk. The health risk assessment indicated that Cd presented low non-carcinogenic risk by corn and rice; however, it showed acceptable carcinogenic risk by corn and unacceptable carcinogenic risk by rice in this study. The sensitivity analysis of health risks showed that the content of Cd was the most sensitive. The SSD inversion showed that the reference values for Cd in dryland soil of pH \leq 5.5, 5.5 < pH \leq 6.5, 6.5 < pH \leq 7.5, and pH > 7.5 had HC₅ values of 0.491, 0.382, 0.376, and 0.588 mg·kg⁻¹, respectively, and that for Cd in paddy soil had an HC₅ value of 0.807 mg·kg⁻¹. The reverse analysis showed that the HC₅ of Cd in dryland soil (pH \leq 7.5) was lower than the soil risk screening values, which showed that the current standard was relatively loose. However, the HC₅ of Cd in dryland soil (pH > 7.5) was lower than the soil risk screening values, which showed that the current standard was relatively strict. It is suggested that the current soil standard could be adjusted in this area.

Key words: gangue heap of coal mine; soil; cadmium(Cd); species sensitivity distributions(SSD); corn; rice

煤炭是中国主要的一次能源,约占全国能源消费总量的70%,但在开采和洗选过程中会产生大量废弃的煤矸石,占煤炭总产量的10%~15%^[1~3].据统计,我国煤矸石堆存量超过70亿t,已成为排放最多的工业废弃物之一,这不仅造成大量土地资源浪费,也对周边环境造成一定危害^[4~6].有研究表明,煤矸石长期露天堆放,在淋溶、风化和渗滤等作用下,会导致矸石中重金属析出,并扩散和迁移至周边土壤环境中,对土壤-植物系统产生毒性,最终通过食物链危害人体健康^[7~10].近年来,煤矸山周边土

壤重金属污染问题得到学者关注,主要研究集中在小尺度区域土壤污染评价^[11~13]、农产品污染评价^[14,15]和污染源解析^[16,17]等方面,且土壤和农产品污染特征总体表现为距煤矸山越近,污染程度越重.

对农产品污染而言,不仅关系到我国粮食安全问题,更关系到人体健康. 2013 年开始,我国粮食安

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全的目标确定为"谷物基本自足、口粮绝对安全", 其中谷物以水稻、小麦和玉米为主^[18].有研究表明,农产品经食用后其含有的污染物易于被人体直接吸收,应引起重视^[19].目前大多采用 USEPA 推荐的模型,根据污染物含量和相关暴露参数对人体健康进行等级划分^[20,21],但由于个体的差异性,利用固定参数无法准确识别对人体影响较大的因素,导致低估或高估健康风险水平^[22].因此,相关学者在健康风险评估中引入蒙特卡罗模拟开展敏感性分析,确定风险控制的优先要素^[23,24].

对土壤污染评价而言,大多采用《土壤环境质 量农用地土壤污染风险管控标准(试行)》(GB 15618-2018)中的风险筛选值和管制值作为评价限 值. 理论上, 当土壤重金属含量低于筛选值时, 农产 品的安全风险可忽略; 当土壤重金属含量高于管制 值时,农产品存在不同程度安全风险,应采取相应管 控措施. 但由于不同地区土壤、地质和气候等自然 条件迥异,不同农作物对重金属的富集能力也存在 差异,导致现行的土壤环境质量标准在实际应用中 出现土壤和农产品超标不对应的情况[25]. 如朱志军 等[26]研究发现,广西桂平市土壤 Cd 含量不超标但 水稻 Cd 含量超标占比 9.2%; 王旭莲等[27] 研究发 现,黔西北地质高背景区土壤 Cd 含量超标但马铃 薯 Cd 含量不超标占比 82.9%. 因此,相关学者运用 物种敏感性分布法(species sensitivity distributions, SSD) 反演计算不同农作物下的土壤环境基准 值[28]. 如刘海等[20] 通过小麦和水稻反演得到皖江 经济带土壤 Cd 环境基准值均为 0.13 mg·kg⁻¹: 徐 梦琪等[21] 通过玉米和水稻反演得到黔西北山区旱 地和水田土壤 Cd 环境基准值分别为 0.67 mg·kg⁻¹ 和 2. 42 mg·kg⁻¹; 韩东锦等^[29] 通过水稻反演得到 西南碳酸盐区不同土壤 pH下 Cd 环境基准值,pH≤ 5.5、5.5 < pH ≤ 6.5、6.5 < pH ≤ 7.5 和 pH > 7.5 下 Cd 环境基准值分别为 0.22、0.34、0.68 和 0.80 mg·kg⁻¹. 可见,不同地区农作物对应的土壤重金属 环境基准值存在较大差异,应因地制宜结合实际情 况进行调整.

重庆作为西南地区历史最悠久的煤炭工业基地,探明资源储量 2.1 亿 t,主要集中在渝南地区綦江、万盛和南川等区县,产业结构转型前,煤矿的过度开采,遗留下较多煤矸山^[30,31].目前,针对重庆市煤矸山周边农产品重金属健康风险的研究相对缺乏,且土壤重金属基准值的研究鲜有报道.马杰等^[14,16,17,32]研究发现,重庆市煤矸山周边土壤和农产品重金属污染以 Cd 为主,影响范围主要为煤矸山周边 1km 范围内,易受地表径流影响的区域.因

此,本文选取重庆市 12 座煤矸山周边农用地土壤和农产品(玉米和水稻)为研究对象,测定土壤和农产品(玉米和水稻)中 Cd 含量,基于蒙特卡罗模拟评估摄入农产品(玉米和水稻)对人体的潜在健康风险,并运用 SSD 方法推导土壤环境基准值,以期为重庆市煤矸山周边土壤重金属 Cd 污染防治和农业种植结构优化提供科学支撑.

1 材料与方法

1.1 研究区概况

研究区位于重庆市南部,属亚热带湿润季风气 候,雨热同季,以丘陵山地为主,主要地层包括:三叠 系雷口坡组、嘉陵江组和飞仙关组,二叠系梁山组、 栖霞组、茅口组、龙潭组和长兴组,志留系韩家店 组. 含煤地层集中在龙潭组,龙潭组自上而下分为五 段,其中一、三和五段为含煤段,主要发育海陆过渡 相和浅海碳酸盐沉积,聚煤区以障壁海岸沉积环境 为主,对煤储层的整体封盖能力较强[33,34].区内排 查近50座煤矸山,最终选取12座历史遗留无人管 理,且堆存时间较长、堆存量较大的煤矸山,其中5 座位于綦江区、3座位于万盛区和4座位于南川区 (图1和表1).由于煤矸山多位于煤矿开采区,因受 采煤影响,区域水土流失较为严重,大量水田改为旱 地[35]. 因此,煤矸山周边农用地土地利用类型以旱 地为主,土壤类型为黄壤或紫色土,主要农作物为玉 米, 其次是水田, 土壤类型为水稻土, 主要农作物为 水稻.

1.2 样品采集和测定

在煤矸山周边 1 km 范围内,易受地表径流影响的区域布设土壤和农产品采样点位.根据现场耕地和农产品种植情况,按照 150 m×150 m的网格进行点位布设,每座煤矸山布设 5~16 个采样点位(表1).每个点位土壤和农产品协同采样,采样方法为双对角线 5 点混合法.采集 0~20 cm 的表层土壤样品,混匀后质量不低于 1 kg;采集农产品样品,质量不低于 1 kg,土壤和农产品采集后分别装入聚乙烯塑料密封袋,并贴好样品标签带回实验室.2021 年 7 月共完成土壤样品采集 132 个,并协同采集 113 个玉米和 19 个水稻样品.

土壤样品经自然风干后,将测定 pH 的土壤过 2 mm 孔径筛,按 HJ 962-2018 要求^[36],用酸度计 (SevenExcellence)测定;将测定 Cd 的土壤过 0.15 mm 孔径筛,按照 GB/T 17141-1997 要求^[37],经盐酸-硝酸-氢氟酸-高氯酸全消解后,用石墨炉火焰原子吸收分光光度计(ZEEnit700P)测定.将玉米脱粒和水稻去壳后,籽实用自来水冲洗 3 遍,再用去离子

水冲洗3遍,放入带鼓风的专用烘箱,在60℃以下烘干,用谷物粉碎机加工至过0.42 mm 孔径筛,Cd按照 GB 5009.268-2016 微波消解进行前处理

后^[38],用电感耦合等离子体质谱仪(Xseries II)测定.每批次土壤和农产品样品均设置1组平行样,每组平行样分析测试结果的相对偏差均在8%以内.

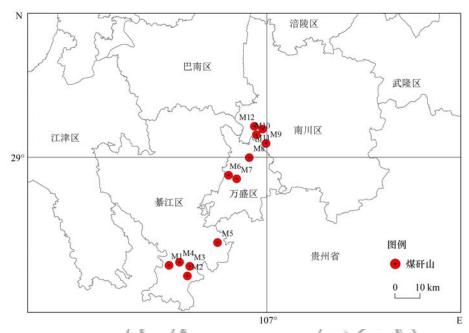


图 1 研究区煤矸山区位分布示意

Fig. 1 Location of coal gangue heap in the study area

表 1 研究区煤矸山基本信息及周边点位布设情况

1.3	Table 1	Characteristic	es of coal gangue	heaps and su	rrounding sampling sites	in the study area	~	100
编号	位置	占地面积 堆存时 堆存量 影		影响耕地且种植	土壤点位	农产品点位/个		
- H 5	W. L.	/万 m ²	间/a	/万 m³	农产品面积/万 m²	/个	玉米	水稻
M1	綦江打通镇	4. 0	13	50	15	8	8	0
M2	綦江石壕镇	8. 0	30	200	12	6	4	2
M3	綦江打通镇	0.7	20	10	30	16	16	0
M4	綦江打通镇	2. 9	13	80	25	12	12	0
M5	綦江赶水镇	1.0	42	150	15	7	7	0
M6	万盛南桐镇	10.0	10	110	15	8	4	4
M7	万盛万东镇	6. 2	32	100	25	12	12	0
M8	万盛丛林镇	1.5	18	3. 5	10	5	3	2
M9	南川南平镇	1.0	20	40	20	10	10	0
M10	南川南平镇	0.4	30	10	20	9	5	4
M11	南川南平镇	0.3	20	3	20	10	6	4
M12	南川南平镇	0.5	20	5	20	10	7	3

1.3 评价方法

1.3.1 潜在生态风险指数法

潜在生态风险指数法综合考虑了重金属性质、 生物毒性和生态效应等因素,并能定量评估重金属 生态危害程度^[39,40],如公式(1):

$$E_{\rm r} = T_{\rm r} \cdot C_i / C_{\rm s} \tag{1}$$

式中, E_r 为土壤重金属潜在生态风险指数; T_r 为土壤重金属毒性响应系数(Cd 取 30) $^{[41]}$; C_i 为土壤重金属实测值; C_s 为土壤背景值(Cd 取 0.11 $\text{mg}\cdot\text{kg}^{-1}$) $^{[42]}$. 潜在生态风险指数分为轻微生态危害(E_r < 40)、中等生态危害($40 \le E_r$ < 80)、较高生态危害($80 \le E_r$ < 160)、高生态危害($160 \le E_r$ < 320)

和极高生态危害($E_r \ge 320$).

1.3.2 健康风险评价

基于 USEPA 推荐的健康风险评价模型对人体 摄入农作物的致癌和非致癌健康风险进行评价^[43]. 农产品主要通过食用摄入,如公式(2):

$$EDI = \frac{C_p \times IR \times EF \times ED}{BW \times AT}$$
 (2)

式中,EDI 为农产品籽实重金属日均摄入量, C_p 为农产品籽实重金属实测值,其他参数含义见表 2.

非致癌风险计算如公式(3):

$$HQ = EDI/RfD$$
 (3)

式中,HQ 为重金属非致癌风险指数,RfD 为重金属

参考剂量[Cd 取 1.0 × 10⁻³ mg·(kg·d)⁻¹]^[44]. 非致癌风险分为无风险(HQ < 1)和有风险(HQ ≥ 1). 致癌风险计算如公式(4):

$$CR = EDI \times SF$$
 (4)

式中,CR 为重金属致癌健康风险指数.SF 为癌症斜率因子(Cd 取 6.1) $^{[44]}$. 致癌风险分为无风险(CR < 10^{-6})、人体可耐受风险($10^{-6} \le CR < 10^{-4}$)和人体不可耐受风险($CR \ge 10^{-4}$).

与传统的健康风险模型相比,蒙特卡罗模拟的健康风险评价模型首先要确定变量的分布函数,然后从变量分布中随机取样,并输出仿真结果的概率分布^[45].本研究采用 Oracle Crystal Ball 11.1.2.4 软件进行数据处理,每次运行的迭代次数设置为10 000,置信水平确定为 95%,求出风险评价的近似解.蒙特卡罗模拟的相关参数分布和取值见表 2.

表 2 基于蒙特卡罗的健康风险模型参数取值

Table 2 Calculation parameters and values used in health risk assessment model based on Monte Carlo simulation

参数	含义	单位	分布类型	取值	文献
EF	暴露频率	d•a ⁻¹	单点	350	[44]
IR	摄人率	g•d -1	三角	1.93、18.5和38.6(玉米) 263.5、279.1和293.9(水稻)	[46]
ED	暴露期	a	单点	24	[44]
BW	体重	kg	三角	42.4、56.4和73.7	[46]
AT	平均暴露时间	d	单点	365 × ED(非致癌) 365 × 70(致癌)	(J44) P

1.4 土壤环境基准值推导

土壤环境基准值推导一般采用敏感性分布曲线法(SSD),通过测定土壤和农产品重金属含量,计算BCF,公式如式(5). 然后将 1/BCF 作为概率分布指标,利用公式(6) 拟合 SSD 曲线^[28]和公式(7) 反演种植农作物土壤中 Cd 的基准值^[20]:

BCF =
$$C_p/C_i$$
 (5)

$$y = a + \frac{b}{1 + (x/x_0)^c}$$
 (6)

$$C_s = C_p/BCF$$
 (7)

式中,BCF 为富集系数, C_p 为农产品籽实重金属实测值, C_i 为土壤重金属实测值,x 为 1 /BCF;y 为作物样品对应的累积概率,a、b、c 和 x_0 为常数, C_s 为农作物 Cd 限值,参照《食品安全国家标准食品中污染物限量》(GB 2762-2022)规定玉米和水稻中 Cd 限值分别取 $0.1 \text{ mg} \cdot \text{kg}^{-1}$ 和 $0.2 \text{ mg} \cdot \text{kg}^{-1}$.

2 结果与讨论

2.1 土壤 Cd 含量及污染特征

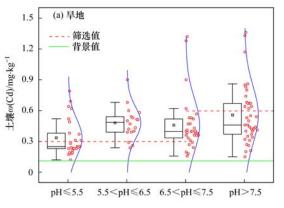
煤矸山周边农用地土壤 Cd 含量如表 3 所示. 旱

地和水田土壤 ω (Cd) 均值分别为 0.47 mg·kg⁻¹和 0.52 mg·kg⁻¹. 从变异系数(variable coefficient, CV) 来看,旱地和水田土壤 CV 分别为 0.54 和 0.49,均属 于中等程度变异(0.1 \leq CV <1)^[44],说明旱地和水田土壤 Cd 含量分布不均匀,受外界人为因素影响较大.

与现行土壤标准(GB 15618-2018)相比(图 2),旱地土壤 Cd 含量超风险筛选值的样品有 63 个,占旱地土壤样品总数的 55.8%,其中 pH≤5.5 的样品超标率为 40.0%,5.5 < pH≤6.5 的样品超标率为 77.8%,6.5 < pH≤7.5 的样品超标率为 80.6%,pH > 7.5 的样品超标率为 22.2%.水田土壤 Cd 含量超风险筛选值的样品有 6 个,占水田样品总数的 31.6%,其中 pH≤5.5 的样品超标率为 25.0%,5.5 < pH≤6.5 的样品超标率为 50.0%,6.5 < pH≤7.5 的样品超标率为 50.0%,6.5 < pH≤7.5 的样品超标率为 50.0%,pH > 7.5 的样品均未超标,说明旱地和水田在中性和酸性土壤中 Cd 超标率较高.与重庆市土壤背景值相比(图 2),旱地和水田土壤 Cd 含量均高于土壤 Cd 背景值,说明旱地和水田土壤 Cd 存在明显累积.与相关研究相比(表3),重庆煤矿区周边土壤Cd含量均值明显高于煤

表 3 土壤 Cd 含量统计情况

		Table 3	Statistical characte	eristics of Cd in se	011			
研究对象		最小值	最大值	平均值	标准差	变异系数	文献	
		$/\mathrm{mg} \cdot \mathrm{kg}^{-1}$	/mg⋅kg ⁻¹	/mg·kg ⁻¹	/mg·kg ⁻¹	文开示奴	文帆	
重庆煤矸山周边	旱地土壤(n=113)	0.12	1.36	0.47	0.25	0.54	本研究	
	水田土壤(n=19)	0.18	1.07	0.52	0.25	0.49	平明九	
重庆渝北区农用地土壤(n=71)		0.06	0. 58	0. 26	0.11	0. 43	[51]	
重庆黔江区农用地土壤(n=118)		0.09	11.4	0.34	1.05	3. 05	[52]	
重庆垫江县农用地土壤(n=44)		0. 16	0.46	0. 28	0.71	0. 26	[53]	
三峡库区(重庆段)农用地土壤(n=276)		0.06	0. 73	0. 25	0. 14	0. 54	[54]	
重庆永川煤矿区土壤(n=8)		1. 91	4. 44	2. 97	0. 91	0. 31	[55]	
重庆煤矿区周边土壤(n=27)		0.05	8. 91	0. 98	1.64	1. 67	[56]	



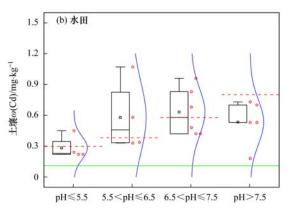


图 2 不同 pH 区间土壤 Cd 含量统计

Fig. 2 Statistical characteristics of Cd in soil with different pH conditions

矸山周边土壤,这可能是因为煤矿周边土壤受矿业活动影响,导致重金属累积^[47~50]. 而煤矸山周边土壤 Cd 含量均值高于重庆一般农用地区域,说明煤矸石长期露天堆放,对周边土壤环境会造成一定污染^[7~10,16,17,32].

潜在生态风险指数法能重点反映 Cd 等生物毒 性相对较高的重金属元素对土壤环境质量的影 响[19]. 煤矸山周边土壤 Cd 潜在生态风险指数评价 结果如图 3 所示. 旱地土壤 Cd 评价为极高生态危害 的样品 $(E_r \ge 320)$ 占比 6.2%, 高生态危害 $(160 \le E_r$ <320)占比37.2%,较高生态危害(80≤E_r<160) 占比 46.0%, 中等生态危害 $(40 \le E, < 80)$ 占比 10.6%. 水田土壤 Cd 评价为极高生态危害(E,≥ 320)的样品占比 5.3%, 高生态危害(160 ≤ E_r < 320) 占比 36.8%, 较高生态危害(80≤E_r<160) 占比 47.4%, 中等生态危害 (40 $\leq E_{c} < 80$) 占比 10.5%. 综上,旱地和水田土壤评价结果基本一 致,以较高生态危害(80≤E, <160)和高生态危害 $(160 ≤ E_r < 320)$ 为主. 有研究表明 Cd 的活性和迁 移能力较强,易被植物吸收,可能对农产品安全及 人体健康造成威胁[57]. 因此,应加强对煤矸山周边 农用地土壤 Cd 的污染防治.

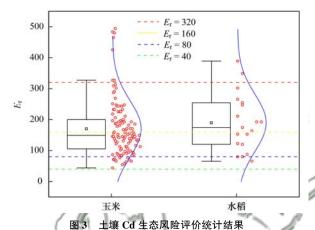


图 3 上壤 Cu 主意风险计训练时 结未 Fig. 3 Ecological risk assessment results of Cd in soil

2.2 农产品 Cd 含量及污染特征

煤矸山周边农产品 Cd 含量如表 4 所示. 玉米和 水稻 ω (Cd) 均值分别为 0.021 mg·kg⁻¹ 和 0.026 mg·kg⁻¹. 从 CV 来看,玉米 CV 为 1.64,属于强变异 (CV > 1);水稻 CV 为 0.71,属于中等变异(0.1~1),说明玉米 Cd 含量差异性更明显. 与现行农产品标准(GB 2762-2022) 相比,玉米 Cd 含量超标的样品有 5 个,占玉米样品总数的 4.4%,水稻 Cd 含量均未超标. 与重庆相关研究相比(表 4),煤矸山周边玉米和水稻 Cd 超标率处于相对较低水平.

表 4 农产品 Cd 含量统计情况¹⁾

Table 4 Statistical characteristics of Cd in corn and rice

研究	对象	最小值 /mg·kg ⁻¹	最大值 /mg·kg ⁻¹	平均值 /mg·kg ⁻¹	标准差 /mg·kg ⁻¹	变异系数	超标率 /%	文献
重庆煤矸山周边	玉米(n=113)	0.002	0.263	0.021	0.035	1.64	4.4	本研究
	水稻(n=19)	0.003	0.067	0.026	0.019	0.71	0	
重庆江津区	玉米(n=64)	0.003	0.258	0.033	0.053	1.61	9.4	[58]
	水稻(n=63)	0.005	0.246	0.037	0.051	1.38	4.8	
渝东南汞矿区	玉米(n=32)	0.006	0.202	0.058	0.051	0.88	15.6	[59]
	水稻(n=45)	0.004	1.38	0.202	0.339	1.68	20	
重庆梁平区	玉米(n=12)	0.005	0.017	0.012	0.015	1.29	0	[60]
	水稻(n=25)	0.002	0.365	0.060	0.106	1.77	8	

¹⁾参照《食品安全国家标准 食品中污染物限量》(GB 2762-2022), 玉米和水稻 Cd 超标限量分别为 0.1 mg·kg⁻¹和 0.2 mg·kg⁻¹

2.3 农作物 Cd 摄入健康风险评价

人体食用煤矸山周边玉米和水稻摄入 Cd 的非致癌风险指数如图 4 所示. 食用玉米和水稻摄入 Cd 非致癌风险指数(HQ)均小于 1,均值分别为0.0007和 0.132. 说明食用玉米和水稻的非致癌健康风险可忽略. 人体食用煤矸山周边玉米和水稻摄入 Cd

的致癌风险指数如图 5 所示. 食用玉米摄入 Cd 致癌风险指数 (CR)大于 1.00×10⁻⁶的点位占比31.9%,均值为 1.46×10⁻⁶,说明食用玉米存在可耐受致癌风险;食用水稻摄入 Cd 致癌风险指数(CR)大于 10⁻⁴的点位占比84.2%,均值为 2.76×10⁻⁴,说明食用水稻存在不可耐受致癌风险.

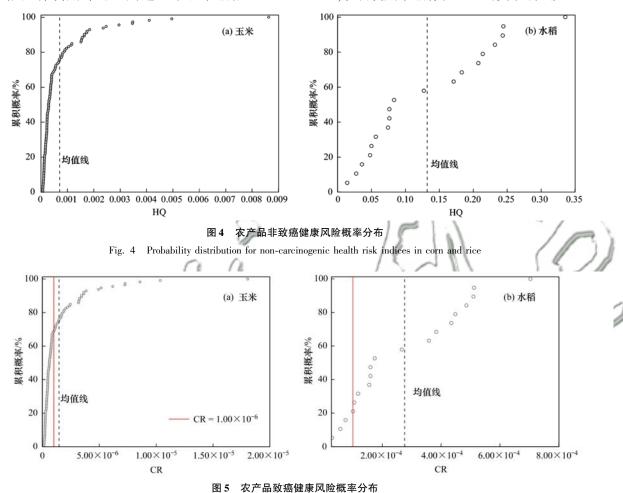


Fig. 5 Probability distribution for carcinogenic health risk indices in corn and rice

人体食用煤矸山周边玉米和水稻敏感性结果表明玉米和水稻 Cd 含量敏感度最高,分别为 95.1% 和 97.4%,敏感度值越大则其对风险结果的影响越大,且敏感度为正值,则表示与风险结果正相关^[61],说明玉米和水稻 Cd 含量是影响健康风险水平的主要因素,此外玉米和水稻摄入量和体重两个参数也具有一定敏感度.有研究表明,农产品中 Cd 摄入人体是构成致癌健康风险的关键因子,会诱发肾脏受损、神经毒害、糖尿病等器官损伤和疾病^[20,62].因此,煤矸山周边土壤和农产品应将 Cd 作为首要污染元素,进一步开展土壤污染防治和农产品安全利用研究.

2.4 土壤 Cd 环境基准值预测

SSD 模型一般包括 log-normal、log-logistic、Burr III、Weibull 和 Gamma 累积分布概率分布函数 等^[63,64],本研究选取 log-logistic 分布模型拟合玉米和水稻中 Cd 敏感性分布(SSD) 曲线,如图 6 所示. SSD 曲线拟合度 R^2 在 0.979 ~ 0.989,拟合度较高,说明模型结果可靠.有研究表明,点位集中位于 SSD 曲线上端,说明农产品对重金属的吸收富集能力较弱,敏感性较低,反之亦然^[20,21]. 煤矸山周边玉米在不同土壤 pH 区间 Cd 富集特征存在一定差异,其中土壤 pH \leq 5.5 和 5.5 < pH \leq 6.5 时,曲线上端点位较少,说明富集 Cd 能力较强,敏感性较高;土壤6.5 < pH \leq 7.5 和 pH > 7.5 时,曲线上端点位较多,说明富集 Cd 能力较弱,敏感性较低. 这与已有研究的结果基本一致,因为重金属在碱性土壤环境中容易失活,较高的 OH 含量会降低 Cd²⁺的活性,使其不易被农作物吸收富集^[65,66]. 鉴于水稻点位较少,故不划分土壤 pH 区间进行分析,研究发现水稻在

曲线上端点位较少,说明富集 Cd 能力较强,敏感性较高. 玉米和水稻相比,玉米大多数点位介于 0~100之间,水稻大多数点位介于 0~50之间,水稻曲线斜率较玉米陡,说明水稻富集 Cd 能力较玉米强.有研究表明水稻根系中 Cd 的运输速率显著高于玉米[67],这与徐梦琪等[21]研究的结果基本一致.

基于 log-logistic 分布模型拟合获取保护 95% 的作物安全临界值 (hazardous concentration for 5% of species, HC_5),即可保障区域 95% 种植农产品可食部位的重金属含量低于安全限值,该方法已被多个

国家确立为制定环境质量基准的方法 $^{[63,64]}$. 煤矸山周边土壤 Cd 环境基准值结果如表 5 所示,旱地土壤在 $pH \le 5.5$ 、 $5.5 < pH \le 6.5$ 、 $6.5 < pH \le 7.5$ 和 pH > 7.5 的环境基准值分别为 0.491、0.382、0.376 和 0.588 $mg \cdot kg^{-1}$,水田土壤 Cd 的环境基准值为 0.807 $mg \cdot kg^{-1}$. 与现行土壤标准 $(GB\ 15618-2018)$ 相比,煤矸山周边除旱地土壤 pH > 7.5 时,较现行土壤标准 $(GB\ 15618-2018)$ 略偏宽松外,对于旱地土壤 $pH \le 7.5$ 和水田土壤均偏严,建议根据土壤和农产品 Cd 污染状况对环境基准值进行适当调整.

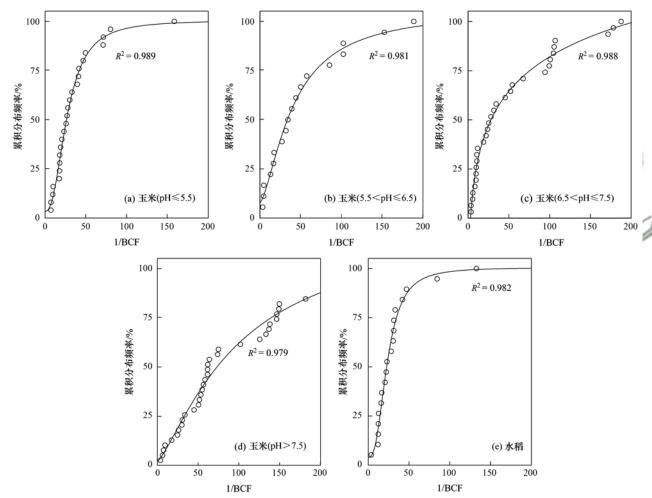


图 6 玉米与水稻中 Cd 敏感性分布(SSD)曲线

Fig. 6 Species sensitivity distributions (SSD) of Cd in corn and rice

表 5 基于 log-logistic 分布模型下农用地土壤 Cd 环境基准值划分/ $mg\cdot kg^{-1}$

Environmental benchmark of Cd in farmland soils based on log-logistic model/mg·kg⁻¹ log-logistic 模型临界值(HCs) GB 15618-2018 风险筛选值 农用地 农产品 土壤 pH pH≤5.5 0.491 0.3 5. 5 < pH ≤ 6. 5 0.382 0.3 旱地 玉米 6. 5 < pH ≤ 7. 5 0.376 0.3 pH > 7.50.588 0.6 水田 水稻 未划分区间 0.807 $0.3 \sim 0.8$

3 结论

(1) 重庆煤矸山周边农用地土壤 Cd 含量均高

于背景值,旱地和水田土壤 ω (Cd)均值分别为 0. 47 mg·kg⁻¹ 和 0. 52 mg·kg⁻¹. 与现行土壤标准 (GB 15618-2018)相比,旱地土壤 Cd 含量超风险筛选值

的点位有63个,占比55.8%,水田土壤Cd含量超风险筛选值的点位有6个,占比31.6%.旱地和水田土壤Cd以较高生态危害和高生态危害为主,分别占47.4%和36.8%.应加强对煤矸山周边农用地土壤Cd的污染防治.

- (2)重庆煤矸山周边玉米 Cd 超标的样品有 5 个,占玉米样品总数的 4.4%,水稻 Cd 含量均未超标.因食用玉米和水稻摄入 Cd 的非致癌健康风险可忽略,HQ 均值分别为0.0007和 0.132.因食用玉米摄入 Cd 存在可耐受致癌健康风险,CR 均值为1.46×10⁻⁶.因食用水稻摄入 Cd 存在不可耐受致癌健康风险,CR 均值 2.76×10⁻⁴.玉米和水稻 Cd 含量敏感度最高,分别为 95.1% 和 97.4%,应开展对煤矸山周边农产品安全利用研究.
- (3) SSD 推导出煤矸山周边旱地土壤 Cd 在 pH \leq 5.5、5.5 < pH \leq 6.5、6.5 < pH \leq 7.5 和 pH > 7.5 的环境基准值分别为 0.491、0.382、0.376 和 0.588 mg·kg⁻¹, 水田土壤 Cd 的环境基准值为 0.807 mg·kg⁻¹. 水田土壤和旱地土壤 pH \leq 7.5 时,现行土壤标准(GB 15618-2018)相对偏严;旱地土壤 pH > 7.5 时,现行土壤标准(GB 15618-2018)相对偏严;旱地土壤 pH > 7.5 时,现行土壤标准(GB 15618-2018)相对偏宽松. 应根据土壤和农产品 Cd 污染状况对土壤环境基准值进行调整.

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