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基于源导向的农用地土壤重金属健康风险评估及优先控制因子分析

马杰^{1,2}, 葛淼^{1,2}, 王胜蓝^{1,2}, 邓力^{1,2}, 孙静^{1,2*}, 蒋月^{1,2}, 周林^{1,2}

(1. 重庆市生态环境监测中心, 重庆 401147; 2. 有机污染物环境化学行为与生态毒理重庆市重点实验室, 重庆 401147)

摘要: 以重庆市农产品主产区为研究对象, 运用正定矩阵因子分解(PMF)受体模型对土壤重金属进行源解析, 运用蒙特卡罗模拟的健康风险评估(HRA)模型, 探析土壤重金属对人体的健康风险, 并将PMF受体模型和HRA模型结合, 探讨不同污染源影响下的土壤健康风险, 确定优先管控要素。结果表明, 研究区土壤Cd均值含量远高于背景值, Cr均值含量低于背景值, As、Pb、Cu、Ni和Zn均值含量与背景值基本持平。PMF受体模型源解析结果表明, 研究区土壤受自然源、工业源和农业源影响, 贡献率分别为35%、24%和41%。HRA模型评估结果表明, 研究区土壤重金属对儿童和成人存在可耐受致癌健康风险($1.00E-6 < TCR \leq 1.00E-4$), 非致癌健康风险可忽略($HI \leq 1$), 经口摄入是主要的暴露途径。土壤重金属、污染源与健康风险关系的分析结果表明, 研究区工业源和As为首要管控要素, 农业源和Cd为次要管控要素。研究结果为研究区有针对性开展土壤污染防治和提供降低土壤污染管理成本提供科学支撑。

关键词: 土壤; 重金属; 正定矩阵因子分解(PMF)模型; 蒙特卡罗; 健康风险评估; 重庆

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Health Risk Assessment and Priority Control Factors Analysis of Heavy Metals in Agricultural Soils Based on Source-oriented

MA Jie^{1,2}, GE Miao^{1,2}, WANG Sheng-lan^{1,2}, DENG Li^{1,2}, SUN Jing^{1,2*}, JIANG Yue^{1,2}, ZHOU Lin^{1,2}

(1. Chongqing Ecological and Environmental Monitoring Center, Chongqing 401147, China; 2. Key Laboratory of Organic Pollutants in Environmental Chemical Behavior and Ecological Toxicology of Chongqing, Chongqing 401147, China)

Abstract: To analyze the source apportionment and health risk of heavy metals in agricultural soils of major producing areas of agricultural products in Chongqing, a positive matrix factorization (PMF) model and health risk assessment (HRA) model based on Monte Carlo simulation were used. Meanwhile, both the PMF and HRA model were combined to explore health risks of heavy metals in agricultural soils by different pollution sources in order to determine the priority control factors. The results showed that the average values of Cd concentration were higher than its corresponding background value; the average values of Cr concentration were lower than its corresponding background value; and the average values of As, Pb, Cu, Ni, and Zn concentration were basically consistent with their corresponding background values. Using PMF model analysis, natural sources, industrial sources, and agricultural sources were identified as the determinants for the accumulation of heavy metals in agricultural soils, with the contribution rates of 35%, 24%, and 41%, respectively. Using the HRA model based on Monte Carlo simulation analysis, carcinogenic risks of adult and children were tolerable ($1.00E-6 < TCR \leq 1.00E-4$), whereas non-carcinogenic risks were acceptable ($HI \leq 1$). Oral ingestion was the main exposure pathway. The analysis results of the relationship among heavy metals, pollution sources, and health risks showed that industrial pollution and As were identified as priority control factors, and agricultural pollution and Cd were identified as secondary control factors. Our findings provide scientific support for decision makers to control soil pollution and reduce the management costs of soil pollution.

Key words: soil; heavy metals; PMF model; Monte Carlo; health risk assessment; Chongqing

根据全国土壤污染状况调查公报显示, 我国土壤以无机型污染为主, 无机污染物超标点位占全部超标点位的82.8%, 其中Cd、As、Pb、Cr、Cu、Ni和Zn超标率分别为7.0%、2.7%、2.1%、1.5%、1.1%、0.9%和4.8%, 不同土地利用类型中耕地污染占比最高^[1]。因土壤重金属污染具有毒性大、隐蔽性强、持久性和降解难等特点, 对农业生产、粮食安全和人体健康等方面已构成潜在威胁^[2-5]。有研究表明, 土壤重金属直接经口、皮肤和呼吸摄入是影响人体健康的3种重要暴露途径^[6-8], 同时, 土壤重金属也易被农作物根部吸收, 并在农产品中积累, 通过食物链进入人体, 威胁人体健康^[2,9]。长期摄入Cd、As、Pb、Cr和Ni等重金属会影响人体肾、肺和其他器官功能, 造成

神经系统、心血管和呼吸道等疾病, 诱发癌症几率增高^[13-16]。然而土壤中重金属来源广泛, 除部分受成土母质等自然源影响外, 更多的受人为源影响, 如化石燃料燃烧、采矿、冶炼、交通排放、废水灌溉和化肥农药过度使用等^[17-20]。因此, 掌握土壤重金属污染水平, 开展源解析和健康风险评估, 对土壤污染管控、耕地安全利用和保障人体健康安全至关重要。

当前土壤污染源解析方法较多, 其中正定矩阵因子分解(positive matrix factorization, PMF)受体模型

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作者简介: 马杰(1986~), 男, 硕士, 高级工程师, 主要研究方向为土壤和农村生态环境监测与评价, E-mail: pony312@qq.com

* 通信作者, E-mail: 58000915@qq.com

是应用最为广泛的源解析方法之一^[16,21,22],其优点在于不需要提前构建污染源成分谱,方便高效,能对因子分解矩阵进行非负约束,获得更准确的源解析结果^[23,24]。土壤健康风险评估则主要采用USEPA推荐的健康风险(health risk assessment, HRA)模型,模型一般选取固定暴露参数评估,但因个体的差异性,固定参数会导致健康风险水平被低估或高估^[14]。因此,学者们引入蒙特卡罗不确定性分析模型,在确定暴露参数先验分布下生成随机数进行迭代计算,并通过概率分布的形式表达,提高评估结果的准确性,已被广泛用于土壤健康风险评估^[25~28]。当前大量研究集中在土壤污染源解析或者健康风险评估上,但两者相结合,开展以污染源为导向的土壤健康风险评估则较少^[29]。鉴于土壤污染主要来自自然源和人为源,而自然源存在不可控性,管控的关键应放在降低人为源对土壤的污染上。因此,在资源和资金等投入有限的情况下,开展源导向的土壤健康风险评估,确定优先管控要素,对有针对性制定相应管控措施,降低土壤对人体的健康风险具有重要的现实意义。

重庆作为西南地区唯一的直辖市,集“大城市、大农村、大山区、大库区”于一体,区域性发展差距较大^[30]。根据重庆市自然资源保护和利用“十四五”规划,将全市38个区县划分为重点生态功能区、农产

品主产区和城市化发展区^[31]。本研究选取忠县和垫江县2个农产品主产区为研究对象,测定土壤重金属Cd、As、Pb、Cr、Cu、Ni和Zn的含量,分析土壤重金属污染特征,运用PMF受体模型开展土壤重金属源解析,蒙特卡罗模型对土壤重金属健康风险进行概率评估,并将PMF受体模型和HRA模型相结合,探讨不同污染源影响下的土壤健康风险,以期为研究区有针对性开展土壤污染防治和降低土壤污染管理成本提供科学支撑。

1 材料与方法

1.1 研究区概况

研究区位于长江中上游地区,重庆东北部,包括忠县和垫江县(图1),面积为3 705 km²,介于107°13′~108°14′E,29°38′~30°53′N之间。东邻万州区,南接涪陵区、长寿区和石柱县,西连四川省大竹县、邻水县,北靠梁平区;地貌以丘陵为主,属川东褶皱带平行岭谷区;气候属亚热带湿润季风气候,全年四季分明,雨量充沛,年均温约17.5℃,年均降水量约1 200 mm,农用地面积约为1 500 km²,土壤以紫色土和水稻土为主,垫江县以种植水稻和玉米为主,忠县以种植水稻、玉米和特色产业柑橘为主^[32~34]。垫江县和忠县城区(工业区)分别位于研究区中东部和中西部。

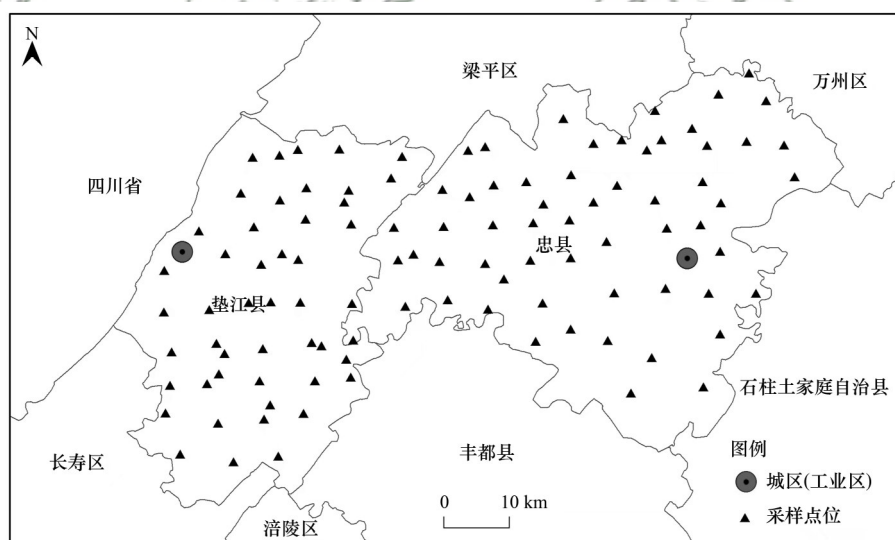


图1 研究区区位及采样点位分布示意

Fig. 1 Location of the study area and distribution of sampling sites

1.2 样品采集和测定

研究区按网格法(6 km × 6 km)布点,对农用地面积超过50%的网格布设1个采样点,共布设106个土壤采样点位(图1),每个采样点选取面积较大的旱地田块,且距周边高速公路、国道等主要交通干道、工矿企业等潜在污染源1 km以上。现场采用5点混合法采集0~20 cm表层土壤,剔除动植物残体和碎石等

杂物,装入聚乙烯塑料密封袋,贴好标签,并用GPS记录采样点信息。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^[35],用酸度计(SevenExcellence)测定;将测定Cd和As的土壤过0.149 mm孔径筛,Cd按GB/T 17141-1997要求^[36],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;As按GB/T

22105.2-2008 要求^[37],经硝酸-盐酸混合试剂在沸水浴中加热消解后,用原子荧光光度计(AFS-9750)测定;将测定 Pb、Cr、Cu、Ni 和 Zn 的土壤过 0.075 mm 孔径筛,按照 HJ 780-2015 要求^[38],用 X 荧光光谱仪(S8 TIGER)测定.测试过程同步测定 3 个平行样和 3 个土壤成分分析标准物质(GSS-8),平行样中各重金属含量相对偏差均在 8% 以内,标准物质各重金属含量也均在限值范围内,符合质量控制要求.

1.3 PMF 受体模型

利用 USEPA 推荐的 PMF 受体模型进行土壤源解析,其原理是将重金属元素含量矩阵分解为因子贡献和因子残差矩阵,然后基于各个重金属污染源的 特征确定不同因子的贡献率^[39],如公式(1):

$$X_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (1)$$

式中, X_{ij} 为第*i*个样本中*j*种重金属含量, f_{kj} 为*j*种重金属在源*k*中的含量, g_{ik} 为源*k*对第*i*个样本的贡献, e_{ij} 为残差矩阵.

同时,通过加权最小二乘法进行限定和迭代计算,不断分解矩阵*X*,选择最优的矩阵*g*和*f*,得到最小目标函数*Q*,如公式(2):

$$Q = \sum_{j=1}^m \sum_{i=1}^n \left(\frac{X_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right)^2 \quad (2)$$

表 1 基于蒙特卡罗的健康风险模型参数取值¹⁾

Table 1 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
				儿童	成人	
EF	暴露频率	d·a ⁻¹	三角	180、345 和 365		[41]
R _{ingest}	土壤颗粒摄入速率	mg·d ⁻¹	三角	66、103 和 161	4、30 和 52	[14]
SL	皮肤黏附系数	mg·cm ⁻²	对数正态	0.65 和 1.2	0.49 和 0.54	[41]
R _{inhal}	土壤颗粒吸入速率	m ³ ·d ⁻¹	单点	8.6	19	[42]
ED	暴露期	a	单点	6	24	[15]
PEF	颗粒物释放因子	m ³ ·kg ⁻¹	单点	1.36×10 ⁹		[15]
BW	体重	kg	正态	16.68 和 1.48	56.4 和 11.9	[42,43]
AT	平均暴露时间	d	单点	365×ED(非致癌)和 365×70(致癌)		[15]
SA	皮肤暴露面积	m ²	单点	0.23	0.54	[42]
ABF	皮肤吸收因子	—	单点	0.001(非致癌)和 0.01(致癌)		[15]

1) “—”表示无量纲

通过日均土壤摄入量计算非致癌风险指数和致癌风险指数,分别如公式(7)和公式(8):

$$HI = \sum HQ = \sum \frac{ADD_i}{RfD_i} \quad (7)$$

$$TCR = \sum CR = \sum ADD_i \times SF_i \quad (8)$$

式中,HQ 和 HI 分别为单一和综合非致癌风险指数;CR 和 TCR 分别为单一和综合致癌风险指数;RfD_{*i*}为土壤重金属*i*的参考剂量;SF_{*i*}为土壤重金属*i*的斜率

根据重金属含量与 MDL 关系, u_{ij} 计算如公式(3):

$$u_{ij} = \begin{cases} \frac{5}{6} \times MDL & (C_{ij} \leq MDL) \\ \sqrt{(RSD \times X_{ij})^2 + MDL^2} & (C_{ij} > MDL) \end{cases} \quad (3)$$

式中, u_{ij} 为第*i*个样本中*j*种重金属含量的不确定性大小;RSD 为重金属含量的相对标准偏差; C_{ij} 为第*i*个样本中*j*种重金属含量;MDL 为方法检出限,Cd、As、Pb、Cr、Cu、Ni 和 Zn 检出限分别为 0.01、0.01、2.0、3.0、1.2、1.5 和 2.0 mg·kg⁻¹.

1.4 蒙特卡罗模拟的健康风险评估

利用 USEPA 推荐的 HRA 模型对成人和儿童的健康风险进行评估^[40].主要暴露途径(经口摄入、皮肤摄入和呼吸摄入)下日均土壤摄入量的计算分别如公式(4)~(6):

$$ADD_{i, ingest} = \frac{C_i \times R_{ingest} \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (4)$$

$$ADD_{i, dermal} = \frac{C_i \times SA \times SL \times ABF \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (5)$$

$$ADD_{i, inhal} = \frac{C_i \times R_{inhal} \times EF \times ED}{PEF \times BW \times AT} \quad (6)$$

式中,ADD_{*i, ingest*}、ADD_{*i, dermal*}和 ADD_{*i, inhal*}分别为土壤重金属*i*经口、皮肤和呼吸日均摄入量; C_i 为土壤重金属*i*实测值.其他参数含义见表 1.

因子.具体取值见表 2.当 HQ/HI ≤ 1 时,说明非致癌风险可忽略,反之则存在非致癌风险.当 CR/TCR ≤ 1.00E-6 时,说明致癌风险可忽略,1.00E-6 < CR/TCR ≤ 1.00E-4 时,说明存在可耐受致癌风险,CR/TCR > 1.00E-6 时,说明存在不可耐受致癌风险.

在 HRA 模型中引入蒙特卡罗不确定性分析,首先确定各变量的分布函数,然后从变量分布中随机取样,并输出仿真结果的概率分布^[44,45].本研究采用

表 2 参考剂量和斜率因子取值¹⁾
Table 2 Corresponding reference dose and slope factor values

元素	RfD _i /mg·(kg·d) ⁻¹			SF _i /mg·(kg·d) ⁻¹			文献
	经口摄入	皮肤摄入	呼吸摄入	经口摄入	皮肤摄入	呼吸摄入	
Cd	1.00E-3	1.00E-5	1.00E-5	6.1E+0	—	6.30E+0	[15]
As	3.00E-4	1.23E-4	1.23E-4	1.5E+0	3.66E+0	1.51E+1	[15]
Pb	3.50E-3	5.25E-4	3.52E-3	8.50E-3	—	—	[27]
Cr	3.00E-3	6.00E-5	2.86E-5	8.50E-3	—	4.20E+1	[15]
Cu	4.00E-2	1.20E-2	4.02E-2	—	—	—	[27]
Ni	2.00E-2	5.40E-3	2.06E-2	—	—	8.4E-1	[15]
Zn	3.00E-1	6.00E-2	3.00E-1	—	—	—	[27]

1) “—”表示无数据

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 1.

1.5 源导向的健康风险评估

结合 PMF 受体模型解析获取的不同污染源上各重金属贡献率和 HRA 模型获取的致癌和非致癌风险指数,计算不同污染源对致癌和非致癌风险的贡献率,如公式(9)~(12):

B_{i,HQ} = ∑ C_{in} × HQ_n (9)

B_{i,CR} = ∑ C_{in} × CR_n (10)

D_{i,HQ} = C_{in} / B_{i,HQ} (11)

D_{i,CR} = C_{in} / B_{i,CR} (12)

式中,B_{i,HQ}为第*i*类污染源的非致癌风险指数;B_{i,CR}为第*i*类污染源的致癌风险指数;C_{in}为第*i*类污染源中

第*n*种重金属的贡献率(%);HQ_n为第*n*种重金属的非致癌风险指数;CR_n为第*n*种重金属的致癌风险指数;D_{i,HQ}为第*i*类污染源在非致癌风险指数的贡献率(%);D_{i,CR}为研究区域第*i*类污染源在致癌风险指数的贡献率(%).

2 结果与讨论

2.1 土壤重金属含量特征

研究区土壤重金属含量如表 3 所示. 土壤 pH 值在 4. 63~8. 72 之间,ω(Cd)、ω(As)、ω(Pb)、ω(Cr)、ω(Cu)、ω(Ni)和 ω(Zn)平均值分别为 0. 27、5. 78、26. 2、63. 2、23. 6、32. 8 和 77. 3 mg·kg⁻¹. 与重庆土壤背景值^[46]相比,研究区土壤 Cd 均值含量是背景值的 2. 46 倍;土壤 Cr 均值含量是背景值的 0. 79 倍;As、Pb、Cu、Ni 和 Zn 均值含量与背景值基本持平. 与 GB 15618-2018 风险筛选值^[47]相比,17. 9% 的点位土壤 Cd 超风险筛选值,其他 6 项重金属含量均未超风险筛选值. 说明研究区重金属污染以 Cd 为主.

表 3 土壤重金属含量统计情况¹⁾
Table 3 Statistical characteristics of heavy metal concentrations in soil

统计	pH	Cd	As	Pb	Cr	Cu	Ni	Zn
最小值/mg·kg ⁻¹	4.63	0.08	2.00	11.6	24.5	5.0	16.0	20.3
最大值/mg·kg ⁻¹	8.72	0.60	12.4	42.6	149	40.2	59.9	118
平均值/mg·kg ⁻¹	—	0.27	5.78	26.2	63.2	23.6	32.8	77.3
变异系数/%	—	38.1	40.6	21.9	27.8	25.5	25.7	27.5
重庆土壤背景值/mg·kg ⁻¹	—	0.11	5.00	26.0	80.0	26.0	32.0	80.0
GB 15618-2018 风险筛选值/mg·kg ⁻¹	pH > 7.5	0.6	25	170	250	100	190	300
	6.5 < pH ≤ 7.5	0.3	30	120	200	100	100	250
	5.5 < pH ≤ 6.5	0.3	40	90	150	50	70	200
	pH ≤ 5.5	0.3	40	70	150	50	60	200

1) “—”表示无数据

变异系数(coefficient of variation,CV)能反映变量的分散程度,常用于反映土壤重金属受人为活动影响的程度^[48]. 研究区重金属变异程度均为中等变异水平,具体表现为 As (40. 6%) > Cd (38. 1%) > Cr (27. 8%) > Zn (27. 5%) > Ni (25. 7%) >

Cu (25. 5%) > Pb (21. 9%). 说明研究区 As 和 Cd 受人为活动影响相对更大.

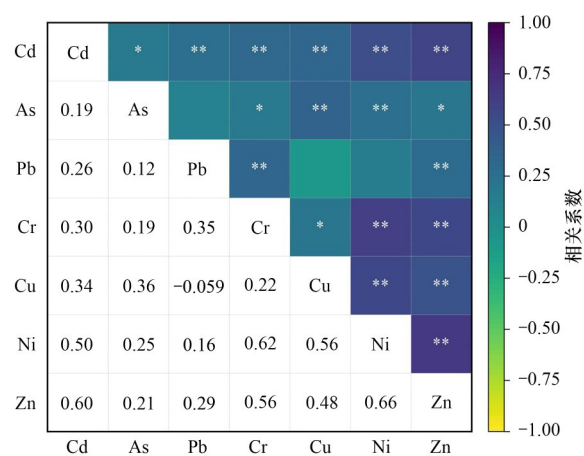
2.2 土壤重金属源解析

相关性分析能在一定程度上反映土壤重金属之间的同源性,相关系数越大,说明来自同一污染源的

可能性就越大^[49]. 研究区土壤重金属相关性结果如图2所示,除Pb分别与As、Cu和Ni不存在显著相关性外,其他元素之间均存在一定程度的显著相关性,其中Cd、Ni和Zn之间相关性系数在0.50以上,说明这3项元素存在部分同源性;Cr-Ni和Cr-Zn相关性系数在0.50以上,说明Cr分别与Ni和Zn存在部分同源性;Cu-Ni和Cu-Zn相关性系数在0.50左右,说明Cu分别与Ni和Zn也存在部分同源性.而Cd、Cr和Cu之间相关性系数在0.3左右,同源性相对较弱.综上,说明研究区污染来源相对复杂,重金属元素受不同污染源的复合影响.

利用EPA PMF 5.0对研究区土壤重金属进行源解析,选择因子数为3~5进行迭代运算20次,结合研究区可能源成分谱信息和相关性分析结果,最终确定3个因子数.经运算,模型运行结果在第36次最佳, Q_{Robust} 和 Q_{true} 值分别为1530.6和1632.2,残差分析结果显示所有样本残差均在-3~3之间,除Cr拟合度 R^2 为0.60外,其他6项重金属拟合度 R^2 在0.72~0.99之间,说明模型结果较好.

研究区土壤重金属源解析结果如图3所示,因子1中Pb和Cr贡献率最高,分别为57.9%和50.1%,远高于其他5项重金属元素.由于Cr均值



*表示在0.05水平上显著相关,**表示在0.01水平上显著相关

图2 土壤重金属含量相关性分析结果

Fig. 2 Correlation between the heavy metals in soil

含量是背景值的0.79倍,远低于背景值,Pb均值含量与背景值基本持平,且变异系数为21.9%,相对最低,说明Pb和Cr受人为活动影响较小,可能受自然因素影响较大.这与大量研究认为Cr主要受成土母质等自然源影响一致^[22,50,51],同样有研究表明Pb也可能受自然源影响^[20,52].结合研究区因子1贡献值空间分布相对均匀[图4(a)],推断因子1为自然源.

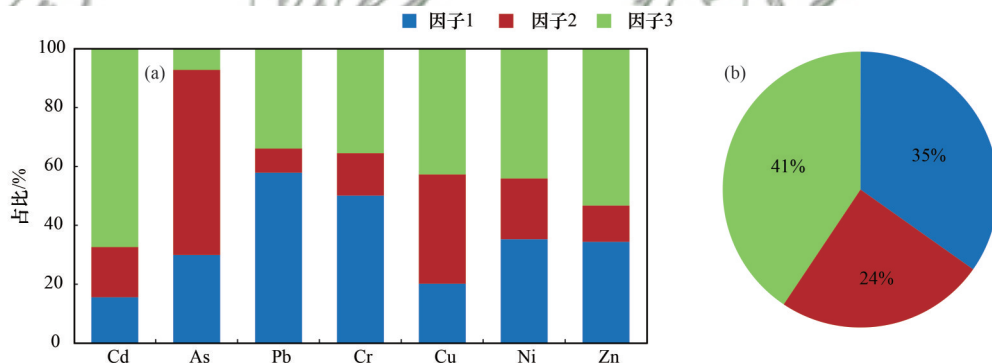


图3 PMF模型土壤重金属污染源贡献率

Fig. 3 Source contribution ratios of heavy metals of soil based on PMF receptor model

有研究表明,土壤重金属污染主要受农业和工业等影响.如长期施用有机肥、复合肥和农药等可能导致Cd、As、Cu、Ni和Zn等重金属在土壤中累积^[15,20,53].矿业开采、金属冶炼、电镀、机械制造等行业也可能导致Cd、As、Cu、Ni和Zn等重金属在土壤中累积^[56~59].结合研究区土壤重金属贡献值空间分布[图4(b)和图4(c)],因子2的贡献值总体上在城区(工业区)相对较高,呈点状,距城区(工业区)越远,贡献值越低,推断因子2为工业源.因子3贡献值总体上东南部高于其他区域,呈面状,其原因可能是研究区东南部为忠县南部,近十余年忠县在传统种植业的基础上,大力推动柑橘产业发展,主要集中在双

桂、新立、永丰和拔山等南部乡镇^[60].同时,通过对研究区标准化柑橘园调查发现,柑橘园均施用了大量有机肥对土壤进行改土培肥,可能导致土壤重金属偏高^[20,55].因此,推测因子3为农业源,受农药化肥等面源污染.

2.3 蒙特卡罗模拟的土壤重金属健康风险评估

2.3.1 非致癌健康风险评估

研究区农用地土壤非致癌健康风险评估结果如表4和图5所示.7项重金属非致癌健康风险指数(HQ)最大值均小于1,均值大小表现为:Cr > As > Pb > Ni > Cu > Cd > Zn,儿童非致癌健康风险高于成人,摄入风险均表现为:经口摄入 > 呼吸摄入 > 皮肤

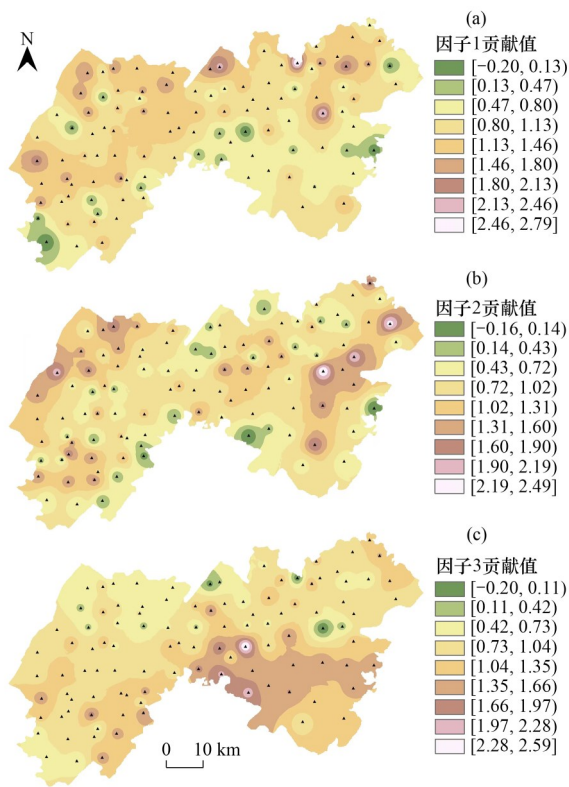


图4 PMF模型土壤重金属污染源贡献值空间分布
Fig. 4 Spatial distribution of source contribution value of heavy metals in soils based on PMF receptor model

摄入. 综合非致癌健康风险指数(HI)表明,成人和儿童HI均值分别为 $2.55\text{E-}2$ 和 $2.94\text{E-}1$,最大值均小于1,说明研究区农用地土壤重金属对儿童和成人的非致癌健康风险可忽略.

2.3.2 致癌健康风险评估

研究区农用地土壤致癌健康风险评估结果如图6所示. 5项重金属致癌健康风险指数(CR)均值大小表现为:As>Cd>Cr>Pb>Ni,其中As、Cd、Cr和Pb对儿童致癌健康风险高于成人. Cd在成人的CR最大

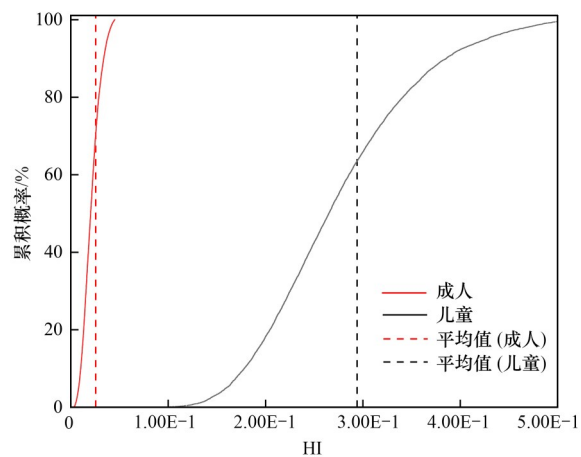


图5 土壤非致癌风险概率分布
Fig. 5 Probability distribution for non-carcinogenic risk in soil

值为 $6.24\text{E-}7$ ($\leq 1.00\text{E-}6$),说明Cd对成人的致癌风险可忽略,Cd在儿童的CR均值为 $8.25\text{E-}7$,95%分位值为 $1.36\text{E-}6$ ($> 1.00\text{E-}6$),说明Cd对儿童存在可耐受致癌风险;As在成人和儿童的CR均值分别为 $1.48\text{E-}6$ ($> 1.00\text{E-}6$)和 $4.34\text{E-}6$ ($> 1.00\text{E-}6$),最大值均小于 $1.00\text{E-}4$,说明As对成人和儿童均存在可耐受致癌风险;Pb、Ni和Cr的CR最大值均小于 $1.00\text{E-}6$,说明Pb、Ni和Cr对成人和儿童的致癌风险可忽略. 综合致癌健康风险指数(TCR)表明,成人和儿童TCR均值分别为 $2.11\text{E-}6$ 和 $5.63\text{E-}6$,最大值均小于 $1.00\text{E-}4$,说明研究区农用地土壤重金属对儿童和成人存在一定程度的可耐受致癌健康风险,且儿童致癌风险高于成人,这与儿童因生理和行为等特征,对污染物敏感性更高有关^[15]. 主要致癌因子为As和Cd,这与已有研究的结论一致,其原因在于Cd和As对人体多个系统功能造成危害相对更大,模型参数中致癌斜率因子(SF)较大有关^[15,28,29,61].

表4 土壤非致癌健康风险评估
Table 4 Non-carcinogenic health risk indices in soil

元素	非致癌风险指数(儿童)				非致癌风险指数(成人)			
	HQ _i	HQ _{i, ingest}	HQ _{i, dermal}	HQ _{i, inhal}	HQ _i	HQ _{i, ingest}	HQ _{i, dermal}	HQ _{i, inhal}
Cd	1.61E-3	1.60E-3	2.32E-7	9.80E-6	1.42E-4	1.36E-4	1.20E-7	6.34E-6
As	1.12E-1	1.12E-1	3.98E-7	1.68E-5	9.59E-3	9.58E-3	2.06E-7	1.09E-5
Pb	4.37E-2	4.37E-2	4.23E-7	2.67E-6	3.72E-3	3.72E-3	2.19E-7	1.72E-6
Cr	1.24E-1	1.23E-1	8.92E-6	7.92E-4	1.10E-2	1.05E-2	4.62E-6	5.11E-4
Cu	3.45E-3	3.45E-3	1.67E-8	2.11E-7	2.94E-4	2.94E-4	8.64E-9	1.36E-7
Ni	9.57E-3	9.57E-3	5.15E-8	5.71E-7	8.16E-4	8.16E-4	2.66E-8	3.69E-7
Zn	1.50E-3	1.50E-3	1.09E-8	9.23E-8	1.28E-4	1.28E-4	5.65E-9	5.96E-8

2.3.3 敏感性分析

敏感性分析用于反映各参数对风险结果的影响程度,敏感度值越大则对风险结果的影响越大^[28]. 研究区农用地土壤致癌和非致癌风险敏感性结果如图

7所示. 土壤颗粒摄入速率(R_{ingest})敏感度最高,其中致癌风险中儿童和成人土壤颗粒摄入速率(R_{ingest})敏感度分别为64.3%和46.1%;非致癌风险中儿童和成人土壤颗粒摄入速率(R_{ingest})敏感度分别为33.4%

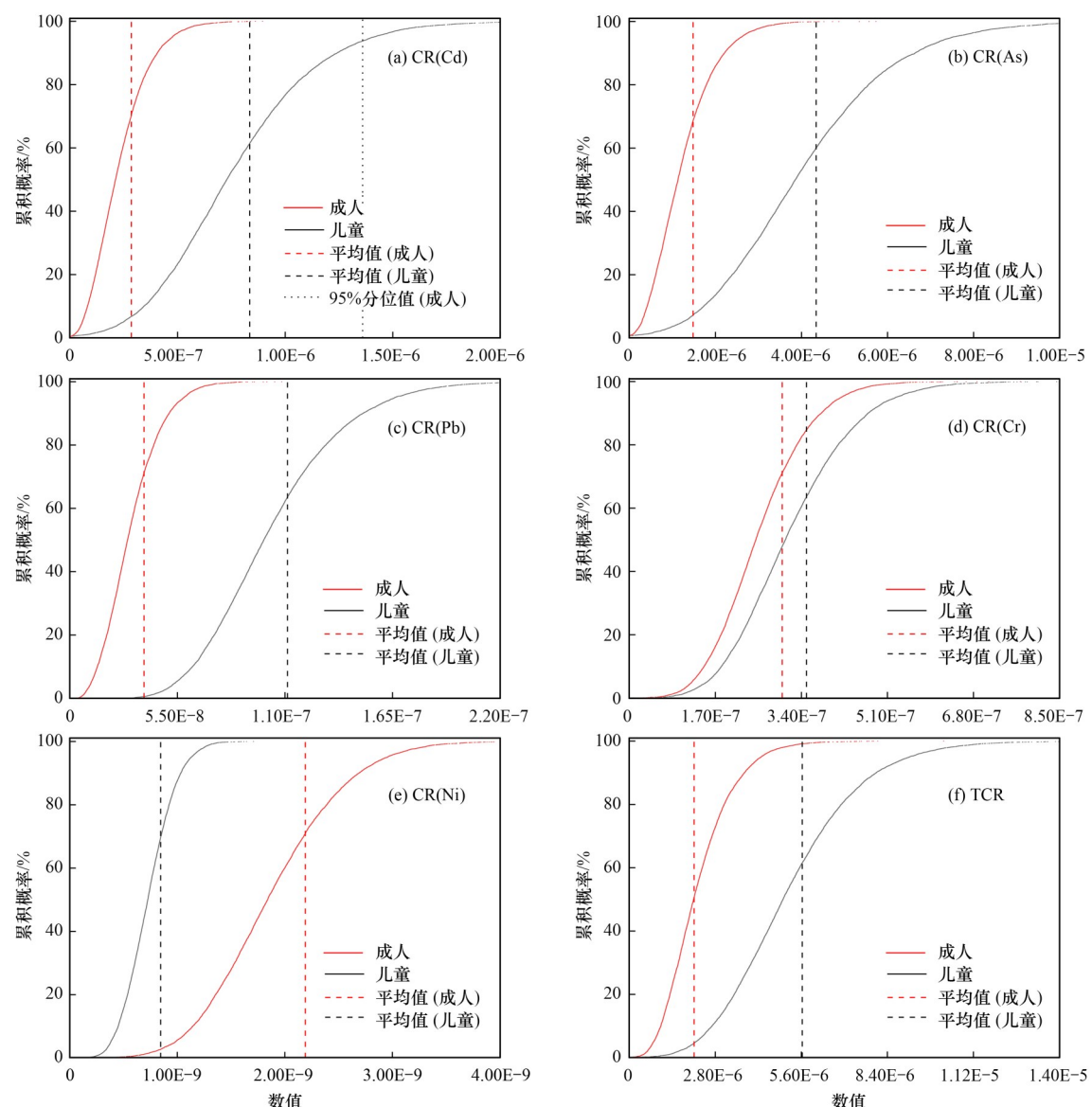


图 6 土壤致癌风险概率分布
Fig. 6 Probability distribution for carcinogenic risk in soil

和 62.5%. 这与马杰等^[22]研究的结果一致,说明减少经口摄入能较大程度降低健康风险,日常应做好防护措施. 对 7 项重金属而言,As 含量敏感性最高,应

作为优先管控元素,其次是 Cd 和 Cr. 此外,体重 (BW) 的敏感度均为负值,说明增长体重一定程度上有助于降低健康风险^[15,62].

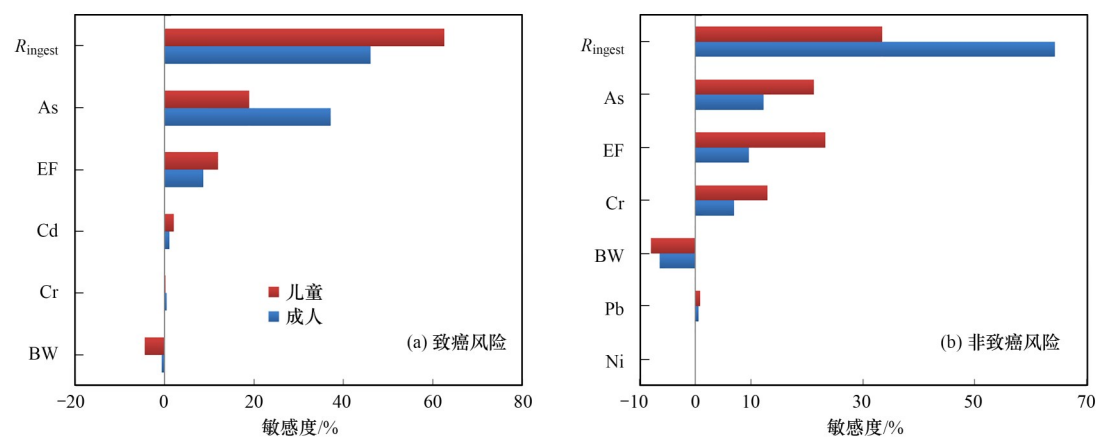
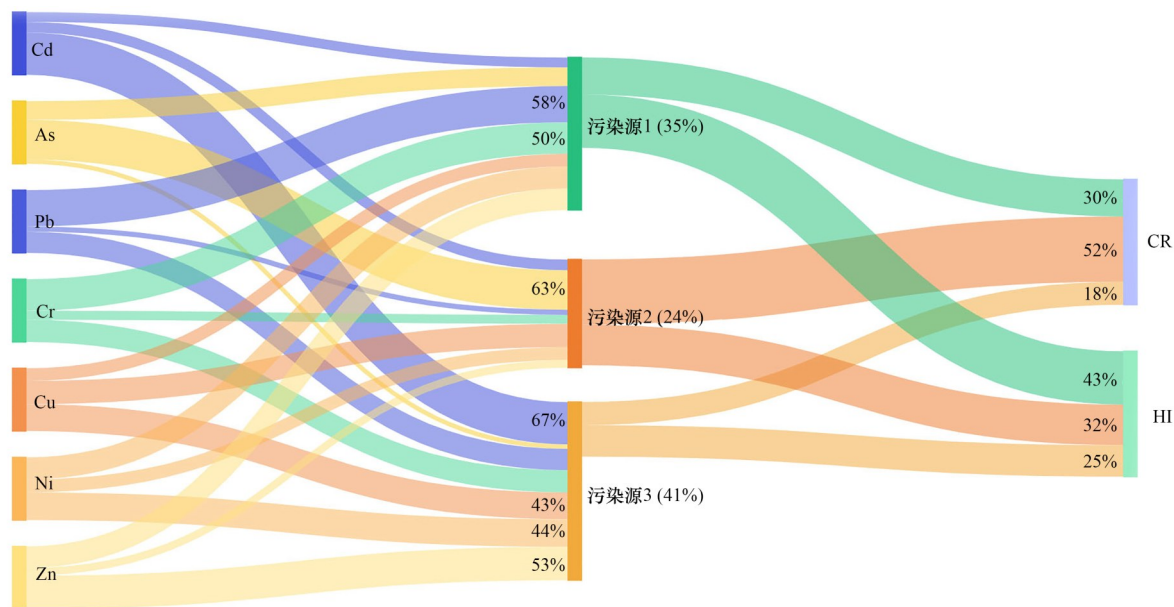


图 7 土壤致癌和非致癌风险敏感度分析
Fig. 7 Sensitivity analysis of health risks for non-carcinogenic risk and carcinogenic risk

2.4 基于源导向的土壤重金属优先控制因子分析

鉴于上述研究表明儿童健康风险高于成人,因此,以儿童HRA模型结果为例,结合PMF受体模型源解析,研究区农用地土壤重金属、污染源与健康风险关系如图8所示。人为源是导致研究区土壤重金属累积的主要因素,贡献率为65%,也是影响人体健康的主要因素,人为源引起的致癌和非致癌风险贡献率分别为70%和57%。其中农业源在3类污染源中贡献率最高(41%),污染因子以Cd、Cu、Ni和Zn为主。有研究表明,有机肥(鸡粪、鸭粪等畜禽粪便)中Cd、Cu、Zn和Ni相较其他肥料含量偏高,长期施用会增加农田土壤重金属污染的环境风险^[55,63]。如前所述,标准化柑橘园的建设施用了大量有机肥对土壤进行改土培肥,可能导致土壤Cd、Cu、Ni和Zn等重金属

偏高。农业源对致癌和非致癌风险的贡献率最低,分别为18%和25%,其原因在于相比As和Cr而言,Cd、Cu、Ni和Zn对人体的影响相对较小,模型中参考剂量(RF_i)和斜率因子(SF_i)取值较小^[15,27],因此,Cd、Cu、Ni和Zn致癌和非致癌风险较低。工业源在3类污染源中贡献率最低(24%),污染因子以As为主。有研究表明,As主要来源于工业生产(如化学工业、冶炼工业和电子工业等废水排放)和燃料燃烧^[64-66],这与前述研究区城区(工业区)周边的工业源贡献值较高一致[图4(b)],推断As主要来自城区(工业区)生产活动。工业源对致癌风险的贡献率最高(52%),其原因在于As的毒性更高,人体长期摄入As可导致肾、肝、膀胱等内脏器官的癌变^[15,67]。因此,应加强在产企业监管,避免造成对周边土壤的持续污染。



曲线的宽度表示贡献率大小,重金属曲线和污染源曲线用彩色标注

图8 土壤重金属、污染源与健康风险的关系

Fig. 8 Relationship among heavy metals, pollution sources, and health risks in soil

从土壤重金属含量特征看,虽然研究区重金属污染以Cd为主,对农业源的贡献最大,同时农业源是研究区首要污染源,然而对人体的健康风险却是最低的,因此,研究区农业源和Cd并非研究区农用地土壤中影响健康风险的首要管控要素。相反,工业源在研究区污染源中贡献率最低,但在人为源中对人体的健康风险最高,且As对工业源的贡献率最大,因此,研究区应将工业源和As作为土壤中影响健康风险的首要管控要素。综上,本研究综合探讨了土壤重金属含量特征、污染源和人体健康风险的关系,更有助于管理者或决策者制定有针对性的管控措施,降低管理成本,从而减少土壤对人体健康的风险。

3 结论

(1)结合相关分析和PMF受体模型源解析表明,研究区确定自然源、工业源和农业源为3大污染源。其中自然源占35%,污染因子以Cr和Pb为主;工业源占24%,污染因子以As为主;农业源占41%,污染因子以Cd、Cu、Ni和Zn为主。

(2)基于蒙特卡罗模拟的健康风险评估表明,研究区农用地土壤重金属对儿童和成人存在可耐受致癌健康风险,非致癌健康风险可忽略,经口摄入是主要的暴露途径。

(3)基于源导向的优先控制因子分析表明,工业源和As应作为农用地土壤中影响健康风险的首要管控要素,农业源和Cd应作为次要管控要素。

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