

张妍,赵新雷,冯雪珍,等.河南荥阳市耕地土壤重金属分布特征及来源解析[J].岩矿测试,2024,43(2):330–343. DOI: 10.15898/j.ykcs.202306300084.

ZHANG Yan, ZHAO Xinlei, FENG Xuezhen, et al. Distribution Characteristics, Ecological Risks, and Source Identification of Heavy Metals in Cultivated Land in Xingyang City[J]. Rock and Mineral Analysis, 2024, 43(2): 330–343. DOI: 10.15898/j.ykcs.202306300084.

## 河南荥阳市耕地土壤重金属分布特征及来源解析

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**摘要:**耕地质量关系着人民生活,而重金属是影响耕地质量的重要因素之一。根据全国土壤污染状况调查显示,中国耕地环境状况不容乐观,对耕地的重金属调查分析迫在眉睫。但仅简单地对重金属含量水平及来源类型进行判断已不足以作为区域土壤重金属污染治理提供支持,而通过对各类污染源贡献率的定量计算,不仅可以明确农田土壤重金属分布特征,同时可判别污染源类别及来源,从而识别优先控制的污染元素,为重金属污染精准管控提供关键信息。本文采集河南荥阳市耕地表层土壤样品(0~20cm),应用电感耦合等离子体质谱和发射光谱法(ICP-MS/OES)、原子荧光光谱法(AFS)及离子选择电极法(IES)对As、Cd、Cr、Cu、Hg、Ni、Pb、Zn等8种重金属进行测试和pH分析;利用多元统计、绝对因子分析-多元线性回归(APCS-MLR)受体模型探讨研究区8种重金属污染含量空间分布特征及来源,利用富集因子和地累积指数开展土壤污染评价。结果表明:①耕地土壤中重金属含量整体偏高。除Cr外,其他元素为郑州市土壤背景值的1.04~1.40倍,其中Cd的累积效应较明显。②研究区重金属高值区主要分布于荥阳市城区周边。③基于富集因子法、相关性分析、主成分分析及APCS-MLR源解析结果显示,研究区重金属主要有三个来源:自然源对Ni、As、Cu、Cr的贡献率分别为98%、94%、80%及63%;工业源对Cd的贡献率为78%;其他源则主要是农业化肥源、燃煤源的混合源,对Cr、Pb、Hg的贡献率分别为37%、35%及33%。④地累积指数表明,研究区各重金属以无污染为主,而Cd超标率最高,其中度、中-重度污染、重度污染样点数分别为19个、5个及3个,并存在1个极重度污染样点。综上,Cd在研究区耕地中富集较明显,为潜在的主要污染元素;工业源、自然源、农业化肥源及燃煤源是重金属的主要来源,表明人类活动已对研究区耕地产生影响,需采取措施避免该影响进一步加剧。

**关键词:**耕地土壤重金属;来源解析;绝对因子分析-多元线性回归(APCS-MLR)受体模型;风险评价;荥阳要点:

- (1)除Cr外,研究区各重金属元素在表层土壤中呈富集趋势。
- (2)研究区各重金属元素以无污染为主;Cd元素重度污染样点数为3个,极重度污染样点数1个,为主要的潜在污染物。
- (3)工业源、自然源、农业化肥及燃煤源的方差贡献率分别为44.45%、21.93%、11.34%,是研究区重金属的三个主要来源。

中图分类号: S151.93; X820.4

文献标识码: A

收稿日期: 2023-06-30; 修回日期: 2024-01-14; 接受日期: 2024-02-01

基金项目: 河南省地质研究院院管财政科研项目(2023-901-XM002-KT02)

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耕地土壤是农业生产的重要载体,其质量关系着人民的生活,而重金属是对耕地环境质量产生不利影响的主要污染物。土壤重金属具有难降解、持久性强和累积性等特点,被作物吸收后可通过食物链进入人体,从而对人体健康造成风险<sup>[1]</sup>。2014年发布的《土壤污染状况调查公报》指出,中国土壤总的超标率为16.1%,中度、重度污染点位比例为1.5%及1.1%,污染类型以无机污染为主,重金属镉、汞、砷及铅等在不同区域呈升高趋势<sup>[2]</sup>。目前,中国受到重金属污染的农业土壤占比约17%<sup>[3]</sup>,农药、农肥、农用化学品、矿物开采、工业排放及污水灌溉等是农业土壤重金属的主要来源<sup>[4-8]</sup>。

土壤重金属来源机制复杂,可分为自然来源及人为来源<sup>[9]</sup>,污染源解析是土壤重金属污染评价、防控及治理的重要前提<sup>[10]</sup>,具有落实国家“科学治污”和“精准治污”的重要现实意义。近年来对耕地土壤重金属污染的研究呈现增长趋势,许多学者在不同地区开展了大量工作。例如,李文明等<sup>[11]</sup>在青海省典型高山农业区域开展研究发现,表层土壤中As元素污染趋势较明显,自然、交通、冶炼和大气沉降为重金属的主要污染源;而陈林等<sup>[12]</sup>对宁夏引黄灌区农田进行研究发现,8种重金属(As、Cd、Cr、Cu、Hg、Ni、Pb、Zn)含量相对于宁夏土壤背景值均有一定的富集现象,且Hg和Cd存在中、较高等级生态风险点位,而人为活动为主要的影响因素。

河南省是中国主要的小麦产地,其种植面积及小麦产量均居全国首位,当地农产品及生态环境安全已受到广泛关注。在河南省开展的耕地土壤中8种重金属研究表明,部分地区重金属含量超过了农田土壤风险筛选值,且在农作物中累积明显<sup>[13-15]</sup>。但是土壤重金属污染成因来源较复杂且存在多源叠加作用,仅简单地对重金属含量水平及来源类型进行判断已不足以区域土壤重金属污染治理提供支持。因此,对耕地重金属污染源进行解析,定量计算各类污染源对元素累积的相对贡献率,明确河南省农田土壤重金属分布特征及污染状况,对农业生态环境评价、保障食品及居民安全具有重大意义。本文以河南省荥阳市为研究区,以土壤中As、Cd、Cr、Cu、Hg、Ni、Pb及Zn为研究对象,采用多元统计分析、空间分布制图等方法研究各重金属的含量及空间分布特征,运用富集因子法(EF)、地累积指数( $I_{geo}$ )对土壤污染程度进行分析,通过主成分分析(PCA)、绝对因子分析-多元线性回归(APCS-MLR)受体模型等方法对重金属来源进行解析并定量计算

各影响因素贡献率,从而确定研究区的主要污染源,以期为区域生态风险评价、土壤污染防治及人体健康管控提供科学依据,充实区域重金属评价的理论基础。

## 1 研究区概况

荥阳市位于河南省省会郑州市西部,是河南省距省会最近的县级市(图1)。荥阳市土地以农用地及建设用地为主,分别占土地总面积的62.88%、21.82%;而农用地则主要为耕地,其次为园地。荥阳市地处豫西黄土丘陵向豫东过渡地带,地形类型可分为西、南部丘陵区,北部邙山丘陵区及黄河滩地,中、东部平原区。荥阳市土壤分布主要受地貌类型影响,砂质潮土、壤质潮土分布于黄河滩地,褐性潮土多分布于研究区南部、北部丘陵区,而褐土为主要土壤类型(图1b)。荥阳工业分为汽车、煤电铝工业及阀门制造等七大主导产业,被誉为“中国阀门之乡”和“中国建筑机械之乡”(图1a)。

## 2 实验部分

### 2.1 样品采集与分析测试

本次工作在荥阳市全域开展,工作面积为365.78km<sup>2</sup>。样点布设以1:5万地形图为底图,以每平方米为一大格进行网格化布置,主要布设于农用地,采样密度为4个样/km<sup>2</sup>。各样点位置如图1所示。表层土壤样品的采样深度为0~20cm,由5个子样等份组合成1个混合样品,分析样品数量共计2113件,并保证均匀性每个样品采样总质量达到1.5~2.0kg。样品在阴凉处自然风干后,用10目尼龙筛过筛,保证总质量大于300g,测试土壤pH值。后采取缩分法取100g粉碎后过200目尼龙筛,用于重金属含量分析。

为确保各重金属元素分析质量,针对不同分析项目,其分析方法如下:①Cd、Cu、Ni、Pb采用电感耦合等离子体质谱法(ICP-MS, iCAP RQ型,美国ThermoFisher公司);②Cr、Zn、Fe采用电感耦合等离子体发射光谱法(ICP-OES, PRODIGY SPEC型,美国Leeman公司);③As、Hg采用原子荧光光谱法(AFS, AFS-3100型,美国ThermoFisher公司);④测量pH值采用离子选择电极法(IES, PXSJ-216型,美国ThermoFisher公司)。

### 2.2 实验测试数据质量控制

土壤样品分析工作由河南省地质调查院实验室完成,分析过程中严格执行《生态地球化学评价样品

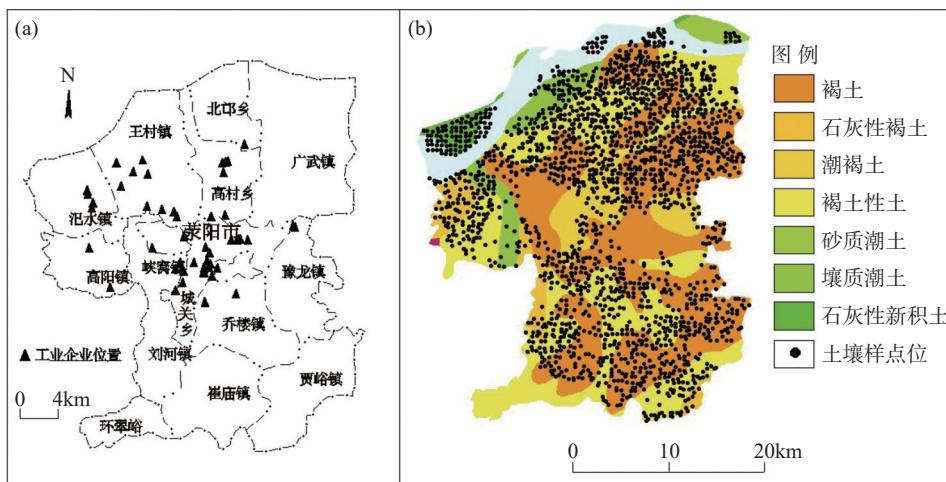


图1 研究区(a)工业企业分布和(b)土壤类型及采样点位

Fig. 1 (a) The geographic location of industrial enterprise; (b) Soil types and sampling site of the study area.

分析技术要求(试行)》(DD2005-03)、《生态地球化学评价样品分析技术要求补充规定》及《土地质量地球化学评价规范》(DZ/T 0295—2016)等相关技术标准。为保证分析测试质量,每50件样品中随机插入4个国家一级标准物质及重复样,各元素分析检出限、准确度、精密度(RSD)及报出率见表1。分析质量指标均达到或优于技术标准,分析数据质量可靠。

表1 分析方法质量监控

Table 1 Quality control of analysis method.

元素	分析方法	检出限 (mg/kg)	准确度 (△lgC)	RSD (%)	报出率 (%)
As	AFS	0.2	0.008	2.84	100
Cd	ICP-MS	0.03	0.010	4.64	100
Cr	ICP-OES	2	0.011	2.58	100
Cu	ICP-MS	0.3	0.006	5.39	100
Hg	AFS	0.0005	0.003	5.53	100
Ni	ICP-MS	0.3	0.005	5.19	100
Pb	ICP-MS	0.3	0.014	3.75	100
Zn	ICP-OES	1	0.004	4.17	100
Fe <sub>2</sub> O <sub>3</sub>	ICP-OES	0.05(%)	0.006	1.88	100
pH	IES	0.1(无量纲)	0.009	4.91	100

### 2.3 实验测试数据处理

数据处理及平均值、最大值、最小值、变异系数等参数统计工作、相关性分析、主成分分析由 SPSS Statistics16 及 Excel 软件完成, 图件制作由 Mapgis6.2、Origin2021 完成。

### 2.4 富集因子分析重金属来源

富集因子(Enrichment factor)最初是用来估算人为来源对沉积物影响的一种常用方法, 本次研究

用于分析重金属来源。第一步对重金属测量值进行标准化处理, 由于 Fe 具有相对较高的自然含量且受人为来源影响相对较小<sup>[16]</sup>, 因此在本次研究中将 Fe 作为标准化元素。计算公式如下:

$$EF = (C_i/\text{Fe})_{\text{实测值}} / (C_i/\text{Fe})_{\text{背景值}} \quad (1)$$

式中:  $(C_i/\text{Fe})_{\text{实测值}}$  为研究区 i 元素与 Fe 的比值;  $(C_i/\text{Fe})_{\text{背景值}}$  为土壤地球化学背景值中 i 元素与 Fe 的比值。本文中采用的背景值参考《河南省农耕区地球化学基准值与背景值研究》所提供的数据, As、Cd、Cr、Cu、Hg、Ni、Pb、Zn 的背景值 (mg/kg) 分别为 9.42、0.15、68.82、19.20、0.045、24.71、22.17、56.73; Fe 数据由本次研究中的分析测试项 Fe<sub>2</sub>O<sub>3</sub> 换算而来。EF 分级标准如下: 当  $EF < 0.5$  时, 表明重金属元素主要来源于地壳; 当  $0.5 \leq EF \leq 1.5$  时, 表明重金属元素主要来自成土母质或自然风化过程; 当  $EF > 1.5$  时, 表明重金属元素来自非成土母质或非自然风化过程, 如点源、非点源污染或人为影响等。

### 2.5 地累积指数评估土壤重金属污染水平

采用地累积指数 ( $I_{\text{goe}}$ ) 对土壤重金属污染水平进行评估。该方法可以同时反映自然条件下各种地质作用及人为活动对重金属积累的影响<sup>[17]</sup>。计算公式如下:

$$I_{\text{goe}} = \log_2(C_i / 1.5B_i) \quad (2)$$

式中:  $C_i$  为土壤样品重金属 i 元素的测试值;  $B_i$  为重金属 i 元素土壤背景值; 1.5 为常数。 $I_{\text{goe}}$  的分级标准:  $I_{\text{goe}} < 0$ , 无污染;  $0 \leq I_{\text{goe}} < 1$ , 轻度污染;  $1 \leq I_{\text{goe}} < 2$ , 中度污染;  $2 \leq I_{\text{goe}} < 3$ , 中-重度污染;  $3 \leq I_{\text{goe}} < 4$ , 重度污染;  $4 \leq I_{\text{goe}} < 5$ , 重-极重污染;  $5 \leq I_{\text{goe}}$ , 极重污染。

## 2.6 APCS-MLR 受体模型判断元素来源

主成分分析(Principal component analysis, PCA)是将多个变量通过线性变换从而提取或归类为一种或者多种相似变量群的方法。它通过各指标间的关联度来描述各指标或因素之间的联系,通常相关性显著的元素间可能具有同源性<sup>[18]</sup>。APCS-MLR受体模型则是通过对PCA进行相应改进而得到的一种判断元素来源的新定量分析方法,它可以计算出不同来源对同一元素的贡献率<sup>[19]</sup>。计算步骤如下<sup>[20]</sup>。

对所有元素引入1个浓度为0的人为样本,计算0浓度样本的因子分数,公式为:

$$Z_{i0} = (0 - \bar{C}_i) / \sigma_i \quad (3)$$

式(3)中: $Z_{i0}$ 为各元素的0含量因子;  $\bar{C}_i$ 为元素*i*含量的平均值;  $\sigma_i$ 为元素*i*含量的标准差。

由因子分析得到的主因子得分减去0浓度样本的主因子分数可得到每个样本的APCS;后将APCS作为自变量,重金属元素含量为因变量,进行多元线性回归分析,得到的回归系数可将APCS转化为主因子对应的污染源对每个样本的浓度贡献,公式为:

$$C_i = b_{i0} + \sum_{p=1}^n (b_{pi} \times APCS_p) \quad (4)$$

式(4)中: $C_i$ 为元素*i*的实测值;  $b_{i0}$ 为回归方程常数项;  $b_{pi}$ 表示源*p*对元素*i*的回归系数;  $APCS_p$ 为因子*p*的绝对主因子分数。 $b_{pi} \times APCS_p$ 为因子*p*对于 $C_i$ 的含量贡献,所有样本的 $b_{pi} \times APCS_p$ 的平均值即为因子*p*对应的污染源平均绝对贡献量。

表2 研究区表层土壤重金属含量统计

Table 2 Heavy metal concentrations in surface soil of the study area.

统计项目	pH	As (mg/kg)	Cd (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	Hg (mg/kg)	Ni (mg/kg)	Pb (mg/kg)	Zn (mg/kg)
算术平均值	8.22	10.61	0.21	61.21	20.74	0.047	26.15	23.70	65.70
几何平均值	8.22	10.52	0.20	60.94	20.54	0.041	26.00	23.32	64.29
中位数	8.21	10.40	0.20	60.80	20.20	0.041	25.60	23.10	62.90
众数	8.25	10.20	0.19	59.60	19.80	0.034	24.70	23.40	57.40
算术标准差	0.22	1.45	0.19	5.86	3.23	33.30	2.99	8.05	20.34
几何标准差	1.03	1.14	1.36	1.10	1.14	1.62	1.11	1.16	1.20
最大值	9.17	21.00	7.29	120.00	70.10	0.87	47.30	344.50	645.00
最小值	6.85	4.80	0.08	39.50	11.60	0.0089	14.80	16.40	37.70
变异系数	0.03	0.14	0.89	0.10	0.16	0.71	0.11	0.34	0.31
偏度	-0.0056	1.28	27.08	1.08	3.74	9.71	1.71	30.88	14.47
峰度	0.72	4.68	955.90	7.36	34.81	193.50	5.73	1202.63	345.98
郑州市背景值	8.11	9.42	0.15	68.82	19.20	0.045	24.71	22.17	56.73
风险筛选值	6.5~7.5	30	0.3	200	100	2.4	100	120	250
	pH>7.5	25	0.6	250	100	3.4	190	170	300

## 3 结果

### 3.1 耕地土壤中重金属含量总体分布特征

荥阳市耕地土壤重金属含量统计特征见表2,其中郑州市土壤背景值取自河南省地质调查院2020年度项目《河南省农耕区地球化学基准值与背景值研究》,土壤环境质量标准采用《土壤环境质量农用地土壤污染风险管理标准(试行)》(GB15618—2018)中其他土地利用类型在 $6.5 < pH \leq 7.5$ 及 $pH > 7.5$ 情况下的农用地土壤污染风险筛选值。研究区表层土壤pH为 $6.85 \sim 9.17$ ,算术平均值(均值,下同)为8.22,呈弱碱性。8种重金属元素含量的平均值分别为As(10.61mg/kg)、Cd(0.21mg/kg)、Cr(61.21mg/kg)、Cu(20.74mg/kg)、Hg(0.047mg/kg)、Ni(26.15mg/kg)、Pb(23.70mg/kg)及Zn(65.70mg/kg)。与郑州市土壤背景值相比,研究区重金属含量整体偏高,比值范围为1.04~1.40,而Cr含量较低,仅为郑州市背景值的0.89倍。结果表明,荥阳市耕地部分重金属存在一定程度的积累现象。As、Hg、Ni元素原始数据呈对数正态分布,其他元素则呈对数偏态分布,且Cd、Pb、Zn的峰度及偏度值较高,表明其存在部分尖峰值数据。

变异系数(CV)是一种反映重金属空间变异程度的方法,其值越大,表明重金属分布越不均匀<sup>[21]</sup>。研究区8种重金属的变异系数由小到大顺序为:Cr<Ni<As<Cu<Zn<Pb<Hg<Cd。Cd、Hg元素呈高度变异水平( $CV \geq 0.36$ ),变异系数分别为0.89、0.71;Cu、Pb、Zn呈中度变异水平( $0.16 \leq CV < 0.36$ );而As、Cr、Ni则属于低变异水平( $CV < 0.16$ ),特别

是 Cr, 其变异系数仅为 0.10, 表明其空间分布较均匀, 受人为影响较小。

与农用地土壤污染风险管控标准相比, 研究区 8 项重金属含量均值都低于风险筛选值, 但 Cd、Pb、Zn 仍存在部分样点值高于风险筛选值, 其大于筛选值样点数排列顺序为: Cd(13个) > Zn(3个) > Pb(1个), 并且有1件样品 Cd 含量超过管制值, 表明研究区耕地土壤 Cd 存在一定的污染风险。

### 3.2 土壤重金属空间分布特征

基于 Mapgis 软件绘制的研究区土壤重金属空间分布图(图 2)显示, 受不同地质环境及人为因素影响, 8 种土壤重金属含量在空间分布上存在显著差异性。Cd(图 2b)、Hg(图 2e)、Pb(图 2j)、Zn(图 2h) 等 4 种元素分布较相似, 在研究区中部即荥阳市周边存

在一高值区, 其与荥阳市部分企业分布相重合, 因此工业生产活动可能导致 Cd、Hg 累积, 而较频繁的交通运输可导致 Pb、Zn 含量升高; Ni 元素(图 2f)的高值区仅分布于研究区北部, 该元素主要受到成土母质影响, 故受人类扰动较少, 而 Cr 元素(图 2c)分布与其相似; As 元素(图 2a)、Cu 元素(图 2d)高值区零星分布于研究区北部及荥阳市周边。因此, 人口密度增加、工业生产活动及能源消耗是导致研究区表层土壤重金属富集的原因之一。

## 4 讨论

### 4.1 土壤重金属污染评价

#### 4.1.1 富集因子计算

研究区表层土壤不同重金属元素富集因子

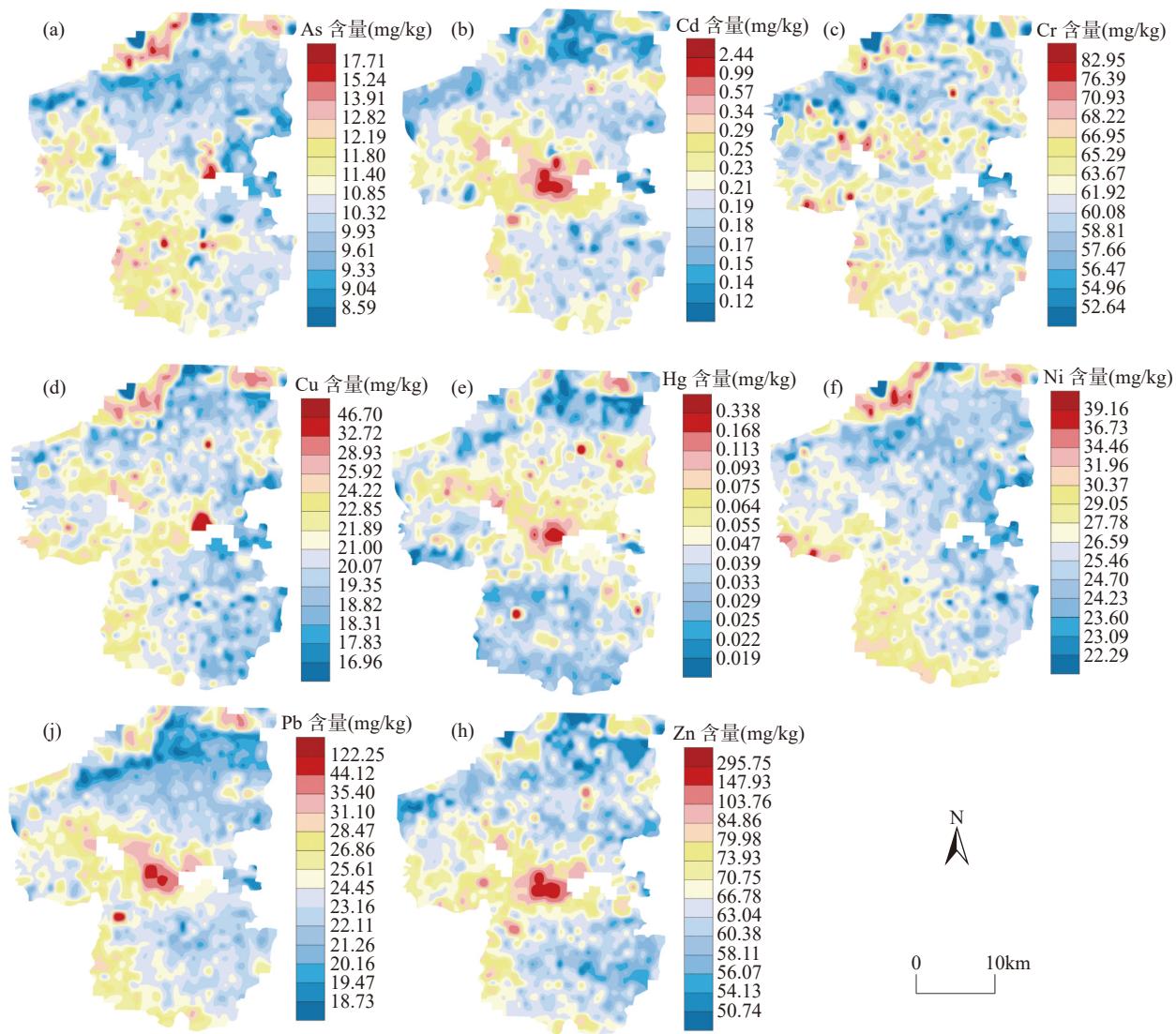


图2 研究区土壤重金属空间分布

Fig. 2 Spatial distributions of heavy metals in soil of the study area.

(EF) 趋势见图3。由图可知,As、Cd、Cr、Cu、Hg、Ni、Pb、Zn的EF值范围分别为0.86~2.80、0.68~6.72、0.81~2.01、1.08~3.53、0.27~7.98、1.15~3.10、1.04~2.47、0.96~6.62。其中,Cr、Cu、Ni、Pb等4种元素富集因子主要分布在0.5~1.5之间,样点数分别为2027、1415、1681及1522个,但仍存在部分点位的富集因子大于1.5,表明此类点位受到外源因素的影响。Hg富集因子变化范围较大,且存在小于0.5的点位,样点数为67个,表明其除来源于成土母质外仍存在点源污染;As、Zn两元素富集因子箱图上边缘均超过1.5,表明其受到人为来源不同程度地影响;而Cd元素箱图下边缘均超过1.5,其25%分位值为1.59,表明人为来源对其扰动强烈。

#### 4.1.2 地累积指数评价

地累积指数结果(表3)表明研究区土壤As、Cd、Cr、Cu、Hg、Ni、Pb、Zn的 $I_{geo}$ 值范围分别为-1.56~0.57、-1.45~5.02、-1.39~0.21、-1.31~1.28、-2.92~3.69、-1.32~0.35、-1.02~3.37、-1.17~2.92。其中As、Cr、Ni三元素各样点处于无污染和轻度污染,As元素无污染样点数为2059个(97.44%),轻度污染样点数54个(2.56%);Cr元素仅存在1个轻度污染样点;Ni元素无污染样点数2090个(98.91%),轻度污染样点数23个(1.09%);Cu元素的无污染、轻度污染、中度污染样点数分别为2057个(97.35%)、55个(2.60%)及1个;Pb、Zn、Hg元素样点分布与Cu相似,只Pb、Hg各存在1个重度污染样点;Cd超标率最高,其无污染样点数为1556个,均低于其他元素;而中度、中-重度污染、重度污染样点数分别为19个(0.90%)、5个(0.24%)及3个(0.14%),并存在1个极重度污染样点。

表3 研究区表层土壤重金属地累积指数

Table 3 Geo-accumulation index of heavy metals in soil of the study area.

重金属元素	地累积指数( $I_{geo}$ )范围	污染程度的样点数(个)						
		无污染	轻度污染	中度污染	中-重度污染	重度污染	重-极重污染	极重污染
As	-1.56~0.57	2059	54	0	0	0	0	0
Cd	-1.45~5.02	1556	529	19	5	3	0	1
Cr	-1.39~0.21	2112	1	0	0	0	0	0
Cu	-1.31~1.28	2057	55	1	0	0	0	0
Hg	-2.92~3.69	1841	248	20	3	1	0	0
Ni	-1.32~0.35	2090	23	0	0	0	0	0
Pb	-1.02~3.37	2079	32	1	0	1	0	0
Zn	-1.17~2.92	2000	105	7	1	0	0	0

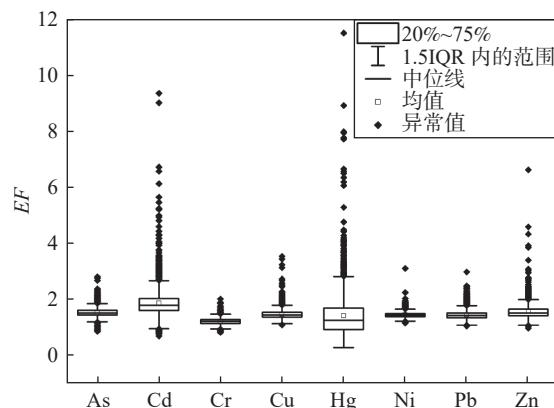


图3 研究区土壤重金属富集因子分布箱式图

Fig. 3 Box diagram of enrichment factor distribution for heavy metals in soil of the study area.

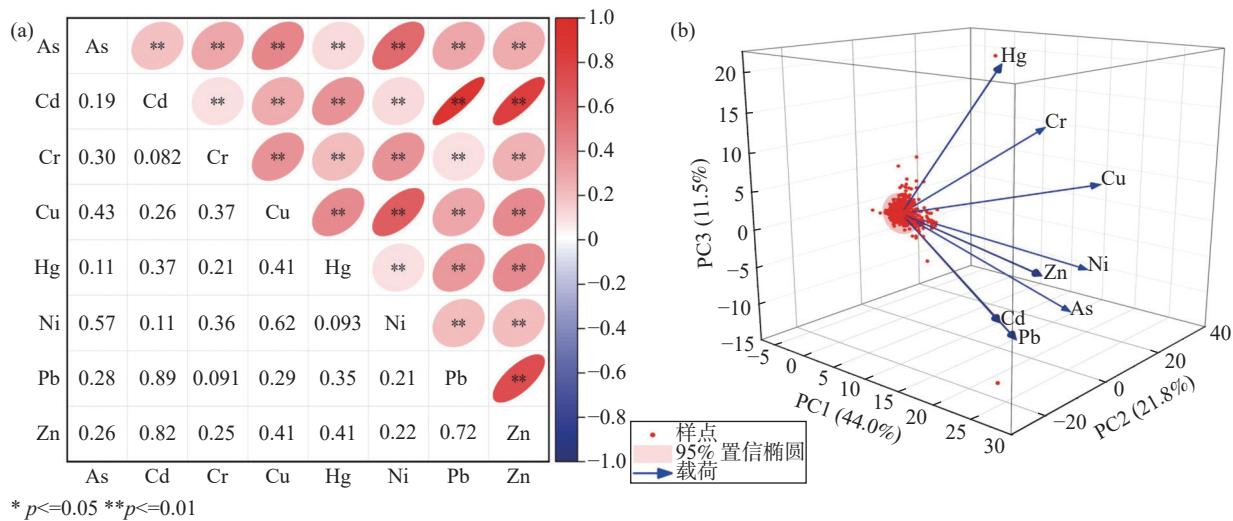
## 4.2 土壤重金属来源解析

### 4.2.1 重金属间相关性分析

通过Pearson相关分析(图4a)可知,As-Ni、As-Cu、Cd-Pb、Cd-Zn、Cu-Ni、Cu-Zn及Pb-Zn之间存在极显著正相关( $P<0.01$ ),其中Cd-Pb、Cd-Zn相关系数更高达0.89、0.82,表明元素间地球化学行为相似,赋存状态相同;在PCA成分图解中(图4b),载荷变量线越接近则具有越强的相关性,因此研究区土壤8种重金属可分为Cd-Pb-Zn、As-Ni-Cu-Cr及Hg三类组合。组合内当元素间相关系数值越大时表明其两者间的关系越强,其可能具有相似的污染源<sup>[22]</sup>;而这三类组合相互间相关系数较低,不存在显著相关关系,表明这三类元素组合之间污染来源存在较大差异,可能具有多种来源。

### 4.2.2 主成分分析

对研究区8种重金属进行主成分分析后可知,其KMO值(Kaiser-Meyer-Olkin检验统计量)为



\*  $p \leq 0.05$  \*\*  $p \leq 0.01$

图4 研究区土壤重金属富集相关性系数矩阵和PCA成分

Fig. 4 Correlation plot and PCA plot of heavy metals in soil of the study area.

0.727, 大于 Kaiser<sup>[23]</sup>给出的 KMO 标准值 0.7; Bartlett 球形检验 Sig.(概率  $P$  值)为  $0.000 <$  显著性水平  $\alpha(0.05)$ , 分析结果能够反映元素间的联系<sup>[24]</sup>, 可在研究区开展主成分分析。由表 4 可知, 对 Kaiser 标准化后因子进行 Varimax 正交旋转, 特征值大于 0.9 的主成分有三个: 3.556、1.755、0.907; 其方差贡献率为 44.45%、21.93%、11.34%, 累积贡献率为 77.72%, 可以解释重金属的大部分信息。

表 4 研究区表层土壤重金属主成分分析矩阵

Table 4 Principal component analysis matrix of heavy metals in surface soil of the study area.

重金属元素及指标	变量在各主成分上的因子载荷		
	第一主成分	第二主成分	第三主成分
As	0.565	0.496	-0.383
Cd	0.775	-0.563	-0.157
Cr	0.451	0.479	0.325
Cu	0.701	0.431	0.194
Hg	0.553	-0.148	0.696
Ni	0.565	0.653	-0.228
Pb	0.790	-0.455	-0.237
Zn	0.832	-0.349	-0.017
初始特征值	3.556	1.755	0.907
方差贡献率(%)	44.45	21.93	11.34
累计方差贡献率(%)	44.45	66.38	77.72

第一主成分 (F1) 方差贡献率为 44.45%, 载荷较高的重金属元素是 Zn(0.832)、Pb(0.790)、Cd(0.775)、Cu(0.701), 该组元素均属于中度及高度变异, 表明部

分受人类活动影响。由相关性分析及元素空间分布图可知, 此四元素间具有空间相关性; Cd 是一种汽车轮胎的重要添加剂<sup>[25]</sup>, 而车辆润滑油氧化后生成的有机化合物能够腐蚀含 Cd、Zn 等元素的金属部件及油泵, 导致其向环境中释放重金属元素<sup>[26]</sup>。汽车产业是荥阳市的支柱产业之一, 因此第一主成分主要受工业生产活动影响。

第二主成分 (F2) 的方差贡献率为 21.93%, 载荷较高的重金属元素是 Ni(0.653)、As(0.496)、Cr(0.479)。田江涛等<sup>[27]</sup>研究表明, Ni、Cr 主要富集于超基性、基性火山岩中, 且两元素变异系数分别为 0.11、0.10, 属低变异水平, 表明受人类活动影响较小, 因此第二主成分主要受母岩风化影响。

第三主成分 (F3) 的方差贡献率为 11.34%, Hg 是主要的组成元素, 其载荷为 0.696。由相关性分析和载荷距离可知, Hg 元素的来源有别于其他元素, 且其高度变异, 高值点多呈点状分布, 由于 Hg 主要通过大气干湿沉降在土壤中富集<sup>[28]</sup>, 因此该成分可归为“远源大气传输”。

#### 4.2.3 APCS-MLR 受体模型

研究区 8 种重金属 APCS-MLR 受体模型如表 5 所示, 相关系数( $R^2$ )是用于衡量模型与实际观测值的相关性, 当其数值越接近 1 时, 其线性拟合度越高, 模拟结果也越好<sup>[29]</sup>。As、Cd、Cr、Cu、Hg、Ni、Pb、Zn 的相关系数( $R^2$ )分别为 0.553、0.917、0.406、0.673、0.327、0.739、0.831 和 0.813, 除 As、Cr、Hg 三元素外, 其他元素拟合效果较好。根据 APCS-MLR 受体模型, 分别计算出主成分 F1、F2 及其他类

对各元素的贡献率(图5),F1中占比较高的元素Cd,其贡献率为78%。研究区Cd具有强的变异性,空间分布差异性显著,表明其受人类活动干扰较为严重。有研究表明,Cd的外部来源主要为工业源与化石燃料燃烧<sup>[30]</sup>、汽车尾气与交通粉尘<sup>[31]</sup>和农药化肥<sup>[32-33]</sup>等。研究区Cd高值区与工业企业分布重叠,故F1可识别为工业源。

表5 绝对因子分析-多元线性回归(APCS-MLR)受体模型

Table 5 Absolute principal component score-multiple linear regression (APCS-MLR) receptor model.

重金属元素	受体模型	$R^2$
As	$C(As)=-0.498+0.219APCS_{F1}+1.058APCS_{F2}$	0.553
Cd	$C(Cd)=0.016+0.181APCS_{F1}+0.003APCS_{F2}$	0.917
Cr	$C(Cr)=22.498+0.404APCS_{F1}+3.719APCS_{F2}$	0.406
Cu	$C(Cu)=-5.596+0.967APCS_{F1}+2.47APCS_{F2}$	0.673
Hg	$C(Hg)=-43.78+17.641APCS_{F1}+7.262APCS_{F2}$	0.327
Ni	$C(Ni)=-0.452+0.159APCS_{F1}+2.566APCS_{F2}$	0.739
Pb	$C(Pb)=8.283+7.291APCS_{F1}+0.856APCS_{F2}$	0.831
Zn	$C(Zn)=4.034+17.793APCS_{F1}+4.422APCS_{F2}$	0.813

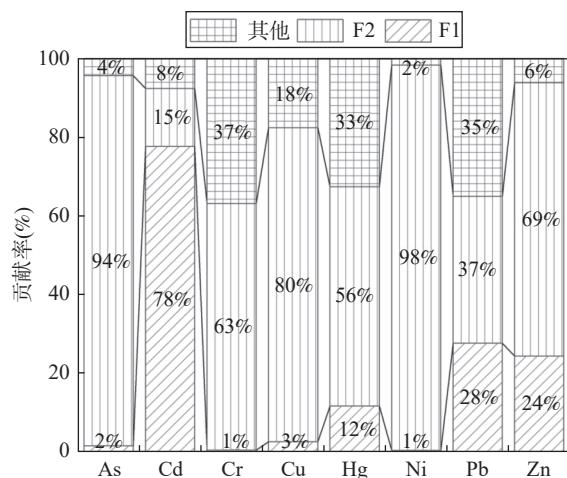


图5 研究区土壤重金属污染贡献率

Fig. 5 Source contribution ratios of heavy metals in soil of the study area.

此外,Cr、Pb、Hg也受到其他源的控制,其贡献率分别为37%、35%、33%。Zhao等<sup>[34]</sup>研究发现,化肥和粪肥的使用可导致重金属(Cd、Cu、Pb、Zn)含量的年增长率约为3%,研究区土壤多为农业用地,大量使用的有机肥中Pb含量偏高可能造成土壤中

Pb升高<sup>[35]</sup>。Hg的变异系数为0.71,属强变异元素,表明其受人类扰动较大。研究认为,大气Hg的干湿沉降是土壤Hg含量超标的主要原因之一<sup>[36]</sup>,研究区冬季取暖做饭等主要燃料来源为燃煤,Hg含量空间分布高值点分散且多分布于城镇周边;因此其他源可认定为农业化肥源及燃煤源的混合源。

主成分F2中Ni、As、Cu、Cr四元素的贡献率分别为98%、94%、80%、63%。Cr、Cu、Ni受地球化学成因影响较大,主要为地质自然来源<sup>[37-38]</sup>。研究区As、Cr和Ni空间变异性较小,其中Cr平均值低于郑州市土壤元素背景值,且Cr、Cu、Ni元素富集系数主要分布在0.5~1.5之间,表明它们在表层土壤中基本保持了原始背景状态,主要受土壤地球化学作用和成土母质控制,受人类活动影响较小或基本未受影响<sup>[39]</sup>。因此,可认为F2表示自然源。

## 5 结论

通过多元统计法分析荥阳市耕地区表层土壤重金属含量分布特征,运用富集因子、地累积指数法对其污染水平进行评估,并最终结合相关性系数、主成分分析及APCS-MLR受体模型确定重金属污染来源。结果表明:研究区耕地表层土壤重金属除Cr外其他7种重金属含量均高于郑州市土壤背景值,重金属在耕地中存在一定的积累现象;空间分布表明Cd、Cu、Hg、Pb、Zn等元素高值区主要分布于荥阳市城区周边,受人口密度增加、工业生产活动及能源消耗影响;富集因子计算结果表明,Cr、Cu、Ni、Pb元素主要来自成土母质或自然风化过程;Hg则呈两极分化,67个点位来源于成土母质,其他存在点源污染点位;As、Zn、Cd受人为扰动强烈;而地累积指数显示研究区重金属整体以无污染样点为主,Cd污染程度最高且存在极重污染样点,后续应对其持续关注。通过相关分析、主成分分析的定性分析及APCS-MLR受体模型定量计算表明研究区重金属来源主要为工业源、自然源、农业化肥源及燃煤源。

研究区部分重金属元素存在污染风险且受人类活动影响较大,本次工作对重金属的其他来源及成土母质影响的研究较少,下一步工作中应对其进行持续关注以防止污染程度的进一步加剧。同时,需加强人类活动对元素地球化学特征的影响研究,以防止生态环境恶化,保障生态环境安全。

# Distribution Characteristics, Ecological Risks, and Source Identification of Heavy Metals in Cultivated Land in Xingyang City

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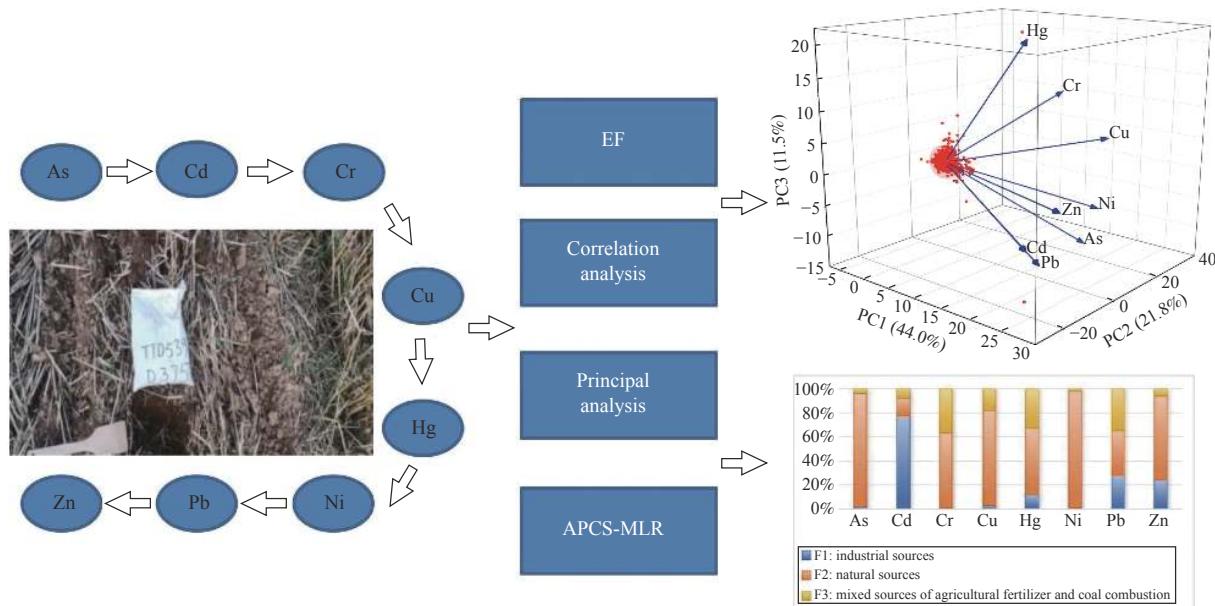
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## HIGHLIGHTS

- (1) All heavy metal elements except Cr exhibit an enrichment trend in surface soil.
- (2) The predominant status for each heavy metal element in the study area is uncontaminated. However, the Cd element has 3 points with heavy contamination and 1 point with extremely heavy contamination, making it a primary potential pollutant.
- (3) The variance contribution rates of industrial sources, natural sources, as well as agricultural fertilizer and coal burning sources are 44.45%, 21.93% and 11.34%, respectively, which are the three main sources of heavy metals in the study area.



**ABSTRACT:** The quality of arable land is closely related to people's livelihoods, and heavy metals are one of the significant factors affecting arable land quality. The spatial distribution characteristics and sources of eight heavy metal pollutants in the cultivated land of Xingyang City were investigated by multivariate statistical analysis and absolute principal component score-multiple linear regression (APCS-MLR) receptor model, and soil pollution assessment was carried out by enrichment factor and land accumulation index. The results show that the heavy metal content in cultivated soil was higher as a whole, and the accumulation effect of Cd was more obvious. The heavy metals in the study area were mainly distributed around Xingyang City. Industrial, natural, and the mixed sources of agricultural fertilizer and coal-burning are the main sources of heavy metals. The accumulative index shows that the heavy metals in the study area are mainly unpolluted, and the Cd exceeding standard rate is the highest. Therefore, it indicates that human activities have affected the cultivated land in the study area, and measures should be taken to avoid further aggravation. The BRIEF REPORT is available for this paper at <http://www.ykcs.ac.cn/en/article/doi/10.15898/j.ykcs.202306300084>.

**KEY WORDS:** heavy metals in cultivated soil; source analysis; absolute principal component score-multiple linear regression (APCS-MLR) receptor model; risk assessment; Xingyang

## BRIEF REPORT

**Significance:** Arable soil serves as a crucial medium for agricultural production, with its quality directly impacting people's livelihoods. Heavy metals represent the primary pollutants that adversely affect the environmental quality of arable land. Characteristics such as non-degradability, strong persistence, and accumulation make soil heavy metals a significant threat, as they can be absorbed by crops and subsequently enter the human body through the food chain, posing risks to human health<sup>[1]</sup>. The sources of soil heavy metals are complex, encompassing both natural and anthropogenic origins<sup>[9]</sup>. Analyzing the origins of pollution sources is a crucial prerequisite for the assessment, prevention, and control of soil heavy metal pollution<sup>[10]</sup>. This has practical significance in implementing the national strategies of "scientific pollution control" and "precision pollution control". Henan Province is the main wheat producing area in China, with both its planting area and wheat yield ranking at the top nationally, so local agricultural products and eco-environmental security have been widely concerned. In the research, the distribution characteristics and pollution status of heavy metals in farmland soil in Henan province was clarified, and the relative contribution rate of various pollution sources to the accumulation of elements was calculated. It is of great significance to the assessment of agricultural ecological environment and the safety of food and residents.

**Methods:** The current study was conducted throughout the entire Xingyang City, with a working area covering 365.78km<sup>2</sup>. Surface soil samples were collected at a depth of 0-20cm, and a total of 2113 samples were analyzed. Surface soil samples (0-20cm) were collected, and eight heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, Zn) and pH were analyzed using inductively coupled plasma-mass spectrometry (ICP-MS), inductively coupled plasma-optical emission spectrometry (ICP-OES), atomic fluorescence spectrometry (AFS), and ion-selective electrode method (IES). Multiple statistical analyses, spatial distribution mapping, and methods such as enrichment factor (EF) and geo-accumulation index ( $I_{geo}$ ) were employed to analyze the degree of soil pollution. We also utilized principal component analysis (PCA), an absolute principal component score-multiple linear regression (APCS-MLR) receptor models, and other methods, to quantify the contribution rates of various influencing factors, thus identifying the main pollution sources in the research area.

**Data and Results:** (1) Overall distribution characteristics. The average concentrations of eight heavy metal elements (Table 2) were as follows: As (10.61mg/kg), Cd (0.21mg/kg), Cr (61.21mg/kg), Cu (20.74mg/kg), Hg (0.04mg/kg), N (26.15mg/kg), Pb (23.70mg/kg), and Zn (65.70mg/kg). In comparison with the soil background values of Zhengzhou City, the overall heavy metal concentrations in the study area were relatively high, with ratio

ranges from 1.04 to 1.40. Notably, the concentration of Cr was lower, at only 0.89 times the background value of Zhengzhou City. These results indicate a degree of accumulation of some heavy metals in the arable land of Xingyang City.

Coefficient of variation ( $CV$ ) reflects the degree of spatial variability of heavy metals. The larger the  $CV$  value, the more uneven the distribution of heavy metals<sup>[21]</sup>. The order of  $CV$  of 8 heavy metal elements in the study area is: Cr<Ni<As<Cu<Zn<Pb<Hg<Cd. Cd and Hg were highly variable ( $CV \geq 0.36$ ), with  $CV$  of 0.89 and 0.71, respectively. Cu, Pb and Zn were moderately variable ( $0.16 \leq CV < 0.36$ ), while As, Cr and Ni were low variable ( $CV < 0.16$ ), the  $CV$  of Cr was only 0.096, which indicates that the spatial distribution of Cr is uniform and less affected by humans. Compared with the soil pollution risk control standard of agricultural land, the average content of 8 heavy metal elements in the study area was lower than the risk screening value, but the values of Cd, Pb and Zn were still higher than the risk screening value. The order of the number of samples exceeding the risk screening value was as follows: Cd (13 samples)>Zn (3 samples)>Pb (1 sample), showing that there was a certain risk of Cd pollution in cultivated soil in the study area.

**(2) Spatial distribution characteristics.** Among the eight soil heavy metals, Cd, Hg, Pb, and Zn exhibited a similar distribution pattern, forming a high-value zone in the central part of the study area, particularly around the periphery of Xingyang City. The high-value zone for Ni was exclusively in the northern part of the research area. Cr and Ni were primarily influenced by parent material, thus showing less disturbance by human activities. Sporadic high-value zones for As and Cu were scattered in the northern part of the research area and around Xingyang City.

**(3) Pollution assessment.** The Enrichment Factors (EF) were ranked from highest to lowest as follows (Fig.3):  $EF_{Cd} (1.86) > EF_{Zn} (1.57) > EF_{As} (1.53) > EF_{Cu} (1.46) > EF_{Ni} (1.44) = EF_{Pb} (1.44) > EF_{Hg} (1.40) > EF_{Cr} (1.21)$ . This indicates that Cu, Ni, Pb, Hg, and Cr were primarily influenced by natural soil process. Cd, As, and Zn showed enrichment, especially with Cd being significantly impacted by anthropogenic disturbances.

The Geo-accumulation Index reveals that the number of non-contaminated sample points for As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were 2059, 1556, 2112, 2057, 1841, 2090, 2079, and 2000, respectively (Table 3). The majority of heavy metal samples in the study area were non-contaminated, with Cd exhibiting the highest pollution level and the presence of extremely contaminated sample points, making it a primary potential pollutant in the research area.

**(4) Source analysis.** Through Pearson correlation analysis, it is evident that there were highly significant positive correlations ( $P < 0.01$ ) between As-Ni, As-Cu, Cd-Pb, Cd-Zn, Cu-Ni, Cu-Zn, and Pb-Zn, with Cd-Pb and Cd-Zn reaching as high as 0.89 and 0.82, respectively (Fig.4). After Kaiser normalization and Varimax orthogonal rotation of the factors, three principal components with eigenvalue greater than 0.9 were identified, measuring 3.556, 1.755, and 0.907, respectively. The variance contribution rates were 44.45%, 21.93%, and 11.34%, resulting in a cumulative contribution rate of 77.72%. The results of both correlation and principal component analyses indicate that heavy metals in the study area can be categorized into three groups: F1 (Zn, Pb, Cd, Cu); F2 (Ni, As, Cr); F3 (Hg). F1 is mainly affected by industrial production activities; F2 is mainly affected by weathering of parent rock and F3 can be classified as “remote atmospheric transport”.

The results of the APCS-MLR receptor model indicate that Cd constitutes a relatively high proportion, with a contribution rate of 78% (Fig.5). Previous studies suggested that external sources of Cd primarily include industrial emissions and fossil fuel combustion<sup>[29]</sup>, automobile exhaust and traffic dust<sup>[30]</sup>, as well as pesticides and fertilizers<sup>[31-32]</sup>. The high-value areas of Cd in the study area overlap with the distribution of industrial enterprises, thus identifying F1 as an industrial source. The deposition of atmospheric Hg, through both dry and wet processes, is considered one of the major contributors to excessive soil Hg content<sup>[35]</sup>. In the study area, winter heating and cooking predominantly rely on coal combustion, with high-value points of Hg spatially scattered, especially around urban areas. Therefore, other sources can be identified as a mixed source of agricultural fertilizers and coal combustion.

Cr, Cu, and Ni are significantly influenced by geochemical factors, mainly originating from geological and natural sources<sup>[36-37]</sup>. Thus, F2 is considered to represent natural sources. Cr is greatly influenced by geochemical genesis, mainly from geological natural sources<sup>[38]</sup>. The spatial variability of Cr elements in the study area is small and the enrichment coefficient is mainly distributed between 0.5 and 1.5, indicating that they basically maintain the original background state in the surface soil, mainly controlled by the biogeochemistry of soil environment and soil-forming parent materials, and are little or basically unaffected by human activities<sup>[39]</sup>.

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