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半干旱亚湿润干旱沙区樟子松根内真菌群落结构和功能时空动态特征



宁夏引黄灌区农田土壤重金属生态风险评价及来源解析

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摘要:农田土壤环境直接关系到农产品质量安全,为了解宁夏引黄灌区农田土壤重金属特征及主要影响因子,在2017~2021年连续5年采样分析了重金属分布特征和相关关系,评价了重金属污染状况和潜在生态风险,解析了农田重金属主要来源.结果表明,宁夏引黄灌区土壤 $\omega(Pb)$ 、 $\omega(As)$ 、 $\omega(Zn)$ 、 $\omega(Ni)$ 、 $\omega(Cu)$ 、 $\omega(Hg)$ 、 $\omega(Cr)$ 和 $\omega(Cd)$ 的平均值分别为19.74、11.67、66.88、29.09、22.55、0.03、62.27和0.19 mg·kg⁻¹,相对宁夏土壤环境背景值存在一定程度的富集,无中度及以上污染等级.其中,Hg和Cd有中、较高等级生态风险点位,但均未超过农用地土壤污染风险管控值,所有样点均无高风险和极高风险等级.正定矩阵因子分解模型(PMF)与相关分析结合得出的源解析结果表明,研究区农田土壤重金属来源有自然来源、工矿活动及居民生产生活的混合源、交通运输源、农业生产活动源和工业源等5个主要来源,贡献率分别为26.54%、25.59%、22.52%、15.63%和9.72%.综合来看,宁夏引黄灌区农田土壤重金属均无超标现象,且没有较高等级生态风险,农田土壤生产环境条件良好,但人为活动对土壤重金属的贡献率较大.

关键词:宁夏引黄灌区;农田;重金属;生态风险评价;源分析

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Risk Assessment and Sources of Heavy Metals in Farmland Soils of Yellow River Irrigation Area of Ningxia

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Abstract: The farmland environment is directly related to the quality and safety of agricultural products. In order to understand the characteristics and main influencing factors of heavy metals in farmland soil in the Yellow River irrigation area of Ningxia, sampling and monitoring were conducted for five consecutive years from 2017 to 2021, and the distribution characteristics and correlation of heavy metals were analyzed. The pollution status and potential ecological risks of heavy metals were evaluated, and the main sources of heavy metals in farmland were analyzed. The results showed that the average values of Pb, As, Zn, Ni, Cu, Hg, Cr, and Cd in the soil of the Ningxia Yellow River irrigation area were 19.74, 11.67, 66.88, 29.09, 22.55, 0.03, 62.27, and 0.19 mg·kg⁻¹, respectively, which were enriched to some extent compared with the background values of the soil environment in Ningxia. Among them, Hg and Cd had middle- and high-grade ecological risk points; however, none of them exceeded the control value of agricultural land soil pollution risk, and all sampling sites had no high-risk or extremely high-risk levels. The results of source analysis based on positive matrix factorization (PMF) and correlation analysis showed that there were five main sources of heavy metals in farmland soil in the study area; natural sources, mixed sources of industrial and mining activities and the production and life of residents, transportation sources, agricultural production activities sources, and industrial sources, with contribution rates of 26.54%, 25.59%, 22.52%, 15.63%, and 9.72%, respectively. On the whole, the heavy metals in farmland soil in the Ningxia Yellow River irrigation area did not exceed the standard, and there was no high-level ecological risk. The production environment of the farmland soil was good, but the contribution rate of human activities to soil heavy metals was large.

Key words: Yellow River irrigation area of Ningxia; farmland; heavy metal; risk assessment; source apportionment

土壤是农业生产过程中不可或缺的自然资源,掌握农田土壤重金属的空间分布特征及污染水平,对于农田生态系统安全和人类健康具有重要意义^[1].重金属作为土壤环境中一种具有潜在危害的污染物,通常不能被微生物所降解,具有易积累、难挥发、毒性大和隐蔽性强等特点^[1,2].在土壤化学组成中重金属对土壤的理化性质影响最大,当重金属大量累积时会导致土壤性质发生变化,使土壤肥力下降,造成农作物产量和质量下降,更为严重的会导致重金属在食物链中富集,最终影响人类的健

康^[3]. 我国有 19.4%的耕地土壤重金属超过了其背景值^[4],土壤总的超标率为 16.1%,镉(Cd)、汞(Hg)、砷(As)、铜(Cu)和铅(Pb)这5种无机污染物点位超标率分别为7.0%、1.6%、2.7%、2.1%和1.5%.因此,对于土壤重金属的潜在生态风险和

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来源进行研究是有必要的,其中对于重金属的污染源进行解析,是污染治理的基础^[5].准确确定农田土壤重金属特征和污染状况,对于评价农业生态环境质量、保障食品安全和人类健康等具有重要意义,目前已受到管理部门和相关学者极大地关注.

宁夏引黄灌区素有"塞上江南"的美誉,是全国12个商品粮基地之一,有悠久的灌溉耕种历史,是中国西北重要的商品粮基地和特色农业基地.在人口不断增加和经济承载力持续提高的同时,引黄灌区已成为支撑和保障宁夏地区经济社会发展的重要基础,同时也是我国西部重要的生态屏障,对于维系黄河中上游及华北、西北地区的生态安全有着重要的作用.为了解决宁夏引黄灌区农田土壤重金属土壤污染掌握不清、潜在生态风险认识不足和污染来源判断不明的问题,本文开展了宁夏引黄灌区农田重金属潜在生态风险和来源的研究,以期为司法定责、生态补偿和环境管理提供理论指导,对未来农田污染制定有效的防治策略提供科学依据.

1 材料与方法

1.1 研究区概况

宁夏引黄灌区南接萧关与关中平原,北近乌兰布和沙漠,东临鄂尔多斯台地,西靠贺兰山天然屏障,南北长320 km,东西最宽40 km,是中国最古老的大型灌区之一,主要包括银川市、石嘴山市、吴忠市和中卫市的引黄灌溉和扬水灌溉区域,涉及11个县(市)和20多个农、林场,耕地面积约46.7万hm²,年平均气温为8~9℃,年均降水量为190~230 mm,地貌类型为黄河冲积平原,广泛分布有灌淤土,地势平坦,海拔为1100~1300 m^[6].

1.2 样品采集与测试方法

于2017~2021年的6月或9月(作物成熟期), 用五点采样法在宁夏引黄灌区采集农田0~20 cm 表层土壤,用四分法混合后获得约1 kg 的样品, 2017年采集163个,2018年采集104个,2019年采 集58个,2020年采集52个,2021年采集64个,共 441个土壤样品,装入聚乙烯袋内,贴上内外双标签 后运回实验室备用. 将采集的土壤样品置于通风处自然风干,避免阳光直射,去除砾石和根系等,一部分研磨过2 mm 尼龙筛用于测定 pH、有机质和阳离子交换量,另一部分研磨并过筛(200 目)用于测定重金属.

根据不同的测试指标,选取合适的分析方法,其 中 pH 的测定采用电位法(PHS-3C/NMIE-0421),有 机质采用滴定法(50 mL/HJ-DA1-4-A),阳离子交换 量采用滴定法(25 mL/HJ-DA2-4-A),机械组成采用 比重计法(NMIE-0462), Pb 采用石墨炉原子吸收分 光光度法(6A7CB24AD6491C3E/NMIE-0375), Cd 采用石墨炉原子吸收分光光度法 (6A7CB24AD6491C3E/NMIE-0375), Cr 用火焰原 子吸收分光光度法 (pinAAcle900F/NMIE-0083), Hg、As 采用原子荧光法(SK-2003A/NMIE-0320), Cu、Zn 和 Ni 用火焰原子吸收分光光度法 (pinAAcle900F/NMIE-0083). 为了确保实验结果的 准确性和精密性,所用试剂均为优级纯,水均为超纯 水,而且每批次抽取10%的样品进行平行样测定, 相对标准偏差控制在≤5%. 土壤样品的分析测试均 由内蒙古谱尼测试技术有限公司完成.

1.3 分析方法

1.3.1 地累积指数法

采用地累积指数法(I_{geo})评价宁夏引黄灌区农田中重金属污染程度,该方法是由德国科学家Muller提出用于研究重金属污染程度的定量指标^[7],是目前国内外广泛应用于评价土壤重金属污染程度的一种定量评价方法,不仅能反映自然成岩作用下重金属自然分布特征和累积程度,而且可以判断出人为活动对重金属污染的影响,是区分人为活动影响的重要参数^[1,8],计算公式如下:

$$I_{\text{geo}} = \log_2(C_i/1.5B_n)$$

式中, I_{geo} 为地累积指数; C_i 为实测元素 i 在土壤中的含量, $mg \cdot kg^{-1}$; B_n 为 i 元素 的 地球 化学 背景 值, $mg \cdot kg^{-1}$,采用宁夏的土壤背景值[9](表1);1.5 为考虑成岩作用可能引起背景值波动而设定的校正常数[10].

地累积指数 I_{see}划分标准见表 2.

表 1 宁夏土壤重金属背景值/ $mg \cdot kg^{-1}$

		Table I Baci	kground reference	e values of soil he	eavy metals in N	ıngxıa⁄ mg•kg		
项目	Pb	As	Zn	Ni	Cu	Hg	Cr	Cd
背景值	20. 9	11.9	58. 8	36. 5	22. 1	0. 021	60	0. 112

表 2 地累积指数分级标准

Table 2 Classification standard of soil geo-accumulation index

	- 410-1		a or oon 800 accommand		
污染等级	无污染	轻度污染	中度污染	重度污染	严重污染
$I_{ m geo}$	≤0	(0,1]	(1,2]	(2,3]	>3

1.3.2 潜在生态风险评价

采用 Hakanson 提出的潜在生态风险指数法[11]来综合评价重金属的潜在生态风险,该方法基于元素丰度响应和污染物协同效应,不仅反映了某一种金属潜在的生态风险,而且考虑了多种金属的综合生态效应^[4],是目前应用最为广泛的重金属质量评价方法之一^[12],可从生态毒性角度进行重金属的潜在生态风险评估,并根据结果划分单个重金属指标或综合指标的生态风险等

级[13],计算公式如下:

$$E_{\rm r}^i = T_{\rm r}^i \times C_{\rm f}^i$$

$$RI = \sum_{i=1}^{n} E_{r}^{i}$$

式中, E_r^i 为土壤重金属元素 i 的潜在生态风险指数; T_r^i 为单个土壤重金属的生物毒性因子; C_r^i 为土壤重金属元素 i 的污染指数;RI 为多种金属构成的潜在生态风险指数.

潜在生态风险指数分级标准如表 3.

表 3 潜在生态风险指数和分级标准

Table 3 Ecological risk evaluation index and grading criteria

_			8 8	
-	单因子潜在生态风险指数 $E_{\rm r}^i$	单因子潜在生态风险等级	综合潜在生态风险指数 RI	综合潜在生态风险等级
-	$E_{\rm r}^{i} < 40$	低风险	RI < 150	低风险
	$40 \le E_{\rm r}^i < 80$	中风险	150 ≤ RI < 300	中风险
	$80 \le E_{\rm r}^i < 160$	较高风险	300 ≤ RI < 600	较高风险
	$160 \le E_{\rm r}^i < 320$	高风险	RI≥ 600	高风险
	$E_{\rm r}^i \geqslant 320$	极高风险		COM F

毒性响应系数反映重金属的毒性强度和生物对重金属的敏感程度,本文参考瑞典学者 Hakanson^[11]

提出的重金属毒性水平顺序: Hg > Cd > As > Pb = Cu = Ni > Cr > Zn, 具体数值见表 4.

表 4 宁夏土壤重金属毒性响应系数/mg·kg⁻¹

	/ 11	rable 4	oxicity coefficients of s	on neavy metais in N	ingxia/ mg·kg	100	~) h
项目	/ Pb	As	//Z6	Ni Cu	/ Hg	Cr	Cd
毒性响应系数	% / / 5	10	1 SOV	5 5/	40	2	30

1.3.3 相关性分析

相关性分析是指对两个或多个具备相关性的变量进行分析,从而衡量两个变量因素的相关密切程度^[14].不同重金属之间相关性分析有助于辨识重金属来源^[15],具有显著正相关的重金属元素间具有相似的来源或者富集与迁移等地球化学行为;若具有显著负相关,则说明其来源具有差异,甚至具有一定的拮抗作用^[4].为了解引黄灌区农田土壤8种重金属的来源是否一致,对重金属间及与pH、有机质、阳离子交换量和机械组成进行相关性分析.

1.3.4 正定矩阵因子分解模型

源解析研究最初起源于 20 世纪 60 年代的美国,主要是应用模型进行定性分析或定量的计算^[16]. 计算模型包括以污染源为对象的扩散模型和以污染区域为对象的受体模型,而受体模型以其应用简便、解析比较全面等优势取代了扩散模型成为源解析的重要方法. 本文选用的正定矩阵因子分解模型(positive matrix factorization, PMF)是由Paatero^[17]首次提出的一种基于因子分析原理进行源解析的数据分析方法,是基于主成分分析建立的受体模型^[18],因可以同时将测量不确定性和非负性约束纳入其计算过程,依赖于更具物理意义的假设,而且不需要确定源成分谱,操作方便易行,可以较快

和更好地判断出不同污染来源^[3],因为因子矩阵被限制为非负值,可以获得更有意义的因子^[4],得到了美国环境保护署推荐使用,可以很好地对污染源进行识别和分配污染源对每种重金属的贡献率,是目前广泛应用于土壤、大气和沉积物源解析的受体模型^[3]. 该模型将原始矩阵 X_{ij} 因子化,分解为两个因子矩阵: F_{kj} 和 G_{ik} ,以及一个残差矩阵 E_{ij} ,计算公式如下:

$$X_{ij} = \sum_{i} G_{ik} F_{kj} + E_{ij}$$

式中, X_{ij} 为第 i 个样品的第 j 个重金属元素的含量; F_{kj} 为源 k 中第 j 个重金属元素的含量,即源重金属元素谱矩阵; G_{ik} 为源 k 中第 i 个样品的贡献,即源的分担率矩阵; E_{ij} 是残差矩阵,即随机误差.

PMF 需要通过多次迭代计算分解原始矩阵,得到最优矩阵 G 和 F,使目标函数达到最小值,定义的目标函数 O 为:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} (\boldsymbol{E}_{ij}/\boldsymbol{u}_{ij})^{2}$$

式中, u_{ij} 为第 i 个样品的第 j 个重金属元素的不确定度.

PMF 软件中不仅需要输入含量文件 (concentration data file),还需要不确定度文件 (uncentranty data file).不确定度数据文件的计算方

法如下:

当各个重金属元素的含量小于或等于相应的方法检出限(MDL)时,不确定度 u_{ii} 的计算公式为:

$$\mathbf{u}_{ii} = (5/6) \times MDL$$

当各个重金属元素的含量大于相应的 MDL 时,不确定度 u_{ij} 的计算公式为:

$$\mathbf{u}_{ii} = \sqrt{(a \times c)^2 + (0.5 \times MDL)^2}$$

式中, a 为相对标准偏差; c 为重金属元素含量

 $(mg \cdot kg^{-1}); MDL$ 为 重 金 属 方 法 检 出 限 $(mg \cdot kg^{-1}).$ 本研究中土壤各重金属的检出限值见表 5.

作为模型主要诊断计算的信噪比能够说明在测量值中的可变性是真实的还是数据的干扰,信噪比(S/N)越小,表明这种重金属元素在模型中越不稳定,样品被检出的可能性越小. PMF 的因子数通过多次实验的分析结果、误差和值的相对变化等确定^[10].

表 5 本研究中土壤重金属检出限/mg·kg-1

Table 5	Background refe	rence values and	toxicity coefficier	nts of soil heavy	metals in Ningxia	mg•kg ·	
Pb	As	Zn	Ni	Cu	Hg	Cr	Cd
0.1	0.08	1	3	1	0.002	4	0.01

1.4 数据处理

项目 检出限

所有数据采用 Excel 2019 和 SPSS 21.0 软件进行重金属污染统计、制图、相关性分析、污染评价和潜在生态风险评价, PMF 源解析由 EPA PMF 5.0 软件进行.

2 结果与讨论

2.1 表层土壤中机械组成、pH和阳离子交换量特征研究区农田土壤 pH值范围在7.30~9.08,平均值为8.41,总体呈碱性土壤,一定程度上会抑制重金属的活性,有助于降低土壤重金属的环境风险^[19].土壤阳离子交换量是土壤胶体所能吸附的各种阳离子的总量,可反映土壤保蓄、缓冲阳离子养

分和环境容量,是表征土壤肥力和环境质量的重要指标之一,同时也会影响其他土壤理化性质,在耕地质量调查与评价和土壤污染调查中是必测项目 $^{[20]}$. 研究区土壤阳离子交换量最大值为 27.40 cmol·kg $^{-1}$,最小值为 2.57 cmol·kg $^{-1}$,平均值为 10.02 cmol·kg $^{-1}$,变异系数达到了 0.37(表 6).有机质平均值为 14.36 g·kg $^{-1}$,低于全国有机质平均值(18.63 g·kg $^{-1}$).机械组成中 0.002 ~ 0.02 mm 粒径范围的含量平均值为 322.81 g·kg $^{-1}$; 其次是 0.05 ~ 0.25 mm 粒径的,含量平均值为 257.14 g·kg $^{-1}$; 而 0.5 ~ 1.0 mm 粒径和 1.0 ~ 2.0 mm 粒径的机械组成最少,含量平均值仅为 14.66 g·kg $^{-1}$ 和 7.00 g·kg $^{-1}$.

表 6 表层土壤机械组成、pH 和阳离子交换量特征

Table 6 Mechanical composition, pH and cation exchange capacity in the surface soils

		ω(有机质)	土壤阳离子		不同粒径机械组成含量/g·kg ⁻¹							
项目	pН	ω (有が灰) /g•kg ⁻¹	グ換量 /cmol·kg ⁻¹	<0. 002 mm	0. 002 ~ 0. 02 mm	0. 02 ~ 0. 05 mm	0. 05 ~ 0. 25 mm	0. 25 ~ 0. 5 mm	0. 5 ~ 1. 0 mm	1. 0 ~ 2. 0 mm		
采样数	441	441	441	174	174	174	174	174	174	174		
平均值	8.41	14. 36	10.02	173. 01	322. 81	159. 97	257. 14	65. 38	14.66	7. 00		
最大值	9.08	35. 80	27.40	388. 60	684.00	385. 50	609. 90	739. 30	355.80	255. 40		
最小值	7. 30	2. 15	2.57	2. 70	30.00	6. 00	65. 10	1.40	0.50	0.10		
极差	1.78	33.65	24. 83	385. 90	654.00	379. 50	544. 80	737. 90	355.30	255. 30		
标准偏差	0. 27	5.66	3.74	91. 13	139. 40	67. 08	121. 13	121. 99	40. 36	25.08		
变异系数	0.03	0.39	0.37	0. 53	0. 43	0.42	0.47	1.87	2.75	3.58		
峰度	0.96	0.80	3.37	-0.64	-0.39	0. 73	-0.02	11. 99	42. 22	63. 69		
偏度	-0.48	0. 52	1. 28	0.08	0. 16	0.41	0.71	3. 31	6.11	7. 53		

2.2 表层土壤中重金属含量特征

联合国环境规划署已将 Pb、Cd、Cr、Hg、As、Cu、Zn 和 Ni 等 8 种重金属列为优先控制污染物,我国粮食主产区耕地土壤重金属点位超标率为 21. 49% [21],耕地土壤受重金属污染的研究已成为重中之重 [4]. 宁夏引黄灌区农田 8 种重金属的描述性统计如表 7. ω (Pb)、 ω (As)、 ω (Zn)、 ω (Ni)、

 ω (Cu)、 ω (Hg)、 ω (Cr)和 ω (Cd)的平均值分别为 19.74、11.67、66.88、29.09、22.55、0.03、62.27 和 0.19 mg·kg⁻¹,其中 Pb、As 和 Ni 均低于宁夏土壤背景值,Zn、Cu、Hg、Cr 和 Cd 则分别高于背景值 1.14、1.02、1.43、1.04 和 1.70 倍,说明这 5 种重金属存在不同程度的累积富集效应;与全国表层土壤背景值相比,As、Ni、Cr 和 Cd 分别高于背景值

1.06、1.08、1.02 和 1.96 倍; 而与我国农用地土壤 污染风险筛选值相比,8 种重金属均低于筛选值,更 是远低于管制值,所以研究区土壤相对安全,没有重 金属超标管控风险.

变异系数是概率分布离散程度的一个归一化度量,一般来说离散程度越大,变异系数就越大^[4].变异系数越小,表明该重金属以本地自然背景为主;变异系数越大,则表明其分布受人为因素影响程度较大^[22].根据相关研究对变异系数(CV)的分级^[4][低变异(<0.16)、中变异(0.16~0.36)和高变异(>0.36)],宁夏引黄灌区农田8种重金属的变异系数大小排序为:Hg>Cd>As=Cu>Zn>Ni>Pb

> Cr, 系数范围在 0. 16~0. 43 间, 属于中、高变异, 表明土壤重金属空间分布不均匀, 变异程度较高, 空间离散程度较大, 受人为因素影响的可能性较大. 从各重金属元素的偏度来看, Pb、Ni、Hg、Cr和 Cd 出现了不同程度的正偏现象, 表明这些元素受到人为干扰偏离了原有的正态分布. 因此, 需要关注部分重金属较高或临近标准值的点位, 严格把控污染物输入, 如加强农业灌溉用水水质监测、强化农药化肥施用管理、严禁生活垃圾和生产废物随意堆放或直接用作肥料等, 同时建立长期、规范的农产品产地土壤和农产品质量状况监测是很有必要的管控举措.

表 7 表层土壤重金属特征统计1)

Table 7 Heavy metal characteristics in	the	surface	soils
--	-----	---------	-------

		,			, , , , , ,			
项目	Pb	As	Zn	Ni	Cu	Hg	Cr	Cd.
采样数	441	441	441	441	441	441	441	441
平均值/mg·kg ⁻¹	19. 74	11. 67	66.88	29. 09	22. 55	0.03	62. 27	0, 19
最大值/mg·kg-1	33. 40	18. 90	113. 37	53.00	42. 29	0.06	90.00	0. 33
最小值/mg·kg-1	6. 80	0.30	30.00	13.00	10. 10	0.00	35. 00	0. 03
极差/mg·kg ⁻¹	26. 60	18. 60	83. 37	40.00	32. 19	0.06	55.00	0.30
标准偏差/mg·kg-1	3. 28	2. 69	13. 55	5. 27	5. 10	0.01	10. 21	0.06
变异系数	0. 17	0. 23	0. 20	0. 18	0. 23	0.43	0. 16	0.30
峰度	0.76	0.37	0.47	0.58	-0.06	-0.46	-0.44	-0.44
偏度	0. 39	-0.26	0.00	0. 01	-0.06	0. 11	0.08	0.31
宁夏土壤元素背景值 ^[9] /mg·kg ⁻¹	20. 9	11.9	58. 8	36.5	22. 1	0.021	60	0.112
中国表层土壤背景值 ^[23] /mg·kg ⁻¹	26. 0	11.0	74. 0	27. 0	23. 0	0.065	61	0.097
农用地土壤污染风险 6.5 < pH ≤ 7.5	120. 0	30.0	250. 0	100.0	100.0	2. 400	200	0.300
筛选值 ^[24] /mg·kg ⁻¹ pH > 7.5	170. 0	25.0	300. 0	190. 0	100.0	3.400	250	0.600
农用地土壤污染风险 6.5 < pH ≤ 7.5	700. 0	120.0		_	_	4.000	1 000	3.000
管制值 ^[24] /mg·kg ⁻¹ pH > 7.5	1 000.0	100.0	_	_	_	6.000	1 300	4. 000

^{1)&}quot;一"表示未有该值

2.3 农田土壤重金属污染状况评价

掌握了重金属特征,为了解重金属累积可能引起的环境风险,还需对其进行评价.通过地累积指数法计算结果表明(表8),研究区农田土壤重金属总体较低,污染状况均在无污染和轻度污染等级,其中Pb的无污染等级占总样点数的99.77%,As无污染

等级占 99. 09%, Zn 的无污染等级占 95. 01%, Cu 则有 98. 87% 的样点为无污染等级, Hg 和 Cd 的无污染等级分别占所有采样点的 52. 38% 和 38. 55%, Ni 和 Cr 则全部为无污染等级, 所有采样点中 8 种重金属均没有中、重度和以上等级, 说明宁夏引黄灌区农田具有良好的土壤生产条件.

表 8 研究区农田土壤重金属污染程度

Table 8 Pollution degree of heavy metals in the study area

项目	元素	最大值	最小值	平均值 -	不同污染等级样点数					
坝目	儿系	取入阻	取小阻	十均恒	无污染	轻度	中度	重度	严重	
	Pb	0.09	-2.20	-0.69	440	1	0	0	0	
	As	0.08	-5.89	-0.66	437	4	0	0	0	
	Zn	0.36	-1.56	-0.43	419	22	0	0	0	
I	Ni	-0.05	-2.07	-0.94	441	0	0	0	0	
$I_{ m geo}$	Cu	0.35	-1.71	-0.60	436	5	0	0	0	
	Hg	0.99	-3.98	-0.21	231	210	0	0	0	
	Cr	0.00	-1.36	-0.55	441	0	0	0	0	
	Cd	0.97	-2.53	0.12	170	271	0	0	0	

2.4 潜在生态风险评价

研究区农田土壤潜在生态风险计算结果如表9

和表 10. 单项潜在生态风险指数 (E_r) 中, Pb、As、Zn、Ni、Cu 和 Cr 均为低生态风险; Hg 有 55. 56%

的采样点为中等级生态风险,20.63%为较高等级生态风险;Cd的采样点中较大比例(72.11%)为中等级生态风险,仅有 5.67%的较高等级生态风险.这与王玉等[14]对南方丘陵区和夏子书等[25]对宁南山区土壤重金属的研究结果相一致.虽然研究区农田土壤 Hg 和 Cd 有中等级和较高等级生态风险点位,略高于宁夏农田土壤元素背景值,但远低于农用地土壤污染风险管控值.而且 Hg 和 Cd 的毒性响应系数较大,分别为 40 mg·kg⁻¹和 30 mg·kg⁻¹,因此对 E,的贡献更大,因此对人类危害的可能性较小.所有样点中均无高风险和极高风险等级.根据综合潜在生态风险指数(RI)来看,441 个采样点中占低生

态风险的比例较高(60.77%),中等级风险的样点数为39.23%,无较高和高等级生态风险样点.

整体来看,宁夏引黄灌区农田中无高风险和极高风险等级的样点,说明研究区土壤生产条件状况良好.此外,土壤中重金属总量的高低只是反映了土壤中重金属的含量,然而土壤中重金属具有多种化学形态,重金属不同赋存形态的原生相和次生相的特性不同会对生物产生差异显著的影响.只有综合考虑土壤元素活化迁移行为、生物有效性,才能客观评价土壤重金属元素污染和危害程度.因此,准确判断和评价重金属危害作用,还需进一步深入开展重金属形态和生物有效性方面的相关研究.

表 9 潜在生态风险指数统计

Table 9 Statistical table of potential ecological risk index of soil heavy metals

	Tabl	e 9 Statistical table of	potentiai ecologicai	risk index of soil neavy	metals	
项目	元素	平均值	最小值	最大值	标准偏差	变异系数
	Pb	4. 72	1. 63	7. 99	0. 79	0.17
	As	9. 80	0. 25	15. 88	2. 26	0.23
	Zn	1. 14	0.51	1. 93	0. 23	0. 20
E_r	Ni	3. 98	1.78	7. 26	0. 72	0.18
$L_{\rm r}$	Cu	5.10	2. 29	9,57	1. 15	0. 23
	Hg	59.14	3.81	119. 52	25. 61	0. 43
	Cr	2. 08	1.17	3.00	0.34	0. 16
/ 1	Cd	51.36	7.77	88. 39	15. 40	0.30
3	RI /	137. 33	44. 60	244. 11	35.60	0. 26
1 1	/. " 10	3 B / E-	111	/ // \	J055 5	- Al

表 10 研究区农田土壤重金属生态风险等级样地比例

Table 10 Heavy metal ecological risk index of soil in the study area

5.0% IF 1	. /	Comment.			o o o o o o o o o o o o o o o o o o o	100		oreing access				
指数	元素 -	M	不同生活	态风险等级样	点数/个		不同生态风险等级样点比例/%					
1日 奴	儿系	低风险	中风险	较高风险	高风险	极高风险	低风险	中风险	较高风险	高风险	极高风险	
1	Pb	441	0	0	0	0	100.00	0.00	0.00	0.00	0.00	
-47	As	441	0	0	0	0	100.00	0.00	0.00	0.00	0.00	
	Zn	441	0	0	0	0	100.00	0.00	0.00	0.00	0.00	
$E_{ m r}$	Ni	441	0	0	0	0	100.00	0.00	0.00	0.00	0.00	
<i>L</i> _r	Cu	441	0	0	0	0	100.00	0.00	0.00	0.00	0.00	
	Hg	105	245	91	0	0	23.81	55.56	20.63	0.00	0.00	
	\mathbf{Cr}	441	0	0	0	0	100.00	0.00	0.00	0.00	0.00	
	Cd	98	318	25	0	0	22.22	72.11	5.67	0.00	0.00	
RI		268	173	0	0		60.77	39.23	0.00	0.00		

2.5 重金属来源分析

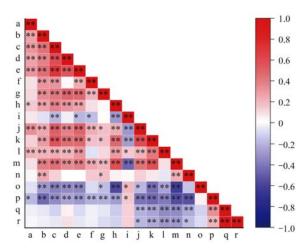
2300

仅评价重金属的风险状况,无法准确识别重金属来源,对重金属污染防控的指导性较差^[18].因此,开展土壤重金属污染的源解析研究,可以明确土壤重金属中不同污染来源及其相对贡献,为土壤重金属污染的风险监控、防治管理和保障农业生产安全提供决策依据^[3].

2.5.1 相关性分析

通过 Pearson 相关分析(图 1),发现有机质与 8 种重金属呈显著正相关关系,即随着有机质的增加, 土壤对重金属的吸附量也会增加,这与杨剑洲等^[18] 对海南岛农用地的研究结果一致.还有研究表明,施 用有机肥会导致土壤重金属增加,且随着有机肥施 用量的增加而增大^[26].

阳离子交换量与土壤有机质、黏粒、As、Zn、Ni、Cu、Hg、Cr 和 Cd 呈显著正相关关系,而与 pH 和粉砂粒呈显著负相关. 主要是因为黏粒是土壤阳离子吸收交换点的主要来源,所以与阳离子交换量有着显著相关性. 土壤交换性阳离子是以土壤胶体(有机质和矿质胶体)为载体,而有机质的阳离子交换量远大于矿质胶体,因此有机质多的土壤阳离子交换量也较高,两者间有着极显著的正相关关系. 农田中人为施入有机肥,加上作物根系分泌有机酸及秸秆还田等,都使有机作用加强,土壤形成更多有机



a. Pb,b. As,c. Zn,d. Ni,e. Cu,f. Hg,g. Cr,h. Cd,i. pH,j. SOM,k. CEC,l. MC(<0.002 mm),m. MC(0.002-0.02 mm), n. MC(0.02-0.05 mm), o. MC(0.05-0.25 mm),p. MC(0.25-0.5 mm),q. MC(0.5-1 mm),r. MC(1.0-2.0 mm),SOM 为有机质,CEC 为土壤阳离子交换量,MC 为机械组成;*表示 $P \le 0.05$,**表示 $P \le 0.01$

图 1 研究区农田土壤重金属的相关性分析

Fig. 1 Pearson's correlation coefficient of soils in the study area

胶体以及有机无机复合胶体,不断增加土壤胶体表面阳离子吸附位点,从而提高了阳离子交换量^[27].

相比之下,土壤 pH 与重金属的相关系数较小, 且与 Zn、Cu、Hg 和 Cd 呈显著负相关关系,与柴磊 等^[4]对兰州耕地的研究结果一致,表明研究区 pH 对土壤重金属富集程度影响相对较弱,可能与研究 区采集样点的土壤 pH 值差异较小,变异系数仅为 3%有关.

当机械组成 < 0.02 mm 时,与大部分重金属呈正相关关系,表明农田土壤重金属富集与土壤黏粒密切相关,这与前人的研究结论相一致^[28]. 在粒径 0.05 ~ 0.5 mm 间,机械组成与大部分重金属间呈显著负相关关系,而较大颗粒(>0.5 mm)则与重金属无显著相关性.

Pb、As、Zn、Ni 和 Cu 这 5 种重金属间具有较高的相关系数,且均为正相关关系,表明它们可能有相似的来源或者富集与迁移等地球化学行为^[4]. Hg与 As、Zn 和 Cu 也具有显著的相关性,但与 Pb和 Ni 无相关性. Cr 除与 Pb 无显著相关外,与其他 7种重金属同样有显著相关,而 Cd 仅与 Hg 和 Cr 间无显著相关性. 无显著相关的重金属间表明各自具有不同的来源^[18].

2.5.2 正定矩阵因子分析

目前,源解析技术按研究对象的不同可以分为 扩散模型与受体模型^[3].由于受体模型的研究方法 不受环境因素多变的限制,并且不依赖于排放源的 排放条件,也不用考虑污染物的迁移过程,因此得到 了更为广泛的应用^[29]. 其中, PMF 模型是一种利用相关矩阵对变量进行降维分析的受体模型, 对源谱信息的依赖性较低, 能够有效识别污染源和定量获得非负源贡献率^[30], 主要原理为利用因子分解分析压缩数据维度, 用较少的代表性因子表征原始数据的信息, 而特征污染物通常属于这一类代表性因子, 包含较多的信息, 对源解析的结果影响较大, 而其他非特征污染物在土壤中的来源相对单一, 受人类活动影响较小, 其数据特征包含的信息量较少, 在源诊断和源识别过程中的辅助判别作用有限, 其有无对源解析结果影响较小^[31], 该模型被广泛用于各种环境介质中污染物的源解析^[32].

由于富集因子分析对重金属元素来源具有不确定性,为进一步明确重金属的准确来源,对研究区内的重金属元素进行 PMF 定量源解析. 运用 EPA PMF 5.0 软件,设定因子数(3~9),运算次数设为 20 次,随机选择初始点依次运行 PMF 模型.

通过信噪比(S/N)检查数据质量,S/N>1的表明该元素是强信号,1>S/N>0.5的是弱信号,S/N<0.5的则是差信号 $[^{32]}$,而且如果 Q_{Robust} (PMF 模型在 Robust 模式下得到的目标函数 Q 的最优解)和 Q_{True} (目标函数 Q 的真值)具有较差的匹配性,那么该元素也被划定为差信号,同时结合观测值与预测值之间的相关性(R^2),确定最佳因子数. 本研究中,当选取 5 个因子时,除 Hg 元素外,其他 S/N 均>2,被定义为"strong"变量,且大部分残数值在 $-3\sim3$ 之间. 当 5 个因子情景时,实测值与模型预测值拟合系数 R^2 均>0.80(表 11),说明模型总体拟合效果较好,所选因子数可以充分地解释原始数据中所包含的重金属源信息,能满足源解析的需要.

研究区 8 种重金属污染源解析结果如图 2 所示. 因子 1 的主要金属元素是 Pb, 贡献率为54.60%.有研究表明, Pb 是交通排放的主要标志物,因为 Pb 主要来源于交通工具的燃料燃烧、汽车引擎、汽油添加剂、刹车片和轮胎摩擦等^[33,34].引黄灌区拥有相对便利的交通,公路密集度较高、铁路运输量较大,大量农田沿着交通道路分布,交通运输是土壤 Pb 积累的主要原因^[35],而且研究区农业的机械化程度较高,同样也会导致 Pb 进入农田中的机会大大增加,这与车凯等^[36]的研究结果一致. 尽管自 2000 年禁止生产、销售和使用含 Pb 汽油,但 Pb 在土壤中的累积仍然存在^[37]. 所以,可以认为因子 1 代表交通运输源.

因子 2 的主要金属元素是 Cd, 贡献率为 47.88%. Cd 是农业活动的标志元素,常作为农药的有效成分被广泛使用,易挥发和迁移^[38],在我国

表 11 不同输入变量下实测值与 PMF 模型预测值拟合结果

Table 11 F	Fitting results of meas	ed values and pred	cted values of Pl	MF model with	different input variables
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			1			
元素 -	3 个因子情景		4 个因子情景		5 个因子情景	
儿系	斜率	R^2	斜率	R^2	斜率	R^2
Pb	0. 83	0.72	0. 87	0. 90	0.88	0. 92
As	0. 83	0.80	0. 98	0. 91	1. 01	1.00
Zn	0.80	0.76	0.82	0. 78	0. 83	0.80
Ni	0. 75	0.78	0. 78	0.79	0. 81	0.81
Cu	0. 67	0.77	0.76	0.81	0. 78	0. 83
Hg	0. 51	0.43	0.51	0. 43	0. 92	0. 90
Cr	0. 83	0.76	0.86	0.80	0. 92	0. 85
Cd	0.66	0.60	0.77	0. 77	0. 85	0. 85

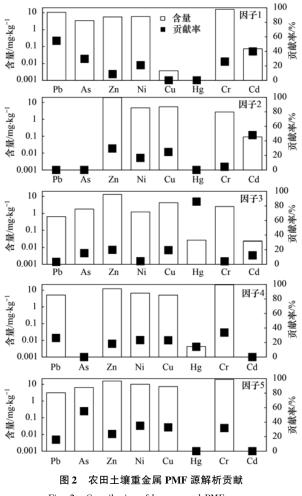


Fig. 2 Contribution of heavy metal PMF source analysis in farmland soil

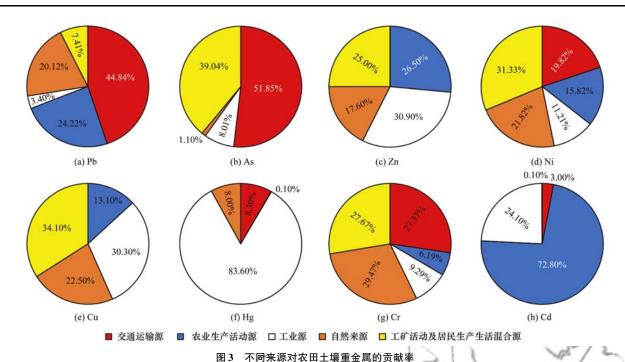
利用率较低,仅约有30%被吸收利用,而且化肥、杀虫剂的使用和污水灌溉均会增加Cd^[39,40].有研究发现长期施用磷肥会增加土壤Cd污染,一定程度上造成了重金属Cd的积累^[41].因此该因子应该表示农业生产活动源.

因子 3 的主要金属元素是以 Hg 为主要载荷, 贡献率高达 85. 81%,远高于其他元素的贡献率,可 以反映该因子的主要信息. 研究区中 Hg 的变异系 数最大(0.43),说明受到的外部干扰强度最高,该 元素变化范围和空间变异程度最大.有研究得出全球约60%~80%的 Hg 是来自于人为排放^[42],如燃煤发电、石油生产和废弃物燃烧等通过大气干湿沉降进入农田^[40].研究区分布有较多的煤矿、化工厂、矿石厂、水泥厂和砖厂等工厂企业,因此,因子3可以认为主要是工业源.

因子 4 的主要载荷金属元素是 Cr(贡献率为33.79%),虽然平均值(62.27 mg·kg⁻¹)略高于宁夏土壤元素背景值(60.00 mg·kg⁻¹),但其大部分监测点的 Cr 值低于宁夏土壤元素背景值,结合 Cr 的来源多与成土母质相关^[40,43],因此,认为因子 4 中Cr 主要受土壤背景值控制,说明主要受自然因素影响,而受人类干扰较小,可以将因子 4 定义为与成土母质和成土过程相关的自然来源.

因子 5 中 As 是主要载荷元素,贡献率为55.05%.在农田中,农业活动是导致土壤中 As 富集的主要因素而被广泛研究^[18],可能的来源是长期施肥、喷打农药和人工灌溉^[18,44,45].此外,化石燃料的燃烧也会造成大量的重金属沉降到农田土壤之中^[4],As 常作为煤燃烧的示踪物^[46].贺兰山沿南向北分布有较多的煤矿、矿石厂和砖厂等企业,加之北方之前有多年烧煤取暖的生活方式,常年燃烧产生大量飞灰所携带的重金属通过大气沉降到农田土壤中.此外,在部分调查点的周围,居民生活垃圾任意堆放,也会经雨水冲刷或渗漏进入农田导致土壤As 的积累^[47].因此,可以将因子 5 认定为工矿活动及居民生产生活的混合源.

综上所述,对宁夏引黄灌区农田土壤重金属贡献率较高的是因子4自然来源(26.54%)和因子5工矿活动及居民生产生活混合源(25.59%),其次是因子1交通运输源(22.52%)和因子2农业生产活动源(15.63%),而因子3工业源贡献率最低,为9.72%(图3).总体来看,人为活动对农田土壤重金属的贡献较大,占所有贡献率的73.45%,起着主导作用.



3 Contribution rates of different sources based on PMF to heavy metals in farmland soil

3 结论

- (1) 宁夏引黄灌区农田土壤重金属 ω(Pb)、ω(As)、ω(Zn)、ω(Ni)、ω(Cu)、ω(Hg)、ω(Cr) 和 ω(Cd) 的平均值分别为 19.74、11.67、66.88、29.09、22.55、0.03、62.27 和 0.19 mg·kg⁻¹,除Pb、As 和 Ni 外,Zn、Cu、Hg、Cr 和 Cd 均高于宁夏土壤背景值,但8 种重金属均低于我国农用地土壤污染风险管控值.
- (2)地累积指数法结果表明, Hg 和 Cd 的无污染等级分别占所有采样点的 52.38% 和 38.55%, 而其他重金属的无污染等级占总样点数的比例均在 95%以上, 均无中度、重度和以上等级的样点. 单项潜在生态风险指数(E_r)中,除 55.56% 和 20.63% 的 Hg 采样点、72.11% 和 5.67% 的 Cd 为中等和较高等级生态风险, Pb、As、Zn、Ni、Cu 和 Cr 均为低生态风险. 从综合潜在生态风险指数(RI)来看, 所有采样点中占低生态风险的比例较高(60.77%),中等级风险的样点数为 39.23%, 无较高和高等级生态风险样点.
- (3)有机质与 8 种重金属呈显著正相关关系. 阳离子交换量与土壤有机质、黏粒、As、Zn、Ni、Cu、Hg、Cr 和 Cd 呈显著正相关关系,而与 pH 和粉砂粒呈显著负相关. pH 与重金属的相关系数较小,农田土壤重金属富集与土壤黏粒密切相关. 用 PMF 法进行来源解析,结果表明宁夏引黄灌区农田土壤重金属贡献率较高的是自然来源(26.54%)和工矿活动及居民生产生活混合源(25.59%),其次是交

通运输源(22.52%)和农业生产活动源(15.63%)。 而工业源贡献率最低,为9.72%.

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