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Geo-accumulation Index (I_{geo})

The geo-accumulation index method considers both the influence of natural geological processes on background values and the effects of human activities on heavy metal contamination. [1] It allows for the assessment of deviations in soil heavy metal concentrations relative to geological background levels, which provides a robust indicator for evaluating the degree of heavy metal pollution in soils and sediments. The formula for calculating the geo-accumulation index is as follows [1,2]:

$$I_{geo} = \log_2 \frac{C_s^i}{K \times C_n^i} \quad (S1)$$

In the equation, I_{geo} represents the geo-accumulation index, C_s^i represents the measured concentration of heavy metal i in soil (mg/kg), and C_n^i represents the background value of heavy metals in soil (mg/kg) which in this study was taken as the background value for soils in Ningxia. K represents the correction factor (accounting for variations in soil heavy metal background values due to regional rock differences, typically set at 1.5). Pollution levels based on I_{geo} can be classified into seven categories: $I_{geo} \leq 0$ indicates unpolluted, $0 < I_{geo} \leq 1$ indicates unpolluted to moderately polluted, $1 < I_{geo} \leq 2$ indicates moderately polluted, $2 < I_{geo} \leq 3$ indicates to moderately to heavily polluted, $3 < I_{geo} \leq 4$ indicates heavily polluted, $4 < I_{geo} \leq 5$ indicates heavily to extremely polluted, and $I_{geo} > 5$ indicates extremely polluted.

Positive matrix factorization (PMF) model

PMF, proposed by Paatero, is a typical receptor model capable of identifying pollution sources without the need for source profiles.[3] It can weight data uncertainties and imposes non-negative constraints to ensure that source contributions are positive, thereby yielding more meaningful factors. Owing to these advantages, PMF has become the US Environmental Protection Agency's recommended general-purpose source apportionment tool and is widely used in source apportionment studies of air, water, soil and particulate matter. According to the EPA PMF 5.0 User Guide, PMF uses an iterative least-squares algorithm to decompose the matrix of sample concentration data into two matrices: the factor contribution matrix (G) and the factor component matrix (F), and then identifies the source types contributing to the sample. The basic formula is as follows:

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (S2)$$

In the equation, x_{ij} represents the concentration of heavy metal j in sample i ; g_{ik} represents the contribution of source k to sample i ; f_{kj} represents the concentration of heavy metal j in source k ; e_{ij} represents the residual of heavy metal j in sample i .

Source contributions and source composition profiles can be determined by minimising the objective function Q of the PMF model:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right)^2 \quad (S3)$$

In the equation, u_{ij} represents the uncertainty of the j heavy metal in sample i .

When the concentration of each element is less than or equal to the corresponding method detection limit (MDL), the uncertainty is as follows:

$$u_{ij} = \frac{5}{6} \times MDL \quad (S4)$$

When the concentration of each element exceeds the method detection limit (MDL), the uncertainty is as follows:

$$u_{ij} = \sqrt{(RSD \times X)^2 + (0.5 \times MDL)^2} \quad (S5)$$

In the equation, RSD represents the relative standard deviation of the heavy metal concentration; X represents the heavy metal concentration; and MDL is the detection limit of the method used to determine the heavy metal concentration.

The limits of detection for various heavy metals in soil are shown in Table S1.

PMF model configuration and solution evaluation

In this study, the PMF model was applied to apportion sources of heavy metals in the soils of the study area. The concentrations and corresponding uncertainties of As, Hg, Pb, Cd, and Cr in 515 samples were input into EPA PMF 5.0, and all elements had S/N ratios exceeding 7, so they were all classified as "strong." The measurement uncertainty was calculated using Equations (4) and (5), and the element-specific method detection limits are listed in Table S1.

To determine the optimal number of factors for the PMF model, the diagnostic results of 3-factor and 4-factor solutions were compared. The optimal factor number was determined by comparing $Q_{\text{robust}}/Q_{\text{expected}}$ values [4]. The $Q_{\text{robust}}/Q_{\text{expected}}$ ratio decreased from 2.93 to 1.99 when the number of factors increased from 3 to 4. Except for a few residuals of Pb data points falling outside the -3 to 3 range, all other data residuals were within this range. Moreover, the R^2 values of measured versus predicted concentrations for the four elements other than Pb all exceeded 0.9, indicating that the model fit was optimal when four factors were selected.

Table S1. Limits of detection for heavy metals and metalloid in soils

Element	Detection limits(mg/kg)
As	0.01 [5]
Hg	0.002 [6]
Pb	2.1 [7]
Cd	0.03 [7]
Cr	1.0 [7]

Table S2. The fit between measured and model-predicted concentrations of soil heavy metals in the PMF model.

Species	S/N	R²	Intercept	Slope
As	9.0	0.99565	0.14508	0.98511
Hg	8.4	0.99972	0.00002	0.99939
Pb	8.1	0.32790	11.98975	0.47597
Cd	7.3	0.98285	0.01192	0.95415
Cr	9.0	0.90157	6.78473	0.88321

Table S3. Compilation of Cd concentration and isotope results of soil samples and end - members

Sample Category	Source Type	Sample Size (n)	Cd (mg/kg)	$\delta^{114/110}\text{Cd}$	References
Soil in the study area	Paddy soil	1	0.28	0.40 ± 0.07	This study
	Paddy soil	1	0.51	0.57 ± 0.08	This study
	Paddy soil	1	0.44	0.63 ± 0.04	This study
	Paddy soil	1	0.20	0.39 ± 0.07	This study
	Paddy soil	1	0.45	0.32 ± 0.04	This study
	Paddy soil	1	0.23	0.42 ± 0.02	This study
	Paddy soil	1	0.22	0.18 ± 0.06	This study
Natural sources	Uncontaminated soil	30	-0.12 ± 0.02	-0.06 ± 0.18	Yin et al., 2025[8]
	Uncontaminated soil	1	0.66	-0.22 ± 0.04	Bo et al., 2025[9]
	Soil parent material	1	0.27	-0.45 ± 0.08	Feng et al., 2025[10]
Agricultural sources	Mixed fertilizers	2	0.23 ± 0.08	-0.53 ± 0.04	Xia et al., 2024[11]
	Mixed fertilizers	1	2.1	-0.26 ± 0.03	Liao et al., 2022[12]
	Mixed fertilizers	1	0.22	0.035 ± 0.03	Wu et al., 2025[13]
	Mixed fertilizers	1	0.41	-0.51 ± 0.06	Feng et al., 2025[10]
	Urea	1	5.1	-0.46 ± 0.02	Liao et al., 2022[12]
	Phosphate fertilizer	1	0.55	0.34 ± 0.01	This study
	Phosphate fertilizer	1	3	-0.36 ± 0.02	Liao et al., 2022[12]
	Phosphate fertilizer	1	2.8	-0.11 ± 0.06	Liao et al., 2022[12]
	Phosphate fertilizer	1	2.03	-0.15	Yan et al., 2021[14]
	Chemical fertilizer	1	0.08	-0.07	Liu et al., 2022[15]
	Cow dung	1	0.21	0.34 ± 0.03	This study
Coal combustion	Coal	1	0.39	-0.39 ± 0.06	Bo et al., 2025[9]
	Coal fly ash	1	16.49	-0.86 ± 0.01	Zhong et al., 2020[16]
	Coal fly ash	1	0.72	0.82 ± 0.06	Bo et al., 2025[9]
Industrial sources	Dust	1	0.40	0.03 ± 0.06	This study
	Dust	1	18772	-0.15 ± 0.14	Zhang et al., 2020[17]
	Dust	1	538	-0.11 ± 0.01	Yan et al., 2021[14]
	Dust	2	6813 ± 7611	-0.64	Cloquet et al., 2006[18]

	Dust	1	1.17	-0.64	Zhang., 2020[17]
	Smelting fly ash	1	108.79	-1.42 ± 0.06	Bo et al., 2025[9]
	Smelting fly ash	1	42280	-0.568	Zhong et al., 2021[16]
	Smelting slag	1	2.13	1.74 ± 0.06	Bo et al., 2025[9]
	Slag	1	2541	1.042	Zhong et al., 2021[16]
	Slag	1	1.5	0.01 ± 0.02	Feng et al., 2025[10]
	Slag	2	727 ± 53.74	-0.48 ± 0.06	Wen et al., 2015[19]
	Cement	1	0.53	0.09 ± 0.06	Bo et al., 2025[9]
	Atmospheric deposition	1	8.00	-0.003 ± 0.06	This study
	Road deposition	1	0.20	0.31 ± 0.07	This study
Traffic emissions	Automobile exhaust	2	3.26 ± 1.72	-0.21 ± 0.03	Yan et al., 2021[14]
	Automobile exhaust	1	2.21	-0.19 ± 0.06	Bo et al., 2025[9]
	Automobile exhaust	2	4.15 ± 0.44	-0.40 ± 0.01	Zhang., 2020[17]

Table S4. Compilation of Pb isotope results of soil samples and end - members

Sample Category	Source Type	Sample Size (n)	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{207}\text{Pb}$	References
Soil in the study area	Paddy soil	1	2.0768	1.1980	This study
	Paddy soil	1	2.0801	1.1938	This study
	Paddy soil	1	2.0762	1.1997	This study
	Paddy soil	1	2.0807	1.1954	This study
	Paddy soil	1	2.0777	1.1960	This study
	Paddy soil	1	2.0736	1.1987	This study
	Paddy soil	1	2.0687	1.1952	This study
Natural sources	Uncontaminated soil	30	2.1205 ± 0.04	1.1618 ± 0.03	Li., 2007[20]
	Uncontaminated soil	24	2.0708 ± 0.01	1.1944 ± 0.01	Wu et al., 2011[21]
Agricultural sources	Mixed fertilizers	2	2.0817 ± 0.03	1.186 ± 0.03	Xia et al., 2024[11]
	Phosphate fertilizer	1	2.0677	1.1741	This study
	Chemical fertilizer	1	1.9851	1.2396	Yuan et al., 2021[22]
	Chemical fertilizer	1	2.0070	1.2107	Chen et al., 2018[23]
	Chemical fertilizer	3	2.0157 ± 0.01	1.2113 ± 0.01	Liu et al., 2019[24]
	Chemical fertilizer	1	2.0058	1.2256	Li et al., 2025[25]
	Cow dung	1	2.0780	1.18965	This study
Coal combustion	Coal	51	2.0869 ± 0.04	1.1879 ± 0.03	Bi et al., 2017[26]
	Coal	2	2.1000 ± 0.04	1.1345 ± 0.06	Zhao et al., 2015[27]
	Coal	2	2.1395 ± 0.01	1.137 ± 0.01	Liu et al., 2015
	Coal	3	2.0784 ± 0.02	1.1903 ± 0.02	Gao et al., 2004[28]
	Coal	4	2.1230 ± 0.02	1.1611 ± 0.02	Lu et al., 2011[29]
	Coal	4	2.0693 ± 0.02	1.1930 ± 0.01	Peng et al., 2020[30]
	Coal cinder	2	2.1415 ± 0.01	1.1531 ± 0.003	Lu et al., 2011[29]
	Coal fly ash	1	2.1425	1.1464	Lu et al., 2011[29]
	Coal fly ash	1	2.1111	1.1633	Tan et al., 2006[31]
	Coal fly ash	1	2.0920	1.1906	Wang et al., 2024[32]
Industrial sources	Dust	4	2.0901 ± 0.01	1.1812 ± 0.01	This study
	Smelting fly ash	7	2.1146 ± 0.03	1.1636 ± 0.02	Zhu et al., 2020[33]

Traffic emissions	Slag	2	2.1051	1.1758	Wen. et al., 2015[19]
	Road deposition	1	2.0762	1.1967	This study
	Automobile exhaust	2	2.1125 ± 0.01	1.1655 ± 0.01	Gao et al., 2004[28]
	Automobile exhaust	3	2.0982 ± 0.02	1.1604 ± 0.004	Zhu et al., 2001[34]
	Automobile exhaust	2	2.1204 ± 0.01	1.1538 ± 0.01	Peng et al., 2020[30]
	Automobile exhaust	1	2.1344	1.1433	Yuan et al., 2021[22]
	Automobile exhaust	1	2.1110	1.1570	Wang et al., 2021[35]
	Gasoline	1	2.1400	1.1220	Gai et al., 2014[36]
	Gasoline	1	2.1236	1.1475	Bi et al., 2017[26]
	Gasoline	5	2.1828 ± 0.10	1.1148 ± 0.03	Lu et al., 2011[29]
	Gasoline	2	2.1589 ± 0.05	1.1287 ± 0.03	Tan et al., 2006[31]
	Diesel	3	2.0857 ± 0.01	1.1603 ± 0.003	Zhu et al., 2001[34]
	Diesel	2	2.1196 ± 0.002	1.1439 ± 0.01	Lu et al., 2011[29]

Table S5. Posterior summary statistics of Cd source contributions estimated by MixSIAR.

Source	Mean (%)	SD (%)	95% credible interval (%)
Agricultural	33.1	26.3	0.6–84.9
Industrial	34.3	24.8	0.7–80.1
Transportation	19.1	20.5	0.4–74.8
Natural	13.5	11.2	0.5–40.9

Table S6. Posterior summary statistics of Pb source contributions estimated by MixSIAR.

Source	Mean (%)	SD (%)	95% credible interval (%)
Coal emissions	19.2	11.4	1.3–44.1
Fertilizer	38.1	9.8	18.8–58.0
Industrial	18.3	11.7	1.2–44.7
Natural	16.9	10.8	1.0–41.2
Transportation	7.5	5.8	0.3–21.5

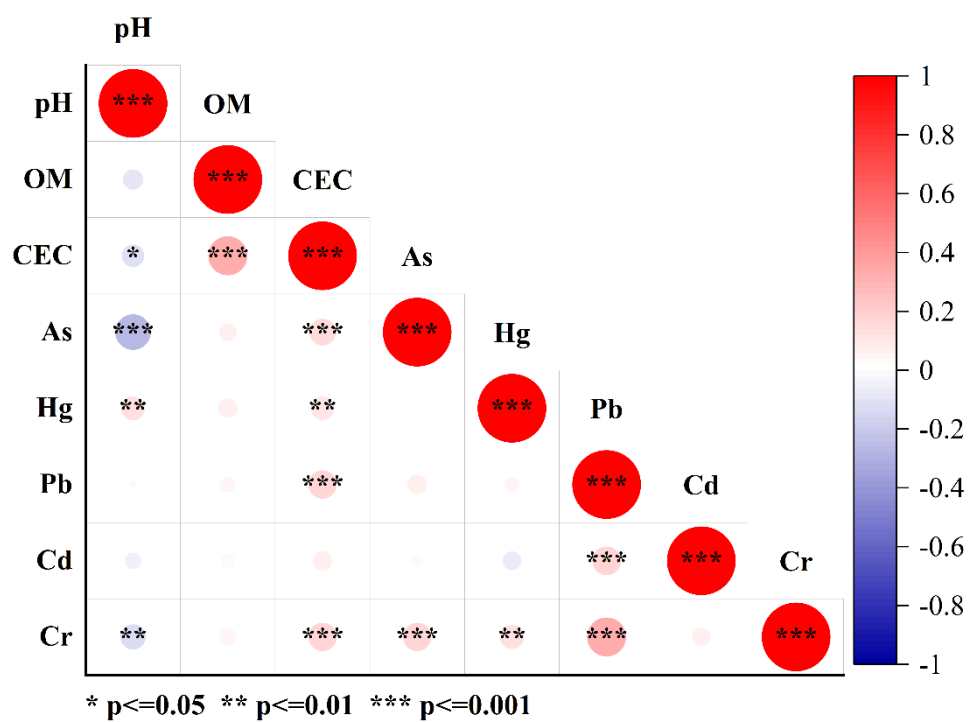


Figure S1. Pearson correlation matrix among soil physicochemical properties and heavy metal concentrations in paddy soils.

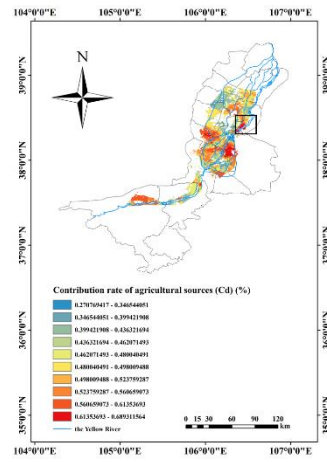


Figure S2. Distribution of agricultural source contribution rate of Cd in paddy soil based on PMF model (Black box area: Yonggu Village)

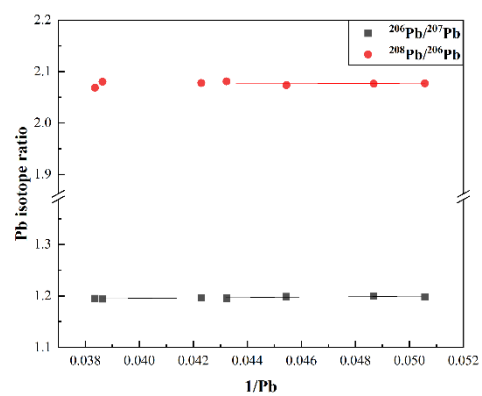


Figure S3. Correlation between Pb isotope ratios and Pb concentrations

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