

Article

Deciphering Heavy Metal Sources in Intensive Agricultural Soils of the Yangtze–Huaihe Watershed: Insights from High-Resolution Sampling and the APCS-MLR Modeling

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Abstract

Identifying the specific sources of heavy metal accumulation in intensive agricultural landscapes is essential for ensuring soil sustainability and food security. In this study, we independently carried out a high-density regional geochemical survey and high-resolution field sampling in the Yangtze–Huaihe Watershed, Eastern China, and used the original sample dataset to distinguish between geogenic backgrounds and anthropogenic enrichments. By employing the APCS-MLR model, four distinct pollution sources were quantitatively identified: natural pedogenesis, agricultural activities, traffic emissions, and industrial inputs. Results demonstrated that while most heavy metal concentrations remained below national safety thresholds, Cd and Hg exhibited significant topsoil enrichment, signaling potential ecological risks. Source apportionment revealed that natural sources primarily controlled As, Cr, Ni, and Pb, with the contribution ranging from 41% to 70%. In contrast, traffic emissions (e.g., tire wear and fuel combustion) emerged as the dominant source for Cd (68%), Zn (55%), and Cu (34%), while industrial activities accounted for a substantial 89% of Hg accumulation via atmospheric deposition. Notably, despite the region's intensive cultivation, agricultural practices played a surprisingly minor role in heavy metal accumulation. These findings highlight that the accumulations from traffic and industry now account for approximately 50% of the total heavy metal load in the region. Our results underscore the critical importance of high-resolution spatial data for precise source identification and suggest that implementing vegetative buffer zones and stricter industrial emission controls are imperative to mitigate further soil degradation in similar agricultural watersheds.

Keywords: heavy metal; source apportionment; agricultural soil; receptor model; Yangtze–Huaihe Watershed



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1. Introduction

Soil has commonly been recognized as a crucial yet vulnerable component of the terrestrial ecosystem, performing biogeochemical processes essential to life [1]. As a dynamic natural resource for shelter and food production, soil also acted as a primary reservoir for both chemical elements and compounds, and especially as a sink for heavy metals and other pollutants from various sources [2–4]. With the acceleration of urbanization and the

intensification of agriculture globally, heavy metal contamination in soils has become an increasingly serious environmental concern that threatens ecological security and human beings. Compared with other soil contaminants, heavy metals are characterized by their persistent toxicity, non-biodegradability, and tendency for long-term accumulation [5]. Heavy metal accumulation in agricultural soils may directly or indirectly pose potential threats to ecology, food safety, and human health through bioaccumulation and the food chain [6,7].

The concentrations of heavy metals in agricultural soil exhibit significant regional heterogeneity, which is jointly regulated by natural and anthropogenic factors [8–10]. Natural factors, including parent rock composition, soil particle size, organic matter content, and pedogenic processes, determine the background concentrations of heavy metals in soils [11,12]. In contrast, anthropogenic activities have become the dominant drivers of heavy metal accumulation in agricultural soils in recent decades, especially in intensively cultivated regions. These human-induced inputs mainly include industrial emissions, vehicle exhaust, fossil fuel combustion, and, most notably, agricultural practices such as the application of chemical fertilizers, pesticides, sewage sludge, and soil amendments [13–15]. For instance, Li et al. (2020) reported that the long-term application of phosphate fertilizers, which often contain impurities of Cd, has significantly exacerbated heavy metal accumulation in cultivated soils of Northern China [16]. Similarly, Pan et al. (2016) demonstrated that human inputs of Cd and Pb in some intensive agricultural areas have exceeded natural background contributions by 2–5 times [17]. Moreover, heavy metals accumulated in soils can migrate to adjacent ecosystems, such as surface water and groundwater, further expanding the scope of environmental pollution and health risks [18,19]. Consequently, investigating the concentrations, spatial distribution, and sources of heavy metals in cultivated soils is fundamental for identifying pollution hotspots, quantifying source contributions, and formulating targeted pollution control strategies [20–22].

Accurate source apportionment of heavy metals in cultivated soils is a prerequisite for developing targeted remediation strategies and ensuring sustainable soil management [23,24]. Multiple receptor models have been widely adopted as robust tools for this purpose due to their ability to quantify source contributions without prior knowledge of pollution source characteristics, including absolute principal component scores–multiple linear regression (APCS-MLR) [25], mathematical receptor model (UNMIX) [26], chemical mass balance (CMB) [27], and positive matrix factorization (PMF) [28]. Among these receptor models, the APCS-MLR model has emerged as one of the most commonly used and reliable methods in soil heavy metal source apportionment studies [29]. Distinguished from other receptor models, the APCS-MLR model requires no prior information on source profiles, simplifies computational complexity while maintaining high interpretability, and performs robustly even in low-pollution level scenarios where other models may be susceptible to noise interference [30]. The model has been successfully applied in source apportionment of heavy metals in various environmental media, including soils and sediments, and has proven effective in distinguishing natural sources and anthropogenic sources in complex ecosystems [31].

The Yangtze–Huaihe Watershed, located in Eastern China, has a long history of agricultural cultivation spanning centuries and serves as one of the most important grain production bases in China [32]. Over the past few decades, agricultural intensification in this watershed has significantly increased in response to population growth and land scarcity, characterized by high input of fertilizers, pesticides, and other agricultural chemicals. Unlike many other industrialized regions, the selected study area within the Yangtze–Huaihe Watershed is a typical intensive agricultural zone with minimal industrial disturbances. This unique setting makes it an ideal study area to investigate the effects of agricultural

activities on soil heavy metal accumulation, as the primary sources of heavy metals are expected to be agricultural practices rather than industrial or traffic-related inputs. However, there is a lack of published data on source apportionment of heavy metal contamination in this specific region, particularly studies that employ high-resolution sampling to accurately characterize pollution and reduce the uncertainty caused by sparse sampling.

To address this gap, a systematic investigation based on a high-density regional geochemical survey is urgently required. Against this backdrop, we conducted a comprehensive study in the intensive agricultural soils of the Yangtze–Huaihe Watershed, using topsoil and subsoil samples to distinguish geogenic background from anthropogenic enrichment and adopting the APCS-MLR model. The specific objectives are as follows: (1) to determine the baseline status of heavy metal accumulation in the cultivated soils of the study area; (2) to investigate the spatial distribution characteristics of heavy metals based on vertical differences between topsoil and subsoil using high-resolution sampling data; and (3) to identify the primary natural and anthropogenic sources of heavy metals and quantitatively assess their contribution ratios via the APCS-MLR model. We hypothesized that (1) heavy metals in the study area are predominantly derived from the natural pedogenesis of the uniform Xiashu loess parent material and that (2) among anthropogenic inputs, traffic emissions and industrial atmospheric deposition contribute more significantly to topsoil heavy metal enrichment, especially for Cd, Hg, and Zn. To test these hypotheses, we analyzed the geochemical characteristics of heavy metals and soil physicochemical properties, characterized the vertical and spatial distribution patterns of heavy metals, and combined correlation analysis, principal component analysis with the APCS-MLR model to quantitatively partition the contributions of different heavy metal sources. This research aims to provide a scientific basis for soil environmental protection and targeted pollution control in the Yangtze–Huaihe Watershed and other similar intensive agricultural watersheds worldwide.

2. Materials and Methods

2.1. Study Area

The study area, covering 5247 km², is located in the middle of Anhui, Eastern China (between 31°30′–32°28′ N and 116°40′–117°53′ E), which mainly includes Hefei city, Feidong county, Feixi county, and Changfeng county (Figure 1). This region is situated in a climate transition zone in China, where there is semi-humid weather in the southern warm temperate zone and humid monsoon weather in the northern subtropical zone. Thus, the ecology here is very fragile, the climatic and ecological types are diversified, the soil erosion is quite serious, the droughts and floods occur frequently, and the agricultural production is extremely unstable. This area has four distinct seasons, with an annual average temperature and precipitation ranging from 14.5 °C to 16 °C and from 900 mm to 1100 mm, respectively. These suitable climate environments provide superior natural conditions for agricultural economic development. The geomorphic features are mainly hills and plains. The parent material is dominated by Xiashu loess from the Late Pleistocene. paddy soil and yellow cinnamon soil are the major soil types, followed by skeletal soil and purple soil. There are other soil types like lime-concretion black soil, yellow–brown soil, limestone soil, as well as alluvial soil, sporadically distributed in the study area. With the rapid development of intensive agriculture here in recent years, the dominant land use type has been paddy–upland rotation, and there is still a portion of paddy land, with dryland, forest land, industrial land, and residential land sporadically distributed in this intensive agricultural area.

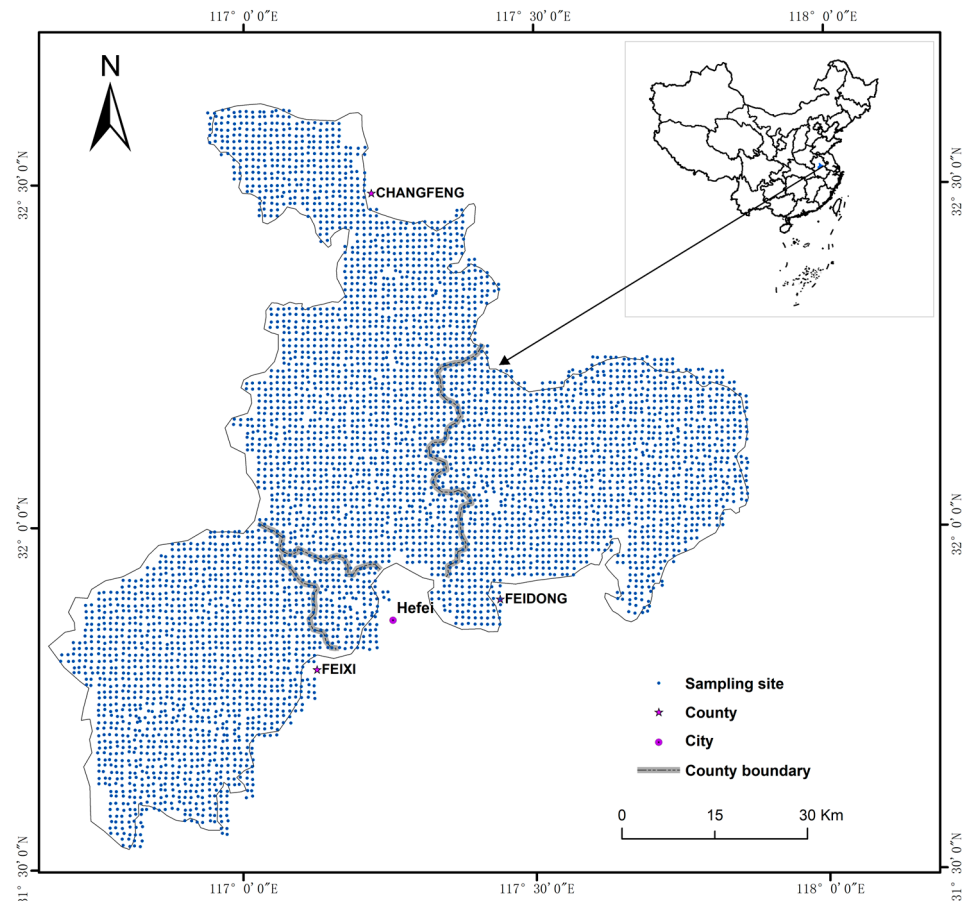


Figure 1. Locations of the study area and sampling sites.

2.2. Sampling and Chemical Analysis

Soil sampling was part of a national project named the National Multi-purpose Regional Geochemical Survey [33]. A total of 5207 topsoil samples (0–20 cm) and 1311 subsoil samples (150–180 cm) were collected from the study area in October 2003. During the sampling procedure, a square grid (1 km × 1 km) for topsoil and a grid of 2 km × 2 km for subsoil were used, covering an area of 5250 km² approximately. Among them, about 2–3% of the sampling sites were re-sampled to assess sampling variation. Soil sample locations are presented in Figure 1. Geographic coordinates of all the sample sites were recorded using a global positioning system. The surface of each sample location was cleaned of superficial debris and vegetation. Prior to physical and chemical analysis in the laboratory, all soil samples were air-dried at room temperature. Following disaggregation, samples were pulverized and sieved through a 2 mm nylon mesh. The experiments were conducted in 2003 and 2004 by Anhui Institute of Geological Experiment, located in Hefei, Anhui Province, China. The oxides/elements (Al₂O₃, TFe₂O₃, Na₂O, SiO₂, Cr, Cu, Ni, Pb, and Zn) were analyzed by an X-ray fluorescence spectrometer (XRF). The elements (As, Hg) were determined by atomic fluorescence spectrometry (AFS). Cadmium was determined by non-flame atomic absorption spectrometry (AAN). Soil organic carbon (SOC) was determined by volumetry (VOL), and soil organic matter (SOM) was calculated from SOC using a conversion factor of 1.724. Total nitrogen (N) was measured by the Kjeldahl digestion method, and total phosphorus (P) by alkali fusion–molybdenum blue spectrophotometry. Soil sulfur (S) was determined using the combustion iodometric method. Soil pH was measured potentiometrically with an ion-selective electrode (ISE): a 10.0 g subsample of sieved soil was placed into a 50 mL polypropylene beaker and mixed with 25.0 mL deionized water at a soil-to-water ratio of 1:2.5 (mass/volume). All detailed analytical procedures

were described in Bao's work (2000) [34]. Both accuracy and precision of the analytical work met the demands of the National Multi-purpose Regional Geochemical Survey [33].

2.3. APCS-MLR Model

The APCS-MLR model was used to quantify the contributions of the major sources to the concentrations of heavy metals in this study. The application of the APCS-MLR method in source analysis was first found in the study of Thurston and Spengler (1985) concerning source analysis of inhalable particulate matter in the atmosphere [35]. The model assumed that there is a linear relationship between the concentration of soil or air pollutants and the contribution rate of various sources. The principle of this model involves obtaining the rotated factor loading matrix and eigenvalues to calculate the principal factor eigenvectors through principal component analysis (PCA). Subsequently, multiple linear regression is performed using the eigenvectors and the absolute principal component scores (APCS) derived from standardized data to compute the contribution values of identified pollution sources to the substance in the receptor. The formula is as follows:

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

where C_i is the standardized value of the concentration of the element, b_{oi} is the intercept of MLR for the i th heavy metal, b_{pi} is the regression coefficient of source p on heavy metal i , and $APCS_p$ is the contribution of source p to C_i .

2.4. Relative Enrichment Coefficient Calculation

To explore the enrichment characteristics of heavy metals in topsoil, two enrichment coefficients of K_1 and K_2 were calculated in this study. Specifically, K_1 was used for comparison with national background soil concentrations [36], and K_2 for comparison with elemental concentrations in subsoil in the study area. The formulas are defined as follows:

$$K_1 = \frac{C_{topsoil}}{C_{national}} \quad (2)$$

$$K_2 = \frac{C_{topsoil}}{C_{subsoil}} \quad (3)$$

where $C_{topsoil}$ is the mean element concentration in the topsoil in the study area, $C_{national}$ denotes the corresponding arithmetic mean background concentration for the A horizon of soils across China, $C_{subsoil}$ is the mean element concentration in the subsoil in the study area.

2.5. Multivariate and Geostatistical Analysis

Basic descriptive statistics were calculated using Excel 2021. Correlation analysis was used to identify relationships between different heavy metals and physicochemical properties in topsoil samples. It is regarded as a very useful tool for the determination of multi-element relations. Principal component analysis (PCA), as an extension of correlation analysis, has frequently been used to identify underlying dimensions or factors that may describe relationships among sets of interrelated variables [37,38]. Variables can be divided into groups that are consistent with pedogenic processes or anthropogenic processes. The multivariate analysis was performed using SPSS 26.0, Microsoft Excel 2021, and Origin 2021. Ordinary kriging was applied to obtain spatial distribution patterns of heavy metals based on vertical differences in the platform of ArcGIS 10.2.

3. Results and Discussion

3.1. Concentrations of Heavy Metals and Physicochemical Properties

Table 1 summarizes the descriptive statistics for heavy metals and physicochemical properties in the study area. The coefficient of variation (CV) values of elements dominated by natural sources will be relatively lower, while CV values affected by anthropogenic sources will be quite higher [14]. As shown in Table 1, the CVs of these elements were less than or close to 25%, which illustrates that the contents of most elements in this area are derived from a single and uniform parent material. Although some regions were affected by parent material, landform, human activity, and other factors, most elements still showed a uniform distribution. Compared with the national level, the contents of elements in topsoil, such as Cr, were much higher as K_1 was higher than 1.1. Meanwhile, the concentrations of elements in topsoil, such as Hg, Zn, and others, were much lower than the national level due to the fact that K_1 was lower than 0.9. The contents of the remaining elements were comparable to those of the national level's contents. Apart from that, combined with some existing subsoil data, this study computed K_2 to reflect the elements' enrichment or depletion during pedogenesis. It is worth noting that elements with K_2 values ranging from 0.8 to 1.1 constituted the dominant proportion. This suggests that most elements in the topsoil inherit geochemical characteristics from the subsoil. Accordingly, pedogenesis and anthropogenic activities exert only a minor influence on the majority of topsoil elements. Elements which were evidently enriched in the soil consist of dispersion elements Cd, together with Hg, reaching the highest ratio up to 2.51 among these elements.

Table 1. Descriptive statistics for heavy metals and physicochemical properties in the study area.

Analyte	Unit	Min	Max	Mean	SD	CV (%)	K_1	K_2
Al ₂ O ₃	g/kg	7.49	17.79	12.26	1.32	10.8	0.98	0.83
Na ₂ O	g/kg	0.20	4.12	1.20	0.169	14.1	0.87	1.10
SiO ₂	g/kg	49.44	79.71	71.84	2.91	4.1	1.10	1.08
TFe ₂ O ₃	g/kg	1.54	11	4.37	0.71	16.3	1.04	0.75
pH	-	4.5	8.5	6.02	0.66	11.0	-	-
SOM	g/kg	1.38	49.65	17.33	4.46	25.7	-	-
N	mg/kg	0.04	0.16	0.11	0.012	10.9	-	-
P	mg/kg	0.015	0.290	0.049	0.016	32.7	-	-
S	mg/kg	51.7	441.4	192.65	45.46	23.6	-	-
As	mg/kg	3.1	16.5	9.87	2.25	22.8	0.88	0.78
Cd	mg/kg	0.029	1.27	0.10	0.02	20.0	1.03	1.61
Cr	mg/kg	14.7	197.3	68.74	8.8	12.8	1.13	0.82
Cu	mg/kg	4.9	74.3	24.71	3.11	12.6	1.09	0.89
Hg	mg/kg	0.004	1.10	0.04	0.01	25.0	0.62	2.51
Ni	mg/kg	5.7	152.7	24.98	5.04	20.2	0.93	0.70
Pb	mg/kg	13.6	69.2	25.44	2.4	9.4	0.98	1.00
Zn	mg/kg	22.5	207.9	52.12	7.77	14.9	0.7	0.83

SD: standard deviation; CV (%): coefficient of variation; K_1 : relative enrichment coefficient = mean content in study topsoil/national arithmetic mean of topsoil in China; K_2 : vertical enrichment coefficient = mean content in topsoil/mean content in subsoil; “-”: not available.

The heavy metal concentrations in agricultural soils sampled in the study area are also compared with data reported for other agricultural areas in China (Table 2). The mean heavy metal concentrations in the study area were generally lower than or close to those in several other regions in China. In general, heavy metals in the study area were slightly higher than those in Songnen Plain [39] and Taihang Piedmont Plain [40], but much lower than those in Huanghuai Plain [41] and Pearl River Delta [42]. The mean concentrations of Cr, Cu, and Ni in the study area were found to be a little higher, while Pb

and Zn were lower than those in Dehui [43], an area with traditional agricultural activity and intensive agriculture.

Table 2. Summary of heavy metal concentrations in agricultural topsoils across China.

Region	n	Mean Concentrations (mg/kg)								Reference
		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	
Huanghuai Plain	224	11.8	0.17	79.0	-	0.04	35.3	25.3	73.8	[41]
Songnen Plain	2410	7.16	0.08	50.86	41.39	0.02	21.99	24.7	51.5	[39]
Taihang Piedmont Plain	100	6.16	0.15	52.8	21.2	0.08	25.0	18.8	69.9	[40]
Pearl River Delta	1384	12.81	0.18	48.07	-	-	24.4	50.1	-	[42]
Dehui, Jilin	73	6.47	0.103	55.81	19.82	0.123	24.38	29.24	63.25	[43]
Tibet	239	19.27	0.14	155.5	24.27	0.04	55.86	32.15	75.59	[44]
Taiyuan	139	10.7	0.25	74.10	32.1	0.09	29.74	27.87	90.76	[45]
Inner Mongolia	163	12.11	0.08	61.77	17.07	0.05	26.77	12.61	85.71	[46]
Topsoil in this study area	5207	9.87	0.099	68.74	24.71	0.039	24.98	25.44	52.12	This study
Subsoil in this study area	1311	12.43	0.061	82.51	27.29	0.014	35.50	25.77	61.24	This study
Anhui Province	37,427	10.0	0.161	69.1	26.4	0.059	28.2	26.7	65.4	[47]
Background value, China	4095	11.2	0.097	61.00	22.60	0.065	26.90	26.00	74.20	[36]
EQSS	-	30	0.30	250	50	0.30	40	250	200	[48]

n: number of samples; EQSS represents Environmental Quality Standard for Soils in China [48]; “-”: not available.

Compared with background values in China [36], the mean concentrations of As, Hg, Ni, Pb, and Zn were not higher than the background, while those of Cr and Cu were slightly higher than the national background values. The mean concentration of Cd in topsoil was 0.099 mg/kg, which was marginally higher than the background. Statistical results from 5207 samples showed that there were approximately 71.5% for As, 57.2% for Cd, 92.0% for Hg, 66.0% for Ni, 59.4% for Pb, and 98.6% for Zn, lower than the corresponding background values in China [36]. Therefore, there were insignificant human inputs. In addition, the average concentrations of all eight metals were all below the limits of the Chinese Environmental Quality Standard for agricultural soils [48]. However, some of the maximum values (Table 1) greatly exceed the values in the standard (e.g., 1.27 versus 0.3 mg/kg for Cd, 152.7 versus 40 mg/kg for Ni, 1.1 versus 0.3 mg/kg for Hg, and 74.3 versus 50 mg/kg for Cu). It can be inferred that this area has not been suffering contamination risks from these heavy metals. Compared with previous studies in Anhui Province, the mean concentrations of all eight heavy metals in this study were found to be lower than the corresponding provincial averages reported in the literature [47]. Meanwhile, the mean concentrations of the heavy metals in topsoil were all lower than those in subsoil, except Cd and Hg in this study, which illustrated that Cd and Hg were very likely to derive from human inputs, while the other metals were inherited from parent materials.

3.2. Spatial Distributions of Heavy Metals Based on Vertical Differences

The spatial distribution of heavy metal concentration is a useful tool to identify the possible sources of enrichment and the hotspots with high metal concentrations [49]. In the present study, to effectively interpret the enrichment characteristics and potential sources of heavy metals in soils, we employed the concentration differences between topsoil and subsoil to produce the spatial distribution maps of heavy metals. The vertical differences in the selected heavy metals, including As, Cd, Cu, Cr, Hg, Ni, Pb, and Zn, were presented in Figure 2. Three distinct spatial patterns were identified from the geochemical maps across this intensively cultivated agricultural area. For As, Cr, Cu, Ni, and Zn, concentrations in topsoil were generally lower than those in subsoil, indicating that these heavy metals were mainly controlled by soil parent material and exhibited weak anthropogenic influence. In addition, the high concentrations of As, Cr, Cu, Ni, and Zn in subsoil were found to the north and east of the study area. In contrast, Cd and Hg showed significantly higher

concentrations in topsoil than in subsoil, suggesting obvious enrichment in topsoil and potential ecological risks. This pattern reflects the significant impact of anthropogenic activities such as agricultural practices and atmospheric deposition. For Pb, surface concentrations were slightly higher than those in subsoil in the northern and southwestern areas but relatively lower in the eastern region, demonstrating that the accumulation of Pb was jointly regulated by soil-forming processes and human activities.

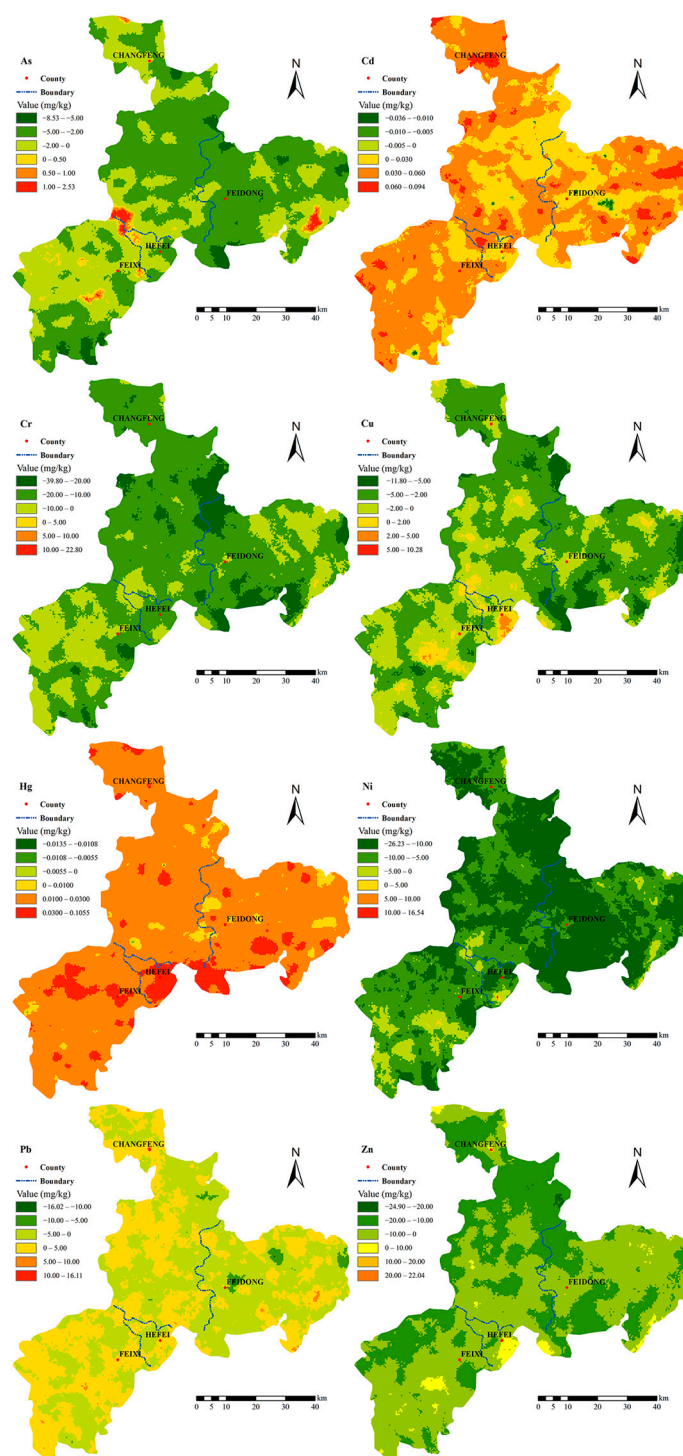


Figure 2. Spatial distribution of vertical concentration differences (topsoil–subsoil) of heavy metals in the study area (positive values indicate topsoil enrichment; negative values indicate topsoil depletion relative to subsoil).

3.3. Correlation Analysis and Principal Component Analysis

Pearson's correlation coefficients between heavy metal concentrations and physicochemical properties in topsoil are depicted in Figure 3. The correlation coefficients among 17 variables showed a considerable degree of linear relationships between each pair of variables. All the metals exhibited positive relations except for Hg, which seemed to be an isolated element, showing low correlations with other variables and a negative correlation with Al_2O_3 and TFe_2O_3 . Arsenic, Cr, Cu, Ni, Pb, and Zn were significantly and positively correlated with Al_2O_3 and TFe_2O_3 , while negatively associated with SiO_2 and Na_2O at the 0.01 significance level, which indicated their probable common origin (Table S1). Al_2O_3 and TFe_2O_3 of the soils are regarded as important products of parent rock weathering. Therefore, it could be inferred that these metals may have a lithogenic source. Cadmium showed no significant correlations with Al_2O_3 and TFe_2O_3 , while significantly associated with N, P, S, and SOM, indicating that the source of Cd is very likely to be associated with fertilizer uses. It was also observed that Pb, Cu, and Zn are significantly positively correlated with Cd apart from the strong association with Al_2O_3 and TFe_2O_3 (Table S1), which suggests they are likely to derive from both natural and anthropogenic resources.

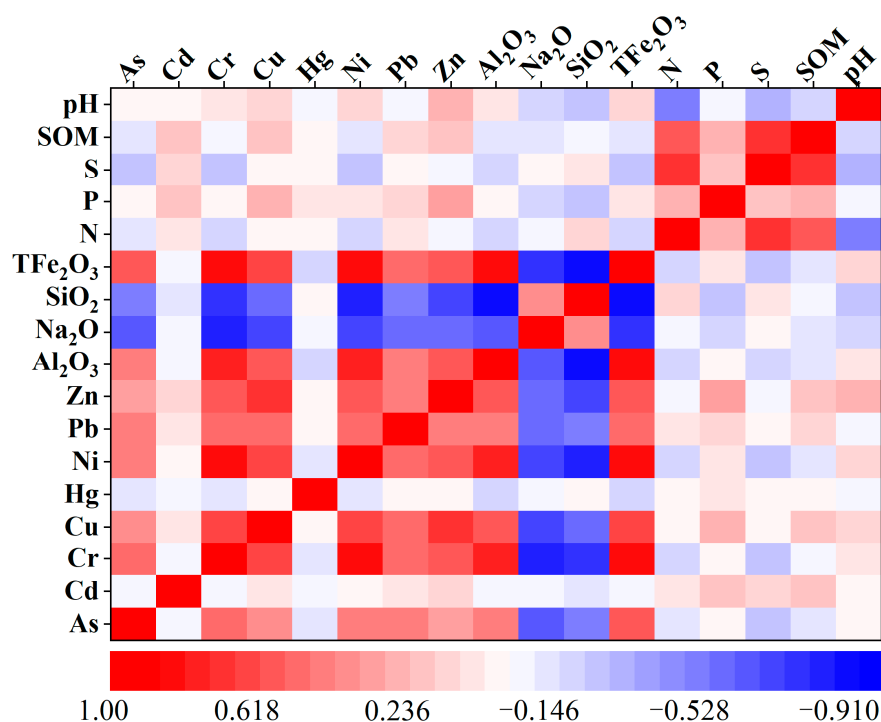


Figure 3. Heat map of Pearson's correlation coefficients between heavy metals and physicochemical properties.

To better quantify the relationships and identify the source of these heavy metals, PCA was performed. In the present study, the first four factors were chosen on the basis of the initial eigenvalues being greater than 1.0 (Table 3). These factors explained 73.38% of the total variance using the 17 variables in the analysis. Factor 1 explained 40.03% of the total variance and was the most important factor, which was strongly negatively loaded with SiO_2 (−0.863), positively loaded with Al_2O_3 , TFe_2O_3 , Cr, and Ni (0.883–0.957), and partly loaded with As, Cu, Pb, and Zn (0.656–0.817). Consistent with prior geochemical studies, the correlation of these heavy metals can be interpreted by an anthropogenic or geogenic origin [50,51]. Factor 1 is thus interpreted as a dominant geogenic component, as Al_2O_3 , TFe_2O_3 , SiO_2 , Cr, and Ni exhibit the highest loadings in the agricultural soils of this study. Previous studies have indicated that Al_2O_3 and TFe_2O_3 of the soils are regarded

as important products of parent rock weathering, and the concentrations of Cr and Ni are dependent on their concentrations in lithological units, and also in basic and ultrabasic rocks. The lithogenic nature of Al_2O_3 , TFe_2O_3 , SiO_2 , Cr, and Ni has been clearly demonstrated by Esmaeili et al. (2014) and Martín et al. (2013), where Al_2O_3 , TFe_2O_3 , SiO_2 , Cr, and Ni were also grouped in the same component [50,51]. In addition, the parent material in this region is uniform with Xiashu Loess, hence these heavy metals in this study are very likely to originate from the soil parent materials and might be related to loess-deposited parent rocks as the natural sources, which is in accordance with previous works [52]. Although geogenic origins were the primary sources of heavy metals in the study area, the role of anthropogenic activities cannot be ignored, especially in the long term.

Table 3. Total variance explained and component matrix (4 factors selected) for agricultural topsoil.

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	7.06	41.53	41.53	7.06	41.53	41.53	6.80	40.03	40.03
2	3.00	17.67	59.20	3.00	17.67	59.20	2.87	16.90	56.93
3	1.38	8.12	67.32	1.38	8.12	67.32	1.69	9.92	66.85
4	1.03	6.06	73.38	1.03	6.06	73.38	1.11	6.53	73.38
5	0.82	4.81	78.19						
6	0.74	4.36	82.55						
7	0.70	4.14	86.70						
8	0.48	2.84	89.53						
Component matrix									
Parameters	Component matrix				Rotated component matrix				
	1	2	3	4	1	2	3	4	
As	0.656	−0.064	−0.224	0.251	0.706	−0.045	−0.160	0.148	
Cd	0.108	0.376	0.470	−0.239	−0.008	0.240	0.610	−0.033	
Cr	0.928	−0.082	−0.145	0.004	0.936	−0.079	0.039	−0.070	
Cu	0.817	0.230	0.139	0.062	0.775	0.140	0.330	0.118	
Hg	−0.052	0.180	0.335	0.829	−0.048	0.020	0.075	0.909	
Ni	0.917	−0.085	−0.041	−0.054	0.900	−0.107	0.147	−0.095	
Pb	0.680	0.253	−0.109	0.235	0.704	0.225	0.032	0.215	
Zn	0.791	0.195	0.353	−0.068	0.698	0.058	0.548	0.052	
Al_2O_3	0.883	−0.113	−0.111	−0.158	0.873	−0.105	0.103	−0.217	
Na_2O	− 0.754	−0.023	0.184	−0.202	− 0.789	−0.026	0.067	−0.125	
SiO_2	− 0.863	0.095	−0.030	0.210	− 0.821	0.123	−0.246	0.223	
TFe_2O_3	0.957	−0.102	−0.145	−0.032	0.961	−0.097	0.051	−0.108	
N	−0.096	0.862	−0.317	−0.008	−0.038	0.918	−0.086	0.036	
P	0.240	0.530	0.366	0.049	0.165	0.389	0.493	0.230	
S	−0.115	0.870	−0.129	−0.137	−0.104	0.883	0.120	−0.029	
SOM	0.060	0.861	0.066	−0.104	0.032	0.809	0.317	0.053	
pH	0.256	−0.321	0.703	−0.114	0.106	− 0.507	0.637	0.042	

Extraction method: principal component analysis; rotation method: varimax with Kaiser normalization, rotation converged after 25 iterations. Bold values indicate high factor loadings with absolute value ≥ 0.5 , representing the dominant variables of each principal component.

Factor 2, including Cd, Pb, N, P, S, and SOM, explains 16.90% of the total variance. Cadmium is considered to be the most critical toxic metal threatening food safety and agricultural sustainability in China, and special emphasis should be placed on Cd [53]. It is easy to connect N, P, S, and SOM with soil fertilizer uses, especially phosphate fertilizer, in which the concentration of Cd was relatively high. The mean concentration of Cd in the phosphate fertilizers can be as high as 0.61 mg/kg in China [54]. The higher loading of N, P, and S is mainly associated with inorganic fertilizers used in agricultural soils.

Thus, the enrichment of Cd is supposed to be related to fertilizer use. There are still other anthropogenic sources, such as the application of dust, pesticides, sewage irrigation, and industrial activities, as has been found in previous studies in other areas [55,56]. Factor 3 accounted for 9.92%, including Cd, Cu, and Zn. Factor 4 accounted for 6.53% of the total variance, and was dominated exclusively by Hg (0.909). The spatial maps of the four PCA factor scores were presented in Figure S1. The spatial distribution of Factor 1 exhibited minimal heterogeneity across the entire study area (Figure S1a), which aligns with the uniform geochemical properties of the Xiashu loess parent material and strongly supports its interpretation as the natural pedogenic source. Factor 4 scores (Figure S1d) showed distinct high-value clusters around Hefei City, matching the spatial concentration of industrial activities in this urban core region and corroborating the industrial input source attribution. Therefore, factor 4 was assumed to be related to industrial activities.

3.4. Source Apportionment by APCS-MLR Model

To quantitatively delineate the contributions of various potential origins to soil heavy metal accumulation, the APCS-MLR model was implemented based on the results of PCA. Prior to the contribution assessment, the reliability of the model was verified by comparing the predicted values with the measured concentrations. The high correlation coefficients ($R^2 > 0.7$) and the ratios of measured-to-predicted mean values (close to 1.0) for most elements confirmed that the APCS-MLR model provides a robust and applicable framework for source apportionment in this intensive agricultural landscape.

Four distinct factors were identified, and their contributions were exhibited in Figure 4, including natural, agricultural, traffic, and industrial origins. Factor 1 is identified as natural sources, with contributions of 49%, 70%, 55%, and 41% for As, Cr, Ni, and Pb, respectively. These elements exhibited high positive loadings on the first principal component and demonstrated a strong geochemical affinity with Al_2O_3 and TFe_2O_3 (Table 3). As these oxides are fundamental products of parent rock weathering, it can be inferred that these metals are intrinsically linked to the mineralogical composition of the Xiashu loess [57]. Meanwhile, the relatively low CVs for Cr and Ni, lower than 25%, and the lack of topsoil enrichment lower than 1.0 further exemplify that their distribution is controlled by pedogenic processes rather than exogenous disturbance (Table 1). This aligns with the natural background profiles often observed in uniform loess-deposited regions where geogenic inputs overshadow anthropogenic signals for these specific transition metals.

Factor 2, characterized by associations with Cd, Pb, and soil nutrients (N, P, and SOM), was defined as the agricultural source, specifically the application of fertilizers. While the overall contribution to total heavy metal load was relatively slight, this factor specifically targets the accumulation of Pb and Cd through long-term soil amendments. Previous studies have highlighted that the long-term application of organic fertilizers, such as livestock manure (e.g., pig and chicken manure) and sewage sludge, is a significant pathway for Pb and Cd accumulation in soils [58]. For instance, long-term field experiments in red soil regions have demonstrated that manure-treated soils exhibit significantly higher Pb concentrations compared to those receiving only chemical fertilizers [59]. Similarly, studies in greenhouse vegetable production areas have confirmed substantial accumulation of both total and available Pb following organic amendments [60]. In this study, the significant correlation between Cd and SOM suggests that organic matter may act as a carrier, facilitating the retention of these metals in the topsoil. However, the relatively lower contribution ratio compared to other anthropogenic sources suggests that modern fertilization practices in the Yangtze–Huaihe Watershed might be more regulated than in other heavily contaminated plains in China.

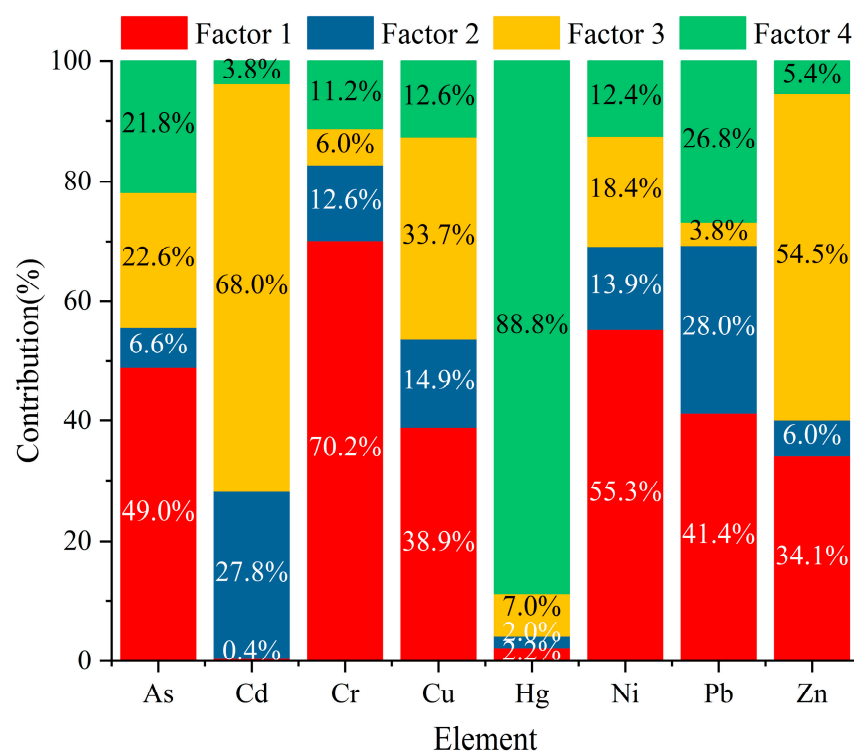


Figure 4. Quantitative source contributions of heavy metals in agricultural soils calculated by the APCS-MLR model. Factor 1: natural pedogenesis; Factor 2: agricultural activities; Factor 3: traffic emissions; Factor 4: industrial inputs.

Factor 3 represents traffic emissions, which emerged as the primary contributor to Cd (68%), Zn (55%), and Cu (34%). These metals are typically derived from gasoline combustion, tire abrasion, and the wear of brake pads and clutch linings [61]. In particular, Cd and Zn are often associated with tire wear and road construction materials (e.g., asphalt and sealants), while Cu is a primary component of brake pads and clutch linings [62]. During vehicle operation, mechanical friction and tire-road abrasion release metal-bearing microparticulates into the atmosphere. Given the dense road network traversing this agricultural watershed, these traffic-derived particles are dispersed via wind and subsequently deposited onto surrounding agricultural land through atmospheric deposition [63]. Our results identified that this factor accounted for 68% of Cd, emphasizing that traffic emissions, rather than conventional agricultural inputs, have become the dominant driver of Cd accumulation in the topsoil of this region.

Factor 4 was exclusively dominated by Hg, with a contribution of 89% (Figure 4). The “isolated” statistical behavior of Hg in the PCA and correlation analysis suggests a source distinct from both the soil parent material and other metallic elements. Industrial flue gas is widely acknowledged as the primary source of Hg, which accumulates in topsoil through dry and wet atmospheric deposition [20]. The study area is characterized by dense populations and a robust industrial base, with coal mining, coal washing, and equipment manufacturing serving as economic pillars [3]. Hg released during industrial production processes settles into the soil, confirming Factor 4 as an industrial source. Previous studies have demonstrated the anthropogenic sources of Hg, where Hg was also classified in an isolated group [64]. Gosar et al. (2016) reported that the main sources of mercury were all from human inputs and could be divided into the following categories: the use of mercury in industrial processes, consumer products containing mercury, waste incineration, cremation, and hazardous waste disposal [65]. The result in this study underscores the

vulnerability of agricultural soils to regional industrialization, even in areas primarily used for cultivation.

In summary, heavy metal accumulation in the study area is driven by a combination of natural parent material and anthropogenic activities (agriculture, traffic, and industry), with the latter two accounting for approximately 50% of the total contribution. However, in our study, it seems reasonable to tentatively conclude that agricultural activities play insignificant roles in heavy metal pollution. The results are not consistent with the previous findings of Songnen Plain [39], Huanghuai Plain [41], and Dehui [43], where the intensive agricultural practices are important sources exerting profound influences on the heavy metal concentrations. However, what needs to be emphasized here is that although agricultural activities are likely to be the source of some heavy metals, they may just play a very slight role in heavy metal accumulation for agricultural soils. And it is noticed that the sampling density in this study proved to be much higher than the abovementioned research. In addition, this selected region is mostly covered by cultivated land with few disturbances from industrial activities. Hence, the results we drew might be much more reliable for agricultural soils. Further studies should be carried out to find inter-parameter associations existing between different elements or combine with more specific data, such as land use, irrigation, etc. Additionally, to mitigate further contamination, it is recommended that local authorities implement protective measures, such as establishing vegetative buffer zones to intercept traffic dust and adopting stricter emission controls for industrial facilities.

4. Conclusions

This study conducted a high-resolution geochemical survey and source apportionment of heavy metals in intensive agricultural soils of the Yangtze–Huaihe Watershed based on 5207 topsoil and 1311 subsoil samples. By integrating a high-density regional geochemical survey with the APCS-MLR model, we have achieved a precise understanding of the complex interplay between natural pedogenesis and anthropogenic pressures. The core findings and their implications are summarized as follows:

(1) All eight typical heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, Zn) had mean concentrations below the limits of the Chinese Environmental Quality Standard for Agricultural Soils, indicating no overall soil contamination risk. Cr and Cu showed slight enrichment relative to national soil background values, and Cd had negligible enrichment; notably, Cd and Hg exhibited significant topsoil enrichment ($K_2 = 1.61$ and 2.51), with their maximum concentrations far exceeding the national standard, posing potential ecological risks to the regional agroecosystem.

(2) As, Cr, Cu, Ni, and Zn were dominated by soil parent material, with lower topsoil concentrations than subsoil and homogeneous spatial distribution. Cd and Hg were driven by anthropogenic inputs, showing significant topsoil enrichment and obvious spatial heterogeneity. Pb was jointly regulated by natural pedogenesis and human activities, with slight topsoil enrichment only in the northern and southwestern study area and depletion in the east.

(3) The APCS-MLR model identified four heavy metal sources in the study area: natural pedogenesis contributed 41–70% to As, Cr, Ni and Pb, being their dominant source; among anthropogenic sources, traffic emissions were the primary contributor to Cd (68%), Zn (55%) and Cu (34%), industrial atmospheric deposition accounted for 89% of Hg accumulation, and agricultural activities played a minor role in overall heavy metal accumulation. Traffic and industrial sources collectively contributed approximately 50% of the total heavy metal load in the study area.

A key insight of this research is that despite the study area being an intensive cultivation zone, agricultural activities play a relatively minor role in heavy metal accumulation

compared to industrial and traffic sources. This finding challenges traditional assumptions about heavy metal sources in China's farmlands and gains substantial credibility from the high-resolution sampling density employed here, which effectively captured subtle anthropogenic signals that lower-density surveys might overlook. For soil sustainability, we recommend establishing vegetative buffer zones along major traffic corridors to intercept metal-bearing dust and enforcing stricter industrial atmospheric emission standards to mitigate Cd and Hg input. Subsequent research will collect contemporary soil and crop samples to analyze the long-term evolution of heavy metal accumulation and soil–plant transfer characteristics. In conclusion, this study establishes the earliest high-resolution geochemical baseline for heavy metals in the Yangtze–Huaihe Watershed. The findings challenge traditional views on heavy metal sources in intensive agricultural areas and provide a scientific basis for targeted soil environmental management in similar regions globally.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/agronomy16070690/s1>, Table S1: Pearson correlation coefficients between soil heavy metals and physicochemical properties in the study area. Figure S1: Spatial distribution of PCA factor scores for heavy metal sources in the study area.

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References

1. Hartmann, M.; Six, J. Soil structure and microbiome functions in agroecosystems. *Nat. Rev. Earth Environ.* **2023**, *4*, 4–18. [\[CrossRef\]](#)
2. Ahmed, R.S.; Abuarab, M.E.; Ibrahim, M.M.; Baioumy, M.; Mokhtar, A. Assessment of environmental and toxicity impacts and potential health hazards of heavy metals pollution of agricultural drainage adjacent to industrial zones in Egypt. *Chemosphere* **2023**, *318*, 137872. [\[CrossRef\]](#)
3. Jia, X.; Fu, T.; Hu, B.; Shi, Z.; Zhou, L.; Zhu, Y. Identification of the potential risk areas for soil heavy metal pollution based on the source-sink theory. *J. Hazard. Mater.* **2020**, *393*, 122424. [\[CrossRef\]](#) [\[PubMed\]](#)
4. Pulkrabová, J.; Černý, J.; Száková, J.; Švarcová, A.; Gramblička, T.; Hajšlová, J.; Tlustoš, P. Is the long-term application of sewage sludge turning soil into a sink for organic pollutants?: Evidence from field studies in the Czech Republic. *J. Soils Sediments* **2019**, *19*, 2445–2458. [\[CrossRef\]](#)
5. Lv, J.; Liu, Y.; Zhang, Z.; Dai, J.; Dai, B.; Zhu, Y. Identifying the origins and spatial distributions of heavy metals in soils of Ju country (Eastern China) using multivariate and geostatistical approach. *J. Soils Sediments* **2015**, *15*, 163–178. [\[CrossRef\]](#)
6. Sun, W.C.; Ye, J.; Lin, H.; Yu, Q.G.; Wang, Q.; Chen, Z.; Ma, J.C.; Ma, J.W. Dynamic characteristics of heavy metal accumulation in agricultural soils after continuous organic fertilizer application: Field-scale monitoring. *Chemosphere* **2023**, *335*, 139051. [\[CrossRef\]](#)
7. Liu, P.; Wu, Q.; Hu, W.; Tian, K.; Huang, B.; Zhao, Y. Effects of atmospheric deposition on heavy metals accumulation in agricultural soils: Evidence from field monitoring and Pb isotope analysis. *Environ. Pollut.* **2023**, *330*, 121740. [\[CrossRef\]](#) [\[PubMed\]](#)
8. Krishna, A.K.; Mohan, K.R. Distribution, correlation, ecological and health risk assessment of heavy metal contamination in surface soils around an industrial area, Hyderabad, India. *Environ. Earth Sci.* **2016**, *75*, 411. [\[CrossRef\]](#)
9. Liu, Q.; Yang, P.; Hu, Z.; Shu, Q.; Chen, Y. Identification of the sources and influencing factors of the spatial variation of heavy metals in surface sediments along the northern Jiangsu coast. *Ecol. Indic.* **2022**, *137*, 108716. [\[CrossRef\]](#)
10. Xu, Y.; Bi, R.; Li, Y. Effects of anthropogenic and natural environmental factors on the spatial distribution of trace elements in agricultural soils. *Ecotoxicol. Environ. Saf.* **2023**, *249*, 114436. [\[CrossRef\]](#)

11. Zinn, Y.L.; de Faria, J.A.; de Araujo, M.A.; Skorupa, A.L.A. Soil parent material is the main control on heavy metal concentrations in tropical highlands of Brazil. *Catena* **2020**, *185*, 104319. [\[CrossRef\]](#)
12. Liu, T.; Li, J.; Liu, L.; Jiang, Y.; Sun, S.; Deng, T.; Wang, S.; Tang, Y.; Ni, Z.; Lin, Q.; et al. Mobilization of water-dispersible colloidal chromium and nickel in basalt-derived paddy soils: Role of Fe (hydr) oxides and organic matter. *J. Hazard. Mater.* **2025**, *496*, 139188. [\[CrossRef\]](#)
13. Kara, M.; Dumanoglu, Y.; Altok, H.; Elbir, T.; Odabası, M.; Bayram, A. Spatial distribution and source identification of trace elements in topsoil from heavily industrialized region, Aliaga, Turkey. *Environ. Monit. Assess.* **2014**, *186*, 6017–6038. [\[CrossRef\]](#) [\[PubMed\]](#)
14. Karim, Z.; Qureshi, B.A.; Mumtaz, M. Geochemical baseline determination and pollution assessment of heavy metals in urban soils of Karachi, Pakistan. *Ecol. Indic.* **2015**, *48*, 358–364. [\[CrossRef\]](#)
15. Jia, Z.; Zheng, C.; Zhou, S.; Wang, Z.; Xie, Y. Drivers of antagonistic spatial association patterns between soil and rice heavy metals in karst region: Evidence from bioavailability and Pb isotopes. *Plant Soil.* **2026**, *519*, 2071–2088. [\[CrossRef\]](#)
16. Li, H.; Yang, Z.; Dai, M.; Diao, X.; Dai, S.; Fang, T.; Dong, X. Input of Cd from agriculture phosphate fertilizer application in China during 2006–2016. *Sci. Total Environ.* **2020**, *698*, 134149. [\[CrossRef\]](#) [\[PubMed\]](#)
17. Pan, L.B.; Ma, J.; Wang, X.L.; Hou, H. Heavy metals in soils from a typical county in Shanxi Province, China: Levels, sources and spatial distribution. *Chemosphere* **2016**, *148*, 248–254. [\[CrossRef\]](#)
18. Wu, J.; Song, J.; Li, W.; Zheng, M. The accumulation of heavy metals in agricultural land and the associated potential ecological risks in Shenzhen, China. *Environ. Sci. Pollut. Res.* **2016**, *23*, 1428–1440. [\[CrossRef\]](#)
19. Yan, F.; Li, N.; Wang, J.; Wu, H. Ecological footprint model of heavy metal pollution in water environment based on the potential ecological risk index. *J. Environ. Manag.* **2023**, *344*, 118708. [\[CrossRef\]](#)
20. Yang, Q.; Yang, Z.F.; Zhang, Q.Z.; Ji, W.B.; Guan, D.X.; Liu, X.; Yu, T.; Wang, L.; Zhuo, X.X.; Ji, J.F. Transferability of heavy metal (loid)s from karstic soils with high geochemical background to peanut seeds. *Environ. Pollut.* **2022**, *299*, 118819. [\[CrossRef\]](#)
21. Luo, Y.; Yang, C.; Zhou, S.; Zhang, B.; Su, S.; Lu, S.; Yang, Z.; Feng, B.; Liu, S.; Liu, L.; et al. Evaluation of soil heavy metals in major sugarcane-growing areas of Guangxi, China. *Agronomy* **2026**, *16*, 185. [\[CrossRef\]](#)
22. Zhang, X.; Liu, H.; Li, X.; Zhang, Z.; Chen, Z.; Ren, D.; Zhang, S. Ecological and health risk assessments of heavy metals and their accumulation in a peanut-soil system. *Environ. Res.* **2024**, *252*, 118946. [\[CrossRef\]](#) [\[PubMed\]](#)
23. Guan, Q.; Wang, F.; Xu, C.; Pan, N.; Lin, J.; Zhao, R.; Luo, H. Source apportionment of heavy metals in agricultural soil based on PMF: A case study in Hexi Corridor, northwest China. *Chemosphere* **2018**, *193*, 189–197. [\[CrossRef\]](#) [\[PubMed\]](#)
24. Li, W.; Yang, B.; Huang, R.; Yang, J.; Cui, J.; Lu, X.; Xiong, J. Health risk assessment of potentially toxic elements in a Tibetan Plateau Wetland, China, using APCS-MLR, PMF, and Monte Carlo simulation. *J. Soils Sediments* **2025**, *25*, 3905–3925. [\[CrossRef\]](#)
25. Zhang, W.; Yan, Y.; Yu, R.; Hu, G. The sources-specific health risk assessment combined with APCS/MLR model for heavy metals in tea garden soils from south Fujian Province, China. *Catena* **2021**, *203*, 105306. [\[CrossRef\]](#)
26. Mu, T.; Cao, B.; Yang, M.; Gan, X.; Chen, L.; Wang, X.; Li, M.; Lu, Y.; Xu, J. Heavy metals distribution and source identification in contaminated agricultural soils: Isotopic and multi-model analysis. *Agronomy* **2025**, *15*, 812. [\[CrossRef\]](#)
27. Mi, Y.; Zhou, J.; Liu, M.; Liang, J.; Kou, L.; Xia, R.; Zhou, J. Machine learning method for predicting cadmium concentrations in rice near an active copper smelter based on chemical mass balance. *Chemosphere* **2023**, *319*, 138028. [\[CrossRef\]](#)
28. Ma, Y.; Sun, Y.; Wang, J.; Liu, Y.; Guo, M.; Hu, C.; Shui, B. Analysis of heavy metal sources and potential ecological risk assessment of mangroves in Aojiang Estuary. *Ecol. Indic.* **2025**, *173*, 113343. [\[CrossRef\]](#)
29. Wu, J.; Margenot, A.J.; Wei, X.; Fan, M.; Zhang, H.; Best, J.L.; Wu, P.; Chen, F.; Gao, C. Source apportionment of soil heavy metals in fluvial islands, Anhui section of the lower Yangtze River: Comparison of APCS–MLR and PMF. *J. Soils Sediments* **2020**, *20*, 3380–3393. [\[CrossRef\]](#)
30. Cao, Z.; Qian, H.; Gao, Y.; Li, K.; Liu, Y.; Shi, X.; Li, S.; Zhao, W.; Yang, S.; Tian, P.; et al. Ecological risk assessment and source identification of heavy metals in the sediments of the Danjiang River Basin: A quantitative method combining multivariate analysis and the APCS-MLR model. *Ecol. Indic.* **2025**, *175*, 113518. [\[CrossRef\]](#)
31. Guo, Y.; Yang, Y.; Li, Y. Comprehensive perspective on contamination identification, source apportionment, and ecological risk assessment of heavy metals in paddy soils of a tropical island. *Agronomy* **2024**, *14*, 1777. [\[CrossRef\]](#)
32. Zhang, H.; Shan, B. Historical distribution of DDT residues in pond sediments in an intensive agricultural watershed in the Yangtze-Huaihe region, China. *J. Soils Sediments* **2014**, *14*, 980–990. [\[CrossRef\]](#)
33. Liao, Q.; Evans, L.J.; Gu, X.; Fan, D.; Jin, Y.; Wang, H. A regional geochemical survey of soils in Jiangsu Province, China: Preliminary assessment of soil fertility and soil contamination. *Geoderma* **2007**, *142*, 18–28. [\[CrossRef\]](#)
34. Bao, S.D. *Soil and Agricultural Chemistry Analysis*, 3rd ed.; China Agriculture Press: Beijing, China, 2000. (In Chinese)
35. Thurston, G.D.; Spengler, J.D. A quantitative assessment of source contributions to inhalable particulate matter pollution in metropolitan Boston. *Atmos. Environ.* **1985**, *19*, 9–25. [\[CrossRef\]](#)
36. Wei, F.S.; Chen, J.S.; Wu, Y.Y.; Zheng, C.J. Study on the background contents on 61 elements of soils in China. *Environ. Sci.* **1991**, *12*, 12–19. (In Chinese with English abstract)

37. Ha, H.; Olson, J.R.; Bian, L.; Rogerson, P.A. Analysis of heavy metal sources in soil using kriging interpolation on principal components. *Environ. Sci. Technol.* **2014**, *48*, 4999–5007. [[CrossRef](#)]
38. Romano, E.; Bergamin, L.; Croudace, I.W.; Ausili, A.; Maggi, C.; Gabellini, M. Establishing geochemical background levels of selected trace elements in areas having geochemical anomalies: The case study of the Orbetello lagoon (Tuscany, Italy). *Environ. Pollut.* **2015**, *202*, 96–103. [[CrossRef](#)] [[PubMed](#)]
39. Sun, G.; Chen, Y.; Bi, X.; Yang, W.; Chen, X.; Zhang, B.; Cui, Y. Geochemical assessment of agricultural soil: A case study in Songnen-Plain (Northeastern China). *Catena* **2013**, *111*, 56–63. [[CrossRef](#)]
40. Yang, P.; Mao, R.; Shao, H.; Gao, Y. The spatial variability of heavy metal distribution in the suburban farmland of Taihang Piedmont Plain, China. *C. R. Biol.* **2009**, *332*, 558–566. [[CrossRef](#)]
41. Zhou, L.; Yang, B.; Xue, N.; Li, F.; Seip, H.M.; Cong, X.; Yan, Y.; Liu, B.; Han, B.; Li, H. Ecological risks and potential sources of heavy metals in agricultural soils from Huanghuai Plain, China. *Environ. Sci. Pollut. Res.* **2014**, *21*, 1360–1369. [[CrossRef](#)]
42. Li, C.; Li, F.; Wu, Z.; Cheng, J. Effects of landscape heterogeneity on the elevated trace metal concentrations in agricultural soils at multiple scales in the Pearl River Delta, South China. *Environ. Pollut.* **2015**, *206*, 264–274. [[CrossRef](#)]
43. Xu, L.H.; Cui, Z.W.; Wang, Y.; Wang, N.; Ma, J.P. Pollution assessment and source apportionment of heavy metals in farmland soil under different land use types: A case study of Dehui City, Northeastern China. *Agronomy* **2025**, *15*, 2899. [[CrossRef](#)]
44. Sheng, J.; Wang, X.; Gong, P.; Tian, L.; Yao, T. Heavy metals of the Tibetan top soils: Level, source, spatial distribution, temporal variation and risk assessment. *Environ. Sci. Pollut. Res.* **2012**, *19*, 3362–3370. [[CrossRef](#)] [[PubMed](#)]
45. Liu, Y.; Wang, H.; Li, X.; Li, J. Heavy metal contamination of agricultural soils in Taiyuan, China. *Pedosphere* **2015**, *25*, 901–909. [[CrossRef](#)]
46. Xu, L.; Gao, Z.; Jiang, J.; Sun, G. Heavy metal pollution and health risk assessment in black soil region of Inner Mongolia Province, China. *Agronomy* **2025**, *15*, 2717. [[CrossRef](#)]
47. Chen, X.; Chen, F.; Jia, S.; Chen, Y. Soil geochemical baseline and background in Yangtze-Huaihe River basin of Anhui Province. *Geol. China* **2012**, *39*, 302–310. [[CrossRef](#)]
48. GB15618-1995; Environmental Quality Standard for Soils. China Environmental Science Press: Beijing, China, 1995.
49. Zhao, L.; Xu, Y.; Hou, H.; Shangguan, Y.; Li, F. Source identification and health risk assessment of metals in urban soils around the Tanggu chemical industrial district, Tianjin, China. *Sci. Total Environ.* **2014**, *468*, 654–662. [[CrossRef](#)]
50. Esmaeili, A.; Moore, F.; Keshavarzi, B.; Jaafarzadeh, N.; Kermani, M. A geochemical survey of heavy metals in agricultural and background soils of the Isfahan industrial zone, Iran. *Catena* **2014**, *121*, 88–98. [[CrossRef](#)]
51. Martín, J.R.; Ramos-Miras, J.J.; Boluda, R.; Gil, C. Spatial relations of heavy metals in arable and greenhouse soils of a Mediterranean environment region (Spain). *Geoderma* **2013**, *200*, 180–188. [[CrossRef](#)]
52. Maanan, M.; Saddik, M.; Maanan, M.; Chaibi, M.; Assobhei, O.; Zourarah, B. Environmental and ecological risk assessment of heavy metals in sediments of Nador lagoon, Morocco. *Ecol. Indic.* **2015**, *48*, 616–626. [[CrossRef](#)]
53. Zhao, F.J.; Ma, Y.; Zhu, Y.G.; Tang, Z.; McGrath, S.P. Soil contamination in China: Current status and mitigation strategies. *Environ. Sci. Technol.* **2015**, *49*, 750–759. [[CrossRef](#)]
54. Lu, R.K.; Shi, Z.Y. Cadmium contents of rock phosphates and phosphate fertilizers of China and their effects on ecological environment. *Acta Pedol. Sin.* **1992**, *29*, 150–157. (In Chinese with English abstract)
55. Imseng, M.; Wiggenshauser, M.; Keller, A.; Müller, M.; Rehkämper, M.; Murphy, K.; Bigalke, M. Fate of Cd in agricultural soils: A stable isotope approach to anthropogenic impact, soil formation, and soil-plant cycling. *Environ. Sci. Technol.* **2018**, *52*, 1919–1928. [[CrossRef](#)] [[PubMed](#)]
56. Geng, J.; Yu, Y.; Tan, Q.; Li, G.; Fang, H.; Guo, Y.; Cheng, S. Disentangling natural and anthropogenic influences on the spatial variability of soil cadmium contamination. *Environ. Pollut.* **2025**, *384*, 126946. [[CrossRef](#)] [[PubMed](#)]
57. Chen, Y.; Li, X.; Han, Z.; Yang, S.; Wang, Y.; Yang, D. Chemical weathering intensity and element migration features of the Xiashu loess profile in Zhenjiang, Jiangsu Province. *J. Geogr. Sci.* **2008**, *18*, 341–352. [[CrossRef](#)]
58. Zacccone, C.; Di Caterina, R.; Rotunno, T.; Quinto, M. Soil–farming system–food–health: Effect of conventional and organic fertilizers on heavy metal (Cd, Cr, Cu, Ni, Pb, Zn) content in semolina samples. *Soil. Till. Res.* **2010**, *107*, 97–105. [[CrossRef](#)]
59. Xu, F.; Zhu, J.; Zhang, B.; Fu, Q.; Chen, J.; Hu, H.; Huang, Q. Sorption and immobilization of Cu and Pb in a red soil (Ultisol) after different long-term fertilizations. *Environ. Sci. Pollut. Res.* **2019**, *26*, 1716–1722. [[CrossRef](#)]
60. Gul, I.; Manzoor, M.; Kallerhoff, J.; Arshad, M. Enhanced phytoremediation of lead by soil applied organic and inorganic amendments: Pb phytoavailability, accumulation and metal recovery. *Chemosphere* **2020**, *258*, 127405. [[CrossRef](#)]
61. Adamiec, E.; Jarosz-Krzemińska, E.; Wieszała, R. Heavy metals from non-exhaust vehicle emissions in urban and motorway road dusts. *Environ. Monit. Assess.* **2016**, *188*, 369. [[CrossRef](#)]
62. Straffellini, G.; Ciudin, R.; Ciotti, A.; Gialanella, S. Present knowledge and perspectives on the role of copper in brake materials and related environmental issues: A critical assessment. *Environ. Pollut.* **2015**, *207*, 211–219. [[CrossRef](#)]

63. Praburaman, L.; Murugan, P.; Singaravel, V.; Moovendhan, M.; Kolandhasamy, P.; Seralathan, K.K. Heavy-metal contamination in agroecosystems from road transportation: A global threat to food safety and security. *Environ. Qual. Manag.* **2026**, *35*, e70330. [[CrossRef](#)]
64. Lv, J. Multivariate receptor models and robust geostatistics to estimate source apportionment of heavy metals in soils. *Environ. Pollut.* **2019**, *244*, 72–83. [[CrossRef](#)] [[PubMed](#)]
65. Gosar, M.; Šajn, R.; Teršič, T. Distribution pattern of mercury in the Slovenian soil: Geochemical mapping based on multiple geochemical datasets. *J. Geochem. Explor.* **2016**, *167*, 38–48. [[CrossRef](#)]

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