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Source Apportionment and Risk Assessment of Heavy Metals in Agricultural Soils in a Typical Mining and Smelting Industrial Area

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Abstract: Source apportionment and risk assessment are critical for making effective pollution prevention and control policies. The study was carried out to assess source-specific ecological and human health risks associated with heavy metals in farmland soils in Yingtan City based on apportionment results of receptor models. Multivariate analysis and the APCS-MLR model consistently revealed that As, Ni, and Cr in agricultural soils may be mainly derived from natural sources, while the contents of Cu, Zn, Cd, and Pb have been significantly elevated by human activities. According to the outputs of the APCS-MLR model, Cu (34.3%), Zn (67.2%), Pb (75.1%), and Cd (67.9%) primarily originated from the industrial activities related to mineral mining and non-ferrous metal smelting processes. The source-specific ecological risk assessment indicated that industrial sources were the primary contributor to the total ecological risks, posing moderate to high risks in the southern mountainous regions. Natural origins played a significant role in the health risks due to the substantial amounts of As naturally occurring in the soils. The findings could guide the development of effective risk management and pollution control measures for agricultural soils.

Keywords: heavy metals; source apportionment; agricultural soils; APCS-MLR; risk assessment

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1. Introduction

Contamination of agricultural soils with toxic metals (e.g., As, Cd, Pb, Cr, Ni, Cu, and Zn) is one of the most important environmental issues in China, and it has attracted worldwide attention from the public and governments [1–4]. Due to their high toxicity and persistence, heavy metals enriched in agricultural soils could result in the degeneration of soil and crop quality [5,6]. In addition, heavy metals could cause adverse impacts on human health through various pathways, including ingestion, inhalation, skin contact, and crop consumption [6,7]. For instance, chronic exposure to Cd in soils and foods was closely associated with lung cancer, kidney dysfunction, bone fracture, and hematuria [8,9]. Intake of As may result in diseases of the skin, bladder, lung and nerve system [7,9]. Therefore, exploring and controlling the degree of pollution and risk associated with toxic metals in farmland soils are essential to managing and controlling soil pollution.

To date, many researchers have assessed the environmental risks of heavy metals based on various methods, including pollution index (PI) [10], potential ecological risk index (RI) [11], and health risk models [12]. However, very few studies have quantitatively apportioned risks associated with heavy metals in agricultural soil to different sources [9,13,14]. Source apportionment is critical for identifying the primary risk contributors and developing efficient management measures [15–17]. However, source apportionment of heavy metals in soil remains challenging on a regional scale because heavy metals in farmland soils exhibit spatial heterogeneity and are generally contributed by both natural and anthropogenic sources [18,19]. To qualitatively identify the natural and anthropogenic origins,

multivariate analysis methods, such as correlation analysis, cluster analysis (CA), and principal component analysis (PCA), have been widely employed to group heavy metals with similar distribution patterns and qualitatively determine their potential origins [20]. However, these methods fail to find out the amounts of metals contributed by a specific source [21]. Receptor models are effective tools to quantitatively determine the contributions of heavy metals from different sources, including absolute principal component score/multiple linear regression (APCS-MLR) [22], positive matrix factorization (PMF) [23], and Unmix [24]. Among them, APCS-MLR has been extensively applied for source apportionment of air pollutants [25] and was implemented in the field of agricultural soil pollution in recent studies [26] due to its simple manipulation and fast computation [27]. Thus, the source-specific ecological and health risks were expected to be quantified by combining the APCS-MLR model outputs with risk assessments.

Yingtan City is situated in the northeast of Jiangxi Province in southern China (27°35′–28°41′ N, 116°41′–117°30′ E), and the mineral resources in the area are abundant, including gold, silver, copper, lead, zinc, iron, and gypsum. The mining and non-ferrous smelting activities have promoted rapid economic development in Yingtan for recent decades, which is currently well known as the "Copper Capital of China" [8]. However, intensive industrial activities have emitted a large quantity of heavy metals into the farmland soils, eventually posing severe threats to the environment and local residents [28]. Using the surface farmland soil samples collected in Yingtan, the study was carried out to 1) assess the risks associated with heavy metals in surface soil using the potential ecological risk index (PERI); 2) identify the major sources of heavy metals in surface farmland soils and reveal the spatial distributions of source contributions; 3) evaluate the contributions of ecological and human health risks from the different sources. The study aims to provide a scientific foundation for developing appropriate pollution control and risk management policies for agricultural soils in the region.

2. Materials and Methods

2.1. Sample Collection, Preparation, and Chemical Analysis

In the study, 241 surface soil samples covering the agricultural fields throughout the whole region of Yingtan (Figure 1) were collected for analysis. The sampling points are mainly determined according to the distribution of farmland and villages. The number of samples was set based on the village size, with one sample for small villages and additional sampling points for larger villages as appropriate. In addition, the sampling points were at least 500 m away from the main roads to avoid the direct influence of traffic exhaust. For each soil sample, approximately 10 sub-samples, which had roughly the same weights and contained the same amount of soil from all depths (0–20 cm), were randomly collected across an area of approximately 100 m^2 surrounding the pre-determined site. Thus, the sub-samples were mixed thoroughly in a clean container to obtain a representative soil sample (~1.0 kg). Finally, the soil samples were stored in clean polyethylene bags after removing the large debris, such as rocks and plants, and sent back to the laboratory for further preparation and chemical analysis within a week.

After the air-drying and the removal of debris, the soil samples were finely grounded by agate mortars and were sifted through a 100-mesh nylon sieve for further chemical analyses. The heavy metals (As, Cd, Cr, Cu, Mn, Ni, Pb, and Zn) were measured using a NexION 350D Inductively Coupled Plasma–Mass Spectrometer (ICP-MS, PerkinElmer, USA) after the microwave-assisted digestion procedures [29]. The details have been described in our previous research [30] and were given in the Supporting Information (Text S1). To determine the Hg contents in soils, the weighted powdered samples (~0.15 g) were placed in quartz boats and analyzed by a DMA-80 Direct Mercury Analyzer (Milestone, ITA). For quality control and assurance, SRM-2976 (the National Institute of Standards and Technology of the United States) was used as reference materials, and blank controls and duplicate samples (20%) were randomly inserted into the sequence. The limit of detection and limit of quantification (LOD/LOQ) of Hg were 0.13 and 0.43 ng/g, respectively, which

were calculated according to the IUPAC standard [31]. Both the recovery rates (91–106%) and standard deviation (<10%) confirmed that the results were reliable.

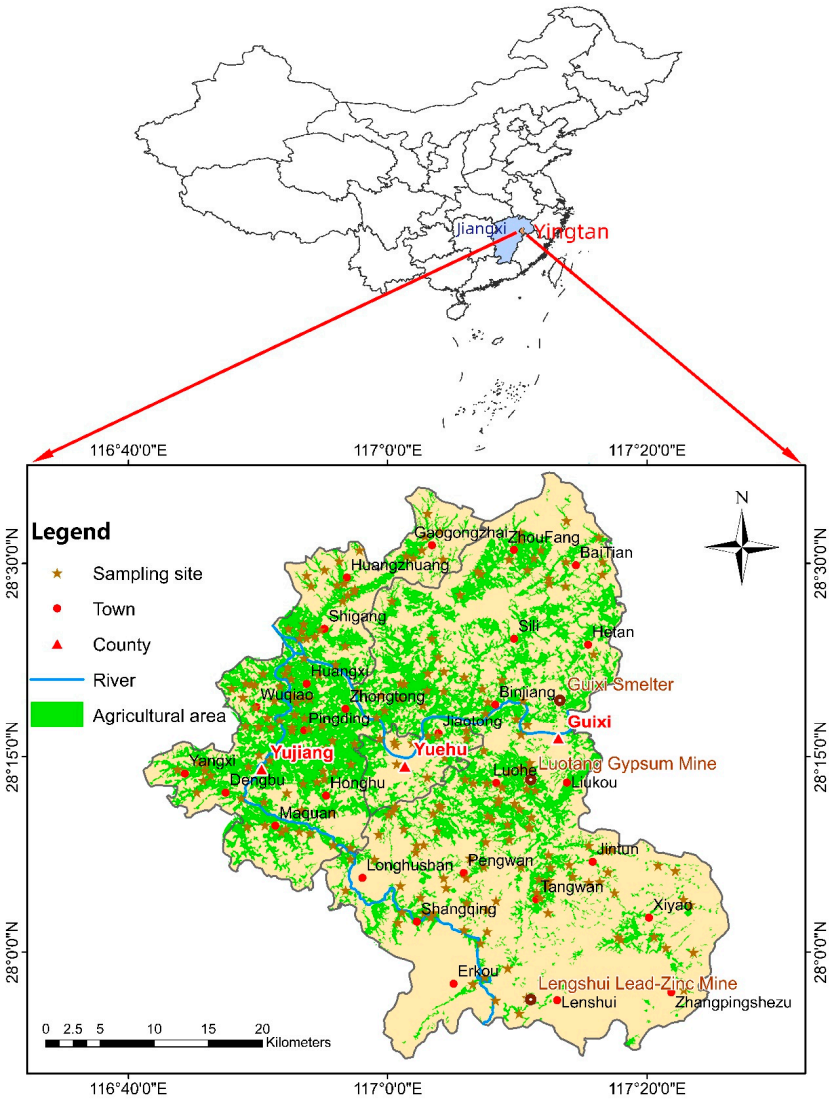

Figure 1. The map of the study area with the locations of sampling sites (n = 241).

2.2. Ecological Risk Assessment

The ecological risk index (RI) was used to assess the potential ecological risk that resulted from the heavy metals in soils [32], which could be calculated as follows:

$$E_i = T_i \frac{C_i}{B_i} \tag{1}$$

$$RI = \sum_{i=1}^{n} E_i \tag{2}$$

where E_i, T_i, and B_i are the single risk factor, the toxic response factor, and soil background value for heavy metals i, respectively. The values of the toxic response factor for Cd, Ni, Cu, Zn, As, Cd, Pb, Mn, and Hg are 2, 5, 5, 1, 10, 30, 5, 1, and 40, respectively [33]. The potential ecological risks of heavy metals in soils are divided into five levels according to the values of E_i and RI (Tables S1 and S2).

2.3. Source Identification and Apportionment

2.3.1. Multivariate Analysis Methods

In this study, multiple multivariate analysis techniques were applied to qualitatively determine the sources of pollutants. A correlation matrix can directly identify the associations between different species. Heavy metals with strong correlations may originate from the same pollution sources. As the most commonly used multivariate statistical methods, PCA and CA have been widely used in environmental research for studying the hidden grouping patterns among soil heavy metals [13]. PCA can obtain a cluster of orthogonal factors with a decreased dimension and show their correlations to the original variables [34–36]. CA results are usually presented as a dendrogram, where elements within a group (or cluster) have a higher similarity than others and may originate from the same sources [37].

2.3.2. APCS-MLR Receptor Model

Based on the factors derived from PCA, the receptor model APCS-MLR was used to quantitatively determine contributions from the identified sources to the receptors [12,27,38]. First, the raw data were normalized as dimensionless standardized forms (Equation (3)). Then, an artificial sample with zero concentrations for all species was introduced to convert the normalized factor scores into un-normalized APCS values (Equations (4)–(6)). The basic idea is that the APCS value $(APCS)_{ki}$ can be calculated by subtracting the absolute zero factor score $(A_0)_k$ from the obtained normalized factor score (A_{ki}). The multiple linear regression, using the heavy metals contents as the dependent variable and $(APCS)_{ki}$ as the independent variable, was performed to estimate the contributions of the factors (Equation (7)). The formulas can be written as the following:

$$Z_{ij} = \frac{C_{ij} - \overline{C}_j}{\sigma_j} \tag{3}$$

$$(Z_0)_j = \frac{0 - \overline{C}_j}{\sigma_j} = -\frac{\overline{C}_j}{\sigma_j} \tag{4}$$

$$(APCS)_{ki} = A_{ki} - (A_0)_k \tag{5}$$

$$(A_0)_k = \sum_{j=1}^{J} S_{kj} \cdot (Z_0)_j \tag{6}$$

$$C_{ij} = \sum_{k=1}^{n} \left(b_{kj} \times APCS_{ki} \right) + (b_0)_j \tag{7}$$

where the subscripts i, j, and k stand for sample i, species j, and obtained factor k, respectively. S_{kj} is the score coefficient of factor k to species j, while Z_{ij}, $(Z_0)_j$ presents standardized concentration of species j in the true and artificial sample, respectively . $\overline{C}_j$ and σ_j are the average content and standard deviation of species j for all samples used in this analysis, respectively. C_{ij} is the concentration of heavy metal j in sample i, b_{kj} is the estimated slope coefficient of source k on heavy metal j, and $(b_0)_j$ is a constant term indicating average contribution of unknown sources. Thus, the contribution from the source k to the heavy metals j in sample i can be quantified as $\left(b_{kj} \times APCS_{ki} \right)$.

APCS-MLR may produce some negative contributions and unknown sources, making it difficult to interpret [22,39]. Thus, the absolute function was utilized to account for the contribution rate (PC_{kj}) of estimated (Equation (8)) and unrecognized origins (Equation (9)) as follows [14]:

$$PC_{kj} = \frac{\left| b_{kj} \times APCS_{ki} \right|}{\left| (b_0)_j \right| + \left| b_{kj} \times APCS_{ki} \right|} \times 100\% \tag{8}$$

$$PC_{kj} = \frac{\left|(b_0)_j\right|}{\left|(b_0)_j\right| + \left|b_{kj} \times APCS_{ki}\right|} \times 100\% \quad (9)$$

In the study, SPSS 26.0 was used to achieve basic statistics information and perform correlation analysis, PCA, CA, and multiple linear regressions. Arc GIS 10.6 was implemented to map the spatial distributions of the factor contributions.

2.4. Source-Specific Risk Assessment

2.4.1. Ecological Risk Apportioned from Different Sources

The source-oriented ecological risk of heavy metals in agricultural soils was quantified by coupling the source apportionment with ecological risk assessment [9,40]. It could be estimated as follows (Equations (10) and (11)):

$$E_{ij}^k = \frac{C_{ij}^k}{B_i} \times T_j \quad (10)$$

$$RI_{ik} = \sum_{j=1}^{n} E_{ij}^k \quad (11)$$

where E_{ij}^k and C_{ij}^k are the ecological risk and concentration of the heavy metals j from the source k in the i sample, respectively. RI_{ik} is the potential ecological risk posed by source k in the i sample.

2.4.2. Health Risk Apportioned from Different Sources

Based on the results of source apportionment, the source-specific health risks for children and adult through three exposure pathways (oral ingestion, dermal contact, and inhalation) were evaluated using health risk assessment method suggested by USEPA [9,41]. The average daily doses (ADD_{ij}^k, mg/kg/day) of heavy metal i from the source k in the j sample via different pathways were calculated as follows [42]:

$$ADD_{ijing}^k = \frac{C_{ij}^k \times IngR \times EF \times ED}{BW \times AT} \times CF \quad (12)$$

$$ADD_{ijdermal}^k = \frac{C_{ij}^k \times SAF \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF \quad (13)$$

$$ADD_{ijinh}^k = \frac{C_{ij}^k \times InhR \times EF \times ED}{BW \times AT} \quad (14)$$

where *ing*, *dermal*, and *inh* represent oral ingestion, dermal contact, and inhalation, respectively. The exposure parameters used in the health risk assessment are defined and listed in Table S3.

For non-carcinogenic and carcinogenic risk, the total hazard quotient (THQ_{ij}^k) and total carcinogenic risk (TCR_{ij}^k) of heavy metal j from the source k in the sample i were calculated as follows:

$$THQ_{ij}^k = \frac{ADD_{ijing}^k}{RfD_{ing}} + \frac{ADD_{ijinh}^k}{RfD_{inh}} + \frac{ADD_{ijider}^k}{RfD_{der}} \quad (15)$$

$$TCR_{ij}^k = ADD_{ijing}^k \times SF_{ing} + ADD_{ijinh}^k \times SF_{inh} + ADD_{ijdermal}^k \times SF_{der} \quad (16)$$

where *RfD* (mg/kg/day) is the reference dose of targeted heavy metal for non-carcinogenic risk, while *SF* (kg·day/mg) is the slope factor of carcinogenic heavy metal. Their values for the heavy metals are shown in Table S4.

No adverse health effects occur if $THI \leq 1$, while there may be potential non-carcinogenic effects when $THI > 1$. Additionally, *TCR* ranging from 1×10^{-6} to 1×10^{-4} is considered to

bean acceptable level, *TCR* less than 1×10^{-6} suggests negligible carcinogenic risk, while *TCR* larger than 1×10^{-4} indicates severe carcinogenic risk [13].

3. Results and Discussion

3.1. Heavy Metals in Agricultural Soil

The total contents of heavy metals in the agricultural soils of Yingtan, together with their soil background values and the risk screening values in agricultural soils, are summarized in Table 1. The ranges of heavy metals in farmland soils were: 5.80–261 mg/kg, 3.59–84.1 mg/kg, 8.00–304 mg/kg, 15.3–278 mg/kg, 1.38–133 mg/kg, 0.0725–4.04 mg/kg, 9.02–208 mg/kg, 26.3–328 mg/kg, and 0.00468–0.455 mg/kg, respectively, for Cr, Ni, Cu, Zn, As, Cd, Pb, Mn, and Hg. Compared with their background values of soil in Jiangxi province [43], the mean and median values of As and Mn were lower, suggesting that they may mainly originate from the weathering of parent materials. In contrast, the higher median and mean values of Cu, Zn, Cd, and Pb than their corresponding background values indicated the great contribution of anthropogenic activities to their contents in soil. In particular, the median and mean values of Cd were three and six times of the background value, respectively, suggesting that Cd in Yingtan farmland soils had been significantly elevated by anthropogenic emissions. Cr, Ni, and Hg had their medians lower and means higher than the corresponding background values, demonstrating that both the natural and anthropogenic sources may greatly contribute to their contents in soils. Moreover, regarding the risk screening values of soil as reference levels, the exceedance rate followed the order of Cd > Cu > Pb > As > Zn > Ni~Cr > Hg. It was noted that the concentrations of Cd exceeded its risk screening value in 63.9% of the agricultural soil samples, confirming that Cd was the major pollutant in the agricultural soils of Yingtan. In summary, the investigated heavy metals were impacted by human activities in the following decreasing order: Cd > Cu, Zn and Pb > Cr, Ni, and Hg > Mn and As.

Table 1. Basic statistical information of heavy metal contents in agricultural soils of Yingtan.

Metal	Concentration (mg/kg)									B.V. [a] (mg/kg)	% of Samples > B.V.	S.V. [b] (mg/kg)			% of Samples > S.V.
	Mean	S.D.	Min	P5	Q1	Median	Q3	P95	Max			pH ≤ 5.5	5.5 < pH ≤ 6.5	6.5 < pH ≤ 7.5	
Cr	56.4	45.4	5.80	12.0	22.2	37.4	77.8	155	261	45.9	43.6	250	250	300	0.41
Ni	21	10.2	3.59	8.47	13.5	18.6	27.3	40.8	84.1	18.9	48.5	60	70	100	0.41
Cu	30.7	23.0	8.00	13.6	20.6	26.7	34.9	50.8	304	20.3	75.9	50	100	100	6.22
Zn	87.2	34.6	15.3	34.6	63.4	86.4	105	138	278	69.4	71.4	200	200	250	1.24
As	9.41	10.8	1.38	2.46	4.90	7.35	11.0	19.2	133	14.9	10.4	30	30	25	1.66
Cd	0.386	0.361	0.0725	0.164	0.260	0.353	0.430	0.667	4.04	0.108	98.3	0.3	0.4	0.6	63.9
Pb	47.3	20.1	9.20	21.0	34.5	44.4	58.2	77.9	208	29.1	85.1	80	100	140	3.22
Mn	186	137	26.3	50.2	91.1	152	238	412	1120	328	10.4	-	-	-	-
Hg	0.0850	0.0499	0.00468	0.0247	0.0547	0.0758	0.109	0.174	0.455	0.084	39	0.5	0.5	0.6	0

Notes: a: B.V. is the soil background value of Jiangxi Province; Percentage (>B.V.) is the percentage of samples exceeding the background value; b: S.V. is the risk screening value for the investigated heavy metals in agricultural soils in China [44]; Percentage (>S.V.) is the percentage of samples exceeding the risk screening value.

Figure 2a displays the contributions of individual heavy metals to the total potential ecological risks at the county level. It suggests that Cd (57.2%) accounted for the majority of potential ecological risk in Yingtan farmland soils due to its high contents and the large toxic response factor values, followed by Hg (24.6%) and the combination of all other investigated heavy metals (As, Cr, Cu, Mn, Ni, Pb, and Zn) contributed less than 20% of the total potential ecological risk. According to Table S2, 0.8%, 6.2%, 61.8%, and 28.2% of soil samples could be recognized as "very high", "high", "considerable", and "moderate" categories owing to the ecological risk posed by Cd, respectively (Figure 2b), whereas 0.4%, 5.0%, and 33.6% (81 out 241) of the samples belonged to the "high", "considerable", and "moderate" categories due to Hg, respectively (Figure 2b). Figure 2c displays the spatial distribution of potential ecological risks in Yingtan, as well as the per capita industrial production value in 2017 for each district and the locations of major industrial sources. The values of the potential ecological risk index ranged from 57.7 to 1222, with a mean value of 180. The highest levels were observed in areas close to Lenshui and Guixi, which were featured with

concentrated mining and smelting activities. Figure S2 depicts the distribution of potential ecological risks resulting from various heavy metals in the surface soils of the study area. The distribution of the overall potential ecological risk was similar to that of Cd. The results consistently suggested that Cd was the priority contaminant and the major risk contributor to the potential ecological risks in Yingtan farmland soils. It is important to identify the sources of Cd for pollution prevention and soil quality management.

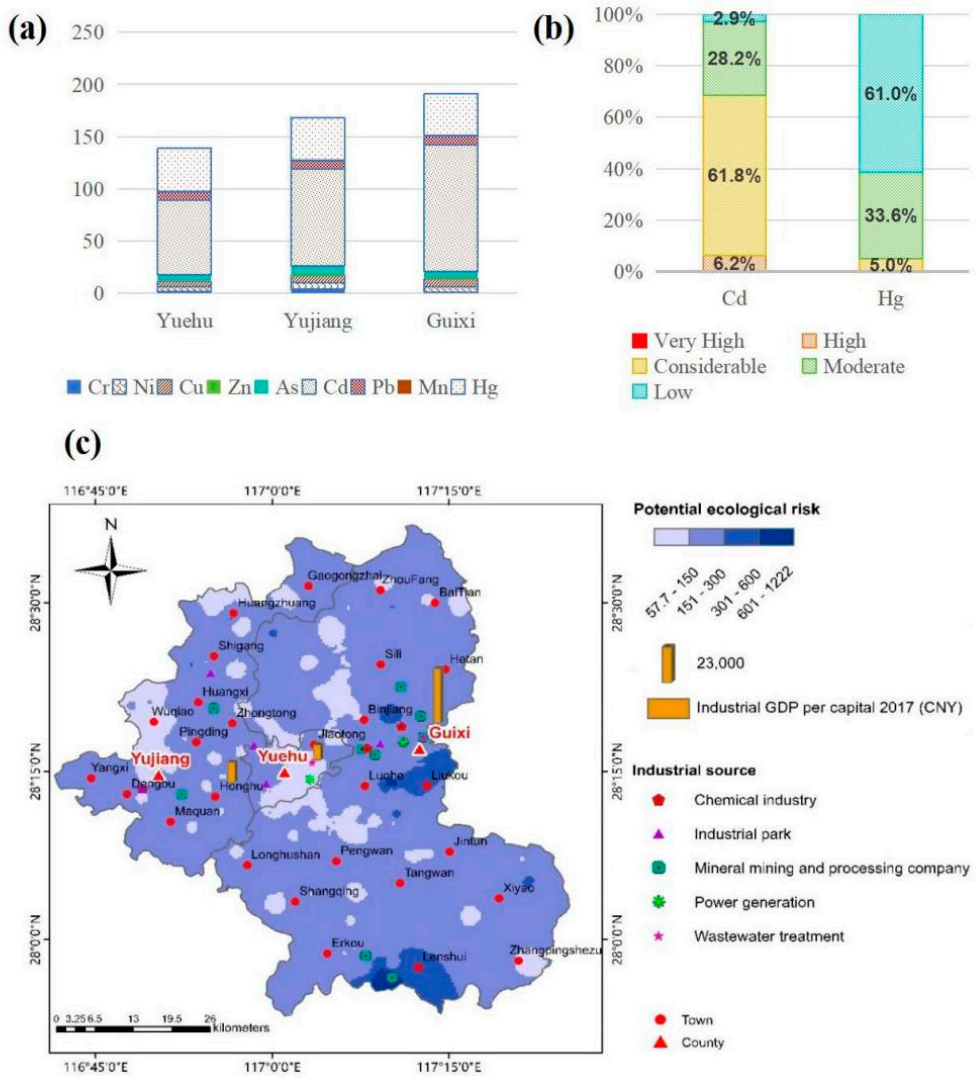

Figure 2. Potential ecological risk resulted from investigated heavy metals in Yingtan farmland surface soils: (**a**) Single and total potential ecological risks resulted from investigated heavy metals in different counties; (**b**) The levels of potential ecological risks caused by Cd and Hg, respectively. The numeric tags are hidden for fractions less than 1%; and (**c**) The spatial distributions of the overall potential ecological risk and major industrial facilities, as well as industrial GDP per capita in different counties.

3.2. Source Identification and Apportionment

3.2.1. Source Identification and Apportionment

Table 2 shows the Pearson's correlation coefficients of the investigated heavy metals in farmland soils. The results indicated that the correlation coefficients between Cr and Ni (0.558), Cu and Zn (0.502), Zn and Pb (0.696), and Cd and Pb (0.507) were relatively high, while those between Mn (or Hg) and other heavy metals were low. The results demonstrated that Cu, Zn, Pb, and Cd could be contributed by the same or correlated sources, and Ni and Cr had similar provenances. In contrast, Mn and Hg had different origins from other heavy

metals. In addition, hierarchical cluster analysis was also performed to identify groups of heavy metals with similar distribution patterns, as shown in Figure 3. It reveals that Cr and Ni formed a cluster, and the cluster of Zn and Pb linked with Cd and Cu at later stages, while Mn and Hg were lack of connections with the other soil heavy metals. PCA was also conducted to identify the source of soil heavy metals, and the outcomes of KMO (0.622) and Bartlett's test ($p < 0.001$) demonstrated that the dataset was appropriate for PCA [45,46]. As shown in Table S5, five principal components (PCs) with a cumulative proportion of 79.9% were kept for further analysis. To make the results more explainable, VARIMAX normalized rotation was performed. Table 3 summarizes the factor loadings of heavy metals at the rotated principal components. In general, factor loadings greater than 0.71 are considered outstanding, while those less than 0.32 are very poor [47]. As is shown in Table S5, Cu (0.418), Zn (0.842), Pb (0.846), and Cd (0.747) had high factor loadings in PC1, indicating that PC1 can explain most of their variances. High factor loading values of Ni (0.868) and Cr (0.842) were found in PC2. PC3 was dominated by As (0.924), followed by Cu (0.495). Mn (0.885) and Hg (0.939) had extremely high factor loadings in PC4 and PC5, respectively. The outputs of PCA were consistent with those of cluster analysis.

Table 2. Pearson's correlation matrix of investigated heavy metals in agricultural soils of Yingtan.

Metal	Cr	Ni	Cu	Zn	As	Cd	Pb	Mn	Hg
Cr	1								
Ni	0.558 **	1							
Cu	0.141 *	0.258 *	1						
Zn	0.170 *	0.443 *	0.502 **	1					
As	0.208 *	0.148 *	0.247 *	0.131 *	1				
Cd	−0.0787	0.0623	0.182 *	0.437 *	0.113	1			
Pb	0.00576	0.166 *	0.167 *	0.696 **	0.0963	0.507 **	1		
Mn	0.184 *	0.00994	0.0603	0.128 *	0.0101	0.120	0.211 *	1	
Hg	0.0395	0.152 *	0.108	0.190 *	0.147 *	0.0552	0.209 *	−0.0508	1

Notes: ** $p < 0.01$; * $p < 0.05$.

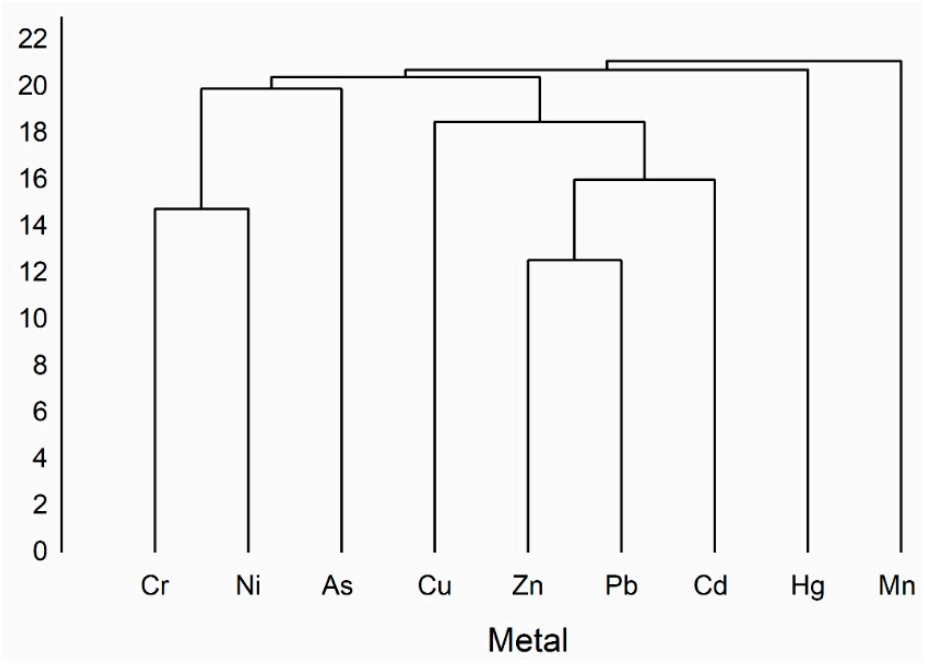

Figure 3. Results of clustering analysis: Hierarchical dendrogram of investigated heavy metals.

Table 3. Results of PCA: The rotated component matrix of heavy metals in Yingtan agricultural soil.

Metal	PC1	PC2	PC3	PC4	PC5
Cr	−0.125	0.842	0.16	0.24	−0.016
Ni	0.178	0.868	0.005	−0.136	0.071
Cu	0.418	0.256	0.495	−0.383	−0.221
Zn	0.842	0.349	0.072	−0.111	0.03
As	0.013	0.065	0.924	0.078	0.147
Cd	0.747	−0.172	0.108	0.1	−0.04
Pb	0.846	0.016	−0.053	0.179	0.201
Mn	0.171	0.083	0.037	0.885	−0.126
Hg	0.108	0.05	0.099	−0.107	0.939

3.2.2. Quantitative Source Apportionment Using APCS-MLR

In this study, APCS-MLR was applied to quantitatively apportion the contributions of heavy metals in soils from various potential sources. Table 4 presents that R^2 of investigated heavy metals varied from 0.612 to 0.916, and the differences between the observed and predicted mean values of heavy metals were minor. It suggested that the APCS-MLR models were reliable for source interpretation [13]. Based on the receptor models simulation, the spatial distribution of APCS is depicted in Figure 4.

Table 4. Results of APCS/MLR: Contributions (%) from each factor to heavy metal contents in Yingtan farmland soils.

Metal	R^2	Contribution (%)					
		Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Unknown
Cr	0.808	9.63	66.6	4.24	9.00	0.863	9.63
Ni	0.809	16.6	68.0	0.136	4.91	4.38	5.96
Cu	0.682	34.3	17.7	13.3	11.6	10.9	12.1
Zn	0.850	67.2	22.8	1.66	3.57	1.58	3.27
As	0.886	2.54	10.8	57.4	6.26	18.3	4.63
Cd	0.612	75.1	1.13	1.33	6.66	11.8	4.02
Pb	0.791	67.9	10.9	2.89	3.54	2.13	12.7
Mn	0.836	18.9	7.67	1.18	49.1	8.60	14.5
Hg	0.916	10.8	5.40	3.23	4.49	71.0	5.10

Notes: R^2 is the coefficient of determination.

Factor 1 had higher contributions to Cu (34.3%), Zn (67.2%), Pb (75.1%), and Cd (67.9%). In general, the elevated content of Pb, Zn, Cu, and Cd in soils would be contributed by industrial activities, such as mining and steel processing, battery production, waste incineration, and coal combustion [19,30,48,49]. According to Figures 4a and S1, the high values of Factor 1 were mainly observed in the southern mountainous region and Guixi County, which featured intensive mining and smelting–processing activities, respectively. In contrast, the central basin with a high density of road network, together with the western part of the central basin with heavy agricultural production activities, had relatively lower levels of contributions from the factor. Therefore, factor 1 could mainly consist of industrial sources associated with mining and smelting processes.

Factor 2 was heavily loaded with Ni (68.0%) and Cr (66.6%) in agricultural soils. Previous studies have recognized that the natural processes, including rock erosion, weathering, and the degradation and mineralization of sediments in wetlands, are the main drivers for their accumulation [50]. In this study, the high contributions of factor 2 were mainly distributed in Yujiang County (Figure 4b), where the industrial system was featured with carving and glass production [51], which hardly released substantial Cr and Ni into soils. Therefore, Cr and Ni mainly originated from the lithogenic process. The results were consistent with the findings in other study areas in China and the world [52–55]. Thus, factor 2 was mainly attributed to a natural source.

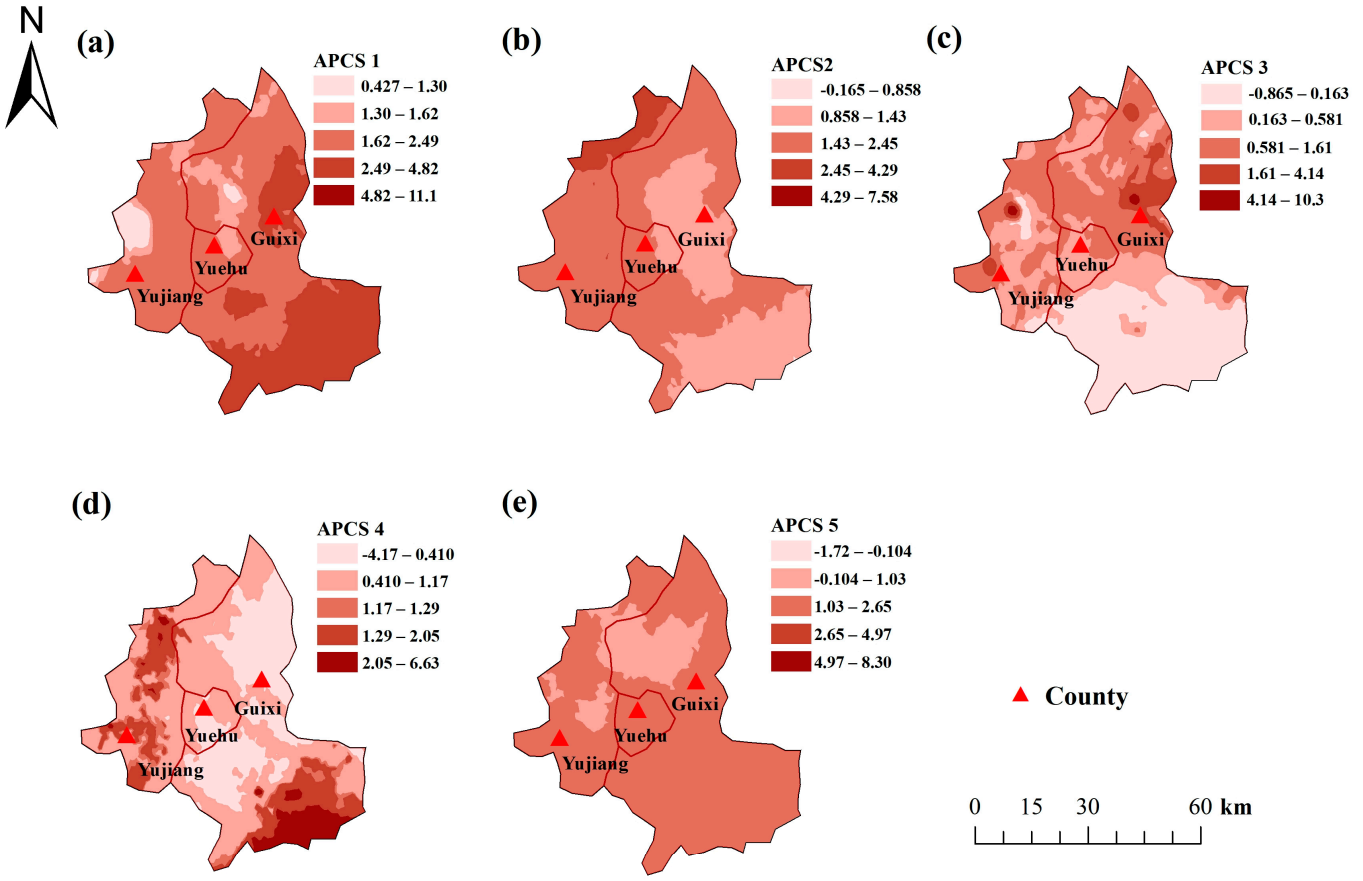

Figure 4. Spatial distributions of APCS: (**a**) Source 1; (**b**) Source 2; (**c**) Source 3; (**d**) Source 4; (**e**) Source 5.

Factor 3 and factor 4 had a main loading of As (57.4%) and Mn (49.1%), respectively. Generally speaking, Mn in soil mainly comes from geological origin, and human activities would not cause significant changes in the content of Mn in the surface soils [56,57], whereas mining activities, coal burning, wood preservative usage, elements smelting, and agrochemical application are major anthropogenic sources contributing to As contamination in soils [58,59]. According to Figures 4c,d, and S1, Guixi and the southern region with intensive mining and smelting activities have relatively low levels observed, indicating that industrial emission would not account for the main component of the factor. In addition, Table 1 shows that Mn and As had lower concentrations than the background value in most samples, indicating that they had little likelihood of being contaminated by anthropogenic sources. Therefore, factors 3–4 could be attributed to natural sources.

The weight of Hg (71.0%) in factor 5 was extremely high. Studies have shown that the concentration of Hg may be significantly elevated by industrial emissions such as non-ferrous metal smelting, coal mining and coal combustion, waste landfill, paper-making, and chemical production [60–62]. Table 1 indicates that Hg concentrations in more than 60% of the samples were below the corresponding background value in the study area, suggesting the important role of natural sources. High contributions from factor 5, as well as the samples with Hg concentrations exceeding 0.1 mg/kg, were frequently found in the areas concentrated with mining and smelting activities, including the mining area located in the southern and northern region, as well as the eastern part of the central basin (Figures 4e, S1 and S3), demonstrating the significant impacts of anthropogenic inputs to the elevated contents of Hg in soils. Thus, factor 5 would be a mixed source of natural origin and industrial emissions.

According to the source identification, the heavy metals in soil were contributed by natural (factor 2–4), industrial (factor 1), and mixed sources (factor 5), which accounted for

34.0%, 49.5%, and 6.73% of total metal contents, respectively (Figure S4). The contribution of unknown sources occupied 9.71%, which may include agricultural sources and traffic sources. The results revealed that the industrial origin exhibited a significant impact on soil quality due to high contribution to the toxic elements input (e.g., Cu, Zn, Pb, and Cd).

3.3. Source-Specific Risk Apportionment

Figure 5 presents the ecological risk contributed by the different sources based on the receptor model outputs. The risk contribution of the different sources followed the sequence: factor 1 (49.1%) > factor 5 (34.3%) > factor 2 (21.7%) > factor 3 (9.29%) > factor 4 (8.25%). Industrial activities (factor 1) posed considerable ecological risks (88.1) due to the high contribution to Cd, whereas the ecological risk caused by mixed sources (factor 5) and natural origins (factors 2–4) were classified as "low" categories (Table S1). It suggested that the industrial activities contributed a remarkable proportion to the entire ecological risk due to high toxicity response coefficients of heavy metals (e.g., Cd and Pb) released from the industrial emission [9,14,63]. Figure S5 shows that the spatial distribution of *RI* resulting from the industrial source was not uniform. Moderate to high levels of risk were observed in the southern mountainous region, which featured intensive mining activities. *RI* posed by factors 2–5 was classified as low risk for the whole study area. The above results consistently revealed that the industrial sources could cause significant harm to the soil environment. Therefore, priority must be given to the management of mining and smelting activities.

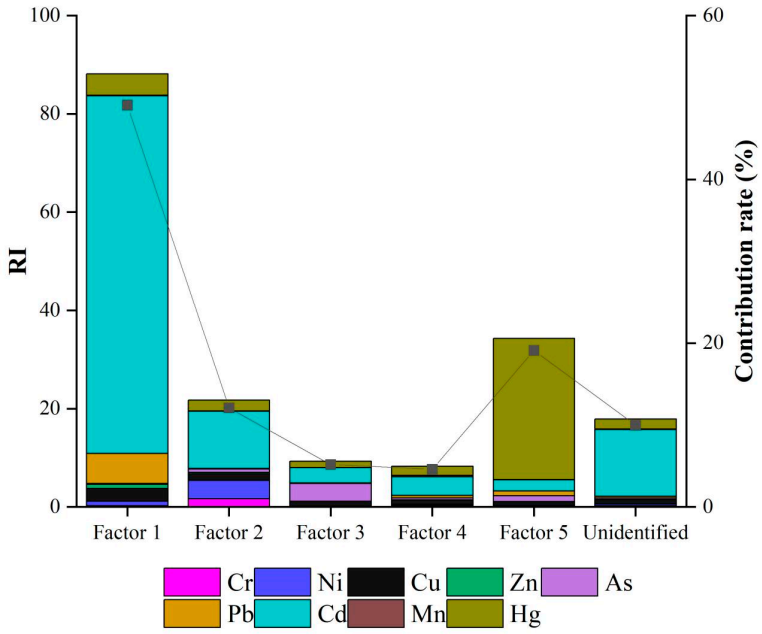

Figure 5. Potential ecological risks and contribution rates from the different sources and heavy metals.

The source-specific health risks for adults and children through three pathways (ingestion, dermal contact, and inhalation), including non-carcinogenic and carcinogenic risks, are summarized in Tables 5 and S6, respectively. For non-carcinogenic risk, *THQ* values for adults and children were 0.101 and 0.409, respectively, suggesting the absence of non-carcinogenic risks. In contrast, the total *TCR* value for adults (1.36×10^{-4}) and children (6.86×10^{-4}) surpassed the acceptable levels of 1.00×10^{-4}, which indicated that significant carcinogenic effects may occur. Compared to the adults, the children suffered more severe health risks, which could be partially attributed to their hand-to-mouth behavior, higher inhalation rates per body weight, and more gastrointestinal absorption of some toxic metals [7,9,64]. *TCR* values were presented in the order of As > Pb > Cd. It was noted that *TCR* values of As for adults and children were both beyond 1.00×10^{-4}, suggesting that As in soil could pose significant health risks to local residents. This may be

explained by the high toxicity response levels and parent material origin of As in soil [7,12]. In addition, Table 5 shows that ingestion was identified as a major exposure pathway for carcinogenic risks, while TCR_{dermal} and TCR_{inh} values for adults and children were much lower than 1.00×10^{-4}. Therefore, the living habits of children in the study area should be considered to reduce exposed health risks.

Table 5. Source-specific carcinogenic risks of heavy metals from different sources.

Item	Source	As	Pb	Cd	TCR_{ing}	TCR_{inh}	TCR_{dermal}	TCR
Adult	Factor 1	3.45×10^{-6}	1.57×10^{-7}	1.01×10^{-10}	3.55×10^{-6}	1.24×10^{-10}	5.46×10^{-8}	3.60×10^{-6}
	Factor 2	1.47×10^{-5}	2.38×10^{-9}	1.63×10^{-11}	1.45×10^{-5}	1.11×10^{-10}	2.33×10^{-7}	1.47×10^{-5}
	Factor 3	7.79×10^{-5}	2.78×10^{-9}	4.32×10^{-12}	7.67×10^{-5}	5.05×10^{-10}	1.23×10^{-6}	7.79×10^{-5}
	Factor 4	8.50×10^{-6}	1.40×10^{-8}	5.29×10^{-12}	8.38×10^{-6}	5.99×10^{-11}	1.35×10^{-7}	8.51×10^{-6}
	Factor 5	2.49×10^{-5}	2.47×10^{-8}	3.18×10^{-12}	2.45×10^{-5}	1.63×10^{-10}	3.95×10^{-7}	2.49×10^{-5}
	Unknown	6.29×10^{-6}	8.44×10^{-9}	1.90×10^{-11}	6.19×10^{-6}	5.94×10^{-11}	9.96×10^{-8}	6.29×10^{-6}
	Total	1.36×10^{-4}	2.09×10^{-7}	1.49×10^{-10}	1.34×10^{-4}	1.02×10^{-9}	2.15×10^{-6}	1.36×10^{-4}
Children	Factor 1	1.74×10^{-5}	7.98×10^{-7}	1.22×10^{-10}	1.80×10^{-5}	1.49×10^{-10}	1.94×10^{-7}	1.82×10^{-5}
	Factor 2	7.41×10^{-5}	1.21×10^{-8}	1.96×10^{-11}	7.33×10^{-5}	1.33×10^{-10}	8.29×10^{-7}	7.42×10^{-5}
	Factor 3	3.93×10^{-4}	1.41×10^{-8}	5.21×10^{-12}	3.89×10^{-4}	6.08×10^{-10}	4.39×10^{-6}	3.93×10^{-4}
	Factor 4	4.29×10^{-5}	7.08×10^{-8}	6.36×10^{-12}	4.25×10^{-5}	7.21×10^{-11}	4.79×10^{-7}	4.29×10^{-5}
	Factor 5	1.26×10^{-4}	1.25×10^{-7}	3.83×10^{-12}	1.24×10^{-4}	1.97×10^{-10}	1.40×10^{-6}	1.26×10^{-4}
	Unknown	3.17×10^{-5}	4.28×10^{-8}	2.28×10^{-11}	3.14×10^{-5}	7.15×10^{-11}	3.54×10^{-7}	3.18×10^{-5}
	Total	6.85×10^{-4}	1.06×10^{-6}	1.80×10^{-10}	6.78×10^{-4}	1.23×10^{-9}	7.65×10^{-6}	6.86×10^{-4}

Due to the insignificant non-carcinogenic effects for local residents, only contributions of various sources to the carcinogenic risks for adults and children are discussed below. Factor 3 had the highest proportion (adult: 57.3%, children: 57.3%) of *TCR* values, owing to the large proportion of As in the factor. Although the industrial sources (factor 1) made the largest contribution to the total ecological risk, their contribution to the total *TCR* values was very low (adult: 2.65%, children: 2.65%). The results were comparable with the previous studies [7,63]. In the study area, the weathering processes of parent materials release substantial As into soil, which has much higher toxicity and carcinogenic effects than other metals [9,12,41]. Despite this, the health risks posed by industrial activities demand long-term monitoring and need to be minimized by the related pollution remediation measures.

Overall, the integration of source apportionment and risk assessment could offer an accurate approach to comprehensively capture source contribution to the ecological and health risks [63,65]. Compared to the traditional risk assessment methods, the source-oriented risk assessment could deliver insight into the impact of natural and anthropogenic sources on soil ecosystems and human health, which is more conducive to providing reliable tools for pollution management and control [64,66]. The in-depth source-oriented risk assessment remains several issues to be addressed, including the influence of soil parent materials, land use type, and bio-availability [63,66], as well as model uncertainty arising from the unidentified component and proper selection of exposure parameters [13]. Hence, further research is required to take the spatial heterogeneity into account and reduce the uncertainty in source apportionment to conduct the comprehensive evaluation of pollution risk in soil.

4. Conclusions

In the study, it was found that Cu, Zn, Cd, and Pb were significantly enriched in agricultural soils in a typical mining and smelting industrial area, and Cd was identified as the most important pollutant in the region. Multivariate statistical methods and the APCS-MLR model were used to qualitatively and quantitatively identify the heavy metal sources. The results showed that Cr (79.8%), Ni (73.0%), As (74.5%), and Mn (58.0%) mainly came from natural sources; Cu (34.3%), Zn (67.2%), Cd (75.1%), and Pb (67.9%) were significantly contributed by industrial activities, while Hg (71.0%) was enriched by the mixed sources of human activities and natural origin. Based on the outputs of the APCS-MLR model,

source-specific risks were estimated, showing that industrial emissions had contributed 49.1% of the overall potential ecological risk, and As naturally occurring in the soils poses a significant threat to human health. The results suggested that there is an urgent need for pollution control and prevention in the region. The findings can offer the scientific foundation for policy development for soil pollution prevention and control.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su16041673/s1, Text S1: Microwave-assisted digestion procedures for determination of total content of heavy metals; Table S1: Potential ecological risk levels based on risk factor of a single trace metal; Table S2: Potential ecological risk levels based on risk index of multiple trace metals; Table S3: Exposure parameter used for health risk assessment to toxic elements; Table S4: Reference dose (*RfD*, mg/(kg · day)) and slope factor (*SF*, $[mg/(kg \cdot day)]^{-1}$) of heavy metals through different pathways [8,67,68]; Table S5: Variance explained by the extracted components of PCA analysis for trace metals in Yingtan agricultural soil; Table S6: Source-specific non-carcinogenic risks of heavy metals from different sources; Figure S1: Distribution of the main mining areas, residential areas, and roads in Yingtan; Figure S2: Spatial distributions of single risk factors of trace metals in Yingtan agricultural soils; Figure S3: Spatial distribution of sampling sites with severe pollution of Hg (the concentration exceeding 0.100 mg/kg); Figure S4: The contribution of different sources to the total metal contents; Figure S5: The spatial distribution of RI from different sources.

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