



Article Ecological Risk Assessment and Source Analysis of Heavy Metals in Farmland Soil in Yangchun City Based on APCS-MLR and Geostatistics

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Abstract: Yangchun City, a typical polymetallic ore distribution area in Guangdong Province (China), was selected as the research region to study the content, distribution, source, and possible impacts of heavy metals (HMs) (Arsenic: As; Cadmium: Cd; Chromium: Cr; Copper: Cu; Mercury: Hg; Nickel: Ni; Lead: Pb; and Zinc: Zn) on the farmland soil of this City. According to our findings, the spatial distribution of HMs in Yangchun City shows higher concentrations in the north and southeast and lower in the west and other regions. Metal content in some sampled sites of the agricultural land exceeded the soil pollution risk screening values, particularly As (7.5%), Cd (12%), Cu (4%), Hg (14.5%), and Pb (3%). Additionally, the average content of As, Cu, Cd, Pb, Hg, and Zn from the studied areas surpassed the soil background value of Guangdong Province for all metals. The absolute principal component score-multiple linear regression (APCS-MLR) was used to identify potential sources of HMs in the soil samples. There are three potential sources identified by the model: traffic emissions, natural sources, and agricultural activities, accounting for 28.16%, 16.68%, and 14.42%, respectively. Based on the ecological risk assessment, the potential ecological risk (E_r^t = 310.77), Nemero pollution index (PN = 2.27), and multiple possible effect concentration quality (mPECQs = 0.23) indicated that the extent of heavy metal pollution in the soil samples was moderate. Three sources were identified: traffic emissions, natural sources, and agricultural activities. We suggest that by combining the above results, a monitoring and early warning system focused on Cd and Hg can be established. The system could utilize geographic information systems and remote sensing technologies to achieve dynamic monitoring and prediction of pollution. Regular testing of soils and sustainable management practices are also recommended to control and remediate contamination.

Keywords: heavy metal; source apportionment; APCS-MLR

1. Introduction

Rapid industrialization worldwide has led to heavy metal contamination in farmland soil and agricultural products. Among various environmental issues, soil pollution has become a matter of global concern [1,2]. The soil is an important environmental component of the ecosystem on the surface of the earth. However, numerous contaminants from industrial or agricultural activities pollute the soil [3]. Therefore, actions to control the source of pollution and minimize the effects of heavy metal contamination are required [4]. For instance, a detailed analysis of the type, distribution, and source of heavy metals (HMs) is critical for preventing and reducing this type of contamination [3].

Several efforts are described in the current literature to accurately assess the toxic effects of HMs using various evaluation indices. Chen et al. used enrichment factors, ecological risk coefficients, and the absolute principal component score-multiple linear regression (APCS-MLR) to describe the seasonal changes in heavy metal risks in Nanchang



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). City [5]. Fan et al. used enrichment factors, ground accumulation indicators, and potential ecological risk indicators to evaluate the heavy metal pollution status of suspended particles in the Minjiang River [6]. Dash et al. used contamination factors, enrichment factors, and the ground accumulation index to analyze the impact of different monsoon conditions on heavy metal pollution [7]. To analyze heavy metal pollution along the Lijiang River, Xiao et al. used the land accumulation index, hidden ecological risk indicator, and multiple possible effect concentration quality [8]. Ustaoğlu et al. used pollution factors, enrichment factors, hidden ecological risk indicators, enrichment factors, hidden ecological risk indicators, and a geo accumulation index to assess the heavy metal pollution in a hazelnut producing area in Turkey [9]. Similarly, in this study, various assessment methods were used to evaluate the toxicity of HMs in the soil in Yangchun.

For the sources of HMs in soil, many methods have been used to classify these qualitatively at present. Multivariate statistical analysis, particularly principal component analysis (PCA), has been extensively applied to determine HM sources in soil. Huang et al. used PCA to analyze the sources of HMs in soil in eastern China and discovered that natural sources have the highest contribution rate to Al, Mn, and Ni, whereas Hg, Pb, and Cd are from two different human sources [4]. Lv et al. [10]. used PCA and other methods to identify heavy metal sources in the soil around Ju State. They found that the main sources of Pb, Cu, Cd, and Zn were related to the parent material, while Hg was related to human activities alone. Furthermore, the distribution of five HMs in the study area has an obvious similarity with the surrounding environment. Rodríguez Martín et al. used PCA to investigate the sources of HMs in cultivated soil in Andalusia, Spain, and discovered that the sources of Cu, Pb, Zn, and Cd were related to agricultural activities [11]. PCA is a linear dimensionality reduction technique that converts various HMs from one source to a small number of principal components. However, the quantitative contribution of PCA to multiple sources of HMs cannot be clearly explained. Multivariate receptor models, such as APCS-MLR, have been developed to address this limitation of PCA. As APCS-MLR is not reliant on the prior source traits of sampling and testing, it affords greater convenience, expediency, and efficacy than traditional approaches. APCS-MLR is currently being used for the source analysis of different pollutants in the atmosphere, soil, and dust. Chen et al. used APCS-MLR to identify the source of HMs in the soil around the Miyun Reservoir, observing that mining activities, agricultural chemicals, and atmospheric sedimentation are all potential anthropogenic sources. They also found that the APC-MLR model can achieve similar results to those of the PMF model [12]; Zhang et al. employed the APCS-MLR and PMF models to discern the origins of polycyclic aromatic hydrocarbons in Taihu Lake's sediments. Their findings evinced that vehicle emissions, coal combustion, and wood combustion were the principal contributors to the pollution. Interestingly, the APCS-MLR model produced results similar to those of the PMF model despite using simpler hardware and software [13].

Therefore, the APCS-MLR model was used in this study to identify the degree of heavy metal pollution sources in the soil of Yangchun City in Guangdong Province, along with the contamination factor (*CF*), land accumulation index (I_{geo}), potential ecological risk index (E_r^i), Nemero pollution index (*PN*), and multiple possible effect concentrations (*mPECQs*). In addition to the Kaiser–Meyer–Olkin (KMO) test, other statistical analyses, including principal component analysis (PCA) and correlation analysis (CA), were also used to test the adequacy of sampling and the difference between the heavy metal contents. The symbiotic relationship between the HMs in the soil was then studied using network reasoning. Geostatistical methods were used to map out the most susceptible locations. Then, the APCS-MLR model was used to subtract the pollution source, contribution level, and percentage. On this basis, reference data on local soil management and recovery were provided.

2. Materials and Methods

2.1. Characteristics of the Study Area

Yangchun City is in the southwestern part of Guangdong Province, China (between 111°16′27″ to 112°09′22″ E and 21°50′36″ to 22°41′01″ N). The city is situated in the Yunwu Mountain range, with a total area of approximately 4037.8 square kilometers, of which arable land covers about 23.7% and forest covers about 60%. Yangchun City belongs to the karst landform zone and has a subtropical maritime monsoon climate. It is one of the cities in China with the richest mineral resources and is also a major production base for high-quality food crops in China.

The research area includes 17 towns, such as Shiwang, Tanshui, and Bajia. It has been discovered that the research area (Figure 1) contains various mineral resources such as toxic sand mines, lead–zinc mines, pyrite, iron ore, tungsten mines, and limestone [14]. With the rapid development of the industry, some of the mineral deposits have already been exploited, and others are currently under development. Yanchun City has 13 well-connected expressways, 3 national highways, 6 provincial roads, and numerous rural roads, making it an important transportation hub connecting to other cities. In recent years, Yanchun City has been planned to be a mountain and water garden livable city. However, research has found that the average levels of heavy metals such as As, Cd, Cu, Hg, Pb, and Zn in the soil of Yanchun City exceed the background values of soil in Guangdong Province. As an important grain production and ecologically livable city, understanding the risks and sources of soil heavy metal pollution is crucial to maintaining the health and sustainability of Yanchun City's agricultural system.



Figure 1. Distribution of farmland soil sampling points and Mineral hotspots in Yangchun City.

2.2. Collection and Preparation of Soil Samples

Soil samples were collected from Yangchun City, Guangdong Province, from 0 to 20 cm deep on the surface of farmland soil. The plum blossom sampling method was used to collect 4 to 8 samples at random within a radius of 20 m; the samples were sent to the laboratory for testing. Each point's longitude and latitude, along with the ambient environmental data, were documented by a GPS. Figure 1 depicts the collection of 200 soil surface samples in their entirety. After drying, the soil sample was ground with an agate mortar, and the part of the sample that could pass the 200-mesh nylon sieve was stored in a plastic bag.

2.3. Physicochemical Properties of Soil

Multiple analytical techniques were employed to ascertain the physical and chemical attributes of the soil. The information regarding chemical reagents is featured in Table 1, and the analytical instrument details are showcased in Table 2. Soil total nitrogen (TN) was determined using an automated Kjeldahl nitrogen analyzer (Kjeltec 8400). The soil samples were digested with sulfuric acid to convert organic nitrogen into ammonium sulfate. The digested samples were distilled and titrated to quantify the nitrogen content. The pH value was measured using a PHS-2F pH meter using the potentiometric electrode method. The alkali diffusion method was used to test for soil alkali-hydrolyzed nitrogen (AN). Soil available phosphorus (AP) was evaluated using flame atomic emission spectrometry (FP650) after extraction of AK was determined using flame atomic emission spectrometry (FP650) after extraction with 1 mol/L ammonium acetate (1:10 soil:extractant ratio). The sample was shaken at 20~25 °C, 150 r/min~180 r/min for 30 min. AK concentrations in the extracts were measured using an air-acetylene flame. For testing of the soil organic matter content (SOM), the oil bath heating potassium dichromate oxidation-volumetric approach was employed [15].

 Table 1. Primary chemical reagent information used in the experiment.

Reagents Name Producers		Purity	Catalogical Number	
Acetic acid	Tianjin Damao Chemical Reagent Factory, China	AR	631-61-8	
Potassium chloride	Tianjin Damao Chemical Reagent Factory, China	AR	7447-40-7	
Sodium bicarbonate	Tianjin Damao Chemical Reagent Factory, China	AR	144-55-8	
Ammonium fluoride	Tianjin Damao Chemical Reagent Factory, China	AR	12125-01-8	
Ammonium molybdate	Tianjin Damao Chemical Reagent Factory, China	AR	12054-85-2	
Potassiumdichromate	Tianjin Damao Chemical Reagent Factory, China	AR	7778-50-9	
Hydrochloric acid	Guangzhou Chemical Reagent Factory Yongda chemical, China	AR	7778-50-9	
Boric acid	Guangzhou Chemical Reagent Factory Yongda chemical, China	AR	10043-35-3	
Hydrofluoric acid	Guangzhou Chemical Reagent Factory, China	AR	7664-39-3	
Perchloric acid	Guangzhou Chemical Reagent Factory, China	AR	7601-90-3	
High purity nitric acid	Guangzhou Chemical Reagent Factory, China	AR	7697-37-2	
Sulphuric acid	Guangzhou Chemical Reagent Factory, China	AR	7664-93-9	

Table 2. The primary analytical instrument information used in the experiment.

Instrument Name	Model	Producers
pH Meter	PHS-2F	Shanghai INESA
Flame photometer	FP650	Shanghai jingke
Automatic Kjeldahl analyzer	Kjeltec 8400	Danish Foss
Inductively Coupled Plasma-Optical Emission Spectrometry	ICP-OES	US Agilent

The heavy metal content analyses were conducted by digesting the samples with HCl-HNO₃ and HF-HCl-HNO₃ solutions. The heavy metal cooking step was carried out according to the national standard "HJ 832-2017" [16] method, and the microwave digestion system (Milestone ETHOS UP) was used for digestion, and the heavy metal content was determined using the Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) method (Agilent 5110). The concentration of Cr, Zn, Ni, and Cu were determined using flame atomic absorption spectrometry. Utilizing graphite furnace atomic absorption spectrometry, we assessed the concentrations of Cd and Pb, while atomic fluorescence spectrometry was employed to measure the levels of As and Hg [17].

For each group of experiments, parallel samples were set up and analyzed in triplicate to mitigate the errors of specific batches. Two to three blanks and two to three groups of environmental sample reference material soil GBW07430 (GSS-16, Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences) were used to enclose quality control samples. The recovery rates of the various heavy metal factors analyzed were in the range of 82.61–106.28%, and the standard deviation of the parallel samples was within 8%.

2.4. Methods for the Evaluation of Heavy Metal Pollution in Soil2.4.1. Methods for Evaluating the Level of Heavy Metal Pollution in Soil

and the Nemero Pollution Index (PN) [18].

The level of heavy metal pollution in soil can be evaluated using three geochemical pollution indices: the Geoaccumulation Index (*Igeo*) [18], the Contamination Factor (*CF*) [19],

$$Igeo = Iog_2(\frac{Cn}{1.5 \times Bn}) \tag{1}$$

$$CF = \frac{Cn}{Bn} \tag{2}$$

$$PN = \sqrt{\frac{CF^2 + CF^2_{\max}}{2}} \tag{3}$$

where, C_n represents the concentration of elements observed in the soil, and B_n denotes the background concentration of heavy metals. *Fn* stands for the evaluation standard value of heavy metal *n*, and the pollution risk screening value from the Standard for Soil Pollution Risk Control of Agricultural Land (Trial) (GB 15618-2018) serves as the evaluation standard. *CF* is the average value of the *CF* pollution element of the investigated metal and *CF*_{max} is the maximum value of the contamination factor in the sample. Among them, Background values (B_n) of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the agricultural soil of the Guangdong Province were 8.9 mg/kg, 0.056 mg/kg, 50.5 mg/kg, 17 mg/kg, 0.078 mg/kg, 14.4 mg/kg, 36 mg/kg, and 47.3 mg/kg, respectively [20].

 I_{geo} consists of seven distinct tiers: $I_{geo} < 0$, uncontaminated; $0 < I_{geo} < 2$, uncontaminated to slightly polluted; $2 < I_{geo} < 3$, slightly polluted to moderately polluted; $3 < I_{geo} < 4$, seriously polluted; $4 < I_{geo} < 5$, seriously polluted to extremely polluted; and $I_{geo} \ge 5$, extremely polluted. There are four pollution levels of CF: CF < 1, no pollution; $1 \le CF < 3$, light pollution; $3 \le CF < 6$, moderate pollution; and $CF \ge 6$, heavy pollution. *PN* has five levels: PN < 0.7 is safe; $0.7 \le PN < 1.0$ is in a warning limit; $1.0 \le PN < 2.0$ indicates light pollution; $2.0 \le PN < 3.0$ refers to medium pollution; and $PN \ge 3.0$ corresponds to heavy pollution.

2.4.2. Evaluation Methods for Ecological Risk of Heavy Metals in Soil

Two ecological risk indices, including ecological risk potential index (E_r^t) and the multiple probable effect concentrations quality (*mPECQs*), are used to assess the ecological risk of the ecosystem, using the following formulas:

$$E_r^i = \frac{T_r^i \times C_n}{C_{ref}} \tag{4}$$

$$mPECQs = \frac{\sum_{i=1}^{n} \frac{C_i}{PEC_i}}{n}$$
(5)

where T_r^i is the toxicity response parameter of heavy metal element *i*, C_n is the measured concentration of HMs, C_{ref} stands for the background value of HMs in the soil. The T_r^i of As, Ni, Cu, Cr, Pb, Zn, Cd and Hg are 10, 5, 5, 2, 5, 1, 30 and 40, respectively [21,22]. C_i is the metal concentration in the soil sample and PEC_i stands for the possible effective concentration of each metal. The predicted effective concentrations of As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg are 33, 4.8, 111, 149, 48.6, 460, 459 and 1.06 mg/kg, respectively. The quantity of metal is given by n. There are five levels of E_r^i : $E_r^i < 40$, denotes negligible risk; $40 \le E_r^i < 80$, indicates a mild risk; $80 \le E_r^i < 160$, implies a moderate risk; $160 \le E_r^i < 320$, signifies a high risk; and $E_r^i \ge 320$, denotes an exceedingly grave risk. mPECQs is classified by three levels: mPECQs < 1 denotes a non-toxic nature or a low prevalence of toxicity, with an incidence rate of less than 25%, $1 \le mPECQs < 5$ signifies an incidence rate between 25% and 75%, and $mPECQs \ge 5$ indicates a highly toxic nature, with an incidence rate surpassing 75%.

2.5. APCS-MLR Model

The APCS-MLR receptor model offers a means of gauging the effects of noxious metal sources on soil, by fusing the APCS and MLR models and concentrating on the PCA model [23–27]. First, the dimension of the data is reduced, the component is derived from a similar variable using PCA, and the rotational factor load is measured as a starting point for determining the metal source [28]. The data obtained by PCA cannot be used to measure source impact, and the APCS model is then adopted as a non-standardized APCS [25,26,29]. In MLR, the soil's metal concentration served as the dependent variable, while APCS validated the model's consistency by juxtaposing the predicted value against the observed one [5,30]. This includes the following four steps:

$$(Z_o)_i = (0 - C_i)/S_i = -C_i/S_i$$
(6)

$$P_{op}^{*} = \sum_{i=1}^{n} B_{pi}^{*}(Z_{o})_{i}$$
⁽⁷⁾

$$[APCS]_{p \times j}^* = [P]_{p \times j}^* - [P_o]_{p \times j}^*$$
(8)

$$C_i = (b_o)_i + \sum APCS_p \times b_{pi} \tag{9}$$

where, C_i represents the mean content of the factor i; S_i refers to the standard deviation; $(Z_o)_i$ means the standardized matrix; P_{op}^* represents the sample factor score when the concentration is supposed to be 0; B_{pi}^* represents the factor score parameter from the rotation matrix; $[P]_{p\times j}^*$ indicates the role of factor P in the concentration of sample *j* in case of the concentration of 0; $[APCS]_{p\times j}^*$ represents the factor mark in the transformed sample *j*, and $(b_o)_i$ represents the constant term in the linear regression; b_{pi} refers to the parameter of linear regression, and $APCS_p \times b_{pi}$ refers to the contribution rate of each source.

2.6. Statistical Analysis

The data were statistically analyzed and computed using SPSS 22.0, and the APCS-MLR model was constructed. To test the suitability of the data for principal component analysis, a Kaiser–Meyer–Olkin (KMO) test was conducted. Additionally, semi-variogram fitting and kriging spatial interpolation were performed using ArcGIS 10.5. The matrix of

Spearman's correlation among soil physicochemical properties and heavy metal content was created using Origin 2022.

3. Results and Discussion

3.1. Physicochemical Properties of Soil and Toxic Metals Contents

In Yangchun city, the soil pH, AP, AN, AK, SOM and TN are 3.77–7.73, 3.62–436.75 mg/kg, 31.15–255.15 mg/kg, 12.00–864.00 mg/kg, 0.65–50.53 g/kg and 0.28–11.20 g/kg, respectively (Figure 1). In addition, the soil in the north, central, and western regions was neutral, whereas the soil in other regions was acidic. Soil pH affects the mobility of HMs in this medium; as pH decreases, heavy metal cations become more active and mobile [15].

The concentration range of HMs (in mg/kg) was considerably wide(Table 3) (Figure 2): As = 0.01-123.29; Cd = 0.01-2.30; Cr = 1.78-118.83; Cu = 2.55-248.18; Hg = 0.01-1.95; Ni = 0.64-69.74; Pb = 3.60-728.36; and Zn = 6.22-227.57. In terms of average concentration, the highest was Zn (57.17 \pm 42.82 mg/kg), followed by Pb (36.66 \pm 54.95 mg/kg), Cr $(36.63 \pm 20.33 \text{ mg/kg})$, Cu $(20.58 \pm 26.78 \text{ mg/kg})$, Ni $(12.93 \pm 7.98 \text{ mg/kg})$, As $(12.69 \pm 14.89 \text{ mg/kg})$, A mg/kg), Hg (0.30 ± 0.43 mg/kg), and Cd (0.22 ± 0.26 mg/kg). The mean levels of As, Cu, Hg, Cd, Pb, and Zn surpassed the soil's baseline values in Guangdong Province [20], indicating that HMs accumulated in the soil over time. Among the six elements with average concentrations higher than their soil background values, Cd and Hg were 3.92 and 3.84 times greater than their background values, respectively. This indicated that these HMs are significantly enriched in the soil; the enrichment may also be influenced by other external factors. As, Cu, Pb, and Zn were, respectively, 1.43, 1.21, 1.02, and 1.21 times more abundant than their background values, suggesting that the presence of these HMs was less related to human activities [15]. In addition, a great portion of the soil samples (21.5–92.5%) surpassed the soil background values of Hg, As, Pb, Cr, Cu, Cd, Ni, and Zn. The levels of Cu, As, Hg, Cd, and Pb found in the soil samples surpassed their thresholds for agricultural soil pollution risk screening, which can be ascribed to the presence of point source emissions. However, Cr and Ni content in Yangchun were not particularly high, and these two metals seem to be the least polluting factors in all cities in China [31]. These results indicated that HMs contaminated the areas where some samples were collected. Therefore, these results indicate that heavy metal contamination has been found in some of the sampling areas, and thus management of the corresponding farmland is required according to relevant standards to reduce heavy metal accumulation in crops. The coefficient of variation of the heavy metal concentration (Pb = 150%, Hg = 144%, Cu = 130%, As = 117%, Cd = 116%, Zn = 75%, Ni = 62%, and Cr = 56%) showed a significant degree of variability, the significant variability indicates that all eight heavy metal elements exhibited a high degree of variability [32], suggesting that these heavy metals have strong spatial heterogeneity at the regional scale, with apparent clustering phenomena in their spatial distribution. Moreover, the elevated levels of As, Cu, and Pb surpassed their thresholds for safe agricultural soil usage, implying that some regional soils may have been seriously polluted by point sources.

Table 3. A statistical synopsis detailing the physicochemical makeup of soil and concentrations of hazardous HMs in Yangchun City.

Elements	Mean (mg/kg)	SD (mg/kg)	Max (mg/kg)	Min (mg/kg)	CV
As	12.69	14.89	123.29	0	117%
Cd	0.22	0.26	2.3	0	118%
Cr	36.63	20.33	118.83	1.78	56%
Cu	20.58	26.78	248.18	2.55	130%
Hg	0.3	0.43	1.95	0	143%
Ni	12.93	7.98	69.74	0.64	62%
Pb	36.66	54.95	728.36	3.6	150%
Zn	57.17	42.82	227.57	6.22	75%
pН	5.36	0.73	7.73	3.77	14%
AP	63.58	46.27	436.75	3.62	73%



Table 3. Cont.

pН

Zn

Figure 2. Box plots of soil heavy metal content in Yangchun City.

AK

3.2. Geostatistical Analysis of Soil Heavy Metals

AN

AP

The results from using ArcGIS 10.5 software for semi-variogram function fitting and spatial structure analysis are shown in Table 4. The nugget values for soil Hg, Cd, Cu, Cr, Ni, As, Zn, and Pb are 0.813, 0.72, 0.662, 0.59, 0.564, 0.558, 0.5, and 0.004, respectively. Among these, the nugget for Pb is less than 0.25, indicating structural variation, with very little variation at a small scale. The nuggets for Zn, As, Ni, Cr, Cu, and Cd are between 0.25 and 0.75, indicating moderate spatial variation. The nugget for Hg is greater than 0.75, indicating a high level of spatial variation, suggesting that the spatial situation of the local agricultural land has been strongly influenced by random factors [33], as the input of heavy metals caused by various human activities has already exceeded the input caused by the original structural factors by a large margin.

OM

TN

According to the semi-variogram function, kriging interpolation was used to plot and analyze the content of eight heavy metals. As shown in Figure 3, the peak values of soil As (Figure 3a) content in the study area are mainly concentrated in a strip from the northeast to the southwest, similar to the distribution of roads in the area. The high-value areas of soil Cr (Figure 3b) content are mainly in the hilly areas in the northeast. The peak values of soil Cd (Figure 3c) content are mainly in the northern and southwestern parts. The high-value areas of soil Cu (Figure 3d) content are mainly near the central region of the study area. The high-value areas of soil Hg (Figure 3e) content are mainly in the northern region. The overall spatial distribution of soil Ni (Figure 3f) content in Yangchun City does not have large high-value areas; the high-value areas are mainly in the northern part. The high-value areas of soil Pb (Figure 3g) are mainly concentrated near the mines in the northern region.



The high-value areas of soil Zn (Figure 3h) are mainly concentrated near the mines in the northern region and the southwestern part.

Figure 3. Spatial Distribution Map of Soil Heavy Metal Content in Yangchun City.

(**g**)

(h)

Element	Semi- Variogram Model	Nugget Value (C ₀)	Abutment Value (C ₀ + C ₁)	R ²	Nugget C ₀ /(C ₀ + C ₁)
Hg	Exponential	0.101	0.541	0.864	0.813
Cd	Exponential	0.0486	0.173	0.729	0.72
Cu	Spherical	264	780	0.709	0.662
Cr	Spherical	249	607.4	0.844	0.59
Ni	Exponential	46	105.62	0.857	0.564
As	Spherical	94.7	214.1	0.724	0.558
Zn	Exponential	1496	2993	0.771	0.5
Pb	Gaussian	1	2666	0.744	0.004

Table 4. Statistical Parameters of Semi-variogram Functions for Eight Heavy Metals.

3.3. Heavy Metal Pollution Level Assessment and Potential Ecological Risk Assessment

3.3.1. Evaluation of Heavy Metal Pollution Level

The ground accumulation coefficient (I_{geo}) can be used to determine toxic metals [34]. The I_{geo} values showed the following ranges: As = -23.67 to 3.21, Cd = -16.36 to 4.78, Cr = -3.84 to 2.22, Cu = -4.89 to 1.71, Hg = -16.83 to 4.06, Ni = -5.08 to 1.69, Pb = -3.91 to 3.75, and Zn = -3.51 to 1.68. Based on the I_{geo} classification, Cd and Hg exhibited levels of pollution ranging from moderate to severe, while As and Pb demonstrated moderate pollution, and Cu, Cr, Ni, and Zn showed mild pollution.

CF demonstrated the dispersion of the trace elements and effectively mitigated the challenge of cross-level comparison [9]. According to the *CF* classification, the *CF* values of the eight heavy metal elements all showed the non-pollution level (*CF* < 1). The average *CF* values were as follows: As (0.42), Cd (0.75), Cr (0.73), Cu (0.13), Hg (0.57), Ni (0.17), Pb (0.38), and Zn (0.28). Although the mean value of the pollution factors of eight HMs is at the non-pollution level, the maximum *CF* values of Cd is 7.68, which is considered a serious pollution level (*CF* > 6). The highest *CF* magnitudes recorded for As and Pb were 4.11 and 4.28, respectively. Furthermore, 5% of the *CF* values of Hg were classified as a moderate pollution level.

The Nemero Pollution Index (*PN*) was used to determine the total pollutants in the soil [35], which revealed that the soil in the area was seriously polluted. Except Ni (*PN* = 0.35), the *PN* value of all other heavy metal elements is greater than or equal to 0.7. Cd (*PN* = 5.46) and Pb (*PN*=3.04) belong to serious pollution; As (*PN* = 2.92) and Hg (*PN* = 2.78) are moderately polluted; Cu (*PN* = 1.76) and Cr (*PN* = 1.17) are slightly polluted; Zn (*PN* = 0.70) is the warning limit.

3.3.2. Evaluation of the Ecological Risk of Heavy Metals

As previously mentioned, multiple possible effect concentration quality (*mPECQs*) had been used to assess the ecological risk of various pollutants using traditional methods to assess soil quality [30]. The *mPECQs* for eight heavy metals are as follows: As (0.38), Cd (0.05), Cr (0.25), Cu (0.28), Hg (0.19), Ni (0.27), Pb (0.28), and Zn (0.12). The mean *mPECQs* value in soil was 0.23 (*mPECQs* < 1), indicating that the toxicity in soil was not strong, and the incidence of toxicity did not exceed 25%.

 E_r^i , proposed by Swedish scientist Hakanson, fully considers the heavy metal content in soil and evaluates the ecological risk degree of heavy metal pollution in soil in combination with the toxicity level and the sensitivity of the environment to heavy metal pollution [36]. The mean E_r^i indices for As, Cd, Cu, Cr, Hg, Ni, Pb, and Zn were 14.25, 120.18, 10.77, 0.82, 153.96, 4.49, 5.09, and 1.21, respectively. The results demonstrated that Cd and Hg exhibited moderate risk, while the other elements showed low risk.

3.4. Correlation Matrix of Soil Physical and Chemical Properties and Heavy Metals in Soil

We used a correlation matrix to represent the Spearman correlations among the heavy metals and physicochemical properties of the soil samples of this study. From Figure 4,

within the heavy metals, Ni is significantly positively correlated with Cd and Cu (p < 0.01), with correlation coefficients of 0.60 and 0.58, respectively. Pb is significantly positively correlated with Zn (p < 0.01), with a correlation coefficient of 0.56. Regarding the physic-ochemical properties, alkaline nitrogen is significantly positively correlated with organic matter and total nitrogen (p < 0.01), with correlation coefficients of 0.62 and 0.78, respectively, and there is also a significant correlation between organic matter and total nitrogen (p < 0.01), with a correlation coefficient of 0.72. Between heavy metals and physicochemical properties, Pb is strongly positively correlated with total nitrogen (p < 0.01), with a correlation coefficient of 0.41. The other heavy metal elements such as Cu and Ni have a good positive correlation with the soil physical and chemical properties, such as TN, OM, AK, and AN.



Figure 4. Correlation matrix of physicochemical properties and heavy metal concentrations in farmland soil.

3.5. Identification of the Source of HMs in Soil Using the APCS-MLR Model

After standardizing the raw data, KMO and Bartlett's tests were used to check the correlation between the variables and the reliability of PCA. The KMO measurement value of the sufficient sampling value was 0.59, with p < 0.001 (Table 5). PCA was used to determine metal composition. According to Table 5, it appears that the three eigenvalues retrieved exceeded 1, implying that a noteworthy 59.26% of the overall variance could be explicated. The first component (PC1) in the rotation component matrix occupies 28.16% of the total sample variance. The second principal component (PC2) occupies 16.69%, and the third (PC3) accounts for 14.42% of the total sample variance (Table 5). In general, PCA exhibited optimal fitting when $a \ge b + 50$, where a denotes the sample count and b indicates the number of metal types [37]. This study met an exemplary standard, indicating that the results of the principal component analysis were trustworthy and amenable to the analysis of the impact of contamination sources. The linear regression coefficient value of HMs was high (R² > 0.75, *p* < 0.05), affirming the validity of the simulation model's outcomes (Table 6).

	Initial Eigenvalue			Rotated Component Matrix			
Feature	Total	Variation (%)	Cumulative (%)	Elements	PC1	PC2	PC3
1	2.253	28.159	28.159	As			0.76
2	1.335	16.686	44.845	Cd	0.71		
3	1.153	14.415	59.260	Cr	0.78		
4	0.924	11.555	70.815	Cu			0.57
5	0.826	10.322	81.137	Hg	0.57		
6	0.675	8.436	89.573	Ni	0.80		
7	0.564	7.047	96.620	Pb		0.68	
8	0.270	3.380	100.000	Zn		0.80	

Table 5. Results of variance and rotational components in heavy metal sources in soils of Yangchun City using APCS-MLR model.

Table 6. Soil Heavy Metal APCS-MLR Receptor Model.

Receptor Model	R ²
$C_{As} = 1.963 + 0.14 \times APCS_{F1} - 2.673 \times APCS_{F2} + 13.712 \times APCS_{F3}$	0.76
$C_{Cd} = -0.198 + 0.216 \times APCS_{F1} + 0.104 \times APCS_{F2} - 0.038 \times APCS_{F3}$	0.764
$C_{Cr} = 8.982 + 15.96 \times APCS_{F1} - 2.247 \times APCS_{F2} + 1.717 \times APCS_{F3}$	0.63
$C_{Cu} = -1.647 - 15.99 \times APCS_{F1} + 10.181 \times APCS_{F2} + 38.9 \times APCS_{F3}$	0.752
$C_{Hg} = 0.191 + 0.308 \times APCS_{F1} - 0.112 \times APCS_{F2} - 0.399 \times APCS_{F3}$	0.791
$C_{Ni} = -3.561 + 6.983 \times APCS_{F1} + 3.518 \times APCS_{F2} + 1.629 \times APCS_{F3}$	0.773
$C_{Pb} = -15.255 - 7.287 \times APCS_{F1} + 65.894 \times APCS_{F2} + 13.079 \times APCS_{F3}$	0.766
$C_{Zn} = 10.572 + 10.534 \times APCS_{F1} + 21.626 \times APCS_{F2} + 2.618 \times APCS_{F3}$	0.751

The APCS-MLR model was employed to ascertain the origins of HMs in every soil specimen. The most influential factors were Cd, Ni, Hg, Cr, and Zn, with contribution rates of 54.06%, 60.28%, 44.31%, 68.99%, and 37.44%, respectively (Figure 5). Cr, Ni, Zn, Cd, and Hg were ranked 22, 24, 25, 63, and 65, respectively, in terms of element abundance. The constituents of the earth's crust, when subjected to exogenous geological processes, wield a profound influence on the elemental composition of the soil [38]. The average chromium and nickel contents are lower than the background values of heavy metals in the soil of Guangdong Province. In addition, it has been discovered that the study area contains various mineral resources such as toxic sand, lead–zinc, pyrite, iron, tungsten, and limestone. Furthermore, from a spatial distribution perspective, the spatial distributions of these three elements all show high concentration ranges around the mining areas. Therefore, based on the above analysis, the first major factor is natural sources.

The second main factor had the greatest impact on Pb, with a contribution rate of 56.65% (Figure 4). The average Pb content was close to the background value, only 1.8% higher; however, the Pb content exceeded the natural background value of 27.5% of the sample points. Moreover, Pb was mostly released into the soil owing to the vehicle fuel combustion [39]. In addition, according to previous analysis, the phenomenon of a small block nugget coefficient and a large coefficient of variation in soil lead content characteristics may be attributed to the emissions from road traffic, which could cause an increase in soil lead content within a certain range on either side of the road, forming a relatively stable marginal effect. This leads to little variation in soil lead content within a small range, exhibiting a strong spatial autocorrelation. In fact, some sampling points are located near the roads in the study area. Therefore, based on the above analysis, the second major factor is traffic emissions.

As for the third primary factor, its impact is most pronounced on As and Cu, with respective contribution rates of 75.52% and 50.34% (Figure 5). The use of fertilizers, pesticides, sewage sludge, or manure increased the As and Cu content in farmland soils [40]. Furthermore, there was a history of using As- and Cu-containing pesticides, and heavy metal-containing feed additives in poultry and livestock breeding (the resulting manure

is subsequently used as land fertilizer), which may cause large-scale agricultural soil contamination [41]. Meanwhile, according to previous analysis, the distribution area of soil As content (Figure 3) highly overlaps with the distribution area of agricultural land in Yangchun City (Figure 1), and there is a positive correlation between soil Cu and soil nutrients (TN, OM, AK, and AN) (Figure 4). As a result of the preceding analysis, the third main factor was related to agricultural activities.



Figure 5. The soil heavy metal pollution sources' contribution rate in the research region.

4. Conclusions and Future Prospects

The present study is based on an investigation of the agricultural soil in Yangchun City, Guangdong Province. Eight heavy metal concentrations were determined in 200 collected soil samples. Subsequently, the descriptive analysis results of the sampling data were compared with the background values of soil in the Guangdong Province. Multiple methods for pollution assessment were applied to evaluate the pollution levels of heavy metals in the study area and to analyze the pollution risks. Correlation analysis and the APCS-MLR model were utilized to distinguish the sources of the eight heavy metals in the region, followed by a quantitative analysis of the main sources of soil heavy metals obtained. Through the comprehensive use of methods such as the semi-variogram variance function, the spatial variation characteristics and distribution of the eight heavy metals (Cr, As, Cu, Hg, Zn, Ni, Cd, and Pb) in the agricultural soil of the study area were analyzed. Furthermore, the uncertainty of potential soil heavy metal pollution risks was assessed. The primary conclusions of this study area as follows:

(1) The average content of the metals studied, in descending order, is Zn (57.17 mg/kg) > Pb (36.66 mg/kg) > Cr (36.63 mg/kg) > Cu (20.58 mg/kg), Ni (12.93 mg/kg) > As (12.69 mg/kg) > Hg (0.30 mg/kg) > Cd (0.22 mg/kg). The soil in Guangdong Province has been found to contain concentrations of As, Cd, Cu, Hg, Pb, and Zn that surpass the natural background levels [20], with a notable trend of enrichment observed. The elevated levels of heavy metals in the soil of Yangchun city, while not necessarily indicating immediate severe risks, do warrant attention due to their potential to influence the environment and human health over time through dietary exposure.

(2)

- and Zn, the *PN* of a solitary metallic element remained within the realm of safety. Of the remaining six heavy metals, Cu and Cr were deemed to cause light pollution, while As and Hg were the culprits behind medium pollution. Cd and Pb, on the other hand, were found to induce heavy pollution and high concentration areas are concentrated near the northern mining area of the study area. Among the evaluation indexes of pollution status, E_r^i of most of the eight heavy metal elements showed low risk ($E_r^i < 40$); however, Cd and Hg were classified as moderate risk ($80 < E_r^i < 160$). In light of the pollution assessment, monitoring Cd and Hg is recommended due to their notable presence and associated risks. The northern mining zone merits special attention for its elevated heavy metal levels, particularly Cd and Pb. It is suggested that people take measures to control agricultural heavy metal pollution in order to maintain a favorable agricultural production environment.
- (3) The APCS-MLR model was used to identify three main components, and the correlation value showed that $R^2 > 0.75$ (p < 0.05), indicating that the model had a statistically significant fit. The three sources identified by the APCS-MLR model were sorted according to natural sources (28.16%), traffic emissions (16.68%), and agricultural activities (14.42%). Based on the findings of this study, it is suggested that efforts to reduce agricultural heavy metal pollution in Yangchun City should primarily focus on minimizing emissions from transportation and agricultural activities, as the combined contribution of these two sources accounts for a significant portion of the total pollution load. At the same time, since natural sources constitute the majority of the total pollution load, it is also imperative to explore measures to address natural pollution.

The study suggests establishing a monitoring and early warning system for heavy metal pollution in farmland soils, with a focus on monitoring Cd and Hg pollution. It is proposed to build a heavy metal pollution monitoring and early warning platform based on geographic information systems and big data technology to achieve dynamic monitoring of the key monitoring areas. The platform database should include data such as soil types, land use, mineral distribution, and heavy metal background values. Technologies such as drones and satellite remote sensing can be used to obtain soil and vegetation parameters as model inputs, and correlation models can be established to predict the pollution status in key areas. The monitoring and early warning results output by the platform can provide a basis for environmental management departments to formulate prevention and control policies.

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