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Spatial Heterogeneity Analysis and Risk Assessment of Potentially Toxic Elements in Soils of Typical Green Tea Plantations

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Abstract: The spatial heterogeneity of potentially toxic elements (PTEs) in a typical green tea-producing area in Zhejiang was investigated with application of geostatistics. The positive matrix factorization (PMF) was conducted for analysis of pollution sources and risk assessment of the soil of the tea garden. The results revealed that 93.52% of the study area did not exceed the PTEs risk screening value in the soil pollution risk control standard of agricultural land. The results of the spatial heterogeneity analysis showed that Cd and Pb had moderate spatial auto-correlation, exhibiting similar spatial distribution patterns. The high-value locations were distributed in the southeast of the study area, while low-value locations were distributed in the southwest of the study area. The Cr, As, and Hg had strong spatial auto-correlation, while Cr and As had similar spatial distribution patterns whose high-value areas and low-value areas were concentrated in the west and center of the study area, respectively. The Cd, Pb, and As originated from the agricultural source, transportation source, and industrial source, respectively, while Cr and Hg were from the natural source on the basis of the results of the PMF model. The results of a potential ecological risk assessment revealed that five PTEs in the study area were of low potential risk. The single-factor ecological risk ranking was Cd > As > Hg > Cr > Pb. The overall ecological risk in the study area was slight. The human health risk model indicates that there was a non-carcinogenic risk for children in the study area, and the high-value area was concentrated in the northwest of the study area. It is concluded that emphasis shall be given to excessive Cd caused by agricultural sources in the southeast of the study area, and control and monitoring will be strengthened in the northwestern part of the study area. The relevant measures for prevention of soil pollution must be conducted.

Keywords: tea plantation soil; PTEs; spatial heterogeneity; risk assessment



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1. Introduction

In recent years, rapid industrial and agricultural development has introduced potentially toxic elements (PTEs) into soils through various pathways such as industrial activities, agriculture, and transportation, posing direct or indirect threats to both ecosystems and human health [1,2]. It has been reported that cadmium (Cd) exposure can damage the kidneys, liver, and immune system [3]; long-term exposure to chromium (Cr) can induce skin eruptions, ulceration, bronchitis, asthma, gastric disturbances, genetic mutations, hepatic damage, kidney failure, and lung cancer [4]. Lead (Pb) can disrupt the hematopoietic system and cause anemia [5]; mercury (Hg) exerts strong teratogenic, neurotoxic, genotoxic, and bioaccumulative effects [6]; and arsenic (As) can cause bladder cancer and skin cancer,

among other ailments [7]. Consequently, to stop the entry of PTEs into the food chain, scientists have emphasized their research on soil pollution with PTEs.

The current research on soil PTEs has primarily focused on spatial variation, risk assessment, and source identification [8,9]. The methodologies, such as geostatistics and multivariate statistics, which reveal the spatial characteristics of soil PTEs, has become a priority area of research. For example, the positive definite matrix factorization (PMF) model, potential ecological risk and human health risk assessment model, and the comprehensive source risk method were used to conduct resource allocation and risk assessment in the densely populated area of molybdenum tailings in Qinling, China [10]. Geostatistical methods, multivariate statistical methods, and PMF models were used to assess and analyze the PTE risk and pollution source allocation in farmland soil in Puning, China [11]. Moreover, geostatistical methods were also used to investigate the spatial distribution characteristics of potential toxic elements in soils near oil production plants in central China, and their potential risks were further analyzed by using potential ecological risk and human health risk assessment models [12]. However, limited research has emphasized soil of tea crops compared to soil of other agricultural crops. As an international tea producer and exporter, China has a profound history and culture of tea production and trade. The Zhejiang Province is a major tea-producing and exporting province in China due to its favorable environment for tea plant growth. Tea plants, which prefer acidic and high-fertility soils, can mobilize PTEs in soils and increase their bioavailability [13]. Additionally, as perennial evergreen plants, tea leaves can adsorb and deposit more pollutants from air compared to annual plants. Furthermore, the soil quality directly affects the growth of tea trees and the quality of tea leaves [14]. Growing tea plants in PTE-contaminated soils transports PTEs from soils to tea leaves, which further enter into the human body through the food chain, posing potential health risks to humans [15].

This study emphasizes the evaluation of Cd, Cr, Hg, Pb, and As content in the soil of a typical tea plantation town in Zhejiang Province. The objectives of our research study were to (1) explore the sources and spatial heterogeneity of soil PTEs in the study area through geostatistical and source analysis methods and (2) evaluate the risk of PTEs in tea plantation soil on human health. This research will generate new ideas and theoretical support for the prevention and control of PTE pollution in tea plantation soil and for the protection of agricultural ecological environments.

2. Materials and Methods

2.1. Study Area, Soil Collection, and Chemical Analysis

The study area is located in the southwestern part of Zhejiang Province, classified as a low-mountainous and hilly-terrain region. It belongs to subtropical monsoon climate with average annual temperature of 17.8 °C and a frost-free period of 236 days throughout the year [16]. The study area is dominated by red and paddy soil types whose parent material primarily consists of acidic magmatic weathering products, resulting in a wide distribution of acidic soils and being suitable for the growth of tea plants. Tea cultivation in the study area can be traced back to the Three Kingdoms period (over 1800 years ago), when it was renowned for its abundant tea production and unique tea quality. Its tea production then peaked during the Ming and Qing dynasties, and still enjoys a prestigious reputation today [17].

The soil and plant samples were collected from an 88 hm² tea plantation area with a grid density of the plots of 100 m × 100 m at the mature stage of the tea plants. The soil sampling method followed an “S”-shaped technique, and soil samples were collected from the surface layer (0–20 cm) at a distance of 5–10 cm from the tea tree fertilization trench. Each soil sample consisted of a composite of five soil cores, totaling approximately 1.5 kg, which were placed into sealed plastic bags. Two top tea leaves and one bud of each tea plant were collected. Finally, a total of 108 soil samples were collected from the tea plantation soil, and the sampling points are depicted in Figure 1.

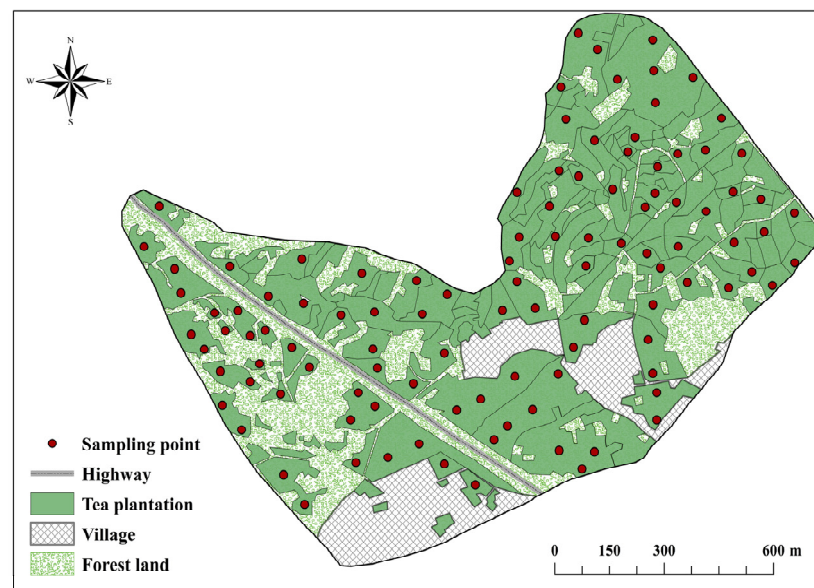


Figure 1. Distribution of soil sampling points in tea plantations in the study area ($n = 108$).

The soil samples were air-dried, ground, and passed through a 20-mesh nylon sieve for determination of the soil physicochemical properties. The soil samples were further passed through a 100-mesh nylon sieve for determination of the total concentration of soil PTEs. The fresh tea samples were rinsed with deionized water to remove dust, then oven-dried at 105 °C for 30 min and 50 °C to a constant weight. The dry tea samples were ground and passed through a 20-mesh nylon screen for a full analysis of the PTEs.

The soil pH was measured using a pH meter (Mettler Toledo, FE28, Ohio, USA) with a soil-to-water ratio of 1:2.5 (w/v). Soil organic matter (SOM) was determined using the potassium dichromate heating method. The soil samples were digested with $\text{HF-HNO}_3\text{-HClO}_4$ (7:5:1, $v/v/v$) at 160 °C, and the tea samples were digested with HNO_3 at 180 °C [18]. Then, the total concentration of PTEs in the soil and tea samples was measured using inductively coupled plasma optical emission spectroscopy (ICP-OES, Leeman Prodigy 7, Ohio, USA). The quality control procedures for PTE concentrations in soil and tea were performed with a standard soil sample (GSS-14) and green tea (GBW10052). The results showed that the recovery rate of PTEs ranged from 93.0 to 105.3% in tea samples and from 93.2 to 108.2% in soil samples. The relative deviations of duplicate samples were controlled to be less than 10%.

2.2. Spatial Analysis Methods

(1) Variogram Function: Variogram functions are used to describe whether a variable within the study area is influenced by random factors or structured factors [19]. They reflect the variation in soil properties between observed values at different distances within the study area and are an effective tool for exploring spatial variation patterns [20].

(2) Kriging Interpolation: Kriging interpolation is a method that utilizes the original data of regionalized variables and the structural characteristics of the variogram function to obtain unbiased optimal estimates within a certain regional range based on a limited number of samples. It is commonly used as a primary method to interpret the spatial distribution of soil indicators by combining field sampling with laboratory analysis.

2.3. Potentially Toxic Elements Source Apportionment Models

The positive matrix factorization (PMF) model is a multivariate factor analysis tool that decomposes the receptor content data matrix into a factor contribution matrix and a factor distribution matrix under non-negative constraints. The goal of PMF is to resolve

pollution sources and their contributions based on the synthesized dataset. The calculation method is as follows [21,22]:

$$X_{ij} = \sum_{k=1}^p G_{ik}F_{kj} + E_{ij} \quad (1)$$

In the equation:

X_{ij} represents the content matrix of the j th PTEs in the i th sample.

G_{ik} represents the contribution of the k th source to the i th sample.

F_{kj} represents the characteristic value of the k th source for the content of PTEs j th, where p is the number of factors. E_{ij} represents the residual.

The PMF model is constrained and iteratively computed based on weighted least squares. The factor contribution and distribution were obtained by minimizing the objective function Q of the PMF model [23]. The calculation method is as follows [21]:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{E_{ij}}{U_{ij}} \right)^2 \quad (2)$$

In the equation:

U_{ij} represents the uncertainty magnitude of the j th element in the i th sample.

The PMF model operates using both concentration data and uncertainty data, which encompass sampling and analysis errors [24]. In this study, the concentrations (C_{ij}) of five soil PTEs, namely, Cd, Cr, Pb, As, and Hg, all exceed the method detection limit (MDL). The uncertainty (Unc) calculation method is as follows [21]:

When the concentration of a chemical element is below or equal to the corresponding method detection limit (MDL), the uncertainty is calculated as:

$$U_{nc} = \frac{5}{6} \times MDL \quad (3)$$

Otherwise, it is calculated as:

$$U_{nc} = \sqrt{(\theta \times C_{ij})^2 + (MDL)^2} \quad (4)$$

In the equation:

C_{ij} represents the concentration of PTEs i in the j th sample.

MDL is the method detection limit for the sample.

θ represents the relative standard deviation.

2.4. Potentially Toxic Elements Risk Assessment Models

(1) Potential Ecological Risk Index Method: The potential ecological risk index method integrates PTE content with environmental ecological effects and toxicological effectiveness. The calculation method for the potential ecological risk index is as follows [25,26]:

$$E_r^i = T_r^i \times \left(\frac{C_i}{S_i} \right) \quad (5)$$

$$RI = \sum_{i=1}^n E_r^i \quad (6)$$

In the equation:

C_i represents the measured value of element i in the sample.

S_i represents the evaluation standard for element i .

E_r^i represents the potential ecological risk coefficient of a certain PTE.

T_r^i represents the toxicity response coefficient for PTEs (Cd, Cr, Pb, Hg, and As are 30, 2, 5, 30, and 10, respectively).

RI represents the comprehensive potential ecological hazard index of multiple PTEs.

The grading criteria for E_r^i and RI are as follows:

$E_r^i \leq 40$ indicates low potential risk; $40 < E_r^i \leq 80$ indicates moderate potential risk; $80 < E_r^i \leq 160$ shows moderate to high potential risk; $160 < E_r^i \leq 320$ indicates high potential risk; $E_r^i > 320$ indicates extremely high potential risk; $RI \leq 150$ indicates a slight ecological hazard; $150 \leq RI \leq 300$ indicates a moderate ecological hazard; $300 \leq RI \leq 600$ indicates a strong ecological hazard; and $RI > 600$ indicates a very strong ecological hazard.

(2) Human Health Risk Assessment: To quantitatively assess the adverse effects of PTEs on human health in the study area, this study has adopted the multi-media, multi-pathway risk assessment model developed by USEPA for health risk assessment. The main routes of entry of PTEs into the human body are through the food chain and through daily contact, inhalation, and skin contact. This study has mainly considered three main exposure pathways: oral ingestion, dermal contact, and inhalation. The calculation of exposure levels for adults and children in the study area is as follows [27,28]:

$$SER_{OI} = \frac{OSIR \times ED \times EF \times ABS_o}{BW \times AT} \times 10^{-6} \quad (7)$$

$$SER_{DC} = \frac{SAE \times SSAR \times ED \times EF \times E_v \times ABS_d}{BW \times AT} \times 10^{-6} \quad (8)$$

$$SER_{PI} = \frac{PM_{10} \times DAIR \times ED \times AF \times (f_{spo} \times EFO + f_s \times EFI)}{BW \times AT} \times 10^{-6} \quad (9)$$

In the equation, SER_{OI} , SER_{DC} , and SER_{PI} represent the exposure doses of PTEs through oral ingestion, dermal contact, and inhalation pathways, respectively. The exposure parameters adopt USEPA's recommended parameters for exposed populations and relevant research results from previous studies [29]. The meanings and values of the relevant exposure parameters are presented in Table 1.

Table 1. Health risk assessment parameters.

Parameter	Parameter Meaning and Unit	Adult	Children
$OSIR$	daily soil intake (mg d^{-1})	100	200
ED	exposure duration (a)	24	6
BW	body weight (kg)	61.8	19.2
SAE	exposed skin area (cm^2)	5374	2848
$SSAR$	soil adherence factor to skin ($\text{mg cm}^{-2} \text{d}^{-1}$)	0.07	0.2
E_v	daily skin contact event frequency (d a^{-1})	1	1
EF	exposure frequency (d a^{-1})	350	350
PM_{10}	particulate matter concentration in air (mg m^{-3})	0.119	0.119
$DAIR$	daily inhalation volume of air ($\text{m}^3 \text{d}^{-1}$)	14.5	7.5
AF	proportion of inhaled soil particles retained in the body	0.75	0.75
f_{spo}	proportion of soil particles in outdoor air	0.8	0.8
f_s	proportion of soil particles in indoor air	0.5	0.5
EFO	outdoor exposure frequency (d a^{-1})	87.5	87.5
EFI	indoor exposure frequency (d a^{-1})	262.5	262.5
AT	non-carcinogenic effect average time (d)	8760	2190
AT	carcinogenic effect average time (d)	27,740	27,740
ABS_o	oral intake absorption efficiency factor	1	1
ABS_d	dermal contact absorption efficiency factor	As: 0.03, others: 0.001	

PTEs can accumulate in the human body, leading to chronic non-carcinogenic risks and carcinogenic risks. In this study, non-carcinogenic and carcinogenic risks were assessed for human health due to exposure to PTEs in farmland soil. These are represented, respectively, by THI and TCR:

$$THI = \sum HQ_{ij} = \sum \frac{SER_{ij} \times C_i}{RfD_{ij} \times SAF} \quad (10)$$

$$TCR = \sum CR_{ij} = \sum SER_{ij} \times SF_{ij} \times C_i \quad (11)$$

In the equation:

SER_{ij} represents the exposure dose of PTEs i through exposure pathway j .

RfD_{ij} represents the reference dose of PTEs i through exposure pathway j .

SAF is the soil exposure adjustment factor, set to 0.5.

HQ_{ij} is the non-carcinogenic risk of PTEs i through exposure pathway j , with THI representing the total non-carcinogenic risk from all PTEs.

SF_{ij} represents the slope factor of PTEs i through exposure pathway j for carcinogenic risk assessment.

CR_{ij} is the carcinogenic risk of PTEs i through exposure pathway j , with TCR representing the total carcinogenic risk from all PTEs.

The values of RfD and SF are based on relevant research findings from previous studies. The meanings and values of the relevant parameters are given in Table 2.

Table 2. RfD and SF take parameters.

PTEs	$RfD/(\text{mg kg}^{-1} \cdot \text{d}^{-1})$			$SF/(\text{mg kg}^{-1} \cdot \text{d}^{-1})$		
	Oral Ingestion	Inhalation	Dermal Contact	Oral Ingestion	Inhalation	Dermal Contact
Cd	1.00×10^{-3}	1.00×10^{-3}	2.50×10^{-5}	0.38	6.1	0.38
As	3.00×10^{-4}	3.00×10^{-4}	3.50×10^{-4}	1.5	18.3	1.5
Cr	3.00×10^{-3}	2.86×10^{-5}	7.50×10^{-5}		42	
Pb	3.50×10^{-3}	3.50×10^{-3}	5.30×10^{-5}			
Hg	3.00×10^{-4}	8.57×10^{-5}	2.10×10^{-5}			

2.5. Statistical Analysis

The data were subjected to descriptive statistical analysis using Excel 2003 and SPSS 22.0 software. For data not following a normal distribution, logarithmic transformation or Box-Cox transformation using Minitab 18 software was employed to achieve normality. Spatial variation structure analysis and the establishment of optimal theoretical models were conducted using GS+ 9.0 software. Source apportionment of PTEs was determined with the PMF model (EPA PMF 5.0). Kriging interpolation analysis was performed using Geostatistical Analyst Tools in the Spatial Statistics module of ArcGIS 10.2 software. Correlation analysis was carried out with Origin 2022 software.

3. Results and Discussion

3.1. Soil Properties and Accumulation of Potentially Toxic Elements in Soils and Tea Leaves

The pH, SOM, and PTE contents in the tea garden soil of the study area are presented in Table 3. The soil pH ranged from 4.06 to 4.78, with a mean value of 4.39, which revealed overall acidity with minimal variation. Soil acidification not only affects plant nutrient absorption, but also influences the migration and transformation of metals such as Cu, Zn, Cd, and Fe in the soil [30]. Attention shall be given to improvement of acidified soil through soil amendments (biochar, attapulgate, etc.) [31,32].

The SOM contents of the soils ranged from 6.45 to 46.60 g kg⁻¹, with a mean value of 32.06 g kg⁻¹, which is in the range of high soil quality for tea gardens (20 g kg⁻¹) according to fertility grading standards of tea garden soil (NY/T 853-2004 which was published by Ministry of Agriculture and Rural Affairs of the People's Republic of China) The average Cd content of the soil was 0.19 mg kg⁻¹, which is approximate to the background value of soil in Zhejiang Province, and is considered as uncontaminated soil according to the soil environmental quality standards of agricultural soils in China [33]. However, its maximum value was 1.49 mg kg⁻¹, which exceeds the risk screening value (0.3 mg kg⁻¹) by 4.97 times. Additionally, 6.48% were polluted sites.

Table 3. Descriptive statistical characteristics of soil pH, SOM (g kg^{−1}), and PTEs (mg kg^{−1}).

Soil Index	Min	Max	Mean	SD	CV	Background Value ^a	Screening Value ^a	Exceedance Rate	K-S
pH	4.06	4.78	4.39	0.19	4.31				0.85
SOM	6.45	46.60	32.06	8.20	25.58				0.99
Cd	0.05	1.49	0.19	0.25	132.43	0.20	0.3	6.48	0.52 *
Cr	23.10	62.50	32.70	9.07	27.74	90	150	0	0.28 **
Pb	16.80	66.60	36.03	9.74	27.04	35	70	0	0.53
Hg	0.02	0.09	0.04	0.02	49.08	0.15	1.3	0	0.26
As	8.09	33.60	12.50	4.64	34.43	15	40	0	0.18 **

Note: * indicates results after logarithmic transformation, ** indicates results after Box–Cox transformation. For Cr, the λ value is -3 , and for As, the λ value is -0.5 . ^a Data from a national standard (GB 15618-2018) which was published by Ministry of Ecology and Environment of the People's Republic of China.

The contents of Cr, Pb, Hg and As in the soil samples ranged from 23.10 to 62.50 mg kg^{−1}, 16.80 to 66.60 mg kg^{−1}, 0.02 to 0.09 mg kg^{−1}, and 8.09 to 33.60 mg kg^{−1}, respectively. All of their maximum values were lower than the risk screening value of the soil environmental quality standards for agricultural soils in China (GB 15618-2018), showing no pollution of Cr, Pb, Hg, or As in the tea garden soil. The PTE concentrations of tea leaves (Table 4) were low and met the tea safety standard published by the Ministry of Agriculture and Rural Affairs of the People's Republic of China.

Table 4. Concentrations of PTEs in tea leaves (mg kg^{−1}).

Plant Index	Min	Max	Mean	SD	CV	Tea Safety Standard *
Cd	0.003	0.058	0.019	0.015	77.72	1.0
Cr	0.037	0.550	0.293	0.106	35.99	5.0
Pb	0.083	0.400	0.220	0.066	29.85	5.0
Hg	0.010	0.015	0.0104	0.001	10.09	0.3
As	0.079	0.210	0.139	0.034	24.64	2.0

Note: * Data from industrial standards (NY5244—2004 and NY659—2003) which were published by Ministry of Agriculture and Rural Affairs of the People's Republic of China.

The coefficient of variation (CV), which is the ratio of the standard deviation to the mean, reflects the variability and dispersion of elements in the soil. The coefficients of variation for Cd, Cr, Pb, Hg, and As were 132.43%, 27.74%, 27.04%, 49.08%, and 34.43%, respectively. The results indicated that the contents of Cd at the sampling points exhibited significant variability, likely attributable to discrete input of human activities, natural factors, or external influences [33].

3.2. Spatial Characteristics of Potentially Toxic Elements in Tea Garden Soil

3.2.1. Spatial Structure Characteristics of Potentially Toxic Elements in Tea Garden Soil

Geostatistical methods were employed for the analysis of the spatial variation characteristics of PTEs in tea garden soil. As presented in Table 3, The K-S values for Pb and Hg were higher than 0.05 (Table 3), showing normal distribution. After logarithmic transformation, the Cd followed a normal distribution after logarithmic transformation, while Cr and As followed a normal distribution after Box–Cox transformation. There was a semi-variogram structure in the PTEs of tea garden soil (Table 5). The nugget variance values for Cr and Hg were close to 0 from the perspective of nugget variance, which indicates that they were less affected by random factors and were mainly influenced by structural factors such as parent material, climate, and topography. Conversely, the nugget variances for Cd, Pb, and As were relatively large, showing a significant influence from random factors such as cultivation and fertilization. The optimal models for Cr, Hg, and As were Gaussian models, with ranges of 95.26, 400.10, and 398.37 m, respectively. The Cd exhibited an exponential model with a range of 657.00 m, while Pb showed a linear

model with a range of 719.56 m. The nugget effect values for Cr, Hg, and As were all less than 25%, which revealed strong spatial correlation and primarily originated from soil parent material, topography, and climatic variations. The nugget effect values for Cd and Pb ranged from 25% to 75%, which demonstrated moderate spatial variation, and this was influenced by structural factors such as soil parent material and topography as well as by random factors such as fertilization and cultivation.

Table 5. Semi-covariance function model for soil PTEs and related parameters.

Soil Index	Model	Nugget	Sill	Range (m)	Nugget/Sill (%)	R ²
Cd	Exponential	37.7	92.4	657.00	40.8	0.574
Cr	Gaussian	0.0055	0.0749	95.26	7.3	0.612
Pb	Linear	29.6031	94.9838	719.56	31.2	0.615
Hg	Gaussian	0.00011	0.00052	400.10	21.2	0.944
As	Gaussian	6.4	26.42	398.37	24.2	0.577

3.2.2. Spatial Distribution Characteristics of Potentially Toxic Elements in Tea Garden Soil

The data for each element conformed to a normal distribution after processing of logarithmic transformation and Box–Cox. The Kriging interpolation was used to spatially interpolate the soil PTE content at each sampling point in the study area. The spatial interpolation results for each element are displayed in Figure 2. The spatial distribution of Cd and Pb reveals that high-value areas were concentrated in the southwest direction and low-value areas were concentrated in the southeast part of the study area, exhibiting a declining trend from high to low values. The spatial distribution of Cr and As displayed that high-value areas were concentrated in the west of the study area and low-value areas were concentrated in the central part of the study area. The spatial distribution of Hg exhibited a patchy pattern overall, with no apparent distribution pattern.

3.3. Source of Potentially Toxic Elements in Tea Garden Soil

The spatial structural characteristics of five PTEs elements in the study area were elucidated; however, their sources and influence factors are still unclear. The aim of this study was to determine whether there was co-pollution or composite pollution among PTE elements by analyzing their interrelationships, thereby further identifying the pollution sources [34,35]. The Pearson correlation analysis in SPSS 22.0 was employed to evaluate the similarities and differences between elements. The PMF model was used to identify sources of PTEs.

The Cd was significantly positively correlated with Pb and showed significant positive correlations with pH and SOM (Table 6). However, they exhibited no correlation with other three PTEs. The Cr and As showed significant positive correlations, but they were significantly negatively correlated with Pb. The Pb and As displayed significant negative correlations. Hg showed no correlation with other PTEs. These results revealed that Cd and Pb may have the same or similar sources and are significantly correlated with soil nutrients, which may possibly be related to agricultural activities. The Cr and As may have the same or similar sources.

The PMF model computations were conducted with factor numbers set from 2 to 4, and the process was repeated 20 times with EPA PMF 5.0 software. The optimal number of factors was determined to be 4 after comprehensive evaluation of the Q values (Figure 3). The Hg was primarily an element in Factor 1, with a contribution of 46.8%. According to the results of the semi-variance function, structural factors primarily contributed to the spatial variability of Hg, which exhibited a patchy pattern without a clear spatial regularity. Moreover, Hg showed no direct correlation with other PTEs, which illustrated that the potential origin of Hg was the soil properties. Therefore, Factor 1 was inferred as having originated from natural sources.

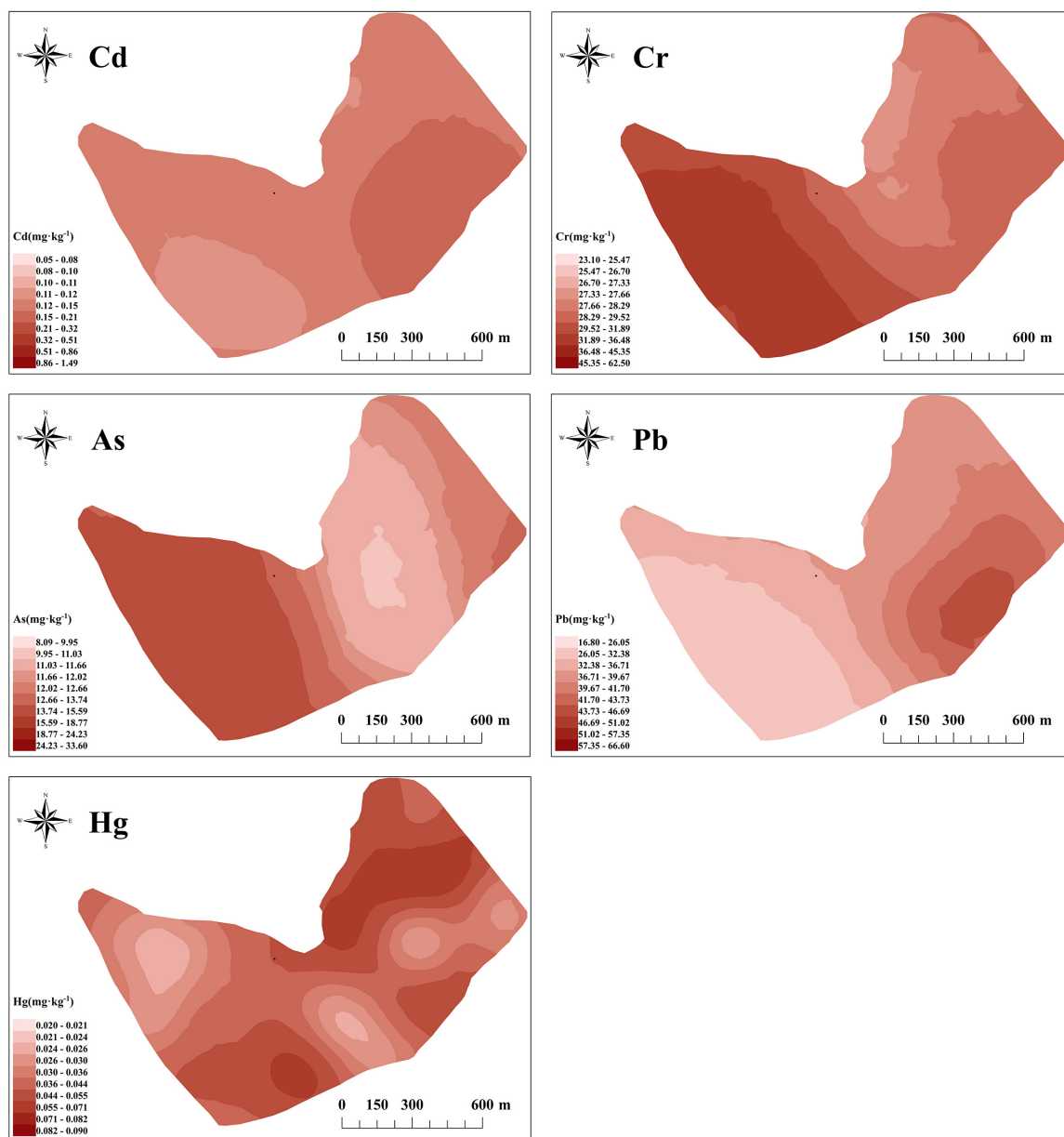


Figure 2. Spatial interpolation of soil PTEs.

Table 6. Correlation analysis of pH, SOM, and PTEs.

Soil Index	pH	SOM	Cd	Cr	Pb	Hg	As
pH	1.000						
SOM	0.189	1.000					
Cd	0.471 **	0.324 *	1.000				
Cr	0.020	−0.555 **	0.063	1.000			
Pb	0.388 **	0.714 **	0.650 **	−0.477 **	1.000		
Hg	−0.022	0.177	0.007	−0.046	0.081	1.000	
As	0.049	−0.625 **	−0.096	0.895 **	−0.536 **	−0.144	1.000

Note: * and ** indicate significant differences at $p < 0.05$ and $p < 0.01$, respectively.

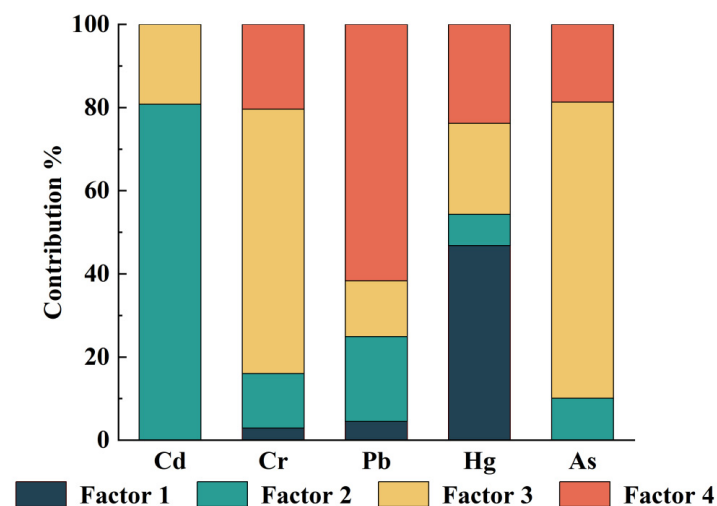


Figure 3. Analysis of soil PTE source factors and contribution rates.

The Factor 2 was dominated by Cd, contributing 80.8% to its presence, which revealed that this factor represents the primary source of soil Cd pollution in the study area. This may be due to the application of fertilizers, especially phosphate fertilizers, having introduced Cd into agricultural soil [36–38]. Moreover, a significant positive correlation was observed between Cd and SOM. Thus, Factor 2 was speculated to have originated from agricultural sources.

The Factor 3 was predominantly loaded with As and Cr, with contributions of 71.2% and 63.6%, respectively. Previous research studies have concluded that As is associated with industrial activities (mining and fossil fuel combustion) and agricultural application (As-containing pesticides) [39,40]. Mining activities were observed in the study area, and As showed a highly significant negative correlation with SOM; therefore, the As came from industrial inputs. The Cr content in the soil was significantly affected by the parent material, and with a block kriging effect of 7.3%, it belonged to structural factor influence [41,42]. Hence, Factor 3 is speculated to be a mixed source of industrial and natural origins.

The Factor 4 was primarily associated with Pb, contributing 61.7% due to its presence. It has been reported that emissions from vehicle exhaust, tire wear, and particle deposition can lead to significant Pb accumulation [43,44]. Moreover, combined with the spatial distribution results, high Pb values were concentrated around villages and areas with heavy traffic flow. Therefore, Factor 4 was inferred to have originated from traffic emissions.

3.4. Assessment of Potentially Toxic Elements Risks in Tea Garden Soil

3.4.1. Potential Ecological Risk Assessment

The average values of the potential ecological risk index for Cd, Cr, Pb, Hg, and As were 28.28, 0.73, 0.07, 8.52, and 8.99, respectively, based on the calculation results of the potential ecological risk index. The single-factor ecological risks were classified as low potential risks of all PTEs. The range of the comprehensive potential ecological index (RI) was 26.24 to 246.59, with an average value of 46.58, which showed a slight ecological hazard in the tea garden (Table 7); however, the maximum value reached a high potential risk level, which should be given attention. The single-factor potential ecological risk values showed the following order: Cd > As > Hg > Cr > Pb, displaying Cd as the highest potential ecological risk. Thus, Cd should be focused on during the evaluation of PTEs in tea garden soil.

Table 7. Evaluation of potential ecological risk indices.

Soil Index	Min	Max	Mean	Ecological Risk
Cd	7.05	223.5	28.28	Low potential risk
Cr	0.51	1.39	0.73	Low potential risk
Pb	0.03	0.10	0.07	Low potential risk
Hg	4.00	18.02	8.52	Low potential risk
As	5.39	22.10	8.99	Low potential risk
RI	26.24	246.59	46.58	Mild ecological hazard

3.4.2. Human Health Risk Assessment

The Human Health Risk Assessment revealed that the average hazard quotients (HQs) of PTEs for both adults and children were all less than 1, indicating that there is no non-carcinogenic risk to human health from individual PTEs in the study area (Table 8). However, the maximum HQ value for children's exposure to As was >1, which suggests a potential non-carcinogenic risk to children in some areas of the study region. For both adults and children, the overall HQ oral intake (HQ_{ois}) values were higher than the HQ dermal contact (HQ_{dcs}) and HQ pulmonary intake (HQ_{pis}) values, which indicates that oral ingestion is the main exposure pathway for non-carcinogenic risk to adults and children in the study area [45,46].

Table 8. Evaluation of non-carcinogenic health risks of PTEs in soil.

Index		HQ _{ois}		HQ _{dcs}		HQ _{pis}		HQ	
		Adult	Child	Adult	Child	Adult	Child	Adult	Child
Cd	Mean	5.85×10^{-4}	3.77×10^{-3}	8.80×10^{-5}	4.29×10^{-4}	4.35×10^{-6}	7.25×10^{-6}	6.77×10^{-4}	4.20×10^{-3}
	Max	4.62×10^{-3}	2.98×10^{-2}	6.96×10^{-4}	3.39×10^{-3}	3.44×10^{-5}	5.73×10^{-5}	5.35×10^{-3}	3.32×10^{-2}
Cr	Mean	3.38×10^{-2}	2.18×10^{-1}	5.09×10^{-1}	2.48×10^{-2}	2.64×10^{-2}	4.40×10^{-2}	6.53×10^{-2}	2.86×10^{-1}
	Max	6.47×10^{-2}	4.16×10^{-1}	9.73×10^{-1}	4.74×10^{-2}	5.05×10^{-2}	8.40×10^{-2}	1.25×10^{-1}	5.48×10^{-1}
Pb	Mean	3.19×10^{-2}	2.06×10^{-1}	7.94×10^{-1}	3.87×10^{-2}	2.38×10^{-4}	3.96×10^{-4}	4.01×10^{-2}	2.45×10^{-1}
	Max	5.91×10^{-2}	3.80×10^{-1}	1.47×10^{-2}	7.15×10^{-2}	4.39×10^{-4}	7.32×10^{-4}	7.42×10^{-2}	4.52×10^{-1}
Hg	Mean	4.41×10^{-4}	2.84×10^{-3}	2.37×10^{-5}	1.15×10^{-4}	1.15×10^{-5}	1.91×10^{-5}	4.76×10^4	2.97×10^{-3}
	Max	9.32×10^{-4}	6.00×10^{-3}	5.01×10^{-5}	2.44×10^{-4}	2.43×10^{-5}	4.04×10^{-5}	1.01×10^{-3}	6.28×10^{-3}
As	Mean	1.39×10^{-1}	8.98×10^{-1}	1.57×10^{-2}	7.67×10^{-2}	1.04×10^{-3}	1.73×10^{-3}	1.56×10^{-1}	9.76×10^{-1}
	Max	3.48×10^{-1}	2.24	3.92×10^{-2}	1.91×10^{-1}	2.59×10^{-3}	4.31×10^{-3}	3.89×10^{-1}	2.43
THI	Mean	2.06×10^{-1}	1.33	2.89×10^{-2}	1.41×10^{-1}	2.77×10^{-2}	4.61×10^{-2}	2.63×10^{-1}	1.51
	Max	4.30×10^{-1}	2.77	5.33×10^{-2}	2.60×10^{-1}	5.32×10^{-2}	8.86×10^{-2}	5.37×10^{-1}	3.12

The mean and maximum values of target hazard index (THI) for adults in the study area were 2.63×10^{-1} and 5.37×10^{-1} , respectively, and the mean and maximum values for children were 1.51 and 3.12, respectively. The acceptable non-carcinogenic risk threshold value was 1 according to USEPA policies; thus, it did not show a non-carcinogenic risk to adults (THI < 1). However, it did demonstrate a non-carcinogenic risk to children (THI > 1) in the study area. This may be due to the high susceptibility of children to toxicity of PTEs in the stages of growth and development, which increases the non-carcinogenic risk to children with long-term exposure [47,48].

The maximum CR values of Cd and As for adults and children in our study area were less than 1.0×10^{-6} , indicating no carcinogenic risk (CR) through oral ingestion, dermal contact, or inhalation pathways according to USEPA (Table 9). The maximum values of Cr for adults and children were 9.57×10^{-6} and 3.98×10^{-6} , respectively. The mean values were 5.01×10^{-6} and 2.08×10^{-6} , respectively, which were greater than 1.0×10^{-6} but less than 1.0×10^{-4} , which belongs to acceptable range according to USEPA's recommended carcinogenic risk levels. The overall values of CR oral intake (CR_{ois}) were higher than the overall values of CR dermal contact (CR_{dcs}). The CR pulmonary intake (CR_{pis}) indicates that oral ingestion was the main pathway for carcinogenic risk of PTEs in the study area. The mean and maximum values of CR for both adults and children were between 1.0×10^{-4}

and 1.0×10^{-6} , which reveals that the carcinogenic risk of the soil in the study area was at an acceptable level.

Table 9. Evaluation of the health risk for cancer of PTEs in soil.

Index		CR _{ois}		CR _{dcs}		CR _{pis}		CR	
		Adult	Child	Adult	Child	Adult	Child	Adult	Child
Cd	Mean	3.51×10^{-8}	5.65×10^{-8}	1.32×10^{-10}	1.61×10^{-10}	4.19×10^{-9}	1.74×10^{-9}	3.94×10^{-8}	5.84×10^{-8}
	Max	2.77×10^{-7}	4.46×10^{-7}	1.04×10^{-9}	1.27×10^{-9}	3.31×10^{-8}	1.38×10^{-8}	3.11×10^{-7}	4.61×10^{-7}
As	Mean	9.91×10^{-6}	1.59×10^{-5}	1.12×10^{-6}	1.36×10^{-6}	8.99×10^{-7}	3.74×10^{-7}	1.19×10^{-5}	1.76×10^{-5}
	Max	2.47×10^{-5}	3.97×10^{-5}	2.79×10^{-6}	3.40×10^{-6}	2.24×10^{-6}	9.33×10^{-7}	2.97×10^{-5}	4.40×10^{-5}
Cr	Mean					5.01×10^{-6}	2.08×10^{-6}	5.01×10^{-6}	2.08×10^{-6}
	Max					9.57×10^{-6}	3.98×10^{-6}	9.57×10^{-6}	3.98×10^{-6}
TCR	Mean	9.95×10^{-6}	1.60×10^{-5}	1.12×10^{-6}	1.36×10^{-6}	5.91×10^{-6}	2.46×10^{-6}	1.70×10^{-5}	1.98×10^{-5}
	Max	2.50×10^{-5}	4.01×10^{-5}	2.79×10^{-6}	3.40×10^{-6}	1.18×10^{-5}	4.93×10^{-6}	3.94×10^{-5}	4.81×10^{-5}

Spatial interpolation of the THI and TCR data for adults and children is presented in Figure 4, showing a similar distribution pattern, with low values concentrated in the central part of the study area and high values concentrated in the northwest region.

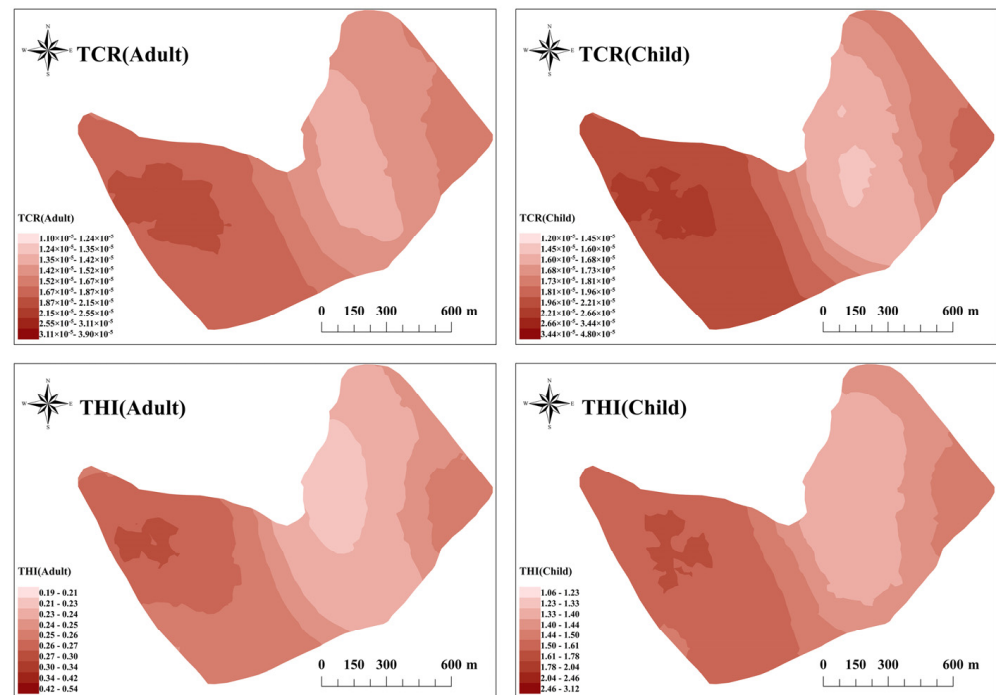


Figure 4. Soil's non-carcinogenic risk and carcinogenic risk.

4. Conclusions

Pollution due to Cr, Pb, Hg, and As was not observed in the tea garden soil of the study area. However, Cd pollution sites comprised 6.48% of sampling points, which exceeded the risk screening value of soil environmental quality standards of agricultural soils in China. The management of soil PTEs in tea gardens shall emphasize the effects of agricultural sources on soil Cd. The Cd and Pb exhibited moderate spatial autocorrelation, while Cr, Hg, and As demonstrated strong spatial auto-correlation. The main sources of PTEs in the study area comprised a mixture of industrial sources, natural sources, agricultural sources, and exhaust gas emission on the basis of the PMF model. The comprehensive ecological risk in the study area revealed slight ecological hazards, and non-carcinogenic risks to children were identified. The high-risk location is located in the northwest of the study area. It is recommended to enhance control and monitoring and to implement relevant safety measures for the area of study.

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