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Original Research

Source Identification and Apportionment of Heavy Metals in Tobacco Farmland Soils via Positive Matrix Factorization and Geochemical Indices

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Abstract

Soil heavy metal contamination in tobacco-growing areas poses a significant threat to ecosystem health and agricultural product safety. Precise identification of pollution sources and assessment of ecological risks are crucial for developing targeted environmental management strategies in such typical agricultural regions. This study conducted a systematic evaluation of pollution levels, ecological risks, and sources of heavy metals in the soils of Zhangping County, a key tobacco-producing region in Fujian Province, China. We collected 70 surface soil samples and analyzed the concentrations of Cd, Hg, As, Pb, Cr, Cu, and Zn. A comprehensive methodology was employed, including the Single-factor Pollution Index (P_i), Nemerow Integrated Pollution Index (P_n), Potential Ecological Risk Index (RI), and Geo-accumulation Index (Igeo) for contamination and risk assessment, coupled with Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF) for quantitative source apportionment. Results revealed that Cd and Hg were the primary pollutants, showing significant enrichment compared to the Fujian background values. Ecological risk assessment identified Cd and Hg as the major risk drivers, elevating the study area to a moderate-to-considerable ecological risk level. Igeo values confirmed the predominance of anthropogenic inputs for Cd and Hg. Source apportionment quantitatively resolved three main sources: industrial-traffic emissions (43.1%, contributing primarily to Pb, Cu, Zn, and Cd), a mixed source of natural pedogenesis and historical agricultural activities (48.6%, contributing predominantly to Cr, As, and Hg), and a specific agricultural source (8.3%, contributing notably to Cd and Hg). Although the total concentrations of heavy metals did not exceed national risk thresholds, inputs of highly toxic elements (particularly Cd and Hg) from agricultural and industrial activities have induced significant potential ecological risks. The synergistic application of PCA and PMF effectively elucidated the complex pollution sources. This study underscores the necessity of prioritizing the control of anthropogenic heavy metal inputs (especially Cd and Hg) in intensive

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agricultural areas and provides a scientific basis for formulating precise and differentiated soil pollution control strategies.

Keywords: heavy metals, source apportionment, ecological risk assessment, positive matrix factorization (PMF), agricultural soils

Introduction

Soil heavy metal contamination represents a pervasive and urgent global environmental challenge, with significant implications for ecosystem integrity, agricultural sustainability, and public health worldwide [1]. Across both developed and developing countries, the rapid expansion of industrial activities, urbanization, and intensive agricultural practices has markedly accelerated the accumulation of heavy metals in soils, transforming previously pristine terrestrial ecosystems into long-term reservoirs of persistent pollutants [2]. Agricultural soils are particularly vulnerable to such contamination, as heavy metals can accumulate through fertilizer and pesticide application, atmospheric deposition, and irrigation practices, posing widespread risks to food security and human health on a global scale.

Within this broader international context, soil heavy metal pollution has become especially pronounced in China. National surveys indicate that a substantial proportion of arable land is affected by heavy metal contamination, drawing significant scientific and regulatory attention to soil quality protection and sustainable agricultural development [3, 4]. Recent studies have further demonstrated that intensively cultivated agricultural soils in China are highly susceptible to heavy metal accumulation due to long-term agricultural inputs [5, 6]. In parallel, environmentally friendly remediation strategies, such as biochar-based amendments, have been increasingly explored for their effectiveness in immobilizing heavy metals, including Cd, Pb, and As, and reducing their bioavailability and ecological risks [7, 8].

The challenge of soil heavy metal contamination is particularly severe in tobacco-growing areas. Compared with many food crops, tobacco cultivation presents a more pronounced risk to soil environmental quality due to several crop-specific and management-related characteristics. Tobacco is a high-input cash crop that requires substantial amounts of fertilizers and pesticides to achieve desirable yield and leaf quality. In particular, the long-term application of phosphate fertilizers, which commonly contain cadmium (Cd) as an impurity, constitutes a major pathway for Cd accumulation in tobacco-growing soils [9]. In addition, tobacco plants exhibit a relatively strong capacity to uptake and accumulate heavy metals such as Cd, Hg, and Pb from soils, intensifying the transfer of contaminants from the soil to plant biomass. Moreover, tobacco leaves are directly used as raw materials for cigarette production, meaning that heavy metals accumulated in soils can ultimately enter tobacco products, thereby amplifying potential environmental and human health risks even when soil contamination levels are moderate [10]. As a result, tobacco-growing systems are generally more sensitive to soil heavy metal pollution than many other agricultural systems.

In this context, Zhangping County, one of the most important tobacco-producing regions in western Fujian Province, represents a highly relevant case study. The region is representative of intensive tobacco-growing areas in Fujian Province and southern China, sharing similar subtropical climatic conditions, red soil types derived from volcanic and granitic parent materials, and a long history of intensive agricultural management. Consequently, the contamination characteristics and source structures observed in Zhangping County can provide valuable insights applicable to other major tobacco cultivation regions at both provincial and national scales. Despite its importance, comprehensive studies that integrate ecological risk assessment with quantitative source apportionment in the tobacco-growing soils of this region remain limited [11, 12].

Accurately diagnosing both the extent and origins of soil heavy metal contamination requires a robust and multifaceted methodological framework. Traditional geochemical indices, including the Single-factor Pollution Index (P_i), Nemerow Integrated Pollution Index (P_n), and Geo-accumulation Index (Igeo), provide essential information on elemental enrichment levels and overall contamination status relative to background values [13, 14]. In addition, the Potential Ecological Risk Index (RI), which incorporates toxicological response factors, offers a more comprehensive evaluation of the ecological threats posed by multiple heavy metals [15]. However, while these indices are effective for characterizing pollution levels and ecological risks, they are limited in their ability to identify pollution sources [16].

To overcome this limitation, advanced multivariate statistical techniques and receptor models are increasingly employed for source apportionment. Among these, the combined application of Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF) has proven to be a powerful and complementary approach. PCA serves as an exploratory tool that reduces data dimensionality and identifies potential pollution sources, thereby guiding the selection of factor numbers for subsequent PMF modeling [17]. PMF, in turn, provides a quantitative solution by resolving source profiles and estimating their contributions to observed heavy metal concentrations without requiring prior source information [18]. The synergistic use of PCA and PMF has been shown to enhance the reliability and interpretability of source apportionment results in complex agricultural and watershed systems [19, 20].

Despite the recognized vulnerability of tobacco-growing regions such as Zhangping, existing studies often remain fragmented, focusing either on descriptive contamination assessments or on single analytical approaches. A systematic investigation that integrates comprehensive pollution assessment, ecological risk evaluation, and quantitative source apportionment is therefore urgently needed. Such an approach is essential not only for identifying contamination levels but also for elucidating pollution origins, thereby providing a scientific basis for targeted and prioritized soil management strategies [21].

Accordingly, this study aims to address these knowledge gaps by conducting a comprehensive investigation of heavy metal pollution in the soils of the tobacco-growing area of Zhangping County. The specific objectives are to: (1) systematically evaluate heavy metal contamination levels and potential ecological risks using a suite of indices (P_i, P_n, RI, and Igeo); and (2) quantitatively apportion heavy metal sources through the integrated application of PCA and PMF models. The findings of this study are expected to support evidence-based soil pollution control and sustainable agricultural management in tobacco-growing regions and similar intensive agricultural ecosystems.

Background of the Study Area

Zhangping County (24°54′-25°47′N, 117°11′-117°44′E) is located in the mountainous western region of Fujian Province, in southern China (Fig. 1). The region experiences a typical subtropical monsoon climate, characterized by abundant precipitation, with an annual average ranging from 1450 to 2100 mm, and a mild annual temperature averaging approximately 20.4°C. This climatic regime, combined with the area's complex geology, featuring Mesozoic volcanic rocks and granitic formations, has resulted in a predominantly hilly topography and well-developed red soils. These soils are classified as Udic Ferrosols under the Chinese soil taxonomy. Together, the favorable climate and rich pedological conditions create an environment highly conducive to agricultural production, with tobacco emerging as a dominant and economically critical cash crop in the region.

The study specifically focuses on the tobacco-growing soils within Zhangping, where tobacco cultivation is not only intensive but also forms a cornerstone

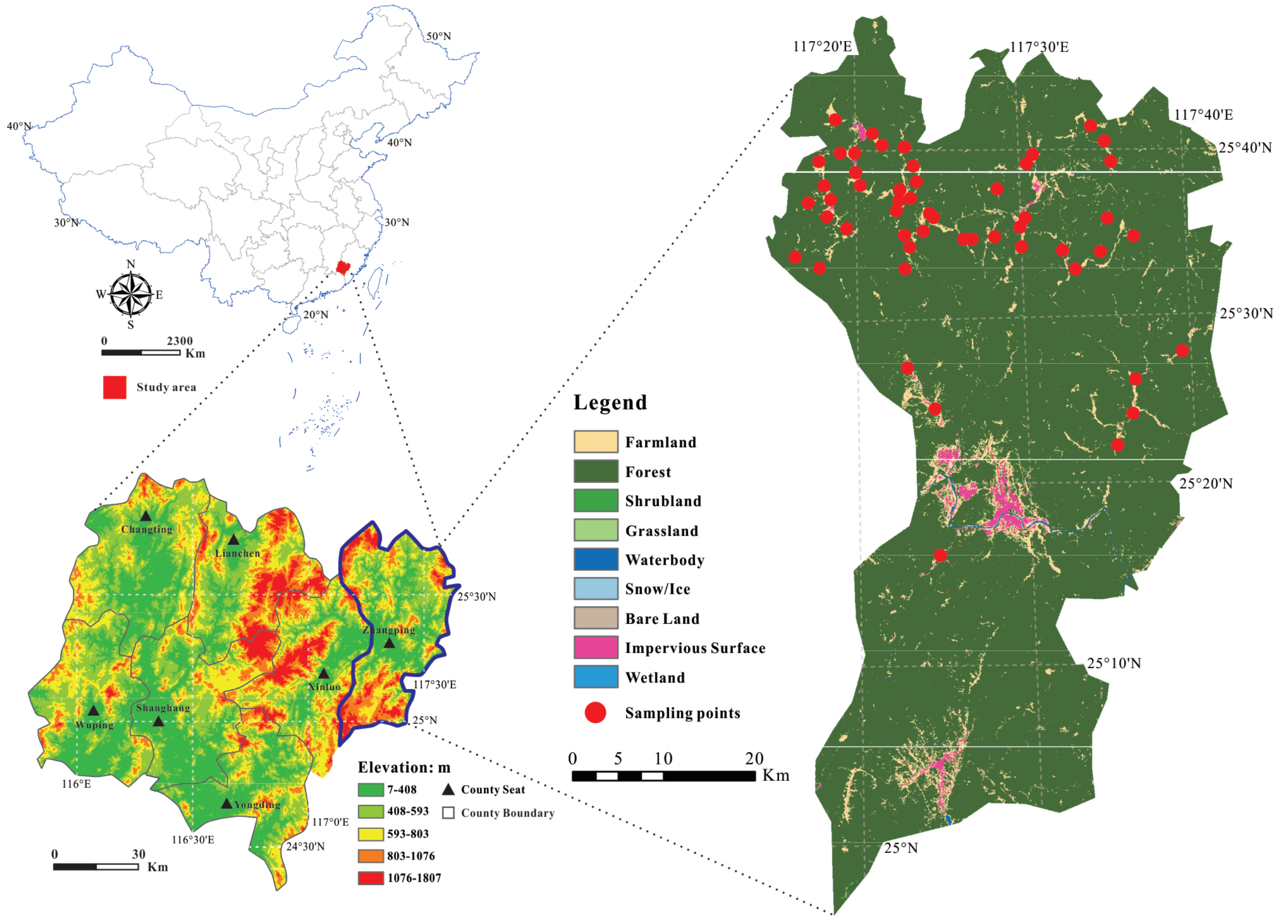

Fig. 1. Location of the study area in Fujian Province, China (map created by the authors using ArcGIS).

of the local agricultural economy. As such, the associated farmlands are subject to significant anthropogenic influences, including the recurrent application of phosphate fertilizers, which are recognized as potential sources of cadmium (Cd), as well as the use of various pesticides that may contain trace amounts of other heavy metals, such as lead (Pb) and arsenic (As). In addition to agricultural activities, the region is also subject to other anthropogenic pressures, such as vehicular emissions from local transportation networks and atmospheric deposition resulting from regional industrial activities.

Given the strategic importance of tobacco to the local economy and the vulnerability of its soil environment to contamination from both natural (geogenic) and human-induced (anthropogenic) sources, Zhangping County provides a compelling and critical case for a detailed environmental assessment. The selection of this region for investigating heavy metal pollution is of considerable practical significance. A comprehensive understanding of pollution levels, ecological risks, and the precise sources of heavy metals in this agro-ecosystem is essential for developing sustainable land management strategies and ensuring the long-term viability of this key agricultural sector.

Materials and Methods

Sample Collection

The soil sampling campaign was carefully designed to ensure that the collected samples were geographically representative of the major tobacco-growing areas in Zhangping County, Fujian Province, China. A total of 70 surface soil samples were collected from the principal tobacco cultivation zones, primarily distributed across the key tobacco-producing towns of Shuangyang, Gongqiao, and Xinqiao. These areas together account for the majority of tobacco production in Zhangping County and exhibit typical agricultural management practices, making them suitable for regional-scale assessment.

The spatial distribution of sampling sites was planned to provide adequate coverage of different cultivation areas and landscape settings while avoiding excessive spatial clustering. Sampling locations were selected within actively cultivated tobacco fields and distributed as evenly as possible across the study area, taking into account land-use patterns, accessibility, and field conditions. This strategy was intended to capture the spatial heterogeneity of soil heavy metal concentrations associated with long-term agricultural inputs and local environmental variability, while minimizing the influence of spatial autocorrelation.

Sampling was conducted after the completion of the tobacco harvesting season and prior to land preparation for the subsequent crop. This timing was chosen to reflect the cumulative effects of a full tobacco growth cycle on soil heavy metal accumulation, while reducing the influence of short-term seasonal disturbances. At each sampling site, surface soil samples were collected from the plow layer (0-20 cm), which represents the soil horizon most directly affected by agricultural practices and root activity.

A standardized plum blossom sampling method (also known as the five-point composite sampling method) was employed at each site. Specifically, 5 subsamples were collected within a 10 m × 10 m plot and thoroughly homogenized to form a single composite sample, thereby reducing small-scale heterogeneity and improving sample representativeness. Each composite sample weighed approximately 1 kg and was placed in a clean polyethylene bag. Geographic coordinates of all sampling sites were recorded using a handheld GPS device, and relevant field information was documented during sampling.

All soil samples were transported to the laboratory under sealed conditions to prevent contamination. Upon arrival, samples were air-dried at room temperature, gently crushed to break up aggregates, and sieved through a 2 mm nylon mesh to remove stones, roots, and other debris. The <2 mm fraction was subsequently ground to a fine powder (<150 μm) using an agate mortar to ensure homogeneity for subsequent chemical analysis.

Overall, this sampling strategy was designed to balance spatial coverage, representativeness, and practical feasibility, providing a robust dataset for evaluating regional patterns of soil heavy metal contamination and supporting reliable ecological risk assessment and source apportionment in the tobacco-growing areas of Zhangping County.

Determination of Total Heavy Metal Concentrations

The total concentrations of 7 heavy metals – cadmium (Cd), mercury (Hg), arsenic (As), lead (Pb), chromium (Cr), copper (Cu), and zinc (Zn) – were determined using well-established analytical protocols to ensure accuracy, reproducibility, and comparability with internationally accepted practices. These elements were selected due to their high toxicity, environmental persistence, and strong tendency to accumulate in agricultural soils, particularly in intensively managed tobacco-growing systems.

Prior to instrumental analysis, soil samples were subjected to a rigorous acid digestion procedure to achieve complete extraction of total heavy metals. For Cd, Pb, Cr, Cu, and Zn, approximately 0.5 g of finely ground soil (<150 μm) was digested using a mixed acid solution of high-purity nitric acid (HNO_3), hydrofluoric acid (HF), and perchloric acid ($HClO_4$) in Teflon vessels with a microwave-assisted digestion system (e.g., CEM MARS 6). This digestion approach is widely applied for silicate-rich soils and has been demonstrated to provide efficient total metal recovery in agricultural and environmental matrices [22-24]. For Hg and As, which are volatile and sensitive to digestion conditions, samples were digested using a microwave-assisted protocol specifically optimized for subsequent atomic fluorescence determination, as commonly adopted in soil heavy metal studies [25, 26].

After digestion, heavy metal concentrations were quantified using established instrumental techniques commonly employed in soil geochemical analysis. Cadmium and lead were determined by graphite furnace atomic absorption spectrometry (GFAAS), chromium, copper, and zinc were measured by flame atomic absorption spectrometry (FAAS), and mercury and arsenic were analyzed using atomic fluorescence spectrometry (AFS). These techniques are widely recognized for their sensitivity, accuracy, and suitability for trace-level determination of heavy metals in soils and have been extensively applied in international studies [22, 27, 28]. All measurements followed the corresponding Chinese national standard methods (GB/T and HJ series), which are technically consistent with internationally accepted analytical principles.

To ensure data quality, a strict quality assurance and quality control (QA/QC) protocol was implemented throughout the analytical process. Certified reference materials (CRMs; GSS-series soils provided by the National Research Center for Certified Reference Materials, China) were analyzed alongside each batch of samples to verify analytical accuracy. Recoveries for all analyzed metals ranged from 85% to 115%, which is within the acceptable range for soil heavy metal analysis reported in international literature [22, 27]. In addition, procedural blanks and duplicate samples were included at a frequency of 10% to monitor potential contamination and analytical precision [29, 30]. The method detection limits for all metals were well below the thresholds required for environmental assessment, ensuring the reliability of the data for subsequent pollution evaluation and ecological risk assessment.

Statistical Analysis

A comprehensive analytical framework, integrating pollution indices, ecological risk assessment, and multivariate receptor modeling, was employed to systematically evaluate the contamination characteristics, potential ecological risks, and sources of heavy metals in the tobacco-growing soils of Zhangping. All statistical computations were performed using Excel for data organization, CorelDRAW 2019 and Origin 2024 for data preprocessing and visualization, and SPSS (version 27.0) for multivariate statistical analysis. The U.S. EPA PMF (Positive Matrix Factorization) model (version 5.0) was utilized for quantitative source apportionment.

Pollution Index (P_i) and Nemerow Integrated Pollution Index (P_n)

To delineate the contamination level of individual heavy metals, the Single-factor Pollution Index (P_i) was employed. This index is defined as $P_i = C_i / S_i$, where C_i denotes the measured concentration of heavy metal i in the soil sample, and S_i denotes the corresponding background or safety threshold value for agricultural soils in Fujian Province. A P_i value greater than 1 indicates that the metal concentration exceeds the reference level, indicating a degree of contamination.

For a comprehensive assessment of the overall pollution status at each sampling site, the Nemerow Integrated Pollution Index (P_n) was applied. This index integrates the impact of all measured heavy metals and is calculated as follows:

$$P_n = \sqrt{\frac{(\max P_i)^2 + (\mathrm{avg} P_i)^2}{2}}$$

The P_n index is specifically designed to emphasize the influence of the most contaminated element, thereby providing a robust and conservative evaluation of the combined pollution level without being unduly diluted by elements with low P_i values. The calculated P_i and P_n values were interpreted using established classification criteria, as summarized in Table 1.

Table 1. Classification criteria for soil pollution levels and potential ecological risk indices.

Grade	P_i	P_n	Igeo	E^i_j	RI	Pollution/Ecological Risk Level
I	$P_i \leq 1$	$P_n \leq 0.7$	Igeo ≤ 0	$E_i < 40$	RI < 110	Unpolluted / Low potential ecological risk
II	$1 < P_i \leq 2$	$0.7 < P_n \leq 1.0$	0 < Igeo ≤ 1	$40 \leq E_i < 80$	110 ≤ RI < 220	Clean to slightly polluted / Moderate ecological risk
III	$2 < P_i \leq 3$	$1.0 < P_n \leq 2.0$	1 < Igeo ≤ 2	$80 \leq E_i < 160$	220 ≤ RI < 440	Light pollution / Considerable ecological risk
IV	$3 < P_i \leq 5$	$2.0 < P_n \leq 3.0$	2 < Igeo ≤ 3	$160 \leq E_i < 320$	440 ≤ RI < 880	Moderate pollution / High ecological risk
V	$P_i > 5$	$P_n > 3.0$	3 < Igeo ≤ 4	$E_i \geq 320$	RI ≥ 880	Severe pollution / Significantly high ecological risk

Note: P_i represents the single-factor pollution index; P_n represents the integrated pollution index; Igeo is the geo-accumulation index; E_i denotes the potential ecological risk factor of individual heavy metals; RI represents the comprehensive potential ecological risk index.

Potential Ecological Risk Index (RI)

The Potential Ecological Risk Index (RI), developed by Hakanson (1980), was applied to quantitatively assess the combined adverse effects of multiple heavy metals on the local ecosystem [31]. This method incorporates both the concentration of each heavy metal and its specific toxicological response, offering a comprehensive measure of ecological risk. The risk factor for an individual element (E_r^i) and the comprehensive Potential Ecological Risk Index (RI) are calculated using the following equations:

$$E_r^i = T_r^i \times P_i = T_r^i \times \frac{C_i}{S_i}$$

$$RI = \sum_{i=1}^{n} E_r^i$$

Here, C_i is the measured concentration of heavy metal i, S_i is its corresponding background reference value in Fujian agricultural soils, and T_r^i is the toxic response factor specific to the metal. The adopted T_r^i values, which reflect the relative toxicity and environmental sensitivity of each element, were as follows: Cd = 30, Pb = 5, As = 10, Hg = 40, Cr = 2, Cu = 5, and Zn = 1 [31, 32].

The RI index aggregates the individual risk indices (E_r^i) of all heavy metals to evaluate the overall ecological threat. Based on the classification scheme proposed by Hakanson (1980), the calculated RI values, along with (E_r^i) and Igeo values, can be categorized into different risk levels as detailed in Table 1. This multi-tiered evaluation provides a nuanced understanding of the potential ecological consequences posed by composite heavy metal contamination in the study area.

Geo-accumulation Index (Igeo)

The Geo-accumulation Index (Igeo) was used to assess the extent of heavy metal contamination in the soils by comparing current metal concentrations with their regional geochemical background values. This index serves as a reliable tool for distinguishing anthropogenic inputs from natural pedogenic processes. The Igeo is calculated as follows:

$$I_{geo} = \log_2\left(\frac{C_i}{1.5 \times B_i}\right)$$

where C_i is the measured concentration of metal i, and B_i is the geochemical background value of that metal in Fujian Province soils. The constant factor of 1.5 is introduced to account for natural variations and potential diagenetic fluctuations in the background data. The resulting Igeo values were interpreted according to the classification criteria presented in Table 1.

Multivariate Statistical Analysis for Source Apportionment

To elucidate the sources of heavy metals, a combination of multivariate statistical techniques was employed. Principal Component Analysis (PCA) was initially conducted on standardized heavy metal concentration data to reduce dimensionality and identify latent pollution sources. The application of Varimax rotation facilitated the interpretation of factors, with components exhibiting eigenvalues greater than 1 retained for further analysis. The resulting factor loadings provided preliminary insights into the types and quantities of potential pollution sources – such as industrial, agricultural, or natural origins – and guided the determination of the optimal number of factors for subsequent Positive Matrix Factorization (PMF) modeling.

PMF, an advanced receptor model, was then utilized for quantitative source apportionment. This model operates without pre-defined source profiles and resolves the contribution of various sources by decomposing the measured data matrix. The fundamental equation of PMF is defined as:

$$X_{ij} = \sum_{k=1} g_{ik} f_{kj} + e_{ij}$$

where X_{ij} is the measured concentration of metal j in sample i, g_{ik} is the contribution of source k to sample i, f_{kj} is the profile of metal j in source k, and e_{ij} is the residual error. The model iteratively minimizes the objective function Q based on measurement uncertainties. The number of factors p was determined by integrating PCA outcomes, factor interpretability, and model stability. Resolved source profiles and contributions were subsequently categorized into specific source types (e.g., agricultural practices, industrial emissions, or natural weathering).

Results and Discussion

Descriptive Statistics and Pollution Characteristics of Soil Heavy Metals

Concentration Levels of Heavy Metals

The heavy metal contents of 70 soil samples from the tobacco-growing area of Zhangping were analyzed in this study (Table 2 and Fig. 2). The average concentrations of the seven heavy metals in the soil, in descending order, were as follows: Zn (104.85 mg/kg) > Pb (47.84 mg/kg) > Cr (38.53 mg/kg) > Cu (25.25 mg/kg) > As (3.13 mg/kg) > Cd (0.26 mg/kg) > Hg (0.20 mg/kg). Notably, the maximum concentrations of all heavy metals did not exceed the risk screening values outlined in the "Soil Environmental Quality-Risk Control

Table 2. Descriptive statistics of soil physicochemical properties and heavy metal concentrations in the tobacco-growing areas of Zhangping (mg/kg).

Parameter	Min	Max	Mean	SD	CV	S_i	B_i
Pb	39.0	55.7	47.8	2.65	0.055	70.0	41.3
Cd	0.185	0.310	0.255	0.027	0.106	0.300	5.78
Hg	0.175	0.244	0.198	0.016	0.081	1.30	34.9
Cr	32.4	43.4	38.5	2.02	0.052	150	82.7
As	2.22	3.40	3.13	0.173	0.055	40.0	21.6
Cu	23.1	26.9	25.2	1.43	0.057	50	0.054
Zn	75.7	113	105	5.91	0.0558	200	0.081

Note: SD denotes Standard Deviation; CV denotes Coefficient of Variation; S_i represents the soil risk screening (safety) value; B_i represents the soil geochemical background value for Fujian Province, China.

Standard for Soil Contamination of Agricultural Land (Trial)" (GB 15618-2018). However, when compared to the soil background values for Fujian Province, Cd and Hg exhibited significant enrichment characteristics, with their average concentrations reaching 8.5 and 2.9 times the background values, respectively. The average concentrations of Pb, Cu, and Zn were 1.17, 1.26, and 1.25 times the background values, respectively, while the average concentrations of Cr and As were below the background values, at 0.82 and 0.47 times, respectively. Additionally, coefficient of variation (CV) analysis revealed that Cd (CV = 0.104) and Hg (CV = 0.082) displayed moderate spatial variability, whereas the CV for the other five heavy metals was below 0.06, indicating relatively uniform spatial distributions.

The heavy metal concentration analysis provides a basic overview of the soil environmental quality in the Zhangping tobacco-growing area. The significant enrichment of Cd and Hg relative to background values signals a clear anthropogenic influence. This finding is consistent with studies conducted in other agricultural regions of China [33]. Wang et al. (2019) and Liu et al. (2022) highlighted the widespread enrichment of Cd and Hg in intensively farmed areas in southern China [34, 35]. Specifically, in tobacco-growing regions, research by Suciu et al. (2022) and Kumar et al. (2023) identified the long-term use of phosphorus fertilizers containing Cd and historical use of mercury-containing pesticides as the primary pathways for the enrichment of these two elements [36, 37]. The differences in the behavior of various heavy metals reflect their distinct sources and environmental characteristics. The pronounced enrichment of Cd and Hg, coupled with moderate spatial variability, suggests uneven external inputs. This finding aligns with Xu et al. (2022), who noted that Cd and Hg typically exhibit high bioavailability and are more prone to human-induced alterations [38]. In contrast, the relatively stable concentrations and lower spatial variability of Pb, Cr, As, Cu, and Zn support the hypothesis that these elements are primarily derived from the parent material, consistent with the volcanic and granitic geological background of Fujian Province [39, 40]. Of particular concern is that the average concentration of Cd is approaching the national risk screening threshold (0.3 mg/kg), indicating that any continued input of Cd could lead to a qualitative shift in soil environmental quality from a clean to a polluted state. Such critical thresholds have been reported in other agricultural regions of China, underscoring the urgent need for management measures to control further Cd inputs [41, 42]. The spatial heterogeneity of Cd and Hg further complicates environmental management, suggesting that differentiated control strategies should be implemented to address hotspots.

Assessment of Soil Pollution Level Using P_i and P_n

The evaluation of soil pollution using the Single-factor Pollution Index (P_i) and the Nemerow Integrated Pollution Index (P_n) offers a systematic overview of the heavy metal contamination status in the study area (Table 3). Among the seven heavy metals, Cd exhibited the highest pollution level, with an average P_i value of 0.855, ranging from 0.717 to 1.033. Notably, the maximum P_i value for Cd exceeded 1.0, indicating slight pollution. The average P_i values for the other heavy metals were all below 1.0, ranked as follows: Pb (0.683) > Zn (0.527) > Cu (0.505) > Hg (0.153) > Cr (0.257) > As (0.079). The comprehensive pollution index, P_n, ranged from 0.578 to 0.804, with an average of 0.679.

The pollution assessment results clearly depict the overall and detailed characteristics of heavy metal contamination in the study area. Cd is identified as the primary pollutant, with its average P_i value approaching 1 and its maximum value surpassing the pollution threshold, establishing its priority in environmental management. This result is consistent with the general trends observed in agricultural soils of southern China. Zhang et al. (2023) conducted a systematic assessment of heavy metal pollution in agricultural soils of South

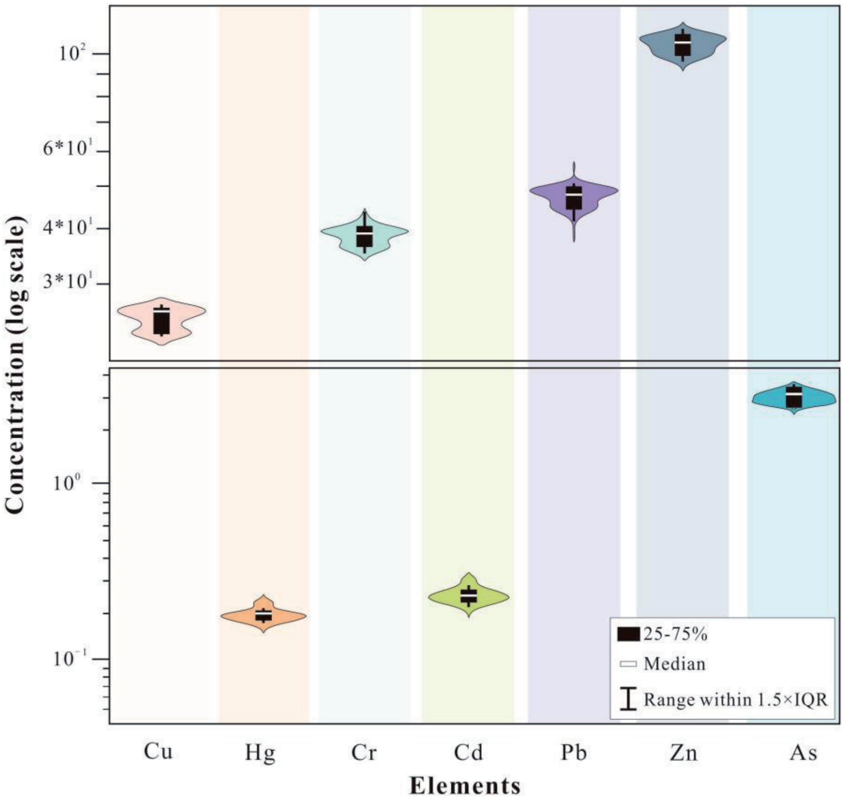

Fig. 2. Concentration levels of seven heavy metals (Pb, Cd, Hg, Cr, As, Cu, and Zn) in surface soils collected from the tobacco-growing area of Zhangping County, based on 70 soil samples.

China and found that Cd is the primary pollutant in more than 60% of the studied areas, closely linked to its high mobility and bioavailability [43]. The maximum P_i value for Cd reached 1.033, indicating that localized pollution hotspots have already emerged in the study area, requiring immediate intervention. The relatively higher P_i values for Pb, Cu, and Zn (all above 0.5), though not reaching the pollution level, show a clear accumulation trend. Similar observations were made by Zhou et al. (2024) in comparable agricultural regions, who attributed this gradual accumulation to the long-term use of agricultural inputs and the persistent impact of atmospheric deposition [44]. The consistently low P_i values for As and Cr further support their dominant natural origin.

The Nemerow Integrated Pollution Index provides an overall assessment of soil environmental quality. The average P_n value of 0.679 places the study area in the “clean” category, though it is in a “mild warning” state, a conclusion consistent with the general status of agricultural soils in the region [45, 46]. However, the maximum P_n value of 0.804, reflecting spatial heterogeneity, carries significant practical implications. It suggests that while the overall regional condition is good, certain local points are approaching the threshold for “mild pollution”. The discrepancy between the average and extreme values underscores the importance of considering both the overall trends and local anomalies in environmental assessments.

The combined analysis of P_i and P_n provides a layered understanding of the pollution status: while the overall soil quality remains acceptable, the prominent role of Cd as the primary pollutant, along with the existence of pollution hotspots, necessitates the formulation of targeted management strategies. This stratified evaluation approach offers scientific guidance for prioritizing remediation efforts and implementing differentiated control measures in the Zhangping tobacco-growing area.

Geo-accumulation Index Evaluation

The evaluation results using the Geo-accumulation Index (Igeo) (Table 3) revealed the accumulation of heavy metals in the soil. The average Igeo values for all heavy metals were below 0. Hg had the highest average Igeo value of 0.658, with a maximum value of 1.26, indicating that some sampling points reached the “uncontaminated to moderately contaminated” critical level ($0<\text{Igeo}\leq 1$). Cd had an average Igeo value of 0.277, with a maximum of 0.789, showing that some points also fell into the “uncontaminated to moderately contaminated” category.

For the other elements, the average Igeo values were negative, indicating no significant contamination. The average Igeo values for Pb, Cr, As, Cu, and Zn were -0.792, -0.557, -0.783, -1.37, and -1.56, respectively. Among these, Cu and Zn showed particularly low

Table 3. Summary of pollution and ecological risk assessment indices for heavy metals in the study area.

Index	Statistic	Pb	Cd	Hg	Cr	As	Cu	Zn
P_i	Min	0.602	0.717	0.135	0.236	0.075	0.462	0.478
	Max	0.725	1.03	0.188	0.289	0.085	0.538	0.567
	Mean	0.683	0.855	0.153	0.257	0.079	0.505	0.527
	SD	0.034	0.087	0.013	0.013	0.003	0.029	0.024
	CV	0.049	0.101	0.082	0.049	0.042	0.056	0.046
P_n	Min	0.578						
	Max	0.804						
	Mean	0.679						
	SD	0.060						
	CV	0.088						
E_i	Min	6.03	119	86	1.71	5.19	5.34	1.16
	Max	7.27	172	120	2.10	5.88	6.23	1.37
	Mean	6.85	143	98	1.87	5.45	5.84	1.27
	SD	0.338	14.4	8.08	0.093	0.231	0.330	0.059
	CV	0.049	0.100	0.082	0.049	0.042	0.056	0.046
RI	Min	230						
	Max	307						
	Mean	262						
	SD	18.4						
	CV	0.070						
Igeo	Min	-0.314	1.41	0.526	-0.808	-1.53	-0.489	-0.375
	Max	-0.046	1.94	1.01	-0.513	-1.35	-0.268	-0.130
	Mean	-0.133	1.66	0.702	-0.684	-1.46	-0.362	-0.237
	SD	0.073	0.143	0.115	0.072	0.061	0.083	0.067
	CV	-0.541	0.086	0.163	-0.104	-0.041	-0.226	-0.280

Igeo values, with minimum values of -1.70 and -2.19, indicating a more prominent background-source characteristic.

Ecological Risk Assessment

Potential Ecological Risk Assessment (RI)

The results of the Potential Ecological Risk Index (RI) assessment reveal the degree of ecological hazards posed by each heavy metal and their spatial variability [47]. As illustrated in Table 3, the individual Potential Ecological Risk Index (E_i) values for the 7 metals exhibit considerable variation. The E_i values for Cd and Hg are substantially higher than those for other metals, with average values of 143 and 98, respectively, and ranges of 119-172 and 86-120. In contrast, the average E_i values for Pb, Cr, As, Cu, and Zn are all below 40, with values of 6.85, 1.87, 5.45, 5.84, and 1.27, respectively. The combined Potential Ecological Risk Index (RI) for the study area ranges from 230 to 307, with an average of 262.

The Potential Ecological Risk Index assessment, which integrates the toxicity response factors of heavy metals, offers a more sensitive ecological risk measure compared to simple concentration analysis [48]. This study identifies Cd and Hg as the principal contributors to ecological risk in the Zhangping tobacco-growing area. This finding is consistent with ecological risk evaluations conducted in several agricultural areas in Fujian Province. Zhou Huang (2024) pointed out in a comprehensive assessment of typical agricultural areas in southern China that Cd, owing to its high toxicity coefficient and bioavailability, is often the dominant contributor to ecological risk [49]. Notably, although Hg concentrations in the study area are relatively low, its high toxicity coefficient significantly elevates its ecological risk, a phenomenon also confirmed by

Proshad et al. (2020) in their study on heavy metal risks in tobacco-growing regions [50]. The average RI value of 262 for the study area is nearing the threshold for "high risk". This contrasts sharply with the concentration-based pollution evaluation, where the average Pollution Load Index (P_n) value of 0.679 falls under the "clean" category. This disparity highlights the forward-looking relevance of the RI index in risk assessment. The difference primarily arises from the RI's consideration of the varying toxicity of heavy metals, suggesting that, even if the total concentration of heavy metals has not yet caused a decline in environmental quality, their potential ecological pressure is already significant. Zaynab et al. (2022) observed similar phenomena in ecological risk assessments, emphasizing the need for targeted management of highly toxic heavy metals [51].

The spatial distribution characteristics of ecological risk for Cd and Hg deserve further attention. The E_i value range for Cd (119-172) indicates significant spatial heterogeneity in its risk distribution, which may be linked to uneven application of fertilizers and pesticides in agricultural practices. Similarly, Hg's E_i value distribution (86-120) also shows moderate spatial variability, reflecting the complex input sources, potentially including historical atmospheric deposition and local anthropogenic activities. These findings suggest that future risk management strategies must account for the spatial variability of heavy metal ecological risks and implement differentiated control measures in response to these spatial differences.

Geo-accumulation Index (Igeo) for Source Implication

The Geo-accumulation Index (Igeo) results provide crucial evidence regarding the degree of anthropogenic influence on heavy metal contamination levels. As presented in Table 3, the Igeo values for each metal exhibit notable differentiation. Cd has the highest Igeo value, with an average of 1.66 and a range of 1.41-1.94. Hg's average Igeo value is 0.702, with values ranging from 0.526 to 1.01. The Igeo values for Pb, Cr, As, Cu, and Zn are all negative, with averages of -0.133, -0.684, -1.46, -0.362, and -0.237, respectively.

The Geo-accumulation Index effectively distinguishes between natural background levels and anthropogenic inputs by comparing current heavy metal concentrations to geochemical background values [52]. In this study, the average Igeo value for Cd was 1.66, categorizing it as "moderately polluted" and providing strong evidence for its anthropogenic sources. This finding aligns with the conclusions of Gao et al. (2021) and Senavirathna et al. (2024), who identified long-term phosphorus fertilizer applications as the primary source of Cd enrichment in agricultural soils [53, 54]. Notably, the minimum Igeo value for Cd was also 1.41, indicating that Cd is influenced by human activities throughout the study area, not just in isolated locations. Hg's Igeo value of 0.702, which falls within the "lightly polluted" category, also points to significant anthropogenic influence. This result is consistent with findings by Austruy et al. (2019), who identified atmospheric deposition and historical industrial emissions as major sources of Hg pollution in industrial areas [55]. Moreover, Hg's maximum Igeo value of 1.01 suggests that localized areas have reached a moderate pollution level, highlighting the need to focus on Hg pollution hotspots. In contrast, the negative Igeo values for Pb, Cr, As, Cu, and Zn strongly suggest that the accumulation of these metals in the study area primarily arises from natural processes, such as weathering of the parent material. This conclusion is consistent with recent findings that concentrations of Pb, Cr, As, Cu, and Zn in similar geological settings are largely controlled by parent material weathering rather than anthropogenic inputs [56].

A comparative analysis of the Geo-accumulation Index and ecological risk assessment results reveals an important environmental geochemical phenomenon: although elements like Pb, Cu, and Zn exhibit relatively high concentrations in the soil, their contribution to ecological risk is limited due to their predominant natural origins [57]. In contrast, even though Cd and Hg have lower absolute concentrations, their intense anthropogenic enrichment and high toxicity make them the primary drivers of ecological risk [58]. This insight has significant implications for regional environmental management, suggesting that limited management resources should be prioritized for heavy metals with significant anthropogenic input and high ecological risk. By integrating the results of both the ecological risk assessment and the Geo-accumulation Index analysis, a comprehensive understanding of the environmental behavior and ecological impact of heavy metals in the Zhangping tobacco-growing area has been developed. This provides a systematic scientific foundation for local soil environmental management and offers a transferable analytical framework for ecological risk control in similar agricultural regions.

Source Apportionment of Heavy Metals Using Multivariate Statistical Analysis

Correlation Analysis and Preliminary Source Identification

The correlation analysis between heavy metal elements provides crucial insights into their potential common sources. The Pearson correlation results (Fig. 3) indicate strong positive correlations between Pb, Cu, and Zn, with values of $r = 0.79$ ($p<0.01$) for Pb-Cu, $r = 0.82$ ($p<0.01$) for Pb-Zn, and $r = 0.75$ ($p<0.01$) for Cu-Zn. A significant positive correlation ($r = 0.67$, $p<0.01$) is also observed between Cd and Zn. In contrast, the correlations between Hg, As, Cr, and the other metals are generally weak or non-significant ($|r| < 0.2$). Notably, As shows a moderate negative correlation with Cd ($r = -0.49$, $p<0.01$).

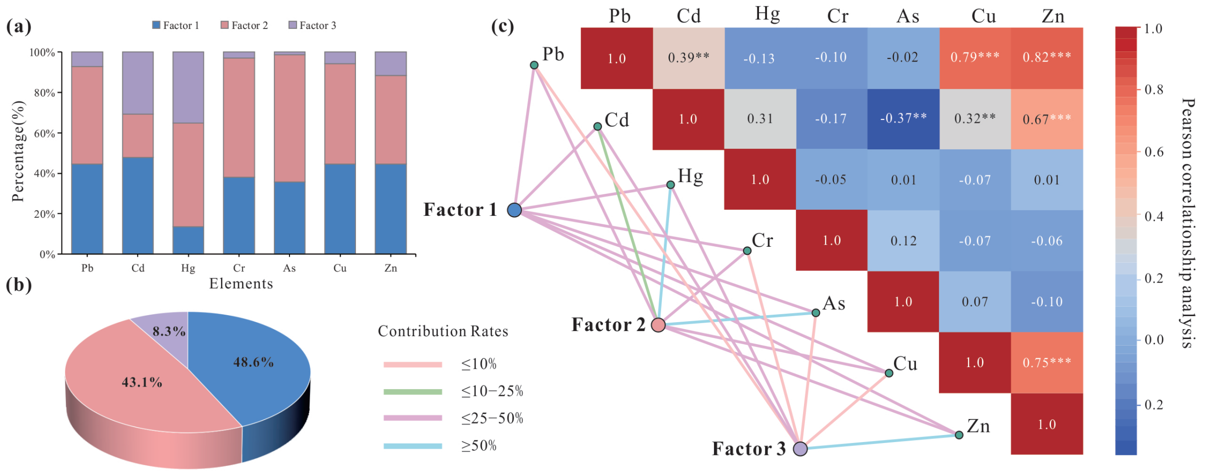

Fig. 3. Source apportionment of soil heavy metals. a) PMF factor profiles of soil heavy metals; b) percentage contribution of each PMF factor to total heavy metal concentration; c) spearman correlation between soil heavy metals and PMF factors.

Principal Component Analysis (PCA) for Source Identification

Principal Component Analysis (PCA) further extracted source-related characteristics of heavy metals [59]. Following Varimax rotation, the Kaiser-Meyer-Olkin (KMO) value was 0.75, which, according to Kaiser's criterion, is considered "acceptable" for factor analysis. Bartlett's test of sphericity was significant ($p<0.001$), strongly rejecting the null hypothesis of variable independence and confirming the suitability of factor extraction. The PCA revealed three principal components (PCs) with eigenvalues greater than 1, and the cumulative variance contribution of the three components reached 80.0% (Table 4). The first principal component (PC1) accounted for 44.4% of the variance and was highly loaded on Pb (0.924), Cu (0.873), and Zn (0.953), with a moderate positive loading on Cd (0.685). The second principal component (PC2) explained 19.5% of the variance and showed a strong positive correlation with As (0.820) and Cr (0.486) and a negative correlation with Cd (-0.523). The third principal component (PC3) explained 16.1% of the variance and was primarily driven by Hg, with a high loading of 0.852.

Quantitative Source Apportionment by Positive Matrix Factorization (PMF)

In this study, the EPA PMF 5.0 software was used for source apportionment. The final model set the factor count to 4. Based on the previous PCA analysis results and multiple basic operation tests, the number of runs was set to 25 to seek a stable solution. The final selected solution has a Q (Robust) value of 48.5,

Table 4. Result of Varimax-rotated PCA for heavy metals in soils.

Element	PCA Loadings			PMF Source Contribution (%)		
	PC1	PC2	PC3	Factor 1	Factor 2	Factor 3
Pb	0.924	0.206	-0.164	44.7%	48.2%	7.10%
Cd	0.685	-0.523	0.274	47.8%	21.6%	30.6%
Hg	0.226	-0.171	0.852	13.5%	51.3%	35.1%
Cr	-0.029	0.486	0.476	38.1%	58.9%	3.00%
As	-0.246	0.820	0.181	35.6%	63.0%	1.40%
Cu	0.873	0.286	-0.204	44.5%	49.70%	5.80%
Zn	0.953	0.181	0.008	44.5%	44.0%	11.6%
Eigenvalue	3.11	1.37	1.13	-	-	-
Variance contribution rate (%)	44.4	19.5	16.1	-	-	-
Cumulative variance (%)	44.4	63.9	80.0	-	-	-
Total contribution (%)	-	-	-	43.1%	48.6%	8.30%

and the fluctuation range of the Q (Robust) value between multiple runs is less than 2%, indicating that the model fits well and the results are stable and reliable [60, 61]. The PMF model quantitatively identified four pollution sources (Table 4). Factor 1, which contributed the most (32%), had a component spectrum dominated by Pb (40%), Zn (36%), and Cr (33%), consistent with PC1 in PCA. Factor 2 was the second largest contributor (30%), with Cd (58%) and Cu (45%) as its main characteristic elements, corresponding to PC2. Factor 3 contributed 23%, with As (48%) as the major element. Factor 4, contributing 15%, was most strongly characterized by Hg, which accounted for 70%, aligning with the result from PCA where Hg independently formed PC3. The PMF model further corroborated the findings from correlation analysis and PCA, providing a quantitative estimate of each source's contribution.

Comparative Discussion and Synthesis of Source Apportionment Results

The comprehensive comparison of multivariate statistical methods revealed consistent source characteristics of heavy metals in the studied soils [62, 63]. PCA and PMF demonstrated remarkable consistency in source identification. PC1 (44.4% variance) from PCA showed clear correspondence with F1 (43.1% contribution) from PMF, both highlighting the significance of Pb, Cu, Zn, and Cd. The information carried by PC2 (19.5% variance) and PC3 (16.1% variance) in PCA was integrated into F2 (48.6% contribution) in the PMF model, which showed predominant contributions to Cr, As, and Hg. PMF additionally resolved a separate minor factor F3 (8.3% contribution) with specific affinity for Cd and Hg.

As synthesized in Fig. 3, three main sources were identified: industrial-traffic emissions (43.1%), predominantly contributing to Pb (44.7%), Cu (44.5%), Zn (44.5%), and Cd (47.8%); a natural pedogenic and historical agricultural mixed source (48.6%), mainly contributing to Cr (58.9%), As (63.0%), and Hg (51.3%); and a specific agricultural source (8.3%), primarily contributing to Cd (30.6%) and Hg (35.1%).

The integrated comparison and synthesis of source apportionment results consolidated evidence from multiple statistical approaches into a coherent understanding of heavy metal origins. The strong agreement between PCA and PMF outputs validated the reliability of source identification. The correspondence between PC1 and F1 firmly established the importance of industrial-traffic emissions in the study area, consistent with observations in other agricultural regions of southeastern China [64]. The notably high contribution of Cd to this source (47.8%) suggests potential Cd emissions associated with industrial activities, a phenomenon also detected in industrial perimeter studies [65]. The merging of PCA's PC2 (natural/agricultural) and PC3 (Hg-specific) into a single mixed source (F2) in PMF has substantial environmental rationale. Statistically, PMF as an uncertainty-weighted receptor model tends to combine sources with similar spatial distributions or strong collinearity [66]. Geochemically, the spatial patterns of pedogenically derived As and Cr may have overlapped with those from historical agricultural applications of As-containing pesticides and Hg-based fungicides over a prolonged cultivation history. This interpretation aligns with findings from historical agricultural regions [67], where long-term anthropogenic disturbance often blurs the boundaries between natural and agricultural sources. The successful identification of F3 (specific agricultural source) by PMF highlights its advantage in resolving subtle source variations. This minor source with specific Cd and Hg contributions likely relates to particular agrochemical applications. Similar phenomena were observed in agricultural source studies [68], where variations in Cd and Hg impurity levels among different phosphate fertilizers and organic amendments could generate such specific contribution patterns. This finding carries significant implications for precision agriculture management, suggesting that optimizing agricultural input selection could effectively reduce Cd and Hg inputs.

Linking source apportionment results with earlier pollution and ecological risk assessments constructs a more comprehensive environmental understanding. The substantial contribution from industrial-traffic emissions (43.1%) explains the widespread yet sub-threshold contamination status of Pb, Cu, and Zn. The dominance of a natural pedogenic and historical agricultural mixed source (48.6%) clarifies why Cr and As maintain relatively low pollution levels despite significant anthropogenic influence. Particularly noteworthy is the multi-source characteristic of Cd and Hg, with significant contributions from multiple pathways, explaining their role as primary drivers of ecological risk.

Methodologically, this research demonstrates the effectiveness and complementarity of multivariate statistical approaches in heavy metal source apportionment. PCA provides superior capabilities in factor identification and preliminary interpretation, while PMF offers enhanced quantitative resolution and minor source detection. The integrated application of both methods yields more comprehensive and reliable results for source apportionment in complex environmental systems.

Although the combined application of PCA and PMF provides a robust framework for heavy metal source apportionment, several inherent limitations should be acknowledged when interpreting the results [69]. PCA is sensitive to data standardization and primarily serves as a qualitative exploratory tool; it may merge pollution sources with similar geochemical characteristics and does not provide quantitative source contributions. PMF, while capable of quantitative apportionment, is influenced by uncertainty estimation, factor number selection, and potential collinearity among sources.

In complex soil systems, particularly in long-term cultivated agricultural regions, natural pedogenic inputs and historical anthropogenic activities may exhibit overlapping spatial and geochemical signatures, which can complicate source separation.

To minimize these limitations, several strategies were adopted in this study. All data were standardized before PCA to reduce scale-related bias, and PCA was used primarily to guide factor selection and preliminary source interpretation rather than as a standalone apportionment tool. PMF modeling was conducted using uncertainty-weighted inputs, and the optimal number of factors was determined through a combination of PCA results, Q-value diagnostics, model stability tests, and source interpretability. The strong consistency observed between PCA-derived source patterns and PMF-resolved source contributions further supports the robustness of the apportionment results. Although the limitations of PCA and PMF cannot be fully eliminated, their integrated application, together with careful model optimization and results cross-validation, effectively enhances the reliability of source identification in complex agricultural soil systems.

Conclusions

This study systematically assessed the contamination characteristics, ecological risks, and sources of heavy metals in soils from the tobacco-growing area of Zhangping County, Fujian Province. Although the overall concentrations of heavy metals remained below national risk screening values, cadmium (Cd) and mercury (Hg) exhibited significant anthropogenic enrichment and were identified as the dominant contributors to potential ecological risk.

Quantitative source apportionment using the PMF model revealed that industrial and traffic-related emissions accounted for 43.1% of total heavy metal inputs, primarily contributing Pb, Cu, Zn, and part of Cd; a mixed source of natural pedogenesis and historical agricultural activities contributed 48.6%, dominating Cr, As, and Hg inputs; while a specific agricultural source contributed 8.3%, with notable inputs of Cd and Hg. These results indicate that ecological risks in tobacco-growing soils are driven less by overall metal accumulation than by targeted inputs of highly toxic elements through specific anthropogenic pathways.

From a broader perspective, this finding has important implications for soil pollution management in tobacco-growing regions. It highlights the necessity of shifting from uniform control strategies to source-oriented and element-specific management approaches, with particular emphasis on Cd and Hg. In practice, priority should be given to reducing Cd inputs from agricultural practices by optimizing fertilizer application, promoting low-Cd phosphate fertilizers, and improving nutrient management efficiency. At the same time, the contribution of atmospheric deposition associated with industrial and traffic activities suggests that strengthened regional emission control and continuous monitoring of atmospheric inputs are also essential. In addition, long-term mitigation strategies should focus on maintaining soil quality and reducing metal bioavailability through appropriate soil amendments (e.g., biochar or liming) and regular soil monitoring. Overall, the integrated application of geochemical indices and receptor models in this study not only provides a robust assessment of ecological risk and source contributions but also offers a methodological framework and practical reference for preventing and mitigating soil heavy metal pollution in tobacco-growing areas and other intensive agricultural ecosystems.

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Conflicts of Interest:

The authors declare no conflict of interest.

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