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Assessment of Heavy Metals in Tea Plantation Soil and Their Uptake by Tieguanyin Tea Leaves and Potential Health Risk Assessment in Anxi County in Southeast China

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Abstract: Evaluating heavy metal pollution in tea plantation soil and conducting potential health risk assessments are crucial for ensuring the safety of tea consumers. However, soil heavy metal pollution levels and dietary exposure risk remain poorly understood, and there is no consensus on how soil physicochemical properties affect heavy metal concentrations. In this study, seventy-three soil samples and corresponding tea leaves from main tea-producing regions in Anxi County were analyzed for arsenic (As), chromium (Cr), lead (Pb) and copper (Cu) concentrations. The results showed that mean concentrations of As, Cr, Cu and Pb in the soil did not exceed respective risk screening values in China (GB 15618-2018). The concentrations of As, Cr, Cu and Pb in the tea leaves were within limiting values of the Chinese National Food Safety Standard, and the bioaccumulation factor of heavy metals in descending order was Cu > Pb > As > Cr. The hazard index values of heavy metals indicated no potential human health risk. Soil pH, EAl, EA and AP were the main controlling factors for heavy metal in soil and tea leaves. Cu and Pb concentrations in tea leaves were positively correlated with soil Cu and Pb concentrations. These results provide a scientific basis for effective monitoring and management in tea plantations and for controlling potential risks in tea leaves.

Keywords: heavy metal; soil pollution; Tieguanyin tea; transfer; health risk assessment

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1. Introduction

Tea plants (*Camellia sinensis*) are mainly cultivated in 45 countries worldwide, such as China, India, Turkey, Sri Lanka and Kenya [1], and they have been cultivated in China for over 2000 years [2], accounting for 60% of global tea acreage [3]. Tea, made from the leaves of tea plants, is one of the most popular non-alcoholic beverages [4]. Tea contains abundant caffeine, amino acids and polyphenols, and it can exert anti-inflammatory and antioxidant effects, which reduce serum cholesterol and lipoprotein oxidation [1,2]. Many trace elements such as Zn and Se also accumulate in tea, so drinking tea benefits human health by decreasing the risk of cardiovascular disease and diabetes [1,4]. However, tea may also contain some hazardous components, the actual effect of tea on human health is still controversial [5] and intake of excessive essential trace elements and very small amounts of nonessential heavy metals can cause deleterious effects [6,7]. Therefore, heavy metals in tea leaves and tea safety have attracted widespread social attention over time.

The safety and health of tea cannot be separated from soil environment quality in tea plantations. Soil heavy metals pollution in agricultural soils is also a very important issue [8] as petrochemical, mining and industrial discharges increasingly contaminate environments [9]. Excessive heavy metals can change soil physicochemical properties, reduce soil productivity and decrease the quantity and quality of tea, causing a long-term toxic impact on human health via entering and transferring along food chains due to their toxicity, persistency and non-biodegradability [10,11]. Arsenic (As), chromium (Cr) and lead (Pb) exposure have been related to many diseases, including cardiovascular and nerve system conditions, and as an essential trace element, excessive copper (Cu) concentrations also affect human health [12]. Several calculation methods of pollution indices have been used to assess soil heavy metals pollution risk; the Nemerow composite index (P) and potential ecological risk index (RI) were frequently applied to reveal heavy metal pollution levels [8,13]. The latest nationwide survey on soil contamination status in China showed that the exceedance of As, Cr, Cu and Pb were 2.7%, 1.1%, 2.1% and 1.5%, respectively, according to the environmental quality standard set by the Ministry of Environmental Protection of China [10]. Peng et al. found that the proportion of the comprehensive pollution degree reaching the alert and light pollution levels were 11.11% and 8.89%, respectively, in the Liu Bao tea production area [14]. Cui et al. pointed out that soils were slightly contaminated with Pb and potential ecological risk was low in Yangtze River Delta [15]. Previous studies also have demonstrated that the risk levels of soil heavy metals were categorized as “medium” and “heavy” levels using overall RI value at tea plantations in Wuyishan [16]. Xie et al. studied heavy metal concentrations in typical tea gardens soil in Yunnan Province, and they found that soils were all lightly polluted and had good soil quality [17]. Soil heavy metals concentrations can affect heavy metals concentrations in tea leaves; in this context, it is of great importance to accurately estimate heavy metal concentrations in the assessment of soil pollution level and potential risk.

Concentrations of heavy metals in tea are very important indicators in tea quality evaluation, because they can be transferred into tea infusions and enter the human body [18]. The standard limit values for concentrations of heavy metals in tea were proposed by the national food safety standard (GB 2762-2017) [19] and the Ministry of Agriculture tea heavy metals limited standards (NY 659-2003, NY/T 288-2012) in China [20,21]. Several studies have reported on the concentrations of heavy metals in tea leaves and possible health risks from elements such as As, Cr and Pb, as well as the phenomena of exceeding the standard limits in some areas especially affected by human activities [22–24]. Grządka et al. pointed out that heavy metal concentrations of tested tea samples imported to Poland were below the limits specified in the regulations issued in their countries of origin [5]. The bioaccumulation factor has been used to analyze the transfer ability of heavy metals from soil to plants, but the response patterns could be different depending on the specific heavy metal, plant species and soil type [4]. For example, previous studies showed that the enrichment ability was Cu > As > Cr > Pb in the tea-producing area in the Qionglai mountains [25], while the bioaccumulation factor values decreased in the order Cu > As ≈ Pb > Cr in Puan County, Guizhou Province [18]. Humans may be exposed to heavy metals through tea intake, resulting in potential health risks [2]. Previous studies have reported that a dose–response relationship was found between tea intake and blood heavy metal concentration [2]. Therefore, the transfer ability of heavy metals to tea leaves from soils and the health risks induced by tea intake need attention.

Numerous studies have demonstrated that heavy metal pollution depends primarily on industrial activities and agricultural activities such as the application of pesticides and chemical fertilizers containing heavy metals [26]. Soil physicochemical properties can affect the heavy metal concentration of soil and tea leaves [4,24]. Soil heavy metal concentrations were correlated with soil texture and soil particle diameter [27], soil H^+ activity [28], soil SOM [29] and pH [4]. The soils of tea plantations become acidified with the increase of tea plants, which contributes to elevated concentrations of bioavailable water-soluble and exchangeable metals [21]. Soil pH is thought of as a key influencing factor in the transfer and accumulation of heavy metals from soil to plant [1]. The Cu concentration in tea leaves was found to be closely correlated with the soil’s H^+ activity [30]. Previous studies have showed that Cr and Pb concentrations in tea leaves were negatively correlated with soil Cr and Pb concentrations, respectively [18], which showed that Pb and Cr in soil will inhibit absorption by tea leaves. Tea leaf Cu and As concentrations showed no significant relation to soil Cu and As concentrations, suggesting that soil was not the only source of tea leaf Cu and As [18]. Zhang et al. found that soil acidification and dissolved organic matter play vital roles in tea leaf heavy metal concentrations [31]. Moreover, Jiang et al. found that no significant relationship between tea leaf heavy metal concentration and soil pH and SOM [32]. These inconsistent results demonstrate that the relationships between soil and tea leaf heavy metal concentrations are still an unaddressed issue and need to be studied further.

Fujian Province is a major center for tea production and export in China. Anxi County, known as "the hometown of China's Oolong tea", is the birthplace of the famous Tieguanyin tea and ranks first among China's key tea-producing counties [33,34]. The tea industry is a dominant agricultural sector crucial for local development, making it essential to maintain soil health and tea quality free from heavy metal contamination. However, most studies have focused either on soil heavy metal concentrations or on heavy metal content in tea leaves, with few addressing both simultaneously. Furthermore, the potential transfer ability of heavy metals from soil to plants and the associated human health risks require further investigation.

In this study, we surveyed heavy metal concentrations in soil and paired tea leaves, as well as soil physicochemical properties, in the main tea-producing areas of Anxi County. The main objectives were to: (1) quantify As, Cr, Cu and Pb concentrations in different soil depth and corresponding tea leaves in major tea plantations in southeast China; (2) analyze the influence of soil physicochemical properties on heavy metal concentrations in soil and tea leaves; and (3) evaluate soil pollution levels and potential risk to human health from heavy metals in tea leaves. Our results provide scientific and comprehensive knowledge for evaluating heavy metal pollution in tea plantations and the associated human health risks. This information can aid in formulating agricultural soil management strategies and controlling heavy metal pollution in tea plantations.

2. Materials and Methods

2.1. Study Area and Field Sampling

The study was conducted within Anxi County (117°36′–118°17′ E, 24°50′–25°26′ N) in southeastern Fujian Province (southeast China). This study area is located in a subtropical oceanic monsoon climate zone and is dominated by mountains and hills. With consideration for the distribution of tea plantations, 73 research sites in nine main growing areas towns were selected, including Futian (FD, n = 11), Gande (GD, n = 20), Hushang (HS, n = 3), Kuidou (KD, n = 4), Longjuan (LJ, n = 4), Taozhou (TZ, n = 5), Xiping (XP, n = 7), Xianghua (XH, n = 13) and Changkeng (CK, n = 6) (Figure S1). A sampling area of 50 m^2 was designated in the central position of the selected tea plantation in each site, and nine soil cores from the upper 20 cm and 20–40 cm soil layers were collected using an auger and mixed to form one soil sample in the same layer. At the same time, five hundred grams of tea leaves with one bud and three leaves based on picking standard of oolong tea were collected and mixed to form one tea sample in each tea plantation. After removing roots and stones, soil samples were air-dried, then sieved through a 2 mm mesh sieve. The fresh tea leaves were blown at 105 °C for 30 min, then dried at 80 °C for the analysis.

2.2. Measurement of Physicochemical Properties and Heavy Metals

Soil pH was measured for a 1:2.5 (w/v) soil:water mixture with a glass electrode meter. Soil organic matter (SOM) was determined using an external heating method with potassium dichromate and sulfuric acid. Soil total N (TN) concentrations were determined using an elemental analyzer (Elementar Vario EL III; Elementar, Langenselbod, Germany). The soil total P (TP) concentrations were determined by using the molybdenum–antimony anti-colorimetric method with a continuous flow analytic system (Skalar San++, Breda, Netherlands). Available P (AP) concentration was determined by extraction with $NaHCO_3$ and the molybdenum–antimony resistance coloration method. Soil available potassium

(AK) was determined using a flame photometer after extracting the soil samples with CH_3COONH_4. Soil alkaline hydrolyzable nitrogen was determined using the method as described in Bao [35]. Soil exchangeable aluminum, exchangeable acidity and exchangeable hydrogen were extracted in 1 M KCl [35,36]. The total metal concentrations of soil and tea leaves samples were measured using inductively coupled plasma atomic emission spectrometry (ICP-AES, Thermo Jarrell Ash Ltd., Corona, CA, USA) and inductively coupled plasma mass spectrometry (ICP-MS, Thermo Fisher XSeries II, Waltham, MA, USA). Briefly, 0.2 g soil sample was accurately weighed, then 5 mL HNO_3, 1 mL $HClO_4$ and 2 mL hydrogen fluoride (HF) were sequentially added to the Teflon tubes. To the temperature procedure was 110 °C for 30 min, 140 °C for 30 min, and finally 180 °C until the samples were digested completely. After digestion, the samples were diluted to 50 mL in a 50 mL volumetric flack, and the heavy metal concentration was determined [37]. After 0.2 g tea leaves sample was accurately weighed, 5 mL HNO_3 was added into a Teflon digestion vessel and digested for 40 min at a power of 1600 W and temperature of 180 °C. The digested solution was then heated and transferred to volumetric flasks, then brought up to volume with 0.2% HNO_3 solution. According to method described in Ma et al. [38], the optimized working conditions for analysis were as follows: the radio frequency powder was 1.3 KW, the carrier gas flow rate was 1.12 L/min and the auxiliary gas flow rate was 0.87 L/min. The oxide ratio of CeO+/Ce+ was $\leq$0.5% and the double charge indices of Ba++/Ba+ was $\leq$2.0%. Quality assurance (QA) and quality control (QC) measures were followed in the experimental process to ensure the accuracy of results. The standard solution was the multielement atomic spectroscopy standard solution V, and all of the determination coefficients of standard curves were higher than 0.99. All samples were selected for parallel processing, the certified reference material (CRM) of green tea (GBW10052) was analyzed to investigate the stability of instrumental response and the relative standard deviation was less than 10%. The recoveries of the studied elements were higher than 90%, suggesting that the accuracy and precision were acceptable.

2.3. Bioaccumulation Factor

To estimate heavy metal transfer from soil to tea leaves, bioaccumulation factor (BAF) was calculated through the following equation:

$$\mathrm{BAF} = \mathrm{C_{plant}}/\mathrm{C_{soil}} \tag{1}$$

where C_{plant} is the individual heavy metal concentration in tea leaves (mg kg^{-1}) and C_{soil} is the corresponding soil heavy metal concentration (mg kg^{-1}). The greater the BAF value, the stronger the migration ability of tea leaves to enrich and absorb heavy metal in soil. BAF > 1 indicates that tea leaves are accumulators for the heavy metals being analyzed [39].

2.4. Soil Pollution Quantification Assessment

2.4.1. Nemerow Composite Index Method

The Nemerow index was evaluated as the pollution level of heavy metal; the formulas are as follows:

$$\mathrm{P_i} = \mathrm{C_s}/\mathrm{C_m} \tag{2}$$

$$\mathrm{P} = \sqrt{\left(Pi\ mean^2 + Pi\ max^2\right)/2} \tag{3}$$

where P_i represents the single factor pollution index, C_s represents soil heavy metal concentration, C_m is the concentration threshold of heavy metals and their risk screening value were set at 40, 150, 50 and 70 mg/kg for As, Cr, Cu and Pb, respectively, in agricultural soil of China (GB 15618-2018) [40]. P represents the Nemerow pollution index. The p-value is classified according to the following ranges: $p \leq 0.7$, indicates safe; $0.7 < p \leq 1.0$, for alert level; $1.0 < p \leq 2.0$ indicates minor pollution; $2.0 < p \leq 3.0$, moderate pollution; and $p > 3.0$ indicates severe pollution [41,42].

2.4.2. Potential Ecological Risk Assessment

The potential ecological risk index (RI) is often used to assess potential risks level of heavy metal pollution combining soil environment and toxicology [43]. It was calculated by the following equation:

$$E_i = T_i \times C_s / C_n \quad (4)$$

$$RI = E_{As} + E_{Cr} + E_{Cu} + E_{Pb} \quad (5)$$

where E_i is the potential ecological risk index of each heavy metal, T_i is the toxicity coefficient of each metal and the following T_i values are used: A_S = 10, Cr = 2, Pb = 5 and Cu = 5 [11]. C_s represents soil heavy metal concentration and C_n is the background value of each metal from Fujian Province [16]. RI is the sum of the potential ecological risk indices for all selected heavy metal. The RI is classified into four categories: RI <110 indicates low risk, $110 \leq RI < 220$ indicates moderate risk, $220 \leq RI < 440$ indicates considerable risk and $RI \geq 440$ indicates very high risk [18].

2.5. Health Risk Assessment

The noncarcinogenic risk for human exposure to heavy metals was assessed based on the target hazard quotient (THQ), which was proposed by the United States Environmental Protection Agency [44]. The hazard index (HI) is the equal to the sum of each THQ value to estimate the total noncarcinogenic health hazard. The calculation formulas of THQ and HI are as follows:

$$THQ = (C_i \times IR \times TR_i)/(B_W \times RfD_i \times 1000) \quad (6)$$

$$HI = \sum\nolimits_{n=1}^{i} THQi \quad (7)$$

where C_i is tea leaf heavy metal concentration (mg kg^{-1}), IR is ingestion rate of tea drinking, which was selected as 11.4 g $person^{-1}$ day^{-1} [45], TR_i is the transfer ratio of the heavy metal from tea leaves to tea infusion [4], B_W is body weight, namely 70 kg for adults [46], RfD_i (mg kg^{-1} bw d^{-1}) is the oral intake amount of heavy metal (i) proposed by USEPA, RfD_i value for As, Cr, Cu and Pb were 0.0003, 1.5, 0.04 and 0.0015 mg kg^{-1} bw d^{-1} [18,24] and THQ_i is target hazard quotient of heavy metal i. THQ values <1 indicate no health effects, while values >1 suggest a potential risk of such effects, with the higher the value, the higher the risk. HI value <1, HI value >1 and HI value >10 indicate a low likelihood of adverse health effects, negative effects on human health and chronic toxicity risk, respectively.

2.6. Statistical Analysis

One-way analysis of variance (ANOVA) was conducted to identify significant differences in soil physicochemical properties, soil heavy metal concentration and tea leaf heavy metal concentration among different regions. Significance was set at $p < 0.05$. To further assess the effects of soil physicochemical properties on soil heavy metal concentration and tea leaf heavy metal concentration, correlation analysis and redundancy analysis (RDA) were performed to evaluate the relationships between soil physicochemical properties and soil and tea leaf heavy metal concentrations. The correlations between soil and tea leaf heavy metal concentrations were examined by Pearson's correlation analysis. RDA analysis was performed using CANOCO 5.0 software (Ithaca, NY, USA) and correlation analysis and figures were drawn using OriginPro software (version 2024, Origin Laboratories, Northampton, MA, USA). In addition, the predictors of heavy metal concentration were determined using random forest analysis with the "rfPermute" package in R 4.4.1.

3. Results

3.1. Soil Physicochemical Properties

The average soil pH of the topsoil in nine regions was KD (3.74) < XH (3.88) < TZ (3.93) < FT (4.00) < CK (4.09) < GD (4.14) < XP (4.23) < HS (4.23) < LJ (4.63), indicating severe soil acidification. LJ had a significantly higher soil pH value in the 0–20 cm soil layer than other regions. SOM in topsoil showed significantly higher values in HS and significantly lower values in FT relative to other regions ($p < 0.05$). EA and EAl in topsoil showed significantly higher values in TZ and significantly lower values in LJ than in other regions ($p < 0.05$). XH had a significantly higher EH and AP in topsoil than the values seen in LJ and XP ($p < 0.05$). No significant differences in TN and TP were found in the topsoil of distinct regions. AN in HS was significantly higher than that in CK (2.025 times) and in LJ (2.279 times), and AK in topsoil was significantly higher in KD than in other regions ($p < 0.05$). In the 20–40 cm soil layer, there were no significant differences in pH value, SOM, EA, EAl and TP EH amongst nine regions; TN and AN in HS were markedly higher than the values seen in TZ and LJ, respectively ($p < 0.05$). KD had significantly higher AK values compared to other regions except for HS ($p < 0.05$) (Table 1).

Table 1. Soil physicochemical characteristics in different soil layer at main tea-producing regions in Anxi County.

Regions	Soil Layer	pH	SOM (g/kg)	EA (cmol/kg)	EAl (cmol/kg)	EH (cmol/kg)
FT	0–20	4.00 ± 0.04 b	21.09 ± 1.75 b	5.89 ± 0.69 ab	5.47 ± 0.68 ab	0.41 ± 0.03 ab
	20–40	4.07 ± 0.05 a	17.22 ± 1.43 a	5.65 ± 0.72 a	5.29 ± 0.72 a	0.36 ± 0.03 a
GD	0–20	4.14 ± 0.07 b	29.06 ± 2.23 ab	5.50 ± 0.62 ab	5.15 ± 0.62 ab	0.35 ± 0.02 ab
	20–40	4.20 ± 0.09 a	22.48 ± 2.18 a	5.23 ± 0.64 a	4.89 ± 0.64 a	0.34 ± 0.03 a
HS	0–20	4.23 ± 0.02 ab	35.96 ± 1.59 a	3.54 ± 0.15 ab	3.09 ± 0.15 ab	0.45 ± 0.17 ab
	20–40	4.19 ± 0.00 a	27.90 ± 3.95 a	3.99 ± 0.52 a	3.62 ± 0.55 a	0.36 ± 0.04 a
KD	0–20	3.74 ± 0.12 b	26.46 ± 2.00 ab	6.56 ± 0.52 ab	6.13 ± 0.44 ab	0.43 ± 0.09 ab
	20–40	3.86 ± 0.09 a	19.42 ± 2.93 a	6.09 ± 0.43 a	5.66 ± 0.43 a	0.43 ± 0.03 a
LJ	0–20	4.63 ± 0.43 a	22.66 ± 1.86 ab	2.87 ± 0.84 b	2.63 ± 0.77 b	0.24 ± 0.06 b
	20–40	4.41 ± 0.26 a	15.20 ± 1.43 a	2.85 ± 0.77 a	2.59 ± 0.70 a	0.26 ± 0.06 a
TZ	0–20	3.93 ± 0.07 b	23.36 ± 1.16 ab	7.81 ± 2.01 a	7.38 ± 1.98 a	0.43 ± 0.02 ab
	20–40	4.08 ± 0.13 a	13.84 ± 2.67 a	7.42 ± 2.35 a	7.112 ± 2.35 a	0.31 ± 0.01 a
XP	0–20	4.22 ± 0.11 ab	27.59 ± 2.27 ab	3.99 ± 0.32 ab	3.73 ± 0.30 ab	0.26 ± 0.02 b
	20–40	4.31 ± 0.11 a	23.16 ± 1.66 a	3.350 ± 0.59 a	3.08 ± 0.57 a	0.26 ± 0.05 a
XH	0–20	3.88 ± 0.03 b	31.18 ± 2.14 ab	6.85 ± 0.65 ab	6.34 ± 0.63 ab	0.50 ± 0.04 a
	20–40	3.95 ± 0.05 a	24.05 ± 2.47 a	6.31 ± 0.55 a	5.94 ± 0.55 a	0.36 ± 0.03 a
CK	0–20	4.09 ± 0.09 b	23.66 ± 3.93 ab	5.34 ± 0.65 ab	4.98 ± 0.61 ab	0.36 ± 0.05 ab
	20–40	4.06 ± 0.06 a	19.85 ± 2.90 a	5.10 ± 0.76 a	4.74 ± 0.74 a	0.36 ± 0.03 a
Regions	**Soil layer**	**TN (g/kg)**	**AN (mg/kg)**	**TP (mg/kg)**	**AP (mg/kg)**	**AK (mg/kg)**
FT	0–20	0.97 ± 0.05 a	121.47 ± 21.00 ab	40.55 ± 41.074 a	194.73 ± 110.20 ab	69.74 ± 5.75 b
	20–40	0.79 ± 0.04 ab	78.39 ± 8.55 ab	199.66 ± 36.88 a	75.35 ± 16.72 ab	59.58 ± 5.73 b
GD	0–20	1.17 ± 0.08 a	99.14 ± 8.55 ab	433.02 ± 139.93 a	166.76 ± 121.46 bc	87.18 ± 6.76 b
	20–40	0.83 ± 0.07 ab	74.49 ± 7.39 ab	268.55 ± 116.58 a	86.24 ± 26.51 ab	72.15 ± 5.36 b
HS	0–20	1.42 ± 0.09 a	161.37 ± 52.27 a	427.51 ± 261.44 a	88.53 ± 59.00 bc	98.86 ± 7.40 b
	20–40	1.08 ± 0.18 a	91.29 ± 8.53 a	270.53 ± 198.68 a	52.72 ± 36.16 ab	99.51 ± 14.96 ab
KD	0–20	1.07 ± 0.11 a	100.71 ± 13.93 ab	325.77 ± 141.46 a	203.16 ± 227.23 ab	136.58 ± 16.65 a
	20–40	0.84 ± 0.15 ab	87.99 ± 15.31 ab	224.45 ± 68.48 a	56.73 ± 30.52 ab	126.47 ± 25.63 a
LJ	0–20	1.05 ± 0.01 a	70.79 ± 5.61 b	100.37 ± 42.82 a	118.35 ± 65.52 bc	97.09 ± 6.01 b
	20–40	0.67 ± 0.10 ab	41.19 ± 9.41 b	107.91 ± 60.21 a	25.98 ± 8.27 b	87.4 ± 4.03 b
TZ	0–20	1.02 ± 0.02 a	99.25 ± 6.94 ab	205.86 ± 49.65 a	233.87 ± 138.77 ab	90.26 ± 16.41 b
	20–40	0.56 ± 0.10 b	52.73 ± 9.13 ab	129.35 ± 67.80 a	57.61 ± 15.64 ab	70.62 ± 12.64 b
XP	0–20	1.233 ± 0.12 a	104.86 ± 8.65 ab	279.74 ± 70.23 a	42.88 ± 31.71 c	68.14 ± 11.50 b
	20–40	0.85 ± 0.11 ab	81.87 ± 6.08 ab	218.89 ± 58.34 a	15.25 ± 5.30 b	56.0364 ± 9.97 b
XH	0–20	1.23 ± 0.09 a	115.68 ± 9.52 ab	511.36 ± 96.49 a	288.93 ± 185.92 a	86.77 ± 4.28 b
	20–40	0.91 ± 0.07 ab	81.52 ± 5.82 ab	329.75 ± 53.51 a	148.74 ± 35.57 a	82.32 ± 5.05 b
CK	0–20	0.99 ± 0.15 a	79.67 ± 14.41 b	233.76 ± 79.32 a	245.72 ± 228.46 ab	84.74 ± 10.26 b
	20–40	0.69 ± 0.12 ab	58.07 ± 9.50 ab	92.27 ± 42.86 a	110.84 ± 48.41 ab	62.91 ± 8.31 b

FT, Futian; GD, Gande; HS, Hushang; KD, Kuidou; LJ, Longjuan; TZ, Taozhou; XP, Xiping; XH, Xianghua; CK, Changkeng. SOM, soil organic matter; EA, soil exchangeable acidity; EAl, soil exchangeable aluminum; EH, exchangeable hydrogen; TN, total N nitrogen concentration; AN, soil alkaline hydrolyzable nitrogen; AK, soil available potassium. Different letters indicate significant differences ($p < 0.05$) among different regions. Values are mean ± standard error.

3.2. Soil Heavy Metals and Pollution Evaluation

The mean total concentration of As, Cr, Cu and Pb in topsoil were 18.754 ± 1.295, 42.796 ± 2.806, 11.478 ± 0.782 and 40.544 ± 3.807 mg/kg, respectively; and they were 20.700 ± 1.443, 43.269 ± 2.995, 11.263 ± 0.891 and 40.232 ± 3.662 mg/kg in the 20–40 cm soil layer, respectively. The highest topsoil As, Cr and Cu concentrations values were observed in LJ, the lowest As concentration appeared in KD and the lowest Cr and Cu concentrations both appeared in TZ. It can be seen that topsoil Pb concentration in HS was significantly greater than that in LJ ($p < 0.05$). In the 20–40 cm soil layer, soil As concentration in XH was significantly greater than that in KD and TZ ($p < 0.05$), soil Cu concentration in LJ was significantly greater than that in TZ ($p < 0.05$) and no significant difference was found in Cr and Pb concentration among different regions (Figure 1).

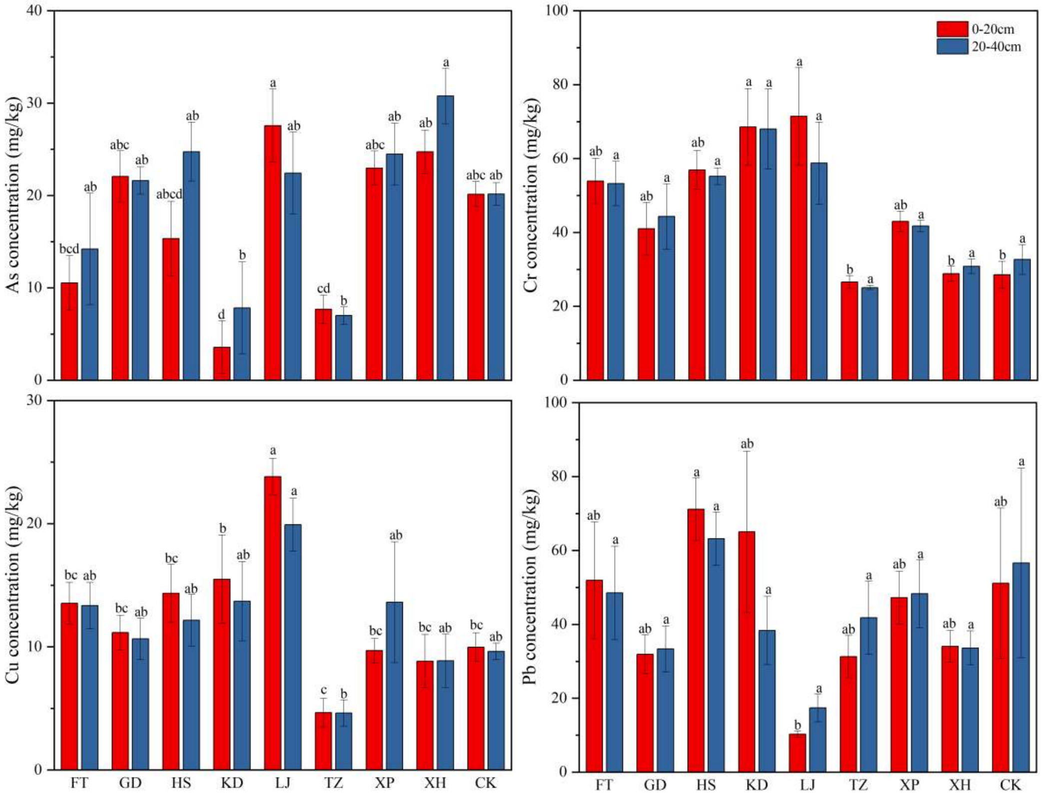

Figure 1. Soil As, Cr, Cu and Pb concentrations in main tea-producing areas in Anxi County. FT, Futian; GD, Gande; HS, Hushang; KD, Kuidou; LJ, Longjuan; TZ, Taozhou; XP, Xiping; XH, Xianghua; CK, Changkeng. Different letters indicate significant differences ($p < 0.05$) among different regions in the same soil layer.

In our study, the Nemerow composite index (P) and potential ecological risk index (RI) were applied to identify heavy metal pollution level. Based on the Nemerow composite index method, topsoil As, Cu and Cr concentrations were less than 0.7, indicating that soils were safe, whereas the Pb pollution index in FT, KD and CK was higher than 0.7 and lower than 1, indicating alert level. The Pb pollution index in HS was higher than 1, suggesting minor pollution. On the whole, the studied soils were not contaminated based on the Nemerow composite index p-value (Table 2). Based on potential ecological risk assessment, soil Cr, Cu and Pb would pose a low ecological risk for environment, because the Ei values of all regions were less than 40. In contrast, the Ei value of As in LJ and XH was higher than 40 and lower than 80, indicating a moderate ecological risk. Assessing the RI value for As, Cr, Cu and Pb in our study, the RI values of the nine regions were all lower than 110, showing low ecological risks.

Table 2. Evaluation results of Nemerow composite index and potential ecological risk index of heavy metals in soils of main tea-producing areas in Anxi County.

Regions	**As**		**Cr**		**Pb**		**Cu**		**P**	**RI**
	P_i	E_i	P_i	E_i	P_i	E_i	P_i	E_i		
FT	0.264 ± 0.073	18.239 ± 5.077	0.359 ± 0.041	2.611 ± 0.299	0.741 ± 0.226	7.435 ± 2.268	0.271 ± 0.034	3.132 ± 0.393	0.662 ± 0.155	31.419 ± 5.742
GD	0.551 ± 0.069	38.160 ± 4.807	0.273 ± 0.212	1.986 ± 0.344	0.456 ± 0.075	4.569 ± 0.756	0.223 ± 0.028	2.581 ± 0.323	0.537 ± 0.067	47.295 ± 5.212
HS	0.383 ± 0.101	26.527 ± 6.991	0.379 ± 0.035	2.756 ± 0.254	1.016 ± 0.122	10.194 ± 1.220	0.287 ± 0.047	3.322 ± 0.544	0.808 ± 0.080	42.798 ± 5.688
KD	0.089 ± 0.072	6.170 ± 4.989	0.457 ± 0.069	3.320 ± 0.501	0.930 ± 0.312	9.322 ± 3.128	0.310 ± 0.072	3.585 ± 0.833	0.744 ± 0.218	22.397 ± 5.058
LJ	0.689 ± 0.099	47.673 ± 6.851	0.476 ± 0.088	3.459 ± 0.640	0.147 ± 0.012	1.473 ± 0.124	0.476 ± 0.030	5.513 ± 0.346	0.606 ± 0.047	58.118 ± 7.025
TZ	0.192 ± 0.038	13.283 ± 2.636	0.177 ± 0.012	1.287 ± 0.084	0.447 ± 0.082	4.484 ± 0.821	0.093 ± 0.023	1.077 ± 0.272	0.369 ± 0.049	20.131 ± 2.258
XP	0.574 ± 0.046	39.740 ± 3.195	0.287 ± 0.019	2.081 ± 0.135	0.675 ± 0.101	6.770 ± 1.021	0.194 ± 0.020	2.243 ± 0.231	0.586 ± 0.067	50.835 ± 4.178
XH	0.618 ± 0.059	42.776 ± 4.061	0.192 ± 0.013	1.397 ± 0.098	0.486 ± 0.061	4.875 ± 0.616	0.177 ± 0.043	2.046 ± 0.501	0.533 ± 0.047	51.093 ± 4.614
CK	0.504 ± 0.034	34.847 ± 2.325	0.190 ± 0.024	1.383 ± 0.174	0.730 ± 0.291	7.322 ± 2.915	0.199 ± 0.023	2.308 ± 0.269	0.686 ± 0.178	45.860 ± 2.073

Note: FT, Futian; GD, Gande; HS, Hushang; KD, Kuidou; LJ, Longjuan; TZ, Taozhou; XP, Xiping; XH, Xianghua; CK, Changkeng. P_i represents single factor pollution index, P represents Nemerow pollution index. E_i is single heavy metal potential ecological risk, RI is comprehensive potential ecological risk.

3.3. Tea Leaf Heavy Metals and Bioaccumulation Factors

The accumulation of heavy metal in tea leaves followed the order: Cu > Pb > Cr > As. No significant difference was found in As and Cr concentration among the nine regions (Figure 2). The mean value of Cu concentration in tea leaves was in order of LJ (9.81 ± 0.54 mg/kg) > HS (8.71 ± 0.10 mg/kg) > KD (8.39 ± 0.39 mg/kg) > XP (8.02 ± 0.43 mg/kg) > FT (7.30 ± 0.40 mg/kg) > CK (6.10 ± 0.87 mg/kg) > GD (6.04 ± 0.24 mg/kg) > XH (4.97 ± 0.37 mg/kg) > TZ (4.24 ± 1.43 mg/kg); Cu concentration in LJ showed the highest ($p < 0.05$). The mean value of tea leaves Pb concentration was in order of HS (2738 ± 199.620 ug/kg) > FT (1040 ± 246 ug/kg) > GD (919 ± 245 ug/kg) > LJ (918 ± 3491 ug/kg) > TZ (766 ± 128 ug/kg) > XH (655 ± 149 ug/kg) > CK (587 ± 269 ug/kg) > KD (585 ±85 ug/kg) > XP (549 ± 76 ug/kg); Pb concentration in HS was markedly higher than that in other regions ($p < 0.05$).

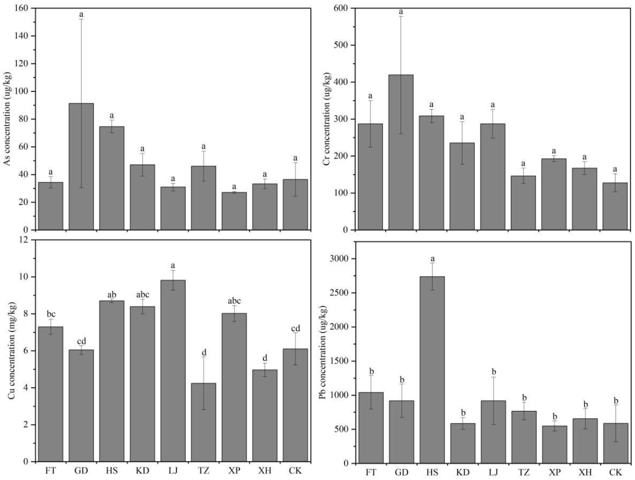

Figure 2. Tea leaves As, Cr, Cu and Pb concentrations in main tea-producing regions in Anxi County. Different letters mean statistical differences at the level of $p < 0.05$.

The mean BAF values for As, Cr, Cu and Pb were 0.001–0.078, 0.003–0.011, 0.410–0.865 and 0.012–0.087, with average values of 0.008, 0.007, 0.685 and 0.029 (Table 3). The BAFs of the four heavy metals in descending order were as follows: Cu > Pb > As > Cr, and the BAFs were all less than one, indicating no significant accumulation of heavy metals from soil to tea leaves.

Table 3. Bioaccumulation factor of heavy metal elements in tea leaves samples.

Index	Bioaccumulation Factor			
	As	Cr	Cu	Pb
FT	0.005 ± 0.001	0.006 ± 0.001	0.635 ± 0.087	0.026 ± 0.005
GD	0.009 ± 0.007	0.011 ± 0.003	0.635 ± 0.047	0.034 ± 0.009
HS	0.006 ± 0.002	0.005 ± 0.0002	0.640 ± 0.091	0.040 ± 0.006
KD	0.078 ± 0.038	0.004 ± 0.0008	0.660 ± 0.187	0.012 ± 0.004
LJ	0.001 ± 0.0003	0.005 ± 0.002	0.410 ± 0.014	0.087 ± 0.033
TZ	0.007 ± 0.002	0.006 ± 0.001	0.860 ± 0.206	0.029 ± 0.006
XP	0.001 ± 0.0001	0.005 ± 0.0003	0.866 ± 0.070	0.013 ± 0.002
XH	0.002 ± 0.0002	0.006 ± 0.0007	0.772 ± 0.108	0.027 ± 0.010
CK	0.002 ± 0.0008	0.005 ± 0.001	0.625 ± 0.083	0.013 ± 0.003

Note: The smaller the coefficient, the worse the ability of tea leaves to absorb and accumulate heavy metals.

3.4. Correlations Between Soil Physicochemical Properties and Heavy Metals in Soil and Tea Leaves

RDA was conducted to identify the main soil physicochemical properties that affected soil and tea heavy metal (Figure 3A). The results showed that the first two axes accounted for 78.01% of the total variation; the first RDA axis accounted for 60.64%, with the strong loadings of AP, pH, EAl and EA, and the second RDA axis accounted for 17.37%. Therefore, they were identified as the dominant factors influencing soil and tea heavy metal. Correlation analysis and random forest analysis showed that the heavy metals concentrations were positively associated with soil pH, while negatively associated with soil EA, EAl and AP (Figure 3B,D). In addition, the correlation analysis of soil heavy metal concentration and tea heavy metal concentration (Figure 3C) showed that soil Cu concentration was positively related to tea Cu concentration, soil Pb concentration was positively related to tea Pb concentration and tea Cu concentration was also positively related to soil Cr and Pb concentration.

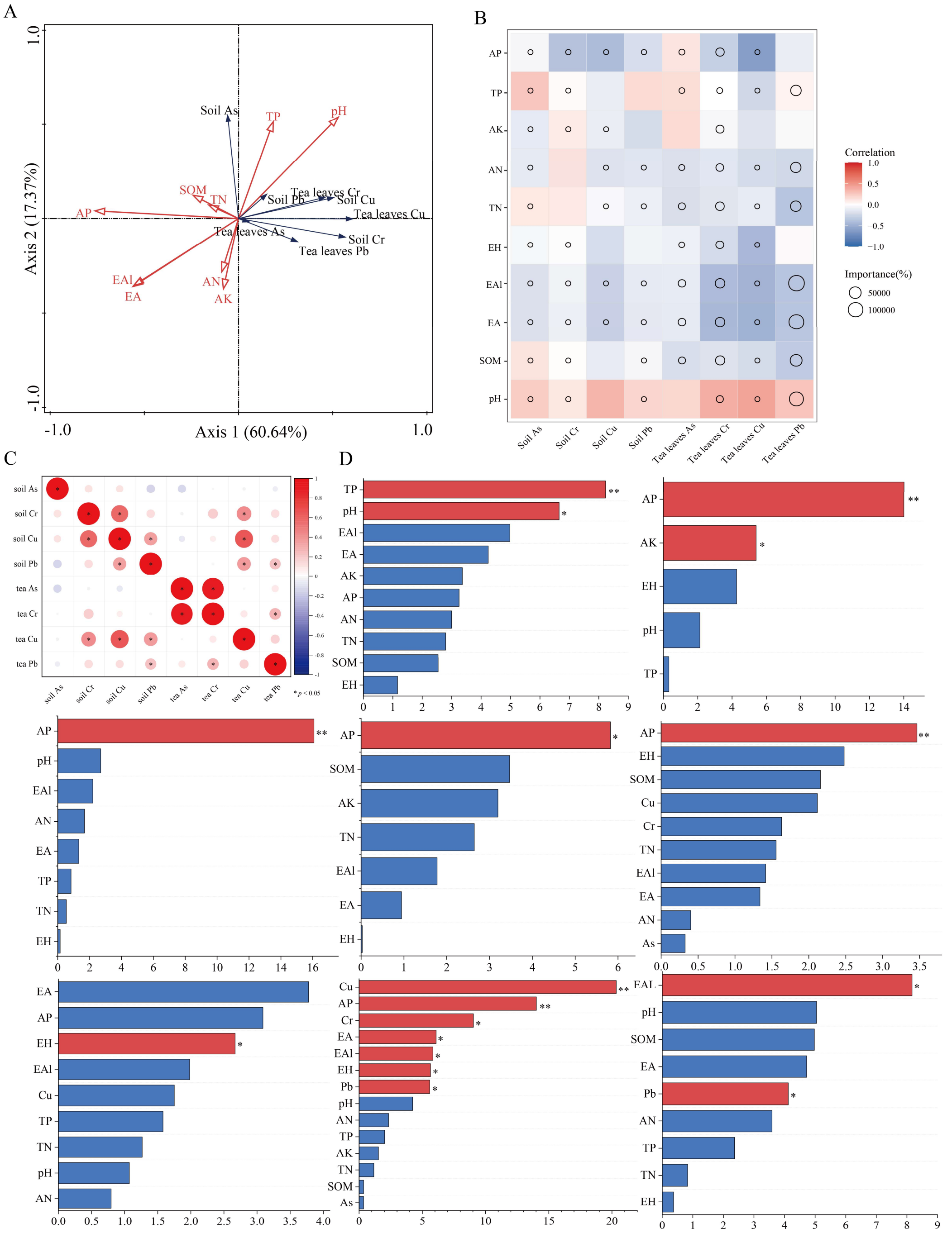

Figure 3. Correlation analysis on soil physicochemical characteristics and heavy metal concentration. Redundancy analysis on soil physicochemical characteristics and soil and tea leaf heavy metal concentration (**A**); correlation analysis between soil physicochemical characteristics and heavy metal concentration (**B**); Pearson correlation analysis between soil and tea leaf heavy metal concentration (**C**); random forest modelling analyses aiming to identify the importance of soil physicochemical characteristics in predicting heavy metal concentration (**D**), * $P < 0.05$, * * $P < 0.01$.

3.5. Human Health Risk Assessment of Heavy Metals in Tea Leaves

The mean THQs of each heavy metal followed the order: Cu (7.659×10^{-3}) > As (7.420×10^{-4}) ≈ Pb (7.410×10^{-4}) > Cr (3.349×10^{-6}); the THQs were all <1, suggesting there was no potential human health via oral intake of single heavy metal. The HI values of four heavy metals in all regions were <1 (Figure 4), indicating no potential human health risk.

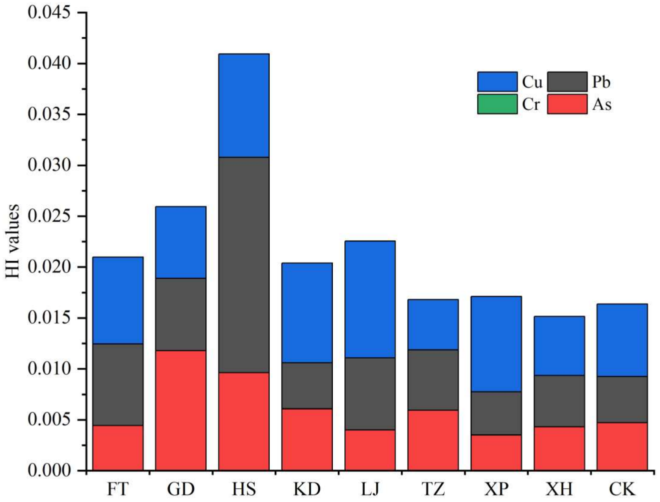

Figure 4. The calculated target hazard quotients (THQ) of As, Cr, Cu and Pb and the accumulative hazard indexes (HI) for adults with tea leaves from main tea plantations in Anxi County in Fujian Province, southeast China.

4. Discussion

4.1. Soil Heavy Metals Concentrations

The environment risk standards of heavy metal proposed are based on soil total heavy metal concentrations so far [4]. The average values of As, Cr, Cu and Pb concentrations in tea plantations were 3.24-, 1.04-, 0.53- and 1.16-fold that of their background values at 5.78, 41.3, 21.6 and 34.9 mg/kg of Fujian Province [32]. At the same time, the average values of As, Cr, Cu and Pb concentration did not exceed their risk screening values of 40, 150, 50 and 70 mg/kg, respectively, in agricultural soil of China (GB 15618-2018) [40]. Pb concentration in HS exceeded the risk screening value, but As, Cr, Cu and Pb concentrations of other regions did not exceed the risk screening value, indicating the soil environment of tea plantations in Anxi County was very good as a whole. General speaking, the mean values of As, Cr, Cu and Pb concentrations were low in this study compared to other main producing areas, such as those observed in some studies in the other six main tea-growing regions (28.3 mg/kg for As, 108 mg/kg for Cr, 43.9 mg/kg for Pb) in Yunnan Province [4] and Mengku tea garden (80.7 mg/kg for As, 110 mg/kg for Cr, 88 mg/kg for Cu) and Jingmai (17.7 mg/kg for As, 87 mg/kg for Cr, 106 mg/kg for Cu) in Yunnan Province [17], but Pb concentration was higher than those in Mengku and Jingmai tea garden [17].

The heavy metal concentrations have strong heterogeneity among the nine studied regions; Pb concentration in LJ showed the lowest value, the highest value in HS was 6.92-fold that of the value in LJ and As, Cr and Cu concentration in LJ showed the highest values, which were 7.73-, 2.69- and 5.12-fold those of the lowest regions in KD and TZ (Figure 1). The differences in the current study may be caused by differences in fertilizers; organic fertilizer is usually employed in LJ and KD. Organic fertilizers can provide essential nutrients to plants, but since the materials used as organic fertilizer contain a large amount of heavy metals, they bring detrimental amounts of heavy metals into the soil–plant system [26].

The Nemerow comprehensive pollution index and potential ecological risk index were used to evaluate the soil pollution level of heavy metals. The mean values of P_i follow the sequence Pb > As > Cr > Cu, suggesting that Pb represents a higher risk compared to other heavy metals. The Pi value of As, Cr and Cu in all regions did not exceed 0.7, and except for HS where the Pi value of Pb exceeded 1, the rest were below 1, indicating serious heavy metal soil pollution has not yet occurred in the studied area. The calculation outcomes and grading criteria for the *p*-value show that the soil heavy metal pollution in HS and KD should be paid great importance, attributed to the higher Pb concentrations [42]. The mean E_i value in the current study followed the following order: As > Pb > Cu > Cr; it was determined that there was low contamination risk according to the categorizing basis. Our data also demonstrated that a low risk of As pollution was identified in LJ and XH; this is attributed to the higher As concentrations, and organic fertilizers and phosphatic fertilizers were important inputs [10]. Given that the RI values in all regions were lower than 110, it is inferred that the area has a low ecological risk. The emphases of different evaluation methods are different, the potential ecological risk index can reflect the toxicity of heavy metals to organisms and there is a need to combine different methods to evaluate soil heavy metal pollution. The results of these two pollution level evaluation indexes were similar, which means that the soils of the studied tea plantations can be considered as not contaminated by heavy metals.

4.2. Tea Leaf Heavy Metals Concentrations

Our results found that As, Cr, Cu and Pb concentration in tea leaves were within the limiting values of Chinese National Food Safety Standard at 2, 5, 30 and 5 mg/kg (GB 2672-2017, NY 659-2003, NY/T 288-2012) [19–21], As and Pb concentration were within the limiting values of WHO at 1 and 10 mg/kg [47] and Pb concentration was also below the standard limits of EU (3 mg/kg) [48], indicating that Tieguanyin tea in Anxi County is very safe. The mean values of As, Cr and Cu concentrations in tea leaves were lower than those in Liubao tea [14], Dayezhong tea [17] and tea from Puan County [18], while Pb concentration in tea leaves was higher than that in other areas, maybe due to higher Pb concentration in HS. Another possible reason is that heavy metal accumulation in tea plants depends on tea cultivars [1], and different tea types could have different physiological absorption mechanisms for different heavy metals [24].

The BAFs of the four heavy metals in descending order were as follows: Cu > Pb > As > Cr, which is similar to tea leaf heavy metal concentrations. Cu is one of the most abundant trace elements in tea leaves, and this study showed that the ability of Cu transfer and accumulation from soil to plants was higher than other heavy metals, indicating strong uptake ability of Cu, which may have been because tea leaf Cu concentration was positively correlated with soil Cu concentration. The tea root is a major organ for the absorption and accumulation of Pb, and soil Pb concentration was the main factor affecting Pb absorption in tea leaves [49]. Previous studies have demonstrated that As is low in mobility, absorbed As is fixed in feeder roots and only a small amount is transported to aboveground organs; this may be the main reason explaining the lower enrichment ability of As in tea leaves [50]. The Cr element enrichment ability of tea leaves was low; our results could be explained by the fact that Cr is mainly accumulated in the root of tea plants after being absorbed from the soil, and the migration ability to the leaves is weak [17]. In addition, accumulation and transfer of heavy metals in tea leaves from the soil were also influenced by other factors such as plant cultivars and geological background [1].

4.3. Factors Influencing Heavy Metals in Soil and Tea Leaves

Soil environmental changes can affect heavy metal concentrations of soil and tea leaves, and previous reports showed that heavy metal concentrations were linked to pH [4,51], soil H^+ activity [28], soil SOM [29] and soil particle diameter [27]. Our data demonstrated that the soil acidification index was an important factor affecting heavy metals concentrations (Figure 3). We also found that the soil of the tea plantations investigated were acidified, and the highest pH value was 4.628 in LJ. Soil acidification could be explained by the fact that the long-term application of nitrogen fertilizer promoted the accumulation of exchangeable

Al^{3+}, and further produced higher concentrations of free H^+; nitrogen fertilizer also leads to the formation of $NH_4{}^+$, which facilitates soil acidification [24]. Heavy metals that exist in the cationic form are prone to be adsorbed and fixed at higher pH; with the process of soil acidification, heavy metals gradually dissociate into free cations, enhancing their toxic effect on plants [52]. In addition, tea root-exuded organic acids, such as oxalic acid and malic acid, also contribute to soil acidification [53].

We found that soil pH was markedly positively correlated with soil As and Cr concentrations, which was consistent with the result of Ju et al. [4]. However, Li et al. [54] revealed that soil pH did not significantly influence soil As and Cr. It is quite possible that this may be due to the degree of soil acidification. The soil from the tea plantations was extremely acidic in our study, with pH values ranging from 3.74–4.63. Previous studies indicated that the phytoavailable As in soil is extremely low under soil acidification conditions in tea gardens [55]. And, As that is absorbed by tea by plant root is localized to the root system, which acts as a buffer and defense, because only a small amount of As is translocated to the aboveground parts of tea plant [24], so As concentrations in tea leaves and soil were not tightly linked in our study. Cu is one of the essential trace elements, but its excessive accumulation can threaten human health [12]. The significantly positive correlations were found between soil pH and soil Cu concentration, soil pH and tea leaf Cu concentration and soil Cu concentration and tea leaf Cu concentration, and random forest analysis also found that soil Cu is main factor of the predictors of tea leaf Cu concentration, which indicated that soil is the main source of tea leaf Cu accumulation. Similar to our results, some findings have demonstrated that soil pH is positive corelated with soil Cu concentration in acid soil [56], probably due to strong adsorption of aluminum oxide and leaching under strong acidic conditions, and the available Cu concentration decreases with the decrease of pH value [56]. Meanwhile, tea leaf Pb concentration was significantly positively correlated with soil pH and soil Pb concentration, which may have been because soil is a substantial source for Pb accumulation in tea leaves, which is consistent with the results of Ju et al. [4].

In this study, AP was also thought to be a key factor influencing heavy metal concentration. Phosphates exerted a greater effect on the adsorption of heavy metal via combining with heavy metals to form phosphate precipitation, increasing the negative charge on the surface of the adsorbent and reducing the increase in surface potential [57], which reduced the mobility and bioavailability of heavy metals in the soil to fix or passivation, and thus reduced the harm caused by heavy metals [58]. In highly phosphorus soil, the formation of metal phosphate precipitation is one of the main mechanisms by which phosphorus reduces the mobility of metal ions in soil [59]. Previous studies have found that the application of more soluble phosphorus in Pb pollution soil can lead to the formation of phosphoric acid lead salt precipitation, thus inhibiting the absorption of Pb in plants and long-distance transport in plants, and reducing the biological toxicity of Pb [59].

4.4. Health Risk Assessment

The potential harm to humans through tea drinking can be evaluated quantitatively, the THQ and HI values have been successfully used to assess the possible non-carcinogenic effects of heavy metals on human health via various food consumption [4,24]. In this study, the THQ values of four heavy metals and HI values were less than 1, indicating there was no potential human health risk. The four heavy metals do not result in health effects via tea intake; this was in consistent with the previous findings in Oolong tea [60] and Yunnan big leaves tea [18]. The mean THQs of each heavy metal were ranked in the order Cu > As ≈ Pb > Cr, which is different from a previous order of Tieguanyin tea, As (0.041) > Cu (0.009) > Pb (0.001) > Cr (0.0008) [61]. Cu showed the highest THQ and HI values, which was attributed to high Cu concentration in tea leaves and TRi value, indicating a high transfer ratio of Cu from tea leaves to tea infusion.

Cr showed the lowest THQ and HI values, which was attributed to high RfDi value, indicating high oral intake amount of Cr proposed by USEPA. In the meantime, our data were based on unprocessed tea heavy metal concentrations, while tea may be contaminated by heavy metals from tea-making machines during production [24]. For instance, Cr concentration of tea probably increased from the rollers at the stage of cutting and Cu concentration increased after rolling by the rotor vane [24].

Although the consumption of tea from this study area is safe, there are other exposure ways on humans health risks to heavy metal. The health risks caused by heavy metal may depend on crop species, with grains exhibiting the highest risk [62]. Previous studies have demonstrated that the health hazard index of tea intake (0.05) was significantly lower than rice (10.44) and vegetables (2.86) [22]. Therefore, more comprehensive investigation into dietary structure and heavy metal concentrations of water, vegetables, fruits and grains are necessary for a comprehensive human health risk assessment.

5. Conclusions

This study investigated the concentration of heavy metals in the soil of tea plantations and the corresponding tea leaves in major tea-producing areas in Anxi County, Fujian Province. Our results indicated that the mean total concentrations of As, Cr, Cu and Pb in the topsoil did not exceed their risk screening values in China (GB 15618-2018). The Pb pollution value in HS suggests minor pollution, while the potential ecological risk assessment of As in LJ and XH indicates a moderate ecological risk. Overall, the potential ecological risk index (RI) for all regions shows low ecological risks. The heavy metal concentrations in tea leaves met the Chinese National Food Safety Standards limits. The accumulation of Cu and Pb in tea leaves was higher than that of As and Cr. The exposure to these four heavy metals through tea consumption does not pose a health risk to humans. The concentrations of heavy metals in both soil and tea leaves were mainly influenced by soil pH, EA, EAl and AP. The Cu and Pb concentrations in tea leaves were positively correlated with the concentrations of Cu and Pb in the soil, respectively. The results of this study contribute to our understanding of heavy metal accumulation in Tieguanyin tea leaves and the factors influencing them in Anxi County. In conclusion, Tieguanyin tea may still be considered beneficial to health.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/agriculture14111907/s1, Figure S1: The study area and distribution of sampling sites in Anxi County, Fujian Province, Southeast China. FT, Futian; GD, Gande; HS, Hushang; KD, Kuidou; LJ, Longjuan; TZ, Taozhou; XP, Xiping; XH, Xianghua; CK, Changkeng.

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