

## Article

# Content and Bioavailability of Hg in a Soil–Tea Plant System in Anxi Area, Southeast China

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**Abstract:** Many studies on the bioavailability of Hg in soil–plant systems have been conducted. However, studies on the soil–tea plant system are still limited. In this study, we collected soil and tea leaf samples from 69 tea gardens in the Anxi area in China. We investigated the Hg contents in tea leaves and soil, and the bioavailability of Hg from soil to tea leaves. The results showed the following: (1) The Hg content in soil was equivalent to the background value in Fujian province and was 2.6–2.9 times higher of the national background value in China. For different geological backgrounds, the Hg contents differed. In addition, we found a negative correlation between the total Hg content and pH value ( $r = -0.196$ ) and a positive correlation between total Hg and soil organic carbon (SOC) content ( $r = 0.116$ ) in surface soil. (2) Compared with young leaves, Hg content in older leaves was higher. The transfer factor (TF) of Hg from soil to tea leaves ranged from 0.25% to 24.76%. (3) The correlation between Hg content in tea leaves and total Hg content in soil was not statistically significant. However, we found a statistically significant positive correlation between the Hg content in tea leaves and the extracted amount of Hg both in surface and subsurface soil. The SOC and pH value were also statistically significantly positively correlated with the Hg content in tea leaves. Our findings show that the Hg content in tea leaves does not only depend on the total Hg content in soil: it is also affected by other factors.



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**Keywords:** Mercury; tea garden; bioavailability

## 1. Introduction

Mercury (Hg) is one of the most toxic heavy metal elements, and it is an environmental pollutant that bioaccumulates in animals and humans [1,2]. The major effects of Hg poisoning manifest as neurological and renal disturbances, as it can easily pass through the blood–brain barrier and affects the brain [3,4]. Food and beverage consumption are the primary paths of Hg intake by humans, which has increasingly attracted attention in recent years [4,5]. Plants, such as crops, vegetables, and tea, are the most common food or beverages in our lives. Most plants mainly take up Hg from the soil [6]. The bioavailability of Hg is defined as the fraction of Hg that is actually taken up by plants, which indicates the ability of plants to extract Hg from the soil, and can be quantified by the effect of Hg on the plants [7]. Therefore, the bioavailability of Hg in soil–plant system has been studied [8].

Studies have been conducted since the 1970s in many areas worldwide [9–11]. For instance, Barnett and Turner [12] reported the bioaccessibility of Hg in soils in the flood-plain of Tennessee; Golow [13] studied the Hg in surface soil and cassava crops near an alluvial goldmine in Ghana. Some studies have been conducted in China [14–16]; however, these studies many considered trees [17,18] or other crops including rice, vegetables, and so on [14,15,19,20]. The results indicated that the Hg content in plants such as tea is influenced by soil conditions [4,21], atmospheric Hg (dry and wet deposition), and other factors [22].

In summary, the factors influencing the Hg content in tea are complex [5]. The tea tree (*Camellia sinensis* L.) originated in China, and tea has been popular globally for hundreds of years. China is currently the largest tea producer in the world, producing 45% of the global total in 2018 [23]. Many studies have reported on the Hg content in different kinds of tea [24–26] and the bioavailability of other metals such as Pb and Cd [27–29]. Nevertheless, studies focusing on Hg bioavailability in the tea plant–soil system remain limited. A study on the Hg bioavailability from 19 tea gardens in Wuyishan showed that the enrichment coefficient of Hg was 0.083–1.236, indicating that tea plants have a strong ability to accumulate the Hg from the soil; a negative correlation was found between the Hg content in tea and the total Hg content in the soil [30]. In China, tea is cultivated on a large scale. The tea gardens in China differ in geological background and environmental conditions. However, the effect of geological background and environmental conditions on the bioavailability of Hg in soil–tea plant systems remains unclear.

In this study, we examined the bioavailability of Hg in the soil–tea plant system in southeast China by sampling and analysis. Our aims in this study were to (1) investigate the distribution and migration characteristics of Hg in soil–tea plant system; (2) to determine the bioavailability of Hg in soil–tea plant system and the influence factors. The results can provide reference for local tea plantation management and similar research.

## 2. Materials and Methods

### 2.1. Study Area

The study area is Anxi area, Fujian province, Southeast China (Figure 1). The longitude and latitude are 117°29′~118°14′ and 24°11′~24°36′, respectively. The terrain is mainly mountainous and hilly, with an average elevation of 500–700 m. The area has a sub-tropical monsoon climate with abundant precipitation. The annual average precipitation is 1600 mm and the annual average temperature is 19–21 °C. Intermediate-acid volcanic rocks are widely distributed in this area, and the strata outcrop is relatively complete. Red soil is the main soil type. Anxi is famous for its tea industry, having a tea production history stretching thousands of years. Nowadays, organic fertilizer is more popular than chemical fertilizer. The cultivated area of tea was approximately 40,000 ha in 2018, with an output of 62,000 t. It is the birthplace of Anxi Tieguanyin and is known as the “tea capital of China”.

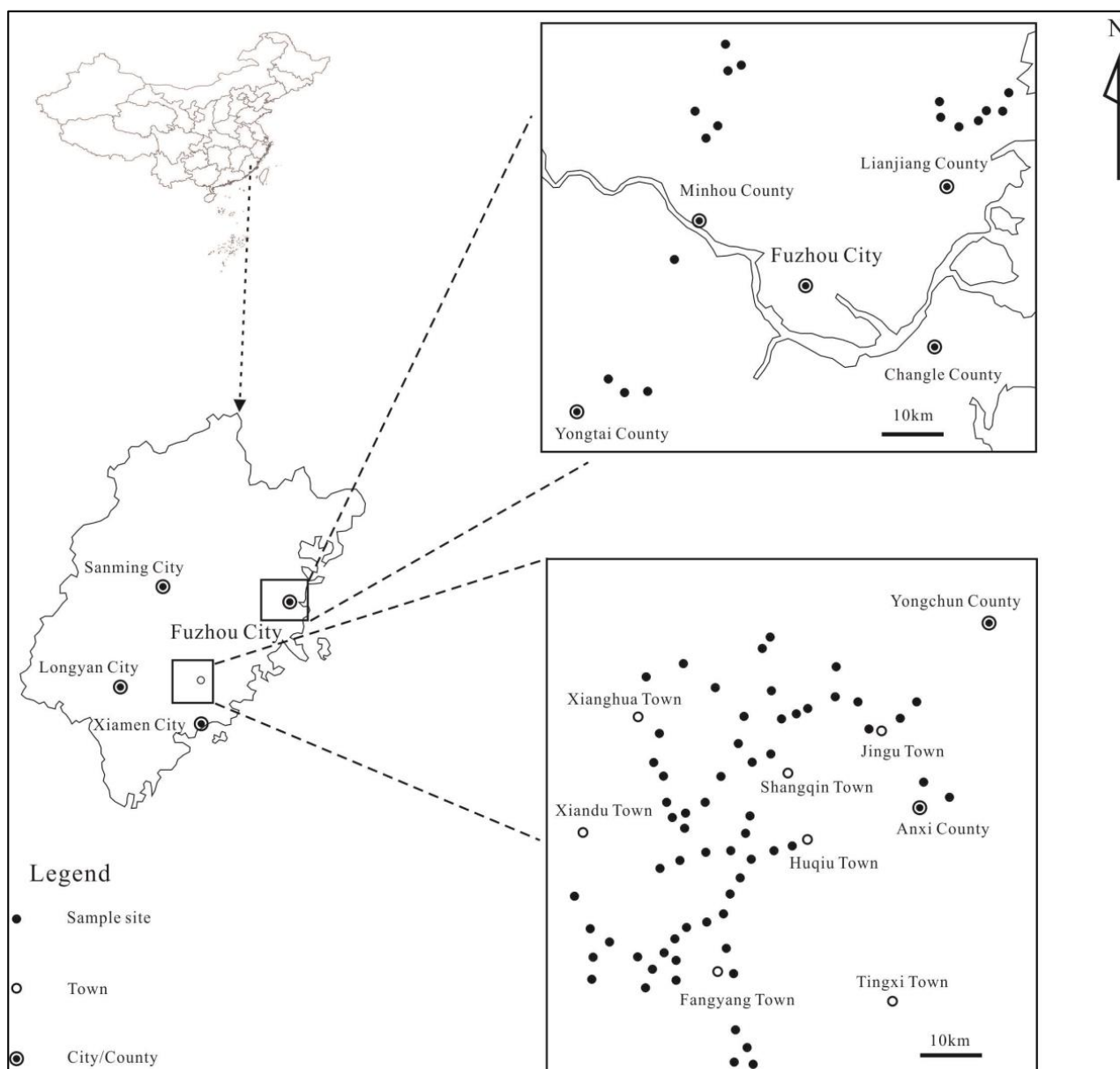
### 2.2. Sample Collection and Pretreatment

In the study area, we selected 69 tea gardens for collecting tea leaf and soil sample (Figure 1). The age of tea trees in all gardens was approximately 15–20 years. We chose 1–2 sampling sites for larger tea gardens (>10 ha) and 1 sampling site for smaller tea gardens (<5 ha); in total, we selected 79 sampling sites. We collected one tea leaf sample and two rhizosphere soil samples (one surface soil sample from a depth of 0–20 cm and one subsurface soil sample from a depth of 20–40 cm) from each sample site.

#### 2.2.1. Tea Leaf Sample

We collected two kinds of tea leaf samples (young and old leaf samples) from study area. For every sample site, we first assessed the quantity of young leaves. If the tea trees had many young leaves, we picked 1 bud and 2–3 young leaves from ten or more tea trees around the sample site as a sample. Otherwise, we collected old leaves as a sample. The fresh weight was above 500 g for each sample.

After collection, we transported all samples back to the laboratory. For each sample, we applied the same process as follows: First, we rinsed the tea leaf samples with tap water, then deionized water 3 times, which we then air-dried for 3–4 h. Then, we placed the samples in a drying baker at 65 °C until dried. Finally, we crushed the samples and passed through a 40-mesh sieve (420 µm) for chemical analysis. We designated this sample as the Group A sample.



**Figure 1.** Location of study area and sample sites.

#### 2.2.2. Soil Sample

We collected one soil sample at the sample site and four soil samples nearby to compose a soil sample. At any sample site, the surface soil sample (depth = 0–20 cm) and the subsurface soil sample (depth = 20–40 cm) shared the same subsample site. The weight of each sample was more than 1.0 kg. After being collected, we naturally air-dried each soil sample, then crushed and sieved it through a 10-mesh (2000  $\mu\text{m}$ ) sieve, which we then mixed thoroughly for later use. The soil samples were designated the Group B sample.

#### 2.3. Screening and Speciation Extraction of Soil Samples

##### 2.3.1. Soil Sample Screening

We randomly selected 30 surface soil samples and 30 subsurface soil samples for sieving into five different soil size fractions. The soil sizes were –10~+20 mesh (<2000~850  $\mu\text{m}$ ), –20~+60 mesh (<850~250  $\mu\text{m}$ ), –60~+120 mesh (<250~125  $\mu\text{m}$ ), –120~+230 mesh (<125~63  $\mu\text{m}$ ), and –230 mesh (<63  $\mu\text{m}$ ). We used a supersonic wave oscillating screen composed of stainless steel for soil sample screening. After screening, we split each sample

into 5 subsamples; thus, we obtained 150 subsamples from 30 surface soil samples and 150 subsamples from 30 sub-surface soil samples in total. They were designated as the Group C sample.

### 2.3.2. Speciation Extraction of Soil Sample

We performed speciation extraction for the Group C sample with a single extraction process (Table 1). The procedure involved aqueous extraction (AE), dilute acid extraction (DAE), salt solution extraction (SSE), and complexing agent extraction (CAE).

**Table 1.** Single extractions used in this study.

Methods	Extractant	Reference
AE	deionized water	[20]
DAE	diluted hydrochloric acid (0.1 mol/L)	
SSE	CaCl <sub>2</sub> (0.1 mol/L)	
CAE	DTPA (0.005 mol/L) + CaCl <sub>2</sub> (0.01 mol/L) + TEA (0.1 mol/L), pH = 7.3	

Notes: AE = aqueous extraction, DAE = dilute acid extraction, SSE = salt solution extraction, CAE = complexing agent extraction, DTPA = Diethylenetriaminepentaacetic acid, TEA = triethanolamine.

### 2.4. Chemical Analyses of Soil and Tea Leaves

We analyzed the Group A sample for total Hg content. We analyzed the Group B sample for total Hg content, soil properties (pH, SOC, TC, and major elements) and other elements; we analyzed the Group C sample for the same indicators as in the Group B sample but with the addition of pH and CEC. After the extractions, we analyzed the Hg content of the extracting solutions.

We determined the soil pH by mixing a 10 g sample with 25 mL of distilled water, which we stirred and left at 25°) for 0.5 h. We determined the suspension pH by using a pH electrode (FE20, Shanghai Mettler Toledo Company, Shanghai, China).

We determined the soil organic carbon (SOC) and total carbon (TC) by the oxidative pyrolysis–potentiometric method (POT); we determined total Hg in soil by cold vapor–atomic fluorescence spectrometry (CV-AFS); and we determined SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, TFe<sub>2</sub>O<sub>3</sub>, MgO, and CaO contents by X-ray fluorescence spectroscopy (XRF). The soil sample analysis information is shown in Table 2.

**Table 2.** The methods and detection limit of soil samples analysis.

Indicator	Method	DL	Unit	Indicator	Method	DL	Unit
SiO <sub>2</sub>	XRF	0.1	%	CEC	Potentiometry	0.25	cmol(+)/kg
Al <sub>2</sub> O <sub>3</sub>	XRF	0.1	%	pH		0.1	/
TFe <sub>2</sub> O <sub>3</sub>	XRF	0.1	%	SOC	POT	0.1	%
MgO	XRF	0.05	%	TC	POT	0.1	%
CaO	XRF	0.05	%	Hg	CV-AFS	2	µg/kg

Notes: XRF = X-Ray fluorescence spectroscopy, POT = Oxidative pyrolysis–potentiometric, CV-AFS = Cold vapor–atomic fluorescence spectrometry, DL = Detection limit.

For the determination of total Hg content in tea leaves, we digested 1 g of sample via the microwave-acid method (GB/T5009-2003) in HNO<sub>3</sub> 65%/HF 40%/HCl 37%, 9:3:2 (v/v). We filtered the digestion solution with a 0.22 µm polytetrafluorethylene filter, which we analyzed by ICP-MS.

We used national standard materials and duplicate samples to monitor the quality of the analytical method. The detection limit, accuracy, and precision of the analyses all met the required criteria in this study.

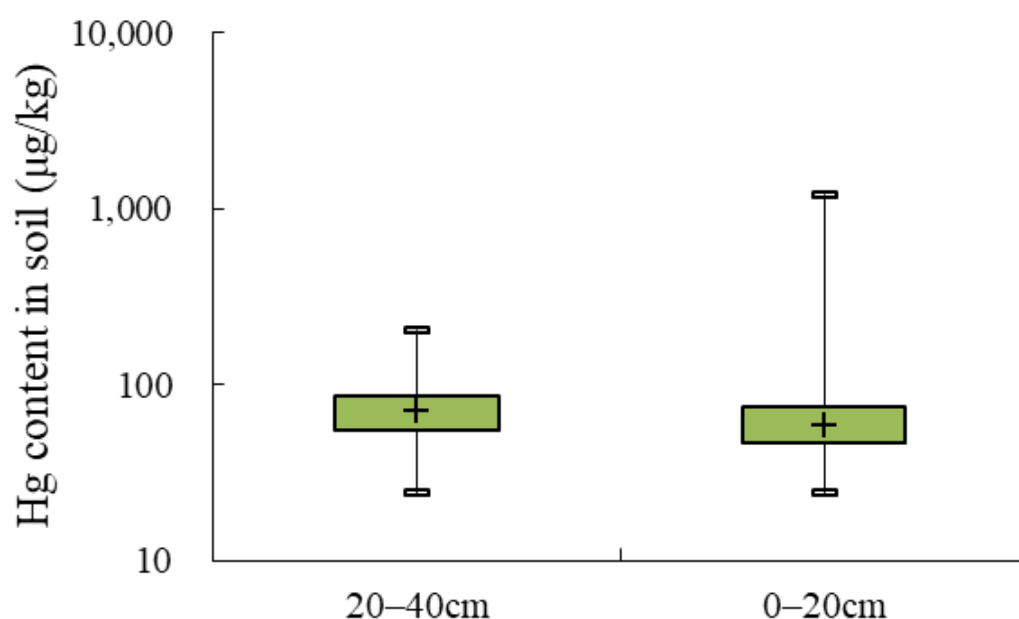
### 2.5. Statistical Analyses and Mapping

We conducted statistical analysis, including analysis of variance and Pearson correlation, with SPSS version 19.0 software (SPSS Inc., Chicago, IL, USA). We mapped the figure of the study location and sample site using CorelDRAW 2018 and other figures using Microsoft Excel.

## 3. Results

### 3.1. Hg Content in Soil

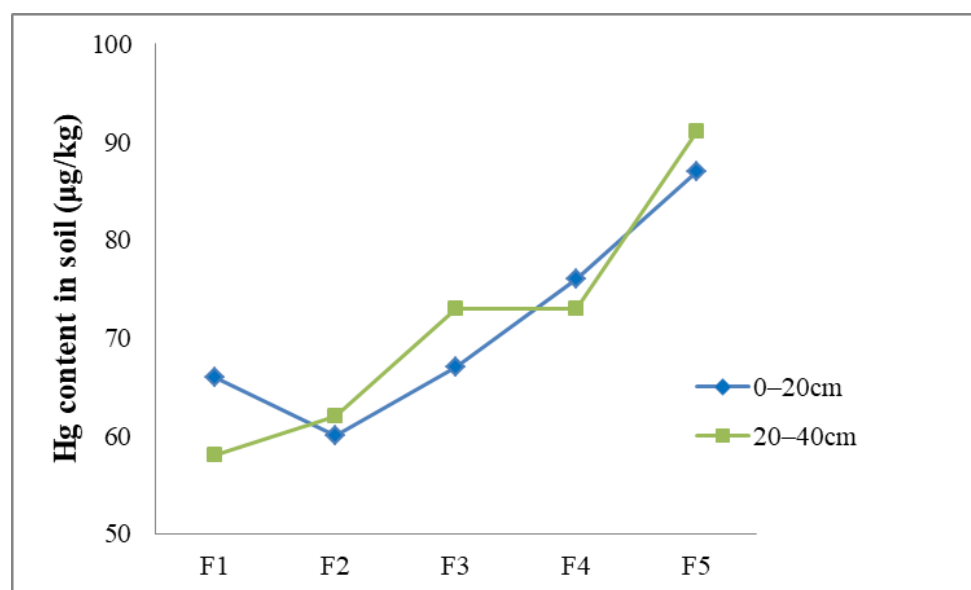
The soil Hg contents from the 79 surface and 79 subsurface soil samples are shown in Figure 2. The Hg content in surface soil ranged from 24.4 to 1201  $\mu\text{g}/\text{kg}$ , with an average of 78.3  $\mu\text{g}/\text{kg}$ . In subsurface soil, the Hg content ranged from 24.4 to 202  $\mu\text{g}/\text{kg}$ , with an average of 75.2  $\mu\text{g}/\text{kg}$ .



**Figure 2.** The Hg content in surface and subsurface soil.

The background Hg value in China and Fujian province is 40 and 74  $\mu\text{g}/\text{kg}$  reported in [31], respectively. The background Hg value in the of whole Fujian province is 81  $\mu\text{g}/\text{kg}$  [32], and that in coastal Fujian is 63  $\mu\text{g}/\text{kg}$  [33]. In this study, the Hg content in soil was almost equal to the background value in Fujian province, and approximately twice higher than that in China. However, the average Hg content in our study area did not exceed the soil screening value listed in GB15618-2018 [34]. The difference among the background values as mentioned above and that in this study is mainly due to soil parent material, parent rock and soil type [33].

We selected 30 surface and 30 subsurface soil samples for sieving into five different soil size fractions (Section 2.3.1). The Hg contents of the surface and subsurface soil in five different soil size fractions ( $-10\sim+20$  mesh,  $-20\sim+60$  mesh,  $-60\sim+120$  mesh,  $-120\sim+230$  mesh, and  $-230$  mesh) are shown in Figure 3. We found a strong correlation between soil Hg content and soil size. Overall, the finer the soil particles, the higher the Hg content. The same result was found in a Hg mining area, which was attributed to the high sorption capacity for Hg in the finest grain size fractions [35]. The Hg contents in the same soil size fraction in the surface and subsurface soils were comparable.



**Figure 3.** Hg content in different soil size fractions. F1 = < 2000–850 µm; F2 = < 850–250 µm; F3 = < 250–125 µm; F4 = < 125–63 µm; F5 = < 63 µm.

We also compared the Hg content in the different soil size fractions after extraction. The results are listed in Table 3.

**Table 3.** Comparison of Hg content in different soil size after single extraction (average value, µg/kg).

Sample	Soil Size	AE	DAE	SSE	CAE
Surface soil	F1 (N = 30)	0.11	0.35	0.65	0.72
	F2 (N = 30)	0.10	0.34	1.00	0.80
	F3 (N = 30)	0.12	0.43	1.10	0.93
	F4 (N = 30)	0.11	0.42	1.31	1.26
	F5 (N = 30)	0.11	0.37	0.81	1.12
Subsurface soil	F1 (N = 30)	0.10	0.67	0.64	0.60
	F2 (N = 30)	0.10	0.67	0.69	0.66
	F3 (N = 30)	0.12	0.73	0.89	0.76
	F4 (N = 30)	0.14	0.59	1.08	0.90
	F5 (N = 30)	0.11	0.72	0.62	0.84

Notes: F1 to F5 represent for different soil size. F1 = < 2000–850 µm; F2 = < 850–250 µm; F3 = < 250–125 µm; F4 = < 125–63 µm; F5 = < 63 µm. AE = aqueous extraction, DAE = dilute acid extraction, SSE = salt solution extraction, CAE = complexing agent extraction.

We found almost no difference among the extracted Hg contents for the different soil size fractions when we used AE and DAE. For SSE or CAE, we observed the maximum extracted Hg content in soil size fraction F4. For the same soil size fractions, the maximum extracted Hg content mostly occurred in CAE. The results showed the difference in the extracted Hg content among the different soil particle sizes and extractants.

### 3.2. Hg Content in Tea Leaves

The average value, median, and range of the Hg content in all tea leaf samples in this study are listed in Table 4. We also collected some data from references about Hg content in different regions or countries for comparison with our results.



**Table 4.** Comparison of Hg content in tea leaves.

Sample	Hg Content ( $\mu\text{g/kg}$ )				Ref
	Min	Max	Ave	Med	
All samples ( $N = 79$ )	1.50	8.25	3.70	3.41	This study
Old leaf ( $N = 23$ )	4.16	8.25	5.59	5.44	
Young leaf ( $N = 56$ )	1.50	4.62	2.93	2.94	
Herbal tea ( $N = 100$ )	0.60–8.50	18.1–49.0	12.0–21.0		[36]
Black teas ( $N = 10$ )	0.82	5.11		2.20	[26]
Green teas ( $N = 14$ )	1.19	4.45		2.73	[37]
Teas ( $N = 8$ )				80	[38]
Green teas ( $N = 8$ )	1.05	2.8	1.77	1.60	[24]
Green teas ( $N = 7$ )	2.20	7.0			[25]
Black teas ( $N = 15$ )	0.94	9.35			
Green teas ( $N = 6$ )	230	690			

Notes: Min = Minimum, Max = Maximum, Ave = Average, Med = Median, Ref = Reference.

In our study, the average Hg content in tea leaves was 2.93–5.59  $\mu\text{g/kg}$ , which is far lower than the limit (300  $\mu\text{g/kg}$ ) provided in China's Agricultural Industry Standards [39]. We also found that the Hg content in old leaves was substantially higher than that in young leaves, indicating the accumulation of Hg in old leaves.

The Hg content in the Nitra region of Slovakia was studied [36]. The minimum, maximum, and average Hg content in the samples from different years were 0.60–8.50, 18.1–49.0, and 12.0–21.0  $\mu\text{g/kg}$ , respectively. All these values are lower than the standard limit (50  $\mu\text{g/kg}$ ) provided in the Codex Alimentarius of The Slovak Republic. The Hg contents in tea collected from China, India, and other places were determined [26]. The results showed that the Hg content in green and black tea was  $2.7 \pm 1.0$  and  $2.2 \pm 1.4$   $\mu\text{g/kg}$ , respectively. Researchers [38] determined the Hg content of eight kinds of green tea collected from a local market. The Hg content ranged from 1.05 to 2.80  $\mu\text{g/kg}$ . The Hg content in tea leaves in a suburban Hg-polluted area in southeast China was  $80 \pm 40$   $\mu\text{g/kg}$  [37]. The Hg levels in green teas from India, China, Japan, and Italy ranged from 230 to 690  $\mu\text{g/kg}$  [25].

Compared with all the values in Table 4, the Hg content in our study is not significantly different, but is significantly lower than in two studies [25,37]. The results showed that Hg content of tea produced in different regions might regionally differ.

### 3.3. Relationship of Total Hg in Soil and Tea Leaves

#### 3.3.1. Correlation

The correlation coefficients of the Hg content between tea leaves and soil are listed in Table 5, none of which were significant.

**Table 5.** The correlation coefficient of Hg content between soil and tea leaf.

	Tea and Surface Soil	Tea and Subsurface Soil	Old Leaf and Surface Soil	Old Leaf and Subsurface Soil	Young Leaf and Surface Soil	Young Leaf and Subsurface Soil
correlation coefficient	−0.097	−0.125	−0.033	−0.093	−0.047	−0.122

The possible reasons for this result are follows: The amount of Hg that can be absorbed by tea leave is not dependent on the total Hg in soil. In other words, the uptake of Hg by plants depends upon the plant species, Hg speciation in the soil, and soil properties [4]. This means that the relationship between the total Hg in the soil and the bioavailable Hg is complex. The total Hg content may not objectively reflect the bioavailable content. The absorption and transport of Hg in plants are also related to crop varieties and genotypes, soil element forms, and element sources.

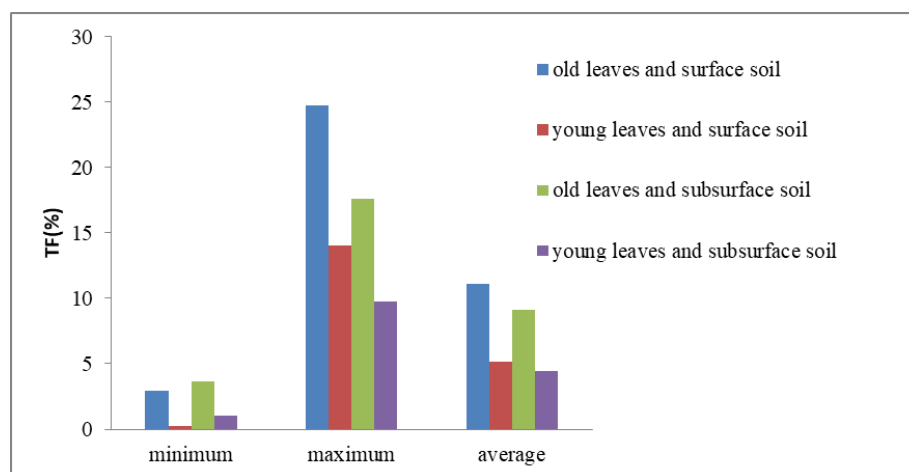
### 3.3.2. Transfer Factor

In this study, we used the transfer factor (TF) between soil and crops to represent the ability of tea leaves to absorb the Hg from soil.

$$TF (\%) = (A_i/B_i) \times 100\% \quad (1)$$

In Formula (1),  $A_i$  is the content of element  $i$  in the crop, and  $B_i$  is the content of element  $i$  in the soil.

The TF values for Hg from soil to tea are shown in Figure 4. Figure 4 shows the following: (1) The range of TF for both old and young leaves was wide. The plant species and physical and chemical properties of soil can affect the absorption of soil elements by plants [19,40]. The concentration, source, and bioavailability of an element in soil can also affect plant absorption of elements from the soil. In this study, we collected the tea leaf and soil samples from 69 tea gardens. The geographical span and altitudes of the sampling area, parent materials of the soil, soil types, and physical and chemical properties of various tea gardens widely differed, resulting in a large variation in TF.



**Figure 4.** TF of Hg from soil to tea leaves.

(2) The TF of Hg was higher in old than in young leaves. Similar to other crops, the transport capacity of Hg from root to leaves in tea is weak, which implies a natural biological barrier to Hg transport from the soil into tea leaves. Because Hg is not an essential nutrient element for plant growth, it mainly enters plants through passive processes such as transpiration. As the tea plant grows, Hg gradually accumulates in tea leaves.

### 3.4. Extracted Hg in Soil and Total Hg in Tea Leaves

In general, the correlation of the content of an extracted element between soil and plant is a basic criterion used for selecting a suitable extraction method simulating the transfer of the element from the soil to the plant. The correlation coefficients of the relationship between the extracted Hg content in the surface soil and the Hg content in young leaves are listed in Table 6. Table 6 shows that all the correlation coefficients between the extracted Hg content in surface soil and the Hg content in young tea leaves with different extraction methods were not in statistically significant. Extraction with dilute acid (DAE) was one of the most effective methods because it most accurately reflected the bioavailability of Hg in young tea leaves despite the lack of observed significant correlations.



**Table 6.** Correlation coefficients between the extracted amount of Hg and the Hg content in young leaves (N = 27).

Soil Size	AE	DAE	SSE	CAE
F1	0.166	0.229	0.160	0.002
F2	−0.288	0.256	0.077	0.020
F3	0.064	0.160	0.111	−0.040
F4	0.001	0.300	0.110	0.064
F5	−0.147	−0.181	0.183	−0.013

Notes: F1 to F5 represent for different soil size. F1 = < 2000~850  $\mu\text{m}$ ; F2 = < 850~250  $\mu\text{m}$ ; F3 = < 250~125  $\mu\text{m}$ ; F4 = < 125~63  $\mu\text{m}$ ; F5 = < 63  $\mu\text{m}$ . AE = aqueous extraction, DAE = dilute acid extraction, SSE = salt solution extraction, CAE = complexing agent extraction.

### 3.5. Correlation between Extracted Hg and Other Indicators

We calculated the correlation coefficient between the extracted amount of Hg and other indicators in surface and subsurface soil (soil size = F1). The results are presented in Table 7.

**Table 7.** Correlation coefficient between extraction amount of Hg and other indicators (N = 30).

	HgS	HgL	TF	SOC	TC	pH	CEC	HgAE	HgDAE	HgSSE	HgCAE
HgS		−0.080	−0.454 *	0.127	0.050	−0.203	−0.032	−0.144	−0.173	−0.125	−0.161
HgL	0.273		0.561 **	0.414 *	0.451 *	0.529 **	−0.017	0.101	0.289	0.472 **	0.413 *
TF	−0.681 **	0.412 *		−0.028	0.015	0.485 **	−0.034	0.005	−0.142	−0.063	−0.009
SOC	0.459 *	0.450 *	−0.219		0.955 **	0.202	0.276	0.031	−0.012	0.394 *	0.688 **
TC	0.440 *	0.462 *	−0.206	0.992 **		0.214	0.445 *	−0.001	0.079	0.454 *	0.708 **
pH	0.053	0.544 **	0.287	0.327	0.326		−0.169	−0.062	−0.026	0.108	0.157
CEC	−0.150	0.004	−0.006	0.225	0.263	−0.265		−0.042	0.117	0.213	0.188
HgAE	0.320	−0.163	−0.284	0.090	0.104	−0.158	0.286		0.127	0.225	−0.015
HgDAE	0.072	0.382 *	0.136	0.024	0.023	0.223	0.211	−0.077		0.413 *	0.175
HgSSE	0.627 **	0.391 *	−0.322	0.415 *	0.371 *	0.268	−0.074	0.090	0.301		0.613 **
HgCAE	0.586 **	0.429 *	−0.269	0.549 **	0.518 **	0.162	0.147	0.142	0.416 *	0.796 **	

Notes: The black text stand for the result in surface soil, while blue text represent the result in sub-surface soil. HgS = Hg content in soil, HgL = Hg content in tea leaf, TF = Transfer Factor, SOC = soil organic carbon, TC = total carbon in soil, HgAE = Hg content extract with AE, HgDAE = Hg content extract with DAE, HgSSE = Hg content extract with SSE, HgCAE = Hg content extract with CAE. \* indicates significantly correlated at  $p = 0.95$  level; \*\* means significantly correlated at  $p = 0.99$  level.

In the surface soil, the Hg content in tea was statistically significantly positively correlated to SOC ( $r = 0.414 *$ ), TC ( $r = 0.451 *$ ), and pH ( $r = 0.529 **$ ). In the subsurface soil, similar correlations were found. The Hg content in tea leaf was statistically significantly positively correlated to SOC ( $r = 0.450 *$ ), TC ( $r = 0.462 *$ ), and pH ( $r = 0.544 **$ ). The results indicated that the soil properties affected the Hg content in the tea leaves.

In addition, the Hg content in tea leaves was statistically significantly positively correlated to HgSSE ( $r = 0.472 **$ ) and HgCAE ( $r = 0.413 *$ ) of the surface soil. For subsurface soil, the Hg content in tea leaves was statistically significantly positively correlated to HgDAE ( $r = 0.382 *$ ), HgSSE ( $r = 0.391 *$ ) and HgCAE ( $r = 0.429 *$ ). This means that the Hg content in tea leaves (both young and old leaves) was closely related to the different forms of Hg in the soil.

## 4. Discussion

### 4.1. Factors Influencing Hg Content in Soil

#### 4.1.1. Effect of Geological Background on Soil Hg Content

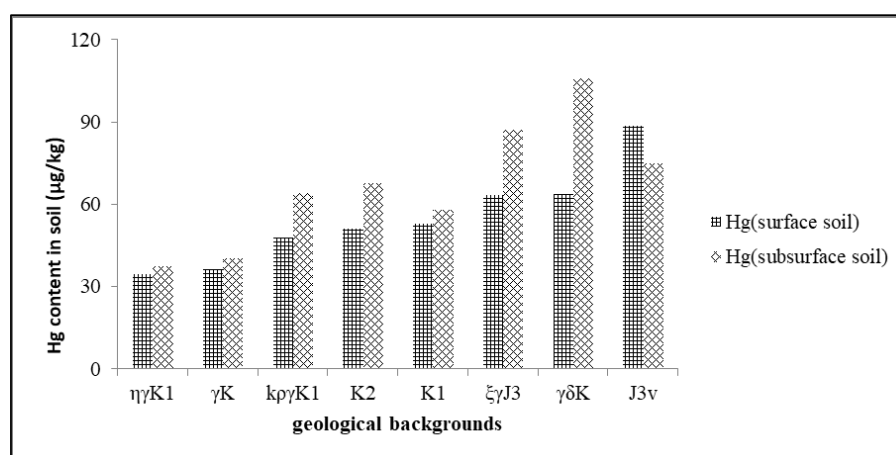
In this study, the main geological backgrounds were  $\text{J}_3^v$  (intermediate-acid volcanic rock and pyroclastic rock in upper Jurassic),  $\xi\gamma\text{J}_3$  (syenogranite in upper Jurassic), and  $\text{K}_2$  (clastic rocks, marlstone and quartz rock in upper Cretaceous); 72 samples had these geological backgrounds. The others five geological backgrounds included  $\eta\gamma\text{K}_1$ ,  $\kappa\gamma\text{K}_1$ ,  $\gamma\delta\text{K}$ ,  $\gamma\text{K}$ , and  $\text{K}_1$  for just seven samples.

We calculated the contents of Hg and other elements in the soil samples according to the geological background (Table 8). As shown in Table 8 and Figure 5, we found that (1): the Hg content in soil with a  $J_3^v$  parent rock (intermediate-acid volcanic rock and pyroclastic rock in upper Jurassic) was higher than in  $\xi\gamma J_3$  parent rock (syenogranite in upper Jurassic). (2) In both the subsurface and surface soil, the element contents widely varied with geological era and parent rock; most of them had a difference of 2–4 times between the maximum and minimum values. For instance, the maximum  $Na_2O$  content was 10 times higher than its minimum.

**Table 8.** Concentration of element/compounds and value of soil property in different geological backgrounds.

Geological Backgrounds		J <sub>3</sub> <sup>v</sup>	ξγJ <sub>3</sub>	ηγK <sub>1</sub>	kργK <sub>1</sub>	γδK	γK	K <sub>1</sub>	K <sub>2</sub>	all	Max/Min
Sample number		54	14	2	2	1	1	1	4	79	
Surface soil	Hg	88.5	63.3	34.7	47.9	63.9	36.4	52.9	51.0	78.3	2.6
	SiO <sub>2</sub>	63.9	64.9	54.9	54.1	51.2	61.4	72.0	57.4	63.2	1.4
	Al <sub>2</sub> O <sub>3</sub>	17.7	17.9	22.2	21.6	25.5	18.7	13.6	19.7	18.0	1.9
	TFe <sub>2</sub> O <sub>3</sub>	4.28	2.65	5.20	5.77	3.55	4.49	3.46	5.32	4.09	2.2
	MgO	0.26	0.16	0.33	0.34	0.18	0.43	0.18	0.34	0.25	2.8
	CaO	0.09	0.11	0.05	0.14	0.04	0.30	0.10	0.09	0.09	7.0
	Na <sub>2</sub> O	0.07	0.19	0.07	0.07	0.02	0.23	0.05	0.10	0.09	10.0
	K <sub>2</sub> O	1.56	2.42	1.32	1.34	0.48	2.41	1.45	1.33	1.69	5.1
	SOC	1.34	0.87	0.73	1.20	0.77	1.40	1.75	1.26	1.23	2.4
	TC	1.50	1.01	0.79	1.28	1.05	1.41	1.84	1.42	1.38	2.3
	pH	4.6	4.9	5.0	4.8	4.5	4.9	4.9	4.8	4.7	1.1
	CEC	8.27	5.94	6.96	6.00	11.2	7.97	8.46	9.95	7.89	1.9
Subsurface soil	Hg	74.9	87.1	37.4	64.2	106	40.4	57.9	67.9	75.2	2.9
	SiO <sub>2</sub>	63.9	65.4	55.5	51.6	49.9	62.0	72.2	57.9	63.3	1.4
	Al <sub>2</sub> O <sub>3</sub>	17.7	17.9	21.8	22.3	25.9	18.3	13.9	20.4	18.2	1.9
	TFe <sub>2</sub> O <sub>3</sub>	4.27	2.79	5.20	6.07	3.41	4.43	3.70	5.37	4.12	2.2
	MgO	0.28	0.15	0.32	0.33	0.19	0.45	0.18	0.35	0.26	2.9
	CaO	0.09	0.11	0.05	0.13	0.04	0.30	0.07	0.09	0.09	7.0
	Na <sub>2</sub> O	0.07	0.18	0.06	0.07	0.02	0.23	0.05	0.10	0.09	10.0
	K <sub>2</sub> O	1.61	2.44	1.14	1.20	0.54	2.44	1.46	1.40	1.72	4.5
	SOC	1.10	0.82	0.52	0.96	0.82	1.13	1.18	1.17	1.03	2.2
	TC	1.18	0.88	0.57	1.06	0.91	1.17	1.35	1.20	1.11	2.4
	pH	4.6	4.9	4.9	4.7	4.4	4.9	4.7	4.7	4.7	1.1
	CEC	8.39	5.69	6.44	7.88	8.22	7.73	5.86	11.4	7.96	2.0

Notes:  $J_3^v$ : Intermediate-acid volcanic rock and pyroclastic rock in upper Jurassic,  $\xi\gamma J_3$ : syenogranite in upper Jurassic,  $\eta\gamma K_1$ : monzonitic granite in Lower Cretaceous,  $k\rho\gamma K_1$ : alkali-feldspar granite in Lower Cretaceous,  $\gamma\delta K$ : Cretaceous granodiorites,  $\gamma K$ : Cretaceous granite,  $K_1$ : Intermediate-basic-intermediate-acid volcanic rocks and volcanic breccia in Lower Cretaceous,  $K_2$ : clastic rocks, marlstone and quartz rock in upper Cretaceous. Unit:  $SiO_2$ ,  $Al_2O_3$ ,  $TFe_2O_3$ , MgO, CaO,  $Na_2O$ ,  $K_2O$ , SOC and TC are “ $10^{-2}$ ”; CEC is cmol(+)/kg; pH: with no unit; Hg is “ $\mu\text{g}/\text{kg}$ ”.



**Figure 5.** Comparison of Hg contents in soil with different geological backgrounds.

Geological background and the chemical composition of parent rocks are important factors affecting soil element contents.

#### 4.1.2. Effect of Soil Geochemical Properties on Soil Hg Content

Certain chemical indicators in soil affect each other because of their geochemical characteristics. In this study, we calculated the correlation coefficients in surface soil between some major compounds and Hg; the results are shown in Table 9.

**Table 9.** The correlation coefficient between some major compounds and Hg (N = 79).

	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	TFe <sub>2</sub> O <sub>3</sub>	MgO	CaO	SOC	pH	CEC
Hg	0.039	−0.057	0.008	−0.059	−0.012	0.116	−0.196	0.012

As shown in Table 9, we did not find any significant correlation between Hg and other indicators. However, we found a negative correlation between Hg content and pH ( $r = -0.196$ ) and a positive correlation between Hg and organic carbon contents ( $r = 0.116$ ). The results in Table 9 are a little different from those in Table 7 because the original data that we used to calculate the correlation coefficient were different. The original data used for Table 7 were obtained from the analysis of the 30 surface soil samples that we had randomly selected from the 79 surface soil samples. However, the original data used for Table 9 were obtained from the analysis of all the 79 surface soil samples. Ninkov J et al. [41] reported that Hg content is statistically significantly negatively correlated to pH ( $r = -0.062^*$ ) and statistically significantly positively correlated to organic matter content ( $r = 0.191^{**}$ ). Several soil factors, such as organic matter (OM), pH, and microbial activity, affect Hg speciation and bioavailability in soil [4]. Our result is consistent with this, implying that the total Hg content in surface soil may be restricted by pH value and organic carbon content.

#### 4.2. Factors Influencing Bioavailability of Hg in Soil–Tea Plant System

According to our results, we found that the Hg contents in the soil–tea plant system were not significantly correlated. The Hg content in tea leaves positively correlated with the contents of K<sub>2</sub>O, Na<sub>2</sub>O, and Al<sub>2</sub>O<sub>3</sub>, and pH value, but negatively correlated with the SiO<sub>2</sub> content in the soil. This finding indicated that the Hg content in tea leaves was not entirely dependent on the total amount of Hg in soil but was more likely related to the available amount of Hg in the soil. However, the amount of available Hg in the soil depends on the influence of various factors such as soil physical and chemical properties, so the process of Hg absorption from soil to tea plant is complicated. Tangahu et al. [42] reviewed the mechanism through which heavy metals are uptake by plants from soil. In this review, plant species, the properties of the medium, root zone, environmental conditions, chemical properties of the contaminant, and bioavailability of the metal and chelating agent were all considered factors that can affect heavy metal absorption and accumulation in plants.

Some examples prove the statements mentioned above. For instance, one study [43] reported that rape (*Brassica napus* L.) had the highest TF<sub>THg</sub> and TF<sub>MHg</sub> values, whereas the lowest TF<sub>MHg</sub> value occurred in cauliflower (*Brassica oleracea* L. var. *botrytis* L.). Reference [44] reported the distribution and bioavailability of Hg in vegetable soils in Jiulong River, Fujian province. In this study, the Hg contents of 22 crop samples (including 10 crop species) ranged from 0.23 to 2.90 µg/kg, but the highest Hg content was produced from the soil with the lowest Hg content. In surface soil, the Hg content in tea leaves was statistically significantly positively correlated to SOC ( $r = 0.414^*$ ), TC ( $r = 0.451^*$ ), and pH ( $r = 0.529^{**}$ ). In subsurface soil, the Hg content in tea leaves was statistically significantly positively correlated to SOC ( $r = 0.450^*$ ), TC ( $r = 0.462^*$ ), and pH ( $r = 0.544^{**}$ ) (Table 7). This provides evidence that soil properties affect the bioavailability of Hg. Sun et al. [45] studied the bioavailability of 22 heavy metals in 19 Tieguanyin tea plantations in Fujian. The results showed that Cu, Pb, and Cd were highly bioavailable and most easily absorbed by tea trees. This indicated that some differences in bioavailability exist among different heavy metals.

Furthermore, when metals are bound to the soil, the pH, redox potential, and organic matter content all affect the tendency of the metal to exist in ionic and plant-available forms. Plants affect the soil through their ability to lower the pH and oxygenate the sediment, which affect the availability of metals [46].

The Hg concentration in leaves also indicates that the uptake of gaseous Hg by foliage is the predominant pathway through which Hg accumulated in corn plant. Researchers [14] found the highest Hg in corn leaves near the zinc melt area supported this view. In addition, some experimental findings demonstrated that nearly all of the Hg in the leaves of both herbaceous and woody plants derived from the atmosphere [17,18,47]. For example, authors [18] reported that the Hg concentration in the air was the primary factor controlling foliar Hg concentrations. The experimental result indicated that for areas with Hg-enriched air and soil with background Hg concentrations, such as industrial sites, atmospheric Hg as a source of Hg in foliage must be considered. Because of this, we did not expect to find a significant correlation relationship of the Hg content between the soil and tea leaves.

## 5. Conclusions

In this study, we collected soil and tea leaf samples from tea gardens in the Anxi region of China. We studied the Hg content distribution characteristics and bioavailability in the soil and tea plant systems, and the results can be summarized as follows:

(1) The Hg content in the soil in the study area was equivalent to that of the background value in Fujian province, which is about 2.6–2.9 times higher than the national background value in soil. We found a noticeable difference in the Hg content between soils with different geological backgrounds (parent rock of soil formation). In addition, the pH value and organic carbon content in the soil may have restricted the total Hg content in soil.

(2) The Hg content in tea leaves in this study was not much different from that observed in other studies. Compared with young leaves, old leaves had a higher Hg content. The transfer factor (TF) of Hg between the soil and plants ranged from 0.25% to 24.76%, indicating a low bioavailability of Hg in tea leaves.

(3) When we considered all paired soil–tea leaf samples collected from the study area red (N = 79), the correlation coefficients between the total Hg in the surface soil and the Hg content in tea leaves were not significant. However, we found a statistically significant positive correlation between the Hg content in tea leaves and the extracted amount of Hg (such as Hg extracted with SSE or CAE) in both surface and subsurface soil. In addition, we observed a statistically significant positive correlation between the Hg content in the tea leaves and SOC and pH in surface soil (N = 30). This implies a close relationship among the Hg content in tea leaves and the amount of Hg extracted from soil and soil properties.

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