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Original Research

Element Migration in the Bedrock-Soil-Tea System of Metamorphic Regions and Its Implications for Tea Quality

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Abstract

The migration and enrichment of mineral elements within the bedrock-soil-tea system are critical determinants of tea quality. Understanding how lithological background regulates soil nutrient dynamics and tea biochemical traits is essential for optimizing plantation management in metamorphic regions. This study investigated element transfer and enrichment patterns in the "rock-soil-tea" system of metamorphic tea plantations in Guizhou Province, China. Field sampling and geochemical analyses were integrated with redundancy analysis (RDA), random forest (RF), and structural equation modeling (SEM) to evaluate their effects on key quality components. Metamorphic slate exhibited higher concentrations of major, lithophile, and chalcophile elements, whereas metasandstone showed relative enrichment of fluid-mobile elements. Weathering induced significant leaching of base cations, particularly K_2O and MgO, resulting in their depletion in soil profiles. Tea plants demonstrated selective bioaccumulation, enriching beneficial nutrients such as P and Mn while restricting the transfer of toxic metals (As, Pb, Cr, Hg). SEM revealed a strong direct effect of bedrock composition on the soil trace element pool (path coefficient = 0.92, $p<0.001$), which subsequently influenced plant uptake (path coefficient = 0.64, $p<0.001$). RDA and RF consistently identified soil potassium (K) as the primary factor associated with free amino acid and caffeine synthesis. Mass balance calculations further indicated substantial K loss through leaching, suggesting that K supplementation is necessary to enhance tea quality. These findings demonstrate hierarchical controls of lithological inheritance, pedogenic redistribution, and plant selectivity on tea quality, providing a scientific basis for nutrient management in metamorphic tea-growing regions.

Keywords: tea, metamorphic geology, mineral elements, tea quality, agrogeochemistry

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Introduction

Tea (*Camellia sinensis L.*), one of the world's most popular health beverages, possesses high economic value and is widely cultivated across Asia, Africa, and Latin America [1, 2]. The global tea cultivation area is 4.1×10^6 hm^2, with China accounting for 54.6% of this total [3], making it the world's largest tea producer [4]. Guizhou Province, which has the largest tea cultivation area in China, features karst mountainous terrain and a specific climate that enhances tea flavor [5]. As a core production region for high-quality green tea in China [6], the quality and safety of tea from Guizhou are major concerns within the industry.

Tea quality is largely determined by the concentration of secondary metabolites, including free amino acids (FAA), caffeine (CA), tea polyphenols (TP), and soluble proteins (SP) [6-9]. CA, TP, and their derivatives contribute to bitterness and astringency, whereas FAA primarily imparts umami and sweetness [10]. The ratio of TP to FAA (TP/FAA) reflects the balance between astringency and mellowness and is widely used as an indicator of sensory quality [11]. Catechins, amino acids, and tea polyphenols also provide health benefits, including protective effects against neurological diseases, cancer, cardiovascular disorders, and obesity [3, 12]. Mineral nutrients in tea leaves are essential not only for the synthesis of these metabolites but also for human nutrition [13]. For instance, adequate macronutrients and Zn in leaves significantly enhance the accumulation of polyphenols, FAA, and caffeine [14], and exchangeable Cu is recognized as a key factor affecting tea quality [8]. Potassium (K) plays a central role in soil fertility and plant nutrition [1]. It is involved in the Hill reaction associated with NADPH and ATP production, participates in CO_2 fixation within the Calvin–Benson cycle, and regulates carbohydrate synthesis, transport, and macromolecule formation [15]. Therefore, mineral element composition is of critical importance to tea quality and safety [16].

The uptake efficiency of mineral elements by tea plants depends on their distribution and bioavailability in soils, which are fundamentally controlled by mineral release during bedrock weathering [17]. Tea quality is closely linked to the evolution of elements during soil formation from parent rock. Plants accelerate mineral weathering through soil anchoring, alteration of hydrological processes, secretion of organic acids and chelating agents, and reduction of rhizosphere pH, thereby facilitating nutrient acquisition [18]. Elucidating element migration within the "rock-soil-plant" continuum allows tracing the material basis of plant growth and product quality from the source [19]. As a plant highly sensitive to environmental conditions, tea accumulates mineral elements that not only reflect its nutritional value but also record the geological background of its growth environment as a geochemical "fingerprint" [20]. Therefore, systematically clarifying how geological background mediates soil processes and ultimately regulates tea mineral composition and quality formation has both theoretical and practical significance.

Many renowned high-quality tea-producing regions worldwide, such as Wuyi Mountain in China and the Fanjing Mountain and Leigong Mountain areas in Guizhou, considered in this study, are closely associated with the distribution of metamorphic rocks [20]. Metamorphic rocks are formed from pre-existing igneous or sedimentary rocks under solid-state mineralogical and structural transformation induced by temperature and pressure changes during tectonic or magmatic processes. Such metamorphism is often accompanied by element redistribution, migration, and enrichment, resulting in distinctive elemental compositions in both metamorphic rocks and their derived soils [21]. The process may activate or release beneficial trace elements (e.g., Se, Mo, Zn) or generate specific mineral assemblages that gradually release nutrients into the soil solution during weathering, thereby providing a balanced and sustained nutrient supply for tea plants [22]. However, systematic studies on the geochemical behavior of elements within the "rock-soil-tea" system under metamorphic geological settings remain limited worldwide.

Previous studies have extensively investigated mineral elements in soils and tea from carbonate rock regions of Guizhou [23, 24]. In contrast, the relationships between geochemical characteristics of metamorphic regions and tea biochemical quality are still insufficiently understood. Moreover, element transfer studies within soil-crop systems often rely on single-factor correlation analysis and rarely address multi-element interactions and their integrated influence on tea quality. Therefore, this study focuses on the geochemical characteristics of mineral elements in the rock-soil-tea system of metamorphic regions in Guizhou and their systematic control over tea quality. The objectives are: (1) to characterize the geochemical features of mineral elements in bedrock, soils, and tea leaves and to explore their variation with soil depth; (2) to evaluate enrichment factors (EF) from rock to soil and bioaccumulation factors (BAF) from soil to tea, revealing element interactions within the growth environment; and (3) to identify key soil elements driving tea quality based on redundancy analysis (RDA), random forest (RF), structural equation modeling (SEM), and Mantel tests, thereby providing a scientific basis for soil improvement, high-quality tea production, and sustainable development of the tea industry in Guizhou Province.

Materials and Methods

Study Area

The study area is located in the tea-producing regions of Fanjing Mountain and Leigong Mountain,

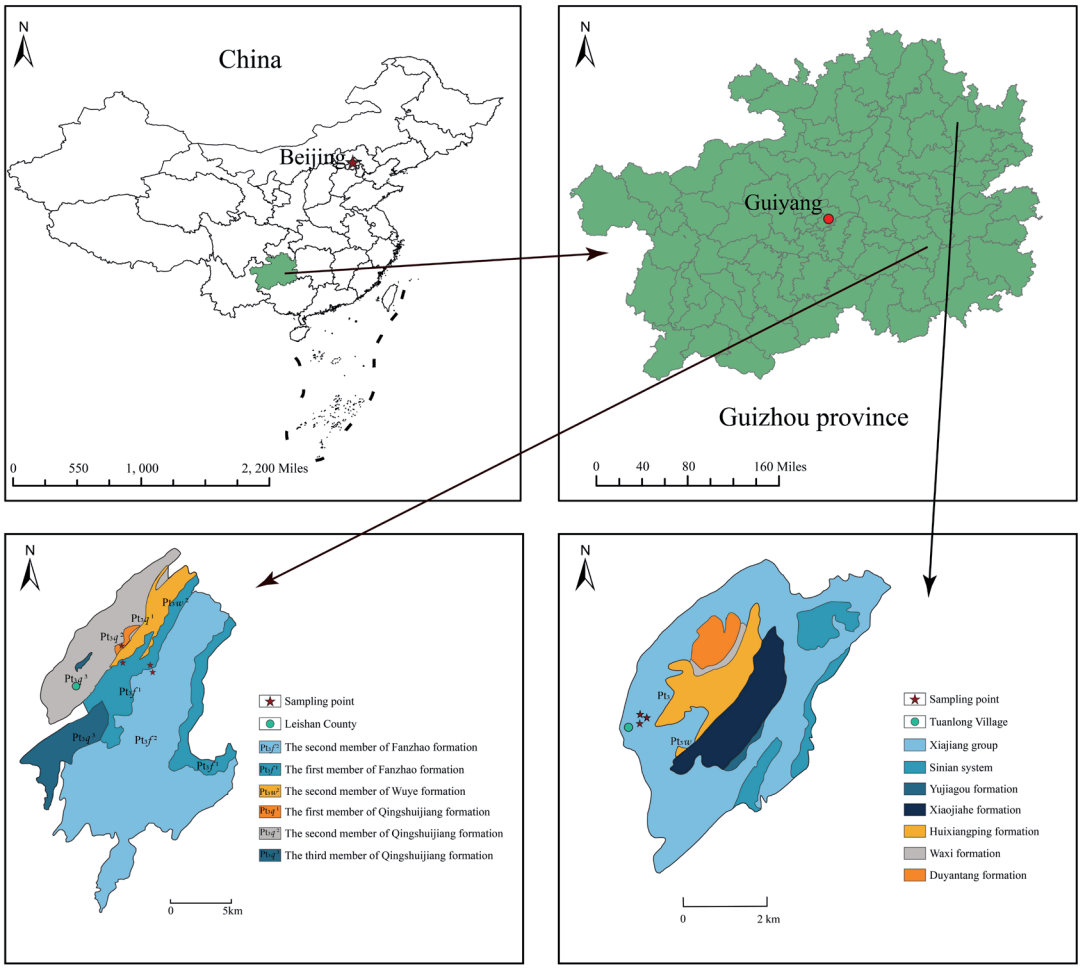


Fig. 1. Geological map of the tea-producing areas in Fanjing Mountain and Leigong Mountain, Guizhou Province.

Guizhou Province, China (Fig. 1). The Fanjing Mountain tea area extends from 108°45′ to 108°48′E and 27°49′ to 28°1′N, whereas the Leishan tea area is situated between 107°55′-108°22′E and 26°02′-26°34′N. The region is characterized by a humid subtropical monsoon climate with warm temperatures and abundant rainfall. The exposed strata in the Fanjing Mountain area mainly belong to the Neoproterozoic Xiajiang Group, including the Jialu (Pt_3j) and Wuye (Pt_3w) Formations. In the Leigong Mountain area, the dominant exposed strata are the Fanzhao (Pt_3f) and Qingshuijiang (Pt_3q) Formations of the Xiajiang Group. Seven soil profiles were collected from tea plantations in Fanjing Mountain (TL, DSX1, DSX2) and Leishan County (JY1, JY2, DL, and WF). Detailed information on geographic coordinates, elevation, bedrock type, soil type, and profile is provided in Table 1.

Sample Collection and Pretreatment

Rock, soil, and tea samples were collected from the same tea plantations to ensure consistency within the rock-soil-tea system. Fresh bedrock samples were obtained from the base of each soil profile, with approximately 1 kg collected per site. Soil samples were collected at 10 cm intervals from bottom to top along each profile, yielding a total of 49 samples. Approximately 1 kg of soil was obtained at each depth after removing visible roots and debris. Tea samples were collected adjacent to each soil profile. At each site, one composite tea sample was prepared by combining three subsamples (500 g each) harvested in spring at the standard “one bud and two leaves” stage. Rock samples were rinsed with deionized water to remove adhering particles and oven-dried at 105ºC. Soil samples were air-dried, ground, and sieved through a 200-mesh nylon sieve. Fresh tea leaves were rinsed with deionized water, air-dried, and steam-fixed to inactivate enzymes. The leaves were first dried at 50ºC for 2 h and subsequently at 30ºC until constant weight. Dried tea samples were ground and sieved (200 mesh). All processed samples were stored in polyethylene bags prior to chemical analysis.

Chemical Analysis

The concentrations of multiple elements in the samples were measured using inductively coupled plasma mass spectrometry (ICP-MS; Visata-MPS, Agilent, USA) and inductively coupled plasma atomic

Table 1. Description of the sampling sites.

Profile	Longitude and latitude	Altitude (m)	Bedrock type	Stratum	Soil type	Section length (cm)
JY1	N26°25′21″	1351	Slate	Pt_3f^2	Luvisol	50
JY2	N26°25′21″	1381	Slate	Pt_3f^1		50
TL	N27°55′18″	1104	Slate	Pt_3w		70
DSX1	N27°57′10″	1232	Metasandstone	Pt_3j		100
DSX2	N27°56′30″	1237	Metasandstone	Pt_3j		180
DL	N26°26′6″	1253	Metasandstone	Pt_3f^1		120
WF	N26°24′15″	1221	Metasandstone	Pt_3q^1		90

Note: Pt_3j, Jialu Formation; Pt_3w, Wuye Formation; Pt_3f, Fanzhao Formation; Pt_3q, Qingshuijiang Formation. Superscripts 1, 2, and 3 indicate the lower, middle, and upper members of the corresponding formation, respectively.

emission spectrometry (ICP-AES; 7700x, Agilent, USA) at the ALS Mineral Laboratory (Guangzhou, China). Detailed operating conditions and analytical procedures for both instruments are described elsewhere [25]. For tea sample digestion, a 0.2 g aliquot of powdered tea is digested with a mixture of concentrated HNO_3 and HF (3:1, v/v) in a polytetrafluoroethylene vessel for 8 h, followed by stepped heating on a hotplate at 50ºC for 1 h, 100ºC for 1.5 h, and 150ºC for 1.5 h. Additional nitric acid is added as necessary to achieve complete digestion. After cooling to room temperature, the residue is dissolved with 2% HCl and made up to a final volume of 25 mL in a volumetric flask. For soil and rock samples, 0.25 g of powder is digested with a concentrated acid mixture ($HClO_4$: HNO_3: HF: HCl = 1:2.5:2:2.5, by volume) at approximately 190ºC for 48 h. The digested sample is then heated on a preheated hotplate (150ºC) to remove excess acids until crystalline solids are formed, before being diluted to a constant volume of 12.5 mL with 2% HCl. The free amino acid (FAA) content in tea was determined by a modified ninhydrin colorimetric method [26]. The tea polyphenol (TP) content was measured according to the Folin-Ciocalteu method [27]. The caffeine (CA) content was quantified using ultraviolet (UV) spectrophotometry [28]. The soluble protein (SP) content was determined by the Coomassie Brilliant Blue G-250 staining method [16].

Methodology

The enrichment factor (EF_i) was used to evaluate elemental migration and enrichment relative to fresh bedrock [29]:

$$EF_i = \frac{(C_i/C_{Al})_{soil}}{(C_i/C_{Al})_{rock}} - 1 \quad (1)$$

Where *soil* and *rock* represent the concentrations of element i in soil and bedrock, respectively, and C_{Al} is the concentration of Al used as a reference element. When EF_i>0, the element is enriched in soil; when EF_i<0, the element is depleted during weathering and pedogenesis; $EF_i = -1$ indicates complete loss [30, 31].

The bioaccumulation factor (BAF) was calculated to quantify elemental transfer from soil to tea leaves [32]:

$$BAF = C_{tea} \diagup C_{soil} \quad (2)$$

Where C_{tea} and C_{soil} denote the concentrations of element i in tea leaves and corresponding soils, respectively. BAF>1 indicates enrichment in tea leaves [33].

Redundancy analysis (RDA) was performed using CANOCO 5 to evaluate relationships between soil elemental concentrations and tea quality parameters (FAA, TP, CA, and SP). Random forest (RF) analysis was conducted in R (version 4.4.2) using the "randomForest" and "rfPermute" packages.

Structural equation modeling (SEM) was carried out using AMOS 24 (IBM, USA) to quantify direct and indirect pathways within the rock-soil-tea system. Model fit was evaluated using the χ^2 statistic, goodness-of-fit index (GFI>0.9), and normed fit index (NFI>0.9) [31].

Mantel tests were performed using the "vegan" package in R to examine correlations between tea elemental concentrations and quality indicators. Statistical significance was set at $p<0.05$.

Quality Control

Strict quality control procedures were implemented throughout the analytical process. Method blanks, analytical duplicates, and certified reference materials (CRMs) were routinely inserted into each analytical batch. Relative deviations and relative errors were maintained within 10%. Tea samples were calibrated using internal reference materials SpL03 and GL03. Soil analyses were validated using internal standards GLG908 and MRGeo08. Recoveries of CRMs generally ranged from 80% to 120%. Analytical precision was evaluated using the standard deviation of replicate samples, which was <11% for tea samples and <15%

for soil samples. Heavy metal concentrations in blank samples were consistently below the method detection limits.

Results

Geochemical Characteristics of Elements in Bedrock, Soil Profiles, and Tea Leaves

Bedrock lithology exerted a primary control on elemental composition. Compared with the metasandstone DL, the slate sample JY2 (both belonging to Pt_3f^1) exhibited higher concentrations of Al_2O_3, Fe_2O_3, MgO, Na_2O, CaO, and most trace elements, including Li, Rb, Cs, Ga, Ni, Co, Mn, Zn, Pb, and V. In contrast, DL contained higher levels of K_2O, As, Hg, Bi, Se, Mo, Sb, and Th. Furthermore, pronounced heterogeneity was observed even within the same lithology and stratigraphic unit. For example, the CaO content in DSX1 reached 0.09%, whereas that in DSX2 (same lithology and formation) was only 0.01% (Table 2), reflecting localized variability in parent material composition.

Distinct vertical distribution patterns were identified in the soil profiles (Fig. S1). Cd, Pb, Zn, Hg, P, and Se were enriched in surface horizons and generally decreased with depth. In contrast, Al_2O_3, CaO, MgO, K_2O, Fe_2O_3, and Mn showed relatively higher concentrations in deeper layers, suggesting leaching from upper horizons and relative accumulation below. Elements such as Cr, Cu, Co, and Ni displayed minor variation along the profiles, indicating limited vertical mobility. Al_2O_3 exhibited low coefficients of variation (<15%), confirming its conservative behavior and suitability as a reference element (Table 3).

Tea leaves contained high concentrations of essential macronutrients, with mean values of K (18,000 mg/kg), P (5,689 mg/kg), S (3,200 mg/kg), Ca (2,614 mg/kg), Mg (1,729 mg/kg), Mn (430 mg/kg), Al (274 mg/kg), and Fe (130 mg/kg). Among trace elements, beneficial elements such as Zn, Cu, and Se exhibited relatively higher concentrations. Notably, the Se concentration in sample TL was higher than in the other samples, reaching 0.16 mg/kg (Table 2).

Element Migration in the Bedrock-Soil System and Element Enrichment in the Soil-Tea System

To quantitatively evaluate elemental mobility during weathering, enrichment factors (EF_i) were calculated relative to fresh bedrock (Fig. 2a) and b)). Potassium (K) exhibited depletion (EF_i<0) in nearly all soil profiles, indicating substantial leaching and nutrient loss during pedogenesis. Calcium (Ca) and magnesium (Mg) also showed partial depletion, whereas sodium (Na) loss was restricted to the lower horizons of DL and WF profiles. In contrast, Be, Hg, Nb, Ag, Mo, Se, W, Sn, Co, and P were significantly enriched in surface soils (EF_i>0).

Table 2. Concentrations of major and trace elements in bedrock and tea leaves.

Rock	JY1	JY2	TL	DSX1	DSX2	DL	WF	Tea	JY1	JY2	TL	DSX1	DSX2	DL	WF
Bedrock type	Sate			Metasandstone					Slate			Metasandstone			
Al	93.40	116.50	91.40	96.6	90.60	99.10	63.60	Al	0.26	0.35	0.26	0.22	0.20	0.35	0.28
Ca	0.30	0.20	1.60	0.90	0.10	0.10	0.10	Ca	2.80	3.10	3.20	2.50	2.40	2.30	2.00
Fe	31.30	35.90	15.80	56.00	39.90	30.80	13.60	Fe	0.10	0.13	0.12	0.09	0.07	0.30	0.11
K	24.30	32.00	33.80	29.10	42.20	34.30	1.50	K	18.90	19.10	18.40	17.10	17.80	16.90	17.80
Mg	3.90	4.20	4.80	5.20	8.30	2.60	0.90	Mg	1.68	2.01	1.66	1.58	1.59	1.77	1.81
Na	0.70	0.90	0.40	0.80	0.60	0.60	0.20	Na	0.10	0.10	0.04	0.02	0.01	0.08	0.07
S	0.10	0.20	0.30	0.30	0.30	0.30	0.30	S	2.93	3.42	3.66	3.16	3.23	2.92	3.08
Ag	0.01	0.01	0.00	0.04	0.03	0.00	0.00	Ag	0.00	0.00	0.00	0.00	0.00	0.00	0.00
As	12.50	7.20	12.00	53.70	40.90	10.05	9.63	As	0.03	0.37	0.14	0.13	0.05	0.12	0.03
Bi	0.12	0.07	0.25	0.65	0.42	0.08	0.16	Bi	0.00	0.00	0.01	0.01	0.01	0.01	0.00

Cd	0.04	0.03	0.05	0.08	0.04	0.02	0.01	Cd	0.05	0.04	0.09	0.06	0.05	0.04	0.03
Co	18.70	6.90	1.00	30.00	10.00	2.00	5.00	Co	0.32	0.15	0.11	0.34	0.34	0.28	0.27
Cr	31.00	28.00	56.00	90.00	47.00	25.00	22.00	Cr	1.57	2.29	0.88	1.31	0.81	9.89	1.78
Cs	7.33	6.18	1.20	4.38	1.94	0.89	1.24	Cs	0.15	0.12	0.15	0.06	0.10	0.11	0.42
Cu	28.20	22.50	14.00	49.00	41.00	8.00	9.00	Cu	17.65	16.10	13.20	14.10	15.70	18.70	14.20
Ga	21.60	24.10	1.64	7.85	3.79	4.46	3.72	Ga	0.06	0.03	0.04	0.04	0.03	0.06	0.05
Ge	0.14	0.14	0.09	0.08	0.13	0.04	0.07	Ge	0.01	0.03	0.03	0.02	0.02	0.03	0.01
Hg	0.02	0.01	0.04	0.20	0.01	0.02	0.03	Hg	0.03	0.13	0.07	0.00	0.00	0.08	0.03
Li	39.50	32.40	70.00	60.00	60.00	20.00	30.00	Li	0.11	0.06	0.05	0.04	0.04	0.19	0.04
Mn	889.00	441.00	68.00	610.00	468.00	87.00	105.00	Mn	843.00	295.00	380.00	361.00	295.00	291.00	544.00
Mo	0.31	0.23	0.35	1.95	0.17	0.54	0.31	Mo	0.03	0.05	0.03	0.02	0.03	0.11	0.03
Nb	12.50	12.20	0.05	0.40	0.02	0.03	0.02	Nb	0.01	0.02	0.01	0.01	0.00	0.08	0.01
Ni	27.20	29.00	9.00	48.00	25.00	5.00	14.00	Ni	7.52	15.30	11.15	19.15	13.15	13.35	13.00
P	140.00	160.00	80.00	530.00	270.00	70.00	230.00	P	5480.00	5950.00	5320.00	5570.00	5600.00	5730.00	6170.00
Pb	55.10	33.00	14.00	45.00	29.00	20.00	16.00	Pb	0.12	0.20	0.98	0.53	0.22	0.36	0.20
Rb	98.00	110.00	160.00	190.00	260.00	130.00	40.00	Rb	79.50	70.30	75.60	45.80	62.80	49.80	31.90
Sb	2.37	0.99	2.29	2.42	1.38	3.49	4.22	Sb	0.03	0.07	0.06	0.02	0.01	0.07	0.05
Se	0.20	0.10	0.30	0.60	0.30	0.20	0.30	Se	0.08	0.12	0.16	0.10	0.10	0.05	0.08
Sn	2.40	2.80	0.09	1.01	0.31	0.11	0.15	Sn	0.06	0.09	0.13	0.05	0.05	0.17	0.05
Sr	21.90	18.30	26.00	38.00	19.00	15.00	14.00	Sr	4.99	11.15	14.30	7.62	6.60	3.94	3.18
Th	9.64	7.99	6.33	13.05	19.30	9.20	2.43	Th	0.00	0.01	0.01	0.01	0.00	0.04	0.00
Tl	0.41	0.49	0.05	0.34	0.12	0.06	0.04	Tl	0.02	0.01	0.01	0.01	0.01	0.01	0.01
U	1.70	1.80	0.30	1.85	2.01	0.98	0.24	U	0.00	0.00	0.00	0.00	0.00	0.01	0.00
W	1.40	1.00	0.03	0.30	0.07	0.01	0.03	W	0.01	0.03	0.02	0.00	0.00	0.03	0.01
Y	10.70	9.00	5.31	2.97	3.78	1.18	2.72	Y	0.01	0.01	0.02	0.02	0.01	0.07	0.03
Zn	76.00	79.00	25.00	143.00	90.00	25.00	27.00	Zn	57.40	62.60	48.50	49.10	48.70	67.90	70.90

Note: Major elements (Al, Ca, Fe, K, Mg, Na, and S) are expressed in g/kg, while all other elements are reported in mg/kg. Differences in elemental concentrations between slate and metasandstone bedrocks were tested using an independent-samples t-test. Significant differences were observed for Ca ($p=0.006$), K ($p=0.008$), and Rb ($p=0.017$), while other elements showed no significant differences ($p>0.05$).

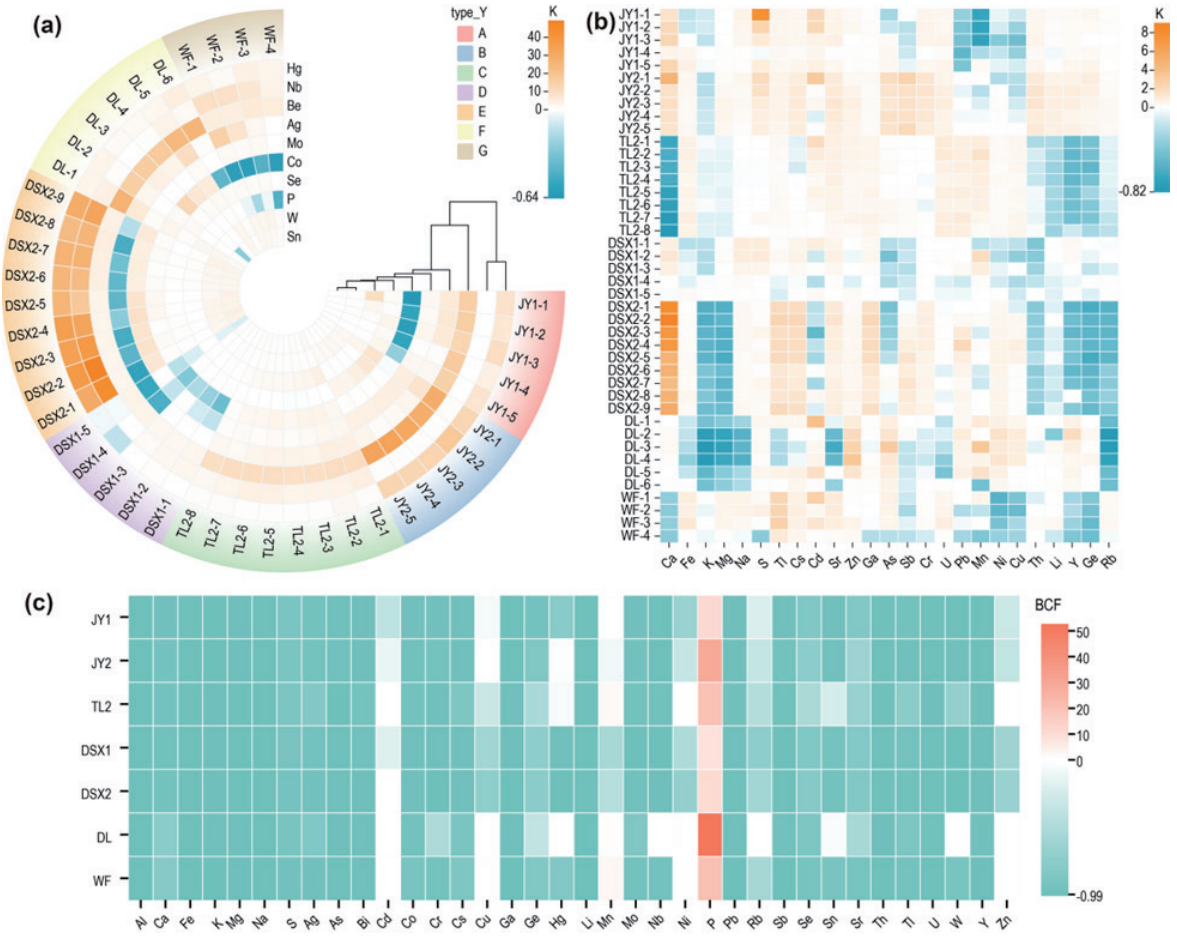


Fig. 2. Geochemical mobilization and soil-tea bioaccumulation patterns of mineral elements. EF_i, enrichment factor of element i; BAF, bioaccumulation factor. EF_i values in soils are shown in a) high-value and b) low-value groups. Color denotes EF_i direction and magnitude. c) BAFs from soil to tea leaves; colors indicate whether BAF>1 or BAF≤1.

To assess plant-mediated elemental redistribution, soil-tea bioconcentration factors (BAFs) were calculated. The BAF values of As, Cr, and Pb were all below 1, indicating negligible bioaccumulation and minimal risk to tea quality. Although P, Mn, Cu, Zn, Cd, and Ni exhibited BAF values exceeding 1 in some samples, P was the only element consistently showing BAF>1 across all sites. Among enriched elements, P displayed the strongest accumulation effect (mean BAF = 22.99), followed by Mn (1.73). By contrast, Cd and Ni showed relatively low enrichment in tea leaves, and Cd concentrations in both soils and tea remained well below established safety thresholds.

Influence of Bedrock-Soil-Tea Geochemical Features on Tea Quality

The concentrations of free amino acids (FAA), caffeine (CA), tea polyphenols (TP), and soluble protein (SP) in tea leaves ranged from 1.86% to 5.58%, 2.07% to 3.13%, 15.46% to 20.22%, and 0.59% to 1.07%, respectively. To investigate the mechanisms linking soil elemental composition to tea leaf biochemical quality, we first performed redundancy analysis (RDA) relating 27 soil mineral elements from seven tea gardens to FAA, CA, TP, and SP (Fig. 3). The first two RDA axes explained 84.2% of the variance in tea quality, demonstrating that the selected soil elements strongly account for observed differences in biochemical traits. Monte Carlo permutation tests (999 iterations) identified seven environmental factors with significant effects ($p<0.05$), ranked by contribution as K (38.5%), Na (21.6%), Cr (11.4%), Fe (8.2%), P (2.9%), Hg (2.3%), and Th (1.6%). Potassium aligned with the positive direction of RDA1 and exhibited the longest vector.

To further disentangle multicollinearity and quantify the contribution of individual elements to specific quality traits, four random forest (RF) regression models were constructed with FAA, CA, TP, and SP as response variables and 27 soil mineral elements as predictors. All models achieved R^2 values exceeding 0.75, confirming their reliability. Variable importance analyses revealed that soil K was the dominant predictor for FAA (Fig. 4a)) and CA (Fig. 4b)). For TP, soil Pb emerged as the most influential predictor (Fig. 4c)), whereas Cr was the key contributor for SP (Fig. 4d)). Collectively, these

Table 3. Mean elemental composition of different soil profiles in the study area.

Sample	JY1	JY2	TL	DSX1	DSX2	DL	WF
Ag	0.04^{a}	0.02^{bc}	0.01^{c}	0.03^{ab}	0.02^{bc}	0.01^{c}	0.03^{ab}
As	12.94^{b}	13.76^{b}	14.45^{b}	37.12^{b}	39.12^{a}	19.90^{b}	20.69^{b}
Bi	0.41^{bc}	0.44^{bc}	0.24^{c}	0.57^{b}	0.94^{a}	0.26^{c}	0.30^{c}
Cd	0.08^{a}	0.04^{b}	0.07^{a}	0.08^{a}	0.04^{b}	0.04^{b}	0.03^{b}
Co	8.16^{cd}	6.78^{cd}	3.25^{d}	19.20^{b}	25.00^{a}	9.50^{c}	3.25^{d}
Cr	40.40^{e}	48.20^{d}	56.50^{c}	82.60^{b}	89.67^{a}	20.17^{f}	36.25^{e}
Cs	9.42^{a}	9.64^{a}	1.22^{e}	4.81^{c}	5.72^{b}	0.96^{e}	3.23^{d}
Cu	18.78^{c}	15.14^{cd}	19.38^{c}	34.60^{b}	59.56^{a}	11.00^{cd}	8.75^{d}
Ga	22.64^{a}	23.08^{a}	2.43^{e}	7.99^{c}	11.07^{b}	4.37^{d}	5.51^{d}
Ge	0.17^{a}	0.17^{a}	0.05^{bc}	0.06^{bc}	0.06^{b}	0.04^{c}	0.06^{bc}
Hg	0.15^{c}	0.14^{c}	0.07^{d}	0.21^{b}	0.31^{a}	0.07^{d}	0.17^{bc}
Li	41.10^{c}	44.18^{c}	50.00^{b}	50.00^{b}	61.11^{a}	18.33^{d}	50.00^{b}
Mn	491.60^{b}	323.40^{bc}	122.38^{c}	801.80^{a}	534.56^{b}	148.33^{c}	160.00^{c}
Mo	0.78^{b}	0.95^{b}	0.88^{b}	1.73^{a}	1.41^{a}	0.78^{b}	1.06^{b}
Nb	15.06^{b}	16.46^{a}	0.23^{c}	0.77^{c}	0.59^{c}	0.08^{c}	0.21^{c}
Ni	20.94^{c}	23.08^{c}	11.25^{d}	37.80^{b}	42.89^{a}	8.33^{c}	11.50^{c}
P	448.00^{abc}	196.00^{d}	250.00^{cd}	588.00^{a}	466.67^{ab}	106.67^{d}	282.50^{bcd}
Pb	28.80^{bc}	25.04^{c}	25.63^{c}	40.60^{b}	54.78^{a}	26.67^{c}	21.25^{c}
Rb	100.30^{c}	107.46^{c}	140.00^{b}	164.00^{a}	155.56^{ab}	41.67^{e}	75.00^{d}
Sb	1.91^{d}	2.15^{cd}	2.59^{bc}	1.75^{d}	2.88^{b}	2.80^{b}	5.33^{a}
Se	0.52^{bc}	0.44^{c}	0.35^{c}	0.64^{b}	0.50^{bc}	0.40^{c}	0.98^{a}
Sn	3.12^{b}	3.42^{c}	0.17^{f}	1.09^{d}	1.52^{c}	0.18^{f}	0.59^{e}
Sr	30.30^{b}	29.30^{b}	42.25^{a}	44.00^{a}	40.44^{a}	8.83^{c}	39.00^{a}
Th	12.70^{b}	13.40^{a}	5.60^{f}	8.49^{d}	14.69^{c}	8.03^{f}	3.90^{e}
Tl	0.55^{b}	0.60^{a}	0.06^{f}	0.32^{c}	0.38^{f}	0.06^{c}	0.17^{e}
U	2.20^{a}	2.40^{a}	0.55^{c}	1.65^{b}	2.31^{a}	0.68^{c}	0.55^{c}
W	1.88^{b}	2.10^{a}	0.05^{de}	0.35^{c}	0.30^{c}	0.02^{f}	0.12^{d}
Y	12.76^{a}	12.04^{a}	2.90^{b}	2.92^{b}	2.46^{b}	1.77^{b}	2.89^{b}
Zn	83.40^{b}	97.40^{b}	36.13^{d}	126.20^{a}	141.22^{a}	57.83^{c}	52.50^{c}
Al	84.92^{bc}	88.22^{bc}	89.83^{b}	82.54^{c}	100.33^{a}	84.13^{bc}	96.50^{a}
Ca	0.76^{b}	0.48^{c}	0.46^{c}	1.20^{a}	0.80^{b}	0.10^{d}	0.10^{d}
Fe	34.32^{cd}	37.36^{c}	26.09^{de}	47.04^{b}	56.18^{a}	21.75^{e}	36.55^{c}
K	20.68^{bc}	20.10^{bc}	31.31^{a}	21.82^{bc}	23.03^{b}	10.98^{d}	17.08^{c}
Mg	3.96^{c}	4.52^{b}	4.40^{bc}	5.08^{a}	4.03^{bc}	1.05^{e}	2.03^{d}
Na	0.86^{b}	0.90^{b}	0.40^{cd}	1.08^{a}	0.89^{b}	0.30^{d}	0.43^{c}

Note: Major elements (Al, Ca, Fe, K, Mg, Na, and S) are expressed in g/kg, while all other elements are reported in mg/kg. The lowercase letters indicate significant differences ($p<0.05$).

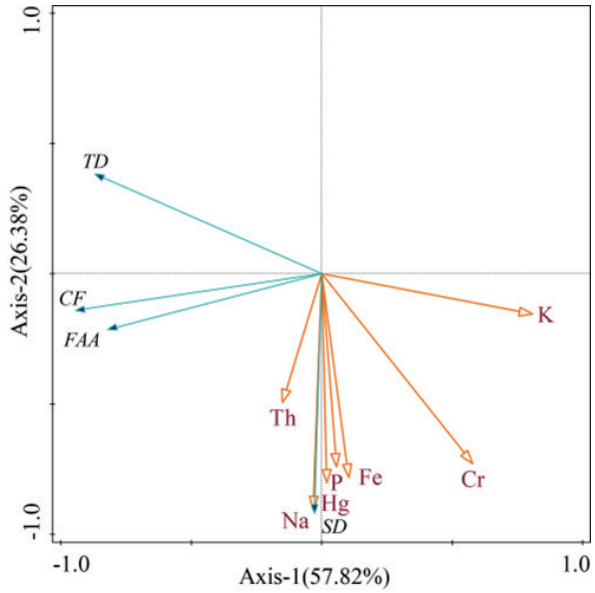


Fig. 3. Redundancy analysis (RDA) ordination of soil mineral elements and tea quality parameters. FAA, free amino acids; CA, caffeine; TP, tea polyphenols; SP, soluble protein.

analyses indicate that soil K serves as the core driving factor for tea leaf biochemical quality, particularly influencing FAA and CA, while elements such as Pb and Cr are associated with TP and SP but with lower predictive strength.

Structural equation modeling (SEM) further quantified the pathway linking bedrock composition, soil elemental pools, and tea elemental uptake (Fig. 5a)). Bedrock composition exerted a significant direct effect on the soil multi-element pool (path coefficient = 0.92, $p<0.001$), which in turn significantly influenced tea elemental uptake (path coefficient = 0.64, $p<0.001$). The direct path from bedrock to tea was not significant. Mantel tests further revealed significant correlations between specific tea leaf elements and quality parameters (Fig. 5b)). FAA was positively associated with Se and Sr ($p<0.05$), CA with Cd, Pb, Se, and Sr ($p<0.05$), TP with Br, Cr, Cu, Ga, Li, Nb, Se, and Zn ($p<0.05$), and SP with Fe, Sn, and U ($p<0.01$), as well as with Ge, Mo, Sb, Th, W, and Y ($p<0.05$).

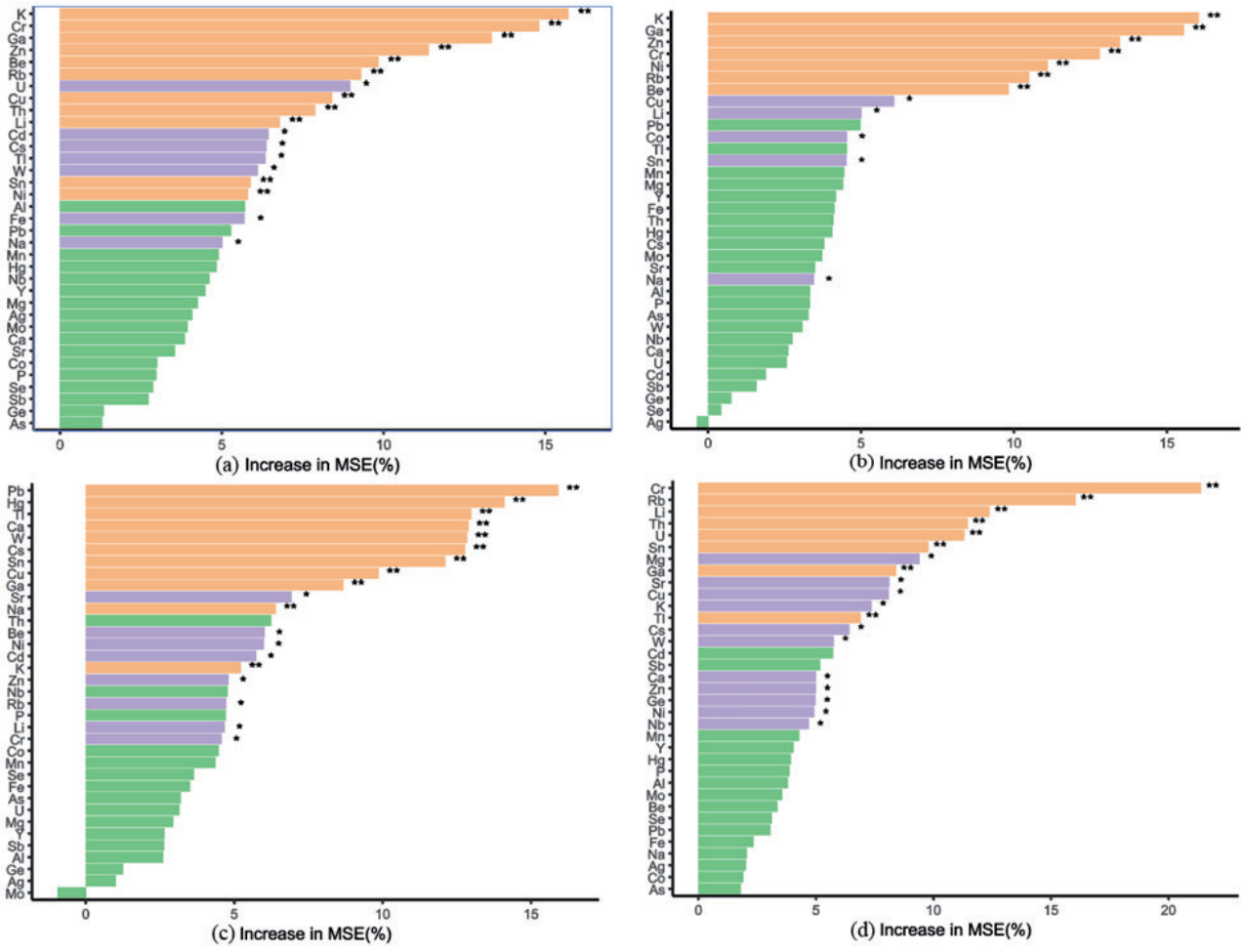


Fig. 4. Random forest (RF) evaluation of soil mineral elements influencing tea quality. The increase in node purity (IncNodePurity) indicates variable importance for predicting a) free amino acids (FAA), b) caffeine (CA), c) tea polyphenols (TP), and d) soluble protein (SP).

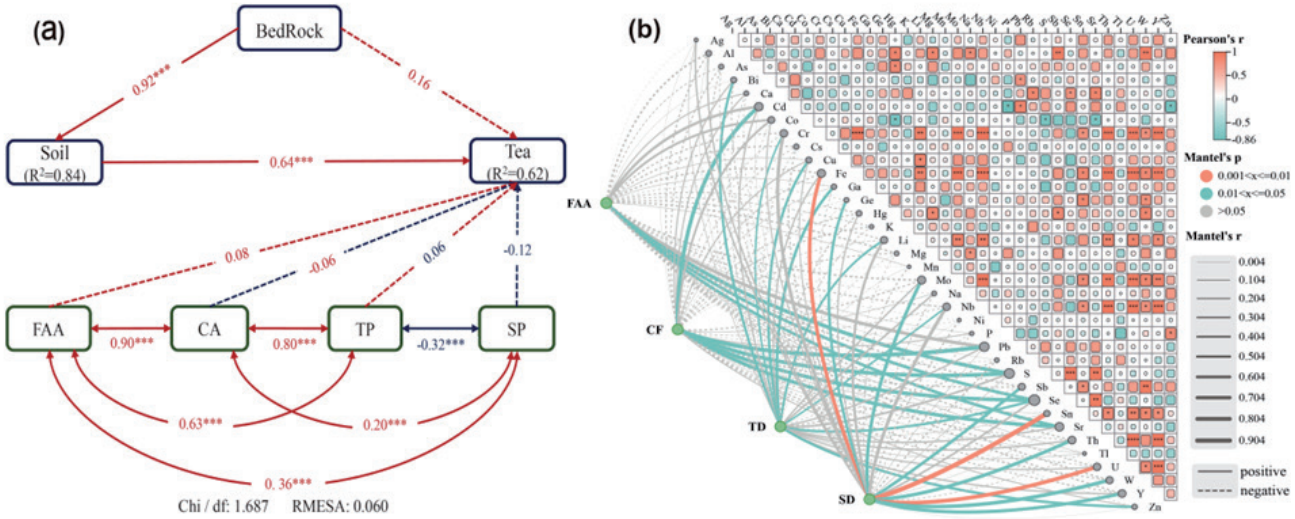


Fig. 5. Direct and indirect effects of mineral elements on tea quality. a) Structural equation model (SEM) of rock-soil-tea pathways. Solid and dashed lines indicate significant and non-significant paths, respectively; *** p≤0.001. R^2 shows explained variance; $\chi^2/df = 1.687$, RMSEA = 0.060. b) Mantel test between tea mineral elements and quality parameters. The heatmap shows Pearson's correlation coefficient (r), and edge width indicates Mantel's r.

Discussion

Geochemical Characteristics of Elements in the Bedrock-Soil-Tea System

The elemental differentiation between metamorphic slate and metasandstone reflects inheritance from their protolith characteristics. Slate retains clay mineral assemblages derived from argillaceous parent rocks [34], and hydrous minerals such as mica and chlorite remain stable during metamorphism [35], effectively preserving major elements as well as lithophile and chalcophile elements [36, 37]. In contrast, the protolith of metasandstone is characterized by higher permeability and is more susceptible to metasomatic alteration under metamorphic fluid activity, leading to relative enrichment of fluid-mobile elements such as As, Hg, Sb, and U [20, 38]. The significant difference in CaO content between DSX1 and DSX2 may be related to variations in parent rock sections of the Pt_3j formation. This intrinsic geochemical heterogeneity of bedrock can be transmitted upward through weathering profiles and subsequently influence soil nutrient status [39].

The vertical distribution patterns of soil elements reflected both pedogenic redistribution and biological cycling [40-42]. Surface enrichment of Cd, Pb, Hg, P, and Se was likely related to atmospheric deposition, litter return, and organic matter complexation [41, 43-47], whereas lower concentrations of K, Ca, and Mg in surface and middle horizons indicated depletion of mobile base cations under humid subtropical weathering [48]. These patterns suggest that weathering leaching and tea plant uptake jointly regulate the vertical redistribution of mineral elements [33, 49].

Mineral nutrient composition is an important indicator of tea quality and economic value [50]. Although Al_2O_3 is abundant in soils, K exhibited the highest mean concentration in tea leaves, suggesting that nutrient bioavailability rather than total soil content controls plant uptake [8]. The relatively high Se concentration in TL may reflect site-specific soil geochemical conditions and plant uptake [51]. The coupling among lithological background, pedogenic redistribution, and biological uptake highlights the integrated nature of the rock-soil-tea system.

Element Migration and Enrichment in the Bedrock-Soil-Tea System

The mass-balance approach revealed distinct enrichment and depletion patterns across the soil profiles, reflecting the interactive effects of parent material weathering, leaching intensity, and biological cycling [52]. Among all elements, potassium (K) exhibited the most consistent and pronounced net depletion ($EF_i<0$) throughout the profiles, a pattern consistent with increasing evidence of widespread K deficiency in tea plantations worldwide [1, 53]. Multiple factors likely contribute to this depletion. Unlike carbonate regions, where enrichment layers commonly develop at the base of weathering profiles, soils derived from metamorphic sandstone and slate are characterized by sandy textures and high permeability, facilitating rapid vertical migration of soluble K [54]. Combined with high annual precipitation (approximately 1200-1600 mm), these properties enhance continuous leaching losses. Furthermore, tea plants are potassium-demanding crops, and repeated harvesting of young shoots removes substantial quantities of K from the system [55]. The combined effects of intensive plant uptake and persistent leaching therefore result

in sustained K depletion, which, if unmitigated, may progressively exhaust both exchangeable and non-exchangeable K reserves [56, 57].

The enrichment of Se, Ag, Be, Hg, and P in surface soils may be related to element-specific weathering behavior, biological recycling, and limited external inputs [29]. In particular, Hg enrichment may partly reflect historical mining activities in eastern and southeastern Guizhou [58-60], whereas P and Se can be redistributed to surface soils through plant uptake and litter return [46, 61].

Bioconcentration factor (BAF) analysis further elucidated plant-soil interactions. Tea plants effectively restricted the uptake and translocation of toxic metals (As, Cr, Hg, and Pb), with BAF values below 1, thereby ensuring product safety [62-65]. Conversely, P and Mn showed preferential accumulation (BAF = 22.99 and 1.73, respectively), along with moderate enrichment of Cu, Zn, and Ni. The preferential accumulation of P and Mn reflects their essential physiological roles in energy metabolism, antioxidant defense, and secondary metabolism [15, 66-68]. Notably, the concentration gradients of P (leaf>soil>bedrock) and Mn (leaf>bedrock>soil) indicate deep root penetration into highly weathered metamorphic strata, enabling nutrient acquisition not only from soil but also directly from weathered bedrock [18, 54, 69]. Although this adaptive strategy may partially alleviate soil nutrient limitations, it is insufficient to compensate for persistent K depletion, highlighting the necessity for proactive potassium fertilization management.

Impact of Element Evolution in the Bedrock-Soil-Tea System on Tea Quality

The combined application of redundancy analysis (RDA) and random forest (RF) modeling enabled a multi-model assessment of soil factors associated with variation in tea biochemical quality in the study area. Both methods consistently identified soil potassium (K) as the most influential predictor of free amino acid (FAA) and caffeine (CA) contents among all measured variables. In the RDA, soil K explained 38.5% of the total variance, exceeding the combined contributions of the next six significant elements (Na, Cr, Fe, P, Hg, and Th). The RF model independently ranked soil K as the most important predictor for FAA and CA, with IncNodePurity values substantially higher than those of other elements. The consistency between linear ordination and non-parametric machine learning reinforces the inference that potassium availability is a primary nutritional factor closely associated with tea quality variation in the metamorphic soils of Guizhou.

From the perspective of plant nutrition, this association is strongly supported by physiological evidence. Potassium acts as a cofactor for multiple enzymes, regulates ATP synthesis and phloem transport, and plays a central role in nitrogen metabolism and amino acid translocation [13, 70]. Adequate K supply promotes the accumulation of theanine and other free amino acids, enhancing the umami taste of green tea while increasing plant tolerance to both abiotic and biotic stresses [1, 33]. However, enrichment factor (EF_i) data indicate that soil K is generally depleted relative to the parent rock in the study area. The alignment between "statistical importance" and "geochemical deficiency" suggests that K may be the key limiting factor for tea quality formation under current soil conditions. These findings support targeted potassium management as a practical strategy to improve tea biochemical quality.

Structural equation modeling (SEM) further elucidated the hierarchical transfer pathway from bedrock composition to tea quality. The path coefficient from bedrock to soil was 0.92, indicating that parent material exerts dominant control over the soil multi-element pool. The significant path from soil to tea (0.64) demonstrates that this geochemical signal can be partially transmitted to the plant, albeit moderated by physiological regulation [71]. In contrast, the direct path from bedrock to tea was non-significant, emphasizing that soil is the critical interface where lithological potential is converted into plant uptake and quality expression. These results indicate that while bedrock composition is fixed, soil nutrient availability, particularly potassium, can still be regulated through environmental and agronomic management. Mantel tests further suggested that several tea leaf trace elements, including Se, Cu, and Zn, were associated with tea quality parameters. These associations indicate that beneficial trace elements may contribute to quality formation [72, 12, 8], although their effects appear secondary to the stronger influence of soil K availability.

Overall, tea quality in the study area reflects the interaction between geochemically inherited bedrock element pools and current soil nutrient availability, with potassium identified as the primary constraint. Addressing potassium limitation while maintaining beneficial trace element backgrounds such as Se, Cu, and Zn offers a practical strategy for enhancing both the biochemical composition and sensory quality of tea. However, detailed historical records of fertilization and agrochemical management were not fully available for all tea plantations before sampling. Therefore, the potential influence of long-term fertilization and plantation management on soil nutrient dynamics cannot be completely excluded. Future studies should integrate long-term management records with geochemical analyses to better distinguish lithological controls from agricultural inputs.

Conclusions

This study systematically investigated the migration and enrichment patterns of mineral elements in the bedrock-soil-tea system of metamorphic rock regions in Guizhou Province and identified the key factors

controlling tea quality. The main conclusions are as follows:

(1) Bedrock geochemistry constitutes the fundamental source of the soil elemental pool. Distinct elemental signatures inherited from slate and metasandstone parent rocks determine the initial availability of nutrient elements and potential contaminants. Structural equation modeling confirmed that bedrock composition exerts a dominant control over the multi-element soil reservoir (path coefficient = 0.92), establishing the lithological geochemical baseline of the system.

(2) Weathering and leaching processes drive significant vertical differentiation of base cations, resulting in systematic potassium depletion. Enrichment factor analysis revealed persistent K loss across all soil profiles, while surface soils also exhibited relative depletion of Ca and Mg. Under humid subtropical climatic conditions and sustained plant uptake, K displays a negative balance within the soil system, indicating gradual exhaustion of its exchangeable reserves.

(3) Tea plants exhibit selective bioaccumulation behavior that simultaneously enhances nutrient acquisition and ensures product safety. Phosphorus and manganese are preferentially accumulated (BAF>1), whereas toxic heavy metals (As, Pb, Cr, Hg) show minimal transfer to leaves (BAF<1). This selective uptake mechanism reflects physiological regulation by the plant and confirms the environmental safety of tea produced in the study area.

(4) Soil potassium is the primary nutrient factor most closely associated with tea quality variation. Redundancy analysis and random forest modeling consistently identified soil K as the most important predictor of free amino acid and caffeine contents, explaining 38.5% of the total variation in quality traits. The statistical importance of K aligns with its geochemical depletion, indicating that K supply may represent the principal nutritional limiting factor for tea quality formation under metamorphic soil conditions.

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Use of AI Tools Declaration

xChatGPT (OpenAI, GPT-5.5) was used solely to improve the English language, grammar, readability, and overall presentation of the manuscript during the manuscript preparation stage. AI was not used for study design, data collection, data generation, statistical analysis, interpretation of results, or formulation of scientific conclusions.

Conflict of Interest

The authors declare no conflict of interest.

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Supplementary Material

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