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Tea quality estimation based on multi-source information from leaf and soil using machine learning algorithm

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ABSTRACT

Mineral nutrients play a significant role in influencing the quality of tea. In order to detect the quantitative relationships between tea quality and mineral elements from the soil and tea plant, samples of soil and tea leaves from 'Baiyeyihao' and 'Huangjinya' cultivars were collected from 160 tea plantations, and these were used to determine 16 types of soil mineral elements, 16 leaf nutrient elements, and 10 key tea quality compositions. Three predictive models including linear regression, multiple linear regression (MLR) and random forest (RF) were applied to predict the main constituents of tea quality. The usage of mineral elements in the soil and tea leaves improved the estimation accuracy of tea quality compositions, the RF performed best for EGCG (R^2 = 0.67–0.77), amino acid (R^2 = 0.61–0.88), tea polyphenols (R^2 = 0.68–0.77) and caffeine (R^2 = 0.59–0.68), while the MLR performed well for predicting the soluble sugars (R^2 = 0.54–0.84). The multi-source information demonstrated a superior accuracy in predicting the biochemical components of tea when compared to individual mineral elements.

Introduction

Tea (*Camellia sinensis* L.), characterized by its distinct flavor and myriad health benefits, ranks among the most consumed beverages globally (Zaman et al., 2022). Tea boasts an array of secondary metabolites, including tea polyphenol (TP), amino acid (AA), soluble sugars (SS), alkaloids, and terpenes. These components are closely tied to tea quality, contributing to its diverse tastes, flavors, and health benefits (Jiang et al., 2019). Past studies suggest that a higher content of these multifaceted components forms the foundation of superior quality tea (Pang et al., 2022). Among the 26 types of AA identified in tea leaves, theanine stands out as a unique component pivotal to the formation of taste and aroma (Guo et al., 2018). TP, caffeine (CAF), and catechins primarily control the bitterness of tea infusion. With enhanced esterification of catechins, a majority of polyphenols undergo oxidation by oxidative enzymes, forming theaflavins (TFs) and thearubigins (TRs) in black tea (Zhao et al., 2020). In particular, high levels of EGCG and low levels of EC in green tea, as observed in 11 Kenyan clones, serve as indicators of the quality potential for ordinary black tea derived from the tea plant (Owuor and Obanda, 2007). Additionally, the contents of TPs, CAF, epigallocatechin gallate, and epigallocatechin show a significant and positive correlation with the sensory evaluation of tea quality (Ma et al., 2022). TPs are potent antioxidants, capable of preventing and treating diseases by neutralizing free radicals and regulating the activity of different oxidases in the body (Yan et al., 2020). Furthermore, during the formation of green tea, an increase in total SS content can amplify sweetness and mellow taste. The SS, along with high boiling volatile compounds, form glycosides that can enhance the broth's flavor (Deng et al., 2022). Hence, the taste of tea is predominantly determined by chemical components such as AA, TPs, CAF, and SS.

Mineral elements have been shown to influence the growth of tea trees and the quality of tea leaves (Yang et al., 2022). For instance, nitrogen (N) is crucial for the growth and development of tea plants, promoting AA accumulation during N metabolism (Wang et al., 2021). Notably, the contents of free AA such as theanine and glutamic acid are negatively correlated with phosphorus (P) and Potassium (K) nutrient content (Wei et al., 2022). A sufficient concentration of zinc (Zn) could boost the synthesis of polyphenols, free AA, and CAF (Tseng & Lai,

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2022). Previous studies have employed multi-factor analysis in acidic soils, revealing that aluminum (Al) negatively affects the concentrations of magnesium (Mg), P and manganese (Mn), thereby influencing tea quality (Dupré et al., 2019). Conversely, galloylated catechins were positively associated with leaf iron (Fe) concentration (Tolrà et al., 2020). Research by Sentkowskar et al. (2009) delved into the antioxidant interaction between selenium and TPs. The application of micronutrients significantly affected the percentages of P, Zn, and copper (Cu). A combination of N and K micronutrients had a notable impact on CAF content. Although lead (Pb) and mercury (Hg) are not essential for the growth of tea trees, they play a significant role in assessing the quality and safety of tea (Ye et al., 2021).

Earlier research has indicated the feasibility of using simple linear regression (LR) to construct crop quality estimation models. For example, Chen et al. (2015) employed a unitary LR model to establish the relationship between the intensity of laser-induced breakdown spectroscopy characteristic line and the concentration of chromium (Cr), enabling the prediction and analysis of Cr content in potatoes. However, since quality parameter formation is influenced by multiple factors, Huang et al. (2021) utilized a multiple linear regression (MLR) equation to establish the relationship between mineral nutrient elements and fruit quality, and the results showed the MLR can more accurately predict the soluble solids, titratable acid content, and the ratio of soluble solids to titratable acid in fruits compared to unary LR models. Soil properties also significantly influence crop quality. For instance, Liu et al. (2021) demonstrated that soil Zn and Cu were the main factors affecting the content of SS in capsicum through stepwise multivariate regression analysis, while available K primarily determined the content of vitamin C in capsicum. Therefore, when constructing quality estimation models, the mineral elements in the soil must also be taken into account. However, the effectiveness of soil properties in improving quality estimation remains under-studied.

The quality prediction has been studied based on the relationship between mineral element content and fruit quality in several crops such as loquat, persimmon, and pepper, while few researches have been reported for quality prediction mechanism of tea (Li et al., 2019). Our goal is to establish the tea quality prediction mechanism based on the concentration of mineral nutrients in leaves or soil. Specifically, our objectives are: (a) to examine the quantitative relationships between tea quality parameters and mineral elements in tender leaves, mature leaves, and soil; (b) to develop an optimal estimation model for tea quality parameters using different regression algorithms; (c) to investigate the optimum range of mineral elements for achieving the optimal levels of tea quality.

Materials and methods

Experimental sites

This study was conducted in Liyang city of Jiangsu province, China (Fig. S1), 31°09′-31°41′N, 119°08′-119°36′E). The climate of the region is subtropical monsoon, with an annual average temperature of 15.4 ℃ and average precipitation of 1149.7 mm. Given these conditions, the area is well-suited for tea cultivation and spans a cultivation area of 4800 ha (Kaleita et al., 2017). The primary tea cultivars were 'Baiyeyihao' and 'Huangjinya' in this experimental area.

Plant and soil sampling

Each sampling location was accurately recorded using a Trimble GeoXH6000 (Trimble, CA, USA) device. At each point, a bud along with two tender leaves, and 4–6 mature leaves were manually collected from a single tea plant. Additionally, five soil samples were collected from a depth of 0–30 cm over an approximately 100 m^2 area surrounding the sampling point. These soil samples were then gathered into a single representative sample for that particular location. The collected tea samples were cleaned with ultrapure water and then dried using a dehydration machine at 80 °C for 12 h. Post-drying, each sample was ground into powder, sieved through a 0.425 mm diameter mesh, and stored in sealed bags at room temperature for subsequent chemical analyses.

Chemical analysis for plant and soil samples

Determination of the tea quality parameter

TPs were quantified using the ferric tartrate colorimetric method (GB/T8313-2002), SS by the anthrone colorimetric method, and AA by the ninhydrin colorimetric method (GB/T8314-2002). ECG, EC、EGCG、EGC、GOG、GC and CAF content was determined via high-performance liquid chromatography (HPLC) under specified conditions: detection wavelength at 280 nm, flow rate at 0.8 mL per minute, column temperature at 40 °C, injection volume at 2 μL, with a mobile phase gradient elution employing 5 % acetonitrile with 0.26 % phosphoric acid (Phase A), and 80 % methanol solution (Phase B).

Determination of mineral elements from tea leaves and soil samples

Total carbon (C) and total N contents in fresh tea leaves were analyzed using the dry combustion method, where in 15 mg of sieved (100-mesh), dried, and ground tea leaves were directly measured with an automated C/N analyzer (Multi EA 5000, Jean, Germany). Approximately 0.1 g of each soil sample was placed into a Teflon digestion tank, combined with 4.5 mL of HCl and 1.5 mL of HNO_3, and then left to stand for 24 h. Following this, the sample was subjected to microwave digestion (Yang et al., 2022). The resulting acid solution was then transferred to a 200 mL volumetric flask and diluted to the 200 mL mark with ultrapure water, yielding the soil sample solution. Similarly, around 0.2 g of tea leaf samples was placed into a Teflon digestion tank, combined with 3 mL of HNO_3, and left for 12 h, after which it was subjected to microwave digestion. The resulting acid solution was transferred into a 100 mL volumetric flask and diluted to the 100 mL mark with ultra-pure water, yielding the tea leaf sample solution.

From each sample solution, 10 mL was filtered through a 0.22 μm membrane and stored in a refrigerator at 4 °C until testing. Concentrations of select metals, including Zn, Ni, Mn, Cr, Cu (quantified using ICP-OES) and Cd, As, Pb, Hg (quantified using ICP-MS), were determined (Brzezicha-Cirocka, Grembecka, & Szefer, 2016). Standard reference materials were utilized during these analytical procedures to measure the concentrations of Zn, Ni, Mn, Cr, Cu, Cd, As, Pb, Hg, and Se (Table S1).

Data processing and statistical analysis

The relationships between quality parameters and mineral elements derived from tea leaves and soil were examined based on the experimental data from 'Baiyeyihao', 'Huangjinya', and the combination of two cultivars. 70 % of the collected experimental data was allocated for model calibration, with the remaining 30 % set aside for model validation. To construct the quality parameter estimation model, three methods were employed: LR, MLR, and the RF algorithm. The 'lm' and 'randomForest' packages in R software (v. 3.4.4., R Development Core Team, 2018) were utilized during the model construction and validation processes. The predictive capacities of each modeling method were evaluated using the coefficient of determination (R^2), the root mean square error (RMSE; equation 1) and relative error (RE; equation 2), the model with larger R^2 and smaller RMSE and RE was selected.

$$\mathrm{RMSE} = \sqrt{\frac{1}{n} \times \sum_{i=1}^{n} (P_i - O_i)^2} \tag{1}$$

$$\mathrm{RE}(\%) = 100 \times \sqrt{\frac{1}{n} \times \sum_{i=1}^{n} \left(\frac{P_i - O_i}{O_i}\right)^2} \tag{2}$$

Where n represents the number of samples, O_i and P_i represent the observed and predicted values, respectively.

Results

The variability analysis of mineral elements

The descriptive statistics of mineral elements from the tender leaf were evaluated, the results showed a significant variations in the two cultivars (Table S2). In the 'Baiyeyihao' cultivar, GCG displayed the highest coefficient of variation (CV) value at 43 %, while in 'Huangjinya', the mineral element GC showed the most variation with a CV value of 75 %. EGCG, EGC, AA, TP, and SS demonstrated similar CV values between the two cultivars. When considering the results from the 'two cultivars combination', GC (55 %) and GCG (53 %) had the highest CV values, followed by EC (49 %), SS (42 %), ECG (31 %), AA (27 %), EGC (25 %), CAF (24 %), EGCG (19 %), with TP displaying the lowest CV value at 14 %.

Correlation analysis between quality parameters and mineral elements

The correlations between the quality parameters and mineral elements from tender, mature leaves and soil were analyzed for 'Baiyeyihao', 'Huangjinya' cultivars, as well as the 'two cultivars combination' (Fig. 1). For the 'Baiyeyihao' cultivar (Fig. 1a), the Cu demonstrated the highest correlation with both EGCG and EGC, with r values of 0.85 and 0.34 respectively. The correlation between K and AA was the highest (r value of 0.74). Cu showed the highest correlation with TPs (r value of 0.49), and the C element correlated most strongly with both CAF and SS, having r values of 0.49 and 0.48 respectively. In the case of the 'Huangjinya' cultivar (Fig. 1b), Cu remained the most correlated with EGCG (r value of 0.85). K displayed the highest correlation with AA (r value of 0.91), while P was most correlated with TPs (r value of 0.92). Lastly, the C element was found to correlate most strongly with CAF (r value of 0.59) and SS (r value of 0.55). Regarding the mineral elements from the tender leaf of the 'two cultivars combination' (Fig. 1c), Cu displayed the strongest correlation with EGCG (r value of 0.82), while the most significant correlation was observed between N and AA (r value of 0.78). Cu also correlated most with TPs (r value of 0.59), and the C element showed the strongest correlation with both CAF (r value of 0.62) and SS (r value of 0.6).

For the mature leaf of 'Baiyeyihao' (Fig. 1d), Cu demonstrated the strongest correlation with EGCG (r value of 0.47). The strongest correlation was seen between K and AA (r value of 0.56), while P and TPs had the highest correlation (r value of 0.38). The C element exhibited the highest correlation with CAF (r value of 0.42), and Cu had the strongest correlation with SS (r value of 0.33). In the mature leaf of 'Huangjinya' (Fig. 1e), Cu showed the correlation with EGCG (r value of 0.47). The correlation was between N and AA (r value of 0.56). The highest correlation was observed between P and TPs (r value of 0.38), and N and CAF (r value of 0.42). Finally, the C element showed the correlation with SS (r value of 0.33). For the mature leaf of the 'two cultivars combination' (Fig. 2f), Cu had the highest correlation with EGCG (r value of 0.47), and the strongest correlation was observed between K and AA (r value of 0.56). The highest correlation was seen between P and TPs (r value of 0.38), and the C element and CAF (r value of 0.42). Lastly, the C element showed the highest correlation with SS (r value of 0.33).

In the combined soil data of 'Baiyeyihao' (Fig. 1g), P showed the highest correlation with EGCG (r value of 0.22), Cu with TPs (r value of 0.24) and P exhibited the highest correlation with CAF (r value of 0.25). For the combined soil data of 'Huangjinya' (Fig. 1h), the strongest correlation was observed between P and EC (r value of 0.31), and Zn had the highest correlation with EGC (r value of 0.28). The C element showed the strongest correlation with GCG (r value of 0.39), Fe with CAF (r value of 0.25), and Zn with SS (r value of 0.27). For the combined soil data from the 'two cultivars combination' (Fig. 1i), the element Fe

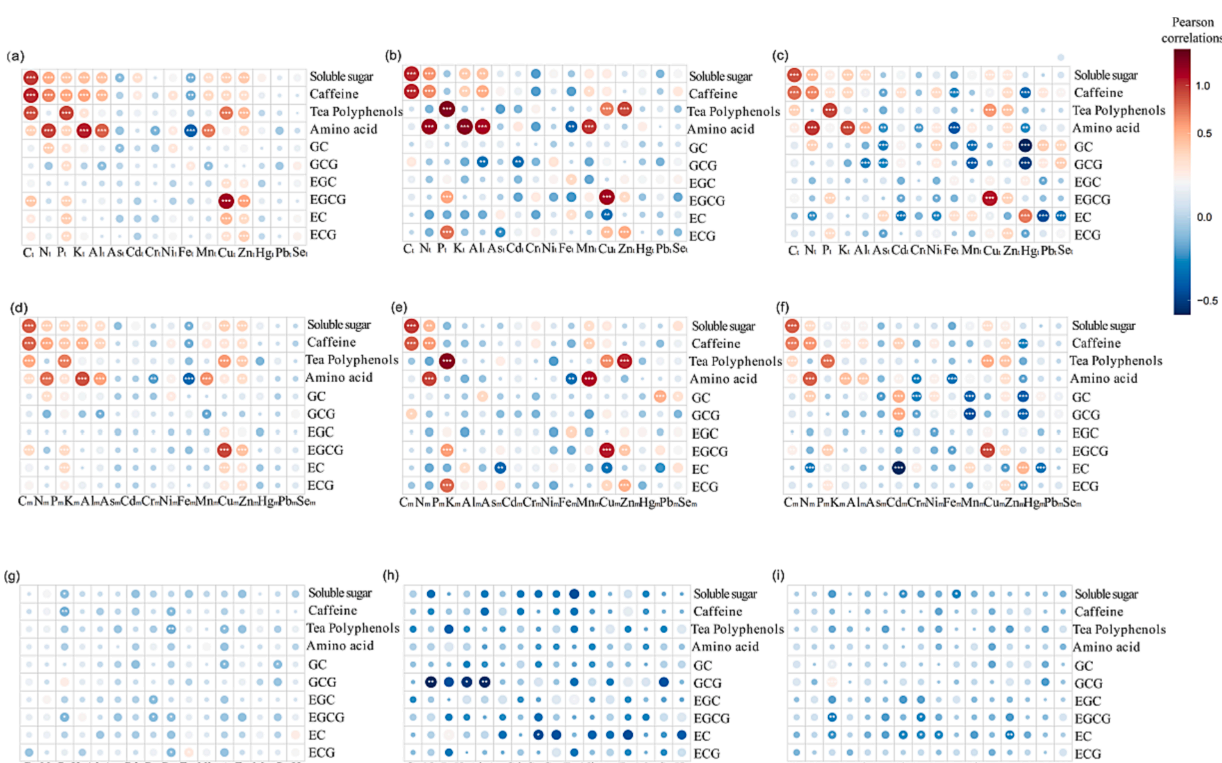

Fig. 1. Correlation of quality parameter and mineral element: from tender leaves of (a) 'Baiyeyihao', (b) 'Huangjinya' and (c) 'two cultivars combination'; from mature leaves of (d) 'Baiyeyihao', (e) 'Huangjinya' and (f) 'two cultivars combination'; from tender, mature leaves and soil combined data of (g) 'Baiyeyihao', (h) 'Huangjinya' and (i) 'two cultivars combination'. The symbols *, ** and **** indicate statistical significance at $p < 0.05$, $p < 0.001$ and $p < 0.0001$, respectively.

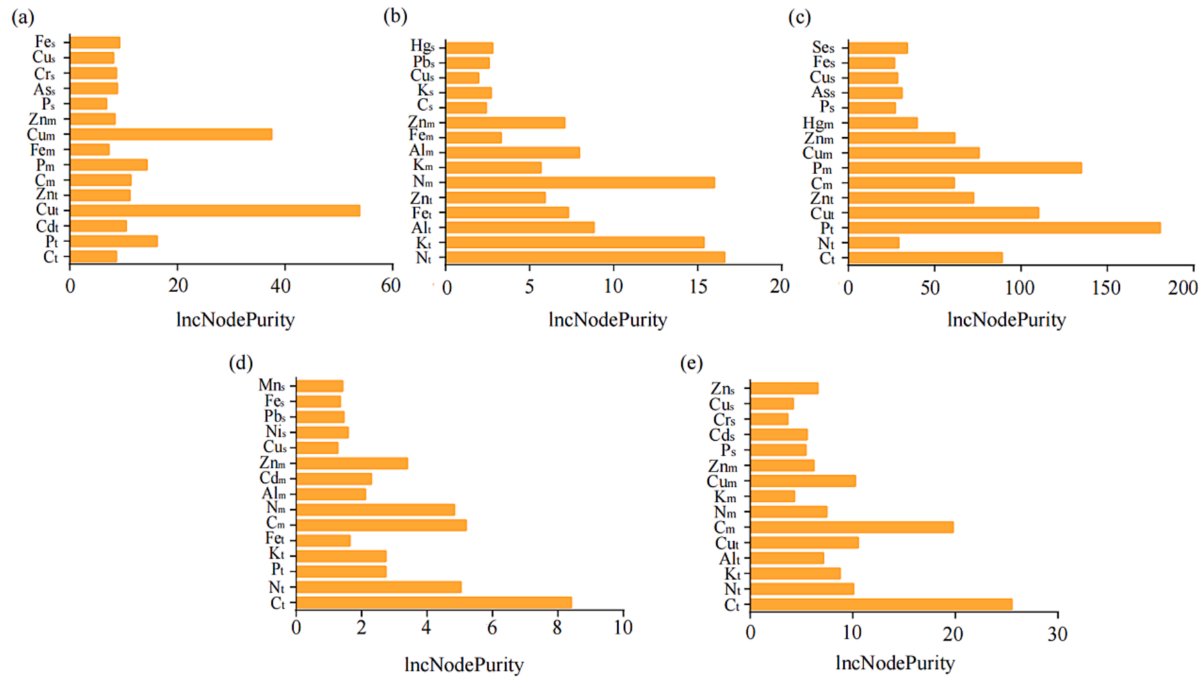

Fig. 2. The Increase in Node Purity (InNoderity) value of each input parameter from the RF model estimation at the (a) EGCG content (b)AA content (c) TPs content (d) CAF content (e) SS content.

exhibited the strongest correlation with EC (r value of 0.21). P demonstrated the highest correlations with EGCG (r value of 0.22), GCG (r value of 0.28), and GC (r value of 0.19). Lastly, Zn was most closely correlated with SS, with an r value of 0.2.

Quality parameter estimation using LR model

LR analyses were conducted between the optimal mineral elements and quality parameters for 'Baiyeyihao', 'Huangjinya' and the combined data (Table 1). The relationships between mineral elements and ECG, EC, EGC, GCG, and GC yielded low R^2 values for both calibration (0.05–0.38) and validation (0.01–0.43) (Table S3). However, mineral elements demonstrated good accuracy for the estimation of EGCG, AA, TP, CAF, and SS. Cu was most strongly correlated with EGCG in both tender and mature leaves, yielding R_c^2 values of 0.18–0.54, 0.57–0.67, and 0.54–0.70 for 'Baiyeyihao', 'Huangjinya', and the combined data, respectively. For the estimation of AA in tender and mature leaves from both cultivars, K (R_c^2 = 0.50–0.66) and N (R_c^2 = 0.59–0.62) performed best. Among all mineral elements, Cu (R_c^2 = 0.44) and P (R_c^2 = 0.32–0.63) yielded the best accuracy for TP estimation in the two cultivars. For CAF estimation in tender and mature leaves, C and N showed high accuracy, with R_c^2 values of 0.48–0.72 and 0.46, respectively. For the estimation of SS, C outperformed all other mineral elements in both cultivars.

Quality parameter estimation using MLR

Five optimal elements from tender and mature leaves were combined to construct the quality parameter estimation models using MLR (Table 2). The results indicated that the MLR model improved the accuracy of ECG, EC, EGC, GCG, and GC estimation compared to the LR model (Table S4). In tender leaves, the AA estimation model achieved the highest accuracy among all quality parameters using MLR for both cultivars, with R_v^2, RMSE, and RE values of 0.69–0.73, 0.16–0.46, and 5.55–13.97 %, respectively. In mature leaves, the TP estimation model exhibited the highest accuracy among all quality parameters using MLR for both cultivars, with R_v^2, RMSE, and RE values of 0.61–0.70, 1.13–1.77, and 5.54–8.91 %, respectively.

Five optimal elements derived from tender and mature leaves, along with soil data, were combined to construct the quality parameter estimation models using MLR method (Table 2). The ECG, EC, EGC, GCG, and GC estimation models demonstrated commendable calibration

Table 1
The calibration and validation results of quality parameter estimation models based on the mineral elements from tender and mature leaf using LR algorithm.

Data source	Quality parameter	Baiyeyihao					Huangjinya					Two cultivars combination				
		Optimal element	R_c^2	R_v^2	RMSE	RE (%)	Optimal element	R_c^2	R_v^2	RMSE	RE (%)	Optimal element	R_c^2	R_v^2	RMSE	RE (%)
Tender leaf	EGCG	Cu	0.54	0.64	0.87	13.16	Cu	0.67	0.66	0.58	8.48	Cu	0.70	0.60	0.91	14.03
	AA	K	0.64	0.63	0.58	17.37	K	0.66	0.53	0.23	8.47	N	0.62	0.55	0.52	17.94
	TPs	Cu	0.44	0.44	2.26	12.21	P	0.63	0.43	0.34	6.10	P	0.68	0.39	0.72	11.23
	CAF	C	0.70	0.37	0.70	24.28	C	0.59	0.34	0.14	6.54	C	0.48	0.32	0.49	18.96
	SS	C	0.62	0.42	0.80	40.18	C	0.61	0.57	0.66	43.49	C	0.54	0.48	0.77	46.51
Mature leaf	EGCG	Cu	0.18	0.52	1.01	15.60	Cu	0.57	0.43	0.65	10.88	Cu	0.54	0.47	0.98	15.41
	AA	K	0.50	0.48	0.59	19.02	N	0.59	0.40	0.40	13.17	K	0.50	0.23	0.81	29.02
	TPs	P	0.32	0.27	1.07	10.56	P	0.56	0.46	1.29	6.15	P	0.32	0.25	2.00	10.33
	CAF	C	0.49	0.34	0.48	19.05	N	0.46	0.31	0.35	14.25	C	0.49	0.32	0.48	20.21
	SS	C	0.52	0.30	0.88	46.81	C	0.53	0.82	0.67	43.33	C	0.52	0.39	0.82	44.16

Note: R_c^2 and R_v^2 represent the calibration and validation R^2, respectively.

Table 2
The calibration and validation results of quality parameter estimation models based on the mineral elements from tender, mature leaf and soil using MLR algorithm.

Data source	Element	Baiyeyihao				Huangjinya				Two cultivars combination			
		R_c^2	R_v^2	RMSE	RE(%)	R_c^2	R_v^2	RMSE	RE(%)	R_c^2	R_v^2	RMSE	RE(%)
Tender leaf	EGCG	0.88	0.65	0.86	12.85	0.83	0.58	0.57	78.14	0.84	0.60	0.91	13.84
	AA	0.89	0.71	0.46	13.97	0.86	0.69	0.16	5.55	0.87	0.73	0.39	11.93
	TPs	0.75	0.62	1.82	10.50	0.77	0.66	1.16	5.39	0.78	0.72	1.51	7.27
	CAF	0.89	0.54	0.80	41.05	0.78	0.51	0.23	10.35	0.83	0.52	0.41	15.79
	SS	0.83	0.50	0.73	36.61	0.76	0.59	0.84	54.45	0.80	0.56	0.74	39.70
Mature leaf	EGCG	0.75	0.51	1.01	14.96	0.72	0.50	0.84	13.37	0.75	0.58	0.94	14.35
	AA	0.85	0.57	0.55	16.91	0.77	0.53	0.61	23.66	0.86	0.50	0.46	15.55
	TPs	0.74	0.61	1.77	8.91	0.69	0.43	0.62	5.54	0.76	0.70	1.13	5.68
	CAF	0.78	0.56	0.38	15.96	0.72	0.45	0.35	14.19	0.81	0.60	0.29	11.81
	SS	0.77	0.48	0.79	43.22	0.72	0.65	0.68	30.28	0.76	0.55	0.78	41.83
T + M + S	EGCG	0.89	0.59	0.93	14.14	0.85	0.69	1.28	19.56	0.86	0.53	0.98	14.86
	AA	0.91	0.74	2.07	23.60	0.88	0.71	0.25	7.97	0.89	0.63	1.39	39.91
	TPs	0.85	0.61	2.36	12.60	0.84	0.76	1.11	5.18	0.85	0.69	1.50	7.22
	CAF	0.91	0.66	2.74	28.96	0.81	0.50	0.35	15.17	0.86	0.61	0.38	16.43
	SS	0.84	0.54	0.77	35.36	0.86	0.84	0.56	36.73	0.84	0.58	0.73	46.78

Note: R_c^2 and R^2v represent the calibration and validation R^2, respectively. T + M + S indicate the combined mineral element data from tender, mature leaf and soil.

accuracy, albeit the validation results were slightly less impressive. For the calibration results of EGCG, AA, TPs, CAF, and SS, the combined model utilizing tender, mature, and soil data showed a higher R_c^2 (0.81–0.91) compared to individual models for tender ($R_c^2 = 0.75$–0.89) and mature leaves ($R_c^2 = 0.69$–0.86).

Quality parameter estimation using RF algorithm

Quality parameter estimation models were also constructed using five optimal elements from tender and mature leaves via the RF algorithm (Table 3). The resulting models for ECG, EC, EGC, GCG, and GC showed slightly reduced validation accuracy, with the exceptions of EC ($R_v^2 = 0.48$–0.60), GCG ($R_v^2 = 0.37$–0.50), and GC ($R_v^2 = 0.53$–0.54) for the combined cultivars (Table S5). The RF algorithm achieved strong accuracy for estimating EGCG, AA, TP, CAF, and SS using data from the tender and mature leaves of both cultivars. In the 'Baiyeyihao' cultivar, the EGCG estimation model ($R_v^2 = 0.72$, RMSE = 0.81, RE = 12.99 %) achieved higher validation accuracy based on tender leaf data compared to other quality parameters, while the amino acid estimation models performed optimally for 'Huangjinya' ($R_v^2 = 0.85$, RMSE = 0.17, RE = 5.84 %) and the combined cultivars ($R_v^2 = 0.78$, RMSE = 0.34, RE = 10.57 %). When mature leaf data was employed, the TP estimation model demonstrated superior accuracy, boasting R_v^2, RMSE, and RE values of 0.61–0.70, 1.22–1.80, and 5.72–9.42 % respectively across the two cultivars.

Utilizing the RF algorithm and a combination of five optimal elements from tender and mature leaves and soil data, quality parameter estimation models were constructed (Table 3). The EGCG, AA, TP, CAF, and SS estimation models all demonstrated robust accuracy. The RF model achieved the highest accuracy in amino acid estimation ($R_v^2 = 0.88$, RMSE = 0.21, RE = 7.13 %) for the 'Baiyeyihao' cultivar, while the TP model delivered the most optimal validation results for both 'Baiyeyihao' ($R_v^2 = 0.68$, RMSE = 1.77, RE = 9.23 %) and the combined cultivars ($R_v^2 = 0.71$, RMSE = 1.53, RE = 7.80 %).

The importance analysis of the governing variables on tea quality

The aforementioned results suggest that the RF models are proficient in predicting the levels of EGCG, AA, TPs, CAF, and SS in tea with high accuracy. As a subsequent step, we sought to identify the input variable in the RF model that exerts the most significant impact on the output variable. In simpler terms, we aimed to discern which mineral elements in tea have the most profound influence on EGCG, AA, TPs, CAF, and SS. Consequently, we carried out an increase in node purity (InNoderity) by incorporating all variables to assess the model's stability.

The results of the importance analysis for the EGCG prediction model are displayed in Fig. 2a. The RF model featuring Cu content in tender tea leaves exhibited the highest importance. The influence of mineral elements in tea on EGCG, in descending order, was: Cu_t, Cu_m, P_t, P_m, C_m, Zn_t, Cd_t, Fe_s, As_s, C_t, Cr_s, Zn_m, Cu_s, Fe_m, and P_s. Fig. 2b presents the importance analysis results for the AA prediction model. The RF model with the highest importance was that with the N content of tender tea

Table 3
The validation results of quality parameter estimation models based on the mineral elements from tender, mature leaf and soil using RF algorithm.

Data source	Element	Baiyeyihao			Huangjinya			Two cultivars combination		
		R_v^2	RMSE	RE(%)	R_v^2	RMSE	RE(%)	R_v^2	RMSE	RE(%)
Tender leaf	EGCG	0.72	0.81	12.99	0.80	0.98	15.87	0.68	0.85	13.83
	AA	0.67	0.48	15.66	0.85	0.17	5.84	0.78	0.34	10.57
	TPs	0.64	1.71	8.68	0.80	1.21	5.4	0.72	1.44	7.40
	CAF	0.52	0.39	15.53	0.55	0.21	9.22	0.65	0.35	13.5
	SS	0.49	0.73	37.29	0.78	0.6	41.51	0.61	0.67	35.27
Mature leaf	EGCG	0.55	1.00	16.05	0.61	0.96	15.96	0.58	0.95	15.23
	AA	0.61	0.49	15.79	0.71	0.29	9.96	0.54	0.49	16.03
	TPs	0.61	1.80	9.42	0.70	1.22	5.72	0.69	1.51	7.76
	CAF	0.62	0.43	18.02	0.51	0.28	12.03	0.55	0.39	15.63
	SS	0.79	0.87	48.79	0.81	0.66	46.01	0.83	0.86	48.03
T + M + S	EGCG	0.69	0.90	14.61	0.77	1.09	17.93	0.67	0.92	14.94
	AA	0.69	0.59	21.46	0.88	0.21	7.13	0.61	0.47	16.37
	TPs	0.68	1.77	9.23	0.77	1.28	5.77	0.71	1.53	7.80
	CAF	0.68	0.45	19.39	0.59	0.24	10.75	0.66	0.36	14.10
	SS	0.48	0.76	37.47	0.77	0.72	51.09	0.55	0.74	40.07

Note: R_v^2 represent the validation R^2. T + M + S indicate the combined mineral element data from tender, mature leaf and soil.

leaves. The mineral elements in tea impacting AA, from highest to lowest influence, were: N_t, N_m, K_t, Al_t, Al_m, Fe_t, Zn_m, Zn_t, K_m, Fe_m, Hg_s, K_s, Pb_s, C_s and Cu_s. The TPs prediction model's importance analysis results are shown in Fig. 2c. The RF model with the P content of tender tea leaves was of the highest importance. The mineral elements in tea influencing TPs, in order of decreasing effect, were: P_t, P_m, Cu_t, C_t, Cu_m, Zn_t, Zn_m, C_m, Hg_m, Se_s, As_s, N_t, Cu_s, P_s, and Fe_s. Fig. 2d illustrates the importance analysis results of the CAF prediction model. The RF model with the highest importance was that with the C content of tender tea leaves. The sequence of mineral elements in tea impacting CAF, from highest to

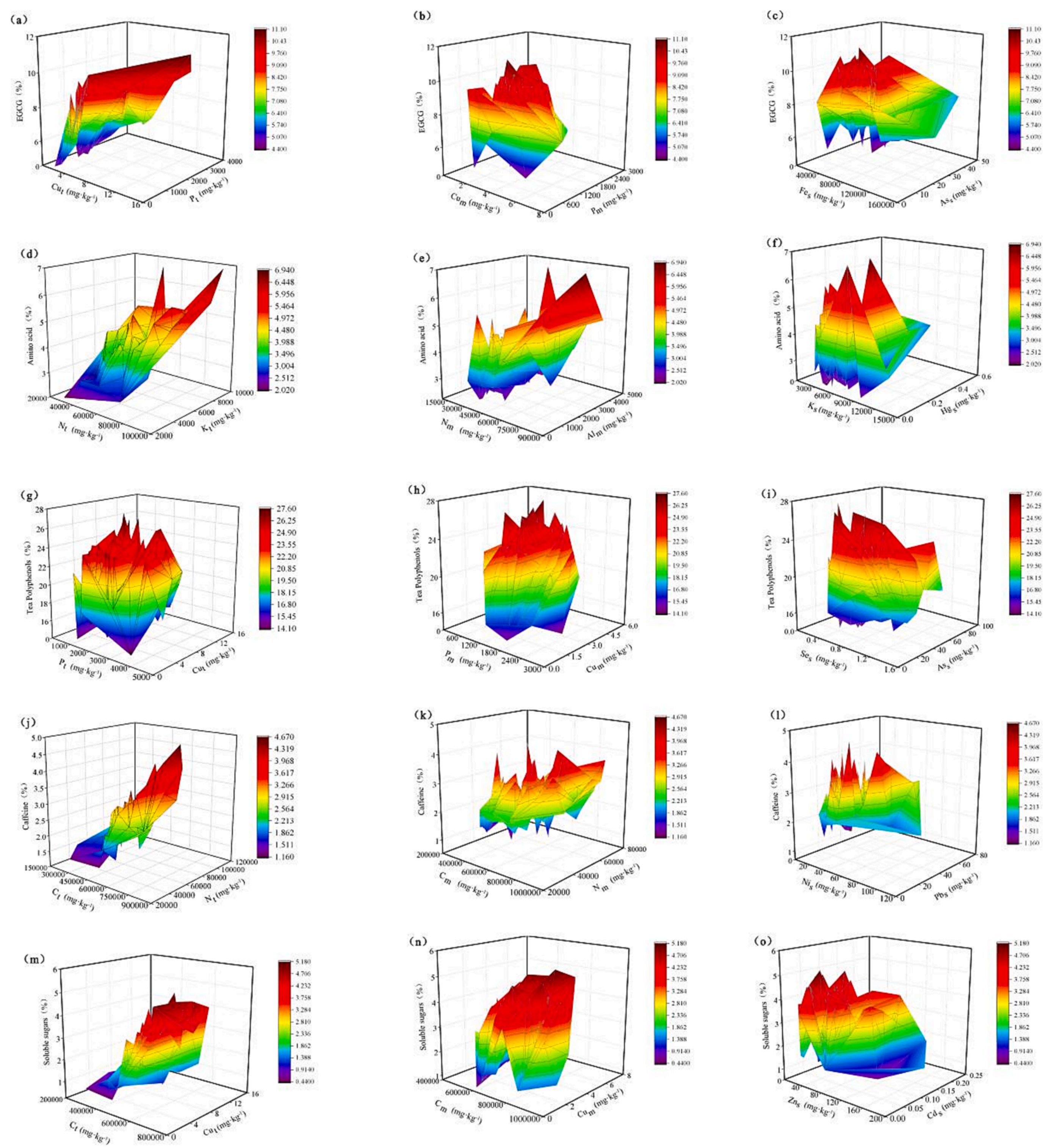

Fig. 3. Response surface plot of the EGCG content, AA content, TPs content, CAF content and SS content under different mineral elements content in tea. (a) Tea Cu_t, P_t content and the EGCG content; (b) Tea Cu_m, P_m content and the EGCG content; (c) Soil As_s, Fe_s content and the EGCG content; (d) Tea N_t, K_t content and Amino acid content; (e) Tea N_m, Al_m content and Amino acid content; (f) Soil K_s, Hg_s content and Amino acid content; (g) Tea P_t, Cu_t content and TPs content; (h) Tea P_m, Cu_m content and TPs content; (i) Soil As_s, Se_s content and TPs content; (j) Tea C_t, N_t content and CAF content; (k) Tea C_m, N_m content and CAF content; (l) Soil Ni_s, Pb_s content and CAF content; (m) Tea C_t, Cu_t content and SS content; (n) Tea C_m, Cu_m content and SS content; (o) Soil Zn_s, Cd_s content and SS content.

lowest, was: C_t, C_m, N_t, N_m, Zn_m, K_t, P_t, Cd_m, Al_m, Fe_t, Ni_s, Pb_s, Mn_s, Fe_s and Cu_s. Lastly, the importance analysis results for the SS prediction model are presented in Fig. 2e. The RF model showcasing the highest importance had the C content of tender tea leaves. The mineral elements in tea influencing SS, from most to least influential, were: C_t, C_m, Cu_t, Cu_m, N_t, K_t, N_m, Al_t, Zn_s, Zn_m, Cd_s, P_s, K_m, Cu_s, and Cr_s.

The analysis of response surface plots

Response surface plots of the highly important parameters were conducted to grasp the influence of these parameters and identify their optimal levels for achieving superior tea quality. The relationships between the contents of C_t, C_m, N_t, N_m, P_t, P_m, K_t, K_s, Cu_t, Cu_m, Al_m, As_s, and other elements in the tea and EGCG, AA, TPs, CAF, and SS were analyzed through these response surface plots (Fig. 3). Fig. 3a-c demonstrated that the impact of P_t, Cu_t, P_m, Cu_m, As_s, and Fe_s content in tea on EGCG. Greater EGCG levels were observed in samples with higher P_t content (2770–3600 mg kg^{-1}), higher Cu_t content (6.46–13.99 mg kg^{-1}), elevated P_m content (1000–2200 mg kg^{-1}), substantial Cu_m content (1.06–4.68 mg kg^{-1}), lower As_s content (17.61–31.68 mg kg^{-1}), and lesser Fe_s content (10800–43700 mg kg^{-1}). The influence of N_t, K_t, N_m, Al_m, K_s, and Hg_s content in tea on AA is displayed in Fig. 3d-f. Greater AA levels were associated with higher N_t content (73800–88900 mg kg^{-1}), increased K_t content (7100–9100 mg kg^{-1}), higher N_m content (29100–78200 mg kg^{-1}), elevated Al_m content (1010–3450 mg kg^{-1}), substantial K_s content (2700–8250 mg kg^{-1}), and lower Hg_s content (0.05–0.23 mg kg^{-1}). However, when the N_t content fell below 60000 mg kg^{-1}, a significant reduction in the tea's AA level was observed. Fig. 3g-i exhibited the effect of P_t, Cu_t, P_m, Cu_m, As_s, and Se_s content in tea on TPs. Lower TPs levels were found in places with lower P_t content (2770–3600 mg kg^{-1}), medium Cu_t content (6.15–7.6 mg kg^{-1}), lower P_m content (650–1800 mg kg^{-1}), medium Cu_m content (1.06–4.68 mg kg^{-1}), high As_s content (17.61–31.68 mg kg^{-1}), and medium Se_s content (0.29–1.01 mg kg^{-1}). When the Cu content exceeded 8 mg kg^{-1}, the TPs level in the tea noticeably increased. Higher levels of CAF were found in samples with increased C_t content (569000–752000 mg kg^{-1}), elevated N_t content (66300–88900 mg kg^{-1}), substantial C_m content (522000–864000 mg kg^{-1}), increased N_m content (32000–78200 mg kg^{-1}), moderate Ni_s content (17.3–43.15 mg kg^{-1}), and diminished Pb_s content (19.96–30.69 mg kg^{-1}). The response surface plot of the impact of C_t, Cu_t, C_m, Cu_m, Zn_s, and Cd_s content in tea on SS is shown in Fig. 3m-o. Higher SS concentrations were observed in instances with elevated C_t content (637000–751000 mg kg^{-1}), increased Cu_t content (6.46–13.99 mg kg^{-1}), substantial C_m content (617000–893000 mg kg^{-1}), increased Cu_m content (3.22–7.12 mg kg^{-1}), low Zn_s content (26.52–53.03 mg kg^{-1}), and medium Cd_s content (0.03–0.14 mg kg^{-1}).

Discussion

Detecting the quantitative relationships between mineral elements and quality indices of tea leaf using different regression methods

A large number of studies have shown that the quality index is affected by mineral elements, so people have tried many times to evaluate the quality of tea by chemical analysis (Jabeen et al., 2019). In this study, by analyzing the correlation between different mineral elements and quality indexes, we selected the mineral elements with the best relationship with quality indexes. For AA prediction, the LR model based on K element in white and 'Huangjinya' buds performed best. Ye et al. (2021) produced the highest free AA increments through potted and field experiments with K nutrient application, and our results are the same. Our results are different from previous studies in which the effect of N element on AA in tea was the most significant. This difference may be due to the improvement of N metabolism by potassium administration, which led to an increase in AA synthesis, and the researchers focused more on the direct effect of N on AA formation. For CAF and SS, the prediction performance of C in tender and mature leaves was the best. This is mainly because the metabolism of CAF is closely related to that of nucleic acid, and the basic skeleton of nucleic acid is element C, so element C is closely related to the composition of CAF (Xia et al., 2017). SS are carbohydrates, which depend on the amount of element C in the tea, similar to the results of this study (Shevchuk et al., 2022).

Tea quality is influenced by a myriad of factors, necessitating the inclusion of a broader range of mineral elements in the quality index prediction model. The MLR model constructed outperformed the LR variant. The prediction accuracy for AA, as measured by the R^2 value of 0.87, was superior when multiple mineral elements were incorporated in the tender leaves of 'Baiyeyihao' compared to a single N element (R^2 = 0.62). This finding echoes the study from Sun et al. (2022), who revealed that incorporating Fe, K, B, and Ca elements heightened the prediction precision for peach quality. In corroborating the assertion from Han et al. (2008) that soil fertility affects tea quality, this study extracted the five soil mineral elements best associated with the quality index. As Table 3 indicates, the model incorporating soil information ($R_v^2 = 0.69$–0.88) outperformed the singular model ($R_v^2 = 0.67$–0.78) in prediction efficiency.

In pursuit of a more universally applicable quality index model, this study amalgamated two tea varieties. Despite the model and verification results being slightly inferior to those from the single-variety model, satisfactory outcomes were obtained for tender leaves ($R_v^2 = 0.49$–0.85), mature leaves ($R_v^2 = 0.51$–0.83), and multi-source data ($R_v^2 = 0.68$–0.88). This study deployed the RF algorithm to construct a tea quality prediction model that incorporated tender leaves, mature leaves, and soil mineral elements. For different quality indices of 'Baiyeyihao' and 'Huangjinya' varieties, the RF model outperformed the LR model. The results were comparable to those from MLR models. However, the RF algorithm performed superiorly in the joint modeling of the two varieties, with its proficiency in handling large datasets standing out. As illustrated by Diaz-Gonzalez et al. (2022), machine learning algorithms have proven highly effective in integrating multi-source information to predict crop quality status. The RF algorithm employed a bootstrap resampling method to generate multiple sample subsets from the original datasets. Each subset was utilized to build an independent decision tree. The amalgamation of predictions from these multiple decision trees formed the final result of the RF model. This unique algorithm renders the RF model more effective and stable when handling massive datasets, as well as dealing with noise and outliers (Wenzl et al., 2021). Several studies have highlighted the consistent performance of the RF algorithm in predicting soil properties, growth, and N status for various crops, including chilli, mango, and cocoa (Islam et al., 2021; Luo et al., 2020).

Relative importance of mineral nutrients to tea quality parameters of tea

Our importance analysis revealed that the Cu and P elements found in tender and mature leaves significantly impact the content of EGCG and TPs. TPs are intricate substances consisting of various compounds, with EGCG, accounting for 9–14 % of green tea dry matter, being the most abundant bioactive component (Tang et al., 2021). Phosphorus plays a pivotal role in tea quality due to its promotion of photosynthesis, thereby increasing sugar content from which polyphenols are derived. A study by Ye et al. (2021) examined the correlation between different forms of phosphorus and tea quality-related components, concluding that phosphorus content can distinguish the quality of tea from different regions, with TPs content being the primary determinant. Furthermore, Ribeiro et al. (2022) established a correlation between Cu elements and polyphenols in green and roasted mate, and Fukushima et al. (2009) discovered that the Cu-reducing ability of green tea was tied to the polyphenols content, a finding consistent with Japanese tea-drinking habits. Our findings corroborate these studies, with our research highlighting a robust correlation between P in mature leaves and EGCG and TPs. This correlation can be attributed to the positive association between the accumulation of Cu and P elements in the tender and mature leaves of tea trees (Han et al. 2020).

The importance analysis also identified N in tender and mature leaves as having the most significant influence on tea's AA content. Several studies have established that the accumulation of AA heavily relies on the N state, and the N element in tea trees determines the formation of AA in the tea's tender leaves (Wang et al., 2021). Additionally, it has been demonstrated that a decrease in total N is linked to a significant reduction in CAF and amino acid concentrations in mature leaves (Li et al., 2016), findings that align with ours.

Our results, from both the linear equation and importance analyses, indicate a close relationship between the formation of theophylline and soluble sugar and the C element in tea. Earlier studies have leveraged the correlation between the C element in tea trees and CAF to classify tea beverages and identify tea tree varieties (Ding et al., 2019). Jean-Pierre et al. (2011) research on the chemical properties of green tea pectin concluded that the C element plays a crucial role in the formation of SS. It has also been shown that the C element can predict SS (Bian et al., 2013). and that the C element in tea roasting forms the foundation for green tea to enhance the sensory quality of SS.

Prior research has demonstrated that N fertilizer can enhance amino acid content in tea plants, potassium fertilizer can stimulate N element absorption and improve tea quality (Xi et al., 2023), and organic fertilizer can boost the C content concentration in soil, enhancing tea quality (Xie et al., 2019). However, no study has delineated the range of elements in high-quality tea. In this study, a range for these elements were determined, whereby an improvement in tea quality was notably observed. Specifically, when the content of C_t was between 637,000–751,000 mg kg^{-1}, C_m between 617,000–752,000 mg kg^{-1}, N_t between 73,800–88,900 mg kg^{-1}, N_m between 59,900–61,000 mg kg^{-1}, P_t between 1,050–1,900 mg kg^{-1}, P_m between 1,200–1,800 mg kg^{-1}, K_t between 7,100–9,100 mg kg^{-1}, and Cu_t between 6.15 and 7.41 mg kg^{-1}, tea quality was significantly enhanced. This research established a theoretical basis for the study of the quantitative relationships between quality parameters and mineral elements from soil and tea leaves. Meanwhile, the research method provided the technical support for predicting the tea quality. In future studies, it is necessary to further explore the effect of multi-source factors on the formation of tea quality, such as soil and meteorological indices, more effective information should be integrated to build the more robust and stable model for tea quality prediction.

Conclusion

In this study, the Hg content was the most crucial independent variable for predicting EC, EGC, GCG, and GC, as indicated by the correlation analysis and LR model. Similarly, the P content emerged as the most vital independent variable for predicting ECG and TPs, the Cu content for predicting EGCG, the N content for predicting AA, and the C content for predicting CAF and SS. According to the RF model, the biochemical composition of tea leaves was most influenced by the contents of C, N, P, and K. A good correlation between tea quality and mineral elements from the soil and tea leaf was found in 'Baiyeyihao' and 'Huangjinya' cultivars. Compared to the LR, the MLR and RF methods performed a more accurate prediction for the tea quality components. The usage of multi-source information improved the estimation accuracy of tea quality compositions, the RF performed best for EGCG, AA, TPs and CAF, while the MLR performed well for predicting the SS. In practical production, the quality of tea leaves can be enhanced by optimizing each main component (AA, TPs, CAF, SS) through the adjustment of independent mineral elements.

Author contributions

Wanping Fang, Xujun Zhu contributed to conception and design of the study, reviewed and approved the final version of all data. Bin Yang and Huan Zhang designed and performed the experiments, analyzed all data conducted and statistical analyses, and wrote the manuscript. Jie Jiang, Salome Njeri Ndombi and Zhaolan Han responsible for sample supply and collection during experiment. Xiaogang Lei, Yao Xiao and Xuejin Chen performed the Chemical analysis experiment.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2023.100975.

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