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Multiple indicators metrological analysis for 5 kinds of tea produced in Yunnan, China

Libing ZHOU[1*], Qin ZHANG[1]

Abstract

Five kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea, from Yunnan Province, China, were selected as the samples for analysis. The combustion heat, differential thermal-thermogravimetric, fat content, crude fiber content, ash content, and trace element content data of the teas we selected were determined, and quality evaluation and classification were carried out by the stoichiometric method from the perspective of food nutrition. In this paper, according to the combustion heat, differential thermal-thermogravimetric, fat content, crude fiber, ash content, and trace element content data of the teas we selected, systematic multi-index comprehensive evaluation systems were constructed through gray pattern recognition, factor analysis, the entropy method and entropy cluster analysis. The multi-index comprehensive evaluation system established in this study provides a new concept for the quantitative control of the quality of tea, a powerful method for research on the large-scale development and classification of tea and basic support for the selection of raw tea materials and the application of a quantitative control mode for assessing the contributions of multi-index ingredients to tea quality.

Keywords: tea; entropy method; thermogravimetric analysis; ICP–OES; trace elements.

Practical Application: The multi-index comprehensive evaluation system established in this study provides a new idea for the quantitative control of the quality of tea, a powerful method for large-scale development and research on classification of tea and basic support for the selection of raw tea materials and the application of a quantitative control mode for evaluating the contributions of multiple ingredients to tea quality.

1 Introduction

Tea (*Camellia sinensis*) is perhaps the most popular and economic beverage globally due to its distinctive fragrance and flavor generated from the leaves of commercially farmed tea plants (Bag et al., 2022). Based on hydrophilic interaction chromatography coupled with triple quadrupole-tandem mass spectrometry, this work developed and validated an efficient (8.5 min per run), sensitive (LOQ: 0.002-0.493 mug/mL) and accurate method. This method was successfully used to determine the contents of 45 hydrophilic constituents in Yunnan large-leaf tea. Umami amino acids and umami-enhanced nucleotides generally exhibited higher contents in green tea and raw pu-erh tea. In contrast, a few amino acids (e.g., proline and gamma-aminobutyric acid) and most alkaloids and nucleosides showed significantly higher contents in black tea or ripe pu-erh tea. By performing orthogonal partial least squares discriminant analysis, classification models for distinguishing four types of tea from each other and for distinguishing green tea from raw pu-erh tea were established (Wang et al., 2022a).

Tea components (tea polyphenols, catechins, free amino acids, and caffeine) are the key factors affecting the quality of green tea (Wen et al., 2021). Aroma is one major aspect of tea flavor and represents an essential criterion for quality evaluation. Herein, we present a novel approach for the rapid, nondestructive and real-time fingerprinting of green tea aromas using ion mobility spectrometry (IMS) without prior sample treatment. Positive photoionization with acetone as a dopant was selected as a suitable technique for tea volatile analysis in comparison to other methods (Li et al., 2019).

Wuyi Rock tea is one of well-known tea in China. The 'Spring Tea Competition' every year is a big event in tea industry of Wuyishan City. In this paper, 667 samples with four grades of Wuyi Rougui teas from 'Spring Tea Competition, 2019' were collected to determine quality indexes. The relationship between the quality index and tea grades was analyzed. The results showed that the higher the grade of teas, the higher the total quality score (TQS) of sensory evaluation (Pang et al., 2022).

Pu-erh tea, native to Yunnan, China, is a highly prized commodity with unique aroma and taste and multiple health effects (Wang et al., 2022b). Public attention in tea quality and nutrition has increased significantly in recent decades, due in part to changes in consumer behavior and the gradually increasing consumption of tea. Demand for high quality tea products obviously requires high standards of quality assurance and process control; satisfying this demand in turn requires appropriate analytical tools for analysis of tea quality and nutrition (Chen et al., 2015).

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[1] *Guangxi Science & Technology Normal University, Laibin, Guangxi, China*
*Corresponding author: zhanyou159@126.com

According to the thermogravimetric parameters, gray pattern recognition was used to determine the combustibility (combustion stability) of 5 teas (Kayisoglu & Coskun, 2021), including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea, from Yunnan Province, China. Research on the combustion heat and thermogravimetric analysis of tea has important theoretical and practical significance. The combustion heat, differential thermal gravimetric values, fat contents, crude fiber contents, trace element contents, and ash contents of 5 kinds of tea were used to construct a systematic multi-index evaluation system by entropy analysis and entropy cluster analysis, which can compensate for the gaps in this area and provide a scientific basis and research significance for the study of tea quality (Zhou et al., 2022a).

Five kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea and pure Banzhang tea, from Yunnan Province, China, were selected as the samples for analysis. The combustion heat (Direktor et al., 2020; Zhang et al., 2022) was measured by an oxygen bomb calorimeter, the combustion stability (Xie et al., 2021; Zhang et al., 2020) of tea was analyzed by thermogravimetric analysis, the fat content was measured by a fat analyzer (Ellers, 1995; Ender et al., 1997; Zhang et al., 2011), and the trace element content was measured by IOP-OES (Jung et al., 2019; Oropeza et al., 2019). According to the combustion heat, thermogravimetric parameters, fat contents, crude fiber contents, trace element contents, and ash contents, a multi-index comprehensive evaluation system (Jia et al., 2017; Chenghua et al., 2016) of five kinds of tea was established, and entropy analysis and entropy cluster analysis were used to evaluate nutritional value. The quality of tea was evaluated by the stoichiometric method from the aspect of food nutrition, which can provide a strong scientific basis for the large-scale development of tea resources and research on tea classification.

2 Materials and methods

2.1 Materials

Five kinds of tea, including camphor-scented palace tea (produced in April 2012, Menghai County, Xishuangbanna Dai Autonomous Prefecture, Yunnan Province, China), fossil glutinous rice tea (produced in September 2010, Menghai County, Xishuangbanna Dai Autonomous Prefecture, Yunnan Province, China), old tree white tea (produced in April 2016, Menghai County, Xishuangbanna Dai Autonomous Prefecture, Yunnan Province, China), Brown Mountain ancient tree tea (produced in April 2014, Mengku Town, Shuangjiang County, Lincang City, Yunnan Province, China) and pure Banzhang tea (produced in March 2016, Menghai County, Xishuangbanna Dai Autonomous Prefecture, Yunnan Province, China), were selected as the samples for analysis. All samples were ground in a mortar and sieved through a 40-mesh sieve. The test was repeated three times for each sample.

2.2 Instruments and methods

Instruments and reagents

Materials used included a BH-series combustion heat experimental measurement device, oxygen cylinder, oxygen meter, grinder, sheet press, ignition wire (nickel-chromium wire, Changsha Changxing Higher Education Instrument Equipment Co., Ltd.), electronic balance (model FA2004, Shanghai Shunyu Hengping Scientific Instrument Co., Ltd.), benzoic acid (AR, Tianjin KERMEL Chemical Reagent Co., Ltd.), medicinal capsules, STA 2500 thermogravimetric analyzer (NETZSCH, Germany), crucible, SE206 fat tester, analytical balance, filter paper, a 100 mL beaker, drying oven, petroleum ether (China Jinan Alva Instrument Co., Ltd.), an F1600 automatic fiber tester, air drying oven, muffle furnace, crucible, filter bag, acid- and alkali-resistant pen, tertiary water, sulfuric acid solution (0.13 mol/L), potassium hydroxide solution (0.23 mol/L), hydrochloric acid solution (0.5 mol/L), petroleum ether (China Jinan Alva Instrument Co., Ltd.), an iCAP7200 ICP–OES (Thermo Scientific, USA), CEM MARS-6 microwave digestion instrument (CEM, USA), Milli-Q ultrapure water preparation system (Millipore, USA), AUY120 millionth electronic analytical balance (Shimadzu Company), nitric acid, hydrogen peroxide (super pure, Guangzhou Chemical Reagent Co., Ltd.) and ultra-pure water prepared in-house.

Methods

The heat of combustion (Libing et al., 2021), thermogravimetric parameters (Junfeng et al., 2021), fat contents (Novokshanova et al., 2019; Pauter et al., 2018), crude fiber contents (Oboh, 2006; Slama et al., 2019), ash contents (Hassid et al., 2022; Krishna et al., 2022; Šimonovičová et al., 2021), and contents of trace elements (Ohata et al., 2009; Vaculovič et al., 2011) for 5 kinds of tea were determined. According to the combustion heat, thermogravimetric parameters, fat contents, crude fiber contents, trace element contents and ash contents, the multi-index entropy method, gray pattern recognition and entropy cluster analysis of 5 kinds of tea were performed in Yunnan, China.

3 Results and discussion

3.1 Calculation of combustion heat values of the samples

1) The first group of experimental samples were of camphor-scented palace tea. The ΔT curve of the Reynolds temperature for camphor-scented palace tea according to the data is shown in Figure 1. The experiment was repeated three times. According to the calculations, $\Delta m_{\text{Camphor-scented palace tea}} = 0.1003$ g, $W_{cal} = 13324.0583$ J/°C, $Q_{capsule} = 18991.5681$ J/g, $m_{capsule} = 0.0971$ g, $\Delta T = 0.321$ °C, $\Delta m_{\text{ignition wire}} = 0.0150$ g, $\Delta m_{\text{Camphor-scented palace tea}} Qv_{\text{Camphor-scented palace tea}} = W_{cal}\Delta T - Q_{\text{ignitionwire}}\Delta m_{\text{ignition wire}} - Q_{capsule}\, m_{capsule}$ and $Qv_{\text{Camphor-scented palace tea}} = 24047.1531$ J/g. The average of $Qv_{\text{Camphor-scented palace tea}}$ was 24769.0499 J/g.

2) In the same way, the combustion heat values for fossil glutinous rice tea, old tree white tea, Brown Mountain

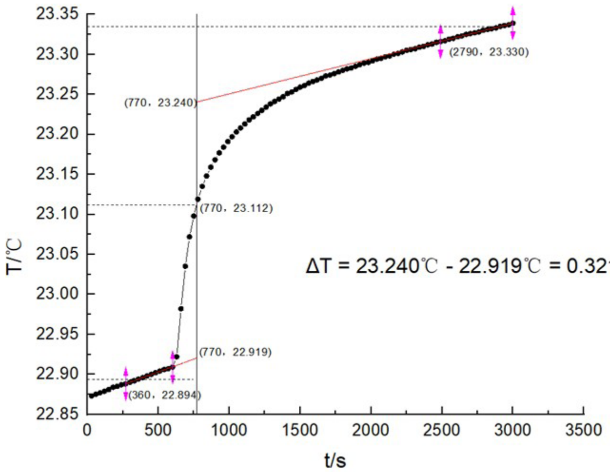

Figure 1. ΔT Curve of Reynolds temperature of Camphor-scented palace tea.

ancient tree tea and pure Banzhang tea were determined, and the tests were repeated three times. The heat of combustion values of the 5 kinds of tea are shown in Table 1. According to Table 1, the 5 kinds of tea were ranked in order of combustion heat magnitude as camphor-scented palace tea > fossil glutinous rice tea > Brown Mountain ancient tree tea > old tree white tea > pure Banzhang tea. The combustion heat of the test samples for 5 types of tea ranged from 21006.8506 to 24769.0499 J/g, CV% < 4.75%. The combustion heat of camphor-scented palace tea was 24769.05 J/g, and the energy was highest for this tea. The combustion heat of pure Banzhang tea was 21006.85 J/g, indicating a relatively low energy value. Combustion heat is regarded as an important physical parameter for measuring the energy content of tea.

3.2 Thermogravimetric analysis

Results of thermogravimetric analysis

1)Thermogravimetric analysis of camphor-scented palace tea

The thermogravimetric data for camphor-scented palace tea are shown in Figure 2 and Table 2. The sample of the camphor-scented palace tea began to decompose at a temperature of 55.6 °C, and after the first stage of decomposition, the loss rate was 4.81%. When the temperature reached 175.3 °C, the sample entered the second stage of decomposition, and at 393.2 °C, the loss rate was 34.03%. The sample continued to decompose, and the final residual mass was 50.27% (Zhou et al., 2022c).

The DTG curve for camphor-scented palace tea displayed two peaks, and the inflection points of the peaks were 91.7 °C and 333.7 °C. The DTA curve for camphor-scented palace tea exhibited a high exothermic peak with a peak value of 106.2 °C, a temperature range of 54.6 °C-181.9 °C, and a peak area of 202.3 J/g.

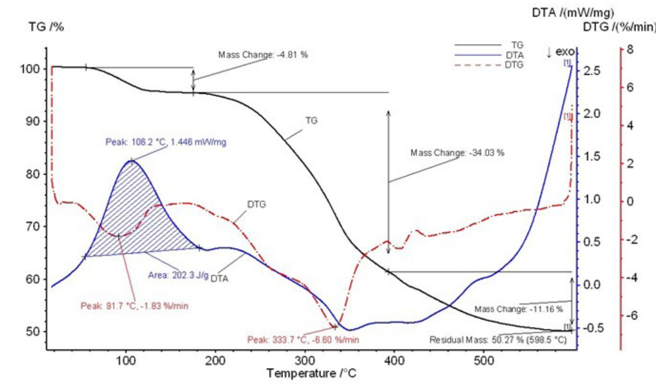

Figure 2. Thermogravimetric (TG %) curve, derivative thermogravimetric (DTG) curve and differential thermal analysis (DT) curve of Camphor-scented palace tea.

Table 1. Combustion heat of 5 kinds of tea (n = 3).

Sample	$Q_{Vaverage}$/(J·g^{-1})	CV/%
Camphor-scented palace tea	24769.0499	3.3732
Fossil glutinous rice tea	24210.9248	4.5321
Old tree white tea	22324.7255	4.7423
Brown Mountain ancient tree tea	24153.8380	3.4710
Pure Banzhang tea	21006.8506	4.5298

Table 2. Thermal analysis data of Camphor-scented palace tea.

Curve	Project	Temperature range/°C	Percentage weight loss/%	Peak area/(J/g)	The fastest weight loss temperature/°C
TG,DTG	Peak1	55.6~175.3	4.81	—	91.7
	Peak2	175.3~393.2	34.03	—	333.7
DTA	Peak1	54.6~181.9	—	202.3	106.2

2)Thermogravimetric analysis of fossil glutinous rice tea

The thermogravimetric data for fossil glutinous rice tea are shown in Figure 3 and Table 3. Figure 3 shows that the fossil glutinous rice tea sample began to decompose at a temperature of 59.9 °C. After the first stage of decomposition, the loss rate was 5.23%; when the temperature reached 178.6 °C, the tea entered the second stage of decomposition. At 413.3 °C, the loss rate was 41.16%; the sample continued to decompose, and the final residual mass was 41.58%.

The DTG curve for fossil glutinous rice tea showed two peaks, and the inflection points of the peaks were 101.5 °C and 333.7 °C. The DTA curve for fossil glutinous rice tea had a high exothermic peak with a peak value of 109.7 °C, a temperature range of 64.1 °C-186.2 °C, and a peak area of 198.2 J/g; there was an endothermic peak with a peak value of 421.7 °C, a temperature range of 315.7 °C to 506.7 °C, and a peak area of 218.2 J/g.

3)Thermogravimetric analysis of old tree white tea

The thermogravimetric data for old tree white tea are shown in Figure 4 and Table 4. Figure 4 shows that the temperature at which the old tree white tea sample began to decompose was 64.1 °C. After the first stage of decomposition, the loss rate was 5.88%. When the temperature reached 174.4 °C, the sample entered the second stage of decomposition. At 395.6 °C, the loss rate was 36.31%. The sample continued to decompose, and the final residual mass was 47.03%.

The DTG curve for old tree white tea showed two peaks, and the inflection points of the peaks were 97.8 °C and 332.5 °C. The DTA curve for old tree white tea exhibited a high exothermic peak with a maximum of 111.6 °C, the temperature range of the peak was 52.5 °C-192.6 °C, and the peak area was 278 J/g.

Table 3. Thermal analysis data of Fossil glutinous rice tea.

Curve	Project	Temperature range/°C	Percentage weight loss/%	Peak area/(J/g)	The fastest weight loss temperature/°C
TG,DTG	Peak1	59.9~178.6	5.23	—	101.5
	Peak2	178.6~413.3	41.16	—	333.7
DTA	Peak1	64.1~186.2	—	198.2	109.7
	Peak2	315.7~506.7	—	218.2	421.7

Table 4. Thermal analysis data of old tree white tea.

Curve	Project	Temperature range/°C	Percentage weight loss/%	Peak area/(J/g)	The fastest weight loss temperature/°C
TG,DTG	Peak1	64.1~174.4	5.88	—	97.8
	Peak2	174.4~395.6	36.31	—	332.5
DTA	Peak1	52.5~192.6	—	278	111.6

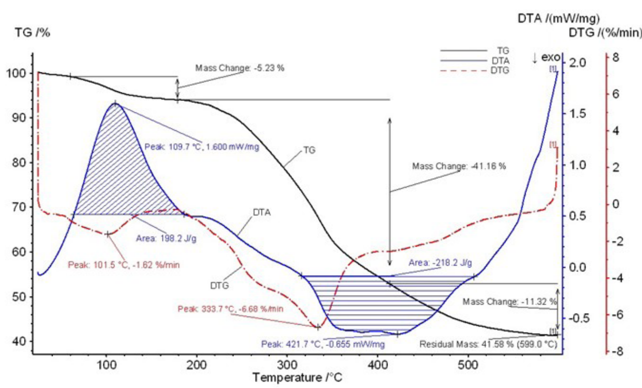

Figure 3. Thermogravimetric (TG %) curve, derivative thermogravimetric (DTG) curve and differential thermal analysis (DTA) curve of Fossil glutinous rice tea.

4)Thermogravimetric analysis of Brown Mountain ancient tree tea

The thermogravimetric data for Brown Mountain ancient tree tea are shown in Figure 5 and Table 5. Figure 5 indicates that the temperature at which the Brown Mountain ancient tree sample began to decompose was 56.6 °C, and after the first stage of decomposition, the loss rate was 7.33%. When the temperature reached 161.9 °C, the tea entered the second stage of decomposition. At 399.2 °C, the loss rate was 59.83%; the sample continued to decompose, and the final residual mass was 15.60%.

The DTG curve for the Brown Mountain ancient tree tea showed two peaks, and the inflection points of the peaks were 98.6 °C and 335.1 °C. The DTA curve for the Brown Mountain ancient tree tea exhibited a high exothermic peak with a peak value of 114.0 °C, a temperature range of 61.4 °C to 175.7 °C, and a peak area of 224.7 J/g; there was an endothermic peak with a peak value of 352.5 °C, a temperature range of 326.2 °C-380.7 °C, and a peak area of 23.68 J/g.

Table 5. Thermal analysis data of Brown Mountain ancient tree tea.

Curve	Project	Temperature range/°C	Percentage weight loss/%	Peak area/(J/g)	The fastest weight loss temperature/°C
TG,DTG	Peak1	56.6~161.9	7.33	—	98.6
	Peak2	161.9~399.2	59.83	—	335.1
DTA	Peak1	61.4~175.7	—	224.7	114.0
	Peak2	326.2~380.7	—	23.68	352.5

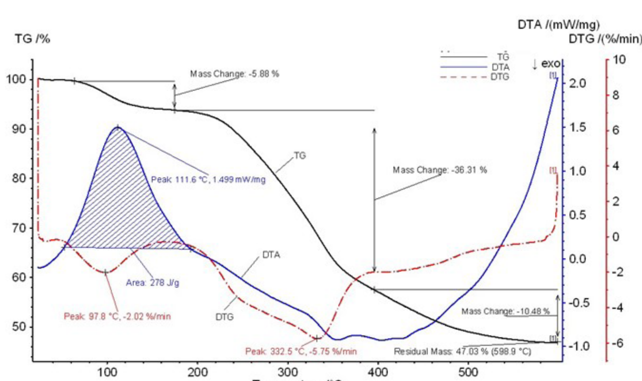

Figure 4. Thermogravimetric (TG %) curve, derivative thermogravimetric (DTG) curve and differential thermal analysis (DTA) curve of old tree white tea.

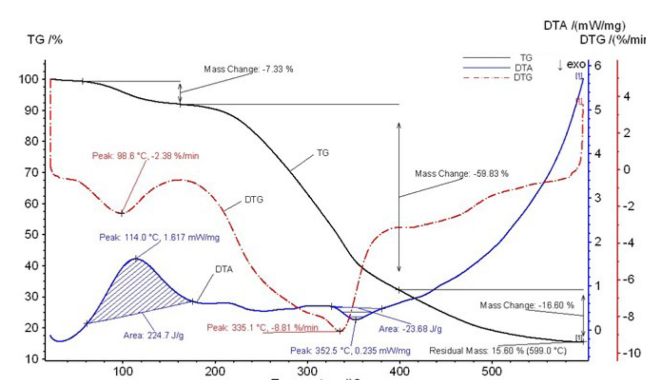

Figure 5. Thermogravimetric (TG %) curve, derivative thermogravimetric (DTG) curve and differential thermal analysis (DTA) curve of Brown Mountain ancient tree tea.

5) Thermogravimetric analysis of pure Banzhang tea

The thermogravimetric data for pure Banzhang tea are shown in Figure 6 and Table 6. Figure 6 shows that the temperature at which the pure Banzhang tea sample began to decompose was 47.3 °C. After the first stage of decomposition, the loss rate was 4.02%; when the temperature reached 175.7 °C, the tea entered the second stage of decomposition. At 402.1 °C, the loss rate was 32.01%; the sample continued to decompose, and the final residual mass was 56.63%.

The DTG curve of pure Banzhang tea displayed two peaks, and the inflection points of the peak shapes were 91.9 °C and 338.7 °C. The DTA curve for pure Banzhang tea showed a high exothermic peak; the maximum was 106.8 °C, the temperature range was 42.0 °C-189.9 °C, and the peak area was 175.1 J/g.

The thermogravimetric data shown in Figures 2-6 indicate that the process of thermal decomposition of the five Yunnan tea samples from room temperature to 599 °C at a constant rate can be divided into three weight loss stages. ① The volatile escape stage: the temperature range is 47.3 °C-178.6 °C, the weight loss rate ranges from 4.02%-7.33%, and the first stage of the DTG curve is an endothermic peak, mainly water, accompanied by volatile aromatic substances and their precursors with the escape of substances that produce more volatile products generating a tea aroma. The sample shows slight weight loss and a small amount of heat absorption, which reflects the slow process of its depolymerization and "glass transition". ② The thermal decomposition stage of deodorization: the temperature corresponds to the range of 178.6 °C-413.3 °C, the weight loss is obvious, and the weight loss rate reaches 32.51%-59.83%, which corresponds to the second stage of the DTG exothermic peak.

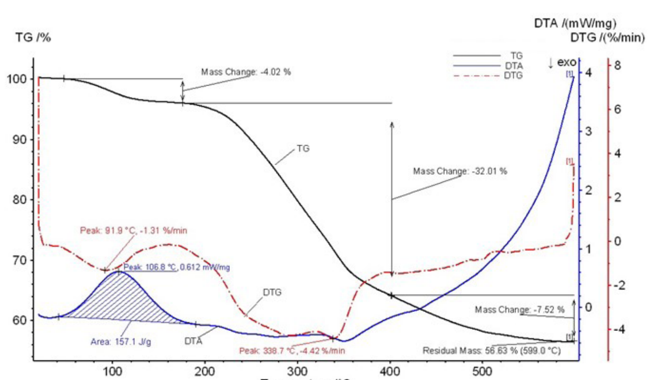

Figure 6. Thermogravimetric (TG %) curve, derivative thermogravimetric (DTG) curve and differential thermal analysis (DTA) curve of Pure Banzhang tea.

Table 6. Thermal analysis data of Pure Banzhang tea.

Curve	Project	Temperature range/°C	Percentage weight loss/%	Peak area/ (J/g)	The fastest weight loss temperature/°C
TG,DTG	Peak1	47.3~175.7	4.02	—	91.9
	Peak2	175.7~402.1	32.01	—	338.7
DTA	Peak1	42.0~189.9	—	175.1	106.8
	Peak2	—	—	—	—

Substances such as sugars, proteins and fats undergo violent thermal decomposition reactions, and the tea flavor has obviously deteriorated, presenting a burnt taste. ③ Carbonization and combustion stage: the temperature range is 413.3 °C-599 °C, the weight loss rate ranges from 7.52%-11.32%, and the thermal weight loss rate is low, because the residue is gradually oxidized and thermally decomposed and the tea is finally burned and ashed. Therefore, to obtain better quality tea, the processing temperature should be maintained under 178.6 °C.

Combustion stability analysis of 5 kinds of tea

Based on the thermogravimetric parameter data of 5 kinds of tea, a multi-index evaluation system for combustion stability was established. Thermogravimetry was applied to study the characteristic combustion index values of tea samples at different heating rates by a thermogravimetric analyzer to judge the combustion stability of tea. The method of gray pattern recognition (Zhou, 2020) was applied to calculate the correlation coefficient between each scheme and the ideal scheme composed of the best indicators, obtain the degree of correlation from each correlation coefficient, and then sort and analyze the results according to the correlation degrees to draw a conclusion. The greater the value of a correlation degree Z is, the better the sample effect is. Finally, all the Z values were compared to evaluate the samples and draw a conclusion. Calculated by Excel, the Z values of 5 kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea, were 0.8153, 0.9039, 0.8394, 0.8677 and 0.8014, respectively. From the analysis of thermogravimetric results and thermogravimetric combustion stability, the 5 kinds of tea were ordered by combustion stability as: fossil glutinous rice tea > Brown Mountain ancient tree tea> old tree white tea> camphor-scented palace tea > pure Banzhang tea.

3.3 Determination of fat, crude fiber, and ash content

Five kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea were examined. The results for determination of fat, crude fiber, and ash contents of the 5 kinds of tea are shown in Table 7. Table 7 shows that the order of fat content (%) of the 5 kinds of tea was fossil glutinous rice tea > Brown Mountain ancient tree tea > old tree white tea > pure Banzhang tea > camphor-scented palace tea. The order of crude fiber content (%) was old tree white tea > pure Banzhang tea > camphor-scented palace tea > Brown Mountain ancient tree tea > fossil glutinous rice tea. The order of ash content (%) was fossil glutinous rice tea > camphor-scented palace tea > old tree white tea > Brown Mountain ancient tree tea > pure Banzhang tea. The energy value of tea can also be reflected by the combustion heat and by fat content to a certain extent. The contents of ash and fat are regarded as important physical data for measuring the quality of tea. Tea quality is evaluated from the aspect of energy, which provides a strong scientific basis for the classification of tea.

3.4 Determination of trace elements

Results of trace element determination (Souza et al., 2022)

A method for the determination of 18 trace elements, including Al, As, Ba, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Na, Ni, Pb, Sc, Sr and Zn, has been established by inductively coupled plasma optical emission spectrometer (ICP–OES) based on microwave digestion. The average value and standard deviation of 18 trace elements in 5 kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea, are shown in Table 8. The element Sc was not detected, and the contents of the heavy metals Pb and Hg and the toxic element As were very low, which is consistent with international regulations (Yao et al., 2021).

Factor analysis of 14 trace elements in 5 kinds of tea

Through factor analysis (Petrov, 2022), the characteristic roots of the factor correlation coefficient matrix and variance contribution rates of trace elements (Liberato et al., 2013; Miele et al., 2015), including Al, Ba, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Sr and Zn, were obtained, as shown in Table 9. According to Table 9, the cumulative contribution rates of the first 3 main factors reached 99.012%, and the eigenvalues of the first 3 main correlation coefficient factors ($\lambda > 1.0$) were larger; that is, the first 3 main factors contributed the most to the explanatory variables. It is most appropriate to extract the first 3 main correlation coefficient factors, which represented 99.012% of the information on 14 trace elements in 5 kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea from Yunnan, China.

The component coefficient matrix after rotation is shown in Table 10. Table 10 shows that the first main factor F_1 of the correlation coefficient mainly contained the original variables Al, Co, Cr, Fe, Li, Mn, Na, Sr, and Zn. The second main factor F_2 of the correlation coefficient mainly contained the information for the original variables Cu, Mg, and Ni. The third main factor F_3 of the correlation coefficient mainly contained the information for the original variable Ba.

The factor scores and the comprehensive factor scores are shown in Table 9. Table 11 indicates that the order of the contents of 14 trace elements in 5 kinds of tea was glutinous rice tea > camphor-scented palace tea > pure Banzhang tea > Brown Mountain ancient tree tea > old tree white tea. In terms of trace element contents, fossil glutinous rice tea contained the highest levels of trace elements in all samples.

Table 7. Determination results of fat, ash content in 5 kinds of tea (n = 3, CV% < 2.0%).

Sample	Crude fiber/%	Fat content/%	Ash/%
Camphor-scented palace tea	9.0708	0.2135	6.4527
Fossil glutinous rice tea	5.7159	1.0544	6.7914
Old tree white tea	32.3535	0.3734	6.1385
Brown Mountain ancient tree tea	6.7044	0.6852	5.6083
Pure Banzhang tea	9.1425	0.2458	5.4570

Table 8. Average values ± standard deviation of 18 trace elements in 5 kinds of tea (μg/g, n = 6, CV% < 5.0%).

Sample	Camphor-scented palace tea	Fossil glutinous rice tea	Old tree white tea	Brown Mountain ancient tree tea	Pure Banzhang tea
Al	327.6664 ± 0.0005	1010.5370 ± 0.0120	18.5864 ± 0.0067	6.9126 ± 0.0061	267.9403 ± 0.0049
As	-	0.1020 ± 0.0002	0.6358 ± 0.0002	0.1791 ± 0.0003	1.2737 ± 0.0018
Ba	7.8382 ± 0.0001	28.1441 ± 0.0003	0.3740 ± 0.0004	-	112.0815 ± 0.0001
Co	0.2147 ± 0.0000	0.2719 ± 0.0000	-	-	0.2911 ± 0.0001
Cr	2.1832 ± 0.0000	11.7947 ± 0.0000	-	-	1.6376 ± 0.0001
Cu	70.4724 ± 0.0005	35.6900 ± 0.0002	3.0666 ± 0.0023	-	31.6230 ± 0.0001
Fe	252.4696 ± 0.0028	623.3855 ± 0.0020	18.2124 ± 0.0086	4.8711 ± 0.0081	190.0655 ± 0.0009
Hg	-	-	0.0374 ± 0.0001	-	-
K	-	-	1777.8609 ± 1.3640	938.7536 ± 1.1920	-
Li	0.1790 ± 0.0000	0.3399 ± 0.0000	-	-	0.1092 ± 0.0000
Mg	1481.7466 ± 0.0230	-	154.2633 ± 0.1006	62.7507 ± 0.0106	1783.1150 ± 0.0380
Mn	734.7888 ± 0.0330	852.4813 ± 0.0160	38.6687 ± 0.0159	14.0401 ± 0.0145	711.7904 ± 0.0180
Na	324.3737 ± 0.0177	337.9674 ± 0.0135	-	-	447.2344 ± 0.0060
Ni	13.6006 ± 0.0003	6.1183 ± 0.0003	0.9723 ± 0.0001	0.3940 ± 0.0003	7.6783 ± 0.0002
Pb	2.1832 ± 0.0003	1.6655 ± 0.0002	0.1122 ± 0.0002	-	0.7642 ± 0.0002
Sc	-	-	-	-	-
Sr	7.5877 ± 0.0001	14.4120 ± 0.0001	0.4488 ± 0.0003	-	6.0408 ± 0.0000
Zn	69.5419 ± 0.0001	63.0184 ± 0.0001	0.3366 ± 0.0001	-	74.6361 ± 0.0011

3.5 Discussion

Construction of multi-index comprehensive evaluation systems for 5 kinds of tea

Construction of a multi-index analysis and comprehensive evaluation system by entropy analysis

According to combustion heat, differential thermogravimetric, fat content, crude fiber content, ash content, and trace element content data for 5 kinds of tea, multi-index comprehensive evaluation systems for 5 kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea, were established by the entropy method (Aide et al., 2021; Álvarez et al., 2020; Arch-Tirado et al., 2020; Bostian et al., 2020; Chen et al., 2020; Cho, 2020).

Table 9. Factor characteristic root and variance contribution rate of 14 trace elements in 5 kinds of tea.

Principal factor	Characteristic root	Contribution rate %	Cumulative contribution rate %
1	9.893	70.664	70.664
2	2.903	20.738	91.401
3	1.066	7.611	99.012
4	0.138	0.988	100.000
…	…	…	…

Table 10. The component coefficient matrix of 14 trace elements in5 kinds of tea.

Element	Component coefficient matrix 1	Component coefficient matrix 2	Component coefficient matrix 3
Al	0.9662	-0.2424	-0.0847
Ba	0.3038	0.2212	0.9258
Co	0.8595	0.3450	0.3763
Cr	0.9122	-0.3892	-0.1274
Cu	0.6454	0.7345	-0.2061
Fe	0.9823	-0.1598	-0.0927
K	-0.8040	-0.4273	-0.2177
Li	0.9903	-0.0293	-0.1356
Mg	0.1345	0.8776	0.4570
Mn	0.9181	0.3584	0.1640
Na	0.7769	0.4548	0.4347
Ni	0.5837	0.8064	-0.0854
Sr	0.9982	-0.0083	-0.0419
Zn	0.8020	0.5223	0.2889

(Deng et al., 2020; Galas et al., 2021; Gupta & Chokshi, 2020; Hu et al., 2020) According to the characteristics of entropy, this paper judges the randomness and degree of disorder of an event by calculating the entropy value and judges the degree of dispersion of an index by using the entropy value. The greater the dispersion degree of the index is, the greater the influence (weight) of the index on the comprehensive evaluation, and the smaller the entropy value. Using the entropy method, 5 kinds of tea were weighted to calculate the comprehensive score S.

The weighted summation formula was used to calculate the evaluation value for each sample. The larger the comprehensive score S is, the better the sample effect is. Finally, all S values were compared; that is, the conclusion regarding evaluation was drawn. The S values calculated in Excel for camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea were 0.3835, 0.6303, 0.2432, 0.2224 and 0.3794, respectively.

According to combustion heat values, combustibility (combustion stability) and fat, crude fiber, ash and trace element contents, entropy analysis indicated that the order of the multi-index comprehensive evaluation of 5 teas was fossil glutinous rice tea > camphor-scented palace tea > pure Banzhang tea > old tree white tea > Brown Mountain ancient tree tea. Therefore, the results of comprehensive evaluation of the quality of tea selected in this study indicated that fossil glutinous rice tea was the best, followed by camphor-scented palace tea.

Construction of a multi-index analysis and comprehensive evaluation system by entropy cluster analysis tree diagram for 5 kinds of tea

Entropy cluster analysis of the 5 kinds of tea was based on the many properties of the samples, and the cluster analysis diagram was obtained from the entropy values. The classification was carried out according to the degree of affinity of the characteristics of the samples. All cases were classified into different classes. Individuals in the same class were more similar to each other, and individuals in different classes were more different (Zhou et al., 2022b). The multi-index comprehensive entropy cluster analysis system of combustion heat, combustibility (combustion stability), fat contents, crude fiber contents, ash contents and trace element contents of 5 teas from Yunnan Province of China was established. A tree diagram of the entropy cluster analysis of the 5 kinds of tea is shown in Figure 7. Figure 7 indicates that 5 kinds of tea, including camphor-scented palace tea, fossil glutinous rice tea, old tree white tea, Brown Mountain ancient tree tea, and pure Banzhang tea, were divided into three categories according to

Table 11. Factor scores and comprehensive factor scores of 14 trace elements in 5 kinds of tea.

Sample	F_1	F_2	F_3	F	Ranking
Camphor-scented palace tea	0.2944	1.4878	-0.9479	0.4488	2
Fossil glutinous rice tea	1.4570	-1.0008	-0.2748	0.8091	1
Old tree white tea	-0.9635	-0.5008	-0.2925	-0.8150	5
Brown Mountain ancient tree tea	-0.9192	-0.5099	-0.1891	-0.7774	4
Pure Banzhang tea	0.1314	0.5237	1.7043	0.3345	3

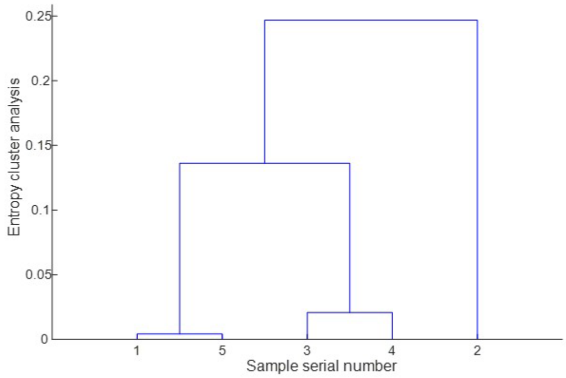

Figure 7. Tree diagram of entropy cluster analysis to 5 kinds of tea.

the results of entropy cluster analysis. Fossil glutinous rice tea constitutes a class, camphor-scented palace tea and pure Banzhang tea form one class, and old tree white tea and Brown Mountain ancient tree tea form a class. Through entropy cluster analysis, we found the degrees of similarity and genetic relationships among the properties of various tea materials, which can facilitate investigation of the classification of tea.

4 Conclusion

According to combustion heat, differential thermogravimetric, fat content, crude fiber content, ash content, and trace element content data for 5 kinds of tea, the results of multi-index analysis constructed through entropy analysis and entropy cluster analysis show that the order of comprehensive evaluation of the 5 teas was fossil glutinous rice tea > camphor-scented palace tea > pure Banzhang tea > old tree white tea > Brown Mountain ancient tree tea. Therefore, the results of comprehensive evaluation of the quality of the teas selected in this study are that fossil glutinous rice tea is the best, followed by camphor-scented palace tea.

In this paper, based on combustion heat, differential thermogravimetric analysis, fat content, crude fiber, ash content, and trace element content data of teas we selected, systematic multi-index comprehensive evaluation systems were constructed through gray pattern recognition, factor analysis, the entropy method and entropy cluster analysis. The multi-index comprehensive evaluation system established in this study provides a new idea for the quantitative control of the quality of tea, a powerful method for large-scale development and research on classification of tea and basic support for the selection of raw tea materials and the application of a quantitative control mode for evaluating the contributions of multiple ingredients to tea quality.

Conflict of interest

The authors declare no conflicts of interest.

Availability of data and material

Data are contained within the article.

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Author contributions

L.B. and Q.Zh. performed the experiments, L.B. was in charge of data processing and writing the paper. All authors reviewed the manuscript.

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