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Evaluation of the quality grade of Congou black tea by the fusion of GC-*E*-Nose, *E*-tongue, and *E*-eye

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ABSTRACT

In the present study, the comprehensive quality of Congou black tea (CBT) including aroma, taste, and liquid color was investigated by a combination of gas chromatography electronic nose (GC-*E*-Nose), electronic tongue (*E*-tongue), and electronic eye (*E*-eye). An excellent discrimination of different quality grades of CBT was accomplished through the fusion of GC-*E*-Nose, *E*-tongue, and *E*-eye combined with orthogonal partial least squares discriminant analysis, with parameters of $R^2Y = 0.803$ and $Q^2 = 0.740$. Moreover, the quantitative evaluation of CBT quality was successfully achieved by partial least squares regression analysis, with the absolute error within 1.39 point, and the relative error within 1.62%. Additionally, 12 key variables were screened out by stepwise multiple linear regression analysis, which significantly contributed to the comprehensive quality score of CBT. Our results suggest that the fusion of multiple intelligent sensory technologies offers great potential and practicability in the quality evaluation of black tea.

1. Introduction

Black tea stands as the second most prevalent tea in China, trailing behind green tea. As a traditional Chinese black tea, Congou black tea (CBT) is renowned for its charming color, mellow taste, and unique aroma (Guo, Long, Meng, Ho, & Zhang, 2018; Peng et al., 2019; Sheikh et al., 2023). The typical processing techniques of CBT generally include withering, rolling, fermentation, and drying. Currently, the quality assessment of CBT predominantly relies on the artificial sensory evaluation, which is challenging to attain satisfactory reproducibility due to substantial individual variations (Hasan et al., 2023; Torrico, Mehta, & Borssato, 2023). In addition, traditional evaluation systems frequently have vague descriptions, making it challenging for consumers to comprehend the quality of tea effortlessly. Therefore, it is urgent to establish an objective and rapid method for evaluating the CBT quality.

Modern instrumental analysis serves as an effective solution for tea quality assessment by offering visual and intuitive data. Intelligent sensory technologies such as electronic nose (*E*-nose), electronic tongue (*E*-tongue), and electronic eye (*E*-eye), which are designed to mimic the human olfactory, gustatory, and visual system, have been developed to obtain comprehensive sensory characteristics of tea products (Cho & Moazzem, 2022; Kubra et al., 2023; Yang et al., 2021). These intelligent sensory technologies generally do not necessitate intricate pre-processing protocols, resulting in remarkable time efficiency and convenience. Unlike the traditional sensor-based electronic nose, gas chromatography electronic nose (GC-*E*-Nose) is developed based on the principle of gas chromatography separation and is capable of acquiring more signal information about the volatile profiles (Xie et al., 2023; Yang et al., 2020; Yang et al., 2022; Yang et al., 2024; Yang, Qian, Deng, Yuan, & Jiang, 2022). For instance, the aroma quality of CBT infusions was successfully assessed by employing GC-*E*-Nose combined with multivariate statistical analysis (Chen et al., 2022). In addition, previous study confirmed the feasibility of *E*-tongue to predict the total contents of free amino acids in black tea (Ouyang et al., 2020). Wei et al. (2022)

Abbreviations: CBT, Congou black tea; GC-*E*-Nose, gas chromatography electronic nose; *E*-tongue, electronic tongue; *E*-eye, electronic eye; CQS, comprehensive quality score; FIDs, flame ionization detectors; VOCs, volatile organic compounds; RT, retention time; RI, retention index; OPLS-DA, orthogonal partial least squares discrimination analysis; R^2Y, cumulative model variation in Y; R^2, cumulative predicted variation; PLSR, partial least squares regression; VIP, variable importance in projection; SMLR, stepwise multiple linear regression; VIF, variance inflation factor..

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proposed a method for tea classification based on *E*-eye and fluorescence imaging. Tea represents a sophisticated system possessing multi-dimensional sensory attributes, making it challenging to comprehensively reflect its quality using a single intelligent sensory. Without complex interventions and professional personnel, multiple intelligent sensory technologies emerge as crucial tools for quality evaluation of tea, which integrate complementary analytics techniques to provide multi-dimensional and reliable information (Awual & Hasan, 2019; Xu, Wang, & Zhu, 2019; Zheng, Shi, Ying, & Men, 2023). Moreover, multiple intelligent sensory technologies coupled with data fusion strategy facilitate the comprehension of disparities in sample traits from diverse perspectives. However, there are few reports on the integration of GC-*E*-Nose, *E*-tongue, and *E*-eye to thoroughly characterize the tea quality.

This study was designed to investigate the feasibility of characterizing different quality grades of CBT by fusion of GC-*E*-Nose, *E*-tongue, and *E*-eye combined with multivariate statistical analysis. In addition, the comprehensive quality of CBT was quantitatively evaluated. The results are expected to provide theoretical support for the quality evaluation of black tea.

2. Materials and methods

2.1. Materials and reagents

Headspace vials (20 mL) capped with 18 mm magnetic PTFE/ silicone were purchased from Agilent Technologies Inc. (Palo Alto, CA, USA). Purified water was obtained from Wahaha Group Co., Ltd. (Hangzhou, China). *n*-Alkane mixtures of C6-C16 (chromatographic purity) were obtained from Restek Co., Ltd. (Centre County, PA, USA). Hydrochloric acid (HCl, analytical purity), sodium chloride (NaCl, analytical purity), and monosodium glutamate (MSG, analytical purity) were obtained from Alpha M.O.S., Co., Ltd. (Toulouse, France).

A total of 56 CBT samples were used in this study, which were purchased from the tea market in 2022. The detailed information is listed in Table S1.

2.2. Sensory evaluation

The evaluation of tea quality was conducted by a well-trained panel according to the Methodology of Sensory Evaluation of Tea (GB/T 23776–2018), in which ethical permission was not required. The panel included three men and three women (aged 28–52) who gave their consent to take part and use their information. Briefly, 3 g of CBT samples were placed into a white cup, and brewed with 150 mL of boiling water for 5 min, then the tea infusion was poured into a white bowl. The aroma was determined by accessing the infused tea leaves left in the cup while the taste and liquid color were determined by accessing the infusion in the bowl. The CBT attributes including aroma, taste and liquid color were evaluated using a 100-point scale, with aroma accounting for 25%, taste accounting for 30%, and liquid color accounting for 10%. The comprehensive quality score (CQS) was attained by weight normalization. During the assessment process, noise levels should not exceed 50 dB, and the room temperature was maintained at approximately 25 °C.

2.3. GC-E-Nose analysis

The volatile profiles were collected using a Heracles II GC-*E*-Nose system (Alpha M.O.S., Toulouse, France), which was equipped with an automatic headspace sampler and two flame ionization detectors (FIDs). The volatile organic compounds (VOCs) were separated by MXT-5 (weak polarity) and MXT-1701 (medium polarity) capillary columns. The method parameters were referenced from our previous study (Wang et al., 2024). Briefly, 0.5 g tea sample and 3 mL water were placed into a 20-mL headspace vial, which was sealed using a silicone cap. Subsequently, it was transferred into an incubator and heated at 60 °C for 20 min. The agitation speed was adjusted at 500 rpm. After incubation, 5000 μL of the headspace gas was inhaled with a sealed syringe, and then injected into an injection system for a duration of 22 s with a flow rate of 300 μL/s. Helium was used as carrier gas with a flow rate of 0.8 mL/min. The VOCs were absorbed by a Tenax trap at 20 °C for 27 s, followed by thermal desorption at 240 °C for 30 s. The adopted temperature procedure was as follows: The initial temperature was maintained at 50 °C for 5 s, ramped up to 80 °C at a rate of 0.1 °C/s, then further increased to 250 °C at a rate of 0.4 °C/s, holding for 10 s. The operational temperature for the two FIDs was set at 260 °C. The signal was collected every 0.01 s, and the total collection time was 740 s. Three replicates were conducted on each sample.

Quality control samples were predominantly obtained from equivalent quantities of each sample, subsequently undergoing comprehensive analysis throughout the entire manipulation procedure to ensure the authenticity and consistency.

2.4. Electronic eye analysis

The tea infusions for *E*-eye analysis were prepared according to the method outlined in Section 2.2, and cooled to room temperature (about 25 °C). Subsequently, the infusions were filtered through a 200 meshes gauze prior to *E*-eye analysis.

The IRIS VA400 *E*-eye system (Alpha M.O.S., Toulouse, France) was employed to capture the images of CBT infusions to obtain the color information. The top and bottom standard light sources (equivalent to D65 light source) were selected as the lighting condition, and a CMOS camera with a 5 mm aperture was utilized to capture the image. Prior to measurement, the following steps were necessary: Preheating for 30 min to obtain a stable light source; Adjusting the lens focal length and exposure; Utilizing a colorimetric card for calibration. Each sample was secured in the same position under a white background, and was captured for three times. The acquired images were preprocessed uniformly, and the feature information of the central circle in the image was extracted. The L*a*b* values, color numbers, and their corresponding area ratios were recorded.

2.5. Electronic tongue analysis

The taste information was collected by the ASTREE *E*-tongue system, equipped with the sixth generation of sensors (Alpha M.O.S., Toulouse, France). The sensor array consists of 7 sensors (AHS, ANS, SCS, CTS, NMS, PKS, and CPS) and a standard reference electrode (Ag/AgCl). Among them, AHS, ANS, SCS, CTS, NMS are sensitive to the sour, sweet, bitter, salty, and umami taste, respectively, while PKS and CPS are accountable for the comprehensive taste. This system converts electrical signals into digital signals based on the sensor's high selectivity for chemicals (ions or neutral molecules) and the potential difference between the sensor and the standard electrode.

Prior to measurement, the sensors were immersed in a beaker for passive activation (1 h), and active activation (0.5 h) in 0.01 mol/L HCl solution followed by a calibration sequence under strict parameters. The sensors were diagnosed using 0.01 mol/L HCl, MSG, and NaCl solution to ensure that they were in a stable working condition. A sample was selected as the standard reference and detected in each round of test to ensure the overall stability. The testing protocol was arranged as follows: cleaning (30 s), testing (120 s), and cleaning (30 s). The average value of the response signal in the period of 110^{th}–120^{th} s was selected as the output value, where the signal was stable and representative. Each sample was repeated three times.

2.6. Statistical analysis

The orthogonal partial least squares discrimination analysis (OPLS-DA) and partial least squares regression (PLSR) of single data set (GC-*E*-Nose, *E*-tongue, and *E*-eye) and fusion data set were performed using

Table 1
The information of volatile components in CBT samples analyzed by GC-*E*-Nose.

No.	Variables	RT (s)	RI	Sources
A1	Acetaldehyde*	55.22	492	MXT-1701
A2	Ethyl alcohol*	63.22	549	MXT-1701
A3	Dimethyl sulfide*	67.06	574	MXT-1701
A4	Trans-1,4-hexadiene	70.39	595	MXT-1701
A5	(4*Z*)-1,4-Hexadiene	74.59	612	MXT-1701
A6	2-Methylfuran	82.70	646	MXT-1701
A7	3-Ethylpentane	87.44	669	MXT-1701
A8	Ethyl acetate*	91.13	680	MXT-1701
A9	Benzene*	94.93	697	MXT-1701
A10	Methyl propanoate	98.51	704	MXT-1701
A11	2-Ethylfuran	104.64	719	MXT-1701
A12	Methyl cyclohexane	107.87	724	MXT-1701
A13	2-Methylbutanal	115.36	740	MXT-1701
A14	Pentanal	131.13	788	MXT-1701
A15	Dimethyl disulfide*	144.96	811	MXT-1701
A16	Ethyl isobutyrate*	151.35	818	MXT-1701
A17	3-Methyl-1-butanol	163.91	844	MXT-1701
A18	3-Hexanone*	175.68	867	MXT-1701
A19	Ethyl 2-methylbutanoate	184.06	887	MXT-1701
A20	Methyl nonyl ketone	318.08	1381	MXT-1701
A21	Acetaldehyde*	47.10	423	MXT-1701
A22	Ethyl alcohol*	49.03	436	MXT-1701
A23	3-Methylbut-1-ene	50.87	450	MXT-5
A24	Pent-1-ene	56.61	487	MXT-5
A25	Dimethyl sulfide*	60.65	514	MXT-5
A26	Cyclopentadiene	65.34	548	MXT-5
A27	Ethyl acetate*	76.39	612	MXT-5
A28	Isovaleraldehyde	88.32	650	MXT-5
A29	(2*E*)-4-methyl-2-Hexene	91.60	661	MXT-5
A30	Benzene*	98.01	679	MXT-5
A31	2-Pentanol	101.90	692	MXT-5
A32	2,3-Pentanedione	105.07	703	MXT-5
A33	3-Penten-2-one	115.11	728	MXT-5
A34	Propionic acid	124.39	741	MXT-5
A35	Pyridine	127.26	747	MXT-5
A36	Dimethyl disulfide*	129.83	752	MXT-5
A37	Ethyl isobutyrate*	135.89	763	MXT-5
A38	3-Hexanone*	144.91	781	MXT-5
A39	Hexanal	153.67	800	MXT-5
A40	Furfural	168.51	825	MXT-5
A41	Citronellal	281.30	1152	MXT-5
A42	Methyl (9*E*)-9-hexadecenoate	409.54	1905	MXT-5
A43	Octadec-9-enoic acid	443.20	2153	MXT-5
A44	Icosanal	457.69	2230	MXT-5

Note: *represented volatile components simultaneously detected on MXT-1701 and MXT-5 columns; No. represented number; RT represented retention time; RI represented retention index.

SIMCA 14.1 software (Umetrics AB, Umeå, Sweden). For quantitative evaluation, the preliminary screening of key variables was accomplished by PLSR, and these variables with variable importance in projection (VIP) > 1 were considered important and advanced to the subsequent step. The final screening of key variables was completed through stepwise multiple linear regression (SMLR) by SPSS 16.0 software (SPSS Inc., Chicago, IL, USA). A strict variable screening criterion was implemented, where the variable admission rule of the final regression equation was set at $p < 0.05$ and the exclusion rule was set at $p > 0.06$.

3. Results and discussion

3.1. Analysis of the results of sensory evaluation

The sensory evaluation of CBT samples was conducted by a panel of six experienced experts. A 100-point grading system was utilized to evaluate the aroma, taste, and liquid color of all CBT samples. The weighted CQS is listed in Table S1. The results showed that the CQS of all CBT samples was ranged from 80.62 to 95.77 points. The classification of quality grades referred to previous study (Chen et al., 2022). With 90 points as the cut-off point, the samples were divided into two categories of grade I and grade II. Specifically, grade I included 15 tea samples with a score of 90 points or higher. In terms of aroma attribute, it was predominantly characterized as floral or sweet aroma. Concerning the taste attribute, the primary performance was sweet, refreshing, and mellow. The attribute of liquid color primarily exhibited orange-red and bright. Grade II was consisted of 41 tea samples with scores below 90. Compared to the samples in grade I, the quality of samples in grade II was diminished to varying extents. In the aroma attribute, some inferior odors such as burnt, grassy, and overcooked odors emerged. The taste presented mixed and unpleasant, and the brightness of the infusion declined somewhat.

3.2. Intelligent sensory characterization analyzed by GC-E-Nose, E-tongue, and E-eye

3.2.1. Volatile profiles analyzed by GC-E-Nose

The GC-*E*-Nose, which is superior to traditional sensor-based *E*-nose, adopts the gas chromatographic separation principle. The analytes are separated by two capillary columns with distinct polarity, facilitating the rapid acquisition of volatile profiles (Hasan et al., 2023; Wang et al., 2023). The radar maps of representative sample are illustrated in Fig. S1. In this study, 44 characteristic peaks were separated and screened out. A total of 36 VOCs were identified by comparing retention index with those in the AroChemBase database, including 8 aldehydes, 6 alkenes, 5 esters, 4 ketones, 3 alcohols, 3 heterocycles, 2 sulfides, 2 acids, 2 alkanes, and 1 aromatic hydrocarbon (Table 1). Notably, 8 VOCs including ethanol, acetaldehyde, dimethyl sulfide, ethyl acetate, benzene, dimethyl disulfide, ethyl isobutyrate, and 3-hexanone were simultaneously detected on the MXT-5 and MXT-1701 columns. In summary, a 44-dimensional characteristic dataset was obtained to characterize the aroma quality of CBTs.

3.2.2. Color information analyzed by E-eye

The *E*-eye system encompasses a built-in autonomous color system, which can obtain the L*a*b* values and corresponding proportions of different colors in recorded images (Table S2). In the L*a*b* color space, L* represents the brightness, ranging from 0 to 100 (from pure black to pure white), a* represents the green-red degree, ranging from −128 to +127 (from green to red) and b* represents the blue-yellow degree, ranging from −128 to +127 (from blue to yellow). The L*a*b* color space is more consistent with human visual perception, and can better reflect the authentic color of the sample. In this study, a total of 40 characteristic colors were extracted from all CBT samples, mainly exhibiting reddish brown, yellowish brown, orange and brown (Fig. S2). In summary, a 43-dimensional characteristic dataset was acquired to characterize the liquid color quality of tea infusion.

3.2.3. Taste fingerprints analyzed by E-tongue

The typical response curve obtained from the *E*-tongue is presented in Fig. S3, which is approximately smooth. During the course of measurement, the signal of each sensor changed continuously in the 0-40th s, began to stabilize around the 40th-110th s, and finally stabilized around the 110th-120th s. Therefore, the average values of the stable response signals in the period of 110th-120th s were selected as the output value (Table S2). A 7-dimensional feature dataset (AHS, ANS, SCS, CTS, NMS, PKS, and CPS) was acquired to characterize the taste quality of tea infusion.

3.3. Discrimination of the quality grade base on OPLS-DA

Accurate identification of the quality grades not only safeguards the interests of consumers, but also contributes to the further development of the quality standard system, which is a potent strategy to augment consumer confidence. In the following study, GC-*E*-Nose, *E*-tongue, and *E*-eye combined with OPLS-DA were used to investigate the quality grades of CBT samples. As a supervised statistical method, OPLS-DA employs partial least squares regression to construct a correlation

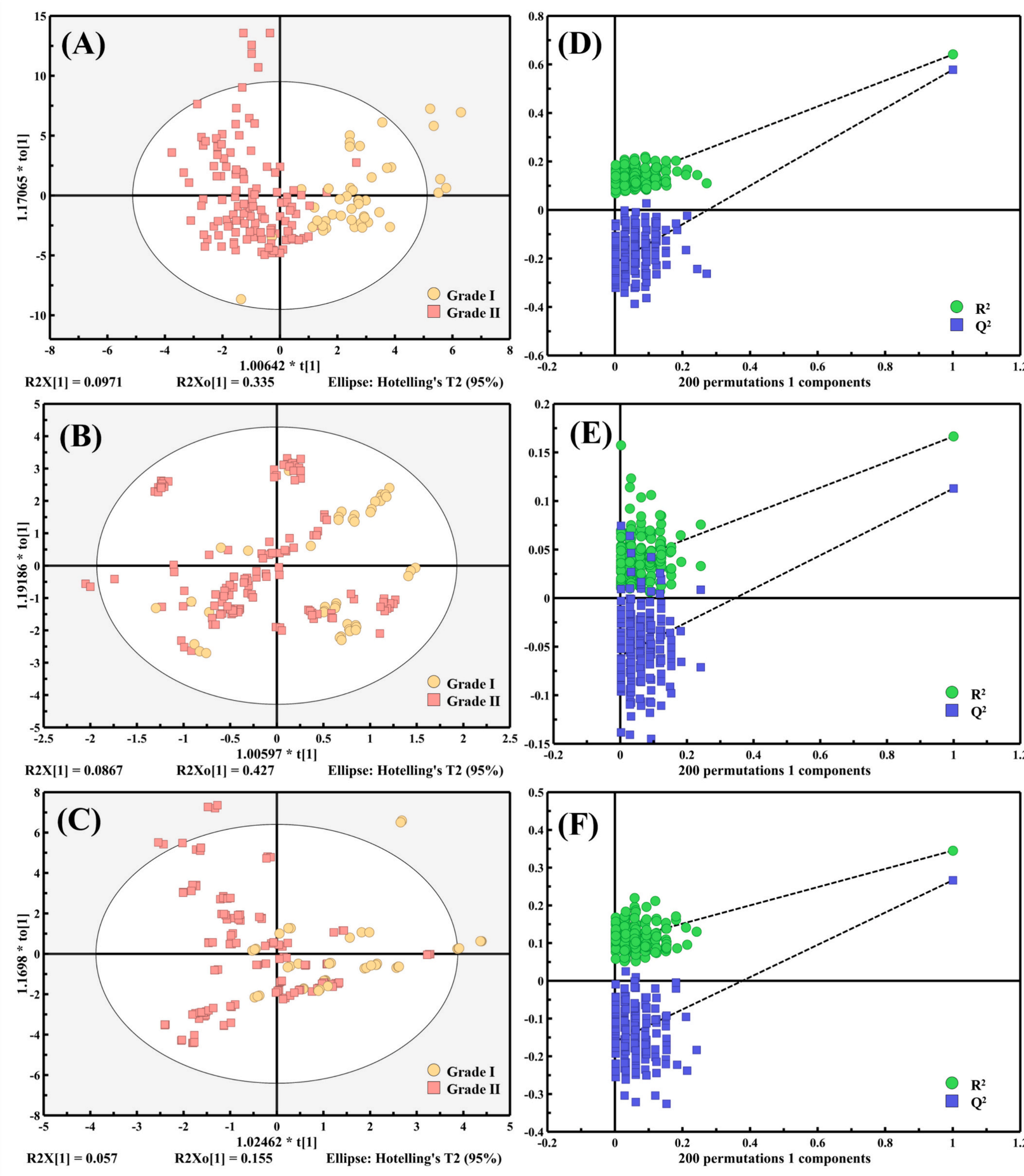

Fig. 1. OPLS-DA analysis based on independent GC-*E*-Nose, *E*-tongue, and *E*-eye. (A) Score plots of GC-*E*-Nose ($R^2Y = 0.641$, $Q^2 = 0.578$); (B) Score plots of *E*-tongue ($R^2Y = 0.167$, $Q^2 = 0.113$); (C) Score plots of *E*-eye ($R^2Y = 0.345$, $Q^2 = 0.267$); (D) Permutation plots of GC-*E*-Nose ($R^2 = 0.105$, $Q^2 = -0.218$); (E) Permutation plots of *E*-tongue ($R^2 = 0.036$, $Q^2 = -0.059$); (F) Permutation plots of *E*-eye ($R^2 = 0.103$, $Q^2 = -0.161$).

model between metabolite expression and sample category, thereby facilitating the judgment and prediction.

In this study, a total of 56 samples (15 samples in grade I and 41 samples in grade II) were used for OPLS-DA analysis. First, the quality grade classification of CBT samples was investigated by independent GC-*E*-Nose, *E*-tongue, and *E*-eye combined with OPLS-DA. As depicted in Fig. 1A, the distributions of two grades of CBT samples exhibited a noteworthy segregation. However, the samples were poorly clustered within groups, indicating that it was a formidable task to effectively distinguish the quality grades based on independent GC-*E*-Nose. For

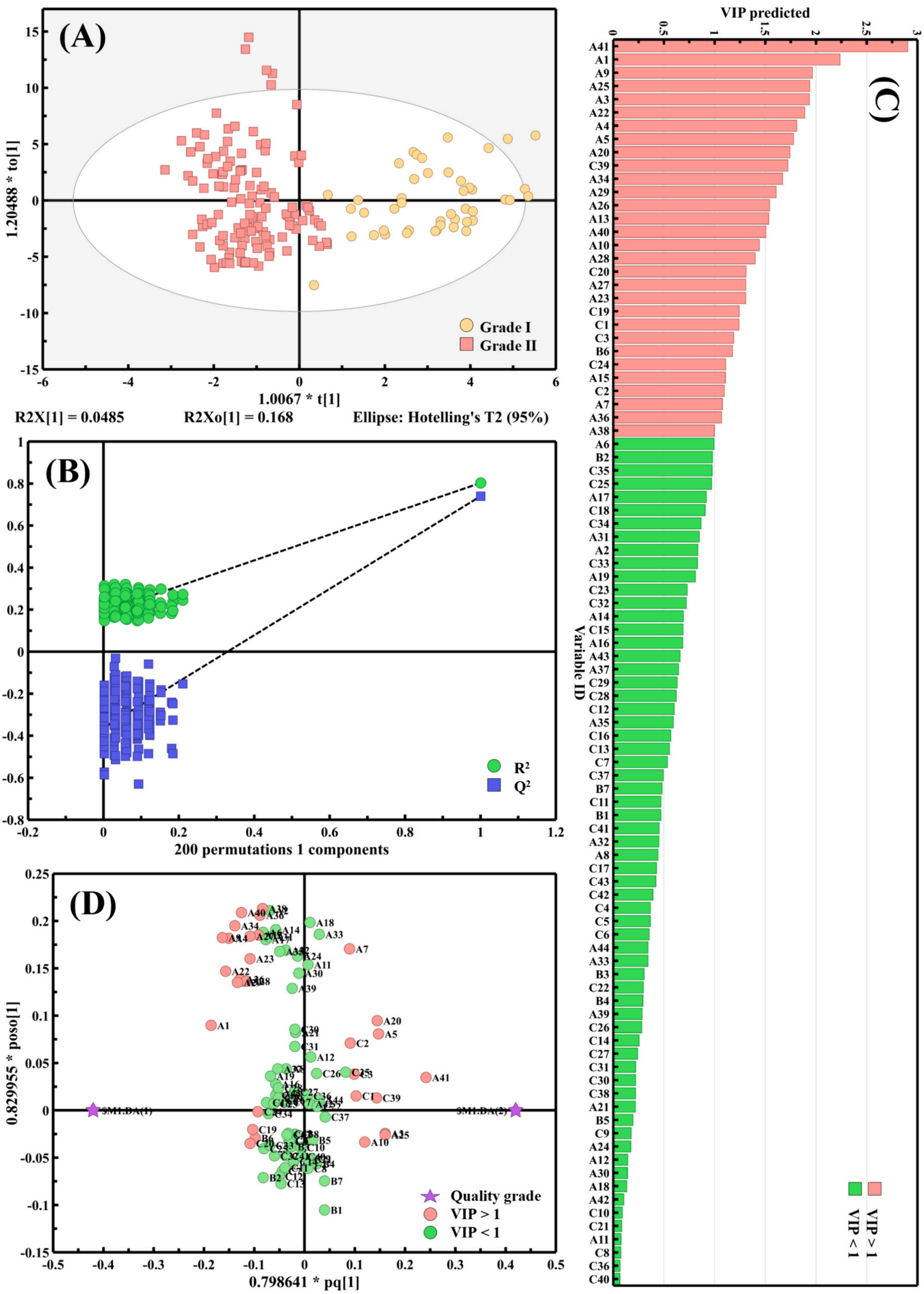

Fig. 2. OPLS-DA analysis based on fusion of GC-*E*-Nose, *E*-tongue, and *E*-eye. (A) Score plots ($R^2Y = 0.803$, $Q^2 = 0.740$); (B) Permutation plots ($R^2 = 0.187$, $Q^2 = -0.367$); (C) VIP values; (D) Loading plots.

independent *E*-tongue, CBT samples from two grades were completely mixed together, and it was also challenging to attain a superior distinction (Fig. 1B). Independent *E*-eye demonstrated a considerable separation tendency for the two groups of CBT samples, but there was still some overlap and a low aggregation degree (Fig. 1C). In addition, permutation tests indicated that there was no overfitting phenomenon in the aforementioned three models (Fig. 1D, E and F). In summary, it is challenging to achieve the accurate identification and differentiation of different quality grades using a single intelligent sensory approach. The application of data fusion strategy provides a promising prospect for the establishment of classification model. Many scholars have underscored the superiority of data fusion strategy (Banerjee, Chattopadhyay, Tudu, Bhattacharyya, & Bandyopadhyay, 2014; Song et al., 2022).

To verify the feasibility of data fusion strategy, a fusion data matrix with integrated GC-*E*-Nose, *E*-tongue, and *E*-eye was constructed for OPLS-DA analysis. All the original data were normalized to mitigate the effects of varying data dimensions from diverse sources. As illustrated in Fig. 2A, a satisfactory discrimination was achieved through the fusion data of GC-*E*-Nose, *E*-tongue, and *E*-eye. Grade I samples were predominantly distributed on the positive X-axis, whereas grade II samples were mainly located on the negative X-axis. The model parameters ($R^2Y = 0.803$, $Q^2 = 0.740$) signified high accuracy and predictive capability. Furthermore, 200 iterations of permutation tests were performed to validate the reliability. As shown in Fig. 2B, the simulated values were all lower than the true values, and the intercept between the Q2 regression line and the Y-axis was < 0, indicating that there was no overfitting phenomenon ($R^2 = 0.187$, $Q^2 = -0.367$). Additionally, the variable importance in projection (VIP) values were introduced to further scrutinize the key differential variables between the two grades of CBT samples. Generally speaking, the higher the VIP value, the greater the contribution. According to the criterion of VIP > 1, 30 variables were identified as the key differential variables (Fig. 2C). Among them, 22 variables were from GC-*E*-Nose, 7 variables were from *E*-eye, and only 1 variable was from *E*-tongue. The specific variables that explained the differences in quality levels were further evaluated by the corresponding loading plots in the OPLS-DA model (Fig. 2D). The distance between the individual variable and the main cluster is positively correlated with its effect on the classification. Representative variables such as A3 (dimethyl sulfide), A5 ((4*Z*)-1,4-hexadiene), A7 (3-ethylpentane), A10 (methyl propanoate), A20 (methyl nonyl ketone), A25 (dimethyl sulfide), A41 (citronellal), C1 (L*), C2 (a*), C3 (b*), and C39 (2912) were positioned nearer to the grade I samples, signifying higher correlations with the quality of grade I. In addition, 19 key variables including A1 (acetaldehyde), A4 (trans-1,4-hexadiene), A9 (benzene), A13 (2-methylbutanal), A15 (dimethyl disulfide), A22 (ethyl alcohol), A23 (3-methylbut-1-ene), A26 (cyclopentadiene), A27 (ethyl acetate), A28 (isovaleraldehyde), A29 ((2*E*)-4-methyl-2-hexene), A34 (propionic acid), A36 (dimethyl disulfide), A38 (3-hexanone), A40 (furfural), B6 (ANS), C19 (2113), C20 (2114), and C24 (2130) were more closer to the grade II samples. Dimethyl sulfide contributes to a sweet aroma, exerting a pivotal influence on the overall sensory profile of black tea (Yang et al., 2024). Additionally, it serves as a vital factor in preserving the freshness of tea (Tao, Guo, Zhang, & Liu, 2022). Furfural imparts a distinct baked and roasted aroma, which exhibits a significant positive correlation with the baking attributes of Oolong tea (Zhu et al., 2015).

Overall, the above results confirmed that data fusion strategy could effectively realize the qualitative classification of different grades of CBTs. In other words, the classification performance of the model based on fusion data was superior to that based on independent GC-*E*-Nose, *E*-tongue, and *E*-eye. This pioneering approach facilitates the integration of datasets from diverse intelligent sensory technologies, enhancing the accuracy and dependability of classification. In conclusion, the data fusion strategy of GC-*E*-Nose, *E*-tongue, and *E*-eye combined with multivariate statistical analysis is a promising method for the recognition of quality grade.

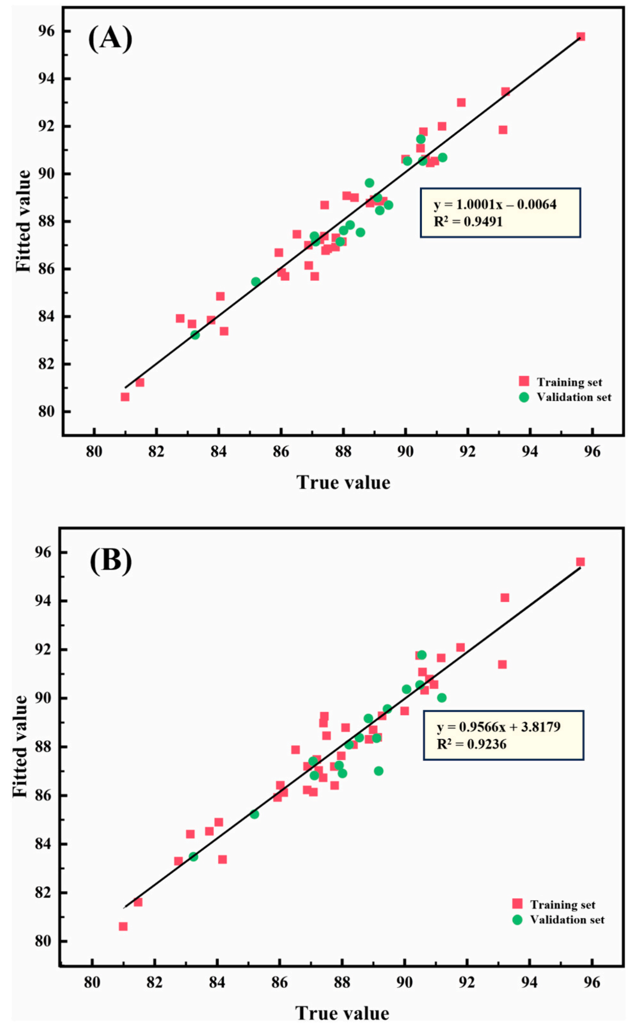

Fig. 3. Quantitative evaluation of the CQS based on PLSR and SMLR analyses. (A) PLSR analysis; (B) SMLR analysis. CQS represented comprehensive quality score. Training set was marked with box; Validation set was marked with circle.

3.3.1. Quantitative evaluation based on PLSR and SMLR analysis

To quantitatively evaluate the comprehensive quality of CBT samples, PLSR was introduced for quantitative assessment of the CQS. As an innovative statistical analysis method, PLSR focuses on the regression modeling of multi-dependent variables to multi-independent variables. It is particularly efficacious when the variables are highly linearly correlated. In this study, 56 CBT samples were randomly divided into 2 subsets: 39 samples as the training set and 17 samples as the validation set. The results of PLSR model based on independent GC-*E*-Nose, *E*-tongue, and *E*-eye as well as their fusion are detailed in Table S3. R^2Y signifies how well the model fits the data, and Q^2 denotes the model's predictive power. Compared with a single intelligent sensory technology, the fitting ability and prediction ability of fusion-based data models have been significantly improved. Subsequently, the correlation between the actual values and predicted values of CQS was further investigated based on fusion analysis. A superior regression line with R^2 of 0.9491 was attained, indicating excellent fitness and reliable predictive ability (Fig. 3A). The absolute error between the actual value and the fitted value was within 1.39 point, and the relative error was within 1.62% (Table 2). The results demonstrated that fusion of GC-*E*-Nose, *E*-tongue, and *E*-eye combined with PLSR analysis could achieve excellent quantitative assessment of CBT quality.

Moreover, stepwise multiple linear regression (SMLR) was employed to further explore the representative variables that significantly affected

Table 2
Error analysis of quantitative prediction of the CQS based on fusion strategy.

No.	True values	PLSR			SMLR			Category
		Fitted values	Absolute errors	Relative errors (%)	Fitted values	Absolute errors	Relative errors (%)	
1	95.77	95.63	−0.14	0.15	95.61	−0.16	0.16	V
2	93.46	93.21	−0.25	0.27	94.14	0.68	0.73	T
3	93.00	91.79	−1.21	1.31	92.09	−0.91	0.98	T
4	92.00	91.17	−0.83	0.91	91.66	−0.34	0.37	T
5	91.85	93.13	1.28	1.39	91.39	−0.46	0.50	T
6	91.77	90.57	−1.20	1.31	91.08	−0.69	0.75	V
7	91.46	90.49	−0.97	1.06	90.55	−0.91	1.00	T
8	91.08	90.48	−0.60	0.66	91.76	0.68	0.74	T
9	90.69	91.19	0.50	0.56	90.02	−0.67	0.74	T
10	90.62	90.00	−0.62	0.68	89.48	−1.14	1.26	T
11	90.62	90.64	0.02	0.03	90.33	−0.29	0.32	T
12	90.54	90.55	0.01	0.01	91.78	1.24	1.37	V
13	90.54	90.06	−0.48	0.54	90.37	−0.17	0.19	V
14	90.54	90.94	0.40	0.44	90.56	0.02	0.02	T
15	90.46	90.79	0.33	0.36	90.79	0.33	0.36	V
16	89.62	88.84	−0.78	0.87	89.17	−0.45	0.50	T
17	89.08	88.11	−0.97	1.08	88.79	−0.29	0.33	V
18	89.00	88.36	−0.64	0.72	88.09	−0.91	1.03	T
19	89.00	89.10	0.10	0.12	88.36	−0.64	0.72	V
20	88.92	88.99	0.07	0.08	88.70	−0.22	0.24	T
21	88.85	89.14	0.29	0.33	88.39	−0.46	0.52	V
22	88.85	89.28	0.43	0.48	89.28	0.43	0.49	T
23	88.77	88.86	0.09	0.10	88.31	−0.46	0.52	T
24	88.69	89.45	0.76	0.86	89.56	0.87	0.98	V
25	88.69	87.40	−1.30	1.46	88.98	0.29	0.33	V
26	88.46	89.17	0.71	0.80	87.01	−1.45	1.64	T
27	87.85	88.22	0.37	0.42	88.09	0.24	0.27	V
28	87.62	88.01	0.39	0.44	86.91	−0.71	0.81	T
29	87.54	88.55	1.01	1.16	88.38	0.84	0.97	V
30	87.46	86.51	−0.95	1.09	87.88	0.42	0.48	T
31	87.38	87.07	−0.31	0.36	87.41	0.03	0.03	V
32	87.38	87.39	0.01	0.01	86.73	−0.65	0.74	T
33	87.31	87.76	0.45	0.52	86.41	−0.90	1.03	T
34	87.23	87.25	0.02	0.02	87.03	−0.20	0.22	T
35	87.23	87.19	−0.04	0.04	87.48	0.25	0.29	T
36	87.15	87.90	0.75	0.86	87.24	0.09	0.10	T
37	87.15	87.97	0.82	0.94	87.63	0.48	0.55	T
38	87.15	87.11	−0.04	0.04	86.82	−0.33	0.38	T
39	87.00	86.88	−0.12	0.13	86.23	−0.77	0.88	T
40	86.92	87.75	0.83	0.96	87.19	0.27	0.32	V
41	86.85	87.50	0.65	0.75	88.46	1.61	1.86	T
42	86.77	87.43	0.66	0.76	89.26	2.49	2.87	V
43	86.69	85.93	−0.76	0.88	85.92	−0.77	0.89	T
44	86.15	86.89	0.74	0.86	87.20	1.05	1.22	T
45	85.85	86.02	0.17	0.20	86.42	0.57	0.66	T
46	85.69	86.13	0.44	0.52	86.12	0.43	0.50	T
47	85.69	87.08	1.39	1.62	86.14	0.45	0.52	T
48	85.46	85.19	−0.27	0.32	85.23	−0.23	0.28	T
49	84.85	84.05	−0.80	0.95	84.90	0.05	0.06	V
50	83.92	82.76	−1.16	1.39	83.30	−0.62	0.74	T
51	83.85	83.75	−0.10	0.12	84.53	0.68	0.81	T
52	83.69	83.14	−0.55	0.65	84.41	0.72	0.87	T
53	83.38	84.17	0.79	0.95	83.37	−0.01	0.01	V
54	83.23	83.24	0.01	0.01	83.48	0.25	0.30	T
55	81.23	81.47	0.24	0.30	81.61	0.38	0.47	T
56	80.62	80.99	0.36	0.45	80.61	−0.01	0.01	T

Note: T represented the training set; V represented the validation set; No. represented number; PLSR represented partial least square regression; SMLR represented stepwise multiple linear regression; CQS represented comprehensive quality score.

the CQS. This involved examining the variables with VIP value > 1 in the PLSR model (Table S4). With the continuous introduction of variables into the model, it was constantly tested and optimized. Finally, the optimal SMLR equation was obtained as follows:

$$\begin{aligned} CQS = {} & 84.629 - 1{*}10^{-3}{*}A1 - 3.600{*}10^{-5}{*}A16 - 4.406{*}C43 \\ & + 1.300{*}10^{-5}{*}A41 - 5.300{*}10^{-5}{*}A40 - 5.790{*}10^{-4}{*}A36 \\ & - 3.000{*}10^{-6}{*}A28 + 1.890{*}10^{-4}{*}A7 - 0.531{*}C20 + 0.133{*}C2 \\ & + 2.000{*}10^{-6}{*}A25 + 1.470{*}10^{-4}{*}A18 \end{aligned}$$

where CQS represented the comprehensive quality score, A1 represented acetaldehyde, A7 represented 3-ethylpentane, A16 represented ethyl isobutyrate, A18 represented 3-hexanone, A25 represented dimethyl sulfide, A28 represented isovaleraldehyde, A36 represented dimethyl disulfide, A40 represented furfural, A41 represented citronellal, C2 represented a*, C20 represented 2114, and C43 represented 3200.

An excellent regression line was attained with an R^2 of 0.9236 (Fig. 3B), indicating a commendable fitting performance. The Dubin-Watson value was 1.999, approximately equal to 2, indicating that there was no autocorrelation in the model. The p values and the variance inflation factor (VIF) values of each regression coefficient were less than 0.05 and 5, respectively, while the tolerance values of independent variables values were > 0.1. The aforementioned results indicated that SMLR exhibited a considerable reliability and stability, and there was no multicollinearity (Table S5). Ultimately, 12 variables were screened out to play a significant contribution to the prediction of CQS, with 10 variables derived from GC-*E*-Nose and 2 variables originated from *E*-eye. Among them, acetaldehyde (A1; fresh and green), ethyl isobutyrate (A16; floral and fruity), 3200 (C43; deep orange yellow), furfural (A40; bread, almond, and sweet), dimethyl disulfide (A36; cabbage-like and sulfury), isovaleraldehyde (A28; nutty, chocolate-like, and floral), and 2114 (C20; strong brown) were significantly negatively correlated with the quality score, while citronellal (A41; floral), 3-ethylpentane (A7), a* (C2), dimethyl sulfide (A25; corn-like and asparagus-like), and 3-hexanone (A18; sweet and fruity) exhibited a significantly positive correlation with the quality score. Notably, the standardized coefficients (SC) of citronellal (A41) and 3200 (C43) were the highest ($|SC| > 0.3$), indicating their substantial contributions to the prediction of CQS. Citronellal has a pleasant floral fragrance, and plays a vital role in black tea (Yao et al., 2023). Meanwhile, it possesses the ability to mitigate the bitterness and astringency caused by caffeine, thereby positively contributing to the formation of high-quality black tea (Suess, Brockhoff, Meyerhof, & Hofmann, 2018).

Overall, our results showed that fusion strategy exhibited superior performances in both qualitative classification and quantitative prediction when compared to independent GC-*E*-Nose, *E*-tongue, and *E*-eye analyses. It is primarily attributed that the comprehensive quality of CBT is composed of aroma, taste, and liquid color, and cannot be fully captured by single intelligent sensory technology.

4. Conclusions

Quality evaluation is of great significance to the high-quality development of the black tea industry. In the present study, different quality grades of CBT were assessed based on the fusion strategy of GC-*E*-Nose, *E*-tongue, and *E*-eye combined with OPLS-DA analysis. The classification performance of the OPLS-DA model based on fusion data was superior to that based on independent GC-*E*-Nose, *E*-tongue, and *E*-eye. This innovative approach allows the integration of data from diverse intelligent sensory technologies, enhancing the precision and dependability of classification. Furthermore, based on fusion strategy combined with PLSR analysis, a satisfactory quantitative prediction of CQS was attained. The absolute error between the actual value and the fitted value was within 1.62 point, and the relative error was within 1.38%. A total of 12 key variables were screened out by SMLR analysis, which contributed significantly to the prediction of CQS. In conclusion, the fusion of multiple intelligent sensory technologies possesses great potential in quality evaluation of CBT. As an efficient and objective method, it is expected to contribute to the improvement of sensory evaluation and the development of authenticity and standards system of tea. In the subsequent study, the number of samples will be expanded for continuous optimization of the model to make the prediction equation more accurate and stable. In addition, the feasibility of monitoring the tea processing will be explored by fusion of multiple intelligent sensory technologies.

CRediT authorship contribution statement

Lilei Wang: Writing – original draft, Visualization, Validation, Software, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jialing Xie:** Methodology, Formal analysis. **Qiwei Wang:** Visualization, Validation. **Jiajing Hu:** Software, Validation. **Yongwen Jiang:** Supervision, Funding acquisition. **Jinjin Wang:** Resources. **Huarong Tong:** Supervision. **Haibo Yuan:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Yanqin Yang:** Writing – review & editing, Supervision, Methodology, Formal analysis, Conceptualization.

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

Data will be made available on request.

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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.2024.101519.

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