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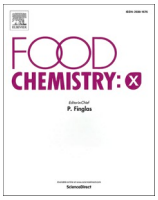

Integration of non-targeted/targeted metabolomics and electronic sensor technology reveals the chemical and sensor variation in 12 representative yellow teas

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epicatechin (PubChem CID 72276)
Gallic acid (PubChem CID 370)
Caffeine (PubChem CID 2519)
Theobromine (PubChem CID 5429)
Theophylline (PubChem CID 2153)
Theanine (PubChem CID 439378)

ABSTRACT

Yellow tea is a lightly fermented tea with unique sensory qualities and health benefits. However, chemical composition and sensory quality of yellow tea products have rarely been studied. 12 representative yellow teas, which were basically covered the main products of yellow tea, were chosen in this study. Combined analysis of non-targeted/targeted metabolomics and electronic sensor technologies (E-eye, E-nose, E-tongue) revealed the chemical and sensor variation. The results showed that yellow big tea differed greatly from yellow bud teas and yellow little teas, but yellow bud teas could not be effectively distinguished from yellow little teas based on chemical constituents and electronic sensory characteristics. Sensor variation of yellow teas might be attributed to some compounds related to bitterness and aftertaste-bitterness (4′-dehydroxylated gallocatechin-3-*O*-gallate, dehydrotheasinensin C, myricitin 3-*O*-galactoside, phloroglucinol), aftertaste-astringency (methyl gallate, 1,5-digalloylglucose, 2,6-digalloylglucose), and sweetness (maltotriose). This study provided a comprehensive understanding of yellow tea on chemical composition and sensory quality.

1. Introduction

Tea, which is made from the leaves of *Camellia sinensis* (L.) O. Kuntze, is one of the most popular non-alcoholic beverages around the world (Zhu, et al., 2017; Zhu, et al., 2020b; Zhu, et al., 2019). Over two billion people in more than 125 countries drink tea (Mei, 2015). Tea is classified into six categories according to their manufacturing process in China: green tea, black tea, oolong tea, white tea, dark tea, and yellow tea. Yellow tea is a lightly fermented tea, which is only manufactured in China. It is mainly produced in Sichuan, Anhui, Hunan, Hubei, and Zhejiang provinces (Fig. 1A). "Yellowing process" is the characteristic process of yellow tea production, which changes the chemical composition of tea leaves by thermochemical reaction and exogenous enzyme. Yellowing process give yellow tea the unique characteristics, including yellow dry tea, yellow infusion, and yellow brewed leaves ("three characteristics of yellowing"). Further, the taste of yellow tea become more smooth, fresh, and mellow after yellowing process. Yellow tea is gradually gaining recognition by consumers around the world, due to its unique aroma, refreshing silky taste, and remarkable health benefits (Kujawska, et al., 2016).

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Yellow tea can be further classified into three categories according to the tenderness of tea leaves: yellow bud tea, yellow little tea, and yellow big tea (Feng, et al., 2023). Yellow bud tea is prepared only using buds or one bud and one leaf. It is mainly produced in Sichuan, Hunan, Anhui, and Zhejiang provinces of China. Yellow bud tea usually has the highest price in these three kinds of yellow tea due to its high-grade raw materials (Zhu, et al., 2016). Yellow little tea is processed using with one bud and one or two leaves. Yellow big tea is manufactured with low-grade raw materials including old tea leaves and stems. The basic processing procedures, which include fixing, yellowing, and drying, is same in these three kinds of yellow tea. However, a significant difference exists in these three kinds of yellow tea in the aspect of technological parameters of yellowing process, including temperature, air humidity, oxygen supply, timing, duration, leaf water content, and leaf tenderness (Fig. 1B). Therefore, the chemical profile and sensory quality of yellow teas in the Chinese market vary considerably, due to the difference in the tea varieties, raw material grade and origin, and processing details.

Researches on the chemical profile and sensory quality of yellow tea has made some progress in recent years. Wei et al. (2020) reported that galloylated catechins and polyphenol-amino acid ratio decreased significantly after yellowing processes. Yellowing process also decrease in the concentration of some compounds casing grassy, floral, and fruity aromas, but increase in the concentration of some compounds having mushroom and sweet aromas (Wei, et al., 2022). However, previous studies focused on the influence of yellowing process on the chemical profile and sensory quality of yellow tea, the chemical composition and sensory quality of representative yellow tea products in the Chinese market have rarely been studied. Only Wang et al. (2021) preliminary reported that the taste properties of yellow tea samples from different regions of China vary considerably according to the human sensory evaluation. 22 chemical components, which belonged to catechins, free amino acids, alkaloids, and flavonoids, had an important contribution to the taste characteristics of yellow tea according to the analysis of colorimetric method and high-performance liquid chromatography (HPLC). Therefore, it is necessary to conduct a systematic and in-depth study on the chemical composition and sensory quality of representative yellow tea products by using the modern analytical techniques, such as electronic sensor technology and metabolomics technology.

Electronic sensor technology includes electronic nose (E-nose), electronic tongue (E-tongue), and electronic eye (E-eye). E-nose, E-tongue, and E-eye are equipped with gas, liquid, and color sensors, which are mainly developed to mimic olfactory, gustatory, and vision systems of human being. Electronic sensor technology provides the global information about the sample, instead of the information on the particular components. Electronic sensor technology is an important method for the sensory evaluation, which provide a more sensitive, digital, and objective description of sensory characteristics in comparison to human (Chang, et al., 2020; Li, et al., 2015). Individual E-nose, E-tongue, or E-eye has been used for the sensory evaluation of the tea grades, tea varieties, and tea origin in recent years. However, tea sensory quality is mainly determined by aroma, taste, and color. Little literature has been reported about the combined application of E-nose, E-tongue, and E-eye for more comprehensively sensory evaluation of tea quality, especially yellow tea (Xu, et al., 2013; Xu, et al., 2019). On the other hand, liquid chromatography–mass spectrometer (LC–MS)-based untargeted and targeted metabolomics has been widely used for the

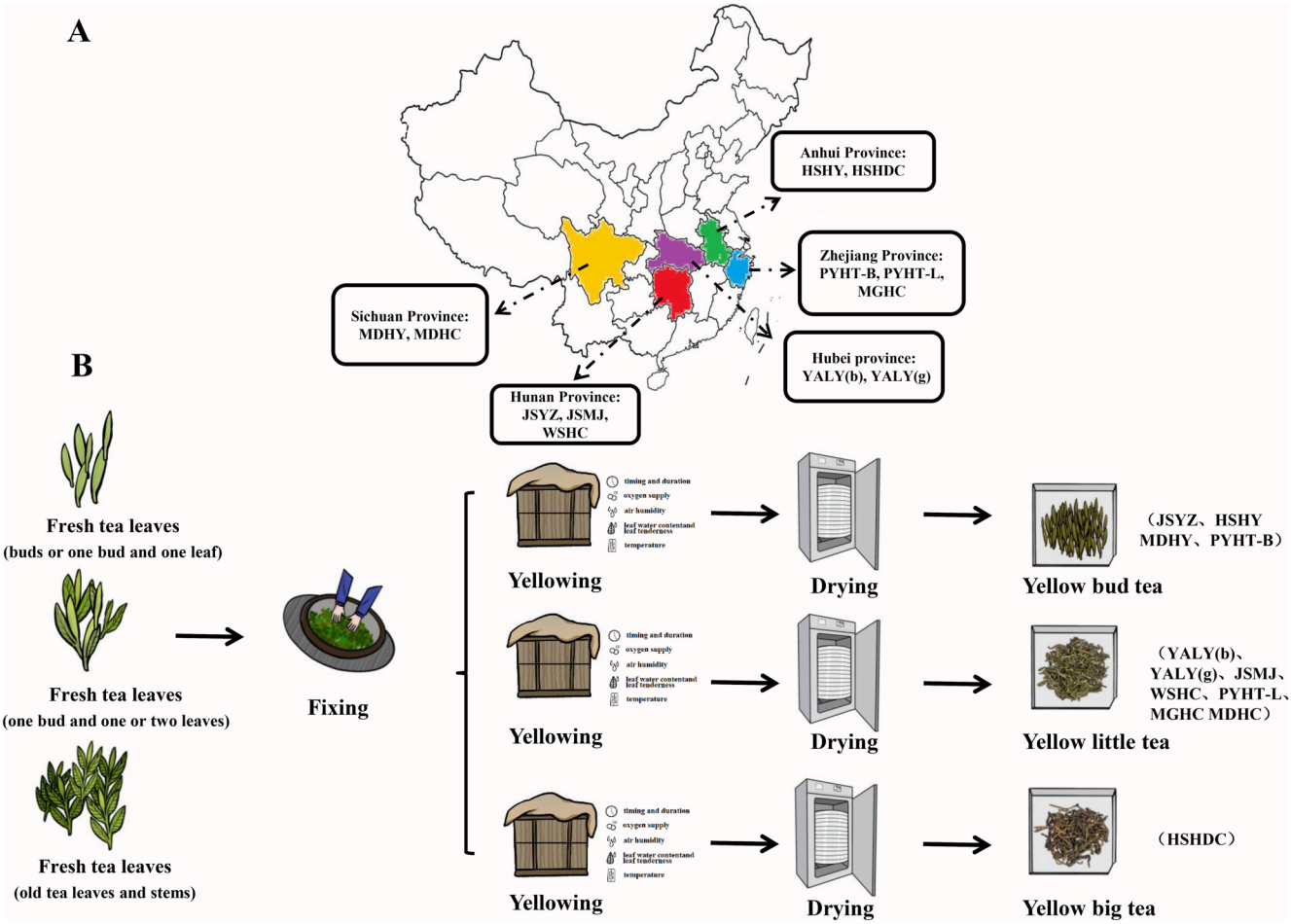

Fig. 1. The place of origin (A) and manufacturing process (B) of 12 representative yellow tea samples. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

metabolic profiling of tea by detecting hundreds of endogenous metabolites (Wang, et al., 2018). Therefore, multiple electronic sensing analysis techniques combined with untargeted and targeted metabolomics can provide a comprehensive investigation of the sensory quality and chemical composition of yellow tea.

In this study, 12 representative yellow teas, which are divided into yellow bud tea, yellow little tea, and yellow big tea and produced in five provinces of China, were used in this study. These 12 representative yellow tea products basically cover the main products of yellow tea in China. These 12 yellow teas were analyzed by untargeted and targeted metabolomics to reveal the chemical variation. Three electronic sensory evaluation methods (E-nose, E-tongue, and E-eye) were used to reveal the sensory quality characteristics of these 12 representative yellow teas. This study provided a comprehensive understanding of yellow tea through the joint research of chemical composition and sensory quality, thus providing a further theoretical basis for the quality control of yellow tea.

2. Material and methods

2.1. Chemicals and materials

Deionized water was produced by a Milli-Q water purification system (Millipore, Billerica, Massachusetts, USA). Acetonitrile of LC–MS grade was purchased from Merck Co., Ltd. (KGaA, Darmstadt, Germany). Formic acid was purchased from Sigma Co., Ltd. (St. Louis, Missouri, USA). Folin-Ciocalteu reagent, aluminum chloride, and anthrone were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). The standards of rutin (≥98 %) and gallic acid (≥98 %) were purchased from Must Biotechnology Co. Ltd. (Chengdu, Sichuan, China). The standards of catechin, epicatechin (EC), epicatechin gallate (ECG), gallocatechin (GC), epigallocatechin (EGC), gallocatechin gallate (GCG), epigallocatechin gallate (EGCG), gallic acid, afzelin, quercetin, kaempferol, and myricetin were purchased from Solarbio Science & Technology Co., Ltd. (Beijing, China).

12 representative yellow teas, which were composed of four yellow bud teas, seven yellow little teas, and one yellow big tea (Huoshan Huangdacha, HSHDC), were used in this study. Four yellow bud teas consisted of Junshan Yinzhen (JSYZ), Huoshan Huangya (HSHY), Mengding Huangya (MDHY), and Pingyang Huangtang bud tea (PYHT-B). Seven yellow little teas were Yuan'an Luyuan (bar shape) (YALY(b)), Yuan'an Luyuan (grain shape) (YALY(g)), Junshan Maojian (JSMJ), Weishan Huangcha (WSHC), Pingyang Huangtang little tea (PYHT-L), Mogan Huangcha (MGHC), and Mengding Huangcha (MDHC). The samples of JSYZ, JSMJ, and WSHC were produced in Hunan province; the samples of MDHY and MDHC were produced in Sichuan province; the samples of HSHY and HSHDC were produced in Anhui province; the samples of PYHT-B and PYHT-L were produced in Zhejiang province; the samples of YALY(b) and YALY(g) were produced in Hubei province (Fig. 1A). These yellow teas were produced according to the typical process of corresponding production area (Fig. 1B). Three production batches of yellow tea samples were collected to avoid artificial variation. The yellow tea samples were milled into powder (100 mesh) and freeze-dried for the future analysis.

2.2. Determination of major chemical components by colorimetric analysis

The contents of total polyphenols and total flavonoids in the yellow tea samples were determined by using Folin–Ciocalteu colorimetric assay (Blainski, et al., 2013) and aluminum trichloride colorimetric method (Wang, et al., 2018; Ye, et al., 2018), respectively. The content of soluble sugars in tea was measured by the anthrone-sulfuric acid method (Wang, et al., 2018).

2.3. LC–MS-based non-targeted metabolomic analysis

The non-targeted metabolomic analysis of yellow teas was performed on an ultrahigh performance liquid chromatography–quadrupole-time-of-flight mass spectrometry (UHPLC–QTOF-MS; Agilent Technologies Inc., Santa Clara, California, USA) equipped with an ACQUITY HSS T3 column (1.8 mm, 2.1 mm × 100 mm; Waters, Milford, Massachusetts, USA), as described previously (Zhu, et al., 2017; Zhu, et al., 2021b). The metabolite profiles were analyzed by using the Mass Profiler Professional 13.0 software (Agilent Technologies Inc., Santa Clara, California, USA).

2.4. Targeted quantitative analysis of catechins, alkaloids, and amino acids

The contents of six catechins (EGCG, EGC, ECG, GCG, EC, and catechin), gallic acid, and three alkaloids (caffeine, theobromine, and theophylline) were measured as described previously (Gong, et al., 2020). Briefly, the HPLC system consisted of a Daojin LC-2010A controller equipped with an Wondasil C 18 Superb column (5 μm, 4.6 × 150 mm), and an SPD-M20A diode array detector (Daojin, Japan). Water (mobile phase A) and a N,*N*-dimethylformamide containing 39.5 % (v/v), methanol and 1.5 % (v/v) glacial acetic acid (mobile phase B) were used for chromatographic elution: 0–10 min, 9 % B; 15 min,14 % B; 27 min, 23 % B; 31 min, 36 % B; 34 min, 9 % B; 34–38 min, 9 % B. The column temperature was kept at 30 ℃. The injected sample (10 μL) was eluted at 1.0 mL/min and monitored at 278 nm. The levels of 18 amino acids in the yellow tea samples were measured by HPLC after Waters AccQ·Tag™ precolumn derivatization (Milford, MA, USA) by using AccQ·Fluor™ Reagent Kit (Waters, Milford, MA, USA) (Gong, et al., 2020).

2.5. Electronic sensory evaluation

2.5.1. E-nose analysis

The smell analysis was carried out by using a FOX4000 E-nose equipment (Airsense Analytics, Alpha, France). This instrument was equipped with 6 different metal oxide semiconductor sensors, including P40/1, T70/2, PA/2, P30/1, LY2/G, and LY2/gCT. A total of 1.0 g crushed yellow tea sample was sealed in a jaw bottle, and then heated at 60 ℃ for 20 min. The pressure value of the carrier gas air generator is set to 0.35 bar. 1 mL gas from the heated yellow tea sample was injected into the E-nose equipment for acquiring the electronic signal. The average acquisition time of electronic nose is 90 s.

2.5.2. E-tongue analysis

The taste evaluation was performed by using an E-tongue system (TS-5000Z, Insent Inc., Atsugi-shi, Japan), which consisted of several sensor probes and a reference probe, including AAE (umami), CAO (sourness), CTO (saltiness), COO (bitterness), AE1 (astringency), and GL1 (sweetness). The detail method was described previously with some modifications (Xu, et al., 2019). Briefly, 3.0 g of yellow tea sample was infused with 150 mL of boiled water for 5 min, and then the tea infusion was rapidly filtered and cooled to room temperature for taste evaluation. The sensor probes were dipped into yellow tea infusion to measure taste strength. The thresholds for sourness and saltiness were − 13 and − 6, respectively, and zero for other tastes. The E-tongue measurement was composed of three phases: sample detection phase (30 s), aftertaste detection phase (30 s), and cleaning phase (120 s). Each yellow tea sample was conducted in triplicate, and each measurement was detected for four times to get an average value. The acquired taste scores were subjected to multivariate analysis to characterize the taste quality of these yellow tea samples.

2.5.3. E-eye analysis

Color of the yellow tea infusion was measured by colorimeter SC-80C

system (Kangguang Instrument Co. Ltd., Beijing, China) in transmission mode using CIE D65/10° illuminant/observer conditions. The color of tea infusion was scored according to sensors, CIE *L*, a*, b**. *L** denoted lightness, *a** denoted redness and greenness, *b** denoted yellowness and blueness. Distilled water was utilized as blank sample, and its color was denoted as L0, a0 and b0. Each sample was performed in triplicates at the room temperature of 25 ± 1℃.

2.6. Multivariate statistical analysis and network visualization

The data related to identified metabolites and electronic sensory characteristics was imported into the OmicShare tools for statistical analysis, which was a free online platform for multivariate statistical analysis (https://www.omicshare.com/tools). The unsupervised principal component analysis (PCA) was applied to obtain an overview of chemical variation among 12 yellow tea samples. Upset plot and heatmap analysis were performed to analyze the similarities and differences of chemical variation. Two-way orthogonal partial least square analysis (O2PLS) and correlation analysis were applied to reveal the taste-metabolite association. Quantification results of identified metabolites were expressed as mean ± standard deviation (SD) for triplicate experiments. The analysis of one-way analysis of variance (ANOVA) was conducted with PASW statistical software (SPSS 26.0, SPSS Inc., Chicago, IL, USA). Duncan's multiple range test was used to compare the metabolite variation.

3. Results

3.1. Non-targeted metabolomics analysis of yellow teas

LC–MS-based metabolomics was applied to characterize the global chemical profiles of 12 representative yellow teas (Fig. 2A). These 12 yellow teas were produced in the main producing areas of China (Hunan, Hubei, Sichuan, Zhejiang, and Anhui provinces) and made by three tenderness levels of tea raw materials (Fig. 1). These 12 representative yellow tea products basically cover the main products of yellow tea in China. A total of 69 metabolites were identified based on a comparison of retention times, MS and MS/MS spectra with standards, metabolome databases, and/or references (Table 1) (Zhu, et al., 2017). In order to analyze the changes of metabolites in different kinds of yellow tea from a macro perspective, we selected JSYZ, which has high raw material tenderness and relatively simple processing technology, as

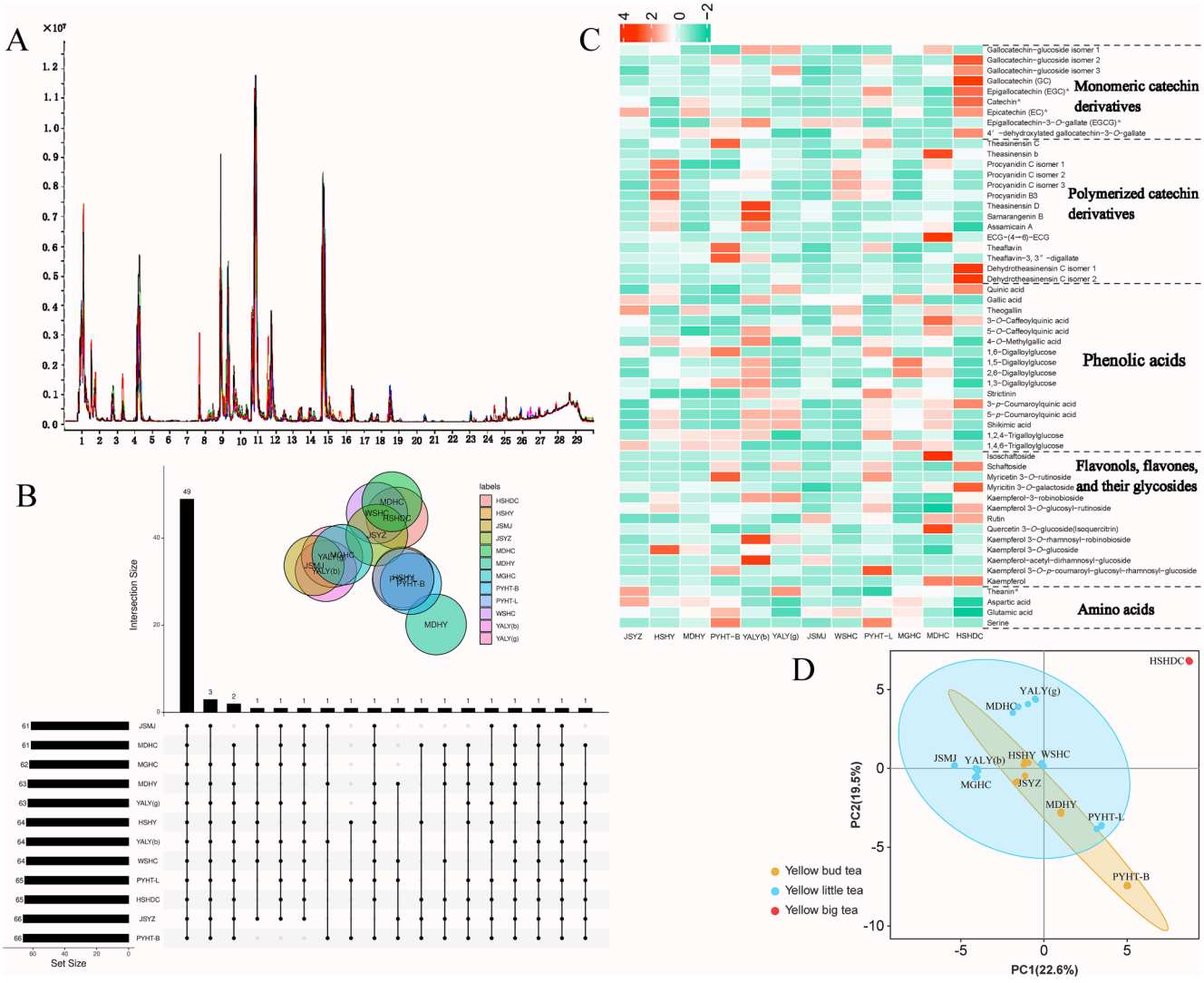

Fig. 2. UHPLC–QTOF-MS-based metabolomic analysis of 12 representative yellow teas: (A) Base peak chromatogram acquired in the negative ionization mode; (B) Upset plot, the numbers representing the detected compounds in yellow tea samples; (C) Heatmap of metabolites according to UHPLC–QTOF-MS and HPLC-UV data. *The heatmap of metabolite was created according to its HPLC-UV data; (D) PCA score plot, $R^2X = 0.975$, $Q^2 = 0.798$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
The metabolite analysis in 12 representative yellow teas according to UPLC-QTOF-MS.

NO	Names	Fold change (11 yellow teas versus JSYZ)[#]											
		JS YZ	HS HY	MDHY	PYHT-B	YALY (b)	YALY (g)	JS MJ	WSHC	PYHT-L	MGHC	MDHC	HSHDC
Monomeric catechin derivatives													
1	Gallocatechin-glucoside isomer 1	1.00^{d}	1.13^{c}	0.61 ^{g}	0.41^{i}	1.52^{a}	1.51^{a}	0.76^{e}	0.49 ^{h}	0.64 ^{g}	1.13^{c}	1.42^{b}	0.69^{f}
2	Gallocatechin-glucoside isomer 2	1.00^{e}	1.41^{c}	1.27^{d}	3.60^{b}	0.81^{f}	1.40^{c}	0.42 ^{g}	0.77^{f}	3.66^{b}	0.48 ^{g}	1.09^{e}	6.59^{a}
3	Gallocatechin-glucoside isomer 3	1.00^{j}	2.35^{e}	1.55 ^{h}	2.47^{d}	2.13^{f}	3.58^{b}	0.86 ^{k}	1.41^{i}	2.47^{d}	1.69 ^{g}	2.72^{c}	4.03^{a}
4	Gallocatechin (GC)	1.00 ^{g}	1.58^{c}	1.20^{e}	1.60^{c}	1.35^{d}	1.87^{b}	0.59 ^{h}	1.09^{f}	1.60^{c}	0.95 ^{g}	1.40^{d}	4.32^{a}
5	Epigallocatechin (EGC) †	1.00^{c}	0.91^{d}	0.80^{e}	0.87^{d}	0.57 ^{g}	0.52 ^{h}	0.61 ^{g}	0.73^{f}	2.01^{b}	0.78^{ef}	0.13^{i}	2.48^{a}
6	Catechin †	1.00^{c}	0.60^{i}	1.13^{b}	0.97^{d}	0.88^{e}	0.73 ^{g}	0.69 ^{h}	0.79^{f}	1.11^{b}	0.88^{e}	0.68 ^{h}	1.53^{a}
7	Epicatechin (EC) †	1.00^{b}	0.67^{hi}	0.97^{c}	0.75^{e}	0.67^{gh}	0.71^{f}	0.67^{gh}	0.70 ^{fg}	0.63^{i}	0.86^{d}	0.62^{j}	1.05^{a}
8	Epigallocatechin-3-*O*-gallate (EGCG) †	1.00^{d}	0.58^{j}	0.64^{i}	1.18^{c}	1.42^{a}	0.93^{c}	1.21^{b}	1.19^{c}	0.64^{i}	0.77 ^{g}	0.69 ^{h}	0.90^{f}
9	4′-Dehydroxylated gallocatechin-3-*O*-gallate	1.00 ^{g}	0.65 ^{h}	1.48^{b}	1.34^{d}	1.16^{f}	0.06 ^{k}	0.02 ^{l}	1.30^{e}	1.39^{c}	0.44^{j}	0.58^{i}	2.21^{a}
Polymerized catechin derivatives													
10	Theasinensin C	1.00 ^{h}	2.68^{c}	1.52^{e}	5.73^{a}	1.47^{ef}	1.39 ^{g}	0.74^{i}	2.03^{d}	4.22^{b}	0.97 ^{h}	1.01 ^{h}	1.41 ^{fg}
11	Theasinensin B	1.00^{f}	0.95^{f}	1.32^{d}	0.87 ^{g}	1.57^{b}	1.22^{e}	0.47^{i}	0.71 ^{h}	1.43^{c}	1.25^{e}	3.31^{a}	1.53^{b}
12	Procyanidin C isomer 1	1.00^{f}	2.54^{a}	0.00^{i}	0.00^{i}	1.32^{d}	1.11^{e}	0.78 ^{g}	1.62^{c}	1.30^{d}	0.35 ^{h}	1.71^{b}	1.10^{e}
13	Procyanidin C isomer 2	1.00 ^{g}	2.61^{a}	0.93^{hi}	1.09^{f}	1.80^{c}	1.13^{f}	0.99^{gh}	2.25^{b}	1.44^{e}	0.60^{j}	1.56^{d}	0.90^{i}
14	Procyanidin C isomer 3	1.00 ^{g}	2.63^{a}	1.44^{e}	1.87^{c}	1.95^{c}	1.08 ^{fg}	1.08 ^{fg}	2.63^{a}	2.09^{b}	1.04 ^{g}	1.73^{d}	1.21^{f}
15	Procyanidin B3	1.00 ^{fg}	2.50^{a}	1.03^{f}	1.34^{e}	1.38^{e}	0.97 ^{g}	0.84 ^{h}	1.90^{b}	1.64^{c}	0.71^{i}	1.46^{d}	1.45^{d}
16	Theasinensin D	1.00^{i}	3.25^{b}	0.75 ^{k}	1.61^{f}	6.66^{a}	1.40 ^{g}	2.35^{c}	0.87^{j}	1.09 ^{h}	1.68^{e}	1.79^{d}	0.75 ^{k}
17	Samarangenin B	1.00 ^{g}	2.71^{b}	0.72 ^{h}	1.55^{d}	5.29^{a}	1.22^{f}	2.03^{c}	0.77 ^{h}	1.06 ^{g}	1.47^{e}	2.09^{c}	0.52^{i}
18	Assamicain A	1.00^{f}	1.29^{b}	0.83 ^{h}	1.12^{d}	1.48^{a}	1.03^{e}	0.98^{f}	0.92 ^{g}	1.13^{d}	1.14^{d}	1.21^{c}	0.71^{i}
19	ECG-(4β → 6)-ECG	1.00^{c}	1.40^{b}	0.00^{f}	0.74^{d}	0.00^{f}	0.00^{f}	0.00^{f}	0.00^{f}	0.00^{f}	0.00^{f}	6.62^{a}	0.62^{e}
20	Theaflavin	1.00^{f}	1.51^{d}	1.53^{d}	3.61^{a}	1.24^{e}	1.49^{d}	0.30 ^{h}	1.21^{e}	2.33^{b}	0.31 ^{h}	0.69 ^{g}	1.85^{c}
21	Theaflavin-3,3″-digallate	1.00^{d}	1.11 ^{cd}	1.19^{c}	2.56^{a}	1.75^{b}	0.53 ^{g}	0.40 ^{h}	1.03^{d}	0.72^{f}	0.33 ^{h}	1.19^{c}	0.82^{e}
22	Dehydrotheasinensin C isomer 1	1.00^{f}	3.87^{bc}	1.16^{f}	4.13^{b}	1.90^{e}	4.00^{bc}	0.00 ^{g}	0.72^{f}	3.60^{c}	0.00 ^{g}	2.46^{d}	20.93^{a}
23	Dehydrotheasinensin C isomer 2	1.00 ^{h}	1.54^{e}	1.10 ^{g}	2.04^{d}	1.19^{f}	3.09^{b}	0.64^{i}	1.55^{e}	2.15^{c}	1.22^{f}	2.02^{d}	10.92^{a}
Phenolic acids													
24	Quinic acid	1.00^{i}	1.96^{c}	1.21 ^{h}	0.86^{j}	1.47^{f}	2.32^{b}	1.47^{f}	1.61^{e}	1.72^{d}	1.27 ^{g}	1.96^{c}	2.63^{a}
25	Gallic acid	1.00^{b}	0.90^{c}	0.00^{f}	0.00^{f}	1.11^{a}	0.41^{e}	0.66^{d}	0.67^{d}	0.00^{f}	1.12^{a}	0.00^{f}	0.00^{f}
26	Theogallin	1.00^{a}	0.00^{f}	0.78^{c}	0.36^{d}	0.00^{f}	0.00^{f}	0.00^{f}	0.85^{b}	0.00^{f}	0.34^{e}	0.78^{c}	0.35^{de}
27	3-*O*-Caffeoylquinic acid	1.00^{c}	0.52 ^{g}	0.52 ^{g}	0.25^{i}	0.66^{f}	0.89^{d}	0.35 ^{h}	1.09^{c}	0.51 ^{g}	0.74^{e}	1.85^{a}	1.31^{b}
28	5-*O*-Caffeoylquinic acid	1.00^{c}	0.68^{d}	0.00 ^{g}	0.37^{f}	1.75^{a}	1.31^{b}	0.71^{d}	1.73^{a}	0.48^{e}	0.67^{d}	1.79^{a}	0.74^{d}
29	4-*O*-Methylgallic acid	1.00^{b}	2.92^{ab}	2.26^{b}	1.39^{b}	3.73^{ab}	0.52^{b}	2.12^{b}	0.89^{b}	2.51^{ab}	7.77^{a}	2.35^{ab}	0.38^{b}
30	1,6-Digalloylglucose	1.00^{d}	0.00^{f}	1.25^{c}	2.23^{a}	0.00^{f}	0.00^{f}	0.00^{f}	0.47^{e}	1.74^{b}	0.00^{f}	0.00^{f}	0.00^{f}
31	1,5-Digalloylglucose	1.00^{i}	3.21^{d}	1.98^{f}	1.76 ^{g}	5.14^{b}	0.74^{j}	2.43^{e}	1.51 ^{h}	3.18^{d}	6.10^{a}	3.58^{c}	0.33 ^{k}
32	2,6-Digalloylglucose	1.00 ^{k}	6.57^{d}	5.19^{e}	4.61^{f}	10.24^{b}	1.40^{j}	4.40 ^{g}	3.08 ^{h}	2.47^{i}	11.85^{a}	8.57^{c}	0.61 ^{l}
33	1,3-Digalloylglucose	1.00 ^{g}	0.85 ^{h}	1.27^{c}	1.79^{b}	1.93^{a}	0.41^{j}	1.14^{f}	0.30 ^{k}	1.23^{d}	0.67^{i}	1.17^{e}	0.12 ^{l}
34	Strictinin	1.00^{e}	0.00 ^{h}	0.00 ^{h}	0.00 ^{h}	1.64^{b}	0.85^{f}	0.99^{e}	1.02^{e}	2.06^{a}	1.36^{c}	1.11^{d}	0.59 ^{g}
35	3-*p*-Coumaroylquinic acid	1.00^{e}	6.76^{bc}	3.16^{de}	2.87^{de}	5.01 ^{cd}	8.20^{b}	1.39^{e}	2.40^{e}	8.34^{b}	5.55^{c}	5.87^{c}	12.20^{a}
36	5-*p*-Coumaroylquinic acid	1.00 ^{l}	8.30^{d}	1.99^{i}	1.79^{j}	10.10^{a}	9.84^{b}	2.80 ^{g}	1.61 ^{k}	7.54^{e}	6.29^{f}	8.60^{c}	2.60 ^{h}
35	Shikimic acid	1.00^{i}	8.84^{c}	2.41 ^{g}	1.63 ^{h}	9.78^{a}	9.95^{a}	2.77^{f}	1.43 ^{h}	7.84^{d}	7.41^{e}	9.31^{b}	2.27 ^{g}
38	1,2,4-Trigalloylglucose	1.00^{i}	1.83^{e}	1.95^{d}	2.00^{c}	2.13^{b}	0.18 ^{k}	1.30 ^{g}	0.89^{j}	2.52^{a}	1.04 ^{h}	1.47^{f}	0.20 ^{k}
39	1,4,6-Trigalloylglucose	1.00^{b}	0.51^{f}	0.94^{c}	0.89^{d}	0.00^{i}	0.09 ^{h}	0.00^{i}	0.00^{i}	0.61^{e}	1.08^{a}	0.89^{d}	0.20 ^{g}
Flavonols, flavones, and their glycosides													
40	Isoschaftoside	1.00 ^{h}	1.35 ^{g}	0.74^{i}	5.91^{c}	0.43^{j}	4.75^{e}	0.00 ^{k}	1.70^{f}	6.15^{b}	0.73^{i}	49.96^{a}	5.40^{d}
41	Schaftoside	1.00^{f}	1.27^{e}	0.75 ^{h}	3.41^{c}	0.51^{i}	3.68^{b}	0.40^{j}	1.96^{d}	3.71^{b}	0.81 ^{g}	0.11 ^{k}	5.26^{a}
42	Myricetin 3-*O*-rutinoside	1.00^{e}	1.00^{e}	2.18^{d}	3.21^{c}	26.90^{a}	1.00^{e}	10.69^{b}	1.00^{e}	1.00^{e}	1.00^{e}	1.00^{e}	1.00^{e}
43	Myricitin 3-*O*-galactoside	1.00^{c}	0.70^{f}	1.06^{c}	0.96^{d}	0.83^{e}	0.53 ^{g}	0.00 ^{h}	0.43 ^{h}	1.06^{c}	0.73^{f}	1.23^{b}	2.28^{a}
44	Kaempferol-3-robinobioside	1.00 ^{g}	2.79^{c}	1.67^{e}	1.25^{f}	4.03^{b}	4.26^{a}	0.95 ^{g}	1.27^{f}	2.76^{c}	0.62 ^{h}	0.00^{i}	2.65^{d}
45	Kaempferol 3-*O*-glucosyl-rutinoside	1.00^{d}	1.27^{c}	0.93^{f}	0.98^{de}	0.96^{e}	0.92^{f}	0.49 ^{h}	0.85 ^{g}	1.37^{b}	0.32^{i}	0.00^{j}	1.48^{a}
46	Rutin	1.00^{i}	1.72 ^{h}	5.10 ^{g}	9.88^{d}	6.22^{f}	0.00^{j}	15.51^{b}	8.11^{e}	10.96^{c}	0.00^{j}	15.55^{b}	17.23^{a}
47	Quercetin 3-*O*-glucoside (Isoquercitrin)	1.00^{e}	1.09^{c}	1.08^{c}	0.36^{i}	0.40 ^{h}	1.01^{e}	1.16^{b}	0.43 ^{g}	1.08^{c}	1.04^{d}	2.82^{a}	0.86^{f}
48	Kaempferol 3-*O*-rhamnosyl-robinobioside	1.00^{gh}	17.21^{e}	19.81 ^{cd}	19.16^{d}	89.52^{a}	46.01^{b}	20.44^{c}	0.00 ^{h}	17.70^{e}	2.07 ^{g}	0.00 ^{h}	13.50^{f}
49	Kaempferol 3-*O*-glucoside	1.00 ^{h}	3.55^{a}	2.05^{b}	0.89^{i}	1.79^{c}	1.20 ^{g}	1.06 ^{h}	1.35^{f}	1.59^{d}	0.79^{j}	0.00 ^{k}	1.47^{e}
50	Kaempferol-acetyl-dirhamnosyl-glucoside	1.00^{e}	1.00^{e}	2.18^{d}	3.21^{c}	26.90^{a}	1.00^{e}	10.69^{b}	1.00^{e}	1.00^{e}	1.00^{e}	1.00^{e}	1.00^{e}
51	Kaempferol 3-*O*-*p*-coumaroyl-glucosyl-rhamnosyl-glucoside	1.00 ^{g}	3.09^{e}	8.37^{d}	43.26^{b}	8.17^{d}	15.44^{c}	2.30^{ef}	1.90^{f}	67.88^{a}	1.00 ^{g}	1.90^{f}	8.04^{d}
52	Kaempferol	1.00^{bc}	0.51^{f}	0.97^{c}	0.24^{i}	0.76^{e}	0.43 ^{h}	0.47 ^{g}	0.85^{d}	0.50 ^{fg}	1.03^{b}	2.83^{a}	2.81^{a}
Amino acids													
56	Theanine †	1.00^{b}	0.58 ^{h}	0.68^{e}	0.59 ^{g}	0.44^{i}	1.05^{a}	0.62^{f}	0.38^{j}	0.22 ^{k}	0.71^{d}	0.74^{c}	0.58 ^{h}
57	Aspartic acid	1.00^{e}	1.06^{d}	1.18^{c}	1.27^{b}	1.18^{c}	0.98^{e}	0.62 ^{g}	1.06^{d}	1.08^{d}	0.67^{f}	1.00^{e}	1.35^{a}
58	Glutamic acid	1.00 ^{g}	1.21^{c}	1.23^{c}	1.24^{c}	1.30^{b}	1.07^{e}	0.68^{i}	1.18^{d}	1.16^{d}	0.76 ^{h}	1.03^{f}	1.43^{a}
59	Serine	1.00^{c}	0.97^{d}	1.07^{b}	1.06^{b}	0.79^{f}	0.62^{i}	0.72 ^{h}	0.73 ^{g}	1.23^{a}	0.92^{e}	0.79^{f}	0.32^{j}
Organic acids													
53	Tartaric acid	1.00^{a}	0.85^{bc}	0.87^{b}	0.85^{bc}	0.57^{f}	0.47 ^{g}	0.64^{e}	0.65^{e}	0.84^{c}	0.87^{b}	0.77^{d}	0.39 ^{h}
54	Malic acid	1.00^{f}	0.85^{i}	1.03^{e}	1.26^{a}	0.91 ^{g}	0.51 ^{k}	1.06^{d}	1.15^{b}	0.89 ^{h}	1.09^{c}	0.72^{j}	0.24 ^{l}
55	Fumaric acid	1.00^{f}	1.11^{d}	1.40^{c}	2.72^{a}	0.95 ^{g}	1.00^{f}	0.76^{i}	1.05^{e}	2.73^{a}	1.78^{b}	1.13^{d}	0.91 ^{h}
Nucleosides													
60	Inosine	1.00^{bc}	0.97^{c}	1.06^{a}	0.76 ^{h}	0.79^{gh}	0.68^{i}	0.81 ^{fg}	0.84^{ef}	0.78^{gh}	0.86^{de}	1.02^{ab}	0.89^{d}

(*continued on next page*)

Table 1 (*continued*)

NO	Names	Fold change (11 yellow teas versus JSYZ)[#]											
		JSYZ	HSHY	MDHY	PYHT-B	YALY (b)	YALY (g)	JSMJ	WSHC	PYHT-L	MGHC	MDHC	HSHDC
61	1-(*sn-Glycero*-3-phoshpo)-1D-myo-inositol	1.00 ^{l}	3.31^{f}	2.69 ^{h}	4.21^{c}	6.18^{b}	1.46^{j}	3.84^{d}	7.55^{a}	3.55^{e}	3.01 ^{g}	1.13 ^{k}	2.50^{i}
62	Uridine 2′-phosphate	1.00^{e}	1.03^{e}	1.21^{c}	1.37^{a}	0.81^{f}	0.66 ^{h}	0.44^{j}	0.53^{i}	1.09^{d}	0.77 ^{g}	0.77 ^{g}	1.28^{b}
63	3′-UMP	1.00^{d}	0.77^{e}	1.23^{b}	1.67^{a}	0.76^{e}	0.53 ^{g}	0.34^{i}	0.44 ^{h}	1.15^{c}	0.66^{f}	0.67^{f}	1.01^{d}
Carbohydrates													
64	Maltose	1.00 ^{h}	1.19 ^{g}	1.91^{d}	4.58^{a}	1.53^{f}	1.74^{e}	0.87^{i}	2.39^{b}	2.41^{b}	1.55^{f}	1.20 ^{g}	2.07^{c}
65	Maltotriose	1.00 ^{g}	1.17^{f}	1.23^{e}	2.69^{a}	1.22^{ef}	0.07 ^{h}	1.00 ^{g}	1.26^{e}	1.99^{b}	1.53^{d}	1.56^{d}	1.67^{c}
66	Gluconic acid	1.00^{c}	1.00^{c}	0.72^{f}	0.54 ^{g}	1.13^{b}	1.25^{a}	0.88^{e}	0.92^{d}	0.71^{f}	0.72^{f}	0.99^{c}	0.86^{e}
67	Ribonic acid	1.00^{a}	0.65^{f}	0.95^{b}	0.82^{d}	0.67^{ef}	0.60 ^{g}	0.57 ^{h}	0.82^{d}	0.88^{c}	0.60 ^{g}	0.82^{d}	0.68^{e}
68	Glucose	1.00^{b}	0.74^{f}	0.66^{i}	0.71 ^{g}	0.77^{e}	1.04^{a}	0.81^{d}	0.79^{d}	0.97^{c}	0.69 ^{h}	0.81^{d}	0.50^{j}
69	Phloroglucinol	1.00 ^{g}	0.66^{i}	2.16^{c}	0.97 ^{g}	1.13^{f}	0.33 ^{k}	0.77 ^{h}	0.52^{j}	1.38^{e}	1.49^{d}	3.95^{b}	6.97^{a}

Data were assessed by Duncan's multiple range test; The values labeled with different letters (a–l) in the same row were significantly different ($p < 0.05$); [#] the full names of 12 yellow teas were as follows: JSYZ, Junshan Yinzhen; HSHY, Huoshan Huangya; MDHY, Mengding Huangya; PYHT-B, Pingyang Huangtang (yellow bud tea); YALY(b), Yuan'an Luyuan (bar shape); YALY(g), Yuan'an Luyuan (grain shape); JSMJ, Junshan Maojian; WSHC, Weishan Huangcha; PYHT-L, Pingyang Huangtang (yellow little tea); MGHC, Mogan Huangcha; MDHC, Mengding Huangcha; HSHDC, Huoshan Huangdacha; [†] Relative fold of the compounds was calculated according to the data of HPLC-UV.

the reference standard to compare the metabolite abundance in various teas. These 69 metabolites were composed of 9 monomeric catechin derivatives, 14 polymerized catechin derivatives, 16 phenolic acids, 13 flavonols, flavones, and their glycosides, 4 amino acids, 6 carbohydrates, 4 nucleosides, and 3 organic acids. An Upset plot was constructed to identify the common and unique metabolites present in these 12 representative yellow teas (Fig. 2B). The numbers of identified chemical compounds in JSYZ (66), HSHY (64), MDHY (63), PYHT-B (66), YALY (b) (64), YALY(g) (63), JSMJ (61), WSHC (64), PYHT-L (65), MGHC (62), MDHC (61), and HSHDC (65) were similar. However, the sample of yellow big tea was completely distinguishable from the samples of yellow bud teas and yellow little teas in the PCA score plot, while the samples of yellow bud tea were not entirely separated from the samples of yellow little teas (Fig. 2D).

Heat map was further applied to visualize the changes of major metabolites in these 12 yellow teas (Fig. 2C). As showed in Fig. 2C and Table 1, yellow big tea (HSHDC) had higher content of monomeric catechin derivatives than yellow bud teas and yellow little teas, except for EGCG, GCG, and gallocatechin-glucoside isomer 1. Additionally, signal intensities of dehydrotheasinensin C isomers 1 and 2 in yellow big tea were much greater (>three times) than those in yellow bud teas and yellow little teas. In contrast, the majority of yellow bud teas and yellow little teas had higher content of phenolic acids compared to yellow big tea, with the exception of quinic acid, 3-*O*-caffeoylquinic acid, and 3-*p*-coumaroylquinic acid. Moreover, yellow big tea had lower levels of flavonols, flavones, and their glycosides than yellow bud teas and yellow little teas, except for schaftoside, myricitin 3-*O*-galactoside, kaempferol 3-*O*-glucosyl-rutinoside, rutin, and kaempferol. Thus, yellow big tea differs greatly from yellow bud teas and yellow little teas in the aspect of metabolite content, rather than metabolite composition. Nevertheless, yellow bud teas cannot be effectively distinguished from yellow little teas based on the above identified metabolites.

3.2. The analysis of major chemical constituents in yellow teas by targeted absolute quantification

The main chemical constituents of 12 representative yellow teas, including polyphenols, catechins, flavonoids, alkaloids, amino acids, and soluble sugars, were measured by targeted absolute quantification. As reflected in Table 2, the content of polyphenols in yellow bud teas (108.76 ~ 127.36 mg/g) and yellow little teas (104.78 ~ 156.17 mg/g) was much higher than that in yellow big tea (HSHDC, 77.41 mg/g). Nonetheless, the content of polyphenols in yellow bud teas and yellow little teas from different origin varied greatly. For example, the polyphenol content of Zhejiang- and Sichuan-produced yellow little teas (PYHT-L, 156.17 mg/g; MGHC, 121.40 mg/g; MDHC, 124.13 mg/g) was higher than that of Zhejiang- and Sichuan-produced yellow bud teas (PYHT-B, 119.92 mg/g; MDHY, 113.76 mg/g); but the polyphenol content of Hunan-produced yellow bud teas (JSYZ, 127.36 mg/g) was higher than that of Hunan-produced yellow little teas (JSMJ, 104.78 mg/g; WSHC, 117.26 mg/g). Tea catechins, which are also known as flavan-3-ols and account for 60 ~ 80 % of tea polyphenols, are the most abundant compounds in tea (Zhu, et al., 2021b). Further, EGCG is one of the main catechins accounting for 6 ~ 8 % of dry weight of tea leaves (Zhu, et al., 2021a). Unlike polyphenol content, the total catechin content of yellow bud teas (39.10 ~ 66.88 mg/g) and yellow little teas (40.52 ~ 75.77 mg/g) was not necessarily higher than that of yellow big tea (HSHDC, 50.82 mg/g), which was consistent with the results of EGCG content. Nevertheless, the content of gallic acid in yellow big tea (HSHDC, 1.22 mg/g) was much lower than that in yellow bud teas (3.32 ~ 5.18 mg/g) and yellow little teas (3.08 ~ 6.57 mg/g).

The flavonoid content of yellow big tea (6.51 mg/g) was obviously higher than that of yellow bud teas (0.75 ~ 3.77 mg/g) and yellow little teas (1.06 ~ 5.85 mg/g). Caffeine is the major alkaloid accounting for 2 ~ 4 % of dry weight in tea (Zhu, et al., 2021a). The caffeine content (24.16 ~ 37.67 mg/g) varies less than polyphenol content in these 12 yellow tea products. Amino acids are the key contributor to fresh taste of tea infusion (Zhu, et al., 2019). The total amino acid content of yellow big tea (HSHDC, 18.10 mg/g) was lower than that of yellow bud teas and yellow little teas except for YALY(b), WSHC, and PYHT-L. But the theanine content of yellow big tea (HSHDC, 9.46 mg/g) was in the middle level of 12 yellow tea products (3.67 ~ 16.32 mg/g). In addition, soluble sugars are also the important contributor to the taste of tea infusion (Zhu, et al., 2020a). Notably, the soluble sugar content of yellow big tea (HSHDC, 79.79 mg/g) was higher than that of yellow bud teas (48.62 ~ 79.31 mg/g) and yellow little teas (51.82 ~ 71.51 mg/g), but yellow bud teas could not be effectively distinguished from yellow little teas based on soluble sugar content. Thus, the major chemical constituents in yellow big tea were greatly different from that in yellow bud teas and yellow little teas, but the yellow bud teas could not be entirely separated from the yellow little teas according to these major chemical constituents.

3.3. Electronic sensory evaluation

E-nose, E-tongue, and E-eye were used to evaluate the aroma, infusion taste, and infusion color of the 12 yellow teas, respectively. Significant variations were observed between yellow big tea and other 11 yellow teas by E-nose (Fig. 3A). The yellow big tea (HSHDC) had the strongest bitterness and aftertaste-bitterness, but the weakest astringency and aftertaste-astringency in 12 yellow teas. PYHT-B showed the strongest sweetness and astringency, but the weakest saltiness and

Table 2
The analysis of major chemical constituents in 12 yellow teas by targeted absolute quantification.

Chemical constituents (mg/g)	JSYZ	HSHY	MDHY	PYHT-B	YALY(b)	YALY(g)	JSMJ	WSHC	PYHT-L	MGHC	MDHC	HSHDC
						Chemical composition						
Polyphenols	127.36 ± 0.06^{c}	108.76 ± 0.06^{i}	113.76 ± 0.06^{g}	119.92 ± 0.47^{e}	136.34 ± 0.00^{b}	127.47 ± 0.47^{c}	104.78 ± 0.47^{j}	117.26 ± 0.06^{f}	156.17 ± 0.06^{a}	109.40 ± 0.47^{h}	124.13 ± 0.12^{d}	77.41 ± 0.49^{k}
Flavonoids	3.77 ± 0.00^{c}	3.85 ± 0.24^{c}	2.26 ± 0.23^{e}	0.75 ± 0.23^{e}	2.23 ± 0.29^{e}	5.85 ± 0.50^{b}	2.84 ± 0.28^{d}	1.06 ± 0.17^{c}	3.96 ± 0.23^{d}	2.87 ± 0.29^{d}	5.54 ± 0.23^{b}	6.52 ± 0.36^{a}
Soluble sugars	48.62 ± 0.28^{i}	55.15 ± 0.44^{g}	58.55 ± 0.16^{f}	79.31 ± 0.48^{b}	57.03 ± 0.28^{f}	64.70 ± 0.16^{e}	53.98 ± 1.70^{g}	71.51 ± 0.00^{c}	64.66 ± 0.16^{e}	69.00 ± 0.32^{d}	51.82 ± 3.26^{h}	79.79 ± 0.29^{a}
						Major catechins, gallic acid, caffeine and alkaloids						
EGCG	33.02 ± 0.15^{d}	19.00 ± 0.26^{j}	20.74 ± 0.63^{i}	39.18 ± 0.05^{c}	46.88 ± 0.15^{a}	30.69 ± 0.09^{e}	40.16 ± 0.08^{b}	39.35 ± 0.08^{c}	21.12 ± 0.07^{i}	25.75 ± 0.33^{g}	22.90 ± 0.22^{h}	29.27 ± 0.47^{f}
EGC	3.13 ± 0.12^{c}	2.72 ± 0.03^{d}	2.43 ± 0.05^{e}	2.61 ± 0.02^{d}	1.76 ± 0.03^{g}	1.52 ± 0.04^{h}	1.84 ± 0.02^{g}	2.21 ± 0.04^{f}	6.09 ± 0.09^{b}	2.36 ± 0.03^{ef}	0.39 ± 0.03^{i}	7.42 ± 0.03^{a}
ECG	16.47 ± 0.09^{a}	10.86 ± 0.09^{g}	9.59 ± 0.04^{h}	14.98 ± 0.06^{c}	15.53 ± 0.31^{b}	11.89 ± 0.04^{f}	16.41 ± 0.16^{a}	12.49 ± 0.43^{e}	4.65 ± 0.06^{j}	11.84 ± 0.04^{f}	13.72 ± 0.03^{d}	7.07 ± 0.15^{i}
GCG	5.93 ± 0.05^{d}	2.74 ± 0.06^{k}	2.94 ± 0.03^{j}	5.29 ± 0.07^{e}	7.18 ± 0.05^{b}	7.12 ± 0.07^{b}	8.40 ± 0.08^{a}	5.15 ± 0.08^{f}	3.87 ± 0.07^{h}	4.01 ± 0.04^{g}	3.24 ± 0.05^{i}	6.08 ± 0.08^{c}
EC	3.70 ± 0.05^{b}	2.44 ± 0.08^{hi}	3.59 ± 0.04^{c}	2.74 ± 0.05^{e}	2.48 ± 0.03^{gh}	2.60 ± 0.02^{f}	2.45 ± 0.04^{ghi}	2.53 ± 0.05^{fg}	2.38 ± 0.03^{i}	3.19 ± 0.03^{d}	2.25 ± 0.07^{j}	3.86 ± 0.07^{a}
Catechin	2.18 ± 0.09^{c}	1.34 ± 0.04^{i}	2.41 ± 0.07^{b}	2.08 ± 0.05^{d}	1.94 ± 0.02^{e}	1.61 ± 0.04^{g}	1.47 ± 0.05^{h}	1.75 ± 0.04^{f}	2.41 ± 0.03^{b}	1.91 ± 0.05^{e}	1.52 ± 0.03^{h}	3.32 ± 0.03^{a}
Total catechins[a]	64.42 ± 0.03^{d}	39.10 ± 0.42^{l}	41.70 ± 0.65^{j}	66.89 ± 0.24^{c}	75.79 ± 0.19^{a}	55.44 ± 0.03^{g}	70.73 ± 0.29^{b}	63.48 ± 0.44^{e}	40.53 ± 0.21^{k}	49.06 ± 0.35^{h}	44.02 ± 0.13^{i}	56.99 ± 0.84^{f}
Gallic acid	3.92 ± 0.06^{f}	5.18 ± 0.04^{d}	3.73 ± 0.06^{g}	3.32 ± 0.05^{h}	3.83 ± 0.03^{fg}	3.08 ± 0.07^{i}	3.33 ± 0.04^{h}	5.82 ± 0.10^{c}	6.44 ± 0.05^{b}	4.63 ± 0.04^{e}	6.57 ± 0.12^{a}	1.22 ± 0.26^{j}
Caffeine	24.16 ± 0.07^{h}	27.33 ± 0.38^{f}	32.24 ± 0.09^{c}	29.12 ± 0.03^{e}	32.92 ± 0.04^{b}	37.67 ± 0.67^{a}	28.83 ± 0.12^{e}	32.17 ± 0.16^{c}	26.13 ± 0.32^{g}	32.12 ± 0.07^{c}	32.54 ± 0.06^{bc}	29.87 ± 0.05^{d}
Theobromine	1.04 ± 0.07^{j}	2.11 ± 0.04^{h}	0.80 ± 0.05^{k}	5.25 ± 0.05^{c}	11.96 ± 0.07^{a}	4.60 ± 0.06^{d}	6.89 ± 0.04^{b}	4.31 ± 0.10^{e}	1.06 ± 0.06^{j}	1.62 ± 0.03^{i}	3.03 ± 0.08^{f}	2.86 ± 0.03^{g}
Theophylline	0.37 ± 0.03^{b}	0.28 ± 0.01^{d}	0.32 ± 0.03^{cd}	0.23 ± 0.02^{e}	0.15 ± 0.03^{g}	0.22 ± 0.03^{ef}	0.22 ± 0.04^{ef}	0.18 ± 0.01^{fg}	0.04 ± 0.01^{h}	0.33 ± 0.01^{c}	0.46 ± 0.03^{a}	0.06 ± 0.01^{h}
						Major amino acids						
Aspartic acid	1.83 ± 0.06^{h}	2.21 ± 0.03^{e}	3.81 ± 0.03^{b}	1.89 ± 0.02^{g}	1.11 ± 0.03^{k}	4.23 ± 0.02^{a}	2.05 ± 0.02^{f}	1.34 ± 0.02^{j}	1.57 ± 0.01^{j}	2.76 ± 0.01^{c}	2.54 ± 0.02^{d}	1.87 ± 0.02^{gh}
Serine	2.52 ± 0.08^{c}	0.94 ± 0.03^{f}	0.92 ± 0.03^{f}	0.82 ± 0.03^{g}	0.37 ± 0.02^{j}	4.26 ± 0.02^{a}	0.73 ± 0.02^{h}	1.14 ± 0.02^{e}	0.66 ± 0.02^{i}	1.26 ± 0.01^{d}	2.82 ± 0.03^{b}	0.78 ± 0.01^{gh}
Glutamate	2.20 ± 0.04^{g}	2.31 ± 0.03^{f}	3.08 ± 0.04^{c}	1.84 ± 0.02^{h}	1.62 ± 0.03^{i}	3.95 ± 0.02^{b}	2.94 ± 0.01^{d}	1.49 ± 0.02^{j}	0.51 ± 0.01^{k}	5.17 ± 0.02^{a}	2.70 ± 0.01^{e}	1.64 ± 0.02^{i}
Glycine	0.02 ± 0.00^{c}	0.04 ± 0.01^{a}	0.02 ± 0.00^{c}	0.01 ± 0.00^{d}	0.01 ± 0.00^{d}	0.04 ± 0.00^{a}	0.02 ± 0.00^{c}	0.00 ± 0.00^{e}	0.04 ± 0.01^{a}	0.03 ± 0.00^{b}	0.04 ± 0.00^{a}	0.02 ± 0.00^{c}
Histidine	1.15 ± 0.03^{d}	0.44 ± 0.04^{i}	0.75 ± 0.03^{f}	1.14 ± 0.05^{d}	0.82 ± 0.02^{e}	1.25 ± 0.01^{c}	1.34 ± 0.04^{b}	0.53 ± 0.01^{h}	0.22 ± 0.02^{j}	0.63 ± 0.02^{g}	0.74 ± 0.01^{f}	1.45 ± 0.02^{a}
Arginine	5.56 ± 0.02^{a}	1.25 ± 0.06^{e}	1.07 ± 0.06^{g}	1.55 ± 0.03^{d}	0.50 ± 0.02^{j}	2.24 ± 0.01^{b}	1.18 ± 0.04^{f}	0.82 ± 0.03^{h}	0.54 ± 0.02^{j}	1.57 ± 0.02^{d}	2.04 ± 0.04^{c}	0.61 ± 0.01^{i}
Threonine	0.83 ± 0.04^{a}	0.53 ± 0.03^{d}	0.53 ± 0.04^{d}	0.52 ± 0.02^{d}	0.42 ± 0.03^{e}	0.70 ± 0.01^{b}	0.45 ± 0.03^{e}	0.42 ± 0.02^{e}	0.16 ± 0.01^{g}	0.62 ± 0.01^{c}	0.80 ± 0.01^{a}	0.24 ± 0.01^{f}
Alanine	0.53 ± 0.01^{a}	0.25 ± 0.01^{f}	0.31 ± 0.02^{d}	0.18 ± 0.01^{i}	0.14 ± 0.01^{b}	0.46 ± 0.01^{h}	0.18 ± 0.01^{i}	0.12 ± 0.02^{i}	0.28 ± 0.02^{e}	0.38 ± 0.01^{c}	0.28 ± 0.01^{e}	0.20 ± 0.01^{g}
Proline	0.74 ± 0.02^{a}	0.37 ± 0.02^{c}	0.32 ± 0.03^{d}	0.23 ± 0.01^{e}	0.14 ± 0.02^{f}	0.73 ± 0.01^{a}	0.25 ± 0.01^{e}	0.22 ± 0.01^{e}	0.17 ± 0.01^{f}	0.37 ± 0.02^{c}	0.65 ± 0.04^{b}	0.22 ± 0.01^{e}
Theanine	16.32 ± 0.04^{b}	9.44 ± 0.03^{h}	11.13 ± 0.08^{e}	9.68 ± 0.01^{g}	7.13 ± 0.02^{i}	17.14 ± 0.04^{a}	10.10 ± 0.05^{f}	6.21 ± 0.02^{j}	3.67 ± 0.02^{k}	11.54 ± 0.01^{d}	12.11 ± 0.03^{c}	9.46 ± 0.02^{h}
Cysteine	0.17 ± 0.02^{b}	0.09 ± 0.01^{f}	0.06 ± 0.01^{g}	0.12 ± 0.02^{cd}	0.06 ± 0.00^{g}	0.13 ± 0.01^{c}	0.11 ± 0.01^{de}	0.09 ± 0.00^{f}	0.06 ± 0.00^{g}	0.11 ± 0.01^{de}	0.19 ± 0.01^{a}	0.07 ± 0.05^{ef}
Tyrosine	0.26 ± 0.02^{a}	0.14 ± 0.01^{cde}	0.13 ± 0.02^{e}	0.15 ± 0.01^{cd}	0.09 ± 0.01^{f}	0.19 ± 0.01^{b}	0.16 ± 0.01^{c}	0.13 ± 0.01^{de}	0.09 ± 0.00^{f}	0.16 ± 0.01^{c}	0.27 ± 0.02^{a}	0.16 ± 0.01^{c}
Valine	0.13 ± 0.02^{e}	0.06 ± 0.01^{g}	0.08 ± 0.01^{fg}	0.03 ± 0.00^{h}	0.49 ± 0.02^{c}	1.27 ± 0.01^{a}	0.03 ± 0.00^{h}	0.04 ± 0.01^{h}	0.04 ± 0.00^{h}	0.09 ± 0.00^{f}	0.23 ± 0.02^{d}	0.58 ± 0.02^{b}
Methionine	0.02 ± 0.00^{b}	0.01 ± 0.00^{c}	0.01 ± 0.00^{c}	0.01 ± 0.00^{c}	0.01 ± 0.01^{c}	0.02 ± 0.00^{b}	0.02 ± 0.00^{b}	0.01 ± 0.00^{c}	0.07 ± 0.00^{a}	0.02 ± 0.00^{b}	0.01 ± 0.00^{c}	0.01 ± 0.01^{c}
Lysine	0.64 ± 0.03^{b}	0.23 ± 0.01^{g}	0.37 ± 0.02^{e}	0.32 ± 0.02^{f}	0.06 ± 0.00^{i}	1.04 ± 0.01^{a}	0.32 ± 0.02^{f}	0.19 ± 0.02^{h}	0.31 ± 0.01^{f}	0.49 ± 0.02^{d}	0.61 ± 0.01^{c}	0.19 ± 0.01^{h}
Isoleucine	0.31 ± 0.02^{b}	0.16 ± 0.02^{d}	0.27 ± 0.02^{c}	0.14 ± 0.01^{de}	0.06 ± 0.00^{f}	0.53 ± 0.02^{a}	0.10 ± 0.07^{ef}	0.18 ± 0.01^{d}	0.09 ± 0.00^{f}	0.33 ± 0.02^{b}	0.51 ± 0.01^{a}	0.14 ± 0.01^{de}
Leucine	0.46 ± 0.04^{c}	0.33 ± 0.02^{e}	0.43 ± 0.02^{d}	0.33 ± 0.02^{e}	0.11 ± 0.01^{h}	0.92 ± 0.03^{a}	0.34 ± 0.02^{e}	0.23 ± 0.01^{f}	0.09 ± 0.00^{h}	0.41 ± 0.01^{d}	0.77 ± 0.02^{b}	0.17 ± 0.01^{g}
Phenylalanine	0.81 ± 0.03^{b}	0.35 ± 0.01^{e}	0.25 ± 0.02^{g}	0.44 ± 0.04^{d}	0.15 ± 0.02^{h}	0.65 ± 0.02^{c}	0.42 ± 0.01^{d}	0.44 ± 0.02^{d}	0.18 ± 0.01^{h}	0.35 ± 0.01^{e}	0.97 ± 0.02^{a}	0.31 ± 0.01^{f}
Total amino acids[b]	34.51 ± 0.19^{b}	19.17 ± 0.11^{h}	23.54 ± 0.21^{e}	19.40 ± 0.04^{g}	13.32 ± 0.08^{k}	39.76 ± 0.05^{a}	20.74 ± 0.22^{f}	13.62 ± 0.08^{j}	8.74 ± 0.04^{l}	26.28 ± 0.09^{d}	28.27 ± 0.02^{c}	18.10 ± 0.06^{i}

Data were assessed by Duncan's multiple range test; The values labeled with different letters (a–l) in the same row were significantly different ($p < 0.05$); the full names of 12 yellow teas were as follows: JSYZ, Junshan Yinzhen; HSHY, Huoshan Huangya; MDHY, Mengding Huangya; PYHT-B, Pingyang Huangtang (yellow bud tea); YALY(b), Yuan'an Luyuan (bar shape); YALY(g), Yuan'an Luyuan (grain shape); JSMJ, Junshan Maojian; WSHC, Weishan Huangcha; PYHT-L, Pingyang Huangtang (yellow little tea); MGHC, Mogan Huangcha; MDHC, Mengding Huangcha; HSHDC, Huoshan Huangdacha.

[a] Total catechins derivatives included EGCG, ECG, EGC, GCG, EC, and catechin. [b] Total amino acids included aspartic acid, serine, glutamate, glycine, histidine, arginine, threonine, alanine, proline, theanine, cysteine, tyrosine, valine, methionine, lysine, isoleucine, leucine, and phenylalanine. Abbreviation: EGCG, epigallocatechin-3-*O*-gallate; ECG, epicatechin-3-*O*-gallate; EGC, epigallocatechin; GCG, gallocatechin-3-*O*-gallate; EC, epicatechin.

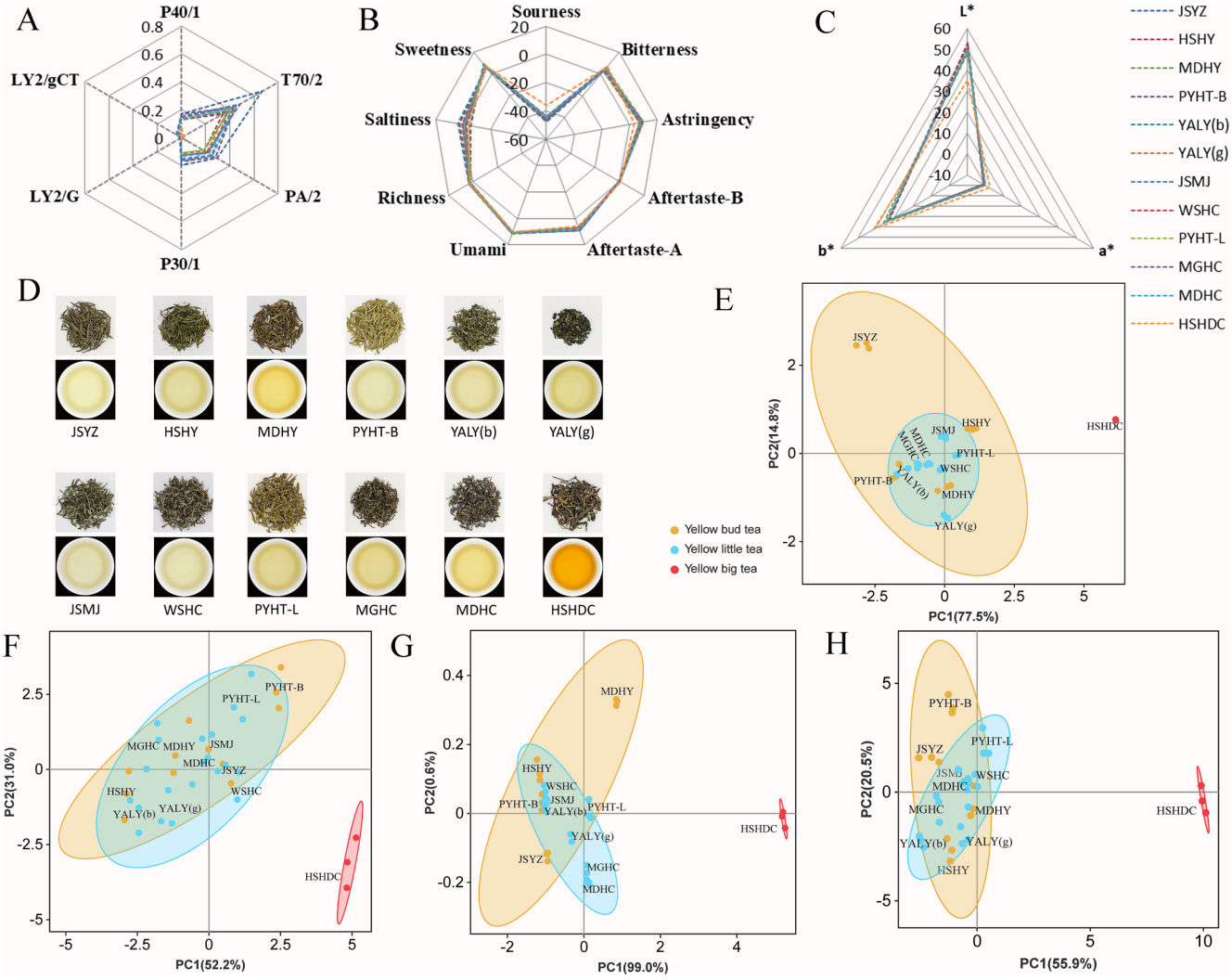

Fig. 3. Sensory evaluation of 12 representative yellow teas: (A), (B), and (C) were the spider plots of E-nose, E-tongue and E-eye, respectively; (D) the appearance and infusion of yellow tea samples; Score plots of principal component analysis based on (E) E-nose signals ($R^2X = 0.868$, $Q^2 = 0.677$), (F) E-tongue signals ($R^2X = 0.832$, $Q^2 = 0.645$), (G) E-eye signals ($R^2X = 0.78$, $Q^2 = 0.457$), and (H) fusion signals ($R^2X = 0.992$, $Q^2 = 0.893$). Abbreviation: aftertaste-A, aftertaste-astringency; aftertaste-B, aftertaste-bitterness. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

richness in 12 yellow teas. However, no statistical difference existed in 12 yellow teas in terms of umami (Fig. 3B). In addition, the noticeable distinction existed in the infusion color between yellow big tea and other 11 yellow teas according to the E-eye evaluation (Fig. 3C). The infusion of yellow big tea also exhibited the deepest hue of tea infusion by naked-eye assessment (Fig. 3D).

Furthermore, PCA score plots were used to reveal the electronic sensory evaluations of 12 yellow teas based on individual E-nose, E-tongue, E-eye signal, and fusion signals (Fig. 3E-H). As depicted in Fig. 3E, yellow big tea exhibited a clear distinction from the other 11 teas. Further, the within-class distances of other 11 yellow teas were large (Fig. 3E). The E-tongue signal and E-eye signal of yellow big tea was also quite different from those of other 11 yellow teas (Fig. 3F-G), but the within-class distances of other yellow 11 teas were small in the PCA score plot of E-tongue signal (Fig. 3F). As expected, the yellow big tea also exhibited a clear distinction from the other 11 yellow teas on the fusion signals of PCA score plot (Fig. 3H). Taken together, yellow big tea could be completely distinguished from the other 12 yellow teas, indicating that the sensory quality of yellow big tea is quite different from that of yellow bud tea and yellow little tea.

3.4. Correlations between chemical constituents and electronic sensory characteristics in yellow teas

The correlations between chemical constituents and electronic sensory characteristics in yellow teas were visualized in Fig. 4A. Further, a O2PLS model, which was based on the variations in tastes and metabolites among 12 yellow teas, was established to explore the taste-metabolite association (Fig. 4B). Moreover, a total of 28 metabolites were identified as critical taste-active metabolites based on correlation coefficients $|r| > 0.7$ and $p < 0.05$ (Fig. 4C). The bitterness and aftertaste-bitterness of yellow teas were positively related to gallic acid, 4′-dehydroxylated gallocatechin-3-*O*-gallate, dehydrotheasinensin C isomer 1, dehydrotheasinensin C isomer 2, myricitin 3-*O*-galactoside, and phloroglucinol. The aftertaste-astringency of yellow tea was positively associated with methyl gallate, 1,5-digalloylglucose, and 2,6-

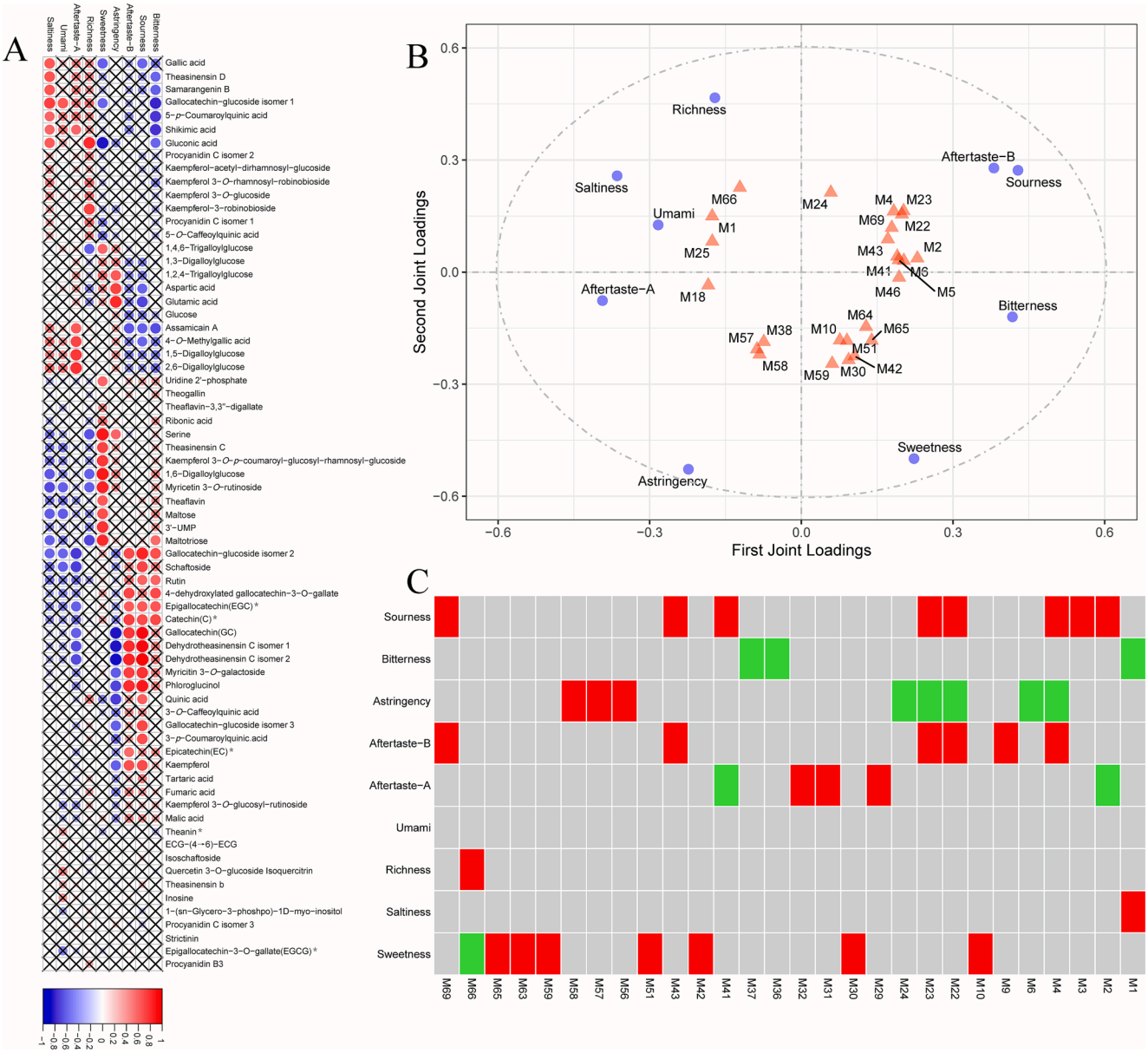

Fig. 4. Taste-metabolite association in 12 representative yellow teas. (A) Correlation coefficient between tastes and metabolites. (B) O2PLS core plot ($R^2X = 0.985$, $R^2Y = 0.836$), the plot showed the top 25 metabolites that were highly correlated with tastes. (C) Heatmap representing Pearson correlation analysis between their taste scores and metabolites ($|r| > 0.7$, $p < 0.05$.). The red, grey, and green colors indicated the positive, no, and negative correlation, respectively. The metabolite ID correspond to that used in Table 1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

digalloylglucose. The sweetness was mainly positively associated with maltotriose. Yellow big tea had the high levels of bitterness and aftertaste-bitterness-related compounds, which led to the strongest bitterness and the weakest astringency in yellow big tea in comparison with other 11 yellow teas. PYHT-B had high content of sweetness-associated compound, which contributed to the strongest sweet taste in yellow tea in comparison with other 11 yellow teas.

4. Discussion

Yellow tea has gained increasing popularity in recent years because of its unique flavor and multiple health benefits. Previous studies have investigated the impact of yellowing processes on chemical composition and sensory qualities of yellow tea (Fan, et al., 2022; Wei, et al., 2021). However, It is also well worth studying the differences in chemical composition and sensory quality of different kinds of yellow tea. The chemical composition and sensory qualities of yellow teas vary greatly, because of the difference in the tea varieties, raw material grade and origin, and processing details. Thus, the chemical composition and sensory qualities of 12 representative yellow teas were analyzed by untargeted and targeted metabolomics and electronic sensory technologies; the correlations between chemical constituents and electronic sensory characteristics in yellow teas were also analyzed.

Yellow tea can be classified into yellow bud tea, yellow little tea, and yellow big tea according the raw material grade. Yellow bud tea is produced by buds or one bud and one leaf; yellow little tea is generally produced by one bud and one or two leaves; yellow big tea is produced by using old leaves and stems (Xu, et al., 2018). We found that the yellow big tea (HSHDC) had the lowest amount of tea polyphenols in these three types of yellow tea. Previous studies have proved that the old leaves and stems of tea had a higher level of tea polyphenols compared with tender tea leaves, which was the main reason of low tea polyphenol

content in yellow big tea (Wan, 2008). Catechins, flavones, and flavonols are crucial contributes to the bitterness and astringency of tea (Zhang, et al., 2020). These components can be degraded, oxidized, and hydrolyzed at longtime yellowing process, which lead the decrease in the intensities of bitterness and astringency (Fan, et al., 2016; Xu, et al., 2018; Zhu, et al., 2021). For instance, the polymeric catechins can be transformed into monomer catechin derivatives during the yellowing process (Fan, et al., 2019; Wei, et al., 2021). Yellow bud tea usually has the longest duration of yellowing in these three types of yellow tea, which lead the maximum intensity conversion of polymeric catechins. This may be one of the main reasons that yellow bud tea had the lowest level of polymerized catechin derivatives but the highest content of monomeric catechin derivatives. The amino acids can be also transformed during yellow tea processing, such as decarboxylation, oxidation, and thermal degradation (Kausar, et al., 2013; Qu, et al., 2019). For instance, the aroma component indole can be formed by the degradation of tryptophan (Shahidi, et al., 1997). Thus, the significant variations in aroma between yellow big tea and other 11 yellow teas might be related to the conversion degree of amino acids. However, the yellow bud teas could not be entirely separated from the yellow little teas according to identified compounds. In addition, the sensory evaluations of yellow tea were mainly carried out by manual evaluation in previous studies (Fan, et al., 2022; Wang, et al., 2021; Wei, et al., 2020). The electronic sensory methods were employed in this study to obtain a more objective assessment of 12 yellow teas. We found that the aroma, infusion taste, and infusion color in yellow big tea differed greatly from that in yellow bud tea and yellow little tea by the comprehensive analysis of E-nose, E-tongue, and E-eye. Previous studies have showed that prolonged yellowing aggravated the degradation of pigment, which affected the leaf color and infusion color of yellow tea (Feng, et al., 2023). Wei et al. (2021) found that the levels of catechins, amino acids, phenolic acids, and glycosidically bound volatiles were significantly decreased as the yellowing time prolonged. Thus, the distinctive sensory phenotype of yellow big tea might be attributed to its longtime yellowing process and low grade of raw materials (Yang, et al., 2013).

Previous studies on black tea, green tea, oolong tea, and white tea have proved that polyphenols, catechins, flavonoids, alkaloids, amino acids, and soluble sugars were closely related to the tea taste (Zhang, et al., 2020). In this study, we found that the polyphenols (e.g., EGCG and EGC) and flavonoids (e.g., rutin) had a positive correlation with the bitterness and aftertaste-bitterness in 12 yellow teas according to the correlation evaluation between chemical constituents and sensory characteristics. Wang et al. (2021) found that rutin was positively correlated with bitter and astringency, and simultaneously reduced the sweet and umami strength in yellow tea. Yellow big tea had the highest amount of rutin, which might be the one of the main reasons for its strongest bitterness and aftertaste among these 12 yellow teas. In addition, previous reports have proved that theanine, glutamic acid, glycine, and alanine synergistically enhanced the umami of tea infusion; the total free amino acid content had a significant positive correlation with sweet of tea infusion (Qi, 2016). We found that the sweetness in 12 yellow teas is mainly positively correlated with amino acids and soluble sugars. The weak tastes of umami and sweetness in yellow big tea might be attributed to its low level of amino acids and soluble sugars. In summary, the chemical constituents and sensory characteristics in yellow big tea were greatly different from that in yellow bud teas and yellow little teas, but the yellow bud teas could not be entirely separated from the yellow little teas according to these chemical constituents and sensory characteristics.

5. Conclusion

In this study, 12 representative yellow teas, which were basically covered the main products of yellow tea, were chosen in this study. The combined analysis of non-targeted/targeted metabolomics and electronic sensor technologies (E-eye, E-nose, and E-tongue,) was applied to reveal the chemical and sensor variation in 12 yellow teas for the first time. The results showed that yellow big tea differed greatly from yellow bud teas and yellow little teas, but yellow bud teas could not be effectively distinguished from yellow little teas based on chemical constituents and electronic sensory characteristics. The sensor variation in 12 yellow teas might be attributed to some compounds related to bitterness and aftertaste-bitterness (4′-dehydroxylated gallocatechin-3-*O*-gallate, dehydrotheasinensin C isomers 1 and 2, myricitin 3-*O*-galactoside, and phloroglucinol), aftertaste-astringency (methyl gallate, 1,5-digalloylglucose, and 2,6-digalloylglucose), and sweetness (maltotriose). This study provided a comprehensive understanding of yellow tea on chemical composition and sensory quality.

CRediT authorship contribution statement

Yuan Li: Data curation, Formal analysis, Software, Writing – original draft. **Yilong Li:** Data curation, Software. **Tian Xiao:** Data curation, Software. **Huimin Jia:** Data curation, Formal analysis. **Yu Xiao:** Data curation. **Zhonghua Liu:** Methodology, Project administration, Writing – review & editing. **Kunbo Wang:** Methodology, Project administration, Writing – review & editing. **Mingzhi Zhu:** Project administration, Writing – original draft, Writing – review & editing.

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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