

# Emerging techniques for determining the quality and safety of tea products: A review

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## Abstract

Spectroscopic techniques, electrochemical methods, nanozymes, computer vision, and modified chromatographic techniques are the emerging techniques for determining the quality and safety parameters (e.g., physical, chemical, microbiological, and classified parameters, as well as inorganic and organic contaminants) of tea products (such as fresh tea leaves, commercial tea, tea beverage, tea powder, and tea bakery products) effectively. By simplifying the sample preparation, speeding up the detection process, reducing the interference of other substances contained in the sample, and improving the sensitivity and accuracy of the current standard techniques, the abovementioned emerging techniques achieve rapid, cost-effective, and nondestructive or slightly destructive determination of tea products, with some of them providing real-time detection results. Applying these emerging techniques in the whole industry of tea product processing, right from the picking of fresh tea leaves, fermentation of tea leaves, to the sensory evaluation of commercial tea, as well as developing portable devices for real-time and on-site determination of classified and safety parameters (e.g., the geographical origin, grade, and content of contaminants) will not only eliminate the strong dependence on professionals but also help mechanize the production of tea products, which deserves further research. Conducting a review on the application of spectroscopic techniques, electrochemical methods, nanozymes, computer vision, and modifications of chromatographic techniques for quality and safety determination of tea products may serve as guide for other types of foods and beverages, offering potential techniques for their detection and evaluation, which would promote the development of the food industry.

## KEYWORDS

tea products, spectroscopic techniques, electrochemical methods, nanozymes, computer vision, chromatographic techniques

## 1 | INTRODUCTION

Originating from China, tea (*Camellia sinensis* (L.) O. Kuntze) is one of the most important economic crops in many countries worldwide. Beginning with drinking tea

made from fresh tea leaves picked in spring, the range of tea products continues to expand; from fresh tea leaves to commercial tea (loose or bag), tea beverages (canned or bottled), tea powder, and tea bakery products (tea bread and cookies), tea products are widely found in the food

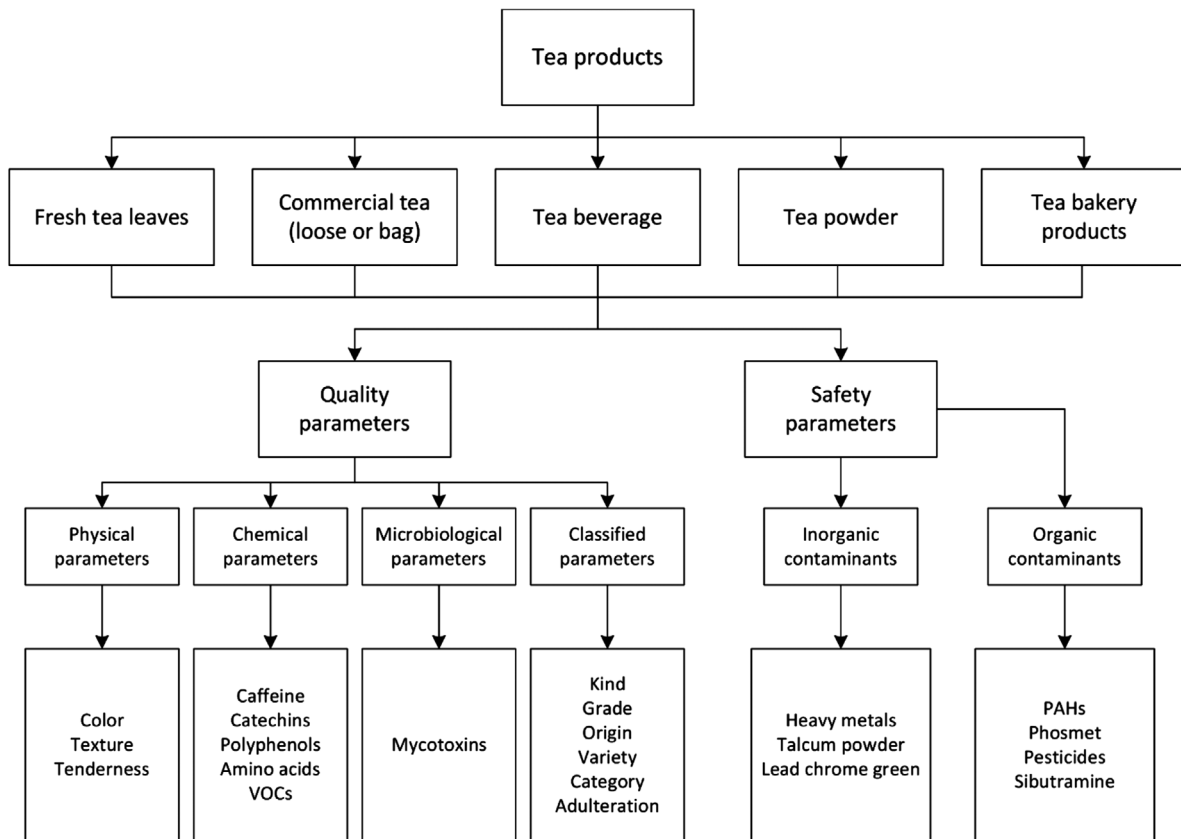


FIGURE 1 Quality and safety evaluations of tea products

industry and are very popular. Similar to other foods, quality and safety are two major aspects of tea products that have unique parameters, such as the tenderness, evenness, and neatness of fresh tea leaves in terms of physical parameters, catechins and volatile organic compounds (VOCs) in chemical parameters, grade, geographical origin, type, and category in classified parameters, lead chrome green in inorganic contaminants, and sibutramine in organic contaminants, as depicted in Figure 1. Most of the parameters listed in Figure 1 have standard detection methods; however, they are time-consuming, labor-intensive, and require operational skills, especially the sensory evaluation of tea, which needs to be performed by professionals who pass the evaluation qualification test and evaluate tea samples in a standard evaluation room. These restrictions have caused researchers to focus on emerging techniques to develop rapid, efficient, and on-the-spot methods for quality and safety determination of tea products.

Ultraviolet-visible spectroscopy (UV-vis) (Diniz, Barbosa, De Melo Milanez, Pistonesi, & De Araújo, 2016; Yu & He, 2018), near-infrared spectroscopy (Chen, Zhao, Zhang, & Wang, 2006, 2007; Zhao, Chen, Huang, & Fang, 2006), Fourier transform infrared (FT-IR) spectroscopy (Li, Sun, Luo, & He, 2015, 2016), Raman spectroscopy (Li et al.,

2015; Zhang et al., 2020), laser-induced breakdown spectroscopy (LIBS) (Yu, Peng, Liu, & He, 2017), and terahertz (Chen, Cao, & Liu, 2011) in spectroscopic techniques, electric nose (E-nose) (Chen, Liu, Zhao, & Ouyang, 2013; Dutta, Hines, Gardner, Kashwan, & Bhuyan, 2003), electric tongue (E-tongue) (Zhi, Zhao, & Zhang, 2017), electric eye (E-eye) (Xu, Wang, & Zhu, 2019), adsorptive stripping voltammetry (ASV) (Guo, Zheng, Mo, & Ye, 2009), cyclic voltammetry (CV) (Kilmartin & Hsu, 2003; Liu et al., 2014), square-wave voltammetry (SWV) (Novak, Šeruga, & Komorsky-Lovrić, 2010), and capillary zone electrophoresis (Horie, Mukai, & Kohata, 1997) in electrochemical methods, nanozymes (Wang, Liu, Qin, Chen, & Shen, 2016; Zhang & Huang, 2015), computer vision (Wang et al., 2015; Xu, Wang, & Gu, 2019; Ye et al., 2019), and modified chromatographic techniques are the five main emerging techniques for quality and safety evaluation of tea products, determining parameters qualitatively and quantitatively, with their distinct advantages and disadvantages, as well as suitable but different detection target. To the best of our knowledge, no study has reviewed the applications of these emerging techniques for the evaluation of different parameters in tea products. Therefore, it is necessary to present a comprehensive description of spectroscopic tech-

**TABLE 1** Current standard techniques for determining the quality and safety of tea products

Parameter	Standard technique	Standard
Moisture	Drying	(ISO 1573:1980 (R2015), 2015)
Crude fiber	Weende analysis	(ISO 15598:1999 (R2016), 2016)
Caffeine	HPLC	(ISO 10727:2002 (R2018), 2018)
	UV spectrophotometry	(GB/T 8312-2013, 2013)
Total polyphenols	Folin–Ciocalteu method	(ISO 14502-1:2005 (R2015), 2015)
Catechins	HPLC	(ISO 14502-2:2005 (R2015), 2015)
Free amino acids	Ninhydrin colorimetric analysis	(GB/T 8314-2013, 2013)
Theanine	HPLC	(ISO 19563:2017, 2017)
Aflatoxin B1	ID-LC/MS/MS, ELISA, TLC	(GB 5009.22-2016, 2016)
	HPLC-precolum derivatization	
	HPLC-post column derivatization	
Lead	GFAAS, ICP-MS, FAAS	(GB 5009.12-2017, 2017)
	Dithizone colorimetric analysis	
Lead chromate	HPLC-ICP/MS, IC	(BJS 201910, 2019)
PAHs	HPLC, GC-MS	(GB 5009.265-2016, 2016)
Pesticides	GC-MS	(GB/T 23204-2008, 2008; GB/T 23376-2009, 2009)
Grade	Human panel test	(GB/T 23776-2018, 2018)
		(ISO 3103:2019, 2019; ISO 8589:2007 (R2017), 2017)
Sensory evaluation		

Abbreviations: ELISA, enzyme-linked immunosorbent assay; FAAS, flame atomic absorption spectrometry; GFASS, graphite furnace atomic absorption spectrometry; IC, ion chromatography; ICP, inductively coupled plasma; ID-LC, Isotope dilution liquid chromatography.

niques, electrochemical detections (ECDs), nanozymes, computer vision, and modifications of chromatographic techniques as emerging techniques for the determination of the parameters of tea products, both for quality and safety, compare their performances, and to propose the challenges and future trends of these emerging techniques.

## 2 STANDARD TECHNIQUES

Standard techniques for determining the quality and safety of tea products are mostly formulated by national standardization organizations or international standardization organizations (ISO). Due to the difference in the history and habits of tea-drinking, there are differences in the main types of tea among tea-consuming countries. For example, black tea and instant tea are the main types of tea in tea-consuming countries outside of China. This difference has led to the current situation of incomplete coverage of ISO standards related to tea to a certain extent. Table 1 demonstrates the vast majority of current standard techniques for determining different parameters of tea products, and it can be clearly found that for common parameters, such as moisture, crude fiber, polyphenols, catechins, and theanine, ISO has laid down standards for their detections, and detection methods in ISO standards for dry matter content (ISO 1572:1980 (R2015), 2015), moisture (ISO 1573:1980 (R2015), 2015), and alkalinity of water-soluble ash (ISO 1578:1975 (R2015), 2015) are adopted by most tea-growing and -consuming countries (e.g., India,

Sri Lanka, Turkey, and the United Kingdom, seen the full lists in the *FOREWORD* of these ISO standards). For the parameters without standards in ISO, such as heavy metals and organic contaminants, their standard techniques are formulated by countries and distinct both in methods and maximum residue limits (MRLs) (Karak & Bhagat, 2010; Ministry of Commerce of the People's Republic of China, 2018), causing trade barriers to tea exports (Ministry of Commerce of the People's Republic of China, 2018). Table 1 also lists the relevant national standards related to tea and three mandatory national food safety standards in China, namely limits of mycotoxins in food (GB 2761-2017, 2017), limits of contaminants in food (GB2762-2017, 2017), and MRLs for pesticides in food (GB 2763-2019, 2019), to present standard techniques for determining them as a supplement.

When researchers actually used the detection methods in these national standards, some problems were revealed. Analyzing and summarizing these problems, it is found that they are mainly in the following three areas: incomplete standardization field, high requirements for sensory evaluation, and the complex matrix effect.

### 2.1 | Incomplete standardization field

From Table 1, it is obvious to see that the coverage of standards set by ISO is not enough; even like China, which

TABLE 2 Parameters of fresh tea leaves and processing tea leaves

Parameters		Definition
Fresh tea leaves <sup>a</sup>	Tenderness	Degree of growth of new shoots
	Evenness	Degree of uniformity
	Neatness	Number of sundries, including related to tea (old leaves) and unrelated to tea
	Freshness	Degree of keeping original physical and chemical properties
	Mechanical damage	Broken leaves and mechanical damage due to mechanical forces
	Deterioration	Buds, leaves, and young stems turn red, or acid gas or fermentation odor appears due to improper handling of fresh leaves after picking
Processing tea leaves <sup>b</sup>	Leaves with rain	Water on the surface of fresh tea leaves due to picking in rainy or foggy days
	Temperature	For withering, fermentation, and drying, usually a range, and decided by tea masters
	Time	For withering, fermentation, and drying, usually a range, and decided by tea masters
	Moisture content	Indicator to decide whether to enter the next processing step, changing as the category of tea changing, usually decided by tea masters

<sup>a</sup>Excerpted from Requirements for fresh leaves of tea, Standard of the People's Republic of China (GB/T 31748–2015).

<sup>b</sup>Excerpted from Classification of tea, Standard of the People's Republic of China (GB/T 30766–2014).

has 109 national standards related to tea at present, they merely focus on both ends of the processing of tea products, that is, the definition of tea categories, the specification of tea processing, and the detection of chemical constituents and contaminants in tea products. Standard techniques for monitoring parameters of fresh tea leaves and processing tea leaves during the picking and processing are lacking. However, multiple parameters (GB/T 30766–2014, 2014; GB/T 31748–2015, 2015) in the picking and processing of fresh tea leaves and processing tea leaves have significant influence on the quality of commercial tea and other tea products, seen in Table 2. The lack of standardized techniques makes the quality of fresh tea leaves and processing tea leaves is assessed by appearance, aroma, and tactile impression by tea masters, thereby depending heavily on human experience rather than standardized indicators. Therefore, it is necessary to evaluate these parameters during the picking and processing of fresh tea leaves and processing tea leaves rapidly and nondestructively to establish a more standardized process for commercial tea and other tea products.

## 2.2 | High requirements for sensory evaluation

Figure 2 demonstrates the basic process of the sensory evaluation of tea according to China's national standard (GB/T 23776-2018, 2018) and international standard (ISO 3103:2019, 2019). In addition to the qualification certificates required for the sensory panelists, as Figure 3 illustrated, to conduct a sensory evaluation of green tea in

China requires standard evaluation equipment, including the color of the evaluation table; material, capacity, and style of the standard cup and bowl; and water used for evaluation; these objective requirements are also applicable in the standard established by ISO for the sensory test of tea (ISO 3103:2019, 2019). Meanwhile, standard operations are demanding, which are strongly dependent on the technical level of the professionals performing the evaluation. Taking the sensory evaluation of tea in China as an example (GB/T 23776-2018, 2018), the brewing time for different categories of tea, and the proportion of the five indicators, namely appearance, color of infusion, aroma, taste, and infused leaf in the score table are different for different categories of tea, as seen in details in Table 3. Appearance in the five indicators is judged by the leaf before brewing in the plate specially designed for tea samples, and infused leaf is referred to the tea leaf after brewing and judged in the plate dedicated for infused leaves. Notably, these standardized evaluation methods cannot make rapid, accurate, and nondestructive identification of tea grades on the spot. It is of great practical significance to identify commercial tea grades with high accuracy on the spot because there are huge differences in the economic values between different grades of commercial tea, especially the renowned teas from China. For instance, for Xihu (West Lake) Longjing tea, one of the top 10 popular commercial teas in China, the price of its highest grade (per 500 g) is 15 to 20 times the lowest grade (Yu, Wang, Xiao, & Liu, 2009; Yu, Wang, Yao, Zhang, & Yu, 2008), and the consistency between price and grade is the most important point considered by consumers when buying famous commercial tea.

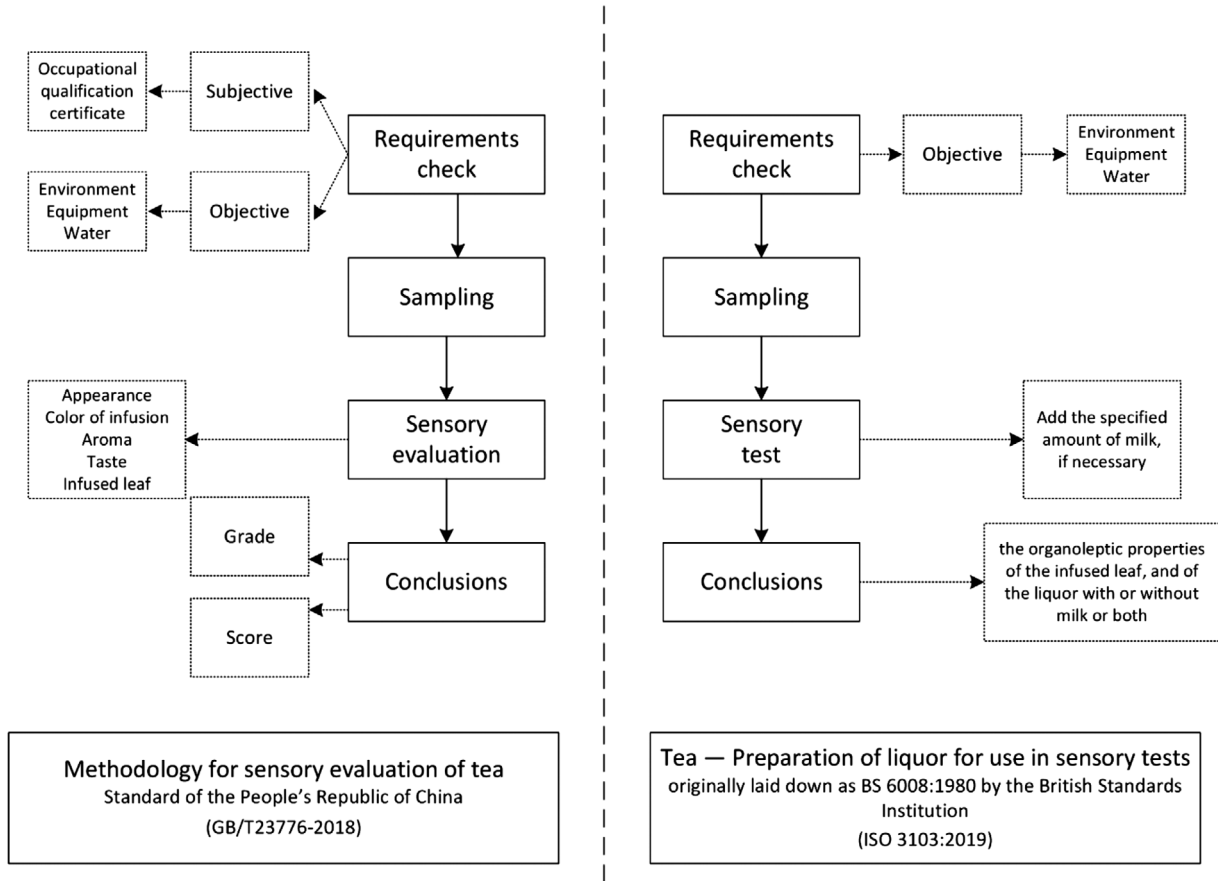


FIGURE 2 Basic process of the sensory evaluation of tea in China and the sensory test in Britain

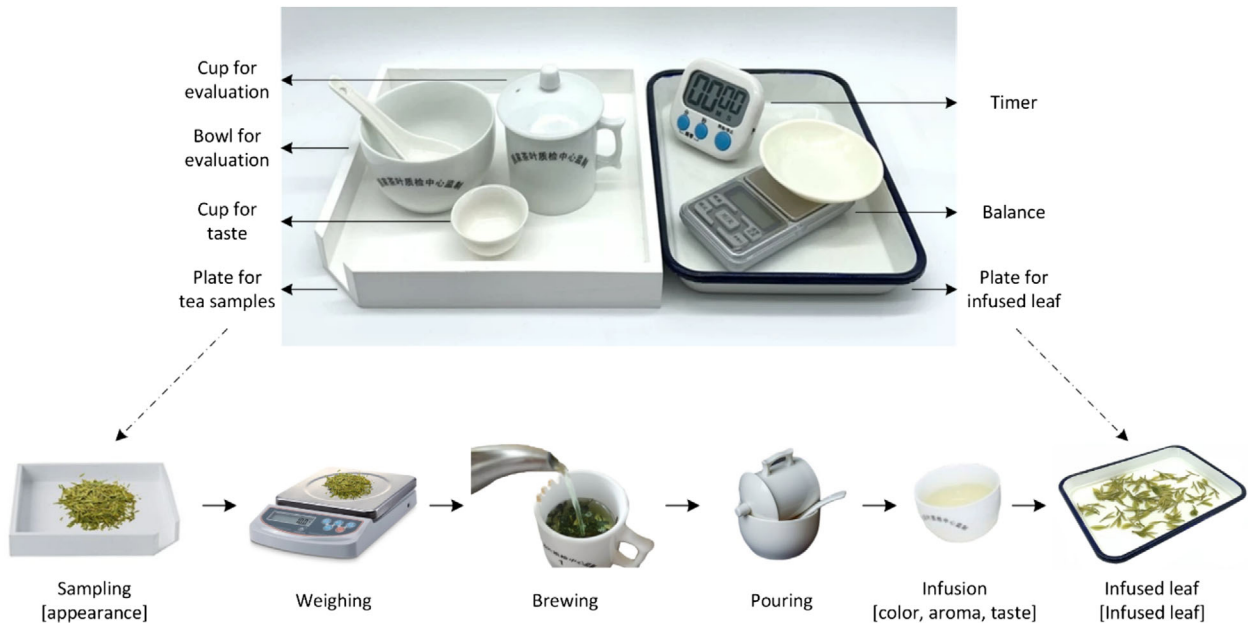


FIGURE 3 Basic equipment and process of the sensory evaluation of green tea in China

TABLE 3 Brewing time and proportions of five indicators for green, black, white, and yellow tea<sup>a</sup>

Category	Brewing time (min)	Appearance (%)	Infusion color (%)	Aroma (%)	Taste (%)	Infused leaf (%)
Green tea	4	25	10	25	30	10
Black tea(leaf)	5	25	10	25	30	10
Black tea(crushed)	5	20	10	30	30	10
White tea	5	25	10	25	30	10
Yellow tea	5	25	10	25	30	10

<sup>a</sup>Excerpted from *Methodology for sensory evaluation of tea*, Standard of the People's Republic of China (GB/T 23776-2018).

## 2.3 | Complex matrix effect

Studies have found that so far, more than 700 known compounds have been isolated and identified in tea leaves (Wan, 2011), leading to a complex matrix for determining contaminants. Due to the high toxicity of contaminants, their MRLs in tea products are usually of the magnitude of 10 mg/kg and lower (GB 2762-2017, 2017; GB 2763-2019, 2019). Thus, the limit of detection (LOD) and the limit of quantification (LOQ) of existing standard detection methods are not sufficient, which needs to be improved to promote the sensitivity and accuracy. Moreover, the process of chemical parameter determination is difficult, consumes large amounts of reagents, and takes a long time; for example, a separation time of 30 min is necessary for high-performance liquid chromatography (HPLC) to determine the contents of caffeine and catechins in one sample, including epicatechin (EC), epigallocatechin (EGC), epicatechin gallate (ECG), and epigallocatechin gallate (EGCG). Such a long determination time is clearly not conducive for the detection of large quantities of samples.

## 3 | EMERGING TECHNIQUES

Emerging techniques have been developed to solve the above problems of standard techniques for quality and safety determination of tea products. Based on trial and analysis for more than a decade, spectroscopic techniques, electrochemical methods, nanozymes, computer vision, and modifications of chromatographic techniques have been applied to evaluate various parameters of tea products qualitatively and quantitatively (Table 4 to 6), acting as emerging techniques. Each detection technique has its own unique advantages and disadvantages according to their different detection principles. At the same time, they are suitable for different detection target. Chemometrics, the art of extracting chemically relevant information from data produced in chemical experiments (Wold, 1995), has been used as an effective method of data extraction and processing, playing an important role together with the

above-mentioned five emerging techniques for the parameter evaluation of tea products.

### 3.1 | Spectroscopic techniques

Spectroscopy is the term used to refer to the measurement of radiation intensity as a function of wavelength, and it is used in physical and analytical chemistry because atoms and molecules have unique spectra. Consequently, these spectra can be used to detect, identify, and quantify information about the atoms and molecules. The focus of this section is spectroscopic techniques applied in the parameter evaluation of tea products, such as UV-vis spectroscopy, NIR spectroscopy, FT-IR spectroscopy, Raman spectroscopy, LIBS, and terahertz. Their basic determination process is shown in Figure 4, usually going through four basic steps before arriving at a conclusion: sample preparation, spectra acquisition, spectra processing, and model establishment. Generally, comparing the results of spectroscopic techniques with those obtained from standard techniques is necessary if the analyst uses a detection method formulated by national standards.

Wavelength is one of the indicators to distinguish spectroscopic techniques, and atoms or molecules behave differently at different wavelengths: in the region of the electromagnetic spectrum (from 380 to 780 nm), atoms and molecules undergo electronic transitions (Skoog, Holler, & Crouch, 2007), while in the near-infrared region of the electromagnetic spectrum (from 780 to 2,500 nm), molecular overtone and combination vibrations are acquired. Combined with infrared spectroscopy, FT-IR spectroscopy collects spectra based on measurements of the coherence of a radiative source, using time-domain or space-domain measurements of the electromagnetic radiation or other types of radiation. Relying upon the inelastic scattering of photons, known as Raman scattering, Raman spectroscopy is commonly used in chemistry to provide a structural fingerprint through which molecules can be identified (Gardiner & Graves, 1989) and lasers in the UV, Vis, and NIR range can be used as excitation light sources for Raman

**TABLE 4** Applications of emerging techniques for determining physical, chemical, and microbiological parameters of tea products

Tea product (form in determination)	Technique	Parameter (analyte)	Accuracy	Remark	References
Fresh tea leaves	Raman	Tenderness (carotenoids)	Same trend with HPLC	Different leaf maturity	Zhang et al. (2020)
Fresh tea leaves (compressed leaves)	NIR	Moisture, total nitrogen, crude fiber	$R^2_p$ 0.791 to 0.830 RMSEP 0.103 to 0.361	PLS	Wang et al. (2014)
Commercial tea (compressed powder)	NIR	Caffeine Total polyphenols	$R^2_p$ 0.939, 0.865 RMSEP 0.0836%, 1.1138%	PLS	Chen et al. (2006)
Commercial tea (powder)	FT-IR	Catechins	$R^2_p$ 0.921 to 0.971 RMSEP 0.103 to 0.361	MC + SNV	Chen, Zhao, Chaitep, and Guo (2009)
Fresh tea leaves (pellet)	FT-IR	Polyphenols	$R^2_p$ 0.707 RMSEP 1.289	RF	Li et al. (2015)
Commercial tea (infusion)	Nanozymes	Tannic acids	LOD 19 nM	Mn <sub>3</sub> O <sub>4</sub> nano-octahedrons output: color	Zhang and Huang (2015)
Commercial tea (infusion)	Nanozymes	Polyphenols	LOD 10 nM	AuNCs-p-h <sup>a</sup> output: color	Wang et al. (2016)
Commercial tea (infusion)	ASV	Polyphenols	LOD 10 mg/L	Modified glassy-carbon electrode	Guo et al. (2009)
Commercial tea (infusion)	CV	Polyphenols	$R^2$ 0.71 to 0.88 <sup>b</sup>	Inert carbon electrode	Roginsky et al. (2003)
Commercial tea (infusion)	CV	Polyphenols	Consistent with HPLC	Carbon electrode	Kilmartin and Hsu (2003)
Commercial tea (infusion)	SWV	EGC, EGCG	Good agreement with RP-HPLC + ECD, but lower	Glassy-carbon electrode	Novak et al. (2010)
Commercial tea (infusion)	CLC-ECD	C, CG, EC, GC, ECG, GCG, EGC, EGCG	LOD 54 to 89 amol		Kotani et al. (2007)
Commercial tea (infusion)	Electro-analytical sensing	EGC, EGCG (aminophenol oxidation)		Modified graphite electrode	Goodwin, Banks, and Compton (2006)
Commercial tea (infusion)	CZE + UV	Caffeine, theanine, ascorbic acid, C, EC, EGC, ECG, EGCG		Quicker than HPLC	Horie et al. (1997)
Commercial tea (powder)	SWV	EGCG		Paraffin-impregnated graphite electrode	Komorsky-Lovrić and Novak (2009)
Commercial tea (infusion)	E-tongue	Sugar content, catechin, EGC	$R^2$ 0.890 to 0.967	PLS	Lvova, Legin, Vlasov, Cha, and Nam (2003)
Commercial tea (infusion)	E-nose + E-tongue + E-eye	Amino acids, catechins, polyphenols, caffeine	$R^2_p$ 0.978 to 0.989 RMSEP 0.076 to 0.669	Fusion signals by RF	Xu et al. (2019)
Commercial tea (infusion)	HPLC + MAE	Caffeine, C, CG, GC, GCG, EC, ECG, EGC, EGCG	LOD 0.11 to 0.29 mg/L	Four times faster about 7 min	Rahim, Nofrizal, and Saad (2014)

(Continues)

TABLE 4 Continued

Tea product (form in determination)	Technique	Parameter (analyte)	Accuracy	Remark	References
Commercial tea (infusion)	HPLC + ECD	EGCG, EGC, EGC3"Me	LOD 10, 10, 40 pmol/mL	Applied voltage of 600 mV	Sano et al. (2001)
Commercial tea (infusion)	RP-HPLC/UV	15 phenolic compounds	LOD 0.06 to 2.92 mg/L	Extraction optimized	Bae et al. (2015)
Commercial tea	HPTLC	Catechins	LOD 10 to 60 ng/band	282, 285 nm	Khan et al. (2011)
Commercial tea (infusion)	HPTLC	Caffeine		275 nm	Shanmuga-sundaram, Manjunatha, Vijayan, Khatwal, and Samanta (2011)
Commercial tea (infusion)	HPTLC	Caffeine	LOQ 0.05 µg	254 nm Fe <sup>3+</sup> cations	Kartsova and Alekseeva (2009)
Commercial tea (infusion)	HPTLC	Caffeine, theobromine, theophylline		275 nm	Kunugi and Tabei (1997)
Commercial tea (extraction)	HPTLC	Caffeine	LOD 10 ng/spot	273, 254 nm	Do, Santi, and Reich (2019)
Commercial tea (extraction)	HPTLC	Theanine, catechins	LOD 5.36, 16.25 ng/spot	480, 254 nm	Kumar et al. (2016)
Commercial tea (extraction)	HPTLC	Caffeine, polyphenols		254 nm	Min and Peigen (1991)
Commercial tea (infusion)	HPLC-UV GC/MS LC/MS, ICP-MS	Polyphenols and caffeine volatile compounds amino acids, minerals		280 nm	Liao et al. (2018)
Commercial tea (infusion)	Enantio-selective GC-MS	Enantiomers of volatile components	LOD 0.21 to 1.88 ng/g		Mu et al. (2018)
Commercial tea (extraction)	UHPLC-PDA	Methylxanthines	LOD 0.05 to 0.1 µg/mL	ASE	Ahmad et al. (2020)
Commercial tea (extraction)	UHPLC	Mycotoxins (aflatoxin B1)	LOD 0.5 to 10.0 µg/kg	5.4 µg/kg in one sample	Martínez-Domínguez et al. (2016)
Tea bakery product (dough)	RP-HPLC + PAD	Tea catechins stability		Relatively stable	Wang and Zhou (2004)

<sup>a</sup>AuNCs-protein(p)-heating(h), AuNCs-p-h.

<sup>b</sup>Compared with the absorbance of the teas at 272 nm.

Abbreviation: CLC-ECD, capillary liquid chromatography with electrochemical detection; MAE, microwave-assisted extraction; ASE, accelerated solvent extraction.

spectroscopy. LIBS uses a highly energetic laser pulse as the excitation source (Anabitarte, Cobo, & Lopez-Higuera, 2012; Cremers & Radziemski, 2006) to form a plasma, which atomizes and excites samples, and then obtains the intensity of various elements in their wavelengths. In spectroscopic techniques, terahertz spectroscopy is referred to as terahertz time-domain spectroscopy (THz-TDS), in which the properties of matter are probed with short

pulses of terahertz radiation. The generation and detection scheme are sensitive to the effect of the sample on both the amplitude and phase of terahertz radiation. By measuring in the time domain, the technique can provide more information than conventional Fourier-transform spectroscopy, which is only sensitive to the amplitude.

The feasibility studies of NIR, FT-IR spectroscopy, and Raman spectroscopy in qualitative and quantitative anal-



TABLE 5 Applications of emerging techniques for determining classified parameters of tea products

Tea product (form in determination)	Technique	Parameter (analyte)	Accuracy	Remark	References
Commercial tea (infusion)	E-nose	Kind	RBF 100%	A tin oxide-base E-nose potentiometric	Dutta et al. (2003)
Commercial tea (infusion)	E-tongue	Kind	PCA effective	All-solid-state	Lvova et al. (2003)
Commercial tea (pellet)	NIR	Grade	PLS 93.10%		Wang et al. (2014)
Commercial tea (infusion)	E-nose	Grade (VOCs)	SVM 95%	4 PCs	Chen et al. (2011)
Commercial tea (infusion)	E-nose	Grade (VOCs)	ANN 90%		Yu and Wang (2007)
Commercial tea (infusion)	E-nose	Grade (VOCs)	PCA 100%		Qin et al. (2013)
Commercial tea (infusion)	E-nose	Grade	KLDA-KNN 100%	Time/frequency domain features fusion	Dai et al. (2015)
Commercial tea (infusion)	E-nose	Grade (VOCs)	CPNN 93.85%		Borah et al. (2008)
Commercial tea	E-nose	Grade (VOCs)	BPNN 88%	3 PCs	Yu et al. (2008)
Commercial tea	E-nose	Grade (VOCs)	SVM 100%	Combined with computer vision	Xu et al. (2019)
Commercial tea	E-nose	Grade	LDA 100%	Feature vectors of average value and integrated value	Yu et al. (2008)
Commercial tea (leaf, infusion, and remain)	E-nose	Grade	LDA and BPNN in accordance	5 PCs infusion (100%) > leaf (96.67%) > remain (93.33%)	Yu et al. (2009)
Commercial tea (infusion)	E-tongue	Grade	ANN 100%		Chen, Zhao, and Vittayapadung (2008)
Commercial tea (infusion)	E-nose + E-tongue	Grade	KNN 91.5%		Chen et al. (2011)
Commercial tea (infusion)	E-nose + E-tongue + E-eye	Grade	SVM/RF 100%	Fusion signals	Xu et al. (2019)
Commercial tea (infusion)	artificial nose + artificial tongue	Grade + type	HCA/PCA 100%		Huo et al. (2014)
Commercial tea (infusion)	E-tongue	Grade + geographical origin	PCA 100%		He et al. (2009)
Commercial tea (infusion)	E-tongue	Geographical origin + picking season	PCA L1 and L2 overlap in seven samples	MLAPV	Tian, Deng, and Chen (2007)
Commercial tea	Computer vision	Grade	MLP 82.33%	Texture analysis	Gill, Kumar, and Agarwal (2013)
Commercial tea	Computer vision	Grade	LVQ 80%	Texture analysis	Borah, Hines, and Bhuyan (2007)

(Continues)

TABLE 5 (Continued)

Tea product (form in determination)	Technique	Parameter (analyte)	Accuracy	Remark	References
Commercial tea	Machine vision	Grade	PCA 96% variance	Darkfield illumination Image texture feature	Laddi, Sharma, Kumar, and Kapur (2013)
Commercial tea (infusion)	UV-Vis	Geographical origin + category	SPA-LDA 100%	251 to 490 nm	Diniz et al. (2016)
Commercial tea (pellet)	Terahertz	Type	SVM 98.75%	5% noise: 86.25%	Chen et al. (2011)
Commercial tea (digestion)	ICP-AES	Category	BP-ANN 95%	Zn, Mn, Mg, Cu, Al, Ca, Ba, and K pattern recognition procedure	Herrador and González (2001)
Commercial tea (digestion)	ICP-MS	Geographical origin + type	LDA 100% 95.2% in detailed mountains	Fe, Mn, Sc, and As	Ma et al. (2016)
Commercial tea (digestion)	ICP-MS	Geographical origin	S-LDA 100%	Na, Mg, Ca, Ni, Rb, Sr, and Pb	Zhao et al. (2017)
Commercial tea (digestion)	ICP-MS	Geographical origin	SLDA 100%	Mg, Ni, Rb, Sr, Cd, and Pb	Zhao et al. (2017)
Commercial tea (digestion)	Isotope signature study	Geographical origin	PCA good differentiation	<sup>87</sup> Sr/ <sup>86</sup> Sr, <sup>13</sup> C, Sr concentration	Lagad et al. (2013)
Commercial tea	GC-MS	Type (VOCs)	S-LDA 100%	HS-SPME	Lin et al. (2013)
Commercial tea (infusion)	Enantioselective GC-MS (Es-GC-MS)	Geographical origin (volatile constituents)	PLS-DA 100%	HS-SPME enantiomeric distributions	Mu et al. (2018)
Commercial tea (infusion)	E-nose	Category (VOCs)	LDA 100%	Odor imaging sensor array	Chen et al. (2013)
Commercial tea (infusion)	E-nose	Category + type	LAPV + staircase		Ivarsson, Holmin, Höjer, Krantz-Rülcker, and Winquist (2001)
Commercial tea (infusion)	CV	Category	SVM 98.75%	Bi <sub>2</sub> O <sub>3</sub> oxide-metallic modified electrode	Liu et al. (2014)
Commercial tea	Computer vision	Category	FSVM + WTA 97.77%	color + texture	Wang et al. (2015)
Commercial tea	Multispectral imaging	Category	RBF-LS-SVM 100%	Entropy image texture	Wu, Yang, Chen, He, and Li (2008)
Commercial tea (extraction)	HPTLC	Batch (flavonoid) Category (polyphenol pattern)	Identify green tea	366 nm	Reich et al. (2006)
Cut tea leaves (under fermentation)	E-nose	Fermentation time (VOCs)	Accuracy method with human panel test		Bhattacharyya et al. (2007)
Fresh tea leaves (under fermentation)	E-nose	Fermentation time + mechanical grading	SITO-MWTS 97.35%		Kaur et al. (2012)

(Continues)

TABLE 5 (Continued)

Tea product (form in determination)	Technique	Parameter (analyte)	Accuracy	Remark	References
Fresh tea leaves (under fermentation)	E-nose	Fermentation process (VOCs)	96% versus human evaluation		Bhattacharyya et al. (2007)
Fresh tea leaves (under fermentation)	Imaging processing	Fermentation	90% with sensory panel	Image, color	Borah and Bhuyan (2003)
Tea powder (matcha)	Vis spectroscopy	Adulteration (steamed green tea powder)	PCA-LDA 100%	Semiquantitative, PLS $R^2_p$ 0.982, RMSEP 4.646	Yu and He (2018)
Tea powder (matcha)	LIBS	Adulteration (green tea powder)	PCA-LDA 100%	Mg, Ca, Fe, and C	Yu et al. (2017)

Abbreviations: FSVM, fuzzy support vector machine; HS-SPME, headspace solid phase microextraction; LAPV, large amplitude pulse voltammetry; MLAPV, multifrequency large amplitude pulse voltammetry; MWTS, moving window time slicing; SITO, social impact theory-based optimizer; SVM, support vector machine; WTA, winner-take-all.

ysis of various processed food products have been thoroughly investigated (Su & Sun, 2018), and most of them performed suitably together with chemometrics. Careful development of a set of calibration samples and application of multivariate calibration techniques (Balabin, Safieva, & Lomakina, 2007) is essential for NIR, FT-IR spectroscopy, Raman spectroscopy, and other spectroscopic techniques for the parameter determination of tea products.

### 3.2 | Electrochemical methods

Electroanalytical methods are a class of techniques in analytical chemistry that are used to study an analyte by measuring the potential (volts) and/or current (amperes) in an electrochemical cell containing the analyte (Bard & Faulkner, 2000; Skoog, West, Holler, & Crouch, 2013). Voltammetry, which measures the current as the potential is varied, and electronic sensing, including the electronic nose (E-nose), electronic tongue (E-tongue), and electronic eye (E-eye), are two major electrochemical methods used in the parameter evaluation of tea products.

Particularly, ASV, CV, SWV, and staircase voltammetry contribute to the quality determination of tea products, with the help of various kinds of electrodes, such as inert carbon electrode (Roginsky, Barsukova, Hsu, & Kilmartin, 2003), glassy-carbon electrode (Guo et al., 2009; Novak et al., 2010), and paraffin-impregnated graphite electrode (Komorsky-Lovrić & Novak, 2009).

The expression of electronic sensing refers to the capability of reproducing human senses using sensor arrays and pattern recognition systems, covering the E-nose, E-

tongue, and E-eye. Previous researches (Xu et al., 2019) found that the fusion signals of the above two or three techniques performed better than the single signal, and chemometrics also played a significant role in the determination. The basic determination process of electronic sensing is illustrated in Figure 5, and the first step, sensors selection, is crucial.

### 3.3 | Nanozymes

Nanozymes, or enzyme-mimetic nanomaterials, possess both enzyme-like properties and nanomaterial features (Manea, Houillon, Pasquato, & Scrimin, 2004), which meet the inspection requirements of high sensitivity, specificity, and reproducibility, and are excellent tools for quality and safety detection in the agri-food sector (Huang, Sun, Pu, & Wei, 2019).  $Fe_3O_4$  nanoparticles (NPs) (Guan et al., 2012),  $Mn_3O_4$  octahedrons (Zhang & Huang, 2015), and protein conjugated gold nanoclusters (protein-Au NCs) (Wang et al., 2016) are three kinds of nanozymes used to determine the contents of ethoprophos, tannic acid, and tea polyphenols in tea products, with the magnitude of LOD at nM.

### 3.4 | Computer vision

From an engineering point of view, computer vision aims to build autonomous systems that can perform some of the tasks that the human visual system can perform (and even surpass it in many cases) (Huang, 1996). Image acquisition, preprocessing, feature extraction, detection, high-level processing, and decision making form the method

TABLE 6 Applications of emerging techniques for determining the safety of tea products

Tea product (form in determination)	Technique	Parameter	Accuracy	Remark	References
Commercial tea (digestion)	Nanozymes	Ag, Cu, Pb, Cd, and Hg	LOD 0.01 to 0.09 ng/mL	Modified Fe <sub>3</sub> O <sub>4</sub> NPs	Mashhadizadeh, Amoli-Diva, Shapouri, and Afruzi (2014)
Commercial tea (powder)	FT-IR	Talcum powder	R <sup>2</sup> <sub>p</sub> 0.927 RMSEP 0.137	ELM	Li et al. (2016)
Commercial tea (infusion)	Raman	Lead chrome green	R <sup>2</sup> <sub>p</sub> 0.876 RMSEP 0.803	PLSR	Li et al. (2015)
Commercial tea (infusion)	HPLC	PAHs	LOD 4 to 145 ng/L	HS-SPME	Viñas et al. (2007)
Commercial tea (leaf and infusion)	HPLC	PAHs	LOD 0.2 to 0.3 µg/kg (leaf) 0.1 µg/L (infusion)	QuEChERS extraction	Pincemaille, Schummer, Heinen, and Moris (2014)
Commercial tea (infusion)	HPLC – FLD	PAHs	LOD 0.15 to 0.25 µg/kg	QuEChERS extraction	Zachara et al. (2018)
Commercial tea (extraction)	HPLC + FLD, DAD	PAHs	LOD 0.01 to 0.7 µg/L		Garcia Londoño et al. (2015)
Commercial tea (extraction)	GC × GC – TOFMS	PAHs	LOQ 0.05 to 0.2 mg/kg	SPE on MIPs	Drabova et al. (2012)
Tea beverage	µ-HPLC	PAHs	LOD 0.1 to 50 ng/L	MWCNTS-AFME	Loh et al. (2013)
Commercial tea (extraction)	HPLC-MS/MS	102 pesticides	LOD 0.03 to 15 µg/kg	QuEChERS extraction	Huang et al. (2019)
Fresh tea leaves (extraction)	UPMC-MS/MS	20 pesticides	LOQ 0.01 to 0.02 mg/kg	QuEChERS extraction, PVPP	Guo et al. (2019)
Commercial tea (infusion)	UPLC-MS/MS	65 pesticides	LOQ 5 to 20 µg/kg	QuEChERS extraction	Chen et al. (2011)
Commercial tea (extraction)	UHPLC	Pesticides	LOD 0.5 to 10.0 µg/kg	Exactive-orbitrap HRMs	Martínez-Domínguez et al. (2016)
Commercial tea (extraction)	HPTLC	Temephos, fenitrothion	LOD 20, 10 ng	290 nm	Fan et al. (2011)
Commercial tea (extraction)	HPTLC	Six pesticides	LOD 3 to 20 ng		Yue et al. (2008)
Commercial tea (extraction)	HTPLC (quality) UHPLC-MS (quantification)	DMBA	LOD 10 ng/mL	Not detected in tea samples	Avula et al. (2015)
Commercial tea (extraction)	GC-ICP-MS/MS	Organophosphorus, organosulfur, and organochlorine pesticides	LOD (lowest) 0.0005, 0.675, 0.144 µg/kg		Nelson et al. (2015)
Commercial tea (extraction)	SERS	Phosmet	LOD 3 µg/L	Sliver nanoparticle	Pan et al. (2015)

(Continues)

TABLE 6 (Continued)

Tea product (form in determination)	Technique	Parameter	Accuracy	Remark	References
Tea beverage	HDEs	Pyrethroids	LOD 0.06 to 0.17 ng/mL	HFIP	Deng et al. (2019)
Commercial tea (pellet)	ATR-FTIR	Sibutramine	HCA, PCA 100%	0.375 to 12 mg in 1.75 g green tea determined	Cebi et al. (2017)

Abbreviations: HDEs, hydrophobic deep eutectic solvents; HFIP, hexafluoroisopropanol; HRMS, high-resolution mass spectrometry; MIPs, molecularly imprinted polymers; MWCNT-AFME, multi-walled carbon nanotube-impregnated agarose film microextraction; SPE, solid phase extraction; TOFMS, time-of-flight mass spectrometry.

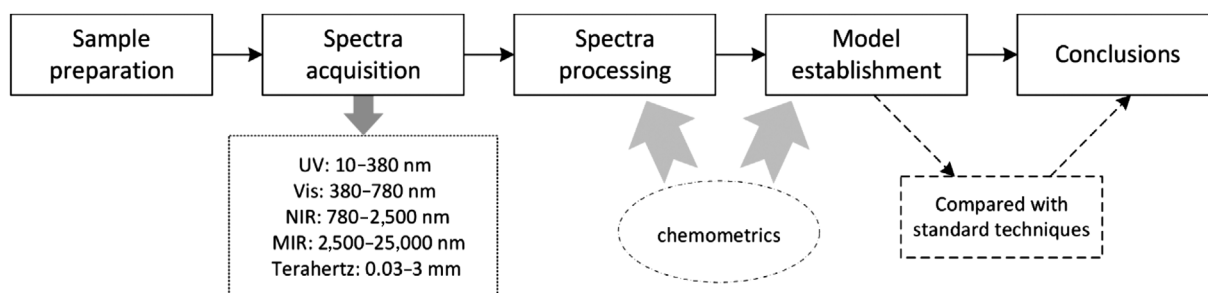


FIGURE 4 Basic determination process of spectroscopic techniques

of computer vision. However, due to its principle, computer vision merely appeared in the evaluation of classified parameters in tea products.

### 3.5 | Modifications of chromatographic techniques

Liquid chromatography/mass spectrometry (LC/MS), HPLC, gas chromatography/mass spectrometry (GC/MS), and thin-layer chromatography (TLC) are basic chromatographic techniques for the determination of chemicals and organic contaminants in tea products; nevertheless, these methods have room for improvement in simplicity, rapidity, and accuracy. Modifications are accordingly developed and used.

QuEChERS, an acronym word formed from “quick, easy, cheap, effective, rugged, and safe,” is a solid-phase extraction method for the detection of pesticide residues in food (Schenck & Hobbs, 2004). It is used in the extraction of pesticide residues and polycyclic aromatic hydrocarbons (PAHs) in commercial tea prior to HPLC to improve the sensitivity for multiple organic contaminant analysis in complex matrices. Reversed-phase HPLC, ultra-HPLC (UHPLC), combining HPLC or UHPLC with ECD (Novak et al., 2010), diode array detection (DAD) (Ahmad, Ahmad, Al-Anaki, Ismail, & Al-Jishi, 2020; Garcia Londoño, Reynoso, & Resnik, 2015), fluorescence detector (FLD) (Garcia Londoño et al., 2015; Zachara, Gałkowska, & Juszczak, 2018), photometric diode array (Ahmad et al., 2020), mass spectrometry (MS) (Guo et al., 2019; Huang et al., 2019), and UV detector (Bae, Ham,

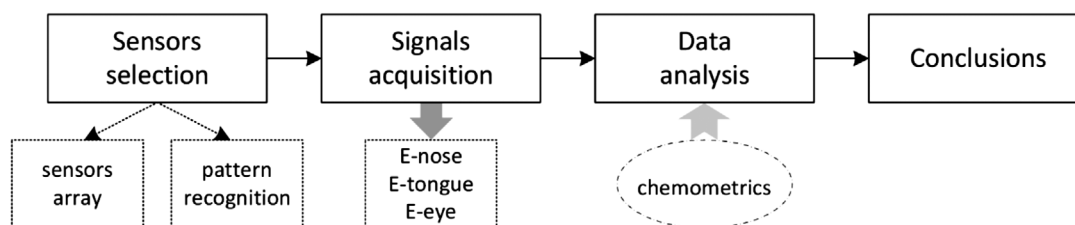


FIGURE 5 Basic determination process of electronic sensing

TABLE 7 Methods of model establishment for determining the quality and safety of tea products

Acronym	Method	Acronym	Method
PCA	Principal component analysis	KNN	K-nearest neighbors
KPCA	Kernel-based principal component analysis	ANN	Artificial neural network
HCA	Hierarchical cluster analysis	BPNN	Back-propagation neural network
PLS	Partial least squares	CPNN	Constructive probabilistic neural network
SPA	Successive projections algorithm	PNN	Probabilistic neural network
ELM	Extreme learning machine	LDA	Linear discrimination analysis
SVM	Support vector machine	S-LDA	Stepwise linear discriminant analysis
LVQ	Learning vector quantization	KLDA	Kernel-based linear discriminant analysis
MLP	Multi-layer perceptron	RBF	Radial basis function
RF	Random frog		

Jeong, Kim, & Kim, 2015) are also applied as modifications of HPLC for determining organic contaminants in commercial tea, and significantly improving the LOD and LOQ of PAHs and pesticides.

As an enhanced form of TLC, high-performance thin-layer chromatography (HPTLC) is an analytical technique based on TLC, but with enhancements intended to increase the resolution of the compounds to be separated and to allow quantitative analysis of the compounds (Attimarad, Mueen Ahmed, Aldhubaib, & Harsha, 2011). Existing literatures demonstrate the feasibility and efficiency of HPTLC in the quantification of chemical parameters, such as caffeine and theanine (Khan et al., 2011; Kumar, Gulati, & Sharma, 2016; Kunugi & Tabei, 1997), and organic contaminants, such as pesticides (Yue, Zhang, Fan, & Tang, 2008) and 1,3-dimethylbutylamine (DMBA) (Avula et al., 2015). Applying HPTLC in the identification of green tea with different batches is also reported (Reich et al., 2006).

### 3.6 | Chemometrics

Spectra processing and model establishment are two main aspects of chemometrics, each contains multiple analysis methods. Standard normal variate (SNV) and multiplicative scatter correction are analysis methods mostly used in spectral processing; methods for model establishment are listed in Table 7, and some of them performed satisfactorily.

The coefficient of determination in prediction ( $R^2_p$ ) and the root-mean-square error of prediction (RMSEP) are two major evaluating indicators in chemometrics.  $R^2_p$  is the square of the correlation ( $r$ ) between the predicted and actual  $y$ -scores, ranging from 0 to 1, and RMSEP is used to evaluate the forecast ability of the model. The smaller the RMSEP value, the higher is the forecast ability of the model.

## 4 | QUALITY EVALUATION OF TEA PRODUCTS

Compared with other foods, in addition to the physical, chemical, and microbiological parameters, the classified parameter is the most unique parameter of tea products, because the quality of commercial tea is not only affected by the cultivar of tea trees, but also by the geographical origin, or other classified parameters. For some famous teas, such as Dongting Biluochun (Ma et al., 2016), the distance of the east mountain and the west mountain in geographical origin will give different qualities of commercial tea even with the same processing technology, which affects its economic value. Owing to the above reasons, the classified parameters are evaluated more than the physical, chemical, and microbiological parameters, as shown in Table 4 and 5.

### 4.1 | Physical, chemical, and microbiological parameters

At present, there is only one study using Raman spectroscopy to perform rapid, *in situ* evaluation of the physical parameter tenderness of fresh tea leaves (Zhang et al., 2020). The application is in its infancy, though, it is instructive. The tenderness of fresh tea leaves is one of the important factors affecting the quality of tea products, and other influential parameters are listed in Table 2. However, the current evaluation of the tenderness of fresh tea leaves is assessed manually by experienced tea farmers or tea workers through its appearance and aroma, which is strongly dependent on the proficiency of tea farmers and tea workers. By measuring the content of carotenoids in fresh tea leaves with different maturity, finding a strong correlation between their contents and the maturity of fresh tea leaves, and indirectly measuring the maturity (tenderness) of fresh tea leaves, the feasibility of this method has enlight-

ened us that it is possible to use metabolites of tea trees as direct measurement analytes to indirectly determine the physical parameters of fresh tea leaves, thereby replacing the current evaluation method, which uses human experience. To achieve this purpose, metabolites of tea trees used as direct analytes should have the following two characteristics: first, there should be unique and easily detectable peaks in the spectrum, such as a characteristic peak of  $1,520\text{ cm}^{-1}$  in the Raman spectra of carotenoids; second, their contents should have a significant positive or negative correlation with the physical parameters of the fresh tea leaves to be measured.

Emerging techniques are widely used for the determination of the chemical parameters of tea products. From the moisture content of fresh tea leaves (Wang, Zhang, Ning, Wei, & Li, 2014) to tea catechins stability in tea bakery products (dough) (Wang & Zhou, 2004), determinations are mainly focused on the rapid detection of tea polyphenols, caffeine, and catechins, namely catechin (C), EC, gallic catechin (GC), EGC, catechin gallate (CG), ECG, gallic catechin gallate (GCG), and EGCG. In these applications, the predicted value of spectroscopic techniques needs to be compared with the measured value of the standard technique, and the LOD of electrochemical methods performs better than spectroscopic techniques. The lowest LOD appeared in the determination of catechins by CLC-ECD (Kotani, Takahashi, Hakamata, Kojima, & Kusu, 2007). With this method, researchers not only determined catechins in the magnitude of attomole,  $10^{-18}$  of a mol, but also determined catechins in human plasma after the ingestion of green tea, with 10  $\mu\text{L}$  of human plasma required. Considering the wide consumption of caffeine at different levels by most segments of the population, the abundance of caffeine in tea products, and the moderate daily caffeine intake at a dose level of up to 400 mg/day for the healthy adult population, as well as  $\leq 300$  mg/day for the reproductive-aged women (Nawrot et al., 2003), applying emerging techniques to develop a rapid and *in situ* method for detecting caffeine contents in tea products, especially tea infusions and tea beverages, deserves a further research with a wide field of applications.

Currently, tea products are not regulated by existing Chinese and European legislations on limits of mycotoxins (Council Regulation, 2006; GB 2761-2017, 2017). Among tea products that apply emerging techniques for microbiological determination, one green tea product was found to contain 5.4  $\mu\text{g}/\text{kg}$  aflatoxin B1 (Martínez-Domínguez, Romero-González, & Garrido Frenich, 2016), which suggests that mycotoxin levels are not high in tea products, and modifications of HPLC are suitable for their determination. Tea products that have been stored for a long time may require more attention on mycotoxins.

## 4.2 | Classified parameters

Before discussing the application of emerging techniques in the evaluation of classified parameters in tea products in detail, it is necessary to understand the exact explanation of each classified parameter in tea products, as shown in Table 8.

Regarding the classified parameters listed in Table 8, there are three key points to pay attention to: first, varieties (Latin), which is a classification level in plant taxonomy, falls under species and subspecies; however, at the same time, variety (English) is selected to describe different commercial teas in existing researches, and usually, when researchers use variety, they refer to different commercial teas, such as Xihu (West Lake) Longjing, Dongting Biluochun, Dahongpao, and so on, whereas, this description is not appropriate, because in plant taxonomy, only three varieties of tea (*Camellia sinensis* (L.) Kuntze) are accepted officially: *Camellia sinensis* var. *assamica* (J.W.Mast.) Kitam., *Camellia sinensis* var. *waldenae* (S.Y.Hu) H.T.Chang, and *Camellia sinensis* var. *dehungensis* (H.T.Chang & B.H.Chen) T.L.Ming; therefore, type is temporarily used in this review to replace variety when it is referred to commercial teas; second, according to China's national standard on tea classifications (GB/T 30766-2014, 2014), there are six categories of tea in China mainly classified by processing technologies, while under each category of tea, different sublevel processing technologies still exist, like pan-fried, roasting, steaming, and sun-drying in green tea, which is classified through the method used for drying, and this classification is temporarily defined as kind in this review; third, the geographical origin is a classified parameter of significant importance for famous teas and traditional teas in major tea-growing countries, because most of them are protected geographical indication products, and their corresponding protection area is specified in their national standards (China) or notification of registration (Japan). Taking Longjing tea (GB/T 18650-2008, 2008) and traditional authentic Yame Gyokuro (Registration (No. 5), 2015) as examples, as shown in Figure 6, Longjing tea produced in Xihu District of Hangzhou can be called Xihu (West Lake) Longjing, Longjing tea produced in Hangzhou (except Xihu District) is called Qiantang Longjing, and Longjing tea produced in Shaoxing, another city in Zhejiang Province, is called Yuezhou Longjing; the producing area of traditional authentic Yame Gyokuro is stipulated as the mid-mountainous area of Yame city and surrounding cities in Fukuoka Prefecture, Japan.

In the existing applications, E-nose accounted for the highest proportion, and the classification of tea products based on geographical origins, grades, varieties, and categories through VOCs pointed to an internal reason: tea

TABLE 8 Explanations of classified parameters of tea products

Classified parameter	Explanation	Example
Species	Biology	<i>Camellia sinensis</i> (L.) O. Kuntze.
Varietas (Latin)	Biology	<i>Camellia sinensis</i> (L.) O. Kuntze. var. <i>sinensis</i>
Cultivar	Agriculture	Longjing 43, Fudingdabaicha, Rougui, etc.
Category	Classified by process technology	Green tea, black tea, yellow tea, white tea, oolong tea, and dark tea (GB/T 30766–2014, 2014)
Type	Classification of commercial tea	Xihu Longjing and Dongting Biluochun, etc.
Kind	Classified by process technology in the same category	Pan-fried, roasting, steaming, and sun-drying in green tea, etc. (Lvova et al., 2003)
Grade	Classified by level in the same type	Grade 1 to grade 5 in Xihu Longjing (Yu et al., 2009)
Geographical origin	Plant area of the cultivar for one type	Xihu (West lake) Longjing, Qiantang Longjing, and Yuezhou Longjing, seen details in Figure 6

products of different categories, varieties, grades, and geographical origins have different qualities, leading to different VOC contents. Therefore, the classified parameters of tea products actually reflect the quality of tea products, which is why there are various but momentous classified parameters of tea products. The quality of tea products is closely related to its economic value; nevertheless, the quality of tea products is too difficult to discern for ordinary consumers without professional training. The occurrence and application of emerging techniques make it possible to identify the quality of tea products quickly, nondestructively, and on the spot. Despite this, a classified parameter corresponds to a specific model establishing method in each research, and the practical application of emerging techniques, such as the E-nose, still needs to overcome a series of problems, such as the robustness and transitivity of models.

Applying E-nose and image processing under the fermentation of cut tea leaves (Bhattacharyya et al., 2007) and fresh tea leaves (Bhattacharyya et al., 2007; Borah & Bhuyan, 2003; Kaur et al., 2012) deserves more attention. The use of emerging techniques in tea manufacturing to monitor the real-time changes in processing tea leaves under fermentation, withering, or drying is helpful to the mechanization of tea processing, resulting in a quicker, cleaner, and more standard tea processing to replace the excessive reliance on human experience in tea processing.

As a newly developed spectroscopic method, THz-TDS had been applied in the research of four different varieties of Chinese green tea, and results showed that support vector machine (SVM) had better classification results in the experiment, demonstrating the feasibility of THz-TDS for the classification of different kinds of tea. Although THz-TDS is still in the research and development stage, it has promising application prospects as a new method for quality and safety determination of tea products, with advantages of no risk of radiation, high transparency for

nonpolar materials, strong spectral fingerprints for crystalline materials and polar molecules, and high sensitivity in both time and frequency domains (Ferguson & Zhang, 2002).

Two studies have utilized Vis spectroscopy (Yu & He, 2018) and LIBS (Yu et al., 2017) to determine the adulteration of green tea powder in matcha qualitatively and semi-quantitatively, whose practical significance still came from the huge gap in the economic values between matcha and green tea powder. These trials broadened the application field of spectroscopic techniques in tea products, as well as reminded researchers and consumers of the adulteration of matcha and its quality evaluation. Notwithstanding, the application of UV-vis spectroscopy and LIBS in the parameter determination of tea products is rare, which is principally caused by the similarity of the color of tea products in the visible ranges to a large extent, and the focus of LIBS on the element rather than the molecule.

On the basis of analyzing inorganic elements, inductively coupled plasma atomic emission spectroscopy (ICP-AES) (Herrador & González, 2001), ICP-MS (Ma et al., 2016; Zhao, Yu, & Li, 2017; Zhao, Zhang, & Zhang, 2017), and isotope signature study (Lagad et al., 2013) successfully differentiate the categories, types, and geographical origins of commercial teas, indicating that commercial teas with different categories, types, and geographical origins contain distinct contents of inorganic elements, covering from macroelements, such as Ca and Mg, to microelements, such as Na, Fe, and Mn; heavy metals, such as Cd and Pb, as well as rare earth elements, such as Sc, also play an important role in the identification.

Enantiomeric distributions could also be used to identify green tea, whose tea trees are planted in different areas (Mu et al., 2018), carrying out a novel but inspiring trial. An HPTLC method presents that the flavonoid fingerprint of green tea can give information about the geographical origin of the material, and the polyphenol pattern can



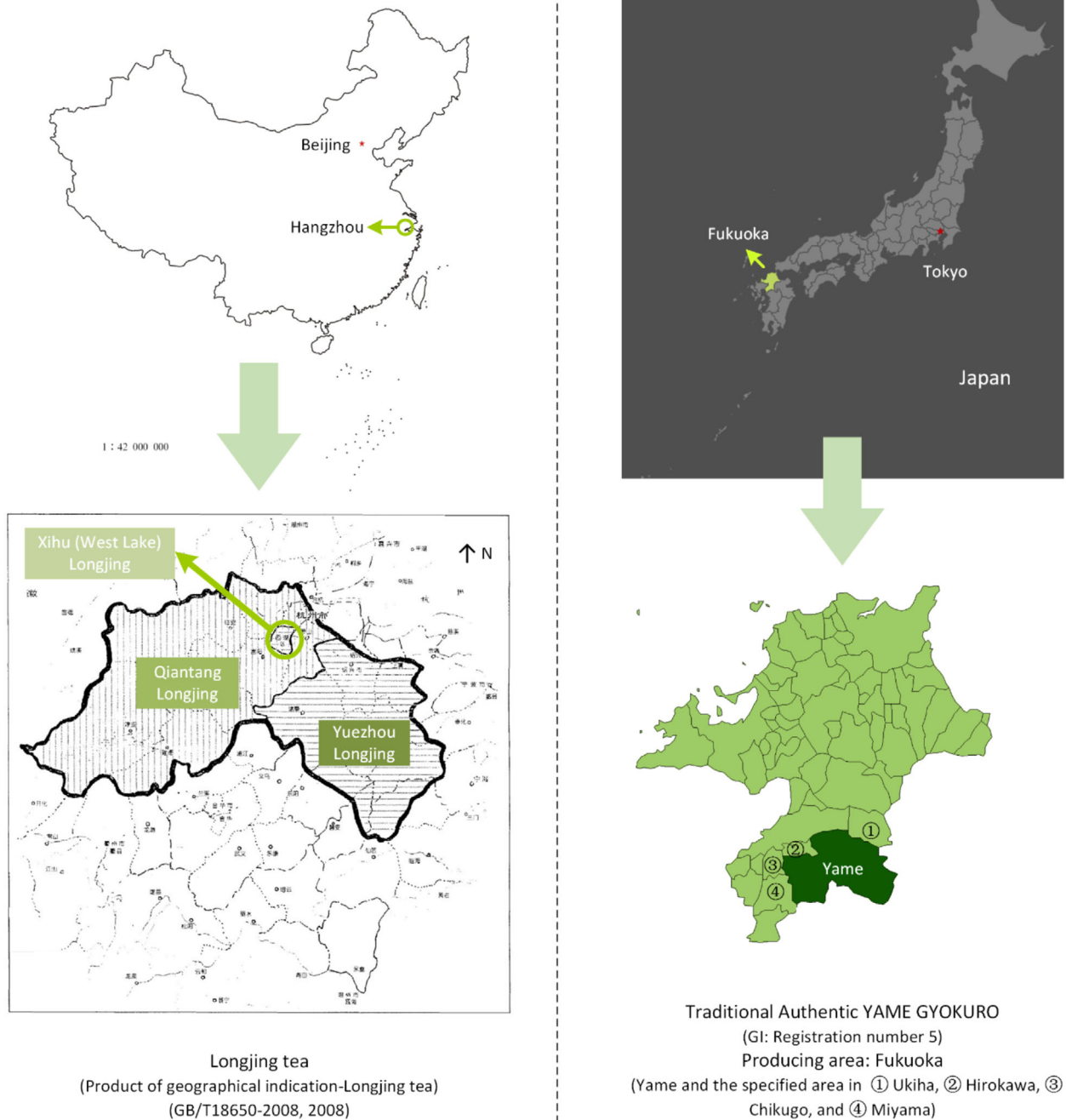


FIGURE 6 The geographical origins of Longjing tea and the producing area of traditional authentic Yame Gyokuro

discriminate green tea from other categories of teas (Reich et al., 2006); with advantages of rapid, cost-effective, and environmentally friendly in comparison to other separation methods, HPTLC demonstrates its capacity in the identification of commercial teas with different categories, though, at this stage, HPTLC could identify a sample, with certainty, as green tea or discriminate it from samples.

## 5 | SAFETY EVALUATION OF TEA PRODUCTS

In addition to conventional contaminants, lead chrome green in inorganic contaminants and sibutramine in organic contaminants are two distinctive contaminants in tea products. The former belongs to industrial pigments,

and its illegal addition makes commercial tea of lower grade, which is usually more yellowish than commercial tea of higher grade, and looks greener to deceive consumers for profit; the latter, sibutramine, is an appetite suppressant that works on the central nervous system, which is added to achieve the purpose of weight loss; however, it has been associated with increased cardiovascular events and strokes (James et al., 2010) and has been withdrawn from the market in several countries and regions, including Canada (Canada Health, 2010), China (China Food and Drug Administration, 2010), the European Union (European Medicines Agency, 2010), the United Kingdom (Medicines and Healthcare products Regulatory Agency, 2014), and the United States (Food and Drug Administration, 2010); therefore, the addition of sibutramine in tea products violates the standards on food safety of the above countries and regions.

## 5.1 | Inorganic contaminants

In previous years, the problem of the excessive residue of lead in tea products was the focus of tea product safety, and the MRL of lead varied among tea-growing and -consuming countries, which are 5 mg/kg in China (GB 2762-2017, 2017), the European Union (EU), and the United Kingdom, 10 mg/kg in Australia, Canada, and India, and 20 mg/kg in Japan (Karak & Bhagat, 2010). The standard lead detection method is relatively complicated to operate, and its LOD and LOQ are on the magnitude of mg/kg or mg/L. However, the LOD of applying nanozymes in heavy metal determination in commercial tea, which also requires digestion, reached the magnitude of ng/mL, as seen in Table 6, showing a considerable progress in comparison with standard detection methods. As more attention was given to the lead content in tea products, the residue of lead in tea products dropped below the MRL of China. Moreover, it is necessary to note that different tea products enter the human body in different ways; commercial teas need to be brewed or boiled, and are different from tea powder, which are consumed entirely by humans. Considering the above facts, the MRL of lead requires consideration separately, for which the extraction ratios or transition rates greatly reduce the content of lead extracted or translated from commercial tea into tea infusions.

Lead chrome green is a pigment prepared by precipitating lead chromate pigment onto Prussian blue pigment dispersion or mixing lead chromate pigment with Prussian blue pigments. The main components of lead chrome green are lead chromate and Prussian blue, and the proportion of lead chromate is not less than 50% (BJS 201910, 2019). At present, there is no limit and standard detection method for lead chrome green in national standards or

ISO standards. Until May 13, 2019, the General Administration of Market Supervision and Administration of China had issued the “Measurement of Lead Chrome Green in Tea” as a supplement inspection method. In this supplement inspection method, lead chromate is quantitatively determined by HPLC-ICP MS and ion chromatography. Compared with the application of Raman spectroscopy in Table 6, it is destructive, time-consuming, and reagent-consuming. The results of qualitative and quantitative measurements of lead chrome green through Raman spectroscopy were also satisfactory with  $R^2_p$ , RMSEP, and LOD of 0.876, 0.803, and 0.651 mg/g, respectively (Li et al., 2015).

Talcum powder is not harmful to the human body; therefore, it is added to food, medicine, and cosmetics to beautify the color and improve the taste or touch. However, because talc is a natural ore, unfiltered natural talcum powder may contain asbestos, which can be dangerous to humans if it enters the lungs, and is not suitable for direct use. Using FT-IR spectroscopy coupled with chemometrics to perform qualitative and quantitative detections of talcum powder added to commercial tea, not only achieved the predicted results of  $R^2_p$  of 0.927 and RMSEP of 0.137 (Li, Zhang, & He, 2016), but also provided guidance of the rapid detection of talcum powder in other foods, medicine, and cosmetics, revealing the application potential of FT-IR spectroscopy in the evaluation of talcum powder adulteration.

## 5.2 | Organic contaminants

Modifications of HPLC are the major emerging techniques used to determine PAHs in tea products, and the modifications are mainly concentrated at the sample pretreatment stage, as demonstrated in Table 6. Infusion in Table 6 indicates that water was utilized to brew commercial tea, while extraction means other solvents were used. QuEChERS, a solid-phase extraction method for the detection of pesticide residues in food, is also utilized in the detection of PAHs, with DAD (Ahmad et al., 2020; Garcia Londoño et al., 2015) and FLD (Garcia Londoño et al., 2015; Zachara et al., 2018) acting as the assistant. The principal purpose of these modifications is to speed up the detection process, reduce the interference of other substances contained in the sample, and improve the sensitivity and accuracy of the technique. In the existing research, the lowest LOD of PAHs is on the magnitude of ng/L (Loh, Sanagi, Wan Ibrahim, & Hasan, 2013; Viñas, Campillo, Aguinaga, Pérez-Cánovas, & Hernández-Córdoba, 2007).

Besides QuEChERS, UHPLC is more commonly applied in the determination of pesticides in tea products. Used as an alternative to HPLC, UHPLC, also called UPLC, has a greater chromatographic resolution and higher

sensitivity, as well as uses less solvent and time owing to faster analysis (Cielecka-Piontek, Zalewski, Jelińska, & Garbacki, 2013), which shows its characteristic advantages in determining multiple pesticides in tea products rapidly and simultaneously. All the above detection methods have good recoveries, and their LOD and LOQ are mostly of the magnitude of  $\mu\text{g}/\text{kg}$ , meeting the requirements of national standards. One of these researches applied polyvinylpyrrolidone (PVPP), a highly cross-linked modification of polyvinylpyrrolidone, to precipitate polyphenols, thus overcoming the matrix effects of tea in order to detect eight neonicotinoid insecticides (Guo et al., 2019), presenting an acceptable demonstration of determining pesticides in polyphenol-rich agricultural samples and worthy of further researches.

Quantification methods of temephos, fenitrothion (Fan et al., 2011), and six pesticides (Yue et al., 2008) in commercial teas are developed by HPTLC, and their LODs are of the magnitude of  $\text{ng}$  or  $\text{ng}/\text{mL}$ , lower than the MRL of pesticides, which not only confirming the feasibility of HPTLC in the quantification of pesticides in commercial teas, but also showing the potential of HPTLC as a complement to GC and HPLC in the determination of pesticides in complex matrixes, having its advantages of simplicity and rapidity.

The application of a GC-ICP-MS/MS method (Nelson et al., 2015) for the selective and sensitive detection of specific heteroatoms of phosphorus, sulfur, and chlorine has been accomplished, and commercial tea (leaf) as well as other food samples (baby food purees and fresh vegetables) are screened for its pesticide contents, following the preparation of triplicate extracts using QuEChERS. The correlation coefficient  $R$  of this application was greater or equal to 0.997, and the lowest LODs of the organophosphorus, organosulfur, and organochlorine pesticides were 0.0005, 0.675, and 0.144  $\mu\text{g}/\text{kg}$ , respectively. The potential of GC-ICP-MS/MS to be used for the ultra-trace analysis of P-, S-, and Cl-containing compounds is clearly presented in this application, and deserves the attention of researchers.

Surface-enhanced Raman spectroscopy or surface-enhanced Raman scattering (SERS), which enhances Raman scattering by molecules adsorbed on rough metal surfaces or by nanostructures, such as plasmonic-magnetic silica nanotubes, is applied to detect 4-mercaptopyridine, rhodamine 6G, and phosmet in commercial tea, based on capturing silver NPs onto the amino-modified (glycidyl methacrylate) –co–(ethylene dimethacrylate) capillary column (Pan et al., 2015). The primary advantages of using SERS for the detection of pesticides in tea products are that it is a simple, highly efficient, sensitive, and affordable technique. The simplicity of sample prepa-

ration, acceptable accuracy, and reliability make SERS columns suitable for on-site environmental monitoring and food analysis, as declared by researchers, with an LOD of 3  $\mu\text{g}/\text{L}$  and a linear response in the 3 to 1,000  $\mu\text{g}/\text{L}$  concentration range ( $R^2 = 0.995$ ) of phosmet, well below the tolerance level prescribed by the National Standard of China. Considering the merits of SERS, further research needs to be carried out to achieve the purpose of practical use, both from a broader range of pesticides and other organic contaminants.

Utilizing hexafluoroisopropanol (HFIP) as hydrogen-bond donor and L-carnitine/betaine as hydrogen-bond acceptor, a series of new hydrophobic deep eutectic solvents were synthesized to serve as extraction solvents for establishing the dispersive liquid–liquid microextraction method to extract pyrethroids, and results showed that after optimization by one-variable-at-a-time and response surface methodology, the enrichment factors were achieved to be 265 to 360 for five pyrethroids, as well as linear ranges of 0.25/0.5/1 to 100/200/400  $\text{ng}/\text{mL}$  ( $r \geq 0.9990$ ) and LOD of 0.06 to 0.17  $\text{ng}/\text{mL}$  (Deng et al., 2019). This novel, simple, rapid, highly efficient, and eco-friendly DLLME method is available for the extraction of pyrethroids in real tea beverages and fruit juices, providing new ideas for the rapid determination of pyrethroids, or extension to other pesticides.

In order to achieve the purpose of enhancing weight loss and seeking profits, sibutramine is illegally added to herbal slimming foods and supplements to deceive consumers (Ariburnu, Uludag, Yalcinkaya, & Yesilada, 2012; Huang, Xiao, Luo, Chen, & Yao, 2008; Popescu & Radu, 2015; Rebiere, Guinot, Civade, Bonnet, & Nicolas, 2012). Obviously, it is illegal and harmful to health and is required to be detected with the assistance of an effective, rapid, and reliable technique. Attenuated total reflection (ATR), a sampling technique used in conjunction with infrared spectroscopy, enables samples to be examined directly in the solid or liquid state without further preparation. This was combined with FT-IR and chemometrics to achieve this goal by determining sibutramine in green tea, green coffee, and mixed herbal tea (Cebi, Yilmaz, & Sagdic, 2017). Results demonstrated that unadulterated and adulterated samples were classified and discriminated with respect to their sibutramine contents with perfect accuracy without any false prediction. Since no solvents or reagents were used during the application of ATR-FTIR techniques, it could be mentioned as “green analytical technique,” bearing other virtues of cost-effective, rapid, easy to operate, and nondestructive, which makes it feasible and acceptable to utilize the ATR-FTIR technique for the detection of organic contaminants like sibutramine and inorganic contaminants.

## 6 | CHALLENGES AND FUTURE TRENDS

By simplifying the sample processing procedure, improving the sensitivity and accuracy of the analytes, and providing real-time detection results on the spot, emerging techniques, including spectroscopic techniques, electrochemical methods, nanozymes, computer vision, and modifications of chromatographic techniques, demonstrate their advantages in the detection of quality and safety parameters in tea products, and show good application prospects and broad application fields when compared with existing standard detection techniques. However, there are challenges of these emerging techniques that need to be solved by further research. The robustness and transferability of models is one of the main challenges of emerging techniques using chemometrics as an assistant, which is also one of the limitations that restrict the wide-scale application of spectroscopic techniques. Establishing an open-source database and uploading and sharing existing research data may play a useful role in promoting the performance of spectroscopic techniques. Increasing the diversity of samples, expanding their sources, and using multiple analytical methods, including spectral processing and model establishment, might be conducive for improving the performance of emerging techniques that are combined with chemometrics during their application, such as spectroscopic techniques, E-nose, E-tongue, and E-eye.

When considering the future trends of emerging techniques for quality and safety determination of tea products, it can be considered from the following three aspects: production, purchase, and detection. For the first aspect, production, the vast majority of application of emerging techniques for the determination of tea product quality in current research is focused on processed tea products, such as commercial tea and tea beverages, and the real-time detection of the corresponding parameters during the picking and processing of tea leaves is rarely seen. Nevertheless, it is of great importance because the use of emerging techniques for rapid, real-time, nondestructive, or slightly-destructive determination during the picking and processing of tea leaves will not only eliminate the strong dependence on professionals, but also help mechanize the picking and processing of tea leaves, making the tea industry considerably more mechanized. For the second aspect, purchase, emerging techniques will play a significant role in the real-time evaluation of quality, such as the geographical origin and the grade of commercial tea because the quality is closely related to price, and the consistency between price and grade is the chief point considered by consumers when purchasing renowned teas. Researchers can develop portable devices that can dif-

ferentiate commercial tea of distinct quality and display the evaluation results. The third aspect, detection, can be divided into laboratory and field detection, for which their focuses are different. Laboratory detection places greater emphasis on accuracy, while field detection places greater emphasis on efficiency and portability. Achieving rapid, nondestructive, real-time, and on-site detection of some of the valuable quality and safety parameters, such as the presence of caffeine, lead, and pesticides in tea infusions or tea beverages, is one of the developmental directions of emerging techniques for field detection. Overall, spectroscopic techniques, electrochemical methods, nanozymes, computer vision, and modifications of chromatographic techniques are expected to be useful and trustworthy techniques for rapid and efficient detection of the quality and safety parameters of tea products, and offer guidance for their applications to other foods and beverages.

## 7 | CONCLUSIONS

In this review, the recent applications of spectroscopic techniques, electrochemical methods, nanozymes, computer vision, and modifications of chromatographic techniques, acting as emerging techniques, have been discussed and underlined as functional techniques for quality and safety determination of tea products, covering various parameters, including physical, chemical, microbiological, and inorganic and organic contaminants. Combined with chemometrics, spectroscopic techniques are widely used to determine the contents of chemical constituents, adulterations, classified parameters, inorganic contaminants, and organic contaminants, while voltammetry in ECD is applied for the determination of chemical parameters, and electronic sensing mostly evaluates the quality of tea products by evaluating classified parameters. Applications of nanozymes are relatively less, but they perform satisfactorily for determining chemical constituents and heavy metals. Computer vision is capable of differentiating commercial tea with different grades and categories, and organic contaminants are mainly determined through modifications of chromatographic techniques. Considering the strong reliance on the expertise of tea professionals in the picking, processing, and sensory evaluation of tea, along with the strong attention of consumers toward the quality of commercial tea, the application of the above techniques in the entire process of commercial tea production, as well as the development of portable devices for the rapid, real-time, and on-site determination of classified and safety parameters requires further research. In light of the continuous understanding and progress in the analysis of multiple variables, the

development of portable devices, and modifications of current techniques, it is undoubtful that these emerging techniques will help in the quality and safety determination of a broader variety of foods and beverages, not only in laboratory detection but also in field detection.

## CONFLICTS OF INTEREST

There are no conflicts of interest to declare.

## AUTHOR CONTRIBUTIONS

Sun and He proposed the outline and reviewed the manuscript. Yu researched prior studies, interpreted the results, and drafted the manuscript.

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