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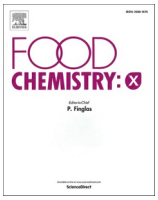

A comprehensive review of the current trends and recent advancements on the authenticity of honey

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ABSTRACT

The authenticity of honey currently poses challenges to food quality control, thus requiring continuous modernization and improvement of related analytical methodologies. This review provides a comprehensively overview of honey authenticity challenges and related analytical methods. Firstly, direct and indirect methods of honey adulteration were described in detail, commenting the existing challenges in current detection methods and market supervision approaches. As an important part, the integrated metabolomic workflow involving sample processing procedures, instrumental analysis techniques, and chemometric tools in honey authenticity studies were discussed, with a focus on their advantages, disadvantages, and scopes. Among them, various improved microscale extraction methods, combined with hyphenated instrumental analysis techniques and chemometric data processing tools, have broad application potential in honey authenticity research. The future of honey authenticity determination will involve the use of simplified and portable methods, which will enable on-site rapid detection and transfer detection technologies from the laboratory to the industry.

1. Introduction

According to the *Codex alimentarius*, honey is defined as "the natural sweet substance produced by honey bees from the nectar of plants or from secretions of living parts of plants or excretions of plant-sucking insects on the living parts of plants, which the bees collect, transform by combining with specific substances of their own, deposit, dehydrate, store and leave in the honey comb to ripen and mature" (Codex Standard, 2001). Thus, honey is a high-value food product widely consumed for its unique flavor, high nutritional value and health benefits, which vary according to its chemical composition. Honey is produced and marketed worldwide with remarkable differences in nutritional value and biological importance. Functional and health-promoting properties of honey are mainly associated with its antioxidant, antibacterial, anti-inflammatory properties, which have been used for therapeutic purposes (X.-H. Zhang, Qing, et al., 2021; X.-H. Zhang, Wang, et al., 2021). Other promising biological properties of honey have also been reported, including but not limited to anticancer, anti-HIV, and wound dresser (Geana & Ciucure, 2020; Tsagkaris, et al., 2021). These biological activities are associated with the composition and content of honey compounds, including flavonoids, phenolic acids, enzymes, ascorbic acid, carotenoids, pigment, and alkaloid, among others.

Honey has unique flavor and above remarkable health-promoting properties, thus verifying its authenticity is the focus of the global honey industry. However, current quality standards and analytical methods are still poorly effective for the determination and evaluation of honey authenticity, which creates challenges for the honey market (China Standard, 2011; Council Directive, 2001; Codex Standard, 2001; Tsagkaris, et al., 2021; L. Wu, et al., 2017). Herein, we discuss the current issues faced by the honey industry related to honey authenticity verification, expounding the difficulties existing in the current detection and market supervision. Direct and indirect methods of honey adulteration were described in detail. In addition, current methods for honey authenticity confirmation are comprehensively reviewed. Furthermore, the most recent advancements in honey authenticity verification are described, with an emphasis on their advantages, disadvantages. Finally,

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innovation trends and novel solutions are discussed, aiming to provide a theoretical guide for honey quality control research and foster the development of the honey industry.

2. Honey authenticity and related issues

Honey is a natural sweetener produced by honeybees, through collecting nectar, secretions, and/or honeydew from plants, which is then mixed with their own secretions, transformed, dehydrated, stored, and brewed. Thus, the addition of known or unknown additives to honey is prohibited (China Standard, 2011; Council Directive, 2001; Codex Standard, 2001; Tsagkaris, et al., 2021; L. Wu, et al., 2017). Herein, we discuss the current issues fac; Codex Standard, 2001). However, the practice of honey adulteration is an ongoing global problem, which has been causing a notorious negative impact on the development of the honey industry. Honey authenticity issues have been reported and summarized, being thus divided into two categories (Fig. 1): i) direct methods related to honey adulteration with addition of sweeteners or mixed honeys, as well as incorrect description as high-quality honey of geographical and botanical origins and organic products; ii) indirect methods related to feeding honeybee colonies, harvesting unripe honey, as well as inappropriate handling and storage.

2.1. Direct methods of honey adulteration

Direct methods of honey adulteration are mainly based on the addition of water, sucrose, invert sugar, caramel, carboxymethyl cellulose (CMC), and dextrin or starch substances. The addition of the above-mentioned substances to honey enables adulterate and increase yield. It has been reported that boiling sucrose could closely resemble natural hues of pure honey by adjusting temperature during heating (X. Zhang, Zhang, Qing, & Lu, 2019). Ciursă et al. developed methods for determining three types of authentic honeys (tilia, sunflower and acacia) were adulterated with inverted sugar in different percentages (5%, 10%, and 20%) using Fourier-transform infrared spectroscopy (FTIR) spectroscopy and multivariate analysis (Ciursă, Pauliuc, Dranca, Ropciuc, & Oroian, 2021). In addition, quality and sweetness of honey mixed with water decrease, being thus prone to fermentation and deterioration. In contrast, honey mixed with sucrose has a relatively flat taste compared to natural honey, which has a sucrose taste and strong transparency. Similarly, honey mixed with caramel has dull glaze and decreased transparency. Moreover, the addition of starch paste substances makes honey crystallize, become rough, and unable to form long "honey silk". The incorporation of CMC increases viscosity and generates transparent agglomerates in honey products. Collectively, the above-mentioned honey adulteration methods can be easily identified by sensory evaluation or relevant honey standards.

In recent years, raw materials and methods used in honey adulteration have become increasingly sophisticated. It is common to use relevant honey standards as a technical reference for performing adulteration, and common honey adulterants include various syrups, of which the composition is similar to honey, such as high-fructose corn syrup (HFCS), corn syrup (CS), maltose syrup (MS), sucrose syrup (SS), rice syrup (RS), among others (Akyıldız, et al., 2022; Cagliani, Maestri, & Consonni, 2022; Huang, Chuang, Kung, & Hsieh, 2021; S. Li, et al., 2017; Qu, et al., 2019; S. Wang, et al., 2015). Among these, HFCS is the most important syrup-based compound used in honey adulteration. According to its fructose content, commercial HFCS is commonly categorized into three types: F42 (42% fructose, 53% glucose), F55 (55% fructose, 42% glucose), and F90 (90% fructose, 10% glucose). In contrast, CS is composed predominantly by glucose and maltooligosaccharides (G2–G16), whereas maltose content in MS falls within the range of 40–60%. Since the ratio of fructose to glucose in these syrups is very close to that in natural honey (1.0–1.2 for pure honey), flavor of honey adulterated with syrups is not only not destroyed, and certain physical and chemical indicators increase. Furthermore, in order to maintain sensory quality of adulterated honey, synthetic pigments and honey flavors are added, which closely reproduce the levels of flavor compound in natural honey (L. Wu, et al., 2017). The continuous refinement of adulteration strategies constitutes a great challenge to

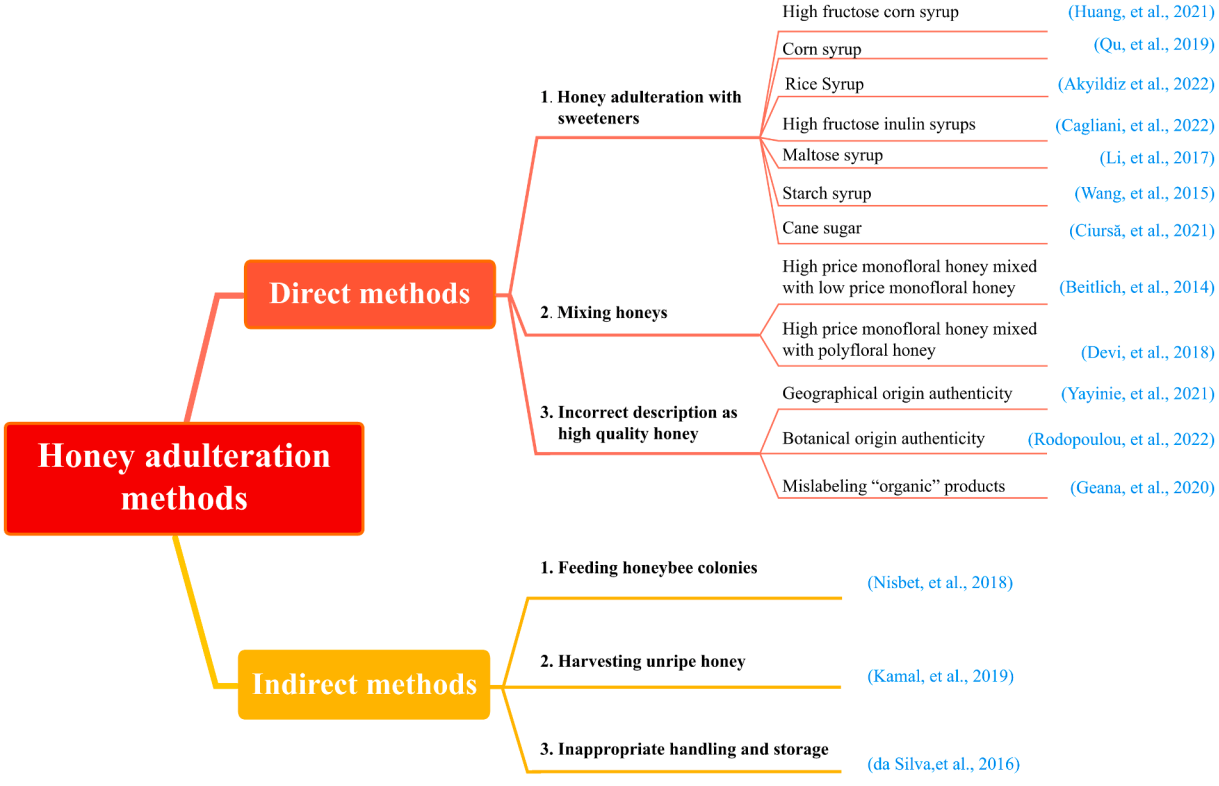

Fig. 1. Schematic representation of the main adulteration methods related to honey.

honey adulteration verification, making it difficult to identify the authenticity based on traditional sensory evaluation and physical and chemical analyses.

Another direct method of honey adulteration is by adding low-quality monofloral or multifloral honey to valuable monofloral honey (Beitlich, Koelling-Speer, Oelschlaegel, & Speer, 2014; Devi, Jangir, & Anu-Appaiah, 2018). The monofloral honey is predominantly from one nectar source, whose pollen proportion should be above 45% for most species, which possesses distinctive organoleptic characteristics, like highly distinguishing aromas, and thus is considered as a premium product (Machado, Miguel, Vilas-Boas, & Figueiredo, 2020; Soares, Amaral, Oliveira, & Mafra, 2017). Contrarily, multifloral honey is obtained when bees collect nectar from different types of flowers, with pollen grains from several plant species, none of which are considered predominant. In recent years, there has been an increasing consumer demand for monofloral honey, which may reach a higher market value than multifloral one due to its particular flavor, taste, and pharmacological properties (Geana & Ciucure, 2020; Tsagkaris, et al., 2021). These honey adulteration methods have been reported worldwide, e.g., decolorization of cheap rape honey to resemble the high-priced acacia honey (Se, Wahab, Syed Yaacob, & Ghoshal, 2019). Other direct honey adulteration methods include confusing labeling, such as misleading geographical and botanical origins and/or organic certification (Geana & Ciucure, 2020; Rodopoulou, et al., 2022; Yayinie, Atlabachew, Tesfaye, Hilluf, & Reta, 2021). According to the geographical origin of production, honey can be linked with specific areas under the labels of Protected Designation of Origin (PDO) and Protected Geographical Identification (PGI) (Machado, et al., 2020; Tsagkaris, et al., 2021). A comprehensive list of the PDO, PGI and "organic" honey products can be found on the website https://ec.europa.eu/info/food-farming-fisheries/food-safety-and-quality/certification_en. The monofloral, PDO, PGI and "organic" honeys, are highly connected with their price, consequently, producers may be tempted to describe different origins and fraudulently mix with lower value and quality honey to increase profit.

2.2. Indirect methods of honey adulteration

An indirect method of honey adulteration involved overfeeding honeybee colonies with sucrose and industrial sugar syrups in the main nectar flow period (Nisbet, Kazak, & Ardali, 2018). In this way, sugars are introduced into honey via a natural process, which negatively impacts consumers and pure honey producers, thus influencing the honey market. Extreme and abnormal climate conditions also severely impact honey production, causing a drastic decrease. For instance, the flowering period is shortened at lower temperatures, and excessive rain and wind affect bees' activities, and planting area of nectar plants has become increasingly smaller, among other factors. Thus, in order to increase honey production, bees are fed with sucrose or syrups. It has been demonstrated that long-term bee-feeding with syrups modifies sugar composition of resulting honey (Nisbet, et al., 2018). Adulterated honey obtained from honeybee colonies fed with different levels of sugar syrups (C_3 and C_4 plants) could be identified by carbon isotope ratio analysis (Guler, et al., 2014). In addition, ultra-high performance liquid chromatography time-of-flight mass spectrometry (UHPLC-TOF-MS/MS) enable the identification of adulteration markers in honey obtained from bees fed with different sugar syrups, among which can be cited 2-acetylfuran-3-glucopyranoside and sorbic acid (Akyıldız, et al., 2022).

Another indirect method of honey adulteration is to use unripe honey to impersonate capped ripe honey. Generally, it takes approximately 7–15 days for bees to brew thin nectar into capped ripe honey. Ripe honey has a relatively high Baume degree, and its water content is below 20% (except for heather honey whose maximum water content is 23%) (China Standard, 2011; Council Directive, 2001; Codex Standard, 2001; Tsagkaris, et al., 2021; L. Wu, et al., 2017). Herein, we discuss the current issues fac; Siddiqui, Musharraf, Choudhary, & Rahman, 2017; Codex Standard, 2001). Unripe honey is obtained if honey is processed prior to reaching its natural maturity, and often has higher water content (above 20%) and lower Baume degree. Thus, unripe honey has low nutritional value and is easily fermented and deteriorated, thus having shorter shelf life. Therefore, honey moisture is a commonly used parameter to confirm honey quality and estimate market value. Chinese and EU honey quality standards stipulate that sucrose content in honey should be below 59/100 g. In addition, unripe honey has a higher sucrose content, and sucrose was not completely converted to glucose and fructose by invertase, thus it can be hypothesized that honey has added exogenous sugars (Kamal, et al., 2019). The gravimetric method can be used to determine honey moisture content, and as an alternative method, the refractive index measurement and Karl Fischer titration can also be used (Geana & Ciucure, 2020). Moreover, proline content in honey is regarded an indicator of honey ripeness, and values below 180 mg/kg are attributed to unripe or adulterated honeys according to the standards preconized by the International Honey Commission.

Due to the low Baume degree of unripe honey, which does not meet honey quality standards, high-temperature concentration equipment is often used to evaporate excess water and concentrate honey for subsequent commercialization. However, honey cannot be thermally treated, such as with high-temperature concentration and pasteurization, since these may cause changes in honey nutritional composition, resulting in the loss of polyphenols and volatile compounds, the reduction of diastase activity, among others. Honey should be sealed and stored in a cool, dry and ventilated place protected from direct sunlight (da Silva, Gauche, Gonzaga, Costa, & Fett, 2016). Freshness and suitability of storage and processing conditions of honey can be assessed by determining the content of 5-hydroxymethylfurfural (HMF) in honey and amylase activity (Geana & Ciucure, 2020). HMF is not intrinsically found in honey, but is produced after treatment at high temperatures, as a result of the transformation of $C_6H_{12}O_6$ in an acid-catalyzed dehydration process. The content of HMF in fresh honey is very low (below 40 mg/kg), but after storage under inappropriate conditions or for a prolonged time, its content will increase (China Standard, 2011; Council Directive, 2001; Codex Standard, 2001; Tsagkaris, et al., 2021; L. Wu, et al., 2017). Herein, we discuss the current issues fac; Siddiqui, et al., 2017; Codex Standard, 2001). HMF can be quantified by high-performance liquid chromatography (HPLC) and spectrophotometric methods as a means for evaluating honey quality and freshness. In addition, HMF can be potentially used as a marker for the adulteration of honey with commercial industrial sugar syrups. Moreover, the diastase uniquely found in natural honey derived from bees, can also be used as an important indicator for evaluating the suitability of storage and processing conditions.

In summary, there are various methods of honey adulteration, which poses great difficulties for market regulation. Honey authenticity is hence the focus of the global apiculture market, which currently represents a challenge for food quality control. Therefore, it is necessary to continuously modernize and improve related analytical methodologies based on the physical, chemical, and biological characteristics of honey.

3. Methods for honey authenticity verification

Due to the factors such as nectar sources, geography/botanical origin, and collection conditions, honey is a complex matrix with varying types and contents of active components, thus creating difficulties for honey authenticity confirmation. It is challenging to effectively ensure that "pure honey" is the natural ripe capped honey of the geographical and botanical origin. Therefore, scientific methods should be established to identify the authenticity of honey, and solve the problems of confusion of varieties, low quality, and adulteration in the honey market. These would in turn foster commercialization of green, safe and healthy pure honey, thus promoting the development of the honey industry.

Detection methods for honey authenticity include physicochemical and sensory analysis, spectroscopy, chromatography, electrochemical

methods, bioanalytical methods, among others. These methods can be classified into three categories: i) conventional analytical methods, including sensory analysis, physicochemical characterization, melissopalinological analysis, etc., can be combined to preliminarily judge of the authenticity of honey, is sometimes not sufficient (Juan-Borrás, Periche, Domenech, & Escriche, 2015; Rodopoulou, et al., 2022; Sharin, et al., 2021); ii) use of large-scale instruments (such as multi-stage mass spectrometry) to comprehensively analyze relevant indicators in honey samples. These methods are mainly based on the determination of volatile compounds, flavonoids, phenolic acids, metal elements and isotopes, etc., and then realize the quality identification of honey (Kato, et al., 2014; Kazalaki, Misiak, Spyros, & Dais, 2015; Osés, et al., 2020). However, they involve costly instruments, time-consuming analysis, expensive reagents, and high costs of operation and maintenance; iii) chromatography or spectroscopy combined with chemometric first-order pattern recognition methods for modeling analysis, such as principal component analysis (PCA), partial least squares (PLS), support vector machine (SVM), among others. These technologies have been widely applied in honey authenticity determination, such as identification and quantification of different adulterants in high-quality honey, geographical and botanical classification of honeys, and identification of unripe honey, etc. (Silva, et al., 2021; Valinger, et al., 2021; Yong, et al., 2022). Among these methods, chemometric tools are mainly used to mine useful information from complex data array, thus providing a novel research strategy for determining honey authenticity. However, the results obtained from these above pattern recognition methods based on chromatographic and spectral information data are a linear combination of the real spectrum of each component, having no relevant physical relevance. The following introduces to the main identification methods of honey authenticity developed in recent years, with a focus on their advantages, disadvantages, and scopes.

3.1. Current criteria of honey and conventional analysis

Similar to other food products, honey quality has to be objectively evaluated by physical, chemical and biochemical methods according to reliable standards. Several relevant international organizations and countries, such as the International Honey Commission, *Codex Alimentarius*, EU Honey Directive, and China National Health Commission, have formulated the quality standards for honey (China Standard, 2011; Council Directive, 2001; Codex Standard, 2001; Tsagkaris, et al., 2021; L. Wu, et al., 2017). Herein, we discuss the current issues fac; Siddiqui, et al., 2017; Codex Standard, 2001). These relevant standards comprise a series of conventional physical and chemical methods, such as moisture, sugar composition, acidity, ash content, electrical conductivity, diastase activity and HMF content to evaluate honey quality. Table 1 shows honey composition based on the Chinese national standards and the EU Directive 110/2001, which indicates that China and EU preconize similar standards for the contents of fructose and glucose, sucrose, moisture, and HMF, but differ in terms of acidity and diastase activity. Although these two standards do not specify the contents of protein, free amino acids, vitamins and elements, as the inherent nutritional components of honey, they should also be included in honey quality control indicators. As a result of refinement of honey adulteration processes, it has become possible to use various non-honey-derived raw materials to produce an "indicator honey" which fully meets the requirements of relevant standards. Therefore, current honey standards cannot be considered a reliable guarantee for honey quality and safety. Currently, the development of detection methods for honey authenticity verification in samples potentially adulterated in various ways has become a research hotspot.

Based on current criteria of honey, the conventional analyses have been widely used to ensure honey authenticity for their direct approach, simple execution, and low cost (Table 2). Honey quality can be evaluated through sensory attributes such as color depth, luster, viscosity, smell, taste, among others. Among these, smell and taste are often the main attributes for identifying honey authenticity. In fact, it is possible to identify by smell and taste high-priced monofloral honey mixed with large amounts of syrup and/or low quality monofloral or multifloral honey. However, it can be challenging to identify adulterated honey that closely resemble pure honey in terms of color, consistency, odor, and taste (Price, Tang, El Kadri, & Gkatzionis, 2019). Moreover, honey authenticity identification requires well-trained and experienced sensory panelists and statistical evaluation of the observations, and the sensory experience is a key factor to ensure results reliability. Therefore, sensory analysis is only suitable for preliminary judgments and should not be used as a definitive assessment of honey authenticity.

During honey collection, bees bring pollen particles to the hive and mix with honey. Thus, honey can be authenticated by identifying and quantifying the percentage of pollen (Juan-Borrás, et al., 2015). However, this melissopalynological analysis alone is sometimes not sufficient, which is highly susceptible to variation due to the influence of factors such as honey collection methods, nectar source, environmental factors, seasons, geographical location, and the morphology and quantity of pollen (Addi & Bareke, 2021; Rodopoulou, et al., 2018). In

Table 1
The composition of honey in Chinese national standard and EU Directive 110/2001.

Honey parameters	Concentration
Chinese national standard	
Moisture content	
litchi, longan, citrus, schefflera and tallow honeys	≤23% w/w
others	≤20% w/w
Fructose and glucose content	≥60% w/w
Sucrose content	
eucalyptus, citrus, alfalfa, litchi and wild osmanthus honeys	≤10% w/w
others	≤5% w/w
Acidity (1 mol/L NaOH)	≤40 mL/kg
w(HMF)/	≤40 mg/kg
Diastase activity (1% starch solution)/	
litchi, longan, citrus, and schefflera honeys	≥2 mL/(g·h)
others	≥4 mL/(g·h)
Ash content	≤0.4% w/w
Zn content	≤25 mg/kg
EU Directive 110/2001	
Fructose and glucose content (sum of both)	
blossom honey	≥60% w/w
honeydew honey, blends of honeydew honey with blossom honey	≥45% w/w
Sucrose content	
in general	≤5% w/w
false acacia, alfalfa, Menzies Banksia, French honeysuckle, red gum, leatherwood and Citrus spp.	≤10% w/w
lavender, borage	≤15% w/w
Moisture content	
in general	≤20% w/w
heather an baker's honey in general	≤23% w/w
bake's honey from heather	≤25% w/w
Water-insoluble content	
in general	≤0.1% w/w
pressed honey	≤0.5% w/w
Electric conductivity	
in general	≤0.8 mS/cm
honeydew honey, chestnut honey, and their blends with other honeys except strawberry tree, bell heather, eucalyptus, lime, ling heather, Manuka or Jelly Bush, tea tree	≥0.8 mS/cm
Free acid content	
in general	≤50 mequiv/kg
baker's honey	≤80 mequiv/kg
Diastase activity	
in general except baker's honey	≥8 (Schale units)
honeys with low natural enzyme content and HMF content ≤15 mg/kg	≥3 (Schale units)
w(HMF)	
in general except baker's honey	≤40 mg/kg
honeys of declared origin from the regions with tropical climate and their blends of these honeys	≤80 mg/kg

Table 2
Some literatures in the past three years relating to the conventional analysis techniques used for monitoring honey authentication issues.

Honey	Authenticity issue	Analytical parameters	Chemometric tools	References
Bracatinga honeydew honeys and blossom honeys from Brazil	Differentiation of honeydew honeys and blossom honeys	Colour analysis	PCA	(Bergamo, et al., 2019)
Fir, chestnut, pine, wild flower, orange blossom, wild thyme from Greek	Evaluation the sensory perception of honey by flash profile and influence of culture	Moisture content and Brix values, pH, rheological properties, apparent viscosity, sensory analysis	/	(Price, et al., 2019)
Evernonia polysphaera, ucalyptus, Citrus, Serjania lethalis, Croton heliotropiifolius, Mimosa scabrella, Wild and Cydonia oblonga honeys from Brazil	Honey authentication	Moisture, pH, ash content, free acidity, diastase activity, HMF, Brix, electrical conductivity and insoluble matter, Color, glucose, fructose, sucrose and fructose/glucose ratio, estimated reducing sugars and total sugar content, Rheological analysis	PCA, CA	(Pereira, et al., 2020)
The imported and locally produced honey from different regions South African	Honey authentication	Sugars (fructose, glucose, sucrose and maltose), reducing sugars (sum of fructose, glucose and maltose), fructose/glucose ratio, pH, total acidity (free acid and lactone), moisture, ash, Lund's precipitate, HMF, and specific rotation	/	(De Beer, et al., 2021)
Cotton, chestnut, sunflower, honeydew, citrus, and canola honey from Turkey	Botanical and geographical origins	Sugar content, phenolic profile, total phenolic content, CIE-color values (L*, a*, b*), oxygen radical absorbance capacity, and Trolox equivalent absorbance capacity	CA, PCA, DA, ANN	(Sogut, et al., 2020)
The fresh honeys from Ethiopia	Honey authentication and geographical origin	Moisture, ash content, pH, free acidity, electrical conductivity, total reducing sugar content, sucrose content, HMF	PCA, LDA	(Yayinie, et al., 2021)
The stingless bee honeys from Malaysia	Discrimination of Malaysian stingless bee honey from different entomological origins	pH, moisture content, ash, total soluble solid, electrical conductivity and volatile compound profiles	PCA, HCA, PLS-DA, SVM	(Sharin, et al., 2021)
The honeys from Zambia and Botswana	Honey authentication	Specific conductivity, moisture/ash contents, pH, acidity, sucrose, fructose, and glucose	PLS	(Padiso, et al., 2021)
Thyme, autumn pine, spring pine, chestnut, knotgrass, strawberry tree, jerusalem thorn, fir, oak, sunflower, cotton, erica, clover, citrus and judas tree honeys from Greece	Botanical origin	Water content, electrical conductivity, HMF content, diastase activity, colour, CIELab parameters, pH and free acidity	MANOVA, PCA, MDA	(Rodopoulou, et al., 2022)

[a]Chemometric tools: cluster analysis (CA), principal component analysis (PCA), hierarchical clustering analysis (HCA), linear discriminant analysis (LDA), partial least squares (PLS), partial least square-discriminant Analysis (PLS-DA), soft independent modeling of class analogy (SIMCA), multivariate analysis of variance (MANOVA), artificial neural network (ANN), support vector machine (SVM), multi-discriminant analysis (MDA).
[b]hydroxymethylfurfural: HMF.

addition, it might be further challenging to identify adulterated or completely fake honey if pollen particles are artificially added to it.

The physicochemical parameters can directly reflect honey quality, being thus used as quality control parameters (Price, et al., 2019). Among these, the most relevant parameters are including color, density, viscosity, rheology, moisture content, sugar composition, pH, acidity, mineral content, ash content, HMF content, electrical conductivity, optical rotation, diastase activity, among others. These parameters can indirectly reflect the authenticity of honey, so they can be used as evaluation parameters for judging the authenticity of honey (Padiso, Keiphetlhetswe, Donald Chinyama, Molwantwa, & Sichilongo, 2021; Rodopoulou, et al., 2022). As moisture content in honey increases, density and viscosity decrease, whilst rheology increases (Pereira, et al., 2020). Some changes in density, viscosity and rheology also occur in honeys when mixed with sugar or industrial syrups (Price, et al., 2019). In addition, color analysis is a valuable parameter for evaluating monofloral honey, since honey from different sources of nectar plants has varying colors (Sogut & Seydim, 2020). Color in adulterated honey changes compared to natural honey and can be assessed using a colorimeter. However, there are many varieties of honey with different colors. In addition, the color of natural honey changes with prolonged storage, thus color judgment cannot accurately determine honey authenticity.

Furthermore, considering that different types of honey have specific optical rotation (generally levorotatory), which changes in honey with different added sugars or even becomes dextrorotatory, being thus useful in honey authenticity determination. Besides, considering that diastase is a bee-derived amylase uniquely found in natural honey, adulterated honey can be identified by measuring stability and activity of diastase. Diastase content of natural honey is generally above 8.0 (China Standard, 2011; Council Directive, 2001; Codex Standard, 2001; Tsagkaris, et al., 2021; L. Wu, et al., 2017). Herein, we discuss the current issues fac; Siddiqui, et al., 2017; Codex Standard, 2001). For the adulterated honey mixed with starch or sucrose, diastase will convert the doping substance into reducing sugar, thus resulting in the value decrease. However, due to the difference of diastase content in natural honey from different sources, it is thus difficult to establish an accurate threshold value for diastase content to be used in the identification of honey authenticity. Moreover, the addition of artificial amylase to boost values and/or isozyme profiles optimization will dampen the ability to distinguish adulterated honey from pure honey. In addition, polyphenols in honey can also be used as a marker to identify honey adulteration combined with chemometrics methods, and a predictive model can be established to distinguish adulterated honey with different nectar sources or syrups (X.-H. Zhang, Qing, et al., 2021; X.-H. Zhang, Wang, et al., 2021).

Sensory characteristics and the amount of pollen of honey varies greatly due to the influence of many factors on bee honey collection environment, and color, aroma, and physicochemical parameters of honey will also change to a certain extent during storage and processing. Moreover, current adulterated honey resembles greatly pure honey in color, taste and properties. Consequently, conventional adulteration detection methods have gradually been abandoned, being thus used exclusively as a preliminary reference. A possibly more efficient strategy for determining honey authenticity could be to combine conventional analytical methods with chemometric analysis. For instance, honey from Malaysian stingless bees could be distinguished from different entomological origins based on physicochemical properties and volatile

compound profiles using chemometrics and machine learning (Sharin, et al., 2021). Moreover, authentication of honey from Spanish avocado (*Persea americana* Mill.) was determined based on its composition, mineral content and sensory attributes (Rodríguez, Cámara-Martos, Flores, & Serrano, 2019). Furthermore, Karabagias et al. showed that antioxidant activity was positively and significantly correlated ($p < 0.05$) with the contents of total phytochemicals, copper, and iron, as well as with color intensity and pH in honey of botanical origin (Karabagias, Karabagias, Papastephanou, & Badeka, 2020).

3.2. High-throughput analysis

Honey is a complex matrix which contains at least 200 compounds among volatile, semi-volatile, and non-volatile compounds, being also a supersaturated solution of sugars mainly composed of glucose and fructose (65–80% of total soluble solids). Therefore, honey authentication is a complex topic and requires an integrated experimental workflow that ideally should involve a carefully selection of the most appropriate sample processing protocols, instrumental methods, reliable chemometric tools, and unambiguous metabolite identification (X.-H. Zhang, Qing, et al., 2021). Sample preparation and related extraction techniques is a critical step which often directly impacts the quality of analytical results and the duration of experiments. Most importantly, instrumental analysis is at the core of honey quality control. The use of emerging technologies in honey quality determination enables more accurate, fast, reliable and convenient results. In this context, chromatography, spectroscopy, electrochemistry, biosensing and other technologies have been widely applied in honey authenticity determination, such as identification and quantification of different adulterants in high-quality honey, geographical and botanical classification of honeys, and identification of unripe honey, etc. Moreover, chemometric approaches can enable mining significant information from complex data array, thus providing a novel research strategy for determining honey authenticity. Thus, improving sample extraction techniques, instrumental analyses, and more robust chemometric approaches will allow a more comprehensive analysis of honey authenticity.

3.2.1. Sample extraction techniques

The sample preparation is a crucial step in honey authenticity analysis, and the quality of the analytical results is often directly related to the extraction technique employed in the experimental layout. Honey background matrix is complex and is a supersaturated solution of sugars, but with a low content of active components. Therefore, compared with other food matrices, efficient sample extraction methods are especially important for the separation of interferents in honey samples, such as carbohydrates, proteins, and enzymes. In addition, a more efficient sample pretreatment enriches the method selectivity and sensitivity, protects the analytical columns and decreases the matrix effect, allowing the collection of better results (Medina, Perestrelo, Silva, Pereira, & Câmara, 2019). Extraction techniques currently applied in honey quality control analysis mainly include liquid–liquid extraction (LLE), solid phase extraction (SPE), column chromatography (CC), microwave-assisted extraction (MAE), ultrasound-assisted extraction (UAE), and stir bar sorptive extraction (SBSE) (Fig. 2). In addition, considering recovered yield, cost, solvent consumption, and environmental impact, various improved sample extraction methods have developed, including liquid–liquid microextraction (LLME), solid-phase microextraction (SPME), aqueous two-phase extraction (ATPS), ionic liquids (ILS), mixed-mode column chromatography (MM-CC), surface-modified column chromatography (SM-CC), and others.

LLE is a generic approach that involves analytes partitioning between two immiscible liquid phases, i.e., organic and aqueous phase. LLE has been widely used in honey sample pretreatment, and ethyl acetate is often used as the extractant since it can extract non-polar analyte such as phenolic acids and flavonoids. Karabagias et al. applied ethyl acetate to extract phenolic compounds from honey samples, which enabled floral authentication of Greek monofloral honey (Karabagias, et al., 2014). However, LLE has certain disadvantages, including

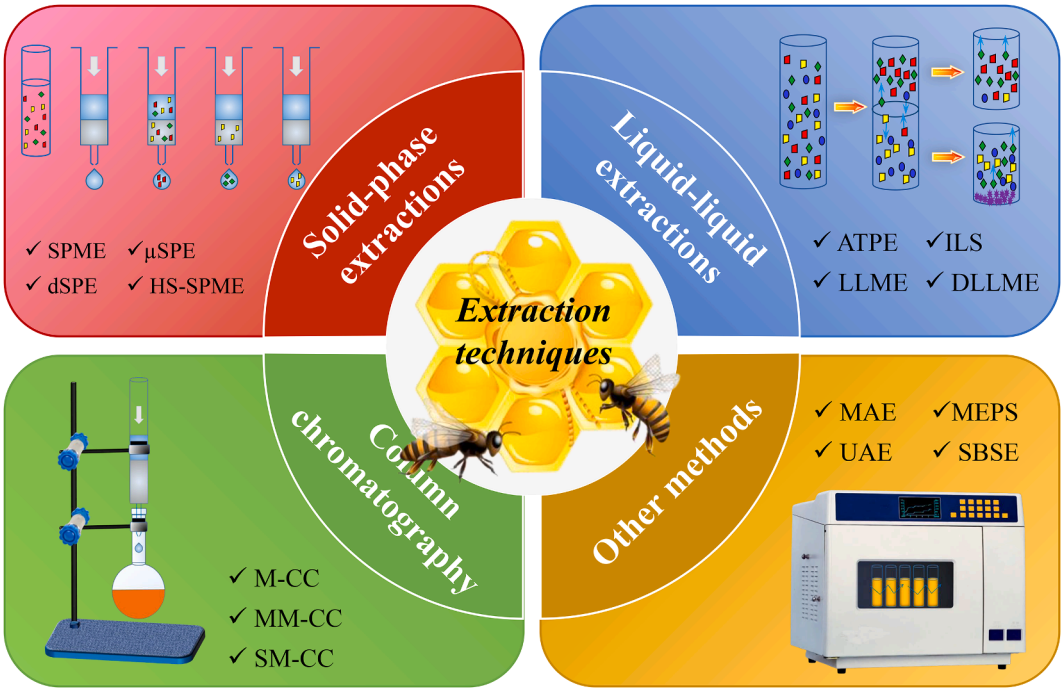

Fig. 2. Various extraction techniques applied in the quality control of honey. SPME: solid-phase microextraction, dSPE: dispersive solid-phase extraction, μSPE: micro-solid phase extraction, HS-SPME: headspace solid-phase microextraction, ATPS: aqueous two-phase extraction, LLME: liquid–liquid microextraction, ILS: ionic liquids, DLLME: dispersive liquid–liquid microextraction, MAE: microwave-assisted extraction, UAE: ultrasound-assisted extraction, MEPS: microextraction by packed sorbent, SBSE: stir bar sorptive extraction, M−CC: multi-column chromatography, MM-CC: mixed-mode column chromatography, SM-CC: surface-modified column chromatography.

complicated processes, the use of large amounts of organic solvents, and relatively low recovery yields. Thus, LLME and similar technologies have been developed. Moniruzzaman et al. evaluated the conditions for dispersive liquid–liquid microextraction (DLLME) for gas chromatography time-of-flight mass spectrometry (GC/TOF-MS) identification of organic compounds in honey (Moniruzzaman, et al., 2014). In addition, vortex-assisted liquid–liquid–liquid microextraction (VALLLME) combined with HPLC was applied for simultaneous determination of fourteen phenolic acids in honey, iced tea and canned coffee drinks (Shalash, Makahleh, Salhimi, & Saad, 2017). However, LLME also has disadvantages, such as long extraction time and low enrichment factor. The use of ionic liquids (ILs) has been suggested as green alternatives to traditional organic solvents for LLME. Vortex assisted-ionic liquid dispersive liquid–liquid microextraction (VA-IL-DLLME) and spectrophotometry were applied to determine quercetin in tea, honey, fruit juice, and wine (Altunay, Bingöl, Elik, & Gürkan, 2019). More recently, a novel LLE-based method, i.e., aqueous two-phase extraction (ATPE), has been successfully applied in the separation and purification of active compounds in natural products. Using ATPS, high extraction rate and product purity can be achieved, whilst maintaining the biological activity of extracted compounds. Compared with conventional LLE method, ATPS has certain advantages of mild extraction conditions, less required volume, various self-adjusting factors, high extraction rate, short phase separation time, easy amplification and operation, as well as suitability for industrial application (Enriquez-Ochoa, et al., 2020).

SPE involves the distribution between liquid and solid phases, and has been gradually used as a substitute for traditional LLE methods, owing to simplified operations, decrease use of organic reagents, and improved quantitative recovery yields. However, SPE has been primarily used for LC analysis, but was ineffective for the extraction of volatile compounds. As a consequence, SPME has emerged based on improvements on SPE, which have certain advantages, such as small aliquot size and solvent volume as well as shorter analysis time. SPME has been successfully applied in combination with gas chromatography (GC) to extract and analyze volatile compounds in honeys, such as alkanes, aldehyde, ketones, alcohols, esters, carboxylic acids, furans, ethers, terpenes, nor-isoprenoids and pyrene derivatives. In recent years, novel SPE strategies have been proposed, e.g., dispersive solid-phase extraction (dSPE), micro-solid phase extraction (μSPE), and headspace solid-phase microextraction (HS-SPME). HS-SPME coupled with gas chromatography-mass spectrometry (GC–MS) has been used more frequently for extraction and analysis of volatile compounds in honey samples for its simplicity, speed, sensitivity and versatility (Karabagias, Karabagias, Nayik, Gatzias, & Badeka, 2022).

CC is a powerful alternative for the removal of impurities and purification of active compounds in honey, which is based on the differences in polarity between molecules. When combined with large-scale instruments such as GC, GC–MS, HPLC, and HPLC-MS, CC has been widely used in the extraction of phenolic acids, flavonoids, abscisic acid, and volatile compounds in honey. For instance, Stanek et al. established a rapid method based on CC and high-performance thin-layer chromatographic (HPTLC) for the qualification and quantification of phenolic compounds and abscisic acid in honey samples (Stanek, Kafarski, & Jasicka-Misiak, 2019). The results suggest that CC is an efficient sample pretreatment technique for sugar-rich matrices like honey. Improved versions of CC have also been reported, such as multi-column chromatography (M−CC), mixed-mode column chromatography (MM-CC) and surface-modified column chromatography (SM-CC). Collectively, these reports suggest that CC can be considered a suitable technique for sample pretreatment of sugar-rich matrices such as honey.

3.2.2. Various instrumental techniques

3.2.2.1. Chromatographic and hyphenated techniques. Chromatographic systems coupled with various detectors have been considered powerful analytical tools and widely used in the determination of honey authenticity by the identification of chemical markers, among which are included carbohydrates, carotenoids, amino acids, phenolic acids, flavonoids, abscisic acid, and volatile compounds. The main advantage of chromatographic methods is that it allows the separation of analytes from the complex honey matrix, improving analytical detection and accuracy. In this context, GC is useful for the analysis of non-polar and semi-polar, as well as volatile and semi-volatile compounds in honey. In contrast, LC is more frequently used in the analysis of polar and non-volatile compounds. In addition, the use of targeted or non-targeted metabolomics combined with chromatographic techniques focusing on a comprehensive analysis of honey has been gaining increasing attention (Table 3).

Qualitative and quantitative analyses of target analytes based on LC have been widely performed, which have several advantages such as robustness, reproducibility, and wide applicability to different target analytes and second-order data output. In order to achieve different detection purposes, LC can be used in tandem with various types of detectors, such as diode array detection (DAD), pulsed amperometric detection (PAD), fluorescence detection (FLD), refractive index detector (RID), electrochemical detect ion (ECD), coulometric electrode array detection (CEAD) and mass spectrometry (MS). For instance, HPLC in tandem with DAD or MS has been widely used to identify polyphenols (flavonoids and phenolic acids) as potential authenticity markers in honey (X.-H. Zhang, Qing, et al., 2021; X.-H. Zhang, Wang, et al., 2021). It has been demonstrated that fructose, glucose, sucrose, and maltose can be quantified using HPLC-RID (Tosun & Keles, 2021). Moreover, HPLC-PAD has been one of the few well-established techniques that enabled identification of C_3 and C_4 sugars, and combined with chemometric tools also enabled detection of adulterants in honey (Akyıldız, et al., 2022). Over the last decade, HPLC systems have evolved towards miniaturization, smaller columns, lower solvent usage, higher pressures, and faster separation of target analytes, which resulted in ultra-high-performance chromatography (UHPLC). In fact, UHPLC has gained popularity in metabolomics applied to honey authenticity (Akyıldız, et al., 2022). In order to obtain higher-way data arrays (e.g., two-way or three-way) and improve analytical capability, two-dimensional liquid–liquid chromatography 2D (LC × LC) can be used as a suitable tool for the analysis of complex food samples. Additional applications of LC in honey authenticity are summarized in Table 3.

GC techniques have been widely used to determine the volatiles of honey samples. Volatiles are important analytes for assessing the authenticity of honey because they are directly related to the botanical species from which honeybees collect nectar or honeydew (Tsagkaris, et al., 2021). There are several reports on honey volatiles and their application to the botanical and geographical origin of honey based on GC coupled with various detectors (Karabagias, et al., 2022; X. Wang, Rogers, et al., 2019; X. Wang, Yang, et al., 2019). Machado et al. published a comprehensive review of volatiles as a fingerprint for botanical origin on monofloral honeys (Machado, et al., 2020). In some cases, more accurate results could be obtained by using a combination of GC and LC techniques (Akyıldız, et al., 2022; Osés, et al., 2020). Apart from volatiles, GC techniques combined with derivatization steps have been used to determine other compounds, such as amino acids and sugars (Azevedo, et al., 2017; Pascual-Maté, et al., 2018). Additional applications of GC in honey authenticity are summarized in Table 3.

3.2.2.2. Spectroscopy. Spectroscopic techniques allow obtaining information related to the structure, physicochemical properties and composition of honey with the purpose of authentication (Table 4). These techniques have been widely used for their speed, non-destructiveness, low cost, automatization, which enable establishing honey sample fingerprints, which in some cases allow simultaneous detection of several analytes. The combination of spectroscopic methods and chemometric tools is necessary for the classification and

Table 3
Some literatures in the past three years relating to the applications of chromatographic and hyphenated techniques coupled to various detectors in honey authentication.

Analytical technique	Purpose of analysis	Markers	Chemometric tools	References
HS-GC-IMS	Discriminate winter honey and sapium honey	Benzaldehyde dimer, phenylacetaldehyde dimer, phenylethyl acetate dimer	PCA, PLS-DA, SIMCA	(X. Wang, Yang, et al., 2019)
HS-GC-IMS, HS-SPME-GC–MS	Untargeted and targeted discrimination of honey collected by Apis cerana and Apis mellifera	Benzaldehyde, heptanal, phenylacetaldehyde, *trans*-linalool oxide, 1-nonanol, phenethyl acetate, 1-heptanol, cyclohexanone	PCA, OPLS-DA, SIMCA	(X. Wang, Rogers, et al., 2019)
CC-HPLC, HPTLC	Authentication of phacelia honeys (Phacelia tanacetifolia)	Gallic acid, 4-hydroxybenzoic acid, caffeic acid, p-coumaric acid, ferulic acid, myricetin, cinnamic acid, naringenin, galangin.	PCA, HCA	(Stanek, Teper, Kafarski, & Jasicka-Misiak, 2019)
SPE-UHPLC-DAD-ESI/MS	Floral classification of honey	Gallic, syringic, p-coumaric, caffeic, chlorogenic, ferulic, phydroxybenzoic, 3,4-dihydroxybenzoic, trans-cinnamic, rutin, quercetin, naringin, hesperitin, myricetin, apigenin, galangin, kaempferol, isorhamnetin, chrysin, pinocembrin, pinostrobin; (+)-catechinand (−)-epicatechin; resveratrol)	ANOVA, PCA, HCA	(Teodora & Elisabeta-Irina, 2019)
HS-GCI-MS	Detection of adulterated honey	Untarget	OPLS-DA	(Arroyo-Manzanares, et al., 2019)
CC-HPLC/GC–MS	Authentication of strawberry tree (Arbutus unedo L.) honeys	Arbutin, norisoprenoids, benzene derivatives, Theobromine, 2,6,6-Trimetyl-4-oxo-2-cyclohexen-1-carboxaldehyde, 3,4,5 trimethylphenol, 2-hydroxycyclopent-2-en-1-one	/	(Osés, et al., 2020)
CC-HPLC-ECD	Identification of acacia honey	Gallic acid, protocatechuic acid, p-hydroxybenzoic acid, chlorogenic acid, caffeic acid, p-coumaric acid, ellagic acid, ferulic acid, rosmarinic acid, syringic acid, vanillic acid, sinapic acid, isochlorogenic acid and cinnamic acid	SIMCA, PCA, OPLS-DA	(Q. Wang, et al., 2020)
HPLC-RID	Detection of honey adulteration with sucrose syrup	Sucrose syrup, moisture, HMF, free acidity, proline, diastase activity, potassium, sodium, pollen, rotation degree, conductivity, sucrose, invert sugar, and total sugar	PCA	(Karabagias, Badeka, & Kontominas, 2020)
MSweEt-UHPLC-MS/MS, UHPLC-PDA, UHPLC-UV, GC–MS, ICP-MS	Identification of the rice syrup adulterated honey	Sorbic acid	/	(Karabagias, et al., 2020)
HS-SPME/GC–MS	Targeted evaluation of the volatile compounds of Quercus ilex honey in relation to its provenance	Heptane, dimethyl-disulfide, octane, nonane, styrene, alpha-pinene, *meta*-cymene, DL-limonene, 1-decanol, tetradecanoic acid ethyl ester, eucalyptol, benzeneacetaldehyde, tetradecanoic acid ethyl ester	MANOVA, PCA, SLDA	(Karabagias, Karabagias, Nayik, Gatzias, & Badeka, 2022)

[a]Sample extraction techniques: column chromatography (CC), liquid–liquid extraction (LLE), solid-phase extraction (SPE), solid-phase microextraction (SPME), headspace solid-phase microextraction (HS-SPME), Modified Swedish ethyl acetate extraction (MSweEt).
[b]Instrument: high-performance liquid chromatography-electrochemical detection (HPLC-ECD), high-performance liquid chromatography-refractive index detector (HPLC-RID), ultra-high performance liquid chromatography -diode array detector-electrospray ionisation mass spectrometry (UHPLC-DAD-ESI/MS), high-performance thin-layer chromatographic (HPTLC), gas chromatography-mass spectrometry (GC–MS), headspace gas chromatography-ion-mobility (HS-GC-IMS), inductively coupled plasma mass spectrometry (ICP-MS).
[c]Chemometric tools: principal component analysis (PCA), hierarchical clustering analysis (HCA), linear discriminant analysis (LDA), partial least squares (PLS), partial least square-discriminant analysis (PLS-DA), supervised orthogonal partial least-squares discriminant analysis (OPLS-DA), stepwise linear discriminant analysis (SLDA), partial least square regression (PLSR), *k*-nearest neighbors (*k*-NN), soft independent modeling of class analogy (SIMCA), analysis of variance (ANOVA), multivariate analysis of variance (MANOVA).

discrimination of honeys. In a study, samples of raspberry, mint, rape, sunflower, thyme, acacia and tilia honey were subjected to authentication using nondestructive FTIR methods coupled with PCA, linear discriminant analysis (LDA) and partial least square regression (PLSR) (Pauliuc, Ciursă, Ropciuc, Dranca, & Oroian, 2021). In another study, UV–Vis spectroscopy and chemometric methods of one-class classification (OCC) were applied to authentication of honey adulterated with sugar syrups (de Souza, Fernandes, & Diniz, 2021).

Vibrational spectroscopy is based on molecular absorption of infrared radiation (infrared spectroscopy) or light scattering (Raman spectroscopy) to study honey authenticity. This technique enables the detection of known compounds by fingerprinting as well as the identification of unknown compounds by determining the properties of functional groups and bonds. Infrared (IR) spectroscopy is more frequently used for detecting adulterants in honey owing to its several advantages such as speed, non-destructiveness, easy sample preparation, low cost, and suitability for field monitoring. Both near-infrared (NIR) and mid-infrared (MIR) spectroscopy have been used to determine botanical and entomological origins of honey. In a study, the syrup-blended adulteration of Manuka honey was detected using NIR spectroscopy combined with aquaphotomics (Yang, et al., 2020). Spectral variance analysis, PCA analysis and PLSR model regression vector analysis were then performed for the spectral region of 1300–1800 nm. The experimental results demonstrated that the detection of syrup-adulterated Manuka honey using NIR spectroscopy combined with aquaphotomics is practical. In addition, convolutional neural networks (CNN) strategy combined with MIR spectra was applied for the identification of sugar adulteration in honey (Q. Li, et al., 2021).

Raman spectroscopy enables establishing molecular fingerprints to detect target analytes at trace amount elements related to the identification of honey authenticity. When combined with chemometric tools, e.g., LDA and soft independent modeling of class analogy (SIMCA), Raman spectroscopy can provide an accurate determination of geographical and botanical origin of honey samples (Magdas, Guyon, Berghian-Grosan, & Muller Molnar, 2021). Nevertheless, considering the complexity of the honey matrix, the application of Raman spectroscopy to honey authenticity verification is still in its infancy. Certain shortcomings in the use of Raman spectroscopy for this purpose have been identified such as weak Raman scattering, fluorescence interference, superimposed spectral information, low reproducibility, redundant dataset, high cost, and relatively low operational speed. In addition, with the development of Fourier transformation (FT) instrumentation, as FT-IR and FT-Raman, the application of vibrational spectroscopy in honey authentication has increased significantly

Table 4
Some literatures in the past three years relating to spectroscopic techniques used for monitoring honey authentication issues.

Analytical technique	Purpose of analysis	Markers	Chemometric tools	References
Vis-NIRS	Identification and quantification of different adulterants in high-quality honey	Untarget	HCA, PLS, LDA	(Aliano-Gonzalez, et al., 2019)
^{1}H NMR	Compositional identification and authentication of Chinese honeys	Proline, xylobiose, uridine, β-glucose, melezitose, turanose, lysine	PCA, LDA, OPLS-DA	(He, et al., 2020)
^{1}H NMR	Detection of adulteration in Chinese monofloral honey	Valine, 2,3-butandiol, ethanol, ethyl acetate, 3-hydroxybutanone, lactic acid, alanine, acetic acid, proline, succinic acid, citric acid, turanose, nigerose, kojibiose, tyrosine, phenylalanine	CDA, OPLS-DA, PLS	(Song, et al., 2020)
NIR	Adulteration detection of Manuka honey	Untarget	PLSR, PCA	(Yang, et al., 2020)
MIR	Sugar adulteration identification of honey	Untarget	LS-SVM, PLS-DA	(Li, et al., 2021)
MIR, Raman	Quantitative analysis of honey adulteration	Untarget	PLS	(Li, et al., 2020)
FTIR, UV	Classification of Cretan thyme, multifloral and honeydew honey	Untarget	HCA, PLS, LDA	(Orfanakis, et al., 2021)
FTIR	Authentication of different monofloral honeys	Untarget	PCA, PLSR, LDA	(Pauliuc, et al., 2021)
Raman	Geographical and botanical classification of honeys	Untarget	SIMCA, ML	(Magdas, et al., 2021)
UV–Vis	Authenticate honey in terms of their individual and simultaneous adulterations with corn syrup, agave syrup, and sugarcane molasses	Untarget	PCA, OC-PLS, DD-SIMCA	(de Souza, et al., 2021)
UV–Vis, NIR	Detection of honey adulteration	Untarget	PLS, ANN	(Valinger, et al., 2021)
IR, FS	Authentication of acacia honey	Untarget	ANOVA, PCA	(Hao, et al., 2021)
EEM–FS	Detection of adulterations in a valuable Brazilian honey	Untarget	PARAFAC, PLS-DA, UPLS-DA, NPLS-DA	(Antonio, et al., 2022)
^{1}H NMR	Detecting adulteration of stingless bee honey	Untarget	SIMCA, PCA, OPLS-DA	(Yong, et al., 2022)

[a]Instrument: nuclear Magnetic Resonance (NMR), ultraviolet (UV), nNear-infrared spectroscopy (NIR), mid-infrared spectroscopy (MIR), visible-near infrared spectroscopy (Vis-NIRS), fluorescence spectroscopy (FS), excitation–emission matrix-fluorescence spectroscopy (EEM-FS).
[b]Chemometric tools: principal component analysis (PCA), hierarchical clustering analysis (HCA), linear discriminant analysis (LDA), partial least squares (PLS), partial least square-discriminant analysis (PLS-DA), supervised orthogonal partial least-squares discriminant analysis (OPLS-DA), partial least square regression (PLSR), soft independent modeling of class analogy (SIMCA), analysis of variance (ANOVA), artificial neural network (ANN), canonical discriminant analysis (CDA), machine Learning (ML), parallel factor analysis (PARAFAC), unfolded PLS-DA (UPLS-DA), multilinear PLS-DA (NPLS-DA), one-class partial least squares (OC-PLS), data-driven soft independent modeling of class analogy (DD-SIMCA), least squares support vector machines (LS-SVM).

(Orfanakis, Markoulidakis, Philippidis, Zoumi, & Velegrakis, 2021; Pauliuc, et al., 2021).

Nuclear magnetic resonance (NMR) spectroscopy has been increasingly applied to food authenticity control to detect adulterations and determine quantitatively target compounds for regulatory control. In a recent study, He et al. combined ^{1}H NMR spectroscopy with chemometric techniques for the identification and authentication of Chinese honeys (He, et al., 2020). In this study, eight honey components, namely proline, xylobiose, uridine, β-glucose, melezitose, turanose, lysine and an unknown component, were screened using volcano plots. Moreover, ^{1}H NMR and ^{13}C NMR were combined for the geographical characterization of honeys including 41 samples (multifloral and acacia) from different countries (Consonni & Cagliani, 2008). Collectively, the advantages of NMR over chromatographic and other spectroscopic techniques are long-term stability of spectra, extensive structural information, and the ability to detect compounds in complex mixtures without prior purification and separation. However, similarly to other high-end techniques, the high costs involved in instrumentation and experiments limit the widespread application.

3.2.2.3. Elemental techniques. Elemental composition of honey is strongly related to the type of melliferous plants, environment, soil, climate, and anthropogenic activity. Thus, elemental analysis techniques are commonly used to differentiate honey botanical and geographical origin as well as adulteration, being hence employed in honey authentication, safety assurance, and quality control. Elemental fingerprinting can be determined inductively by coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectroscopy (ICP-OES), which provide multi-elemental determination in a single run. In a recent study, mineral element chemometrics profiling was applied to differentiate pure honey from honey adulterated by the direct addition of pale syrups (T. Liu, et al., 2021). In this text, five monofloral models were established using sparse partial least squares discriminant analysis (PLS-DA), and discriminative accuracy was above 93%, whereas classification accuracy of the multifloral model was 87.7% in the blind test. In addition, Silva et al. used ICP-MS to analyze 39 elements of bracatinga (*Mimosa scabrella* Bentham) honeydew honey samples produced in three Brazilian regions, i.e., Santa Catarina, Paraná, and Rio Grande do Sul (Silva, et al., 2021). Chemometric methods including clustering analysis (CA), PCA, and LDA have been used for discriminating honey geographical origin.

Isotopic fingerprints have also been commonly used for determining honey authenticity. Geană et al. conducted a comprehensive investigation based on physicochemical properties (water content, Brix, electrical conductivity, free acidity, pH, and HMF level), major sugar composition (fructose, glucose, sucrose, and maltose), and δ^{13}C values of honeys obtained from beekeepers and commercial honeys of different botanical origins (acacia, multifloral, honeydew, sunflower, rape, and linden), and adulteration with various industrial syrups was performed (Geană, Ciucure, Costinel, & Ionete, 2020). The chemometric methods, such as HCA and PCA were applied as statistical analysis tools to effectively distinguish honeys according to their quality. Last but not least, combined statistical analysis tools (i.e. LDA, SIMCA), both stable isotopes and elemental markers ($(D/H)_1$, δ^2H, δ^{18}O, La, Ce and Pr) have been used for effective discrimination of geographical and varietal origin of Romanian and French honeys (>98%) (D. A. Magdas, et al., 2021).

3.2.2.4. Bioanalytical techniques. Bioanalytical techniques targeting

genetic material have been thoroughly used in the field of food authentication. The genetic data encoded in the DNA can provide essential information about the geographical, botanical, and entomological origin (Tsagkaris, et al., 2021). Polymerase Chain Reaction (PCR) is a sensitive technique for amplifying specific DNA fragments, and has been used in gene quantitative analysis, exogenous identification, etc. (Lara Sobrino-Gregorio, Vilanova, Prohens, & Escriche, 2019). In addition, the pollen DNA identification in honey compensates the limitations of pollen morphology and species determination in honey microscopic palynological analysis (El Sheikha, 2019). With the development of high-throughput sequencing technology, novel DNA-based technologies (e.g. high resolution melting (HRM), DNA barcoding and metabarcoding) have been widely used in the detection of honey authenticity due to its advantages of strong specificity, high sensitivity, rapidity, accuracy and simple operation (Bruni, et al., 2015; Kek, Chin, Tan, Yusof, & Chua, 2017). Soares et al. exploits DNA barcoding combined with HRM analysis to differentiate the honey species in three clusters with confidence levels $>$ 99%, being the results well correlated with the sequencing analysis (Soares, et al., 2018). In a recent study, a novel DNA-based tool has been proposed for correct identification of DNA signatures in three economically important honeybee species (i.e., *Apis mellifera*, *Apis cerana*, and *Apis dorsata*) (Moškrič, Mole, & Prešern, 2021).

3.2.2.5. Electrochemical methods. The analytical techniques described above may present certain limitations, such as complex sample preparation procedures, high costs, destructiveness, non-automation, and unsuitable for field monitoring. These shortcomings have been overcome, to some extent with the development of electronic sensors, including the electronic nose (E-nose, gas sensor), electronic tongue (E-tongue, liquid sensor), and electronic eye (E-eye, vision sensor). The combined used of E-sensors and chemometric tools is a promising approach for honey authenticity and adulteration verification (Ciursa & Oroian, 2021; Lozano-Torres, et al., 2022; L. Sobrino-Gregorio, Tanleque-Alberto, Bataller, Soto, & Escriche, 2020). An E-tongue-based multistep pulse voltammetry combined with multivariate statistical techniques (PCA and PLS) has been applied to monitoring honey adulteration with sugar syrups (Lara Sobrino-Gregorio, Bataller, Soto, & Escriche, 2018). More recently, Yin et al. developed a remote E-tongue system combined with variational mode decomposition and Hilbert transformation (VMD-HT) extraction for honey authentication of botanical origin (Yin, et al., 2021). The proposed method resulted in a more accurate classification of honeys (98.2%) compared with other methods.

3.2.2.6. Other and mixed methods. Furthermore, the accuracy of honey authenticity technology is also related to the analysis of other specific components in honey. In particular, protein in honey mainly originates from bees, whereas pollen and/or nectar originate from nectar source plants, being thus a unique internal standard for honey identification. Based on protein specificity and content variation, honey source and hence adulteration can be determined. Bong et al. identified 50 bee-derived proteins in honey, with the most predominant proteins being major royal jelly proteins (MRJPs) (Bong, et al., 2021); these authors also selected twelve candidate peptides as potential authentication markers for New Zealand mānuka (*Leptospermum scoparium*) honey. Muresan et al. evaluated the botanical origin of 42 honey samples from five European countries (Belgium, France, Italy, Romania and Spain) using melissopalynology and specific protein profiles by electrophoresis on SDS polyacrylamide gels (SDS-PAGE) (S. Liu, et al., 2022). The hierarchical clustering analysis (HCA) and 2D heatmap demonstrated the relationship between their protein and melissopalynological profiles for honey authentication. Collectively, many detection techniques can be applied to the detection of honey authenticity. Due to the complex chemical composition of honey, it may be necessary to combine several analytical techniques to improve the selectivity and sensitivity of the methods. In this context, chromatographic techniques (such as GC–MS and LC-MS) and spectroscopic techniques (such as Raman, NIR, MIR, and NMR) combined with chemometric tools have broad application potential. In addition, with the continuous emergence of novel instruments that could be used combined with other equipment enable honey authenticity verification with higher sensitivity and rapidity. Svečnjak et al. proposed an improvement on the discriminating potential by data fusion using physicochemical and spectroscopic and chromatographic techniques (^{1}H NMR, FTIR-ATR, HS-SPME/GC–MS), allowing for a better separation of honey produced in honeycomb constructed in comb foundations adulterated with 90% of paraffin (PF-H) and honey ripened in genuine beeswax (BWF-H) (Svecnjak, et al., 2019). In another study, the analytical power of the combined use of FT-NIR and HPLC-DAD with multivariate data analysis (PLS-DA) was evaluated in the classification of 70 honey samples of seven varieties based on their botanical origin (Ghanavati Nasab, Javaheran Yazd, Marini, Nescatelli, & Biancolillo, 2020). Collectively, the data fusion approach was satisfying, leading to correct classification of 100% of samples belonging to the category of interest. Last but not least, the highly reproducible MIR spectroscopy and highly sensitive MALDI-TOF-MS data were comprehensively applied to the metabolomic profiling of monofloral and multifloral honey samples from three botanical origins (canola, acacia, and honeydew) (Brendel, et al., 2021). In this work, three different chemometric models (PCA-LDA, PCA-*k*NN, and SIMCA) were applied to the data obtained by both techniques to effectively distinguish monofloral and multifloral honey samples.

3.2.3. Chemometric strategies

The emergence and development of instruments employed in high-throughput analysis have enabled easy acquisition of large amounts of data from thousands of analytical channels simultaneously. Thus, researchers and analytical chemists are presented not only with simple scalar or vector data, but also with second-, third-, fourth-order or even higher-order data. The challenge with these large and complex data sets relies on the fact that it contains not only useful chemical information but also large amounts of interference such as interferents response, background, and instrumental noise (H.-L. Wu, Wang, & Yu, 2020). Another considerable challenge is related to the selection of optimal experiment conditions, the performance of optimal mathematical processing of raw data, and effectively mining of chemical composition, structural information and biochemical activities. Chemometrics has emerged and been developing rapidly, paving new ways for efficient chemical determination, mining useful information, and designing less costly and polluting experiments, thus becoming a very promising field in analytical chemistry.

Chemometrics is an interdisciplinary discipline that employs chemical, computational, mathematical and statistical tools to extract useful information from large and complex data sets (H.-L. Wu, et al., 2020). Chemometrics provides powerful tools for targeted and non-targeted analysis to determine honey authenticity or verify its geographical or biological origin. An overview of chemometrics-based methods is shown in Fig. 3. Following analysis by a variety of methods, large data sets derived from honey samples can be combined into high-way arrays, and processing and analysis of high-way arrays have become key to the success of honey authenticity identification. The most commonly used chemometrics methods for honey authentication can be classified into two categories: i) classification and qualitation; ii) calibration and quantitation.

3.2.3.1. Classification and qualitation. Chemical pattern recognition has been improving in the last decades to enable mining useful hidden information from raw complex data sets, thus performing exploratory or predictive data analysis to determine implicit relationships between research objects, thereby providing chemists with useful decision-making information. Based on whether prior knowledge is available of

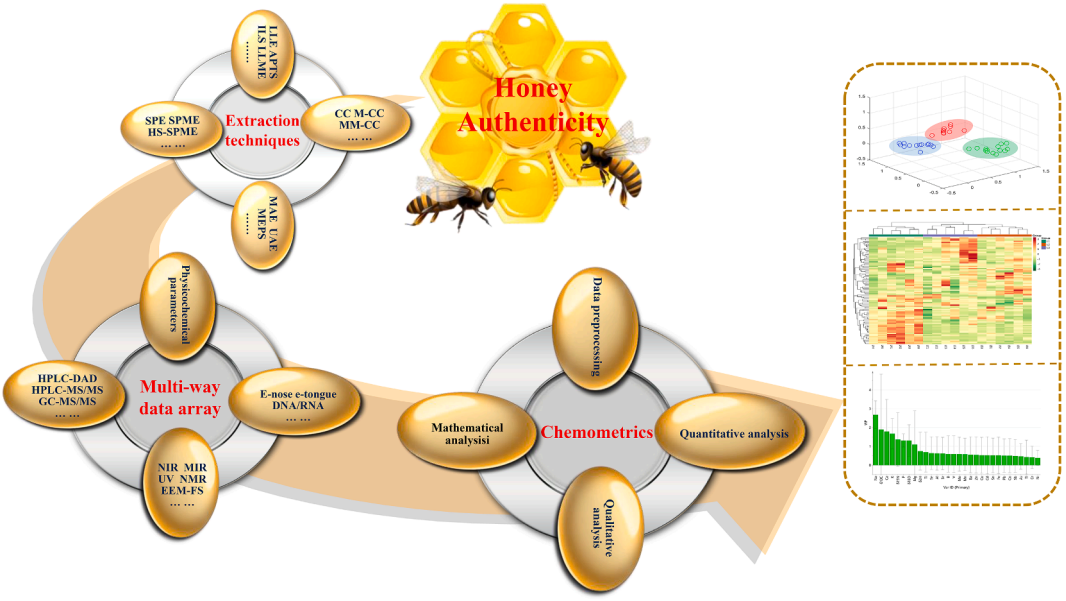

Fig. 3. An overview flowchart of the chemometrics-based methods for the honey authenticity verification.

classes or groups, chemical pattern recognition can be divided into supervised and unsupervised methods.

Unsupervised pattern recognition methods, also known as exploratory analysis techniques, relies on the assumption that samples with similarities group together thus presenting a short distance, whilst dissimilar samples cluster separately hence group with a larger distance from each other in a multi-way space. Thus, the appropriate classification method can be selected by information processing, and discrimination analysis can be conducted. Principal component analysis (PCA) and hierarchical cluster analysis (HCA) are the most common unsupervised pattern recognition methods used in honey authenticity identification. PCA is used for dimension reduction of complex data sets that employs linear combinations of original variables to retrieve new variables called principal components (PCs), while conserving as much information as possible. PCA has been successfully used, for instance, to discriminate geographical and botanical origins (Rodopoulou, et al., 2022; Sogut & Seydim, 2020), as well as detect honey adulteration with sucrose syrup (Karabagias, Badeka, & Kontominas, 2020; Pereira, et al., 2020). In contrast, the basic idea of HCA is that each sample is considered a class, and the distance between samples and the distance between classes are determined; then, the pair with the shortest distance is fused into one class, and classed are progressively created from samples until all samples are classified into one class. HCA is widely employed in honey authentication since it offers an intuitive interpretation of experimental results in the form of a dendrogram. Orfanakis, et al. investigated the classification of Cretan thyme, multifloral and honeydew honey using HCA, PLS, and LDA in combination with FTIR and UV (Orfanakis, et al., 2021). In another study, HCA and PCA were combined to establish a relationship between honey chemical composition and the authentication of phacelia honeys (*Phacelia tanacetifolia*), enabling successful separation of acacia and lime honey samples (Stanek, Kafarski, et al., 2019).

Supervised pattern recognition methods train samples with prior knowledge of classes or groups, thus allowing machines learn from known classes and groups in surveyed samples to enable classification of samples of unknown categories. Among these methods, it can be included linear discriminant analysis (LDA), partial least squares discriminant analysis (PLS-DA), orthogonal projections to latent structures modeling discriminant analysis (OPLS-DA), soft independent modeling of class analogy (SIMCA), support vector machine (SVM), *k*-nearest neighbors (*k*-NN), and artificial neural networks (ANN). LDA, also known as Fisher's linear discriminant analysis, is one of the classic pattern recognition methods; this method employs category information of known training samples to project high-dimensional data into the low-dimensional vector space, in order to enable mining of classification information and reducing dimensionality. LDA has been recently used to confirm the authenticity and geographical origin of honey (Yayinie, et al., 2021). PLS-DA and OPLS-DA are also popular classification methods for distinguishing the botanical and geographical origins of monofloral honeys (X. Wang, Rogers, et al., 2019; X. Wang, Yang, et al., 2019), and detection of honey adulterated with sugar cane or corn syrups (Arroyo-Manzanares, et al., 2019). In addition, SIMCA uses PCA to determine classification of samples; on this basis, the corresponding PCA classification models are established for each sample, and are then used to identify unknown samples (Granato, et al., 2018). SIMCA has been applied to the authentication of commercial honeys (Aykas, Shotts, & Rodriguez-Saona, 2020), and classification of honeys based on geographical and botanical origins (Dana Alina Magdas, et al., 2021). Finally, ANN simulates human thinking based on the working principle of biological neurons, being often used for the classification of nonlinear data due to its strong nonlinear mapping capacity and preliminary self-organization and adaptive ability. ANN has been successfully applied for adulteration identification and quantification of physicochemical proprieties of pure and adulterated honey samples (Valinger, et al., 2021).

Collectively, the above pattern recognition methods present positive and negative aspects. Therefore, it might be challenging to select the most appropriate statistical model due to the complexity of practical problems. In this respect, a combination of multiple pattern recognition methods is recommended to obtain better classification results.

3.2.3.2. Calibration and quantitation. Honey can be adulterated in a variety of ways. Honey adulteration rate ranges from 2 to 27%. In order to determine quality and purity of honey, and to improve accuracy of honey adulteration detection, it is of great theoretical and practical significance to evaluate quantitatively honey authenticity. High-way accurate quantification of chemometrics enables deep exploration of difference between samples, thus providing novel approaches for honey authenticity determination. The following analytical strategies are commonly used in honey authenticity, combining the "second-order advantage of chemometrics multivariate calibration methods and qualitative power of chemical pattern recognition to perform multi-way quantitative analysis: i) targeted quantitative identification; ii) non-

targeted quantitative identification.

Targeted multi-way quantitative analysis for honey authenticity verification is based on two steps: firstly, an accurate and simultaneous quantitative analysis of targeted active components in honey is carried out using multivariate calibration techniques; secondly, according to quantitative results, a bilinear model of honey sample-targeted active components content can be established using chemical pattern recognition method. In a recent study conducted by our group, a chemometric second-order multivariate calibration technique was applied for qualitative and quantitative analysis of seven phenolic acids in honey samples, then PCA and PLS were used based on the contents of major target analytes for honey authentication of different nectar sources (jujube, acacia, linden, cloud and vetch) (X.-H. Zhang, Qing, et al., 2021). In another study, a novel chemometrics-assisted HPLC-DAD method based on alternating trilinear decomposition (ATLD) algorithm and PCA has been developed to rapidly and sensitively quantify phenolic compounds in honey and identify three types of adulteration in honey samples (using HFCS, CS, and TS) (X.-H. Zhang, Wang, et al., 2021).

For non-targeted multi-way quantitative analysis, high-dimensional data sets can be decomposed directly by relevant chemometric algorithms. Based on mathematical separation of second- or third-order calibration techniques, 3D or 4D fingerprint data collected by high-end instruments (HPLC-MS/MS, GC–MS/MS) were analyzed to obtain relevant concentration information (loading matrix). Then, based on data fusion, information fusion was performed, and high-way regression models of chemometrics such as *N*-way partial least square (*N*-PLS) and unfold partial least squares (*U*-PLS) are used to establish the identification and classification methods of honey quality. In 2015, Lenhardt et al. used fluorescence spectroscopy coupled with parallel factor analysis (PARAFAC) and PLS-DA for the characterization and classification of 95 honey samples of different botanical origin (acacia, sunflower, linden, meadow, and fake honey) (Lenhardt, Bro, Zeković, Dramićanin, & Dramićanin, 2015), the obtained PLS-DA classification model, constructed by PARAFAC model scores, discriminated fake honey samples with 100% sensitivity and specificity. Honey samples were also classified using PLS-DA with error rates of 0.5% for linden, 10% for acacia, and about 20% for both sunflower and meadow mix.

4. Conclusions and future perspectives

Honey adulteration and counterfeiting are common practices around the world. Due to the complexity of honey matrix, the technology employed for honey authenticity detection has great limitations. Existing analytical methods can detect honey adulteration based on multiple aspects, which can effectively measure known adulteration practices, but do not enable early identification and a rapid response strategy to tackle the emergence of novel practices of adulteration. Moreover, the development of detection technologies lags behind the adulteration technology, and adulteration surveillance is costly, which makes it difficult to eliminate the practice of honey adulteration. Thus, a quick, accurate and efficient identification of honey adulteration is still highly needed. In the face of these challenges, novel technologies for detecting honey adulteration should reframe the issue, from the past “whether the sample contains adulterants” to “whether all the samples are natural mature capped honey, and labeled with the correct botanical and geographical origins”, the latter is the authenticity detection.

Natural honey has both nectar and bee unique components, while fake honey does not contain these components or the contents are reduced. Therefore, selecting unique natural components found in honey might be a future direction for innovating honey authenticity detection technologies. The application of food metabolomics methods can provide powerful tools for honey authenticity research. Thus, honey authenticity detection based on endogenous components will be one of future research hotspots in the field of honey quality control, and more reliable methods for discriminating honeys need to incorporate a panel of compounds, preferably combined with chemometric tools for data collation, extraction and interpretation. Additionally, due to the complex chemical composition of honey, a combination of several analytical techniques is required to determine honey authenticity. The methods such as chromatography and hyphenated techniques (HPLC, GC, HPLC-MS/MS, GC–MS/MS), spectroscopic techniques (Raman, NIR, MIR and NMR) combined with chemometric data processing tools have broad application potential. Furthermore, new instruments for analytical detection are constantly emerging, and the combination of various instruments is more likely to provide more sensitive and rapid methods to classify authentic or adulterated honey samples. Despite the existing limitations resulting from technical difficulties and/or complexity of the honey matrix, continuous development in analytical instrumentation and chemometric algorithms will propel these technologies from laboratory to industry. Finally, simplification and portability of honey adulteration identification methods, such as kits based on ELISA, are future tendencies in order to enable on-site rapid detection, control honey adulteration at the source, and promote development of the honey industry.

Declaration of Competing Interest

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

Data availability

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

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