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Review Article

# Rapid analysis technologies with chemometrics for food authenticity field: A review

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ABSTRACT

In recent years, the problem of food adulteration has become increasingly rampant, seriously hindering the development of food production, consumption, and management. The common analytical methods used to determine food authenticity present challenges, such as complicated analysis processes and time-consuming procedures, necessitating the development of rapid, efficient analysis technology for food authentication. Spectroscopic techniques, ambient ionization mass spectrometry (AIMS), electronic sensors, and DNA-based technology have gradually been applied for food authentication due to advantages such as rapid analysis and simple operation. This paper summarizes the current research on rapid food authenticity analysis technology from three perspectives, including breeds or species determination, quality fraud detection, and geographical origin identification, and introduces chemometrics method adapted to rapid analysis techniques. It aims to promote the development of rapid analysis technology in the food authenticity field.

## 1. Introduction

In recent years, economically motivated adulteration (EMA) has occurred frequently, which has interfered with the stability of the food industry and damaged the interests of consumers, resulting in a crisis of consumer confidence in the food industry and the government. Therefore, EMA has now been included in the category of food safety supervision. Most countries have taken corresponding countermeasures regarding policies, regulations, and standards while establishing adulteration analysis methods and constructing food fraud databases and traceability platforms to further maintain the stable development of the food market (Manning and Soon, 2016). Continuously exploring analysis technology based on food characteristics provides theoretical basis for the entire regulatory process. Commonly used research methods include stable isotope techniques, mineral element analysis, fatty acid analysis, and traditional DNA techniques (Wang et al., 2022c). Nevertheless, with the complexity of the food adulteration problem, analytical methods need to be continuously updated and improved, while result accuracy and timeliness must be considered (Xing et al., 2019a). Therefore, rapid analysis technologies are necessary to improve the current situation in the food authenticity field.

Recently, spectroscopy, mass spectrometry, electronic sensor technology and DNA technology have been widely used in food analysis. The rapid and non-destructive analytical characteristics of spectroscopic techniques and the low analytical cost and high analytical efficiency of electronic sensor techniques are compatible with the technological needs in the food authenticity field, and have been used to identify the authenticity of a wide range of food products in recent years (Kharbach et al., 2023; Tan and Xu, 2020). Mass spectrometry and DNA analysis have the advantages of high sensitivity and wide range of application (Huang et al., 2010; Scarano and Rao, 2014). Their more mature applications have been reported in food authenticity studies. Recently, under the background of pursuing timeliness in food analysis, scholars have developed many kinds of rapid analysis techniques based on mass spectrometry and DNA technology, which do not affect the analysis effect but improve the efficiency, and these rapid analysis techniques have

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also been gradually explored in the food authenticity field. However, the data collected through these techniques are often characterized by large data volumes, high data dimensions, and complex data structures, so the selection of chemometrics methods appropriate to the analytical techniques to aid authenticity analysis is also crucial for the food authenticity field (Agriopoulou et al., 2022).

The current research on the rapid analytical techniques for food authentication mainly involves breeds or species identification (differentiating between different species or different breeds of the same species), quality fraud identification (identifying illegal addition, food grading, content falsification, food feeding or cultivation methods, food storage or processing methods) and geographical origin identification. Therefore, this paper summarizes the status of the rapid analysis technology for food authentication from these three perspectives, and introduces the representative chemometrics methods adapted to rapid analysis techniques. It aims to provide a reference for the development of rapid analysis technology in the food authenticity field.

## 2. Principle and research progress in rapid analysis technology

### 2.1. Spectroscopic technology

The fast advancement of contemporary optical technology served as the foundation for the creation of spectroscopic technology, which is now comparatively well-established in food analysis. It is commonly used for the non-destructive determination of food authenticity due to its high accuracy, fast response, good stability, and intuitive results (Dong, 2017). The range of applications of spectroscopic technologies varies due to their principle and characteristic differences, which also affect parameters like penetration, accuracy, analytical speed, and organic matter resolution (Table 1). Mid-infrared (MIR), near-infrared (NIR), and Raman spectroscopy and nuclear magnetic resonance (NMR) are frequently used for food authentication, while hyperspectral imagery (HSI) is often employed for breeds or species and geographic origin identification. Terahertz (THz) application for food authentication is still in its infancy.

#### 2.1.1. Breeds or species

Breeds or species identification is most commonly used for food authentication. Most studies on spectroscopic technologies for breeds or species identification focus on plant-derived products, such as fruits, cereals, and oils, while minimal research is available involving animal-derived products, such as meat and dairy products. Extensive research has been performed on NIR and NMR. Relevant literatures are summarized in Table 2.

Recent research has focused on using NIR to identify plant-derived breeds and species in food products, with fewer studies concentrating on determining animal-derived. NIR is mainly applied to oils, fruits, and cereals in plant-derived products. Studies on oils primarily concentrate on differentiating between high-value pure oils and contaminated oil combined with low-value oils. In a study examining pumpkin seed oil for sunflower oil adulteration, NIR combined with orthogonal partial least-squares discriminant analysis (OPLS-DA) completely distinguished pure pumpkin seed oil from the adulterated version, with an accurate discrimination rate of 100% (Balbino et al., 2022). Similarly, the differentiation between olive oil and soy oil was achieved by NIR, and NIR combined with partial least squares regression (PLSR) algorithm could predict the amount of olive oil adulterated with soy oil, with a coefficient of determination ($R^2$) as high as 0.975 (Santos et al., 2020). NIR for fruits has been studied mainly to distinguish their breeds in terms of breeds or species identification. NIR combined with linear discriminant analysis (LDA) distinguished two lemon breeds with 66 % accuracy (Ruggiero et al., 2022), and combined with quadratic discriminant analysis (QDA) identified five apple breeds with 85 %–98 % accuracy (Cortes et al., 2019). The accuracy differences may be related to the chemometrics method used to analyze the data. NIR is also capable of breeds differentiation in cereal-based products. Five wheat breeds were distinguished based on their kernels with an 80 %–100 % accuracy. The wheat flour was also analyzed, distinguishing the five wheat breeds with a 92.4% to 100 % accuracy (Ziegler et al., 2016). The increased accuracy could be attributed to higher sample homogeneity when wheat is milled into flour. Furthermore, NIR was combined with one-class partial least squares (OCPLS) to distinguish between pure almond flour and adulterated samples mixed with peanut flour, yielding an accuracy of 91%–100% (Karacaglar et al., 2019). Although minimal research is available regarding the utilization of NIR for breeds or species identification in animal-derived products, it has been applied to meat, milk, and oil products. In studies reported with meat, the animal origin of both intact and ground meat could be identified by NIR spectroscopy. A study using duck meat instead of beef used FT-NIR spectroscopy to identify raw beef, beef-duck mixtures, and raw duck meat at a 100% accuracy rate (Han et al., 2022). NIR spectroscopy combined with SIMCA identified cattle, pig, and sheep species sources in minced meat at a 100 % discrimination accuracy (Pieszczek et al., 2018). In addition, the substitution of high-value goat milk and butter with low-value products can be recognized by NIR. NIR in combination with PLS-DA can differentiate between cow milk adulterated goat milk with 100% identification accuracy (Teixeira et al., 2021a). Soybean oil doped in butter can also be quantitatively characterized by NIR combined with PLSR (Pereira et al., 2019).

NMR represents the second most popular method for distinguishing food breeds or species, displaying the same application range as NIR. For

**Table 1**
Principles and characteristics of spectroscopic technologies.

| Spectral classification | Generation principle | Wavelength/ Wavenumber | Characteristics | References |
|---|---|---|---|---|
| MIR | Based on the characteristics of light such as scattering, emission and absorption | 4000 $cm^{-1}$–400 $cm^{-1}$ | Simple analysis process, low cost, high speed and high reproducibility | Mehltretter et al. (2017) |
| NIR | Hydrogen-containing groups cause molecular vibration | 14286 $cm^{-1}$–4000 $cm^{-1}$ | High analysis efficiency, wide sample application range, multi-component continuous detection and online analysis | Hao et al. (2019) |
| Raman | Based on the scattering of light and the vibration and rotation energy levels of matter molecules | 4000 $cm^{-1}$–50 $cm^{-1}$ | Simple sample processing, fast detection speed, simple instrument operation and high sensitivity | He and Sun (2018) |
| HSI | Imaging the target area at the same time in tens to hundreds of continuous and subdivided spectral bands. | – | Fast, efficient, non-invasive, accurate results and comprehensive coverage | Cubero et al. (2010) |
| THz | Photoconductive generation of broadband pulse, optical rectification generation of broadband pulse, narrow band continuous terahertz pulse generation technology, etc | 0.03–3 mm | Electronics and optics with dual properties | Moon et al. (2019) |
| NMR | Based on the absorption of radio frequency radiation by atomic nucleus | Low-field NMR in pulse mode | Short test time, easy operation, no radiation, safety, high efficiency, strong penetration and no damage | Lenz and Wilson (2007) |

MIR, mid-infrared; NIR, near-infrared; HSI, hyperspectral image; THz, Terahertz; NMR, nuclear magnetic resonance.

**Table 2**
Application progress of spectroscopic technology in breeds or species identification of food.

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| MIR | Animal | Aquatic product | Tuna, 4 Breeds: yellowfin, skipjack, bigeye and albacore | 4000–700 $cm^{-1}$ | NORM | PLS-DA, FDA | 93.3–100% | Boughattas and Karoui (2021) |
| | | Oil | 2 Species: butter and soybean | 4000–400 $cm^{-1}$ | NORM | PCA, PLSR | – | Pereira et al. (2019) |
| | Plant | Oil | 8 Breeds: olive, sunflower, pumpkin, hempseed, soybean, walnut, linseed and sea buckthorn | 3600–650 $cm^{-1}$ | – | PLS-DA, CA, RF | – | Socaciu et al. (2020) |
| | | | Olive oil, 4 Breeds: Moroccan Picholine, Languedoc Picholine, Arbozana and Arbequina | 4000–700 $cm^{-1}$ | NORM, 2nd, MSC | PCA, FDA | 91.87% | Zaroual et al. (2021) |
| | | Honey | 3 Species: canola, acacia, honeydew | 3050–960 $cm^{-1}$ | BLC | PCA-LDA, SIMCA | 91.3% | Brendel et al. (2021) |
| | | | 3 Breeds: monofloral, polyfloral, and honeydew | 4000–650 $cm^{-1}$ | SG, SNV | PCA, SIMCA, HCA | 100% | Ozbay et al. (2022) |
| | | Cereal | Barley, 8 Breeds: Admiral, Commander, Compass, Fathom, Navigator, Schooner, Buloke and Scope | 4000–375 $cm^{-1}$ | SNV, 2nd | PLS-DA, LDA | 91%–100% | Porker et al. (2017) |
| | | | Quinoa flour, 3 Species: soybean, maize and wheat | 4000–600 $cm^{-1}$ | NORM | PLS-DA | 94%–97% | Rodriguez et al. (2019b) |
| | | Red wine | Grape, 11 Breeds: Sangiovese, Nebbiolo, Nerello Mascalese, Primitivo, Raboso, Cannonau, Teroldego, Sagrantino, Montepulciano and Corvina | 4000–700 $cm^{-1}$ | – | SVM | 96% | Parpinello et al. (2019) |
| NIR | Animal | Meat | 2 Species: beef and duck | 10,000–4000 $cm^{-1}$ | SNV, 2nd | ELM, PCA | 100% | Han et al. (2022) |
| | | | 4 Species: lamb, beef, pork and chicken | 900–1700 nm | NORM, MSC, OSC | SVM | 97% | Dashti et al. (2022) |
| | | | Ground meat, 3 Species: beef, pork and lamb | 960–1960 nm | MSC, ISC | OPLS-DA, SIMCA | 100% | Pieszczek et al. (2018) |
| | | Milk | Goat dairy products, 2 Species: goat and cow milk | 10,000–4000 $cm^{-1}$ | MSC, SNV, SG | PLS-DA | 100% | Teixeira et al. (2021a) |
| | | Oil | 2 Species: butter and soybean | 12,000–4000 $cm^{-1}$ | NORM | PCA, PLSR | – | Pereira et al. (2019) |
| | Plant | Oil | 2 Species: pumpkin seed and sunflower | 904–1699 nm | – | PCA, OPLS-DA | 100% | Balbino et al. (2022) |
| | | | 2 Species: olive and soy | 908–1676 nm | – | PCA, PLSR | – | Santos et al. (2020) |
| | | | 6 Species: sesame, corn, rice, peanut, rapeseed and blend | 10,000–4500 $cm^{-1}$ | – | ECR | – | Chen et al. (2018) |
| | | | 4 Species: camellia, corn, rpeseed and sunflower | 10,000–4200cm-$^{1}$ | SG | PLS-DA | 96.7% | Du et al. (2021) |
| | | Fruit | Apple, 5 Breeds: Fuji, Red Delicious, Royal Gala, Golden Delicious, Golden Rosé | 900–1700 nm | SG | QDA | 85%–98% | Cortes et al. (2019) |
| | | | Lemon, 2 Breeds: Ovale di Sorrento and Sfusato Amalfitano | 10,000–4000 $cm^{-1}$ | – | LDA | 66% | Ruggiero et al. (2022) |
| | | Cereal | Wheat, 5 Breeds: bread, spelt, durum, emmer and einkorn wheat | 1200–2400 nm | SG, 1st | PLS-DA | 80%–100% | Ziegler et al. (2016) |
| | | Almond powder | 2 Species: almonds and peanuts | 1000–2500 nm | SNV | OCPLS | 91%–100% | Faqeerzada et al. (2020a) |
| | | Coffee | 2 Breeds: arabica and robusta; 6 Species: corn, barley, soybean, rice, coffee husks and coffee | 10,000–4000 $cm^{-1}$ | SNV, SG | PCA | – | de Carvalho Couto et al. (2022) |
| | | Honey | 8 Species: Acacia, Bastard indigo, Chestnut, Honeydew, Linden, Rape, Milkweed and Sunflower | 740–1700 nm | SG | PCA, LDA | 99% | Bodor et al. (2021) |
| Raman/FT-Raman | Animal | Milk | 3 Species: goat, ewe, cow | 2000–200 $cm^{-1}$ | BLC | PLS-DA | 93% | Yazgan et al. (2020) |
| | | | Milk fat, 6 Species: 3 vegetable fat blends, sunflower, corn and margarine oil | 2000–200 $cm^{-1}$ | 1st, SG | PCA | – | Karacaglar et al. (2019) |
| | | Oil | Ghee, 2 Breeds: cow and buffalo | 1830–600 $cm^{-1}$ | SG, NORM, BLC | PLSR | – | Ahmad and Saleem (2019) |
| | | | 2 Species: butter and lard | 2000–200 $cm^{-1}$ | 1st, 2nd | PLSR, PCR | 99% | Taylan et al. (2020) |
| | | Aquatic product | Fish, 13 Breeds: Chilean *Salmo salar* L., Norwegian *Salmo salar* L., Danish *Salmo salar* L, Thunnus obesus, *Thunnus alalunga*, *Oncorhynchus keta, Anoplopoma fimbria*, Trichiurus lepturus, Cynoglossus semilaevis, | 3700–500 $cm^{-1}$ | BLC, NORM | CNN | 98.2% | Ren et al. (2023) |

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**Table 2** (*continued*)

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| | | | *Pleuronichthys cornutus*, *Oncorhynchus mykiss*, Pangasius bocourti and *Pagrus major*. | | | | | |
| | | | Salmon, 2 Breeds: atlantic salmon and rainbow trout | 2000–500 $cm^{-1}$ | MSC, BLC, SNV | PLSR | 87% | Chen et al. (2019) |
| | | Meat | Sausage, 6 Species: cattle, sheep, goat, buffalo, chicken and pig | 2000–200 $cm^{-1}$ | 3rd | PCA | 96.26% | Boyaci et al. (2014) |
| | Plant | Cereal | Cereal flour, 4 Species: barley, rye, spelt and wheat | 1750–450 $cm^{-1}$ | SG | PLS-DA | 97.7%–99.2% | Kniese et al. (2021) |
| | | | Rice, 3 Breeds: ndica, japonica and sticky | 3900–100 $cm^{-1}$ | BLC | SIMCA | 100% | Zhu et al. (2018) |
| | | Fruit | Grape, 4 Breeds: Nebbiolo, Barbera and Dolcetto | 4000–200 $cm^{-1}$ | SG | PCA | 90% | Mandrile et al. (2016) |
| | | Coffee | 5 Breeds: BRS Ouro Preto: C73, C83, C91, C79 and C91 | 2000–200 $cm^{-1}$ | SG, MSC | SIMCA | 100% | Luna et al. (2019) |
| | | Honey | 3 Species: fir, pine and thyme | 2000–200 $cm^{-1}$ | SG, BLC | LDA | 92.2%–93.8% | Xagoraris et al. (2021) |
| | | Pistachio nuts | 2 Species: green peas and pistachio nuts | 2000–200 $cm^{-1}$ | NORM | GILS | – | Eksi-Kocak et al. (2016) |
| HSI | Animal | Meat | 3 Species: lamb, beef and pork | 467–693 nm | – | 3D-CNN | 96.9%, 97.1% | Al-Sarayreh et al. (2020) |
| | | | Mutton roll, 3 Species: mutton, pork and dark | 400–1000 nm | SNV, NORM, 1st, 2nd | SPA, PLS-DA | 98.3%–100% | Jiang et al. (2021) |
| | | | Kebab, 4 Species: chicken, duck, pork and mutton | 380–1012 nm | – | PLS-DA | 100% | Jiang et al. (2022) |
| | | Colla coriiasini | 2 Species: colla coriiasini and pigskin gelatin | 388–1045 nm | MSC, SG | GRNN | 92.5% | Wang et al. (2018) |
| | Plant | Cereal | Coarse grain flour, 3 Species: millet, corn and soybean | 865–1711 nm | – | PLS-DA-SPA | 94.8%–100% | Shao et al. (2019) |
| | | | Maize, 5 Breeds: ND633, ND675, ND678, ND689 and XY335 | 382.2–1026.7 nm | SG, MSC | RBF-BPR | 96%–100% | Zhang et al. (2022a) |
| | | Almond powder | 2 Species: apricot and peanut | 900–2494 nm | SG, 2nd | DD-SIMCA | 89%–100% | Faqeerzada et al. (2020b) |
| | | Black pepper | 2 Species: papaya seeds and black pepper | 900–1710 nm | SG, 2nd | SIMCA | 100% | Orrillo et al. (2019) |
| | | Pistachio | 2 Species: green peas and pistachio | 3700–200 $cm^{-1}$ | BLC, 2nd | PLSR | – | Eksi-Kocak et al. (2016) |
| | | | 6 Species: edible and inedible pistachio nuts, pistachio shells, pistachio husks, twigs and stones | 1000–2500 nm | SNV, MC | PCA, KNN | – | Bonifazi et al. (2021) |
| THz | Plant | Cereal | Corn, 2 Breeds: High-oil 5598 and Zhengdan 958 | 0.5–3.5 THz | – | LDA, SVM | 100% | Yang et al. (2021a) |
| | | | Soybean seed, 3 Breeds: glyphosate-resistant and conventional seeds and their hybrid descendants | 0.5–1.5 THz | SNV | LS-SVM | 88.33% | Liu et al. (2016a) |
| | | | Rice, 2 Breeds: non-transgenic and transgenic | 0 ~ 5 THz | 1st | RF | 96.67% | Liu et al. (2016b) |
| | | Honey | 3 Species: Medlar, Vitex, and Acacia | 0.5–1.5 THz | – | PLS-DA | 88.46% | Liu et al. (2018b) |
| NMR | Animal | Milk | Cheese, 3 Breeds: Cheddar, Kefalotyri and Halloumi | – | BLC | OPLS-DA | 90.54% | Tarapoulouzi and Theocharis (2022) |
| | | | 2 Species: caprine and bovine | $^1$H 500.23 MHz | BLC, NORM | OPLS-DA | 80%–100% | Rysova et al. (2021) |
| | | Meat | Frankfurter, 4 Species: beef, chicken, turkey and pork | $^1$H 22.34 MHz | – | PLS | – | Uguz et al. (2019) |
| | | Oil | 3 Species: cod liver, sunflower and canola | $^1$H 400 MHz | BLC | ANN | – | Giese et al. (2019) |
| | Plant | Oil | Olive oil, 3 Breeds: Arbequina, Picual and Verdial | $^1$H 500 MHz | – | LDA | 100% | Sayago et al. (2019) |
| | | | 7 Species: olive, sunflower, high oleic sunflower, hazelnut, avocado, soybean and corn | $^1$H 500.13 MHz | – | PLS-DA | 88%–100% | Alonso-Salces et al. (2022) |
| | | | 2 Species: argan and sunflower | $^1$H 60 MHz | BLC | nearest-neighbor type classifier | – | Gunning et al. (2020) |
| | | | 2 Species: perilla and soybean | $^1$H 43 MHz | BLC | – | – | (Kim et al., 2018), |
| | | | 3 Species: camellia, oriental olive and corn | $^1$H 850 MHz | – | OPLS-DA | 84.1%–90.3% | Xing et al. (2021a) |

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**Table 2** (*continued*)

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| | | | 4 Species: avocado, soybean, corn and rapeseed | $^{1}$H 43 MHz | – | SIMCA | 98% | Jin et al. (2022) |
| | | Fruit | peach puree, 2 Breeds: Spring Lady and Miraflores; pear juice, 3 Breeds: Alejandrina, Conference and Blanquilla | $^{1}$H 400 MHz | BLC | PCA | – | Delpino-Rius et al. (2019) |
| | | | Mango puree, 2 Breeds: alphonso and non-alphonso | $^{1}$H 400 MHz | SG | LOF | 88% | Strecker and Ara (2022) |
| | | | Dry red wine, 3 Breeds: Cabernet Sauvignon, Merlot and Cabernet Gernischt | $^{1}$H 600.23 MHz | NORM | PLS-DA | – | Hu et al. (2020) |
| | | | Grape wine, 8 Breeds: Syrah, Muscat, Xinomavro, Assyrtiko, Malagouzia, Agiorgitiko, Debina and other wine | $^{13}$C 125 MHz | – | KNN, PLS-DA | 83.6% | Mannu et al. (2020) |
| | | Coffee | 2 Breeds: Robusta and Arabica | $^{1}$H 400 Hz | – | LDA | 100% | Badmos et al. (2019) |
| | | Honey | 7 Species: monofloral buckwheat, clover, heather, linden, rapeseed, willow, and polyfloral | $^{1}$H 300 Hz | BLC, NORM | PCA, OPLS-DA | – | Labsvards et al. (2022) |

MIR, mid-infrared; NIR, near-infrared; FT-Raman, fourier transform Raman; HSI, hyperspectral image; THz, Terahertz; NMR, nuclear magnetic resonance; NORM, normalization; SNV, standard normalized variate; 1st, first derivative; 2nd, second derivative; 3rd, third derivative; SG, savitzky-golay; BLC, baseline correction; MSC, multiple scattering correction; MC, mean centering; ISC, inverse scatter correction; PLS-DA, partial least squares discrimination analysis; FDA, factorial discriminant analysis; HCA, hierarchical cluster analysis; LDA, linear discriminant analysis; QDA, quadratic discriminant analysis; OCPLS, one-class partial least squares; PCR, principle component regression; RBF-BPR, radial basis function-biomimetic pattern recognition; PCA, principal component analysis; SVM, support vector machine; OPLS-DA, orthogonal partial least-squares discrimination analysis; RF, random forest; CNN, convolutional neural network; KNN, K-Nearest Neighbor; PLSR, partial least squares regression; SPA, successive projection algorithm; GRNN, generalized regression neural network; DD-SIMCA, data-driven soft independent modeling of class analogy; 3D-CNN, three-dimensional convolutional neural network; LOF, local outlier factor; LS-SVM, least square support vector machine.

example, research on plant-derived foods also focuses on identifying high-value vegetable oils, including perilla oil, argan oil, olive oil, camellia oil, and avocado oil, with discrimination accuracy ranging from 84% to 100% (Table 2). NMR can access differential substances to identify adulterated oils. For example, the primary distinction between argan and sunflower oils is the composition of their mixed triglyceride esters (TAGs) (Gunning et al., 2020). NMR-based non-targeted analysis methods have also been used to differentiate between species or breeds of fruit. Delpino-Rius et al. (2019) developed an identification method for peach puree and pear juice based on primary metabolites and phenolic compounds, identifying two kinds of peaches (Spring Lady and Miraflores) and three kinds of pears (Alejandrina, Conference and Blanquilla). Hu et al. (2020) combined NMR with multivariate statistical analysis to analyze the metabolite content changes in three dry red wines. Although the results showed similar metabolite types in different wines the content levels differed significantly. Therefore, the compound content differences could be used to differentiate between dry red wines. Although studies using NMR to analyze animal-derived food are less common than those involving plant-derived food, this technology has been effectively used for dairy and meat products. In meat products, NMR was used to analyze the pork components in Frankfurt meat products. NMR relaxation was used to identify pork and three otherkinds of meat (beef, chicken, and turkey) using different relaxation times, while their physical properties were measured and analyzed in relation to the relaxation time. The results showed that the optimal frequency for identifying pork in meat products was 22.34 MHz (Uguz et al., 2019). In dairy products, NMR can be used for species identification in raw milk and to analyze cheese samples from different breeds. By combining NMR and Fourier-transform infrared (FTIR) spectroscopic data, the OPLS-DA model showed a 90.54 %t accuracy for identifying halloumi cheese and its two common substitutes (Cheddar and Kefalotyri) (Tarapoulouzi and Theocharis, 2022). Furthermore, a cod liver oil certification study discriminated between cod liver oil and adulterated oils (sunflower and rapeseed oil) with 100% accuracy by combining NMR and support vector machine (SVM) modeling (Giese et al., 2019).

Although MIR is not frequently used, it is mainly utilized for plant-derived products, such as vegetable oil, honey, cereals, and wine. Studies showed that MIR could differentiate between olive oil breeds (Zaroual et al., 2021), while successfully distinguishing between eight edible vegetable oils to address indiscriminate and mislabeled commercial labels (Socaciu et al., 2020). Additionally, MIR can distinguish between different honey and grain species, as well as between different varieties of a single species, with discrimination accuracies ranging from 91% to 100 % (Table 2). MIR also shows potential for species differentiation in aquatic products. A discriminant model capable of distinguishing between four tuna breeds (Yellowfin, Skipjack, Bigeye, and Albacore) was established based on MIR and factorial discriminant analysis. The model assessed the certification of 40 commercial canned tuna products at a discrimination accuracy rate of 93.3–100% (Boughattas and Karoui, 2021). The application of MIR in studying food breeds or species differentiation is relatively mature despite the recent relative paucity of MIR studies.

Raman technology has been used for breeds or species identification in plant-derived products, including grapes, rice, and coffee, and plant sources, such as honey and cereal flour. It is worth mentioning that a study analyzed Raman spectra using SIMCA after pre-processing via multiplicative scattering correction to distinguish between five raw coffee varieties with 100% accuracy. Chlorogenic acid, lipids, and proteins represented the main components responsible for spectral differences (Luna et al., 2019). When applying Raman for cereal flour species classification, spectral differentiation was mainly based on the starch, protein, and arabinoxylan signals, distinguishing between barley, rye, pelt wheat, and wheat flour samples with an accuracy rate of 88% (Kniese et al., 2021). Milk, oil, fish, and meat products are among the animal-derived food products analyzed via Raman spectroscopy for breeds or species differentiation, of which the species origin of milk is the most frequently examined. It can also differentiate milk fats and their low-value substitutes, showing that principal component analysis (PCA) can distinguish between six different non-milk fats. A recent study used Raman spectroscopy to characterize Desi ghee from buffalo and cow milk. The β-carotene, conjugated linoleic acid (CLA), lipid, and fatty acid differences extracted from the spectral data were used to successfully differentiate the Desi ghee species origin (Ahmad and Saleem, 2019). Raman-based aquatic product certification is primarily

concerned with the differentiation of fish breeds. Raman spectroscopy was used to distinguish between two salmon species (rainbow trout and Atlantic salmon) with 87% accuracy (Chen et al., 2019). It was also used to differentiate seven kinds of meat (cattle, sheep, goat, buffalo, chicken, and pork) from salami products in a study on the source identification of raw sausage materials, with an accuracy of 96.26% (Boyaci et al., 2014).

As a special spectroscopic technique, HSI is also gradually being developed in the food authenticity field, primarily employing NIR and VIS-NIR for breeds or species identification in plant-derived products in recent years. Shao et al. (2019) used NIR-HSI to identify millet, corn, and soybean binary mixtures and their pure samples. The PLS-DA results based on the selected effective wavelengths and full spectra showed that the discrimination rates of all the models exceeded 94.8%. Papaya seeds and black pepper can also be identified using NIR-HIS. In a study involving maize seed identification, Zhang et al. (2022a) successfully distinguished five maize breeds using an HSI system based on VIS-NIR spectroscopy, obtaining accuracy rates of 96%–100% after algorithm analysis. VIS-NIR-HSI has achieved varying degrees of success in identifying plant-derived products, including almonds and peanuts, and the identification of the plant origin of honey. Moreover, recent studies have employed spectral modules based on VIS-NIR-HSI to identify breeds or species and the animal origin in meat products, such as pork, pork, lamb, chicken, and duck, with discrimination rates exceeding 95% (Al-Sarayreh et al., 2020; Jiang et al., 2021, 2022).

THz research mainly focuses on plant-derived foods, including corn, rice, and other cereals. In a study identifying maize breeds, the prediction accuracy of the model with a 0.2–1.6 THz spectrum reached 92.08% (Yang et al., 2021a). THz has also been used to distinguish the botanical origin of honey and identify transgenic soybean seeds (Liu et al., 2016a, 2018b). These two studies focused on chemometric method selection. The PLS-DA model was more suitable for identifying the botanical origin of honey, with a verification set accuracy of 88.46%, while the LS-SVM model, was more appropriate for transgenic soybean identification, with a verification set discrimination accuracy of 88.33%. This demonstrates that selecting the appropriate chemometrics techniques also affects the outcome, in addition to selecting the correct analytical technology.

#### 2.1.2. Quality fraud

Quality fraud identification is also a part of practical significance in the food authenticiy field, and the market demand is stronger. Therefore, the application of spectral technology with the main advantage of rapid has been continuously explored in recent years, with specific applications focusing on the identification of illegal additives, farming or seed cultivation methods, and content falsification. MIR, NIR, Raman, and NMR are relative maturity technologies in quality fraud identification. Relevant literatures are summarized in Table 3.

Regarding the quality fraud identification of animal-derived foods, the application of NIR in recent years has been mainly for meat and meat products, but for different purposes of differentiation, including differentiation of storage methods, differentiation of processing methods and differentiation of quality levels. Combining a handheld NIR with the Random Subspace Discriminative Ensemble (RSDE) method effectively differentiated between fresh and frozen chicken and classified chicken fillets according to the different farming conditions, with a classification accuracy exceeding 95% (Parastar et al., 2020). The presence or absence of irradiated samples in sausage samples can also be identified via NIR with 100% accuracy (Varra et al., 2020). Furthermore, NIR shows considerable potential for distinguishing ham grades. NIR was used for the in situ measurements of carcass fat to differentiate between premium and non-premium hams, with a classification accuracy of more than 95% (Piotrowski et al., 2019). In addition, NIR was used to identify the quality of eggs to differentiate the feeding practices of hens. Egg whites and yolks were classified to trace the egg sources (free range and cage) based on the spectral analysis of the protein, carbohydrate, and ash content differences, providing a theoretical reference for evaluating egg quality (Hoffman et al., 2022). Nevertheless, for quality fraud identification of plant-derived foods, NIR focuses on the identification of the growth and feeding system of the product. It was used to trace product cultivation systems, successfully differentiated between conventional and organic rice, tomatoes and bell peppers with classification accuracies of 87.5%, 98.5%, 96.3% (de Andrade et al., 2023; Xiao et al., 2019). Additionally, NIR could distinguish between specialty coffee beans produced using the standard procedure and non-specialty coffee beans produced with inadequate control over the production process, with a classification accuracy of 87% (Manuel et al., 2022).

MIR is a spectroscopic technology second only to NIR for quality fraud studies. Recent food quality fraud identification research involving MIR mainly focuses on plant-derived foods to identify the presence of illegal additives. MIR identified illegal additives in paprika powder, such as Sudan I, Sudan IV, and lead chromate, with a sensitivity and specificity exceeding 80% (Horn et al., 2018). Syrup added to samples as an adulterant was also accurately identified. MIR recognized four types of adulterated syrups in honey (glucose, fructose, sucrose, and high syrup fructose corn syrup)with detection limits as low as 10% (Skaff et al., 2022). The combination of MIR and K-nearest neighbor (KNN) identified syrup or water in Guava pulp with 100% classification accuracy (Alamar et al., 2020). In both these studies, the MIR discrimination accuracy was higher than NIR, suggesting that MIR displayed higher potential for analyzing syrup adulteration scenarios. MIR was also used to classify chocolate samples with different cocoa content levels with 99.67% accuracy (Santos et al., 2021). MIR is mainly used to identify illegal additives when determining the quality of animal products, such as beef and bird's nests. The chemical components, such as NaCl, phosphates, carrageenan, and maltodextrin, doped in beef samples were characterized by selecting specific infrared bands, with the doping identification rate reaching 91% (Nunes et al., 2016). For the study on bird's nests, MIR identification model correctly classified 100% carrageenan adulteration, followed by the nutrient agar class at 98.2%, gelatine class at 97.3%, and collagen class at 94.4% (Ng et al., 2022).

Using Raman, NMR, and Thz techniques to identify quality fraud in animal-derived products focuses on dairy products, such as milk, cheese, and milk powders. These methods aim to identify illegal additions and differentiate feeding practices. Raman spectroscopy successfully identified illegal additives (margarine, corn and palm oils) in cheese, with detection limits as low as 4% (Genis et al., 2021). The H-1-NMR metabolomics method was used to collect the non-volatile metabolite profiles of organic and conventional liquid milk. Metabonomic data analysis indicated that 13 potential biomarkers, such as formate and betaine, could be used to identify liquid milk production systems (Phuenpong et al., 2021). Quality fraud identification in dairy products based on THz focuses onidentifying melamine in milk powder. Combined with the analysis algorithm, the maximum absorption peak reached 2.04 THz, while the absorption coefficient increased at a higher melamine concentration. The mixed logistic regression (MLR) model based on THz displays significant potential or quantitatively analyzing melamine in milk powder (Sun et al., 2019). Furthermore, THz technology has also been applied for quality fraud identification in plant-derived foods. It was combined with PCA and SVM to differentiate edible oils with similar appearance and physical properties from swill dirty oils, displaying a classification accuracy of 100% (Zhan et al., 2016). Compared with THz, Raman spectroscopy and NMR are well-established methods for analyzing plant-derived food samples. Raman spectroscopy can identify diluted coconut water, syrup-doped honey, and rapeseed oil squeezed in different ways, with an accuracy ranging from 93% to 100 % (Aykas et al., 2020; McDowell et al., 2018; Richardson et al., 2019). NMR can identify quality fraud in a variety of plant-derived foods, such as liquid honey, solid coffee, and chili powder. Regardless of whether identifying syrup in honey, coffee from different cultivation systems, or chemical synthetic adulterants in paprika powder, NMR uses baseline correction (BLC) for pre-processing, which at least shows that BLC is the first spectral pre-treatment method that can be considered when using NMR to identify adulterations in

**Table 3**
Application progress of spectroscopic technology in quality fraud identification of food.

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate/limits of detection | References |
|---|---|---|---|---|---|---|---|---|
| MIR | Animal | Meat | Beef, Illegal addition: sodium chloride, phosphate, carrageenan, maltodextrin | 4000–525 $cm^{-1}$ | 2nd, SG, MSC | PCA, PLS-DA | 91% | Nunes et al. (2016) |
| | | Edible bird's nests | Illegal addition: Melamine, karaya gum, nutrient agar, collagen, gelatine | 16,667–2500 nm | – | PCA, OPLS-DA | 94.4% | Ng et al. (2022) |
| | Plant | Vegetable | Paprika, Illegal addition: gum arabic, lead chromate, lead (II, IV) oxide, polyvinyl chloride (PVC), silicon dioxide, Sudan I, and Sudan IV | 4000–400 $cm^{-1}$ | SNV, SG, 1st, 2nd | SIMCA | – | Horn et al. (2018) |
| | | Honey | Illegal addition: glucose, fructose, sucrose, and high syrup fructose corn syrup | 4000–500 $cm^{-1}$ | SG | PCA | – | Skaff et al. (2022) |
| | | Guava pulp | Illegal addition: sugar and water | 4000–400 $cm^{-1}$ | MSC, SNV, SG | KNN | 100% | Alamar et al. (2020) |
| | | Chocolate | Content falsification: cocoa content in chocolates | 4000–600 $cm^{-1}$ | SNV | PLSR | 99.67% | Santos et al. (2021) |
| NIR | Animal | Meat | Chicken, Storage methods: fresh and frozen-thawed | 908–1676 nm | – | RSDE | > 95% | Parastar et al. (2020) |
| | | | Dry fermented sausages, Processing methods: irradiated and non-irradiated | 1000–2500 nm | SNV, SG, 2nd | PCA, OPLS-DA | 100% | Varra et al. (2020) |
| | | | Quality grades: premium and non-premium | 908–1676 nm | 2nd, SG, SNV | LDA, QDA, NPB | > 95% | Piotrowski et al. (2019) |
| | | Egg | Feeding methods: cage and free-range | 950–1600 nm | SG | PCA, LDA | 86%–92% | Hoffman et al. (2022) |
| | Plant | Rice | Cultivated methods: organic and conventional | 12,000–4000 $cm^{-1}$ | SG, SNV, 2nd | PLS-DA | 87.5% | Xiao et al. (2019) |
| | | Vegetable | Tomato and sweet pepper, Cultivated methods: organic and conventional | 900–1650 nm | SG, SNV, MSC | PLS-DA | 98.4%, 96.3% | (de Andrade et al., 2023) |
| | | Coffee | Quality grades: special and non-specialty agroforestry | 937–1655 nm | BLC | PCA, HCA, DD-SIMCA | 87% | Manuel et al. (2022) |
| Raman/FT-Raman | Animal | Milk | Cheese, Illegal addition: margarine, and corn and palm oil | 2000–200 $cm^{-1}$ | 1st, SG | PLS-DA, PLS | 100% | Genis et al. (2021) |
| | Plant | Fruit | Coconut water, Content falsification: coconut water by dilution | 2579–408 $cm^{-1}$ | SG, BLC | PLSR | 97%–99% | Richardson et al. (2019) |
| | | Honey | Illegal addition: molasses, date molasses, grape molasses, high fructose corn syrup, corn syrup, sucrose, and inverted sugar | 2500–200 $cm^{-1}$ | SG, MC | SIMCA | 100% | Aykas et al. (2020) |
| | | Oil | Cold pressed rapeseed oil, Illegal addition: refined rapeseed oil and refined sunflower oil | 1800–800 $cm^{-1}$ | SG, 1st | LDA | 93% | McDowell et al. (2018) |
| HSI | Animal | Meat | Pork, Processing methods: minced jowl and pure | 400–1000 nm | NORM, SNV, MSC, 1st, 2nd | PLSR | 90.63% | Jiang et al. (2020) |
| | | | Porcine dorsi muscles, Storage methods: fresh and frozen-thawed | 900–1700 nm | – | PLS-DA | 100% | Barbin et al. (2013) |
| | Plant | Black tea | Quality grades: 1-7 | 350–1100 nm | SG, SNV | RF | 92.7% | Ren et al. (2021) |
| THz | Animal | Milk | Illegal addition: melamine | 0.75–2.73 THz | – | MLR | 97% | Sun et al. (2019) |
| | Plant | Oil | Illegal addition: Swill-cooked dirty oil | 0.2–1.3 THz | – | PCA, SVM | 97.3% | Zhan et al. (2016) |
| | | Honey | Illegal addition: invert syrup | 0.3–1.5 THz | the complex dielectric constant (Re[ε] and Im[ε]) | PLS | – | Liu et al. (2022b) |
| NMR | Animal | Aquatic product | Little yellow croaker, Illegal addition: carrageen or distilled water | – | – | PLSR | 98.77% | Zang et al. (2017) |
| | | Milk | Feeding methods: organic and conventional | $^{1}H$ 500 MHz | – | HCA, PLS-DA | – | Phuenpong et al. (2021) |
| | Plant | Vegetable | Paprika powder, Illegal addition: azorubine, ponceau 4R, beetroot and sumac powder | $^{1}H$ 400 MHz | BLC | DD-SIMCA | 92% | Horn et al. (2021) |

(continued on next page)

**Table 3** (*continued*)

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate/limits of detection | References |
|---|---|---|---|---|---|---|---|---|
| | | Honey | Illegal addition: barley, rice and corn syrups | $^{1}$H 400 MHz | BLC | PCA, PLS | – | Lozano-Torres et al. (2022) |
| | | Coffee | Cultivated methods: organic and conventional | $^{1}$H 400 MHz | BLC | PLS-DA, OPLS-DA, OSC | – | Consonni et al. (2018) |

MIR, mid-infrared; NIR, near-infrared; FT-Raman, fourier transform Raman; HSI, hyperspectral image; THz, Terahertz; NMR, nuclear magnetic resonance; NORM, normalization; SNV, standard normalized variate; 1st, first derivative; 2nd, second derivative; SG, savitzky-golay; BLC, baseline correction; MSC, multiple scattering correction; MC, mean centering; PLS-DA, partial least squares discrimination analysis; HCA, hierarchical cluster analysis; LDA, linear discriminant analysis; QDA, quadratic discriminant analysis; PCA, principal component analysis; SVM, support vector machine; OPLS-DA, orthogonal partial least-squares discrimination analysis; RSDE, random subspace discriminant ensemble; RF, random forest; ELM, extreme learning machine; PLSR, partial least squares regression; NPB, nonparametric bayes; DD-SIMCA, data-driven soft independent modeling of class analogy; MLR, mixed logistic regression.

plant-derived foods (Consonni et al., 2018; Horn et al., 2021; Lozano-Torres et al., 2022).

Compared to other spectroscopic techniques, there have been relatively few reports in quality fraud identification in recent years. HSI allows more detailed sample study by simultaneously measuring spectral and spatial information, often using NIR as the spectral module. HSI mainly differentiates food processing methods, storage techniques, and quality levels. HSI was combined with VIS-NIR to analyze minced pork in pork products by building a PLSR model using the acquired spectral image data, with an R2 reaching 0.9063 (Jiang et al., 2020). Fresh and freeze-thawed porcine dorsal muscles were analyzed using HSI with integrated NIR, obtaining 100% differentiation accuracy via the PLS-DA model (Barbin et al., 2013). Regarding the identification of quality grades, VIS-NIR-HSI was combined with the random forest (RF) algorithm to identify seven black tea quality grades, achieving an accurate identification rate of 92.7% (Ren et al., 2021).

### 2.1.3. Geographical origin

Generally, the application of spectroscopy in the geographical origin identification of food has been mainly focused on plant-derived foods in recent years. And the regional scope of geographic traceability includes different countries, different regions of the same country, and even smaller regions of the same region. Among them, MIR, NIR, HSI, and NMR have relatively more research results. Relevant literatures are summarized in Table 4.

Similar to other aspects of food authenticity research, NIR is the most commonly used method for geographic origin identification in plant-derived foods, such as cereals and vegetables. The geographic sources of cereal from different countries, including rice (Teye et al., 2019), durum wheat (De Girolamo et al., 2019), grain maize (Schuetz et al., 2022) and mung bean (Qian et al., 2022), were determined with discrimination accuracy values ranging from 90% to 100%. Vegetable product studies have shown that spectral classification accuracy requires selecting suitable, reliable prediction models. NIR was combined with DD-SIMCA to differentiate tomatoes and bell peppers from three Brazilian regions with an accuracy of 82.7 % (de Andrade et al., 2023). The PLS-DA model yielded unsatisfactory results between 61.9 % and 100%, with a high degree of accuracy variability. Combining NIR and SVM successfully classified 97% of European and non-European white asparagus samples (Richter et al., 2019). The research on the geographical origin differentiation of animal-derived food products via NIR mainly concentrates on dairy products and seafood. Zhang et al. (2022b) distinguished the geographical origins of milk from five Chinese provinces by combining NIR with KNN, showing a discrimination rate of 98.67%. Curro et al. (2021) used NIR to analyze the geographical origin of cuttlefish from five FAO (Adriatic Sea, northeastern and eastern central Atlantic Oceans, and eastern Indian and western central Pacific Oceans) fishing regions, with an accuracy reaching 92% after SVM algorithm analysis.

MIR, which has a different wavelength range than NIR, is more mature regarding origin traceability and is mainly used to differentiate the geographical origin of plant-derived foods, such as fruits and vegetables. The study of fruit is primarily concerned with determining the geographical origin of wine. In the study of distinguishing the geographical origin of grapes, MIR combined with PLS-DA was used to model the different geographical origins of Chardonnay grapes in South Australia, and the overall success rate in discriminating geographical origin for samples from different vintages (2014 and 2016) was 83% and 81%, respectively (Gambetta et al., 2019). For vegetables, MIR was used to differentiate lentils from two countries and red chili powder from three countries at 100% accuracy (Innamorato et al., 2019; Kim et al., 2021). Research involving the utilization of MIR for the geographical origin differentiation of animal-derived foods mainly focuses on milk and dairy products, such as goat milk and Alpine milk. Caredda et al. (2017) used fatty acid composition and MIR to analyze goat milk from three regions in Sardinia (north, middle, and south), obtaining excellent classification accuracy of 96% and 99%, respectively, with MIR showing slightly higher efficacy. NMR is also used for dairy origin identification. Haddad et al. (2022) quantified 178 peaks obtained via NMR analysis of cheese. They constructed a multivariate model to quantify a single fatty acid, successfully distinguishing the geographical origin of cheese. The remainder of the NMR research primarily focused on plant-derived foods, with geographical origins mainly represented by different regions in a country, including wines from two Chinese provinces and dark chocolate from four Chinese cities. NMR identified and of quantified various compounds, including 33 metabolites in the wine and 42 chemical components in the chocolate, using the related variation to distinguish the geographical origin of these products (Gougeon et al., 2018; Le Gresley and Peron, 2019).

Since HSI technology can obtain both internal composition and appearance-related information about food products, often revealing sample origin variation, it has been developed for distinguishing geographic origin, mainly of plant-derived products. HSI typically employs NIR as the spectral module. NIR-HIS was used to trace the origin of chia seeds, peaches, foxtail millet, Chinese chestnuts, and other food products, with discrimination accuracy values exceeding 90% (Table 4). This method was also used to differentiate mutton from four Chinese provinces Weng et al. (2021) achieved the best discrimination of mutton by combining RF with NIR-HSI to acquire effective spectral and image information of textural features, obtaining calibration and prediction set classification accuracies of 99.54% and 95.67%, respectively. The study also demonstrated the feasibility of using HSI to trace the origin traceability of animal-derived products. In addition, recent studies on geographical origin identification using Raman and THz techniques only focus on plant-derived products. Although Raman spectroscopy distinguished the geographical origin of rice from 12 Chinese provinces and three Chinese cities, the discrimination rates were different, with the former reaching 100 % and the latter reaching a maximum of 88.9%, possibly because the urban areas were smaller than the provincial regions (Sha et al., 2019; Zhu et al., 2018). And as for the application of THz, Liu et al. (2018a) assessed the feasibility of using THz to quickly distinguish EVOO from four geographical sources. The results indicated

**Table 4**
Application progress of spectroscopic technology in geographical origin identification of food.

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| MIR | Animal | Milk | Goat milk, 3 Regions in Sardinia: North, Central and South | 5011.54–925.92 $cm^{-1}$ | SNV | GA-LDA | 99% | Caredda et al. (2017) |
| | | | 3 Regions: South Tyrol, Tyrol, and EU Regions | 4000–400 $cm^{-1}$ | – | PLS-DA | 95% | Scampicchio et al. (2016) |
| | Plant | Fruit | Grape, wine 4 wine regions in Portuguese: Vinhos Verdes, Lisboa, Açores and Távora-Varosa | 3050–950$cm^{-1}$ | SG, SNV | PLS-DA | 87.7% | dos Santos et al. (2017) |
| | | | Grape wine, 6 Regions: Adelaide Hills, Barossa V alley, Clare V alley, Eden V alley, Langhorne Creek and Riverland | 1500–800$cm^{-1}$ | SG, 2nd | PLS-DA | 81%–83% | Gambetta et al. (2019) |
| | | Vegetable | Lentil, 2 Countries: Italy and Canada | 4000–400 $cm^{-1}$ | SNV, MSC | PCA, WPT-LDA, PLS-DA | 91%–100% | Innamorato et al. (2019) |
| | | | Asian red pepper powders, 3 Countries: Korean, Chinese, and Vietnamese | 4000–400 $cm^{-1}$ | SG, 2nd | CDA | 100% | Kim et al. (2021) |
| | | Oil | Olive oil, 5 Regions in Moroccan: Fez/Meknes, Marrakech/Safi, Eastern, Northern, Beni-Mellal/ Khenifra | 4000–700 $cm^{-1}$ | NORM, 2nd, MSC | PCA, FDA | 91.87% | Zaroual et al. (2021) |
| | | Honey | 2 Regions: Maltese Islands and non-Maltese Islands | 4000–550 $cm^{-1}$ | MSC, OSC, SNV, | PLS-DA | > 95% | Formosa et al. (2020) |
| | | Quinoa grains | 3 Countries: Argentina, Chilean and Postosi Bolivia | 4000–600 $cm^{-1}$ | NORM, BLC | SIMCA | 96% | Rodriguez et al. (2019a) |
| NIR | Animal | Milk | 5 Provinces in China: Heilongjiang, Henan, Hebei, Inner Mongolia, Ningxia | 900–1700 nm | SG | KNN | 98.67% | Zhang et al. (2022b) |
| | | | Cheese, 3 Regions in the State of Bahia: northeast, far west and south | 1100–2500 nm | SNV | PCA, LDA | 90% | Silva et al. (2021) |
| | | Aquatic product | Anchovie, 4 Countries: Morocco, Spain, Tunisia, and Croatia | 1000–2500 nm | MSA, 2nd, SG | OPLS-DA | > 99% | Varra et al. (2021) |
| | | | Cuttlefish, 5 Fishing FAO areas: Adriatic Sea, northeastern and eastern central Atlantic Oceans, and eastern Indian and western central Pacific Oceans | 902–1680 nm | SNV, SG | SVM | 92% | Curro et al. (2021) |
| | | | Sea cucumber, 9 aquacultures–3 Regions: Bohai Sea, Yellow Sea and East China Sea | 10,000–4000 $cm^{-1}$ | – | light GBM | 91% | Sun et al. (2021b) |
| | Plant | Cereal | Rice, 3 Countries: Ghana, Thailand, and Vietnam | 740–1070 nm | MSC | PCA | 90% | Teye et al. (2019) |
| | | | Durum wheat, 9 Countries: Italy, Australia, Canada, France, Greece, Russia, Spain, Turkey, and the United State | 10,000–4000 $cm^{-1}$ | NORM, SNV | LDA | 100% | De Girolamo et al. (2019) |
| | | | Grain maize, 5 Countries: Spain, Ukraine, Slovakia, Peru and the USA | 10,000–4000 $cm^{-1}$ | SNV | SVM | 95% | Schuetz et al. (2022) |
| | | | Mung bean, 4 Counties: Durbert Mongolian Autonomous County and Baicheng, Tailai and Chifeng | 10,000–4000 $cm^{-1}$ | NORM | PLS-DA | 90%–96.67% | Qian et al. (2022) |
| | | Vegetable | Tomato and sweet pepper, 3 Cities in Brazilian: Londrina, Rio de Janeiro, and São Paulo | 900–1650 nm | SG, SNV, MSC | DD-SIMCA | 82.7% | (de Andrade et al., 2023) |
| | | | White asparagus, 2 Regions: German and non-German | 11,500–4000 $cm^{-1}$ | MSC, SG | SVM | 97% | Richter et al. (2019) |
| | | Fruit | Durian, 2 Provinces in Thailand: Prachuap Kiri Khan and Chanthaburi | 12,500–4000 $cm^{-1}$ | SG, 2nd | SIMCA | 100% | Chanachot et al. (2021) |
| | | Coffee | 2 Regions in Vietnam: Dak Lak and non-Dak Lak | 900–1700 nm | SNV | PLS-DA | 92% | Minh et al. (2022) |
| | | Chestnut | 3 Regions in Italy: Viterbo, Vallerano and Solofra | 10,000–4000 $cm^{-1}$ | SNV, 2nd, MC | PLS-DA | 97% | Nardecchia et al. (2020) |
| | | Oil | Olive oil, 19 countries | 11,500–4000 $cm^{-1}$ | 1st, NORM | LDA | 80%–100% | Gertz et al. (2019) |
| | | Honey | 6 Regions in Hungary: Great Plain, Northern Mountains, Small Plain, Transdanubian Hills, Transdanubian Mountains, Western Hungary | 740–1700 nm | SG | PCA, LDA | 99% | Bodor et al. (2021) |
| Raman/FT-Raman | Plant | Cereal | Rice, 3 Cities in China: Wuchang, Yanbian, Panjin | 2339–250 $cm^{-1}$ | SNV, MSC | PCA, SVM | 71.4%–88.9% | Sha et al. (2019) |

(*continued on next page*)

**Table 4** (*continued*)

| Technology | Product category | Food product | Adulterants/classified product | Instrument parameter range | Spectral pretreatment | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| | | | Rice, 12 provinces in China: Heilongjiang, Jiangsu, Jilin, Shandong, Henan, Tianjin, Anhui, Hunan, Guangdong, Guangxi, Hainan and Anhui | 3900–100 $cm^{-1}$ | 1st | SIMCA | 100% | Zhu et al. (2018) |
| | | Oil | Olive oil, 2 Regions: European and non-European | 3600–50 $cm^{-1}$ | MSC | PLS-DA | 80%, 82% | Tena et al. (2019) |
| | | Honey | 2 Countries: Romania and France | 1900–200 $cm^{-1}$ | NORM | SIMCA | 100%, 89% | Magdas et al. (2021) |
| | | Fruit | Grape wine, 3 Regions in Romanian: Transylvania, Banat and Moldova | 3600–1000 $cm^{-1}$ | – | SLDA | 100% | Magdas et al. (2018) |
| HSI | Animal | Meat | Mutton, 4 Provinces in China: Xinjiang, Inner Mongolia, Ningxia and Guangxi | 400–1000 nm | MSC | RF | 99.54% | Weng et al. (2021) |
| | Plant | Fruit | Yangshan peach, 2 Cities in China: Yangshan and Nanjing | 400–1000 nm | – | GSR | 100% | Sun et al. (2021a) |
| | | | Dry Narrow-Leaved Oleaster Fruits, 3 Provinces in China: Gansu, Ningxia and Xinjiang | 874–1743 nm | 2nd | PLS-DA, SVM, KNN | 90% | Gao et al. (2019) |
| | | Cereal | Foxtail millet, 4 Cities in Inner Mongolia: Chifeng, Bayannur League, Hohhot, and Hinggan League | 900–1700 nm | SNV | PCA, SVM | 95% | Wang et al. (2022a) |
| | | | Rice, 2 Countries: South Korean and Chinese | 420–780 nm | SG, 1st, 2nd | PLS-DA | 95% | Kim et al. (2020) |
| | | Chia seeds | 3 Countries: Argentina, Paraguay, and Bolivia | 900–2500 nm | MSC | PLSR | – | Choi et al. (2021) |
| | | Chinese Chestnuts | 3 Chinese provinces: Hebei, Liaoning and Yunnan | 383.4–990.4 nm | SNV | 1D-CNN | 97.12% | Li et al. (2021b) |
| | | Wolfberry | 4 Regions in Ningxia, China: Huinong, Tongxin, Guyuan and Zhongning | 400–1000 nm | CV | 2D-CNN | 97.4%–99.5% | Hao et al. (2022) |
| THz | Plant | Scutellaria baicalensis | 3 Provinces in China: Inner Mongolia, Shanxi and Shaanxi | 0.2–1.7 THz | – | PCA, SVM | 95.56% | Liang et al. (2018) |
| | | Olive oil | 4 Countries: Australia, Spain, Greece and Italy | 0.1–4.0 THz | – | LS-SVM | 96.25% | Liu et al. (2018a) |
| | | Coffee | 3 Regions: Kenya, Tanzania and Yunnan | 0.5–1.9 THz | – | CNN | 90%–100% | Yang et al. (2021b) |
| NMR | Animal | Milk | Cheese Cow from 6 Countries: Bulgaria, France, Germany, Hungary, Italy, and Netherlands; Goat from 2 Countries: France and Spain; Sheep from 2 Countries: Bulgaria and Italy | $^{1}H$ 400.13 MHz | BLC | CDA, LDA | – | Haddad et al. (2022) |
| | Plant | Fruit | Grape wine, 2 Provinces in China: Shanxi and Ningxia | $^{1}H$ 600 MHz | BLC | PCA | – | Gougeon et al. (2018) |
| | | | China's sweet orange, 4 Provinces in China: Hunan, Hubei, Sichuan and Guangxi | $^{1}H$ 600 MHz | NORM | PCA, PLS-DA, OPLS-DA | – | Lin et al. (2021) |
| | | | Coffee, 4 Cities: Minas Gerais, Bahia, São Paulo, and Paraná | $^{1}H$ 600 MHz | – | PCA, DA | – | Toci et al. (2018) |
| | | Vegetable | Asparagus, 6 Countries: Germany, Poland, The Netherlands, Spain, Greece, and Peru | $^{1}H$ 400 MHz | BLC | PCA, SVM | 87.8%–91.5% | Klare et al. (2020) |
| | | Oil | Olive oil, 4 Cities in Huelva (southwest Spain): Beas, Gibraleón, Niebla, Sanlúcar de Guadiana | $^{1}H$ 500 MHz | – | LDA | 100% | Sayago et al. (2019) |
| | | Honey | 2 Regions: Italian and Eastern European | $^{1}H$ 600 MHz | MC | PLS-DA | 100% | Schievano et al. (2019) |
| | | Dark chocolate | 3 Countries: Peru, Madagascar and Venezuela | $^{1}H$ 600 MHz | – | PLS-DA | – | Le Gresley and Peron (2019) |

MIR, mid-infrared; NIR, near-infrared; FT-Raman, fourier transform Raman; HSI, hyperspectral image; THz, Terahertz; NMR, nuclear magnetic resonance; EVOO, extra-virgin olive oil; NORM, normalization; SNV, standard normalized variate; 1st, first derivative; 2nd, second derivative; SG, savitzky-golay; BLC, baseline correction; MSC, multiple scattering correction; MC, mean centering; iVISSA, interval variable iterative space shrinking analysis; PLS-DA, partial least squares discrimination analysis; FDA, fisher discriminant analysis; LDA, linear discriminant analysis; PCA, principal component analysis; SVM, support vector machine; OPLS-DA, orthogonal partial least-squares discrimination analysis; RF, random forest; light GBM, light gradient boosting machine; KNN, K-Nearest Neighbor; PLSR, partial least squares regression; CDA, canonical discriminant analysis; OSC, orthogonal signal correction; CNN, convolutional neural network; 1D-CNN, one-dimensional convolutional neural network; 2D-CNN, two-dimensional convolutional neural network; SLDA, stepwise discriminant analysis; GSR, group sparse representation classifier; WPT-LDA, wavelet packet transform + linear discriminant analysis; GA-LDA, genetic algorithm + linear discriminant analysis; LS-SVM, least square support vector machine.

that combining LS-SVM with a genetic algorithm (GA) achieved better classification, with a prediction concentration accuracy rate of 96.25%.

In conclusion, spectral analysis, as one of the more established categories of rapid analysis techniques, has been frequently seen in the field of food authenticity in recent years. According to the literature survey, the food authenticity issues based on the number of reports are, in descending order, breeds or species identification, quality fraud identification, and geographical origin identification. However, based on the literature survey, spectroscopic techniques have high variability in correct classification. This is because the results of spectroscopic techniques are susceptible to issues such as sample morphology and data processing methods. Therefore, subsequent studies should avoid these issues in the experimental design.

### 2.2. Ambient ionization mass spectrometry

Ambient ionization mass spectrometry (AIMS) can rapidly analyze pristine samples in atmospheric conditions, often with minimal or no sample preparation, and has the unique advantage of directly analyzing intact substances (Huang et al., 2010). It is gradually gaining popularity among experts in various fields due to rapid analysis while ensuring chemical sensitivity and accuracy. Representative AIMS technologies for the analysis of food authenticity mainly include direct analysis in real-time mass spectrometry (DART-MS), rapid evaporation ionization mass spectrometry (REIMS), matrix-assisted laser desorption/ionization-time of flight mass spectrometry (MALDI-MS), and soft ionization by chemical reaction in transfer mass spectrometry (SICRIT-MS) (Fig. 1). Instead of lengthy column separations, these techniques take seconds to minutes to achieve on-site analysis, significantly reducing analysis time and meeting the market demands for authentication. Relevant literatures are summarized in Table 5.

#### 2.2.1. Direct analysis in real-time mass spectrometry

DART-MS is an atmospheric pressure thermal desorption ionization method without a mobile phase and surface contact. During operation, gas flows through the DART-ion source to the sample surface to promote the thermal desorption of the surface analytes, followed by sample ion MS, allowing high-throughput sample measurement in a short time with little or no sample pre-treatment (Qie et al., 2022). DART-MS has attracted increasing research attention for food authentication due to its high efficiency and stability. In the published reports, studies on the identification of varieties or species have been conducted on plant-derived food. Combining DARA-MS with the PLS-DA algorithm differentiated wheat breeds with 90% accuracy (Miano et al., 2018) and cannabis breeds with 99% accuracy (Dong et al., 2019a). The results were compared with LC-MS, showing that DART-MS can obtain discrimination rates close to those of LC-MS in a short time. In the identification of food quality fraud, DART-MS mainly distinguishes the farming or cultivation methods of samples to identify whether there is the substitution of low-value products to high-value products. DART-MS combined with PCA could distinguish between wild and farmed salmon with 100% accuracy (Fiorino et al., 2019). Metabolomic differences in milk were analyzed using a combination of DART-MS and the PLS-DA model, differentiating between cow feeding practices, with 98% accuracy for maize silage and crop silage/hay and 100% accuracy for grassland hay (Riuzzi et al., 2021). In addition, the feasibility of DART-MS to differentiate the geographical origin of animal-derived products was confirmed by Qie et al. DART-MS was used to collect metabolomics data from lambs in four regions. Differences were identified and analyzed in conjunction with LDA, achieving a discrimination accuracy of 82.5% (Qie et al., 2022).

#### 2.2.2. Rapid evaporation ionization mass spectrometry

REIMS enables handheld sampling. During data collection, appropriate sampling equipment and optimal instrument settings are selected based on the nature of the sample. The entire fingerprinting process takes only 1–2s while analyzing landmark components can be achieved combining MS (Balog et al., 2016). Nowadays, REIMS-based

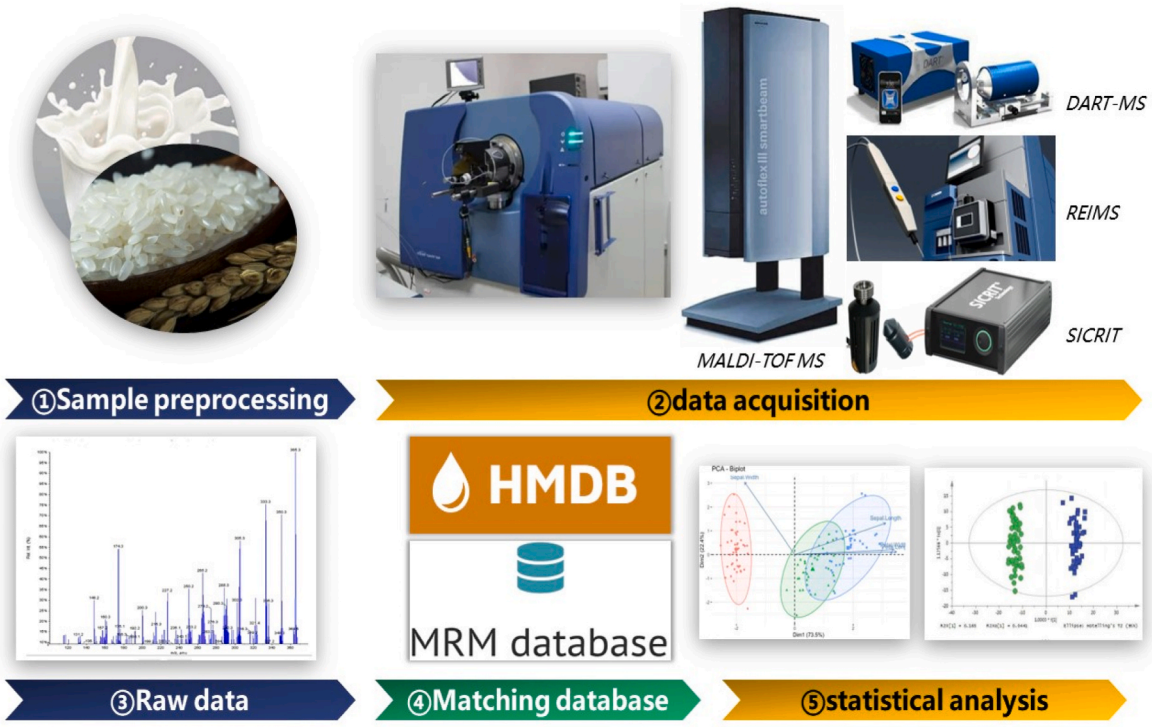


**Fig. 1.** Application of in-situ mass spectrometry in authenticity analysis of food
DART-MS, direct analysis in real time mass spectrometry; REIMS, rapid evaporation ionization mass spectrometry; MALDI-TOF-MS, matrix-assisted laser desorption/ionization time of flight mass spectrometry; SICRIT, soft ionization by chemical reaction in transfer.

**Table 5**
Application progress of AIMS in authenticity analysis of food.

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Instrument mode | mass spectra | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|---|
| Breeds/Species identification | DART-MS | Plant | Cereal | Wheat, 3 Breeds: durum, common and hulled wheat | positive ionization mode | m/z 75–1125 Da | PCA, PLS-DA | 90% | Miano et al. (2018) |
| | | | Cannabis hemp | 4 Breeds: Cherry, Cherry blossom, Eletta and Carmagnola | positive ionization mode | m/z 100–1000 | PCA, PLS-DA | 99% | Dong et al. (2019a) |
| | | | Oregano | 2 Species: oregano and olive leaves | positive ionization mode | m/z 100–1000 | PCA, SIMCA | 90% | Damiani et al. (2021) |
| | REIMS | Animal | Aquatic product | Salmon, 2 Breeds: Salmon and Rainbow Trout | positive and negative ionization mode | m/z 50–1200 | PCA, OPLS-DA | 96.58% | Song et al. (2019) |
| | | | | Fish, 18 Breeds | negative ionization mode | m/z 150–1500 | PCA, LDA | 99% | Rigano et al. (2019a) |
| | | | Meat | 3 Species: horse, cattle, and venison | negative ionization mode | m/z 150–1500 | PCA-LDA | 100%, 97% | Balog et al. (2016) |
| | | | Milk | 3 Species: goat, buffalo, Holstein cow, and Jersey cow milk | positive ionization mode | m/z 200–1000 | PCA, OPLS-DA, LDA | 100% | Cui et al. (2022) |
| | MALDI-MS | Animal | Milk | Cheese, 4 Species: goat, sheep, cow, and buffalo | positive linear mode | m/z, 2000–20,000 Da | – | – | Rau et al. (2020) |
| | | | | Cheese, 2 Species: cow and feta | negative ion mode | m/z 3.5–40 kDa | PLS-DA | 83.5% | Kritikou et al. (2022) |
| | | | Meat | 4 Species: pork, chicken, duck and beef | positive ion mode | m/z, 3000–22000Da | PCA, PLS-DA | 94.7% | Pu et al. (2022) |
| | | | Edible insects | 4 Species: buffalo worms, mealworms, crickets and grasshoppers | positive linear mode | m/z 2–20 kDa | – | – | Ulrich et al. (2017) |
| | | | Aquatic product | Fish, 8 Breeds: Brama japonica, Pampus argenteus, Zeus faber Linnaeus, *Oreochromis mossambicus*, *Mugil cephalus*, Epinephelus rivulatus, Larimichthys crocea and Larimichthys polyactis | positive linear mode | m/z 800–20,000 | PCA | – | Shao and Bi (2020) |
| | | Plant | Cereal | Barley, 8 Species: Kangoo, Laudis, Malz, Marthe, Odyssey, Overture, Sebastian and Wintmalt | positive linear mode | m/z 29–50 kDa | – | – | Hleba et al. (2019) |
| | | | Oil | 3 Breeds: sunflower, refined olive oil and virgin olive oils | positive ion mode | m/z 240–2400Da | PCA | – | Jergović et al. (2017) |
| Quality fraud identification | DART-MS | Animal | Aquatic product | Salmon, Cultivated methods: wild-type and farmed | negative ionization mode | m/z 100–900 | PCA, DA | 100% | Fiorino et al. (2019) |
| | | | Milk | Feeding methods: maize silage, crop silage/hay and grassland hay | positive ionization mode | m/z 75–1125Da | PLS-DA, LDA | 98% | Riuzzi et al. (2021) |
| | | Plant | Vegetable | Leek, Cultivated methods: organic and conventional | positive and negative ionization mode | m/z 100–1000 | PCA, OPLS-DA | 93.8–100% | Birse et al. (2022) |
| | REIMS | Animal | Meat | Processing methods: β-agonist treated livestock | negative ionization mode | m/z 50–1200 | PCA, LDA | 95% | Guitton et al. (2018) |
| | | | | Illegal addition: PS80 protein powder, Naturprotein powder, pork plasma powder and carrageenan | positive and negative ionization mode | m/z 100–1200 | PLS-DA | – | Kosek et al. (2019) |
| | MALDI-MS | Plant | Oil | Processing methods: edible, versus deep fried and gutter | positive ion mode | m/z 280–1860 | PCA | – | Cao et al. (2021) |
| | SICRIT-MS | Plant | Fruit | Orange juice, Processing methods: freshly squeezed and pasteurized | positive ionization mode | m/z 50–1000 | DD-SIMCA | – | Wang and Xu (2022) |
| Geographical origin identification | DART-MS | Animal | Meat | Lamb, 4 Regions: Anhui, New Zealand, Ningxia, and Gansu | positive and negative ionization mode | m/z 50–1000 | OPLS-DA, LDA | 100%,82.5% | Qie et al. (2022) |
| | | Plant | Honey | Chestnut from 2 Countries: Italy and PortugalAcacia from 2 Countries: Italy and China | positive linear mode | m/z 100∼600 | KNN | 96.7%∼100% 90%∼100% | Lippolis et al. (2020) |

(*continued on next page*)

**Table 5** (*continued*)

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Instrument mode | mass spectra | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|---|
| | | | vegetable | Garlic, 3 Countries: Czech Republic, Spain and China | positive and negative ionization mode | m/z 150–2000 | OPLS-DA | 100% | Hrbek et al. (2018) |
| | REIMS | Plant | Pistachio | 5 Countries: Bronte, California, Iran, Turkey and Greece | negative ionization mode | m/z 50–1200 | PCA, LDA | 100% | Rigano et al. (2019b) |
| | MALDI-MS | Animal | Milk | Mozzarella cheese, 2 Origins: Korean and non-Korean | positive linear mode | m/z 1000–20,000 | PLS-DA | 100% | Kandasamy et al. (2021) |
| | | | Aquatic product | *Sparus aurata*, 2 Farms in Madeira Island: Caniçal and Ribeira Brava | positive linear mode | m/z 2–20 kDa | PCA, HCA | – | Freitas et al. (2022) |
| | | Plant | Fermented tea | 2 Regions: Assam and Darjeeling | positive linear mode | m/z 2–20 kDa | PCA, PLS-DA | – | Kaufmann et al. (2022) |
| | | | Fermented-salted vegetables | 2 Countries: China and Korean | positive linear mode | m/z 2000–20,000 | PCA, HCA | – | Yoon et al. (2017) |

AIMS, ambient ionization mass spectrometry; DART-MS, direct analysis in real time mass spectrometry; REIMS, rapid evaporation ionization mass spectrometry; MALDI-TOF MS, matrix-assisted laser desorption/ionization time of flight mass spectrometry; SICRIT-MS, soft ionization by chemical reaction in transfer mass spectrometry; PLS-DA, partial least squares discrimination analysis; LDA, linear discriminant analysis; PCA, principal component analysis; OPLS-DA, orthogonal partial least-squares discrimination analysis; KNN, K-Nearest Neighbor; DD-SIMCA, data-driven soft independent modeling of class analogy.

authenticity studies mostly focus on breeds identification in animal-derived products, such as beef, poultry, fish, and dairy products. For meat identification, REIMS collects the fingerprints of meat products (venison, horse meat, and beef) to identify the animal tissues of different breeds and species, yielding excellent results with 100% accuracy for breeds identification and 97% accuracy for species cognition (Balog et al., 2016). A REIMS-based lipidomic approach was used to identify salmon and rainbow trout in real-time. The OPLS-DA model was used to statistically analyze 12 fatty acids and 37 phospholipids, resulting in an accurate discrimination rate of 96.58% (Song et al., 2019). REIMS was also used to establish a database of 18 marine species from a small area of the Messina Strait. The validation results showed identification rates of over 99% for all 18 species (Rigano et al., 2019a). REIMS combined with OPLS-DA differentiated four types of milk (goat, buffalo, Holstein, and Jersey) with 100 % accuracy (Cui et al., 2022). Furthermore, nine varieties of pistachios from three production areas were analyzed using REIMS (Rigano et al., 2019b). The results showed 98% correct identification based on variety and 100% accurate recognition based on origin. Therefore, REIMS could analyze plant-derived products and showed potential for geographic origin differentiation. In terms of quality fraud identification, combining REIMS and non-targeted metabolomics can directly analyze and identify meat products of livestock treated with β-agonist via the metabolite lipid profile changes at classification accuracy rates exceeding 95%. Therefore, this method can accurately and quickly determine the exposure of animals to ractopamine during reproduction (Guitton et al., 2018).

### 2.2.3. *Matrix-assisted laser desorption/ionization-time of flight mass spectrometry*

MALDI-MS is an ionization method that can directly vaporize and ionize non-volatile samples. The matrix must be mixed with the test solution before analysis to ensure that excessive laser energy does not destroy the test compound. The application purpose of MALDI-MS in food authenticity analysis is mainly to identify breeds or species, and establish a corresponding database based on the obtained data. Hleba et al. (2019) successfully distinguished eight barley breeds using MALDI-MS. They determined that B hordeins was the main substance for distinguishing barley, that B hordeins was the main substance for distinguishing barley, and established a local barley database based on B hordeins. Rau et al. (2020) used MALDI-MS to rapidly and accurately identify the dairy animal species of mozzarella and white brined cheese, establishing a MALDI-MS database for animal species identification in dairy products. MALDI commonly uses the PLS-DA model for breeds or species discrimination. MALDI-MS was used to analyze three other species (pork, chicken, and duck) in beef, obtaining an average discrimination accuracy of 94.7% using the PLS-DA model (Pu et al., 2022). MALDI and PLS-DA was used to identify cow-milk adulteration in feta milk at an accuracy of 83.5% (Kritikou et al., 2022). In addition to breeds or species differentiation, MALDI-MS has also been successfully applied for the geographical origin identification of food. The analysis of animal-derived samples is mainly based on their protein omics data. Kandasamy et al. (2021) successfully differentiated the geographic origin of mozzarella cheese from Korean farms and non-Korean mozzarella cheese using protein profiling data collected via MALDI-MS combined with multivariate statistical analysi. This method can also distinguish cheese from farms and companies within Korea. Protein information in fish mucus varies depending on the growing environment of the fish. Therefore, protein information in the mucus of sparus aurata collected by MALDI-MS, combined with PCA and HCA, can be used to distinguish sparus aurata from two different mariculture farms (Freitas et al., 2022). Besides, Cao et al. (2021) established a method to distinguish edible oil from used cooking oil using MALDI-MS. The results showed that fresh edible oil was successfully separated from deep fried oil and gutter oil. This method quickly identified the authenticity of oil only via visual inspection without complicated calculation and analysis. This study also confirmed the feasibility of

using MALDI-MS for food quality fraud identification.

#### 2.2.4. Soft ionization by chemical reaction in transfer mass spectrometry

SICRIT represents the latest optimal AIMS technology, which can directly analyze gaseous or flavor molecules online. The chemical composition and state of the influx substances can be identified in real-time using the electrode discharge and transient excitation of gaseous chemicals at the entrance of MS. Samples can be analyzed directly, quantitatively and qualitatively without the aid of solvents or auxiliary gases. However, this technology is still in the initial exploration stage in food authenticity studies, with fewer application examples. Wang and Xu (Wang and Xu, 2022) applied SCRIT to analyze orange juice for the first time, successfully determining the differences between orange juice exposed to different degrees of processing, providing a reference for further segmentation of micro-processed juice segmentation. Although SCIRIT is primarily used for volatile compound analysis due to equipment constraints, its efficient and convenient assessment process highlights its promise for common food authentication applications.

In conclusion, AIMS generally presents advantages, such as high efficiency, repeatability, and stability, which are suitable for solving problems related to food authenticity. However, this type of technology is not yet fully mature, and the data acquisition and processing methods easily affect the accuracy of the discrimination rate. Therefore, attempts should be made to optimize the data acquisition procedure and select suitable data processing methods to improve the discrimination accuracy.

### 2.3. Electronic sensors

Electronic sensors include electronic nose (E-nose) and electronic tongue (E-tongue). An E-nose is an artificial olfactory system, while an E-tongue is an artificial gustatory system. They consist of a sample processing system, a chemical sensor array, and a pattern recognition system. The E-nose uses gas sensors to rapidly identify odor components and obtains the overall fingerprint information of the volatile components in a sample (Gonzales et al., 2011). The sample preprocessor of the E-tongue is equivalent to the human taste receptors. It converts the abstract features of liquid samples into visual electronic signals that respond to the tested liquid via the sensor array with low selectivity, non-specificity, and interactive sensitivity. The output signal data can be used to obtain information about the taste characteristics of the sample being tested (Fig. 2). Electronic sensors have been widely used for food authentication. Relevant literatures are summarized in Table 6.

#### 2.3.1. Electronic nose

The E-nose is primarily used to analyze volatile components in samples, and has become popular for analyzing the authenticity of food products. In a 2017 review, 46 applications of electronic noses for food authenticity were demonstrated. Dairy products, vegetable oils and animal fats, as well as meat and alcohol, are all able to achieve authenticity through e-nose technology (Gliszczynska-Swiglo and Chmielewski, 2017). It is worth noting that the research on food authenticity using E-nose technology mostly adopts electronic nose equipment based on fast GC. He et al. (2021) analyzed 65 white wine samples from three regions with six aroma types using the GC-E-Nose. The results showed a total classification accuracy of 91.53% and 93.94% for aroma and region, respectively. Wu et al. (2022) analyzed 41 apple samples from seven regions and three plant sources in China using an E-nose, identifying 29 volatile compounds. The algorithm identification rates were 88.2% and 88.9% for the geographical regions and plant sources, respectively. Therefore, E-nose can be used for breeds or species identification, as well as to determine the geographical origin of food products. An E-nose was also used to determine raw milk quality fraud. Degraded raw milk samples neutralized with sodium hydroxide (NaOH), sodium thiocyanate (NaSCN), sodium carbonate ($Na_2CO3$), and sodium bicarbonate ($NaHCO_3$) were used to simulate the fraudulent product. The degraded raw milk neutralized with chemicals and simulated adulterated samples were examined via flash GC using E-nose and chemometric methods. Analysis indicated that RF could achieve 100% discrimination (Tian et al., 2022).

#### 2.3.2. Electronic tongue

The E-tongue mimics human taste perception and has revolutionized traditional food identification and evaluation methods. Its perception exceeds basic taste recognition. Due to the miniaturization of the E-tongue, it has been widely used for breeds identification, geographical origin determination, and quality fraud evaluation of food products. In breeds identification studies, E-tongue has been applied to meat products such as beef and mutton, and plant-derived foods such as coffee and olive oil, with excellent research results. Suranyi et al. (2021) used conventional methods to analyze the quality indicators of beef, using an E-tongue to predict the indicator parameters. The results showed correct beef breeds classification reaching 100%. The E-tongue can also distinguish different species in meat products, and even different species sources in minced meat. Tian et al. (2019) used E-tongue technology to distinguish mutton, pork, and chicken in minced meat, with the classification rate reaching 100%. The electronic tongue is applied to coffee and olive oil, mainly to distinguish their breeds. In a recent study where electronic sensors were combined with human sensory attributes to

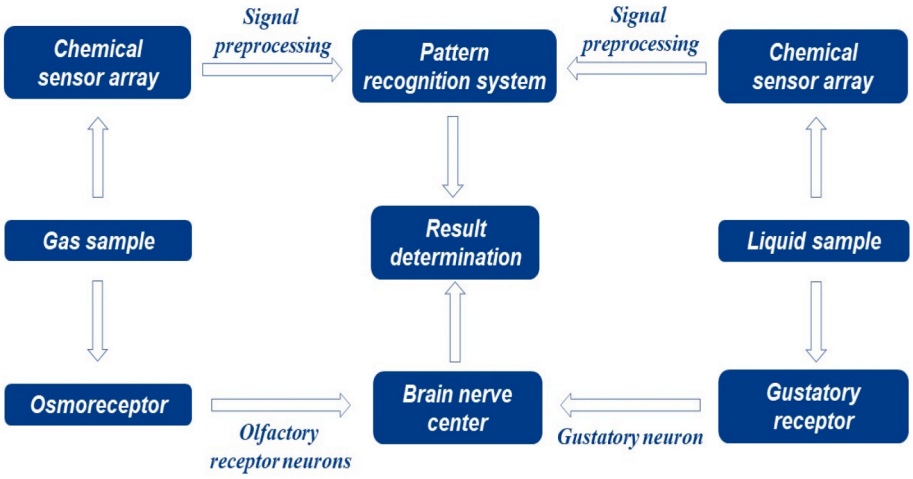


**Fig. 2.** Electronic sensors and human sensory system.

**Table 6**
Application progress of electronic sensors in authenticity analysis of food.

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Instrument configuration | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| Breeds/Species identification | E-nose | Animal | Meat | Minced meat, 2 Species: mutton and pork | a PEN2 E-nose (portable electronic nose II, Airsense Corpora-tion, Germany) | SLDA | – | Tian et al. (2013) |
| | | Plant | Baijiu | 6 Breeds: Strong aroma, Soy sauce aroma, Light aroma, Jian aroma, Feng aroma and Herbal aroma | the fast GC- based electronic nose analysis, Heracles II (Alpha MOS, Toulouse, France) | PLS-DA | 93.94% | He et al. (2021) |
| | | | Fruit | Apple, 3 Breeds: Golden Delicious, Fuji and Ralls | an FGC E-nose (Heracles II, Alpha M.O.S., Toulouse, France) | SLDA | 87.5% | Wu et al. (2022) |
| | | | | Red wine, 3 Breeds: Cabernet Sauvignon, Snake Dragon Ball and Merlot | colorimetric sensors in the E-nose system a E-tongue (Isenso, Shanghai Ruifen International Trading Co., Ltd.) | ELM | 100% | Han et al. (2020) |
| | E-tongue | Animal | Meat | Cattle, 5 Breeds: Angus, domestic buffalo, Hungarian Grey, Hungarian Spotted cattle and Holstein | a potentiometric electronic tongue (e-tongue) with food grade sensors | LDA | 100% | Suranyi et al. (2021) |
| | | | | Minced meat, 3 Species: mutton, pork and chicken | the taste system of a-Astree (Alpha M.O.S, Toulouse, France) | BDA, CDA | 90%–100% | Tian et al. (2019) |
| | | Plant | Cereal | Rice, 2 Breeds: jasmine and white | lab-made E-nose system | BPNN | – | Timsorn et al. (2017) |
| | | | Oil | Olive oil, 8 Breeds (Arbosana, Arroniz, Cornicabra, Frantoio, Manzanilla, Redondilla, Royuela and Zorzal) | the E-tongue included two print-screen potentiometric devices, containing different cross-sensitivity membranes as chemical sensors | LDA-SA | 100% | Dias et al. (2016) |
| | E-nose E-tongue | Plant | Coffee | 7 Breeds: Robusta Xinglong 1, Robusta Reyan 1, Robusta Reyan 2, Robusta Xinglong 24-2, Robusta Xinglong 26, Robusta Xinglong 28, and 'Robusta Chenmai' | a commercial E-tongue (Alpha ASTREE Liquid Taste Analyzer; Alpha M.O.S., Toulouse, France) | PLSR | 100% | Dong et al. (2017) |
| | E-nose, E-tongue, E-eye | Plant | Oil | Olive oil, 3 Breeds: Hojiblanca, Picual and Arbequina | an E-nose (13 MOX sensors, FIS and Figaro) an E-tongue (3-electrode cell) an E-eye (LEDs from 780 nm to 380 nm) | PLS-DA | – | Apetrei et al. (2010) |
| Quality fraud identification | E-nose | Animal | Milk | Illegal addition: acid neutralizers | a flash GC E-nose (Alpha MOS, Toulouse, France) | RF | 100% | Tian et al. (2022) |
| | E-tongue | Plant | Black tea | Quality grades (1–7) | an E-tongue (electrodes made of 6 different cylindrical metal electrodes (platinum, gold, palladium, wolfram, titanium, and silver)) | PLS-DA, SRD | 90% | Chen et al. (2020) |
| | | | Honey | illegal addition: fructose, glucose, inverted sugar, hydrolyzed inulin syrup and malt wort | a voltametric E-tongue (3 electrodes: reference electrode (Ag/AgCl) | PLS-LDA | 83.33% | Oroian et al. (2018) |
| | E-nose, E-tongue | Animal | Aquatic product | Storage methods: different storage times (1–10days) | An E-nose with nine metal oxide semiconductor gas sensors, a commercial E-tongue | RBF-NN | 93.9% | Han et al. (2014) |
| | | Plant | Coffee beans | Processing methods: room-temperature drying, solar drying, heat pump drying, hot-air drying, and freeze drying | an Astree II potentio-metric electronic tongue (Alpha M. O. S., Toulouse, France) an Astree II potentio-metric electronic tongue (Alpha M. O. S., Toulouse, France) | PCA | – | Dong et al. (2019b) |
| | | | Black tea | Quality grades (1–4) | an E-nose (5 MOS sensors, 120 gas sensors, Figaro, Japan), an E-Tongue (5 electrodes made of 5 different noble metals (viz. gold, iridium, palladium, platinum, and rhodium)) | PCA, KNN | 99.75% | Banerjee et al. (2019) |
| Geographical origin identification | E-nose | Plant | Fruit | Apple, 7 Provinces in china: Shandong, Shanxi, Sinkiang, Hebei, Gansu, Liaoning, and Shaanxi | an FGC E-nose (Heracles II, Alpha M.O.S., Toulouse, France) | SLDA | 97.1%–100% | Wu et al. (2022) |
| | | | Vegetable | Ginger, 7 Provinces in China: Yunnan, Sichuan, Henan, Shandong, Fujian, Zhejiang, Guangdong | the Heracles NEO e-nose (Alpha M.O.S., Toulouse, France) | RF | 100% | Yu et al. (2022) |
| | | | Coffee | 4 Regions in Rwanda: northern, southern, eastern, and western | the Heracles NEO e-nose (Alpha M.O.S., Toulouse, France) | PCA, DFA | 95% | Flambeau et al. (2017) |

(*continued on next page*)

**Table 6** (*continued*)

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Instrument configuration | Chemometrics method | Discriminant rate | References |
|---|---|---|---|---|---|---|---|---|
| | | | Cocoa liquor | 10 Countries: the Philippines, Vietnam, Brazil, Cuba, Colombia, Ecuador, Peru, Congo, Ghana and Ivory Coast | an ultra-fast GC electronic nose (Heracles II, Alpha M.O.S., Toulouse, France) | PCA, DFA | – | Rottiers et al. (2019) |
| | | | Baijiu | 3 Provinces in China: Sichuan, Heilongjiang and Jiangsu | a fast GC- based electronic nose analysis, Heracles II (Alpha MOS, Toulouse, France) | PLS-DA | 93.94% | He et al. (2021) |
| | E-tongue | Plant | Honey | 3 Countries: Spain, Honduras and Mozambique | an E-tongue (made of 4 metallic electrodes: Ir, Rh, Pt, Au) | PLS | – | Sobrino-Gregorio et al. (2020) |
| | E-nose, E-tongue | Plant | Fruit | Red wine, 3 Cities in China: Yantai, Langfang and Xianyang | colorimetric sensors in the E-nose system a E-tongue (Isenso, Shanghai Ruifen International Trading Co., Ltd.) | ELM | 100% | Han et al. (2020) |
| | | | Lycium ruthenicum Murray | 5 Provinces in China: Gansu, Inner Mongolia, Ningxia, Qinghai and Xinjiang | an ISENSO® iNose e-nose (New York, 103 USA), an ISENSO ® SmarTongue e-tongue (New York, USA) | PCA, LDA | 92.6% | Wang et al. (2019b) |

PLS-DA, partial least squares discrimination analysis; LDA, linear discriminant analysis; PCA, principal component analysis; KNN, K-Nearest Neighbor; RF, random forest; SLDA, stepwise discriminant analysis; BDA, bayes discriminant analysis; CDA, canonical discriminant analysis; DFA, discriminant factorial analysis; ELM, extreme learning machine; LDA-SA, linear discriminant analysis with simulated annealing selection algorithm; RBF-NN, radial basis function neural network; BPNN, back propagation neural network; SRD, sum of ranking difference.

differentiate between olive oil varieties, eight olive oil varieties were classified correctly with 100 per cent accuracy, a significant improvement over the ability to differentiate using only the electronic tongue or sensory attributes (Dias et al., 2016). And the successful application of the combination of electronic sensors and human sensory attributes confirms the complementary roles of human beings and artificial intelligence. In quality fraud identification studies, E-tongue also showed multiple analytical abilities for quality fraud identification. Although the samples mostly included plant-derived foods, the analysis samples and purposes were diverse, allowing the determination of seven black tea grades (Chen et al., 2020), as well as honey adulterated with additives such as glucose, inverted sugar, and inulin syrup (Oroian et al., 2018). The discrimination accuracies of these two studies were 90 % and 83.3%, respectively. Regarding the research on the authenticity of honey, Sobrino-Gregorio et al. (2020) successfully distinguished honey from three countries using E-tongue technology. Therefore, it is also feasible to use E-tongue technology to examine the geographical origin of food.

#### 2.3.3. *Multi-technology coupling of electronic sensors*

Research on the combination of various sensors, particularly the combination of E-nose and E-tongue, is growing to enhance the use of electronic sensors for food authentication in addition to the combination of electronic sensors with human sensory attributes. The E-nose and E-tongue are mainly combined for quality fraud and geographical origin identification in food authenticity research. Han et al. (2014) analyzed the accuracy of single E-nose or E-tongue use and their combination in distinguishing fish with different levels of freshness. The results showed that the single systems fulfilled the requirements, while combining the two was more accurate, with a discrimination accuracy of 93.9% could be obtained. Furthermore, the combination of E-nose and E-tongue combination traced Lycium ruthenicum Murray from five provinces in China (Guansu, Inner Mongolia, Ningxia, Qinghai, Xinjiang), with 92.6% accuracy (Wang et al., 2019b). Apetrei et al. (2010) combined an E-eye, E-nose, and E-tongue to distinguish olive oil with different bitterness levels, confirming that the discrimination ability of the combined system was superior to the results obtained with the three instruments, respectively.

In general, the reports on the diverse applications of electronic sensor technology in investigating food authenticity concerns exhibit a relatively uniform quantity of data. They primarily use analysis of volatile or odorous compounds in the food matrix to achieve the differentiation goal. This method has the benefits of high efficiency, low cost, and good reproducibility. However, low discrimination accuracy may occur due to the limited number of compounds they can identify. Therefore, it is necessary to effectively avoid similar situations by combining multiple techniques or selecting the appropriate data analysis methods.

### 2.4. *DNA-based technology*

DNA is one of the best indicators for food authenticity analysis and traceability because it is completely consistent in different parts of organisms and has superior consistency and thermal stability throughout the entire life cycle of animals and plants, from farmland to dining table, when compared to other indicators like mineral elements and stable isotopes (Scarano and Rao, 2014). After the horse meat turmoil in Europe in 2003, DNA technology gradually began to be used in the field of food authenticity, primarily for species-derived component analysis in food. The DNA technologies currently used for rapid food authenticity analysis mainly include loop-mediated isothermal amplification (LAMP), recombinase polymerase amplification (RPA), high-resolution melting (HRM), next-generation sequencing (NGS), and DNA barcoding. DNA-based technology is mainly used to distinguish breeds or species. Relevant literatures are summarized in Table 7.

**Table 7**
Application progress of DNA-based technology in authenticity analysis of food.

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Target/Primers/Markers | Detection limit | References |
|---|---|---|---|---|---|---|---|
| Breeds/Species identification | LAMP | Animal | Aquatic product | Identification of skipjack tuna in processed fish products | cytochrome *b* gene | 50 pg | Xiong et al. (2021) |
| | | | | Identification of *Thunnus albacares* in tuna steaks, fillets or canned | cytochrome *b* gene | 540 fg | Ali et al. (2022) |
| | | | | Identification of salmo salar in commercial products | LB-6 | 5 pg | Li et al. (2022) |
| | | | | Identification of Diodon in both cooked and digested samples | cytochrome oxidase I gene | 0.5 pg | Xie et al. (2022a) |
| | | | | Identification of arothron in commercial products | cytochrome *b* gene | 1 ng | Xie et al. (2022b) |
| | | | | Identification of takifugu in commercial products | cytochrome oxidase I gene | 0.1 ng | Xie et al. (2022c) |
| | | | | 8 Breeds: 6 commercial cods and 2 oilfish | cytochrome *b* gene | – | Li et al. (2021a) |
| | | | | European eel, 4 Breeds: *A. anguilla*, *A. rostrata*, *A. australis* and A. japonica | the cytochrome *b*, the 16S ribosomal RNA or the cytochrome oxidase c subunit 1 gene | 500 pg | Spielmann et al. (2019) |
| | | | | Crab, 3 Breeds (blue swimming, crucifix, and three spotted swimming) | cytochrome oxidase I gene | 50 ng | Benjakul and Saetang (2022) |
| | | | Meat | 5 Species: cattle, buffalo, goat, sheep and pork | cytochrome *b* gene | 0.0001 ng | Kumari et al. (2021) |
| | | | | 5 Species: ovis aries, goat, cattle, buffalo and chicken | mitochondrial D loop region | 0.5 ng | Mounika et al. (2021) |
| | | | | 2 Species: horse and donkey | LOC106782588, LOC106825524 | 40 pg | Zhang et al. (2019) |
| | | | | 4 Species: pig, buffalo, sheep, and goat | mitochondrial CO I gene | 10 fg | Jawla et al. (2021) |
| | | | | 10 Species: chicken, duck, pig, cow, horse, goat, rabbit, ostrich, camel and goose | cytochrome *b* gene | 1.5 pg | Yan et al. (2022) |
| | | | | 4 Species: duck, pork, beef, mutton and chicken | a mitochondrial DNA | 30 ng | Shi et al. (2017) |
| | | | | 8 Species: donkey, horse, pork, cow, sheep, chicken, duck and rabbit | cytochrome *b* gene | 1% | Wang et al. (2020a) |
| | | | Milk | 2 Species: cow and goat | mitochondrial cytochrome *b* gene | Cow, 0.1 pg; Goat, 1 pg | Kim and Kim (2018) |
| | | | | 2 Species: milk and goat milk | cytochrome *b* gene | 10 fg | Wang et al. (2022d) |
| | | | | 5 Species: camel, horse, yak, goat and cow | cytochrome *b* gene, cytochrome *c* oxidase subunit 1 gene | 0.05 ng | Yu et al. (2021) |
| | | | Donkey-hide gelatine (DHG) | 7 Species: donkey and horse, cow, pork, goat, sheep or chicken | 12S rDNA | 0.001 ng, 0.1% DHG | Sheu et al. (2020) |
| | | Plant | Vegetable | Eggplant, 12 Bangladesh breeds and 6 Japan breeds | β-fructosidase gene | Senryo-nigou, 50 copies, BARI Begun-4, 50 copies | Yeasmin et al. (2021) |
| | | | | Sweet potato noodles, 2 Species: cassava and sweet potato | – | 1% | Wang et al. (2019a) |
| | | | | R. crustosa, R. rosacea, R. sanguinea, R. variata, R. velenovskyi and edible mushrooms | ITS sequence | 3.2 pg | Wang et al. (2022b) |
| | | | Wheat | 28 durum wheat breeds | Chr 7A | – | Cibecchini et al. (2020) |
| | | | Honey | 4 Breeds: *A. cerana* and four 4 breeds *A. mellifera* | MRJP2 gene | *A. cerana*, 4 ng, *A. mellifera*, 1 ng | Gao et al. (2023) |
| | RPA | Animal | Meat | 4 Species: duck, chicken, cow, sheep and pig | ND2, d-loop, 12S rRNA + 16S rRNA, ND5 and d-loop | 200 fg | Cao et al. (2018) |
| | | | | 3 Species: beef, pork and horse | the porcine mitochondrial ND2, equine ATP 6–8 genes | 1% | Kissenkotter et al. (2020) |
| | | | | 2 Species: beef and chicken | iSp 9 | 0.01% | Liu et al. (2022a) |
| | | | | 2 Species: beef and duck | Beef (ARS-UCD1.2: 23: 10,955,159 : 10,956,296: 1) duck (ENSAPLG00000007071) | 5% | Fu et al. (2020) |
| | | | | 3 Species: mutton, chicken and duck | cytochrome *b* gene | 4 fg | Li et al. (2019b) |
| | | | | Identification of pork in commercial meat products | mitochondrial DNA | 0.001 ng | Zhao et al. (2022) |

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**Table 7** (*continued*)

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Target/Primers/Markers | Detection limit | References |
|---|---|---|---|---|---|---|---|
| | | | Milk | Species: pork and yak milk | cytochrome oxidase subunit I gene | 50 ng | Wang et al. (2020b) |
| | | | Aquatic product | 2 Species: squids and other cephalopod species | cytochrome oxidase I gene and nucleotide sequence modified reverse primers | 50% | Wang et al. (2020b) |
| | HRM | Animal | Aquatic product | Salmonid, 8 Species: *Oncorhynchus keta*, *O. gorbuscha*, *O. kisutch*, *O. nerka* and *O. tshawytscha*, *Salmo salar*, *Oncorhynchus mykiss* and *Salmo trutta* | cytochrome oxidase I gene, cytochrome *b* gene | – | Monteiro et al. (2021) |
| | | | | Mussel, 3 Species: *M. galloprovincialis*, *M. edulis*, and M. chilensis | H1C gene | – | Asorey et al. (2022) |
| | | | Milk | Cheese, 2 Species: bovine and feta | D-loop region and $tRNA^{Lys}$ | 0.1% | Ganopoulos et al. (2013) |
| | | Plant | Grape | 13 Breeds: Alicante Bouschet, Cabernet Sauvignon, Donzelinho Tinto, Merlot, Malvasia Fina, Pinot Noir, Rufete, Tinto Cão, Touriga Franca, Tinta Francisca, Touriga Nacional, Tinta Roriz and Viosinho | Vv1/UFGT, Vv2/F3H, Vv3/UFGT | – | Pereira et al. (2017) |
| | | | Honey | 3 Breeds: A. m. mellifera (M lineage), A. m. ligustica (C lineage) and A. m. carnica (C lineage) | cytochrome oxidase I gene cytochrome *b* gene | – | Soares et al. (2019) |
| | | | Rice | Carnaroli and 35 rice breeds | Alk and Waxy genes | – | Grazina et al. (2022) |
| | | | Panax ginseng | 5 Breeds: P. ginseng, P. quinquefolius, P. notoginseng, *P. japonicus* and P. trifolius | the gene encoding the dammarenediol synthase | – | Grazina et al. (2021) |
| | DNA barcoding | Animal | Aquatic product | Yellow croakers, 7 Species: L. polyactis, L. crocea, L. terengganui, N. albiflora, *C. lucidus*, P. argentata, and *P. macrocephalus*. | cytochrome oxidase I gene | – | Chen et al. (2021a) |
| | | | | Fillet, 2 Breeds: *G. morhua* and *G. macrocephalus* | Cytochrome Oxidase subunit I gene | – | Feldmann et al. (2021) |
| | | | Meat | 15 mammalian and 6 poultry species | 16S rDNA | 0.1% | Dobrovolny et al. (2019) |
| | | | Milk | butter, milk and yogurt, identify plant oil (corn, soybean, rapeseed and sunflower) in products | inner P6 loop | – | Uncu and Uncu (2020) |
| | | Plant | Honey | 3 Breeds: nonItalian, Italian polyfloral and Italian monofloral | trnL | – | Chiara et al. (2021) |
| | | | Cereal | 11 market 3 Species: Black gram, refined wheat flour and white pea flour | rbcL 600 bp, trnH-psbA 380 bp, ITS 680 bp | – | Amane and Ananthanarayan (2019) |
| | NGS | Animal | Aquatic product | Surimi-based products | 16SrRNA | – | Giusti et al. (2017) |
| | | | | Species: DNA from 13 families, 19 genera and 16 species of fish, and from 3 families, 3 genera and 3 species | | | |
| | | | | Bivalve molluscs, 15 species belonging to the bivalve families | 16S ribosomal RNA (16S rRNA) and cytochrome *c* oxidase I (COI) genes | – | Abbadi et al. (2017) |
| | | | Meat | 13 Species: pork and 12 different species | – | 0.1% | Akbar et al. (2021) |
| | | | Milk | 4 Species: cattle, sheep, goat, and buffalo | 12S_Ki, 16S_Ki, 16S_KH | – | Ribani et al. (2018) |
| | | Plant | Honey | 3 Breeds: 6 monofloral honeys, 2 polyfloral honeys and 1 honeydew honey | trnL barcoding fragment | – | Utzeri et al. (2018) |
| | SNP + HRM | Plant | Fruit | Grape, 4 Breeds: Alvarinho, Moscatel Galego, Touriga Francaand Touriga Nacional | UFGT, F3H and LDOX genes | – | Teixeira et al. (2021b) |
| | Bar-HRM | Animal | Aquatic product | Gadoid, 4 Breeds: Atlantic cod, Pacific cod, Alaska pollock and saithe | cytochrome oxidase I gene, cytochrome *b* gene | – | Fernandes et al. (2017) |
| | | | | Hake, 5 Breeds: M. merluccius, *M. productus*, M. hubbsi, *M. capensis* and M. paradoxus | cytochrome oxidase I gene | 0.2–20 pg | Fernandes et al. (2018) |
| | | | | Pufferfish, 4 Species: Takifugu xanthopterus, T. fasciatus, T. flavidus and *T. rubripes* | cytochrome oxidase I gene | – | Chen et al. (2021b) |
| | | Plant | Fruit | Berry, 8 Breeds: Bilberry, Blueberry 'Northblue', Lingonberry, Bog bilberry, Crowberry, Gooseberry, | ITS, rpl36–rps8, trnL–F or trhH–psbA | – | Jaakola et al. (2010) |

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**Table 7** (*continued*)

| Authenticity problem | Technology | Product category | Food product | Adulterants/classified product | Target/Primers/Markers | Detection limit | References |
|---|---|---|---|---|---|---|---|
| | | | | Honeysucle and Mountain shadbush | | | |
| | | | | Juice, 5 Species: orange, mango, peach, pear and pineapple | trnL | – | Faria et al. (2013) |
| | | | | Apricot 35 breeds | ITS1 and ITS2 | – | Hurkan (2022) |
| | | | Walnut | 3 Species: walnut, peanut and soybean | PsbA-trnH gene | 10% | Ding et al. (2020) |
| | | | Processed foods | 3 Species: flax, chia and sesame seeds | rbcL DNA sequences | – | Bruno et al. (2020) |
| | | | Honey | 3 Breeds: L. stoechas subsp., L. penduculata and *L. viridis* | matK gene | – | Soares et al. (2018) |
| | NGS + DNA barcoding | Animal | Meat | 5 mammalian Species: pig, cattle, horse, sheep, and goat; 2 poultry Species: chicken and turkey | Sanger sequencing and matching a ~464 base pair (bp) fragment of the mitochondrial cytochrome *b* gene | – | Dobrovolny et al. (2022) |
| | | | | 27 meat and poultry products | 16S rRNA | – | Xing et al. (2019b) |
| | | | | 12 Species (bovine (cattle and water buffalo), swine (domestic pig), Caprinae (sheep), gallus (domestic chicken), partridge, fish (grass carp, silver carp, blue scad, tile fish, and pomfret), and shrimp (prawn)) | mini COI 136 bp, standard barcode 658 bp | – | Pan et al. (2020) |
| | | | Aquatic product | Identification of salmon in commercial products | 16S rRNA | 1% | Wang et al. (2021) |
| | | Plant | Edible seaweeds | Identification of edible seaweeds in commercial products | cox 1, tufa and LSU | – | Handy et al. (2020) |
| Quality fraud identification | NGS + DNA barcoding | Animal | Aquatic product | Sea cucumber, Processing methods (raw and processed) | COI mini-barcode 257 bp | – | Xing et al. (2021b) |
| Geographical origin identification | SNP + HRM | Plant | Fruit | Grape, 4 Regions: Melgaco, Moncao, Vinho Verdel and Douro | UFGT, F3H and LDOX genes | – | Teixeira et al. (2021b) |

LAMP, loop-mediated isothermal amplification; RPA, recombinase polymerase amplification; HRM, high-resolution melting; NGS, next generation sequencing; Bar-HRM, barcode DNA high-resolution melting.

#### 2.4.1. Loop-mediated isothermal amplification

LAMP is a DNA analysis method that enables nucleic acid amplification in a short time at a constant temperature. The analysis results can be evaluated with the naked eye, meeting the requirements of on-site and grassroots analysis. Currently, the application of LAMP in food authenticity research is based on distinguishing the species or breeds of food. This mostly includes animal-derived foods, such as the three major categories of aquatic products, meat and milk. Studies on aquatic products are mainly divided into species-specific processed product identification and single-species differentiation. LAMP-based fish species-specific identification studies serve two purposes. One is to establish a specific analysis method based on LAMP for high-value species, such as Skipjack tuna, *Thunnus albacares*, and *Salmo salar*, to identify adulteration in the corresponding processed products (Ali et al., 2022; Li et al., 2022; Xiong et al., 2021). Another is the use of LAMP to quickly identify toxic fish species (diodon, arothron and takifugu) in commercially available products, effectively reducing the incidence of poisonings (Xie et al., 2022a, 2022b, 2022c). In relation to the investigation of aquatic product species differentiation, LAMP combined with cytochrome *b* gene successfully differentiated three types of crabs (blue swimming crab, cruciferous crab and three-spotted swimming crab) with a detection limit as low as 50 ng (Benjakul and Saetang, 2022). Recent research on LAMP-based meat identification has aimed to differentiate between the various animal origins of the meat. Meat products with species numbers as low as 2 (horse and donkey) (Zhang et al., 2019) and as high as 10 (chicken, duck, pig, cow, horse, goat, rabbit, ostrich, camel and goose) (Yan et al., 2022) were successfully distinguished, with detection limits as low as 10 fg. LAMP-based dairy research focuses on identifying specific sources of high-value milk, with samples tested on-site in as little as 30 min, effectively avoiding dairy product adulteration (Kim and Kim, 2018). In addition, LAMP specifically recognized donkey DNA in donkey skin gelatine within 1 h, with a relative detection limit of 0.1% for the remaining non-donkey DNA (horse, cow, pig, goat, sheep, or chicken) (Sheu et al., 2020). In the study of plant-derived goods, Wang et al. (2019a) created a LAMP-based technique for rapid analysis of Manihot esculenta in sweet potato noodles in addition to differentiating between breeds. Cassava is often put into production instead of sweet potato because of its high starch content and lower cost of making starch products. The results indicated that the real-time LAMP method could accurately and specifically analyze cassava components in sweet potato noodles with a detection limit of 1%. The gene fragments used by LAMP in animal product research are relatively uniform, with essentially cytochrome *b* and cytochrome oxidase I as the main target genes. Contrarily, the gene fragments of plant-derived products are relatively specific, withminimal duplication.

#### 2.4.2. Recombinase polymerase amplification

The nucleic acid amplification process of RPA technology mainly depends on three kinds of enzymes. Single-stranded nucleic acid recombinases, single-stranded DNA binding proteins (SSB), and strand-substitution DNA polymerases. Since detectable amplification products are usually obtained within 10 min min, so RPA is known as an alternative nucleic acid analysis technology to PCR (Li et al., 2019a). The application of RPA for food authentication is immature, with few related studies. Current research basically distinguishes between the breeds of plant-derived products, which are also divided into three categories: meat, milk, and aquatic products, with meat products being the most dominant. The purpose of all recently reported applications is species differentiation. Cao et al. (2018) established a new method for the visual identification of meat adulteration based on recombinase polymerase amplification (RPA) and SYBR green I (SG), which successfully identified ducks, chickens, cattle, sheep and pigs, and mutton. For seafood (octopus adulteration identification), an analysis method based on RPA and lateral flow analysis (LFA) was developed. The study optimized the

design of primers and the nfo-probe system in the COI region, with the final results acheving100% specificity and sensitivity. The results were validated in eight European laboratories (Velasco et al., 2021). In addition, Wang et al. (2020b) combined RPA and a lateral flow nucleic acid assay (LFNAA) for yak milk authentication, with a detection limit as low as 5%. With its benefits of high efficiency, affordability, and convenience, RPA is thought to have great potential in future food authentication studies, even though it is not widely utilized.

#### 2.4.3. High-resolution melting

Using saturated dyes and the variations in nucleic acid molecule dissolution temperatures, HRM creates distinct dissolution curves that are monitored by real-time PCR technology. This allows for the quick, sensitive, and precise analysis of single-base differences (Ganopoulos et al., 2013). HRM is currently primarily used to distinguish between breeds or species. A comparable number of studies exist regarding using products derived from plants and animals. Interestingly, the HRM studies on plant-based products all distinguish between food varieties, while those on animal-derived products all distinguish between food species. Grazina et al. (2021) used HRM technology to distinguish five kinds of ginseng (ginseng, five-leaf pine, notoginseng, Japanese pine, and three-leaf pine) with a more than 98% confidence. Furthermore, the analysis method based on HRM successfully distinguished 13 kinds of grapes, three kinds of honey, and 36 kinds of rice. For animal-derived products, HRM identified eight species of salmonids in less than 70 min (Monteiro et al., 2021), while accurately determining the authenticity of Greek PDO sheep's milk cheese with a detection limit of 0.1% (Ganopoulos et al., 2013). The research of HRM in distinguishing geographical origin of food needs to be realized by the collection of single nucleotide polymorphisms (SNP). Teixeira et al. (2021b) analyzed SNP in three genes (UFGT, F3H, and LDOX) using a DNA assay in conjunction with HRM analysis to successfully distinguish wines from four regions (Melgaco, Moncao, Vinho Verde, and Douro).

#### 2.4.4. Multi-technology coupling of DNA-based technology

The HRM applied in food authenticity studies is often analyzed in combination with DNA barcoding (Bar-HRM). DNA barcoding is a new technology that can quickly identify species using relatively short DNA fragments. It can rapidly identify a large number of samples simultaneously (Barrett and Hebert, 2005). DNA barcoding can reportedly differentiate species or varieties of aquatic products, meat and milk. The DNA barcodes used to differentiate aquatic products are usually cytochrome oxidase I gene (COI) sequences (Chen et al., 2021a; Feldmann et al., 2021). The 16SrDNA fragments were used as DNA barcodes to differentiate between 15 mammalian and six poultry species (Dobrovolny et al., 2019). The inner P6 loop was used as a DNA barcode to identify vegetable oil adulteration in milk and dairy products (Uncu and Uncu, 2020). It is evident that the desired results can be obtained by selecting a barcode region suitable for differentiation. Bar-HRM mainly distinguishes the breeds or species of aquatic products and fruits. Fernandes et al. (2018) developed a micro-bar code by combining the cytochrome C oxidase subunit I gene with HRM analysis to quickly distinguish hake breeds. The results showed that five hake breeds were completely distinguished with confidence. Bar-HRM can distinguish fruit breeds from raw materials while also tracing plant sources through juice. Faria et al. (2013) combined the trnL DNA barcode with HRM analysis to distinguish fruit breeds (orange, mango, peach, pear, and pineapple) in fruit juice. Bar-HRM has the advantage over DNA barcoding in that it allows for quantitative measurements, and over HRM in that it has the advantage of higher resolution.

In addition to combining with HRM technology, DNA barcode technology is often coupled with NGS technology for food authentication. NGS, also known as high-throughput sequencing (HTS) technology, can sequence hundreds of thousands to millions of nucleic acid molecules at a time and comprehensively and meticulously analyze the transcriptome and genome of a species. NGS has been used in recent years to distinguish between the breeds or species of animal-derived products. Abbadi et al. (2017) used NGS technology to correctly identify 15 species of bivalves, Furnaceae, ostrich, Iridaceae, and salamander, providing a quick, economical method for identifying substitution fraud in seafood products. Akbar et al. (2021) developed an NGS-based method to distinguish pork-derived components from other animal meat products, which could identify mixed samples containing multiple species (up to 12). NGS and DNA barcoding applications for food authentication have gradually increased, with current research focusing on animal-derived foods. The NGS and DNA barcoding combination clearly distinguished five kinds of mammals, two types of poultry, and 12 kinds of animal sources for meat products (Dobrovolny et al., 2022; Pan et al., 2020). This approach can also distinguish seafood species, such as salmon and edible seaweed breeds. Furthermore, the NGS and COI micro-barcode combination was used to identify sea cucumbers, mainly to determine whether commercial sea cucumber products were inconsistent with the labels, such as the substitution of cheap sea cucumbers (Xing et al., 2021b).

With the benefits of high specificity and sensitivity, DNA analysis technologies have proven to be one of the most effective methods for differentiating between food product breeds or species. The DNA-based rapid analysis techniques mentioned in this paper have been gradually applied in the field of food authenticity. Based on the literature research, the current research aims to differentiate food species, which may be related to the low genetic variation of breeds. Therefore, developing more sensitive, multi-targeted, high-throughput DNA technologies is necessary for food authentication. It is also necessary to continuously construct and improve DNA marker databases and prepare more suitable targeting primers to promote new progress of rapid DNA technology in the field of food authenticity.

## 3. Chemometrics methods

With a wide variety of foods and complex ingredients, food authenticity analysis was developed in response to various issues with food quality. Therefore, the popularization and application of rapid analysis technology are inevitable. However, regardless of the changing needs of food authenticity issues, various analytical techniques can only obtain informative datasets characterising different fingerprints, which are often mined and processed in conjunction with chemometrics (Grassi et al., 2023). Moreover, rapid analysis technology acquired more information, highlighting the significance of chemometrics. Selecting appropriate chemometric methods for data analysis is critical for effective results (Tarapoulouzi et al., 2022).

### 3.1. Unsupervised algorithm

Unsupervised learning is typically used for data exploration during the initial data analysis stage to observe the overall data structure, understand information such as data characteristics, variable correlations, and outliers, and further process data such as dimensionality reduction and unsupervised clustering for the purpose of sample classification or complex data simplification (Agriopoulou et al., 2022). PCA and cluster analysis (CA) are unsupervised algorithms commonly used during data exploration. PCA generates independent principal component comprehensive indexes via linear transformation for data dimension reduction. CA uses the similarity principle for data clustering to reduce data processing complexity, while hierarchical cluster analysis (HCA) is typically used for food analysis. PCA and HCA can independently analyze relevant information with significant differences between sample groups and minor differences within groups to classify data. The PCA model can be used to differentiate coffee beans of five geographical origins based on informative data collected by Fourier Transform Infrared Spectroscopy (FTIR) technology (Obeidat et al., 2018). The PCA score plots showed a significant difference between fresh edible oils, frying oils, and gutter oils based on the edible oil data gathered via MALDI-MS (Cao et al.,

2021). Based on the proteomic data collected by MALDI-MS, HCA was able to fully differentiate between fermented-salted vegetables from China and Korea after four weeks of fermentation (Yoon et al., 2017). PCA and HCA are considered the most commonly used models for analyzing IR and Raman spectral data to identify plant-derived products (Kucharska-Ambrozej and Karpinska, 2020). However, in most cases, unsupervised algorithms are mainly used to reduce data dimensionality, which clarifies the data structure and decreases the data processing complexity.

### 3.2. Supervised algorithm

Supervised algorithms are more reliable for data structures with small differences between sample groups during actual sample analysis. Based on the literature survey, the commonly used supervised methods include LDA, partial least squares discriminant analysis (PLS-DA), OPLS-DA, SVM, KNN, and partial least squares regression (PLSR).

#### 3.2.1. Linear discriminant analysis

Since LDA mainly applies to low-dimensional, linear data, it usually needs to be combined with unsupervised algorithms, such as PCA and HCA, and can be implemented using software such as SPSS and MATLAB. It is mainly used to distinguish between species, breeds, and geographical origins in the field of food authenticity. The LDA model was used to analyze the signature markers of the five milk species (cow, goat, camel, soybean, and oat) collected via electrospray ionization mass spectrometry (ESI-MS), with a 100% cross-validated discrimination rate (Hong et al., 2022). The LDA model based on Fourier-transform near--infrared spectroscopy (FT-NIR) yielded an overall discrimination of 97% for wheat from different regions in Italy (Northern, Central, and Southern) and 100% for Italian and non-Italian wheat (De Girolamo et al., 2019).

#### 3.2.2. Partial least squares regression

Three main algorithms are associated with partial least squares, including PLSR, PLS-DA, and OPLS-DA. PLSR, a commonly used multivariate regression method for quantifying food adulterant concentrations, is mainly combined with spectroscopic data. It can be executed in several software programs such as SPSS, The Unscrambler X, and others. Regarding the study on the adulteration of minced pork with jaw meat, the data collected via HSI were optimized using six pre-processing and three wavelength selection methods. Ultimately, it was found that the PLSR model performed best when the data were processed using the standard normal variate (SNV) and regression coefficients (RC) (Jiang et al., 2020).

#### 3.2.3. Partial least squares discriminant analysis

PLS-DA can analyze data from a wide range of techniques, including spectroscopy and mass spectrometry, and is the most commonly used classification method in the field of food authenticity. It can be implemented using SPSS, SIMCA, and MATLAB software. PLS-DA is typically used for data where the predictors are correlated and the number of variables exceeds the number of samples. PLS-DA has been demonstrated for all of the rapid analysis techniques mentioned in this paper, and it is the main classification algorithm currently applied to in situ mass spectrometry data. The PLS-DA model discriminated between Korean and non-Korean cheese samples with 100% accuracy by analyzing the proteomic data obtained via MALDI-MS (Kandasamy et al., 2021).

#### 3.2.4. Orthogonal partial least squares discriminant analysis

OPLS-DA is often employed when the number of variables exceeds the number of samples, resulting in the overfitting of the PLS or PLS-DA model. OPLS-DA has demonstrated validity in authenticity studies for various food products (Agriopoulou et al., 2021). Its implementation software is similar to that of PLS-DA. Based on the data collected from pure and adulterated honey samples via NMR, the PCA-LDA and OPLS-DA models were compared regarding their discriminatory effects, resulting in a high accuracy of 97.6% for OPLS-DA (He et al., 2020). The OPLS-DA model based on the metabolomic data of cocoa collected by REIMS achieved 100% accuracy in distinguishing adulterated cocoa (Chang et al., 2022).

#### 3.2.5. Other algorithms

The adaptability of different analysis methods to different authenticity issues is related to sample analysis technology. SVM is a binary classification model that is more suitable for a moderate amount of data, as well as high dimensional and non-linear data (Lamine et al., 2023). KNN is more suitable for modeling similar class groupings, such as food quality levels. PCA, PLS-DA, and SVM models based on NIR spectral data were developed to identify rice breeds (Indica and Japonica) (Sampaio et al., 2020). The results showed that the SVM model performed best model, with a 97% fitting accuracy, 93% cross-validation, and 91% prediction rate. The KNN and SVM models were established to differentiate rice quality grades. The results showed that the KNN model was more effective, with a 91.81% accuracy (Teye et al., 2019).

## 4. Conclusion

Rapid analysis technology is becoming increasingly popular in the field of food authenticity because of its benefits, which typically include low pre-treatment requirements and high analysis efficiency. Rapid analysis conforms to the needs of the industry and helps improve the frequent adulteration in the food authenticity field. However, these techniques also present various insurmountable shortcomings. (1) The common problem of spectral technology is the relative complexity of data analysis, which requires selecting appropriate pre-processing methods for spectral data to reduce the difficulty of subsequent data analysis. (2) Since AIMS was developed based on MS, data analysis relies heavily on databases, while the overall analysis cost is high. (3) The main disadvantage of electronic sensor technology such as the E-nose and E-tongue is that it is easily influenced by the analytical environment, while its accuracy is generally low. (4) Rapid DNA-based analyses are not yet mature enough to identify varieties with small genetic variation, and DNA marker databases need to be further developed.

Therefore, to select the most appropriate method based on sample differences and specific requirements, it is necessary to continuously explore the potential of various technologies in the future development of rapid analysis techniques. Additionally, a universally standardized model must be established for quick analysis to regulate the analysis of data measured using various instruments or in various conditions. To bring rapid analysis technology closer to practical applications in food certification, further exploration is required regarding smaller, simpler, and more intelligent system equipment.

## CRediT authorship contribution statement

**Zixuan Zhang:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Writing – original draft, preparation. **Yalan Li:** Investigation, Formal analysis, Validation, Project administration. **Shanshan Zhao:** Investigation, Project administration, Jiangxue Wu: Investigation. **Mengjie Qie:** Investigation. **Lu Bai:** Investigation. **Zhiwei Gao:** Writing – review & editing, Funding acquisition. **Kehong Liang:** Conceptualization, Investigation, Methodology, Data curation, Writing – review & editing, Supervision. **Yan Zhao:** Conceptualization, Investigation, Methodology, Data curation, Writing – review & editing, Supervision, Funding acquisition.

## 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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