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Authentication of berries and berry-based food products

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Abstract

Berries represent one of the most important and high-valued group of modern-day health-beneficial "superfoods" whose dietary consumption has been recognized to be beneficial for human health for a long time. In addition to being delicious, berries are rich in nutrients, vitamins, and several bioactive compounds, including carotenoids, flavonoids, phenolic acids, and hydrolysable tannins. However, due to their high value, berries and berry-based products are often subject to fraudulent adulteration, commonly for economical gain, but also unintentionally due to misidentification of species. Deliberate adulteration often comprises the substitution of high-value berries with lower value counterparts and mislabeling of product contents. As adulteration is deceptive toward customers and presents a risk for public health, food authentication through different methods is applied as a countermeasure. Although many authentication methods have been developed in terms of fast, sensitive, reliable, and low-cost analysis and have been applied in the authentication of a myriad of food products and species, their application on berries and berry-based products is still limited. The present review provides an overview of the development and appli-

Nomenclature: 1H-NMR, proton nuclear magnetic resonance; 2D-IR, two-dimensional correlation infrared spectroscopy; ACN, anthocyanin; Bar-HRM, DNA barcoding coupled with high-resolution melting analysis; bp, base pair; CDA, canonical discriminant analysis; CZE, capillary zone electrophoresis; DNN, deep neural network; DA, discriminant analysis; FIMS, flow-injection mass spectrometry; FT-IR, Fourier transform infrared spectroscopy; FT-NIR, Fourier transform near-infrared spectroscopy; GC, gas chromatography; HATR, horizontal attenuated total reflectance; HCA, hierarchical cluster analysis; HPLC, high-performance liquid chromatography; HPLC–DAD, high-performance liquid chromatography with diode array detector; HPLC–UV, HPLC coupled with ultraviolet detection; HRMS, high-resolution mass spectrometry; ICP-MS, inductively coupled plasma mass spectrometry; IR, infrared; IRMS, isotope ratio mass spectrometry; ITS, internal transcribed spacer; LC, liquid chromatography; LC–MS, liquid chromatography–mass spectrometry; LDA, linear discriminant analysis; LS-SVM, least-squares support vector machine; MIR, mid-infrared (spectroscopy); MS, mass spectrometry; MS^2, tandem mass spectrometry; NIR, near-infrared (spectroscopy); NIR-HSI, near-infrared hyperspectral imaging; NMR, nuclear magnetic resonance; NZNG, non-Zhongning wolfberry; PCA, principal component analysis; PCR, polymerase chain reaction; PLS, partial least squares; PLS-DA, partial least squares discriminant analysis; qRT-PCR, quantitative real-time polymerase chain reaction; RAPD, random amplified polymorphic DNA; SCAR, sequence characterized amplified region; SD-IR, second derivative IR; SIMCA, soft independent modeling of class analogy; SNIF–NMR, specific natural isotopic fraction–nuclear magnetic resonance; SNP, single-nucleotide polymorphism; SSR, simple sequence repeat; UHPLC–HRMS, ultrahigh-performance liquid chromatography–high-resolution mass spectrometry; UPLC, ultraperformance liquid chromatography; vis–NIR, visible–near-infrared spectroscopy; ZNG, Zhongning.

cation of analytical chemistry methods, such as isotope ratio analysis, liquid and gas chromatography, spectroscopy, as well as DNA-based methods and electronic sensors, for the authentication of berries and berry-based food products. We provide an overview of the earlier use and recent advances of these methods, as well as discuss the advances and drawbacks related to their application.

KEYWORDS
berries, chromatography, DNA barcoding, food authentication, spectroscopy

1 | INTRODUCTION

Plant-based foods and food products, such as those derived from medicinal plants and wild forest plants, have been recognized to be beneficial for human nutrition and health for thousands of years. Nowadays, due to an increasing amount of health awareness among consumers, numerous health-beneficial foods are gaining more and more attention as part of the human diet. Some of the most important and high-valued plant-based foods are derived from the fruit of different species of berries, which include wild species, such as wild strawberry (*Fragaria vesca* L.), raspberry (*Rubus idaeus* L.), cloudberry (*Rubus chamaemorus* L.), cranberry (bog cranberry, *Vaccinium oxycoccos* L.), lingonberry (*Vaccinium vitis-idaea* L.), and bilberry (*Vaccinium myrtillus* L.), as well as cultivated species, such as American cranberry (*Vaccinium macrocarpon* Ait.), blueberry (*Vaccinium* ssp.), strawberry (*Fragaria* × *ananassa*), sea buckthorn (genus *Hippophaë*), several species of currants (genus *Ribes*), wolfberries (genus *Lycium*), and numerous cultivars of the aforementioned.

Berries are commonly sold and consumed fresh, especially during the season of their ripening. However, due to their generally short shelf life, berries are often stored frozen or dried, and used as ingredients in many types of food products and dietary supplements, such as juices, jams, jellies, and extracts. Their popularity is due to the numerous health benefits they provide (Nile & Park, 2014). Berries are rich in vitamins, carotenoids, dietary fiber, and phenolic compounds, such as flavonoids, phenolic acids, and hydrolysable tannins (Beattie et al., 2005; Jimenez-Garcia et al., 2013; Olas, 2018), and provide the dietary benefits of having a high proportion of nutrients relative to calories (Skrovankova et al., 2015). Berry-rich diet has been shown to be associated with a lower risk of chronic disease development, for example, type 2 diabetes, and can have preventive effects on several serious diseases, such as cancer (Baby et al., 2018; Joseph et al., 2014; Kristo et al., 2016; Seeram, 2008). It should be noted that even though berries are botanically defined as fleshy seed-containing fruits that are derived from a single ovary, do not contain a stone, and whose pericarp is divided into three layers (Hickey & King, 2000), in common usage the word "berry" refers to a soft, juicy, and brightly colored fruit that is sweet, sour, or tart. Within this review, the word "berry" refers to soft fruits commonly recognized as berries. Soft fruits that are berries only by botanical definition are not discussed here because many of them are commonly not recognized as berries. Examples include tomatoes, bananas, and grapes. On the other hand, soft fruits commonly known as berries, but which are not berries by botanical definition, such as strawberries and raspberries, are included.

Due to their numerous health benefits and high value, berries and berry-based products are often subject to adulteration. Though food fraud is not a contemporary phenomenon, it is nevertheless unjust and often committed with the intention to mislead consumers with prospect for financial gain (Spink et al., 2017). The adulteration of berries and berry-based products typically comprises the replacement of high-value and high-quality ingredients with counterparts of lower value and quality, and the consequent mislabeling of product contents or their geographical origin (Lee, 2016; Wu et al., 2018). Common examples include the adulteration of berries from the genus *Vaccinium*, such as the replacement of wild bilberries with cultivated blueberries, and the replacement of wild lingonberries with cultivated American cranberries (Hurkova et al., 2019; Lee, 2016). Because the aforementioned berries are similar in color and appearance, especially in processed form, adulteration by mixing them, or even by the addition of berries of similar appearance from other genera, is feasible (Figure 1). American cranberry, on the other hand, is often adulterated with less expensive berries, such as blueberries, or species that are not even berries, such as red peanuts (Brendler & Gafner, 2017). However, in the berry business, adulteration, especially mislabeling, is not always the result of deliberate action. It may happen unintentionally due to confusion with the names of species or failure to differentiate between two species of similar size and color. As a curious example, mixing the names of bilberries and blueberries appears even in some of the papers

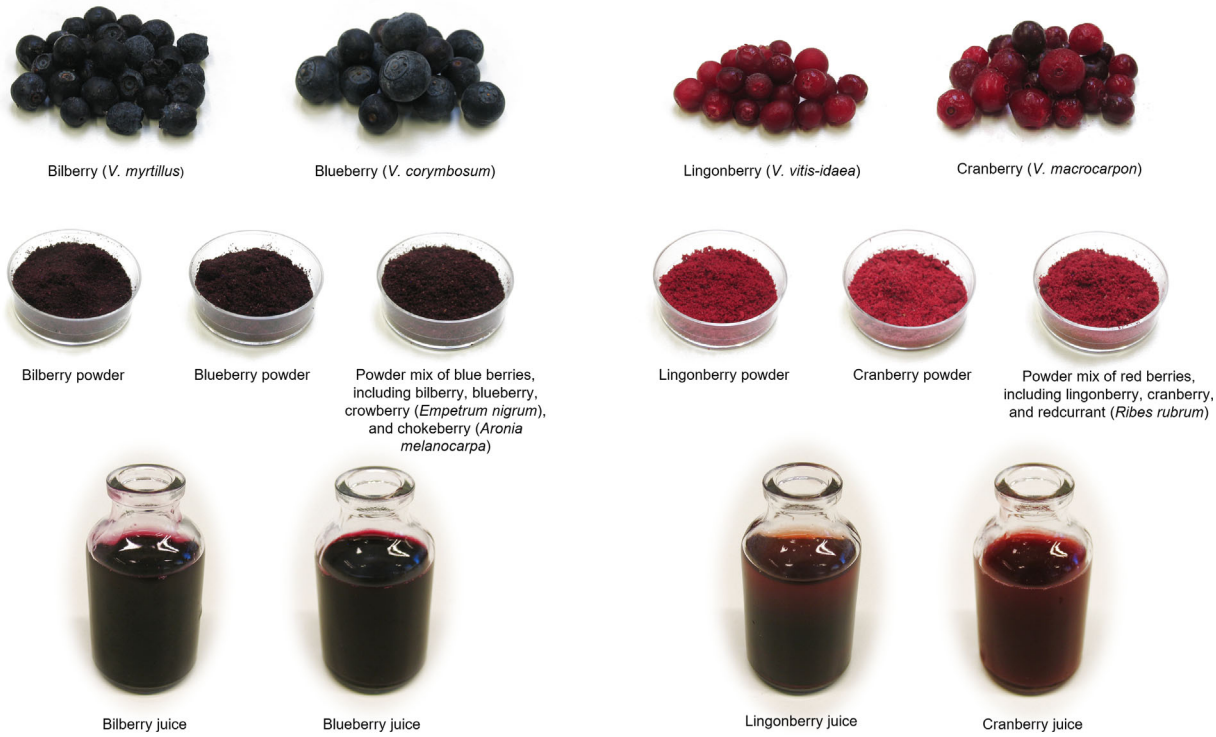

FIGURE 1 Comparison of *Vaccinium* berries of similar color and appearance, namely, bilberry and blueberry, and lingonberry and cranberry, as well as commercially available powders and juice, the former including examples of mixes with species from other genera

related to their authentication. Another issue is the use of one name as an umbrella for many different species, such as cranberry for American cranberry, bog cranberry, and small cranberry (*Vaccinium microcarpum*), both in products and research papers, thus hiding the identity of the actual species.

The authenticity of food products has become a major concern for consumers and the whole food industry from agriculture and manufacturing to distribution and regulation. Therefore, the demand for reliable, rapid, and cost-effective authentication methods as a countermeasure is increasing. In general, food authentication can be defined as a process in which the ingredients of a given product are verified not to be in contradiction with its label description (Danezis et al., 2016). Such methods include analytical chemistry and nucleic acid–based analyses for the identification of ingredients and the detection of adulterants and chemical additives (authentication), as well as the determination of raw material origin (traceability). Even though numerous methods have been applied for the authentication of plant-based food products, the number of authentication studies and method development related to the authentication of berries and berry-based food products, especially those that utilize DNA-based methods, is still scarce.

This review presents a comprehensive summary of the application of analytical chemistry and DNA-based methods, as well as the use of electronic sensors, in research related to the authentication of berries and berry-based food products. We provide an overview for the methods' earlier use, recent advances, assess their future use, and discuss their advantages and limitations.

2 | ANALYTICAL CHEMISTRY APPROACHES

Over the last decades, numerous analytical chemistry approaches, including mass spectrometry (MS), liquid chromatography (LC), gas chromatography (GC), infrared (IR) spectroscopy, and nuclear magnetic resonance (NMR), have been employed for the investigation of metabolite profiles to determine the authenticity of raw materials in berry-based products, as well as the geographical origin of fresh or dried berries. Coupled with chemometric analyses, these methods can characterize the different properties in food samples, provide chemical fingerprints for species or origin determination, and allow the handling of complex and large datasets (Medina et al., 2019). However, because the accuracy of many of these methods is influenced by environmental conditions, combination of multiple methods, or the use of multiple data-processing techniques, is often employed.

2.1 | Mass spectrometry

MS, including isotope ratio mass spectrometry (IRMS), high-resolution mass spectrometry (HRMS), and inductively coupled plasma mass spectrometry (ICP-MS), often used in tandem with chromatographic techniques or applied as tandem MS (MS^2), provides an important analytical tool for different aspects of food authentication (Medina et al., 2019). IRMS, often coupled with other analytical methods, such as LC or GC, is commonly applied to acquire information on the geographical origin of food ingredients. It relies on the detection of isotope ratios for naturally occurring light elements, also known as bioelements (carbon [C], nitrogen [N], sulfur [S], oxygen [O], hydrogen [H]), and heavy elements, also known as geoelements (strontium [Sr], lead [Pb]), whose ratios are indicative of the geology, environmental conditions (sun radiation, light cycles, temperature, and precipitation), and agricultural practices at a given location (Zhao et al., 2014). HRMS, applied either directly or in combination with other methods, such as LC, enables both targeted and nontargeted detection of compounds, as well as provides more comprehensive information on their characteristics, such as molecular mass and elemental composition. It allows the analysis of, for instance, polyphenols (Lucci et al., 2017), which are abundant in the fruit of many species of berries. ICP-MS, on the other hand, allows the fast detection and screening of metals and nonmetals (Drivelos & Georgiou, 2012).

MS-based methods have been applied for the discrimination of the geographical origin of several species of unprocessed and dried berries (Table 1). The studies do not include the analysis of processed products. However, because berries are often sold as fresh, mixes of fresh berries, or dried berries, the development of MS-based methods for the authentication of their origin is important. For instance, by applying IRMS and inductively coupled plasma atomic emission spectrometry, coupled with principal component analysis (PCA) and canonical discriminant analysis (CDA), Perez et al. (2006) discriminated between two different geographical locations and different varieties of several berry species, including strawberry, New Jersey blueberry (*Vaccinium caesariense*), and highbush blueberry (*Vaccinium corymbosum*). Oregon-grown strawberries were successfully separated from those grown in Mexico and Chile by PCA. Furthermore, based on multielement profiling, CDA provided with even better classification. In addition to differences between countries of origin, blueberries showed subregional differences and differences between varieties.

Camin et al. (2009) performed a preliminary evaluation of the effectiveness of isotopic analysis in determining the geographical origin of highbush blueberry. IRMS and specific natural isotopic fraction–NMR (SNIF–NMR) were performed on fresh berries collected from Italy, Poland, and Romania. ^{13}C and ^{15}N values differed between the locations, reflecting the horticultural characteristics and cultivation practices. This work was further continued by Perini et al. (2018) by performing a preliminary evaluation of isotope analysis in determining the cultivation practices, geographical origin, and species identification of strawberry, blueberry, and several species from the *Rubus* and *Ribes* genera collected from Italy, Poland, and Romania. IRMS and SNIF–NMR, coupled with PCA, separated strawberries and currants from the other species irrespective of their geographical origin or farming systems used. However, when all samples were considered together, the different farming systems or the geographical origin of the berries could not be determined.

To develop an approach for tracing the geographical origin of blackcurrant (*Ribes nigrum* L.) in China, Li et al. (2013) analyzed the isotope ratios of C, N, H, and O in blackcurrant fruits and leaves, as well as in soil samples, collected from four different cultivation regions. Based on the isotopic ratios of N, H, and O found in fruit, leaf, and soil samples, IRMS, coupled with discriminant analysis (DA), separated between the four regions. Isotope ratios in soil samples showed highly significant positive correlation with the fruits and leaves from the same location.

Several studies have explored the feasibility of utilizing MS-based methods for tracing the geographical origin of wolfberries. For instance, to trace the geographical origin of wolfberries grown in China and Macedonia, Balabanova et al. (2016) analyzed the major and minor element contents of dried berries by ICP-MS, coupled with PCA for data-analysis. Based on the contents of elements, PCA gave a precise classification of the samples into their countries of origin due to an enrichment of the total element contents in the Chinese wolfberries. Similarly, Zhang et al. (2017) performed an ICP-MS-based study to discriminate between wolfberries of different geographical origin, namely, the valuable Zhongning (ZNG) county wolfberries from lower-value berries originating from other regions in China, that is, from non-Zhongning (NZNG) regions. Nineteen of the 20 analyzed minerals showed significant variation between the regions. Followed by linear discriminant analysis (LDA), a satisfying result was achieved with 95.7% of the ZNG wolfberry samples correctly classified. However, the authors concluded that a larger dataset would be required to achieve a robust classification model. Meng et al. (2019) employed a novel GC–combustion–IRMS coupled with headspace-solid phase microextraction–based strategy to build a discrimination model for the geographical origin of wolfberries from three different provinces in China. Based on three volatile compounds, LDA was successful in separating the samples

TABLE 1 Summary of mass spectrometry–based studies for determining the geographical origin of berries

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Strawberry, blueberry (*V. caesariense/corymbosum*)	Fresh berries	ICP-AES, IRMS	PCA, CDA	Geographical origin	Perez et al., 2006
Blueberry	Fresh berries	IRMS, SNIF–NMR	–	Geographical origin	Camin et al., 2009
Blackcurrant	Fresh berries, leaves, soil samples	IRMS	DA	Geographical origin	Li et al., 2013
Wolfberry	Dried berries	ICP-MS	PCA	Geographical origin	Balabanova et al., 2016
Wolfberry	Fresh berries	ICP-MS	PCA, LDA	Geographical origin	Zhang et al., 2017
Strawberry, raspberry, blackberry (*Rubus fruticosus*), blueberry, whitecurrant, blackcurrant, redcurrant	Fresh berries	IRMS, SNIF–NMR	PCA	Geographical origin	Perini et al., 2018
Wolfberry	Fresh berries	GC–IRMS + HS-SPME	PLS-DA, LDA	Geographical origin	Meng et al., 2019
Wolfberry	Fresh berries	IRMS, ICP-MS, HPLC–DAD–MS	PCA, CA, FSDA	Geographical origin	Bertoldi et al., 2019
Blueberry, bilberry	Fresh berries	IRMS, ICP–OED	PCA	Geographical origin	Klavins et al., 2021

Abbreviations: CA, cluster analysis; DA, discriminant analysis; FSDA, forward stepwise discriminant analysis; GC–IRMS, gas chromatography isotope ratio mass spectrometry; HPLC–DAD–MS, high-performance liquid chromatography diode array mass spectrometry; HS-SPME, headspace solid-phase microextraction; ICP-AES, inductively coupled plasma atomic emission spectroscopy; ICP-MS, inductively coupled plasma mass spectrometry; ICP–OED, inductively coupled plasma spectrometry with optical emission detection; IRMS, isotope ratio mass spectrometry; LDA, linear discriminant analysis; PCA, principal component analysis; PLS-DA, partial least squares discriminant analysis; SNIF–NMR, specific natural isotopic fraction–nuclear magnetic resonance.

according to their origin. Finally, in a study by Bertoldi et al. (2019), the ratios of five light stable isotopes, 57 mineral elements, and 14 carotenoids were successfully employed to distinguish between Asian and Italian goji berries. The study included dried berries from Italy, China, Mongolia, and Tibet, analyzed by a combination of IRMS, ICP-MS, and high-performance liquid chromatography (HPLC). Coupled with forward stepwise DA, the combination of different methods provided with 100% correct classification of the samples, based on light stable isotope ratios, as well as elemental and carotenoid profiles.

Recently, Klavins et al. (2021) employed inductively conducted plasma spectrometry with optical emission detection and IRMS to analyze variation in elemental composition and isotopic ratios of elements in blueberries and bilberries of different origin. Significant differences in element contents were detected in blueberries, whereas IRMS coupled with PCA successfully clustered bilberries from Norway, Finland, Latvia, and Lithuania into distinct clusters.

Based on the aforementioned studies, tracing and verifying the geographical origin of berries is feasible through their chemical profile. However, because no processed products were tested in any of the studies, it remains to be seen whether the use of MS-based methods is limited to fresh and dried berries. On the other hand, IRMS should not be restricted by food processing and has been used for the origin analyses of plant-based foods, for example, orange juice (Rummel et al., 2010). One caveat with some of the studies presented here is a small set of samples. To correctly classify geographical origin, it would be essential to include a large set of samples. Moreover, the number of studies, as well as the number of different species included, is relatively low. Therefore, future studies should focus on the inclusion of a wider range of different species, as well as the establishment of reference databases of isotopic ratios for different regions. Reference values are an absolute necessity for the confirmation of a specific origin and the identification of an unknown origin. However, it should be noted that even though isotope ratios show detectable variation between different regions, they are influenced by climatic conditions, and therefore the isotopic profile of a given area may vary due to changes in climatic conditions even at yearly basis. Moreover, samples may have an identical isotopic signature even if they are of distant origin, but from an area with similar geology and climate.

2.2 | Chromatography

Chromatographic techniques, including GC, LC, and capillary zone electrophoresis (CZE), present with highly sensitive, reproducible, robust, and reliable systematic approaches for the detection of specific marker compounds, as well as contaminants, in foods (Dasenaki & Thomaidis, 2019; Esteki et al., 2018). They are commonly applied for the profiling of specific compounds or for a broader fingerprinting of metabolites (Cuadros-Rodríguez et al., 2016) and are often used in combination with spectrometric or spectroscopic methods, as well as chemometrics, or are applied as two-dimensional chromatography, such as GC × GC or LC × LC (Cortes et al., 2009). GC is suitable for the determination of volatile or semivolatile molecules, such as alcohols, esters, terpenes, aldehydes, terpenoids, hydrocarbons, acids, sulfur compounds, lipids, and ketones (Dewulf et al., 2002), whereas LC is used for the determination of various compounds, such as amino acids, fatty acids, organic acids, sugars, and phenols (La Barbera et al., 2017). CZE refers to a family of separation techniques used in food authentication for the characterization of, for example, simple inorganic ions, small organic molecules, peptides, and nucleic acids (Kvasnička, 2005). Chromatographic methods represent the most utilized group of methods for the authentication of berries and berry-based products (Table 2).

2.2.1 | Gas chromatography

The number of GC-based studies related to the authentication of berries and berry-based products is low. Nevertheless, GC has been applied in the detection of adulterants, as well as discrimination of botanical and geographical origin, regarding both unprocessed and processed products.

Studies on fresh and dried berries have been performed with sea buckthorn, blueberry, and wolfberry. For instance, Socaci et al. (2013) used GC–MS in combination with in-tube extraction in order to develop a fast method for the discrimination of wild and cultivated sea buckthorn (*Hippophaë rhamnoides* L., ssp. *Carpatica*) berries. The chromatographic matrices acquired from mashed berries, and the resulting raw juice, were subjected to cluster analysis and PCA. The wild and cultivated berries were successfully separated based on the qualitative composition of volatile compounds. Kim et al. (2015) studied the possibility of separating cultivars of highbush blueberry using GC–MS-based metabolic profiling coupled with various multivariate statistical analyses. Berries from three cultivars growing at the same farm in the Republic of Korea were measured for their phenolic content and free-radical scavenging ability, but were also subjected to a global metabolite analysis. Although PCA failed to separate all the cultivars, partial least squares discriminant analysis (PLS-DA) and hierarchical cluster analysis (HCA) were successful in differentiating them. More recently, Cossignani et al.

TABLE 2 Summary of chromatography-based studies for determining the authenticity of berries and berry-based food products

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Blueberry (*V. angustifolium*), strawberry, raspberry, blackberry (*R. fruticosus*), blackcurrant	Jam	HPLC	–	Analysis of ACN profiles	García-Viguera et al., 1997
Strawberry	Purée	SPME–GC, GC–MS	PCA, PLSR	Detection of adulteration	Reid et al., 2004
Bilberry	Extracts	HPLC–MS, NMR	–	Detection of adulteration	Penman et al., 2006
Sea buckthorn (different species and subspecies)	Fresh berries	HPLC	–	Differentiation of species	Chen et al., 2007
Cranberry (*V. macrocarpon*)	Dried berries, extract, juice, juice cocktail	HPLC	–	Detection of ACNs	Brown & Shipley, 2011
Strawberry, raspberry, blackcurrant, cranberry (*V. oxycoccos*)	Purée, juice concentrate	HPLC	–	Analysis of ACN and betacyanin profiles	Obón et al., 2011
Blueberry (*V. corymbosum*), blackberry (*Rubus*), blackcurrant, redcurrant, wild strawberry, raspberry	Fresh berries, juice	TLC, HPLC	–	Identification of species by ACNs	Filip et al., 2012
Cranberry (*V. macrocarpon* & *V. oxycoccos*), lingonberry	Frozen berries	UHPLC–UV–MS^2	–	Differentiation of species	Jungfer et al., 2012
Bilberry	Fresh berries	HPLC–DAD	–	Geographical origin	Primetta et al., 2013
Sea buckthorn (ssp. *Carpatica*)	Fresh berries, juice	GC–MS	PCA, CA	Geographical origin	Socaci et al., 2013
Wolfberry	Fresh berries	LC–QTOF-MS	PCA, PLS–DA	Geographical origin	Bondia-Pons et al., 2014
Bilberry, black mulberry, black chokeberry, blackberry (*Rubus nigra*)	Frozen berries, extracts, dietary supplements, juice	UPLC–DAD–MS, UPLC–MS^2	–	Detection of adulteration	Gardana et al., 2014
Bilberry, elderberry, bog bilberry, lingonberry, cranberry (*macrocarpon*)	Extracts	HPLC	–	Detection of adulteration	Govinda raghavan, 2014
Wolfberry (two different species)	Fresh berries	UPLC–MS, FIMS	PLS-DA, PCA	Geographical origin	Lu et al., 2014
Cranberry (*V. macrocarpon*)	Fresh berries, juice, powder capsules, syrup, sachets, extracts	HPLC, UHPLC–HRMS, CZE	PCA	Differentiation of raw material (species)	Navarro et al., 2014
Blueberry (*V. corymbosum*)	Fresh berries	GC–MS	PCA, PLS-DA, HCA	Differentiation of cultivars	Kim et al., 2015

(Continues)

TABLE 2 (Continued)

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Wolfberry (two different species)	Fresh berries	UPLC–MS, FIMS	PCA, SA, HCA	Geographical origin	Lu et al., 2015
Cranberry (*V. macrocarpon*)	Fresh berries, juice, dried berries, extracts, powder capsules, syrup, sachets	LC–ESI–MS^2	PCA	Differentiation of raw material (species)	Puigventós et al., 2015
Blueberry (two different species)	Fresh berries	HPLC–DAD–MS	PCA	Differentiation of cultivars/origin	Dongnan et al., 2016
Wild strawberry	Fresh berries	LC–MS, HPLC	PCA	Geographical origin	D'urso et al., 2016
Cranberry (*V. macrocarpon*), lingonberry, bilberry, blueberry (*V. corymbosum*)	Dietary supplements	HPLC–DAD	–	Detection of adulteration	Lee, 2016
Cranberry (*V. macrocarpon*)	Fresh berries, juice, powder capsules, syrup, sachets, extracts	UHPLC–APPI–MS^2, UHPLC–ESI–MS^2	PCA, CA	Differentiation of raw material (species)	Parets et al., 2016
Blue honeysuckle, bilberry, black chokeberry	Fresh berries, confiture, jam, syrup	HS–SPME/GC × GC–TOF-MS	–	Botanical origin	Chmiel et al., 2017
Blackcurrant, black chokeberry	Juice concentrate	UPLC–TOF-MS	PLSR, rPLSR	Detection of adulteration	Dubin et al., 2017
Bilberry, blueberry (three different species)	Frozen berries, juice, concentrate	UHPLC–MS^2	–	Differentiation of raw material (species)	Heffels et al., 2017
Cranberry (*V. macrocarpon*), blueberry (*V. corymbosum*), raspberry	Fresh berries, dried berries, juice	HPLC–UV	PCA, PLSR	Detection of adulteration	Pardo-Mates et al., 2017
Cranberry (*V. macrocarpon*)	Extracts	HPLC–UV	PCA, PLSR	Detection of adulteration	Puigventós et al., 2017
Cranberry (*V. macrocarpon*), blueberry (*V. corymbosum*), raspberry	Extracts, juices, dried berries, pharmaceutical preparations	UHPLC–HRMS	PCA, PLSR	Detection of adulteration	Barbosa et al., 2018
Bilberry	Frozen berries, jam, juice, liqueur	HPLC–UV/DAD, HPLC–ESI–MS	PCA	Analysis of ACN profiles	Benvenuti et al., 2018
Wolfberry	Dried berries	HRGC–FID/MS	PCA, LDA	Geographical origin	Cossignani et al., 2018
Blueberry (*V. corymbosum*), cranberry (*V. macrocarpon*)	Juice, juice concentrates	LC–QTOF-MS	PCA–DA, OPLS-DA	Detection of adulteration	Zhang et al., 2018
Cranberry (*V. macrocarpon*), blueberry (*V. corymbosum*), raspberry	Fresh berries, juice, raisins, pharmaceutical preparations	UHPLC–HRMS	PCA, PLS-DA, PLSR	Detection of adulteration	Barbosa et al., 2019
Wolfberry	Fresh berries	HPLC–DAD–MS, IRMS, ICP-MS	PCA, CA, FSDA	Geographical origin	Bertoldi et al., 2019

(Continues)

TABLE 2 (Continued)

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Lingonberry, cranberry (*V. macrocarpon*)	Fresh berries, dried berries	UHPLC–HRMS^2	PCA, PLS-DA	Detection of adulteration	Hurkova et al., 2019
Black chokeberry	Frozen berries, pomace, juice, extracts	HPLC–PDA, HPLC–ESI–MS^n	–	Detection of adulteration	Rodríguez-Werner et al., 2019
Bilberry, lingonberry, bog bilberry, crowberry	Fresh berries	GC–MS	PCA	Differentiation of species	Trivedi et al., 2019
Cranberry (*V. macrocarpon*), black chokeberry, elderberry, blackberry (species not defined), blackcurrant, raspberry	Fresh berries, extracts, dietary supplements	UHPLC–DAD–HRMS	PCA	Detection of adulteration	Gardana et al., 2020
Strawberry	Frozen berries	HPLC–UV/RI	PCA, LDA, SIMCA, PLS-DA	Botanical origin (cultivation system)	González-Domínguez et al., 2020
Wolfberry	Fresh berries	HPLC–DAD, MS^2	SA, HCA, PCA, OPLS-DA	Botanical origin (variety/year)	Liu, Wang, et al., 2020
Wolfberry	Dried berries	UHPLC–QTOF-MS	PCA, PLS-DA	Geographical origin	Lv et al., 2020
Bog bilberry, bilberry, cranberry (*V. macrocarpon*), lingonberry, crowberry, gaultheria, rowanberry, hawthorn, blueberry (eight varieties)	Fresh berries	GC-MS	PCA	Differentiation of species	Klavins & Klavins, 2020

Abbreviations: CA, cluster analysis; CZE, capillary zone electrophoresis; FIMS, flow-injection mass spectrometry; FSDA, forward stepwise discriminant analysis; GC–MS, gas chromatography–mass spectrometry; HCA, hierarchical cluster analysis; HPLC, high-performance liquid chromatography; HPLC–DAD, high-performance liquid chromatography with diode array detector; HPLC–DAD–MS, high-performance liquid chromatography diode array mass spectrometry; HPLC–ESI–MS, high-performance liquid chromatography electrospray ionization mass spectrometry; HPLC–ESI–MS^n, high-performance liquid chromatography electrospray ionization sequential mass spectrometry; HPLC–MS, high-performance liquid chromatography–mass spectrometry; HPLC–UV, high-performance liquid chromatography with ultraviolet detector; HPLC–UV/RI, high-performance liquid chromatography with ultraviolet/refractive index detector; HRGC–FID/MS, high-resolution gas chromatography coupled with flame ionization detector/mass spectrometry; HS-SPME/GC × GC–TOF-MS, headspace solid-phase microextraction combined with two-dimensional gas chromatography time-of-flight mass spectrometry; ICP-MS, inductively coupled plasma mass spectrometry; IRMS, isotope ratio mass spectrometry; LC–ESI–MS^2, liquid chromatography electrospray ionization tandem mass spectrometry; LC–MS, liquid chromatography mass spectrometry; LC–QTOF-MS, liquid chromatography quadrupole time-of-flight mass spectrometry; MS^2, tandem mass spectrometry; NMR, nuclear magnetic resonance; OPLS-DA, orthogonal partial least squares discriminant analysis; PCA, principal component analysis; PCA–DA, principal component analysis–discriminant analysis; PLS-DA, partial least squares discriminant analysis; PLSR, partial least squares regression; rPLSR, recursive weighted partial least squares regression; SA, similarity analysis; SIMCA, soft independent modeling of class analogy; SPME–GC, solid-phase microextraction gas chromatography; TLC, thin-layer chromatography; UHPLC–APPI–MS^2, ultrahigh-performance liquid chromatography atmospheric pressure photoionization tandem mass spectrometry; UHPLC–DAD–HRMS, ultrahigh-performance liquid chromatography coupled with diode array detector and high-resolution mass spectrometry; UHPLC–ESI–MS^2, ultrahigh-performance liquid chromatography electrospray ionization tandem mass spectrometry; UHPLC–HRMS, ultrahigh-performance liquid chromatography–high-resolution mass spectrometry; UHPLC–HRMS^2, ultrahigh-performance liquid chromatography–tandem high-resolution mass spectrometry; UHPLC–MS, ultrahigh-performance liquid chromatography–mass spectrometry; UHPLC–QTOF–MS, ultrahigh-performance liquid chromatography coupled with quadrupole time-of-flight mass spectrometry; UHPLC–UV–MS^2, ultrahigh-performance liquid chromatography–ultraviolet tandem mass spectrometry; UPLC–DAD–MS, ultraperformance liquid chromatography diode array mass spectrometry; UPLC–MS, ultraperformance liquid chromatography mass spectrometry; UPLC–MS^2, ultraperformance liquid chromatography tandem mass spectrometry; UPLC–TOF-MS, ultraperformance liquid chromatography time-of-flight mass spectrometry.

(2018) performed a high-resolution GC–based study to discriminate between wolfberries originating from China, Italy, and Mongolia. Albeit the study was performed on a small sample set, PCA and LDA separated the berries of different origin based on their fatty acid and sterol lipid fractions.

As an application of GC, lipid profiles could be utilized as biomarkers in authenticity testing due to their resistance to degradation during food processing. For instance, plant cuticular waxes, being composed of lipophilic compounds, have been widely used as biomarkers in paleogeological studies as well as chemotaxonomic studies to classify plant species (Schwark et al., 2002). In a recent study by Trivedi et al. (2019), GC–MS-based analysis showed significant differences in the composition of cuticular wax layer between four different berry species, namely, bilberry, lingonberry, bog bilberry, and crowberry. PCA on wax constituents clearly separated the species into distinct clusters indicating that wax analysis has potential in species authentication. Differences were found from the quantities of several classes of compounds, such as alkanes, fatty acids, and triterpenoids. Furthermore, adriaticol, an isoarborinol derivative and a potential biomarker, was found from the cuticular wax layer of lingonberry. In another recent study by Klavins and Klavins (2020), the analysis of cuticular wax composition of different berry species and cultivars of blueberry showed that different species, and even cultivars of the same species, can be separated based on the fatty acid profile of their cuticular wax layer.

Applications of GC on processed berries are very few. Reid et al. (2004) studied the potential of solid-phase microextraction–GC coupled with chemometrics to detect adulteration of raspberry purée with apple purée at different percentages (v/v). PCA was successful in separating samples with higher percentages of adulteration (≥70%), whereas partial least squares (PLS) regression showed separation capability at ≥40% adulteration. It was concluded that even though the method has potential for the detection of adulteration percentages, it would need further optimization to extend its capabilities. In another study, Chmiel et al. (2017) successfully applied headspace solid-phase microextraction in combination with two-dimensional GC–time-of-flight MS in order to develop an analytical procedure for the determination of aroma-active monoterpenes in samples of fresh berries from blue honeysuckle (*Lonicera caerulea* L.), bilberry, and chokeberry (*Aronia melanocarpa* Michx.), as well as in lightly processed berry products, including jam, confiture, and syrup. The results indicated that monoterpene contents could be used as an indicator for the geographical and botanical origin of berries as they reflect these even in processed products.

Considering the studies summarized here, GC could provide with effective means for the authentication of unprocessed berries, especially with the novel addition of lipid analysis. However, because the number of studies is low and the analysis of processed products is very scarce, the application of GC-based analyses on berry-based products should be explored further, especially considering lipid-based differentiation. Moreover, it is noteworthy that metabolites in plants might be influenced by environmental changes and hence large datasets would be needed to verify the feasibility of GC-based methods for authentication purposes, especially considering the determination of geographical origin.

2.2.2 | Liquid chromatography

Applications of LC, especially HPLC and ultra-HPLC (UHPLC), are the most numerous of all the methods used for berry authentication (Table 2). Because a significant number of the applications of LC are based on the analysis of anthocyanin (ACN) contents and profiles in ACN-containing berries, especially species from the genus *Vaccinium*, we focus on the analysis of ACN profiles by LC in the first part of this section. The analysis of ACNs is followed by the analysis of other compounds in *Vaccinium* berries, which in turn is followed by the use of LC in the authentication of wolfberries. Finally, LC-based studies regarding other berry species are discussed.

Authentication by ACN profiling

ACNs are a class of water-soluble pigments responsible for the red, blue, and purple color of plant tissues. They belong to a class of molecules known as flavonoids, and have numerous health-promoting properties (Smeriglio et al., 2016). Especially species from the genus *Vaccinium* are rich in ACNs (Moyer et al., 2002), and are included in all the published studies regarding authentication of berries by ACN analysis.

Because ACNs are considered beneficial for human health, they are sought after, and therefore ACN-containing products present a potential target for adulteration. For instance, Penman et al. (2006) reported the adulteration of a commercial bilberry powder that contained less ACNs than stated in its label. Based on an HPLC analysis, the adulterated extract did not contain bilberry at all. Instead, analysis by NMR revealed that the extract contained a coloring agent known as amaranth that gives a spectrophotometric profile similar to ACNs and masks the detection of adulteration by means of single-wavelength spectroscopic assays.

The recognition of species by ACN analysis is based on qualitative differences in their ACN profiles, that is,

ACN fingerprints. Some studies have focused solely on the characterization and detection of ACN profiles in berry products. For instance, Obón et al. (2011) developed an HPLC-based method for the determination and detection of ACNs and betacyanins, as well as natural and synthetic pigments in purées and juice concentrates, including purées of strawberry and raspberry, and juice concentrates of blueberry and blackcurrant. Brown and Shipley (2011) used HPLC coupled with ultraviolet light (HPLC–UV) for the detection and quantification of five of the six predominant ACNs in American cranberry products, including freeze-dried berries, powdered extracts, juice-cocktails, and juice, demonstrating the applicability of HPLC–UV for the authentication of products containing ACN-rich berries.

In order to determine the authenticity of berry jams by ACN analysis, García-Viguera et al. (1997) analyzed the ACN profiles of blueberry, strawberry, raspberry, blackberry (*Rubus fruticosus* L.), and blackcurrant jams by HPLC. They observed that the profiles of ACNs were characteristic to the corresponding fresh fruit and that the manufacturing process had no effect on the qualitative profiles of ACNs in the studied species. Filip et al. (2012) used HPLC and thin-layer chromatography to obtain ACN and anthocyanidin fingerprints from berry fruits of six different species, and to use the fingerprints to determine the authenticity of commercial berry juices. The juices had ACN and anthocyanidin profiles with matching characteristics to the profiles of their listed ingredients, therefore confirming their authenticity. In a study by Gardana et al. (2014), ultraperformance LC (UPLC)-based profiling and quantitation of ACNs in bilberry fruit from seven different countries, as well as in extracts of black mulberry (*Morus nigra* L.), chokeberry and blackberry (*Rubus nigra* L.), was used as reference for the authentication of bilberry-based products. Adulteration by mulberries and chokeberries was detected in extracts and capsules, whereas the content of ACNs was lower than declared in approximately 60% of the extracts and 33% of the food supplements. Some of the tested extracts, capsules, and juices did not contain ACNs at all. Govindaraghavan (2014) analyzed the levels of ACNs as well as the ACN profiles of bilberry extracts and extracts of suspected adulterant species by HPLC. Adulteration was detected in the studied extracts based on their ACN profiles. There were lower-than-stated contents of ACNs, as well as wrong profiles, which indicate adulteration by other species, some of which remained unknown. Lee (2016) analyzed the ACN profiles of marketplace dietary supplements of *Vaccinium* species including American cranberry, lingonberry, bilberry, and blueberry. Some of the products were observed to contain no ACNs at all, and according to the ACN profiles, over 30% of the analyzed products did not contain the berries listed as ingredients.

Even though the occurrence of species-specific qualitative ACN profiles can be used to distinguish between different ACN-rich species of berries, they do not allow the determination of geographical origin. Moreover, quantitative differences in ACNs do not allow the determination of raw material origin in processed products because the amounts of ACNs are influenced by product processing, as shown by Benvenuti et al. (2018). However, quantitative differences in ACNs could be used as an origin indicator for wild berries because ACN contents in bilberry show variation between different geographical locations with higher quantities at northern latitudes and higher altitudes (Åkerström et al., 2010; Burdulis et al., 2007; Lätti et al., 2008; Pepkolaj et al., 2017; Zoratti, Jaakola, et al., 2015). In this regard, in a study by Primetta et al. (2013), ACN profiles of ripe bilberries collected from Finland and Turkey were analyzed by HPLC coupled with diode array detector (HPLC–DAD). The study showed that although the ACN profile of bilberries is a consistent feature, there are significant differences in the proportions of sugar moieties between berries from the different countries. A total of 96.7% of the samples were correctly classified into their geographical origin by a logistic regression model. In another study, Dongnan et al. (2016) analyzed ACN contents in different varieties of blueberries (*Vaccinium corymbosum* L. and *V. corymbosum* L./*V. angustifolium* Ait.) originating from four different regions in China. PCA based on differences in ACN proportions was successful in separating the cultivars and regions.

As shown by the aforementioned studies, determining ACN contents in fresh berries can be used to separate samples of different geographical origin. Although this may be feasible when comparing specific locations within a smaller area, such as country level, the establishment and use of extensive reference databases for typical region-specific ACN values would be required to trace fresh berries whose origin is unknown or mislabeled. Moreover, berries originating from areas of similar geographical and climatic conditions, as well as daylength, would likely produce very similar ACN profiles even when not in close proximity, making them undistinguishable from each other.

LC-based authentication of Vaccinium *species*

Numerous studies related to the authentication of berries and berry-based products by means of LC-based analysis have focused on species from the genus *Vaccinium*, including cranberries, lingonberries, blueberries, and bilberries. Some of these studies were already described in the ACN section. However, besides analyses based on ACNs,

Vaccinium species have been target for LC-based authentication studies involving other compounds as well, including other flavonoids and polyphenolics. Most of these studies include cranberry.

Jungfer et al. (2012) analyzed and compared the procyanidin profiles and concentrations of frozen berries from three *Vaccinium* species, namely, American cranberry, European cranberry, and lingonberry, by UHPLC–UV–MS^2. Based on differences in procyanidin patterns, the authors concluded that these compounds could be suitable for the determination of authenticity regarding the studied species. In another study regarding American cranberries and lingonberries, Hurkova et al. (2019) utilized UHPLC coupled with high-resolution MS^2 for a metabolomic fingerprinting of the two species in order to discriminate between them, and to detect the adulteration of lingonberry with cranberry. Based on polyphenol and phospholipid markers, fresh berries were discriminated by PCA and PLS-DA. Dried berries, on the other hand, were separated by a different marker, myricetin, which is not present in lingonberries.

Several studies have been performed to develop means for the detection of adulteration with grape in American cranberry-based products. First, multiple studies were conducted in order to develop straightforward methods for the characterization and separation of cranberry and grape-based products based on their polyphenolic composition, that is, polyphenolic fingerprints. These studies included analyses by HPLC and CZE (Navarro et al., 2014), LC–MS^2 (Puigventós et al., 2015), and UHPLC–atmospheric pressure photoionization–MS^2 (Parets et al., 2016). Each method, coupled with PCA, was successful in separating cranberry and grape-based products, including dried berries, juices, extracts, powder, capsules, syrup, and sachets, from each other. However, because only samples of 100% cranberry or grape were analyzed in the aforementioned studies, Puigventós et al. (2017) went further and used an HPLC–UV-based polyphenolic fingerprinting approach to detect grape adulteration in cranberry extracts at different percentages ranging from 2.5% to 50%. Coupled with either PCA or PLS, clear separation was achieved even at the lowest percentages of adulteration. The analysis of polyphenolics was further expanded by Pardo-Mates et al. (2017) by focusing on the detection of adulteration in cranberry extracts supplemented with grape, blueberry, or raspberry. HPLC–UV coupled with PCA provided a reasonable classification regarding the kind of fruit involved. Coupled with PLS, prediction errors in the quantitation of adulterants were below 4.3% even at very low adulterant levels (2%). In another study from the same research group, Barbosa et al. (2018) developed a UHPLC–HRMS-based method for the analysis of polyphenolic profiles in cranberry, blueberry, raspberry, and grape products. Coupled with PCA, samples were separated according to the raw material species. Moreover, in mixes of cranberry extract with the adulterant species at different percentages, very good quantitation of their contents was achieved by PLS. Finally, Barbosa et al. (2019) used the same species and setup in a nontargeted UHPLC–HRMS analysis and aimed at sample classification and authentication based on chemical descriptors. Combined with PLS-DA, UHPLC–HRMS provided acceptable discrimination between products of the different species. In addition, PLS showed good performance in detecting the level of adulteration in cranberry extracts. However, because polyphenolic profiles may not be sufficient alone to detect adulteration, especially if it happens by the addition of raw materials devoid of polyphenolic compounds, Gardana et al. (2020) focused on finding other markers for detecting adulteration in cranberry extracts. By using ACN fingerprints, as well as epicatechin/catechin, procyanidin A2/total anthocyanidin, and procyanidin/ACN ratios, adulteration was detected in four commercial extracts and six food supplements. Adulterant species included black mulberry and *Hibiscus*.

Zhang et al. (2018) identified robust biomarkers by targeted and untargeted LC/MS-based metabolomics analysis for the authentication of blueberry and cranberry juices, as well as juice concentrates. Adulteration was simulated by adding apple and grape juices to blueberry and cranberry juices at different percentages (10%–50% v/v). Based on 41 metabolites, significant differences were detected between authentic berry fruit juice and their adulterants. Moreover, PCA–discriminative analysis clearly separated juices from different species.

Besides studies related to the traceability and authentication of cranberries, LC has been applied in the analysis of other *Vaccinium* species as well. In a study including fruit, juice, and juice concentrate from bilberry, highbush blueberry, lowbush blueberry (*V. angustifolium*), and bog bilberry (*V. uliginosum*), Heffels et al. (2017) analyzed the qualitative and quantitative profiles of iridoids. Significant differences were observed in iridoid profiles between the species. For instance, not all iridoids present in bilberries and bog bilberries could be found in the two blueberry species analyzed. Because iridoids are stable and found even in highly processed products such as juice, and may be exclusive to specific species, they present a potential authenticity marker for *Vaccinium* species. Future studies should explore the applicability of iridoid analysis in the separation of species in more detail.

LC-based authentication of wolfberries

Because wolfberry is an important species in China and has been introduced in Europe as well, there are numerous studies regarding its authenticity, some of which have been performed using LC-based methods. The studies

focus mainly on the discrimination of wolfberries of different origin or different cultivars. For instance, Bondia-Pons et al. (2014) performed metabolite profiling of wolfberry fruit from Tibet, northern China, and Mongolia by LC coupled to quadrupole time-of-flight MS. Followed by PCA and PLS-DA, clear differences were observed between berries of different origin, showing that metabolite profiling combined with multivariate statistics would be a useful tool for wolfberry origin classification. However, because the study included samples from only three different locations, two of which were in China, Lu et al. (2014) did a follow-up study by performing LC–MS and flow-injection mass spectrometry (FIMS) analyses to separate the more expensive wolfberry fruit grown in Ningxia from berries grown in five other provinces in China. PLS-DA based on data from both methods was successful in separating Ningxia berries from the other regions. In a similar study by the same authors, wolfberry fruit from five different provinces in China, including Ningxia with four different cultivars, were characterized by UPLC–MS and FIMS (Lu et al., 2015). Even though HCA grouped samples from Ningxia together, no perfect separation was achieved because one sample from Gansu was grouped with Ningxia, and the different cultivars were not separated. As a follow-up for the previous studies, Liu, Wang, et al. (2020) used HPLC–DAD and MS^2 combined with similarity analysis, HCA, PCA, and orthogonal partial least squares discriminant analysis (OPLS-DA) to distinguish between wolfberry samples of different varieties and different growing years. OPLS-DA could clearly and systematically identify all five varieties and the six different growing years albeit on a small sample set. Finally, to provide tools for the separation of the valuable ZNG wolfberries grown in Ningxia from NZNG wolfberries, Lv et al. (2020) performed an LC–MS-based nontargeted metabolomic approach, coupled with PLS-DA, on dried berries of the two types. Distinction between ZNG and NZNG was achieved with the use of two sets of combinative biomarkers.

These studies show that LC-based methods combined with multivariate statistical analysis tools provide with means suitable for the identification of wolfberries of different origin. However, some of the studies were performed on small sets of samples and therefore should be considered only preliminary. Moreover, the identification of origin based on biomarkers is only feasible when comprehensive databases of typical values for different regions are available.

LC-based authentication of other berry species

Besides studies performed with species from the *Vaccinium* and *Lycium* genera, there are several studies dealing with the traceability/authenticity analysis of berries by LC-based methods. For instance, fruit from both wild strawberry and strawberry have been analyzed by LC to find discriminating factors for their geographical origin. D'urso et al. (2016) compared the phytochemical content of wild strawberries growing in two different areas in Italy. By applying targeted and nontargeted LC–MS-based techniques and PCA, the two different sampling locations were separated based on phenolic compounds. In a recent study by González-Domínguez et al. (2020), multichemical profiling and several chemometric analyses were applied to determine the effect of genotype and cultivation conditions on the chemical composition of fruit from garden strawberry. The content of ACNs, phenolic acids, sucrose, and malic acid discriminated different cultivars, whereas climatic conditions were responsible for slight changes in polyphenolic profiles. Also, cultivation conditions induced minor changes in the concentrations of ACNs and phenolic acids. The results indicate that large reference datasets would be needed to exclude the effects of climatic conditions between different years and cultivation conditions. In general, geographical origin tracing through metabolic profiling suffers from changes influenced by environmental conditions, such as weather and soil conditions.

For the detection of chokeberry adulteration in blackcurrant juice, Dubin et al. (2017) performed a preliminary study by analyzing mixes of blackcurrant and chokeberry juice concentrates at different percentages. Albeit performed on a small set of samples, LC–MS-based analysis coupled with PLS regression models was successful in detecting even low levels (5%) of chokeberry in blackcurrant concentrate. Interestingly, although chokeberry is used as an adulterant in blackcurrant products, chokeberry-based products, on the other hand, are also subject to adulteration. To tackle this issue, Rodríguez-Werner et al. (2019) analyzed the phenolic composition of chokeberries and related products, including juice, pomace, and commercial extracts. After the identification and characterization of phenolic composition by HPLC–electrospray ionization–MS and HPLC–DAD, a fingerprint was established for authenticity determination. Based on the analysis, one of the four tested commercial extracts was chokeberry, whereas the three other extracts pointed toward adulteration with blackberry.

In a study including different species and subspecies from the genus *Hippophaë*, commonly known as sea buckthorn, HPLC was used to identify and compare flavonoid profiles in berries originating from different sites in the province of Sichuan in China (Chen et al., 2007). The flavonoid profiles were suitable for species discrimination but variation in growth environments did not have a profound effect on them. Moreover, even though all the other species and subspecies were distinguishable from the commercially most important *H. rhamnoides* ssp. *sinensis*, its

subspecies *yunnanensis* had a very similar chemical profile.

The GC and LC-based studies described in this section show that the analysis of chemical profiles combined with complementary statistical tools can provide useful tools for the authentication of berries, both in terms of species identification and origin traceability. However, their use in tracing geographical origin cannot be considered definitive because the metabolite profiles of genetically identical plant materials can be affected not only by variety, growth conditions, time of harvest, and storage conditions, but also by the preparation and extraction processes used (Heffels et al., 2015). Chromatography-based analyses are therefore better suited for the differentiation of species.

One downside of HPLC- and GC-based methods is that they are time-consuming, expensive, destructive of the sample, and require a highly specialized operator. On the other hand, they can be applied for the identification of potential markers, which can then be analyzed with more user-friendly methods. However, targeted chromatographic analyses cannot guarantee the absence of other not-known adulterants (Pawar et al., 2017). Furthermore, the analysis of ACNs, for instance, cannot be used to identify adulterant species devoid of these compounds. On the other hand, nontargeted analytical methods that determine the entire composition of food metabolites without the need for individual marker identification are becoming more and more popular (Dasenaki & Thomaidis, 2019).

2.3 | Spectroscopy

A wide array of spectroscopic techniques, including near-infrared (NIR), mid-infrared (MIR), Raman spectroscopy, surface-enhanced Raman spectroscopy (SERS), and NMR, have been employed for the detection of adulterants and the authentication of food products during the past decades (Hatzakis, 2019; Lohumi et al., 2015). These methods benefit from being fast, sensitive, nondestructive, noninvasive, automatic, and inexpensive, and coupled with chemometric tools (Biancolillo et al., 2020), they provide with effective alternatives for the determination of food properties. Authentication analyses regarding berries and berry-based products have been performed mostly with IR-based methods and NMR (Tables 3 and 4).

2.3.1 | IR spectroscopy

IR-based methods, including NIR, MIR, and far-infrared (FIR), as well as Fourier transform IR (FT-IR), produce a spectrum of fingerprints by absorbing the incoming IR radiation at a specific wavelength in liquid, solid, and gaseous samples. These methods can be used to determine a unique "fingerprint" of a given sample based on its functional groups and structural characteristics and to use this information in sample identification (Cozzolino, 2012). Studies on berries have mainly focused on the detection of adulterants in strawberry and raspberry jams and purées, and tracing the geographical origin of wolfberries. However, there are a few studies related to the authentication of other species as well.

Authentication of strawberry and raspberry jams and purées

The adulteration of strawberry and raspberry jams and purées by the addition of cheaper ingredients, such as apple and plum, is a common adulteration. Therefore, multiple studies based on FT-IR, horizontal attenuated total reflectance (HATR), and visible–near-infrared spectroscopy (vis–NIR) have been performed for the differentiation of fruit type, as well as for the detection of adulterants in strawberry and raspberry jams and purées. For instance, Defernez et al. (1995) applied FT-IR and HATR in combination with PCA and DA for the differentiation of strawberry, raspberry, and apple purées at 800–2000 cm^{-1}. In combination with DA, FT-IR and HATR were 100% successful in differentiating the purées of different species. Continued by Defernez and Wilson (1995), the FT-IR spectra of jams were analyzed to differentiate strawberry-containing jams from those without strawberry. Diffuse reflectance IR Fourier transform spectroscopy combined with DA classified the samples to strawberry and nonstrawberry with almost 100% success. To provide a method for the detection of adulterants in raspberry purée, Kemsley et al. (1996) used FT-IR coupled with PLS on self-made purée adulterated by the addition of sucrose, apple, and plum at different percentages (w/w). Apple and plum adulterations were detected at ~20% (w/w), whereas the addition of sucrose was detected at ~4% (w/w). Even though the detection limits for apple and plum were quite high, the method provided with a rapid protocol for the detection of these adulterants. Similarly, in a study by Holland et al. (1998), FT-IR in combination with PLS was applied to develop a fast method for the detection of adulterants in strawberry purées, including commercial products. Detection of adulteration was evaluated by adding apple and plum to strawberry purée at different percentages (w/w). With PLS regression analysis, all adulterants were detected down to levels that would be expected for adulterated products on the market, and approximately 96% of industrially manufactured purées were correctly classified by the method in a blind test as either strawberry of raspberry.

Contal et al. (2002) developed a vis–NIR-based method for the detection and quantification of apple adulteration in strawberry and raspberry purées. Rapid detection of

TABLE 3 Summary of spectroscopy-based studies for determining the authenticity of berries and berry-based food products

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Strawberry, raspberry	Purée	FT-IR, HATR	PCA, DA	Differentiation of raw material (species)	Defernez et al., 1995
Strawberry, raspberry	Jam	DRIFTS, HATR	PCA, PLSR	Differentiation of raw material (species)	Defernez & Wilson, 1995
Raspberry	Purée	FT-IR	PLSR	Detection of adulteration	Kemsley et al., 1996
Strawberry, raspberry	Purée	FT-IR	PLSR	Detection of adulteration	Holland et al., 1998
Strawberry, raspberry	Purée	vis–NIR	PLSR	Detection of adulteration	Contal et al., 2002
Strawberry, raspberry	Purée	vis–NIR	PCA, SIMCA, PLSR	Detection of adulteration	Downey & Kelly, 2004
Cranberry (species not defined), blueberry (species not defined)	Juice	FT-IR	PCA, SIMCA, HCA	Differentiation of raw material (species)	He et al., 2007
Wolfberry (eight different species)	Dried berries	FT-IR, SD-IR, 2D-IR	–	Differentiation of species	Yao et al., 2010
Wolfberry	Dried berries	FT-IR	PCA, SIMCA, DA	Geographical origin	Gao et al., 2015
Wolfberry	Fresh berries	NIR	PCA, LS-SVM, BP-ANN, KNN, Si-PLS	Geographical origin	Tingting et al., 2016
Wolfberry (*L. ruthenicum*)	Fresh berries	NIR	PCA, CA, LDA, BP-ANN, LS-SVM, KNN, Si-PLS	Geographical origin	Yahui et al., 2017
Wolfberry	Fresh berries	NIR-HSI	PCA, SVM, NN-RBF, ELM	Geographical origin	Yin et al., 2017
Bilberry	Extracts	FT-NIR, HPLC–DAD	PCA, PLSR, MD	Detection of adulteration	Gardana et al., 2018
Sea buckthorn (five subspecies)	Powder	FT-IR, SD-IR, 2D-IR	PCA, PLS-DA	Differentiation of raw material (species)	Liu et al., 2018
Blackcurrant, black chokeberry, strawberry, raspberry	Juice, nectar, syrup	TFS, TSFS	PARAFAC, PLS-DA	Differentiation of raw material (species)	Sikorska et al., 2020
Wolfberry	Fresh berries	NIR-HSI, NIR	SVM, LDA, Softmax	Geographical origin	Wang et al., 2020
Strawberry	Purée (dataset)	FT-IR	GRU, LSTM, TCN	Detection of adulteration	Zheng et al., 2020

Abbreviations: 2D-IR, two-dimensional correlation infrared spectroscopy; BP-ANN, back propagation artificial neural network; CA, cluster analysis; DA, discriminant analysis; DRIFTS, diffuse reflectance infrared Fourier transform spectroscopy; ELM, extreme learning machine; FT-IR, Fourier-transform infrared spectroscopy; FT-NIR, Fourier-transform near-infrared spectroscopy; GRU, gated recurrent unit; HATR, horizontal attenuated total reflectance; HCA, hierarchical cluster analysis; HPLC–DAD, high-performance liquid chromatography with diode array detector; KNN, K-nearest neighbors; LDA, linear discriminant analysis; LS-SVM, least squares support vector machine; LSTM, long short-term memory; MD, Mahalanobis distance; NIR, near-infrared spectroscopy; NIR-HSI, near-infrared hyperspectral imaging; NN-RBF, neural network with radial basis function; PARAFAC, parallel factor analysis; PCA, principal component analysis; PLS-DA, partial-least squares discriminant analysis; PLSR, partial least squares regression; SD-IR, second derivative infrared spectroscopy; SIMCA, soft independent modeling of class analogy; Si-PLS, synergy interval partial least squares; SVM, support vector machine; TCN, temporal convolutional network; TFS, total fluorescence spectroscopy; TSFS, total synchronous fluorescence spectroscopy; vis–NIR, visible–near-infrared spectroscopy.

TABLE 4 Summary of nuclear magnetic resonance–based studies for determining the authenticity of berries and berry-based food products

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Blueberry (*V. corymbosum*)	Fresh berries	SNIF–NMR, IRMS	–	Geographical origin	Camin et al., 2009
Blackberry (*two different species*)	Dried berries, leaves	^{1}H-NMR, RAPD	PCA, PLS-DA, HCA	Differentiation of species	Park et al., 2012
Blueberry (two species), cranberry (*V. macrocarpon*)	Leaf extracts	^{1}H-NMR	PCA	Differentiation of species	Markus et al., 2014
Sea buckthorn (two subspecies)	Frozen berries	^{1}H-NMR	PCA, PLS-DA	Differentiation of species/origin	Kortesniemi et al., 2014
Sea buckthorn (seven species and seven subspecies)	Fresh berries	^{1}H-NMR, qNMR	PCA, PLS-DA	Differentiation of species/cultivars	Liu et al., 2017
Strawberry, raspberry, blackberry (*R. fruticosus*), blueberry, whitecurrant, blackcurrant, redcurrant	Frozen berries	SNIF–NMR, IRMS	PCA	Geographical origin	Perini et al., 2018

Abbreviations: 1H-NMR, proton nuclear magnetic resonance; HCA, hierarchical cluster analysis; IRMS, isotope ratio mass spectrometry; PCA, principal component analysis; PLS-DA, partial least squares discriminant analysis; qNMR, quantitative proton nuclear magnetic resonance; RAPD, random amplified polymorphic DNA; SNIF–NMR, specific natural isotopic fraction–nuclear magnetic resonance.

adulteration was achieved for levels greater than 20% (w/w) in strawberry, and between 10% and 20% in raspberry. With PLS regression, prediction errors of 5.5% (strawberry) and 3.4% (raspberry) were achieved with the most accurate models. However, because the study did not include sulfited purées, Downey and Kelly (2004) performed a follow-up study by detecting and quantifying apple adulteration in strawberry and raspberry purées into which sodium metabisulfite had been added as a preservative. Vis–NIR coupled with PCA, soft independent modeling of class analogy (SIMCA), and PLS provided detection limits of approximately 18% and 23% (w/w) of apple adulteration in strawberry and raspberry purées, respectively. More recently, Zheng et al. (2020) used a publicly available dataset of MIR spectra from almost 1000 fruit purées, divided in two classes, strawberry samples and nonstrawberry samples, the latter including common adulterants such as apple and plum, in order to employ deep neural networks (DNNs) for the classification of pure and impure strawberry purées. Of the three different DNNs tested, best performance for the separation of strawberry and nonstrawberry purées was achieved with a temporal convolutional network, providing a classification accuracy of 98.65%.

Although these studies provide with quick and nondestructive methods for the detection of adulteration, they have a very narrow range of applications. However, they prove that the use of IR-spectra can provide with efficient tools for the detection of adulteration as long as the adulterants are known. Their applicability on other species should be explored further.

IR-based authentication of wolfberries

FT-IR and NIR have been used for the analysis of species from the genus *Lycium* mainly for the geographical traceability of fresh and dried berries of *L. barbarum* and *L. ruthenicum* Murr. For instance, the cultivation region of mature ZNG wolfberries (*L. barbarum*) from Ningxia, China, was successfully differentiated from four NZNG wolfberries, also from the Ningxia region, by FT-IR coupled with DA (Gao et al., 2015). For calibration sets, recognition rates of 93.98% for ZNG and 90.91% for NZNG were achieved, whereas for the validation sets, recognition rates were 100% for both ZNG and NZNG. In turn, Tingting et al. (2016) developed a method for the differentiation of *L. barbarum* berries originating from four different topographical regions in China. NIR at 10,000–4000 cm^{-1} for the test sets, when coupled with least squares support vector machine (LS-SVM) and back-propagation artificial neural network pattern recognition algorithms, provided satisfactory discrimination rates of 96.67% and 94.50%, respectively.

Though the aforementioned studies provided fast means for origin determination, they were destructive of the sample. Therefore, to provide a nondestructive IR-based method for tracing the geographical origin of *L. barbarum*, the feasibility of near-infrared hyperspectral imaging (NIR-HSI) was tested on berries originating from four different areas in China (Yin et al., 2017). Best recognition rates were achieved with an extreme learning machine model using characteristic wavelengths from PCA loadings, giving accuracies of 93.25% and 90% for the calibration and prediction sets, respectively. Similarly, Wang et al. (2020) performed an NIR-HSI-based analysis to discriminate between wolfberries originating from five different locations, four in China and one in Inner Mongolia. Best results were achieved by using zero-phase component analysis whitening for spectral preprocessing, followed by PLS-based dimension reduction, and Softmax Regression for data analysis. The samples of different origin were separated with 96.4% accuracy albeit on a small set of samples.

NIR coupled with multiple chemometric models was used for the rapid determination of ACN contents, as well as the origin of samples of *L. ruthenicum*, also known as black Goji (Yahui et al., 2017). The study included wild and cultivated Chinese black Goji berries from one location, inferior and superior quality berries from another, and adulterated berries from the market. The best results for correct classification of sample origin were achieved with LS-SVM. Recognition rates for the calibration and prediction sets were 100% and 98.18%, respectively, showing excellent performance.

Besides studies related to the geographical origin of wolfberries, Yao et al. (2010) successfully differentiated eight different species of *Lycium* based on their IR spectra and variation in peak intensities. The study was conducted using FT-IR, second derivative IR spectra, and two-dimensional correlation infrared spectroscopy (2D-IR) on air-dried berries.

IR-based authentication of other berry species

IR-based studies comprise the analysis of bilberries, blueberries, cranberries, and sea buckthorn. The studies are few, but focus on the detection of different ingredients, that is, species, in processed products, including juice, powders, and extracts. For instance, He et al. (2007) performed an FT-IR-based analysis coupled with several chemometric tools to develop a simple, rapid, and reproducible method for the differentiation of juice manufactured from small fruits, including cranberry and blueberry, and the detection of adulterants. The method allowed for the differentiation of juices from different species through the analysis of different fractions in the MIR range. Hundred percent correct classification was obtained from phenol-rich fractions by a SIMCA model, as well as HCA. Pure juice, as well as blends, was clustered accordingly.

More recently, Liu, Zhang, et al. (2018) performed a multistep IR coupled with multivariate data analysis for the quick and nondestructive classification and identification of sea buckthorn berry powders from five different subspecies of *H. rhamnoides*. The multistep IR consisted of FT-IR, further analysis by second derivative IR (SD-IR), and final analysis by 2D-IR to amplify and validate the FT-IR and SD-IR spectra. By performing PCA and PLS-DA on characteristic IR signals, clear separation of all the subspecies was achieved.

In another recent study, Gardana et al. (2018) studied the feasibility of Fourier transform-NIR (FT-NIR) as a rapid, low-cost, and nondestructive routine method for the detection of adulteration in commercial bilberry extracts, and the determination of their ACN contents. A preliminary detection of ACNs, as well as possible adulteration, was performed with HPLC–DAD, followed by the comparison of FT-NIR prediction models to the preliminary analysis. HPLC detected adulterations with mulberry, chokeberry, and blackberry, which were then confirmed by FT-NIR and PLS regression. Moreover, the technique allowed for the quantitation of total ACNs in bilberry extracts.

2.3.2 | Nuclear magnetic resonance

NMR spectroscopy is based on the magnetic properties of spinning nuclei. It presents a rapid and nondestructive method applied in metabolomics, food analysis, quality control, and research for determining the molecular structure of organic compounds in a given sample. What makes NMR ideal for food analysis is that its application does not require tedious sample preparation. On the other hand, its use is hindered by high costs and relatively low sensitivity (Hatzakis, 2019). NMR has been applied in several studies for the differentiation of berry species, as well as for the determination of their geographical origin (Table 4). However, because some NMR-based studies concerning the latter were accompanied with IRMS analysis, those studies are described in the IRMS section of the article.

Park et al. (2012) used proton NMR (^{1}H-NMR) along with random amplified polymorphic DNA (RAPD) technique to discriminate between two different species of black raspberry, namely, *Rubus coreanus* and *Rubus japonicus*, and to separate samples of different geographical origin. RAPD analysis was performed using raspberry leaves and NMR using berries, collected from different provinces in Korea. Classification of the samples into two species by NMR-based metabolic profiling matched the result obtained from RAPD. Moreover, PLS-DA score plot based

on NMR showed clustering of samples according to their geographical origin.

NMR has been applied for the comparison of metabolic profiles of different sea buckthorn (*Hippophaë*) species, as well as berries of different geographical origin. For instance, Kortesniemi et al. (2014) performed ^{1}H-NMR-based fingerprinting to distinguish between berries of wild *H. rhamnoides* ssp. *rhamnoides* collected from three different locations in Finland and *H. rhamnoides* ssp. *sinensis* from six different locations in China. NMR coupled with PLS-DA showed excellent performance in separating Finnish berries from Chinese berries. The discriminative factors were related to different metabolites that are affected by different growth conditions. In another study, Liu et al. (2017) used ^{1}H-NMR and quantitative ^{1}H-NMR coupled with PCA and PLS-DA to identify potential discriminating metabolites between the seven species and subspecies of sea buckthorn native to China. Primary metabolites, such as organic acids, sugars, L-quebrachitol, and secondary metabolites, such as flavonoid aglycones and flavonoid glycosides, were identified as discriminative metabolic markers. Successive PCA and PLS-DA both discriminated between the species. Furthermore, separation of subspecies was successful when they were considered separately from the main species.

To distinguish between three different *Vaccinium* species, namely, lowbush blueberry, Alaskan blueberry (*V. ovalifolium*), and American cranberry, Markus et al. (2014) performed an ^{1}H-NMR-based fingerprinting analysis. NMR spectra coupled with PCA clearly clustered the species into three distinct clusters without any overlap. However, the study was performed using leaf extracts. Therefore, its applicability on fresh berries should be explored further.

Though the aforementioned studies show potential for the application of NMR in the identification of berry species and their geographical origin, they were all performed by using unprocessed berries or leaves. Therefore, their application for the authentication of processed products remains to be examined. On the other hand, these studies prove that NMR-based analyses provide with good nondestructive tools for origin identification and authentication of species that are sold as fresh berries. However, the routine use of NMR-based methods to trace geographical origin would require the establishment and use of reference databases.

2.3.3 | Fluorescence spectroscopy

The analysis of berry-based products with spectroscopic methods besides IR and NMR is very limited. However, recently, total fluorescence spectroscopy and total synchronous fluorescence spectroscopy were used to characterize commercial berry beverages to develop classification models for products manufactured from chokeberry, blackcurrant, raspberry, and strawberry (Sikorska et al., 2020). Both methods showed spectra characteristic for the studied species. Parallel factor analysis extracted four fluorescent components that showed differences among the beverages from the different species and were assigned to different groups of phenolic compounds. Finally, PLS-DA successfully classified the beverages based on their fluorescence.

3 | DNA-BASED METHODS

DNA-based methods rely on the detection of variation present in specific DNA sequences that allows for the discrimination of different species (Böhme et al., 2019). Even though numerous methods have been developed for the sensitive, rapid, cost-effective, and repeatable discrimination of species and subspecies, the number of authentication studies regarding berries is low (Table 5). In general, a limiting factor for the use of DNA-based methods in food authentication is the difficulty of obtaining good-quality DNA from highly processed products (Gryson, 2010). Moreover, many species of berries contain substances that may hamper DNA extraction, such as polysaccharides and phenolic compounds, including ACNs. Another challenge lies in the difficulty of finding enough discriminative differences in DNA sequences between closely related species. However, unlike metabolite profiles or isotopic ratios, the information contained within DNA molecules is not influenced by short-term variation in environmental factors.

DNA-based methods can be divided into hybridization-based, polymerase chain reaction (PCR)-based, and sequencing-based techniques. Despite some trials with hybridization methods, such as restriction fragment length polymorphism (Clarke et al., 2008), authentication studies that focus on berries have been performed mainly with PCR- and sequencing-based methods.

3.1 | PCR-based techniques

Unlike many other PCR-based methods, RAPD, sequence characterized amplified region (SCAR), and analysis of simple sequence repeats (SSRs) do not require prior sequence information for the amplification of DNA. RAPD utilizes a PCR with short and arbitrary oligonucleotide primers to generate a high number of DNA fragments. Despite problems related to its reproducibility, RAPD profiling has been, and still is, widely used in species identification from food and feed. To circumvent

TABLE 5 Summary of DNA-based studies for determining the authenticity of berries and berry-based food products

Berry species	Material type	Technique	Chemometric methods	Purpose of analysis	Reference
Wolfberry (different species and varieties)	Fresh berries	RAPD	–	Differentiation of species	Zhang et al., 2001
Raspberry	Yogurt, jam	DNA barcoding	–	Detection of adulteration	Ortola-Vidal et al., 2007
Blueberry (*V. corymbosum*), elderberry	Leaves, fresh berries, juice concentrate, smoothie	PCR-RFLP	–	Identification of species	Clarke et al., 2008
Wolfberry (two different species)	Dried berries	SCAR	–	Differentiation of species	Sze et al., 2008
Raspberry, blueberry (*V. corymbosum*), blackberry (species not defined), redcurrant, blackcurrant, strawberry	Juice, yogurt, jam, baby food	qRT-PCR	–	Differentiation of raw material (species)	Palmieri et al., 2009
Bilberry, lingonberry, bog bilberry, blueberry (species not defined), crowberry, gooseberry, blue honeysuckle, serviceberry	Fresh berries, leaves	Bar-HRM	–	Differentiation of species	Jaakola et al., 2010
Wolfberry (three different species)	Dried berries, leaves	DNA barcoding	–	Differentiation of species	Xin et al., 2013
Sea buckthorn (seven species and seven subspecies)	Fresh berries	DNA barcoding	–	Differentiation of species	Liu et al., 2015
Sea buckthorn (seven species and seven subspecies)	Dried berries, commercial products	Bar-HRM, DNA barcoding	PCA	Detection of adulteration	Liu et al., 2018
Wolfberry (two different species)	Leaves, dried berries, seeds	ARMS, DNA barcoding	–	Differentiation of species	Wetters et al., 2018
Blueberry (four different species), cranberry (*V. macrocarpon*), lingonberry, blackcurrant, white mulberry, raspberry (three different species), mock strawberry, wolfberry (*L. chinense*), lantern fruit	Fresh berries, jam, juice, pulp	DNA barcoding	–	Detection of adulteration	Wu et al., 2018
Black chokeberry, blackberry (*R. fruticosus*), cranberry (*V. macrocarpon*), strawberry	Powder, tea, cookie	qRT-PCR	–	Detection of adulteration	An et al., 2019
Raspberry (four different species)	Leaves, dried berries	SNP markers	–	Detection of adulteration	Mohanan et al., 2019
Raspberry (three different species), cranberry (three different species), blueberry (four different species)	Leaves, dried berries, pulp, juice, jam, purée	qRT-PCR	–	Detection of adulteration	Yang et al., 2019
Wolfberry (two different species)	Leaves	SCAR	–	Identification of species	Liu, Cheng, et al., 2020

Abbreviations: ARMS, amplification refractory mutation system; Bar-HRM, DNA barcoding coupled with high resolution melting; PCA, principal component analysis; PCR-RFLP, PCR-restriction fragment length polymorphism; RAPD, random amplified polymorphic DNA; qRT-PCR, quantitative real-time PCR; SCAR, sequence characterized amplified region; SNP, single-nucleotide polymorphism; SSR, simple sequence repeat.

problems associated with RAPD, species-specific RAPD fragments can be converted into the more reliable SCAR markers. RAPD and SCAR markers have been used for the fingerprinting of *Vaccinium* berries, such as blueberry and cranberry cultivars (Carvalho et al., 2014; Cho et al., 2017; Debnath, 2005; Levi & Rowland, 1997; Polashock & Vorsa, 1997, 2002). However, along with more economical sequencing services and the accumulation of specific sequencing data from different species, methods utilizing random primers are likely to get less popular in the future.

As mentioned in the NMR section, Park et al. (2012) successfully used RAPD to confirm the identity of two different species of black raspberry collected from different provinces in Korea. Other applications of RAPD, as well as SCAR, have focused on the discrimination of different species of wolfberry. For instance, Zhang et al. (2001) used RAPD markers to distinguish between wolfberry and its related species from the genus *Lycium*. Ten arbitrary primer pairs produced species-specific fingerprints with which eight different species were distinguished. In order to further improve the reliability of RAPD-based authentication of *Lycium* species, Sze et al. (2008) performed an RAPD fingerprinting analysis followed by SCAR. By using DNA from dried berries of *L. barbarum* and its potential adulterant *L. chinense* var. *potaninii*, the newly developed species-specific SCAR primers were successful in distinguishing between the species. Moreover, when the method was applied on 10 commercially available dried berries, nine of them produced the amplification product specific to *L. barbarum*. This supports the applicability of the method for the distinction of different species, and moreover, reveals adulteration within commercial products. Recently, Liu, Cheng, et al. (2020) continued the aforementioned studies by developing a novel RAPD-based SCAR marker specific for the medicinal *L. chinense* Miller in order to separate it from other *Lycium* species, as well as from its common adulterant species *Nitraria sibirica* Pall. The newly designed primers were specific to only *L. chinense* and therefore allowed to distinguish it from the other species. Despite the low number of applications of RAPD in berry authentication, the studies described herein support that it presents a rapid and cost-effective method for species authentication, especially when combined with the downstream application of SCAR.

Microsatellites, often referred to as SSRs, are tracts of repetitive DNA in which certain DNA motifs of 2–6 base pairs (bp) are repeated variable times. SSRs are highly reproducible, polymorphic, and have been used to identify different cultivars of berry species, including blueberry, blackberry, strawberry, and cranberry (Bassil et al., 2009, 2010; Boches et al., 2006; Honjo et al., 2011). Also, inter SSRs, genomic regions between microsatellite loci, have been applied to access genetic diversity among geographically distant populations, for example, among wild lingonberry clones (Debnath, 2007) and within bilberry populations (Zoratti, Palmieri, et al., 2015). However, the application of these markers for authentication purpose needs to be explored further. The studies mentioned here were performed on DNA from berries, leaves, and shoot tips. Therefore, the applicability of these markers on processed products remains to be tested.

Applications of single-nucleotide polymorphism (SNP)-based markers in the authentication of berry-based products are also few. To authenticate bokbunja (*Rubus coreanus*) from three other *Rubus* species (*R. coreanus*, *R. crataegifolius*, and *R. occidentalis*) available in the Korean peninsula, Mohanan et al. (2019) developed a simple multiplex PCR-based method utilizing SNP markers in primer design. Primers based on two regions of DNA, namely, *rpl16* and *trnG-trnS*, successfully separated the species based on the length of the PCR products. The method was tested on commercially available dried berries labeled as bokbunja and revealed that only a few of the 31 products actually contained bokbunja. The other products consisted of one of the other species, or a combination of two. In another SNP-based study, Wetters et al. (2018) performed an amplification refractory mutation system analysis for the identification of the two closely related but distinctive species of wolfberry, *L. barbarum* and *L. chinense*. By using two diagnostic primer pairs in combination with universal *psbA* and *trnH* primers, the species were differentiated due to a single nucleotide difference. Even though the application of these markers was reproducible in fresh and dried wolfberries, the amplified sequences are likely to be too long (>600 bp) for applications with processed products. On the other hand, because wolfberries are usually sold as dried berries and not processed further, the use of long amplicons might present with a sufficient method for their authentication.

Besides conventional PCR techniques, quantitative real-time PCR (qRT-PCR) presents a very sensible method for the quantification of very small amounts of DNA, even from highly processed food products. As an application of qRT-PCR, Palmieri et al. (2009) used rRNA 5S and anthocyanidin synthase regions with species-specific primers to identify different species of berries in juices, yogurt, jam, and baby food. The presence of correctly informed berry DNA in commercial products was confirmed. Moreover, by using relative standards, the proportion of blueberry in baby food, as well as in a juice mix containing blueberry and wine grape, could be determined. The dual-labeled probes had good specificity even in multiplexed PCR when used on juice mixes. More recently, Yang, Liu, et al. (2019) developed a Taqman probe-based qRT-PCR method for the authentication of raspberry, blueberry, and cranberry

products. By using species-specific primer pairs and PCR products of less than 250 bp, they were able to detect the different species, as well as adulteration in food products, even in mixed juices and after DNA-damaging thermal treatments. Finally, An et al. (2019) used SNP/indel-based species-specific primers in order to authenticate four different berry species, namely, black chokeberry, blackberry, cranberry, and strawberry, in commercial products by qRT-PCR. The analysis was based on short (94–224 bp) amplicons from the chloroplast *matK*, *rbcL*, and *trnL-F* regions, often deployed in DNA barcoding. The amplicons were capable of detecting the target species in commercial products. All of the 18 products included were found to be consistent with their listed ingredients.

3.2 | Sequencing-based techniques

Because there is demand for accurate and reproducible authentication methods amenable in high-throughput form, methods based on sequence data are more or less replacing the PCR-based methods described above. The increase in accumulation of DNA sequence information, also on many berry species, has led to the search for DNA barcodes applicable for food authentication. DNA barcodes are universally amplifiable short sequences of DNA between 300 and 800 bp located in mitochondrial, chloroplast, or nuclear DNA, and used for species differentiation. However, because there is no single DNA barcode that could identify all plant species (Hollingsworth et al., 2011; Kress, 2017), approaches combining two or more barcoding regions have been commonly used for higher efficiency.

DNA barcoding has been applied in several studies regarding the authentication of berry-based products. For instance, the internal transcribed spacer (ITS) barcoding sequences, which are commonly applied in herbal plant identification (Michel et al., 2016; Newmaster et al., 2013), have also been used to discriminate between different species of berries. In a study by Xin et al. (2013), three different species of wolfberry, namely, *Lycium barbarum*, *L. chinense*, and *L. ruthenicum*, were successfully distinguished from each other by using the ITS2 sequence and subsequent analysis by phylogenetic tools. The DNA used was extracted from leaves and dried berries. To separate between berries of medicinal and nonmedicinal sea buckthorn species and subspecies, Liu et al. (2015) used the ITS2 and *trnH–psbA* barcoding regions separately and as a combination for effective species discrimination. All species were clearly identified by using the barcoding regions as a combination. Moreover, the approach was successful in identifying four subspecies of *H. rhamnoides*. DNA barcoding was also applied by Zhang et al. (2018) to verify the species authenticity of blueberry and cranberry juices prior to further analysis by LC.

To overcome the challenge of detecting degraded DNA in processed products, DNA mini-barcoding, which utilizes barcodes of only 100–200 bp long, can be applied (Little, 2014). However, on the downside, their use may considerably reduce the discriminative information contained within the barcoding regions and pose problems for the identification of closely related species. A mini-barcoding approach was used by Ortola-Vidal et al. (2007) to identify species in mixes of raspberry and rhubarb (*Rheum* × *cultorum*) yogurt at different ratios. Barcoding by pyrosequencing only a 104 bp region of the *rbcL* gene resulted in successful identification of species even at a mix of 2% (w/w) of rhubarb adulteration in raspberry yogurt. In another study, mini-barcodes of *rcbL*, *matK*, *psbA–trnH*, ITS, and *trnL* regions were employed in the authentication of several unprocessed and processed commercial products manufactured from cranberry, lingonberry, raspberry, and several species of blueberries (Wu et al., 2018). The *rbcL* and *psbA–trnH* barcodes discriminated between the species in highly processed products with 1%–10% sensitivity. Furthermore, the study showed that species content in nearly half (45.4%) of the tested products did not match their label descriptions.

DNA barcoding coupled with high-resolution melting analysis (Bar-HRM) is a relatively new method that combines DNA barcoding with post-PCR melting analysis. It provides with an accurate, low-cost, and rapid method ideal for mutant screening and discrimination of different species. Detection of genetic variants by HRM is based on a gradual increase in temperature and monitoring the consequent denaturation of DNA. Essentially, very small differences, such as SNPs, or small indels in short DNA sequences between different species, can be visualized by the shape of their melting curves (Druml & Cichna-Markl, 2014). Bar-HRM has been applied for the authentication of berries in two instances. A preliminary Bar-HRM analysis was performed by Jaakola et al. (2010) for the differentiation of eight different wild berry species, focusing primarily on the differentiation of bilberry from similarly colored berries. The analysis utilized the *ITS*, *rpl36-rps8*, and *trnL-F* barcoding regions and included species from the families Ericaceae, Grossulariaceae, Caprifoliaceae, and Rosaceae. In addition to separating species of different families, the method was successful in separating two different cultivars of blueberries. More recently, Liu, Xiang, et al. (2018) used the ITS2 barcode in combination with HRM analysis and PCA for the differentiation of seven sea buckthorn species, their subspecies, and the detection of adulterants in commercial sea buckthorn products. The different sea buckthorn species were successfully discriminated from each other, and the study revealed

severe adulteration in several commercial products. Subsequent sequencing revealed adulteration with four different species from an entirely different genus. All in all, even though Bar-HRM presents a promising method for simple, rapid, and cost-effective authentication of species, its sensitivity could pose a limitation for its accurate application in products containing mixed species.

The DNA-based papers described within this section show that they hold great promise for the detection of different species of berries even in processed products. The number of studies, however, is small, and therefore future studies should aim to expand their application, especially regarding methods that provide quick detection of species, such as Bar-HRM. Moreover, in addition to the emerging new nucleic acid amplification technologies, such as droplet digital PCR and loop-mediated isothermal amplification, the fast development in high-throughput sequencing technologies, as well as the emerging third-generation sequencing, has much potential in increasing the sensitivity and accuracy of food authentication methods in the future. For instance, DNA metabarcoding, that is, the use of high-throughput sequencing methods to simultaneously barcode all species in a mixed sample, has in recent years been proven to be an effective approach for species determination in different types of commercial plant product (Bruno et al., 2019).

4 | ELECTRONIC SENSORS

Electronic sensors are human-sense-mimicking electrochemical devices based on simple nonspecific sensors and a pattern recognition software system. Two types of electronic sensors, namely, the electronic nose (E-nose), which consists of an array of gas sensors for the detection of volatiles, and the electronic tongue (E-tongue), which consists of an array of sensors designed for the analysis of liquids (Peris & Escuder-Gilabert, 2016), have been used for traceability analyses of berries. The studies are few, but all recent, and focus on the discrimination of wolfberries from different regions in China.

By using a portable E-nose coupled with multivariate statistical analyses, Li et al. (2017) managed to discriminate the valuable ZNG wolfberries from NZNG wolfberries. By using E-nose on fresh berries, LDA had the best identification rate and provided a feasible method for discriminating berries originating from three different regions, one of which was ZNG. However, discrimination between berries originating from the two NZNG regions was not achieved.

A voltammetric E-tongue (VE-tongue) has been used for the discrimination of wolfberry origin in two studies. A self-developed VE-tongue coupled with chemometric tools was used by Yin et al. (2018) to provide with a quick and simple method for the differentiation of dried wolfberries from four different provinces, one of which was Ningxia, the origin of the most valuable wolfberries of China. Although PCA and LDA failed to classify the samples, a 100% classification accuracy was achieved with a support vector machine model that was optimized with a particle swarm optimization algorithm. Yang, Wang, et al. (2019) combined VE-tongue with deep learning algorithms to discriminate between dried berries from four different regions in China. Best results for origin classification were achieved with a Convolutional Neural Network at 98.27% accuracy.

Based on the studies described above, electronic sensors could provide a viable option for the authentication of berries. However, it should be noted that these studies were performed with freshly picked or dried wolfberry fruits and therefore provide methodology suitable for the origin identification of berries in either form. In the case of wolfberry, and other berries sold fresh or dried, these methods may provide with an efficient way to determine their origin. However, it remains to be seen how these methods perform in the analysis of processed products, and whether they are suitable for the analysis of other species as well. Moreover, akin to analyses of origin by other methods, reference databases would be essential for routine use. Nevertheless, methods based on electronic sensors merit from being fast and convenient with good repeatability.

5 | CONCLUSIONS AND FUTURE PERSPECTIVES

In order to serve as plausible detectors for food fraud in the future, and to respond to consumer demand, commonly deployed food authentication methods must be sensitive, accurate, repetitive, and reliable, as well as scientifically valid. From this review, it is clear that even though a number of both analytical and DNA-based methods have been applied for the authentication of berries and berry-based products, so far no universal catch-all method exists for all species and for all aspects of authentication. For instance, even though DNA-based methods have been demonstrated to be accurate and not affected by environmental factors, their ability to analyze highly processed food products, complex mixtures of species, and to discriminate on intraspecies level is still limited. Therefore, combinations of different analytical and DNA-based methods are increasingly used to complement each other in order to improve the quality assurance of authenticity analyses. The need for further method development is emphasized by the fact that many important berry species are not included in any authentication studies. In addition to broadening the applicability of the current

methods for a broader spectrum of species and samples types, future research should focus on testing the new emerging sequencing and sensor technologies, as well as the establishment of comprehensive databases to support the use of analytical methods, such as IRMS, HRMS, and NMR.

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AUTHOR CONTRIBUTIONS

Heikki Salo conceptualization-equal; investigation-lead; Writing-original draft-Lead; writing-review and editing-Lead. Nga Nguyen conceptualization-Equal, investigation-supporting; writing-original draft-supporting; writing-review and editing-supporting. Emmi Alakärppä conceptualization-equal; funding acquisition-Supporting; investigation-supporting; project administration-supporting; writing-original draft-supporting, writing-review and editing-supporting. Linards Klavins Investigation-supporting, writing-review and editing-supporting. Anne Linn Hykkerud investigation-supporting; writing-review and editing-supporting. Katja Karppinen investigation-supporting; writing-review and editing-supporting. Laura Jaakola investigation-Supporting; writing-review and editing-supporting. Maris Klavins funding acquisition-Supporting; investigation-supporting; supervision-Supporting; writing-review and editing-supporting. Hely Häggman conceptualization-equal; funding acquisition-Lead; investigation-supporting; project administration-lead; supervision-lead, writing-review and editing-supporting.

CONFLICTS OF INTEREST

The authors declare no conflicts of interest.

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