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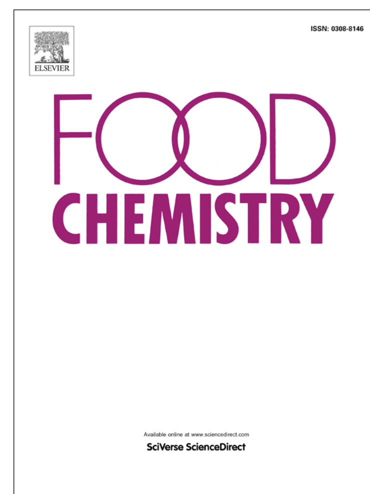

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PII: S0308-8146(17)31809-5
DOI: https://doi.org/10.1016/j.foodchem.2017.11.007
Reference: FOCH 21984

To appear in: *Food Chemistry*

Received Date: 4 May 2017
Revised Date: 16 October 2017
Accepted Date: 2 November 2017

Please cite this article as: Abbas, O., Zadravec, M., Baeten, V., Mikuš, T., Lešić, T., Vulić, A., Prpić, J., Jemeršić, L., Pleadin, J., Analytical methods used for the authentication of food of animal origin, *Food Chemistry* (2017), doi: https://doi.org/10.1016/j.foodchem.2017.11.007

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Analytical methods used for the authentication of food of animal origin

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Abstract

Since adulteration can have serious consequences on human health, it affects market growth by destroying consumer confidence. Therefore, authentication of food is important for food processors, retailers and consumers, but also for regulatory authorities. However, a complex nature of food and an increase in types of adulterants make their detection difficult, so that food authentication often poses a challenge. This review focuses on analytical approaches to authentication of food of animal origin, with an emphasis put on determination of specific ingredients, geographical origin and adulteration by virtue of substitution. This review highlights a current overview of the application of target approaches in cases when the compound of interest is known and non-target approaches for screening issues. Papers cited herein mainly concern milk, cheese, meat and honey. Moreover, advantages, disadvantages as well as challenges regarding the use of both approaches in official food control but also in food industry are investigated.

Keywords:
Food authenticity; adulteration; geographical origin; ingredients; chromatography, spectroscopy, DNA-based techniques

1. Introduction

Authentication of food products involves procedures capable of verifying that the product matches the label statements and that it conforms to the provisions of applicable laws and regulations. Different types of food fraud, including adulteration, counterfeiting, substitution and deliberate mislabelling of goods, can occur for a variety of reasons, but are often linked to financial profit achieved by adulteration intended to improve the perceived quality of products, mimic an established brand, reduce manufacturing costs or enableshelf life extension. Research conducted in this area aims to prevent food adulteration and other practices that may mislead consumers, who are entitled to truthful information about the food they consume (Reid, O'Donnell, & Downey, 2006), as stated under the Regulation 178/2002 (EC, 2002). Even though regulations transposed into national and international legislation mandate the trueness of label information, they are unfortunatelyunable to prevent food fraud (Ballin, 2010).

The ability to trace and authenticate food products is of the outermost importance for the food industry, not only for economical, but also for safety reasons (Cubero-Leon, Peñalver, &

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Maquet, 2014). In order to protect consumer interests and public health, in addition to combating the growing problems of food fraud and adulteration, scientific expertise and technologies are constantly being developed and advanced to test the authenticity of different food products. Furthermore, depending on the nature of adulterants, admixtures can also represent a health risk for the consumer. The most common type of food fraud, reported in 95 % of publications, is substitution (Stamatis et al., 2015) of an original ingredient with a similar cheaper one, difficult to recognise by the consumer and difficult to detect by routine analytical techniques.

The main fraudulent practices met in the meat industry sector are substitutions of meat ingredients with other animal species, breeds, tissues, proteins; faking of meat origin and animal feeding regime (especially when it comes to traditional/ regional meat products); modifications of processing methods, and the addition of non-meat components such as water (Ballin, 2010). Other important food frauds concern the food of animal origin such as honey, milk and dairy products, fish and seafood (Cubero-Leon, Peñalver, & Maquet, 2014). Identification of the species' origin is also important to consumers due to the economic loss arising on the grounds of fraudulent substitution, as well as for health-related (food allergies) and religious reasons (Asensio, Gonzalez, Garcıa, & Martın, 2008).

Testing of authenticity includes the analysis of ingredients, determination of geographical origin, and the production technology analysis. The use of rapid, effective and reliable analytical methods, when correctly applied to verify authenticity and traceability of a product, represents a valuable and irreplaceable tool for the authorities which aim to establish control over food products in market circulation. The authenticity and the origin of ingredients have to be labelled on the final product. The feeding regime of livestock is fundamental for the properties and safety of food of animal origin, but this regime is often hidden from the consumers.

Analytical methods that can be used for authentication of products labelled as having a Protected Designation of Origin (PDO), Protected Geographical Indication (PGI) and Certificate of Specific Character (CSC) can be divided into several categories. Tools and methodologies coming as a result of scientific innovation and technological evolution can help to quickly locate particular sophisticated frauds and adulterations. These methodologies include targeted approaches in cases when the compound of interest is known and non-target approaches for screening issues. The large-scale study by Stamatis et al. (2015), which comprised a total of 348 food products (meat, poultry and fish, milk, pet food, cheese), made use of a universal 16S rDNA marker. The results were alarming, since showing a number of mislabelled food products adulterated so as to prolong their shelf life.

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It is clear that it becomes important for food safety to apply extensive, rapid, inexpensive, but reliable food authentication methods. One of the solutions is the use of nano-materials or nano-sensors, but the safety of their use is still questioned due to the uncertainties about their bioaccumulation potential and human health impact. However, the recent report by Patra, Roy, Madhuri, & Sharma (2017) brings new details about the advancements in nano-sensor technologyused for trace-level determination of various food contaminants, offering therefore new and more beneficial prospects of this technique. However, this review shall be mainly concerned with analytical techniques most commonly used with food of animal origin.

2. Application of analytical techniques

To obtain results that allow for a reliable judgment, food authentication employs analytical techniques such as liquid chromatography (LC) and gas chromatography (GC) tandem mass spectrometry (MS), vibrational spectroscopic techniques such near-infrared (NIR) and mid-infrared (MIR) spectroscopy, Raman spectroscopy, hyper-spectral imaging (HSI), nuclear magnetic resonance spectroscopy (NMR), in addition to techniques as optical and infrared microscopy, electronic spin resonance spectroscopy (ESR), polymerase chain reaction (PCR) and enzymatic assays (ELISA). Di Stefano et al. (2010) authored the review article on the application of liquid chromatography–mass spectrometry (LC/MS) for food analysis. It contains an exhaustive list of works dealing with the characterization of food quality (authentication and adulteration). A focus on non-targeted fingerprinting for authentication of food in official control has been investigated by Esslinger, Riedl, & Fauhl-Hassek (2014). Some examples of application of analytical techniques to the end of food authentication, displayed per type of food of animal origin and the analytical purpose, are given in Table 1.

Articles on the advances in authentication of food of animal origin continue to be published. Cajka, Showalter, Riddellova, & Fiehn (2016) proposed mass spectrometry-based omics sciences as high-throughput methods permitting the assessment of food adulteration. Spizzirri and Cirillo (2016) published the book on innovative analytical tools used for food safety assessment, in which current and official analytical methods used for the analyses of sweeteners, lipids, allergen markers, antioxidant compounds and genetically modified organisms (GMOs) in food are described.

Recently, reviews on milk adulteration were published by Nascimento, Santos, Pereira-Filho, & Rocha (2017) and Attrey (2017). Siddiqui, Musharraf, Choudhary, & Rahman (2017) published the review on the use of NMR spectroscopy for authentication of honey, its botanical

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and geographical origin, and adulteration by sugar syrups. Wu et al. (2017) published the review that covers known syrup adulterants and analytical methodologies adopted for their detection in honey, such as TLC, C-isotope, HPAEC, GC, HPLC, IR, NMR, and the Raman spectroscopy in addition to metabolomics-based detection methods such as Q-TOF-MS, which are becoming ever more interesting given an increased use of different adulterants whose detection is getting more and more difficult.

This review focuses on analytical approaches exploited within the frame of authentication of food of animal origin, with an emphasis put on techniques used for the determination of specific ingredients, geographical origin or substitution of original ingredients. Papers cited in this review mainly discuss milk, cheese, meat and honey authentication by the use of most common analytical methods as spectroscopy (non-target approach) and chromatography, or DNA-based techniques (target approaches). As will be observed in this review depending on the kind of authentication of food of animal origin, analytical techniques are less or more used.

2.1. Determination of specific ingredients

Authentication of food of animal origin by the determination of specific ingredients is widely studied. Chromatographic techniques make it possible to identify food composition and then its characterization, spectroscopic techniques permit to differentiate rapidly food based on its composition while DNA-based analyses allow the detection of origin of food ingredients.

2.1.1 Chromatographic techniques

Determination of specific ingredients in different food products often makes use of chromatographic methods such as liquid (LC) and gas (GC) chromatography. These techniques are capable of separating a large number of compounds, allowing therefore for their identification using different types of detectors. Data resulting from these techniques are then compared to the information stored in databases, or to the results of analysis of authentic standards undertaken in order to identify the contents of food and screen for a given adulterant. The main difference between LC and GC is that GC is more suitable for detecting volatile and semi-volatile compounds.

The application of LC was found useful, for example, in the detection of origin of different cheeses by virtue of measuring ß-lactoglobulin (Ferreira & Caçote, 2003). To prevent possible cheese frauds, it is necessary to control whether the milk employed in the production matches the one appearing on the label. To that effect, high-performance liquid chromatography with diode-

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array detection (HPLC-DAD) can be very useful in defining the protein profile of a dairy product, and consequently also the type of milk used (Rodríguez, Ortiz, Sarabia, & Gredilla, 2010). Cow milk is also a subject to adulteration. Even though a soymilk contains a cheaper protein quite similar to that of cow milk, it has not gained high popularity, mainly due to its beany flavour and astringency. In India, vendors do not declare its addition into cow milk, which should be viewed upon as an unethical practice consumers have to be protected from. A rapid detection technique is required to detect and quantify soymilk in cow/buffalo milk. It is important that the milk present in dairy products corresponds to that advertised on the label. Rodríguez, Ortiza, Sarabiab & Gredillacet (2010) proposed the use of protein chromatographic profiles of cheese and milk extracts combined with chemometric treatments to detect adulterations.

The study by Jiye et al. (2010) showed that GC can be utilized for the detection of mechanically separated meat (MSM). MSM can be detected in raw meat mixtures down to 10 % by detecting specific metabolites using GC-MS. The study conducted so as to discriminate between intensive and extensive pig breeding was performed by Gallardo, Narváez-Rivas, Pablos, Jurado, & León-Camacho (2012). To determine triacylglicerols extracted from subcutaneous fat, analyses making use of GC coupled with FID detector were performed. Proton transfer reaction mass spectrometry coupled with time-of-flight mass analyser (PTR-TOF-MS) is successfully applied for the fast discrimination of PDOs (pectin-derived oligosaccharides) (Sanchez del Pulgar et al., 2011). However, studies have also shown that GC and LC have some disadvantages due to the fact that the target compound has to be extracted, meaning that the duration of analysis is prolonged and the laboratory throughput limited. Besides, the application of GC or GC coupled with mass spectrometry (MS) can serve the purpose of determination of fatty acids as target analytes for the species identification, while the percentage of saturated, monounsaturated and polyunsaturated fatty acids can be used as an indicator of animal species. However, an everyday practice has shown that the scale of variations is too large and can lead to less reliable results when it comes to species identification (Schwägele, 2005).

It has been found that, in order to check the trueness of food label claims, different food additives can be traced using liquid chromatography tandem mass spectrometry (LC-MS/MS). LC-MS/MS was developed to determine the species origin of bovine and porcine blood plasma in meat products (Grundy et al., 2007; Grundy et al., 2008). Fibrinopeptides released during the blood clotting process differ in mass depending on species, and thus those derived from bovine blood can be differentiated from those derived from porcine blood by determining their mass. Certain meat-binding products or ‘glues’ used to bind minced meat or off-cuts and trimmings of

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high-value meats in 'steak-like' products are often derived from blood. The blood plasma protein fibrin can be mixed with meat and, during the addition of the blood-clotting enzyme thrombin, clot to bind the meat together. This particular food-binding process therefore raises the issue of an ingredient of animal origin being added as an undeclared ingredient during manufacturing of other meat and fish products. Furthermore, since binding agents are permitted for use as food ingredients, there exists the concern that some producers might seize the opportunity to use these agents so as to fraudulently increase the declared meat content of the final product.

The use of vegetable proteins in various types of meat products is a common practice. To control food composition and specification and to avoid an allergic response, a sensitive screening method HPLC-MS/MS was established by Hoffmann, Münch, Schwägele, Neusüß, & Jira (2017) in order to detect lupine (*Lupinus angustifolius*), pea (*Pisum sativum*), and soy (*Glycine maxima*) in meat products. The limits of detection of the method were about 5 mg/kg, 4 mg/kg and 2 mg/kg of meat product for pea protein, soy protein and lupine protein, respectively, with no false-positive or false-negative results at all. The presence of soy protein was also checked for in 10 commercial brands of beef hamburgers using the isotope method (δ13C and δ15N). Only 3 % of the controlled products complied with the Brazilian legislation. The rest contained > 4 % of the soy protein, which was indicative of an extensive adulteration.

2.1.2 Spectroscopic techniques

Vibrational spectroscopic techniques used in conjunction with chemometrics have shown high-class analytical performance when it comes to food authentication. Advantages of these techniques are low costs and a little or no need for sample preparation before analysis. NIR (near infrared) signals are associated with molecular vibrations, specifically with overtones and combinations of fundamental vibrations. In general, chemical bonds between light atoms, such as C-H, O-H and N-H, have high vibrational frequencies detectible in the NIR region of 780-2,500 nm. Due to its robustness and simplicity of instrumentation and the advantage of deep sample penetration, NIR spectroscopy has been used in food research for decades. Low sensitivity related to the high signal-to-noise ratio is the most prominent disadvantage of NIR spectroscopy. Some of the applications of NIR in the field of food fraud come down to the assessment of meat adulteration using processed animal proteins (PAPs). A very useful technique used for proving food authenticity is FT-IR (Fourier transform infrared) spectroscopy. FT-IR spectroscopy focuses on the MIR (mid-infrared) region (4,000-400 cm^{-1}) of the electromagnetic spectrum. The technique provides a greater amount of chemical information regarding the scanned sample than

NIR does, because FT-IR spectroscopy measures relevant fundamental vibrations instead of overtones and combination bands that get to be measured in the NIR region.

Alike MIR spectroscopy, the Raman spectroscopic method provides structural information about proteins, water and lipids in a muscle. The Raman spectroscopy was implemented so as to predict sensory qualities like texture, tenderness and juiciness of beef samples and the fatty acid composition of different types of meat (Beattie, Bell, Claus, Fearon, & Moss, 2006), as well as to determine the fat content of a fish muscle (Marquardt and Wold, 2004), and to reveal changes occurring during frozen storage (Herrero, Carmona, & Careche, 2004). The Raman spectroscopy is a versatile, non-destructive analytical technique, which provides unique spectral fingerprints of many analytes. The main advantages over the NIR spectroscopy are that a sample can be profiled through a variety of transparent materials and that analytes can be detected in solutions with minimal water interferences. Although the Raman spectroscopy is mainly used for detecting adulteration of oils and juices, it has a potential of determining meat and meat products' freezingbased on changes inmyofibrillar and connective tissue proteins.

The Raman spectroscopy was investigated by Ellis, Broadhurst, & Clarke (2005) in order to discriminate between very close poultry species (chicken and poultry), but also between muscle groups (breast and leg). The combination of this technique with the cluster analysis has shown that the latter differentiation is more reliable than the discrimination between species. Gelatine, a water-soluble protein, is widely used in the food industry. For different reasons, its authentication has become an important issue across Muslim, Jewish, Hindu, vegan and vegetarian communities. For this purpose, ATR-FTIR (attenuated total reflection - Fourier transform infrared) method has been proposed as an economic andrapid method ofdetermination of both gelatine presence and its origin (Cebi, Durak, Toker, Sagdic, & Arici, 2016). It was successfully applied to discriminate between gelatine sources (bovine, porcine or fish) by the use of the hierarchical cluster and principal component analysis (PCA). Spectral ranges associated with amide-I (1,700–1,600 cm^{-1}) and amide-II (1,565–1,520 cm^{-1}) have been revealed to bevery significant for the above discrimination.

In their study, Hammami et al. (2010) showed the potential of fluorescence spectroscopy in combination with the factorial discriminant analysis in identifying sheep milk coming from sheep fed on different feeding regimes. The discrimination was made possible based on the content in specific intrinsic probes (aromatic amino acids and nucleic acids, tryptophan, vitamin A and riboflavin). Characterization can also be based on metabolites due to their association with dairy animal breeds or species. Metabolites can help in evaluation of milk traits and the detection of

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milk adulteration. Yang et al. (2016) used them as biomarkers when identifying differences in milk produced by the Holstein cows and other, minor dairy animals. Combination of nuclear magnetic resonance (NMR) spectroscopy and liquid chromatography–tandem mass spectrometry (LC-MS) with multivariate analysis has permitted the identification of some metabolites and has contributed to better understanding of differences in synthesis of milk coming from the Holstein cows and other dairy animals.

Jaiswal et al. (2015) proposed the use of ATR–FTIR, which revealed the differences between cow milk, soymilk and adulterated cow milk samples in the spectral region of 1,680–1,058 cm^{-1}. This discrimination range includes the bands of amide-I, amide-II, amide-III, beta-sheet protein, a-tocopherol and Soybean Kunitz Trypsin Inhibitor. The detection limit was as low as 2 %. Another kind of milk adulteration is the addition of whey prohibited by the Brazilian law. De Carvalho et al. (2015) developed a MIR spectroscopic method to detect and quantify whey adulteration of milk powder through the measurement of glycomacropeptide protein. The results indicated the successfulness of the above detection with the Pearson's coefficient of 0.9885 and a root mean square error of cross-validation RMSECV of 0.24.

Recently, NIR hyper-spectral imaging (HIS), a powerful analytical technique that uses vibrational spectroscopy for food quality and authenticity identification, has emerged. HIS attains spectral and spatial characterization of complex heterogeneous samples, whereas spectral features allow for a vast range of multi-constituent surface and subsurface features to be identified. This technique is a point-based scanning technique able to examine a very small area of a specimen. HSI has instrumental flexibility and can be used to collect hyper-spectral data on specimens of different sizes and shapes. Furthermore, the spectral region collected, the spatial resolution and the field-of-view can be adjusted depending on application. This technique has been used for the determination of water-holding capacity of fresh beef (ElMasry, Sun, Allen & 2011) and melamine adulteration of milk powder (Fu et al., 2014).

Various methods can be combined so as to determine the authenticity of different types of honey, including a direct measurement of specific marker compounds, such as methylglyoxal (MGO) and dihydroxyacetone (DHA). A NMR technique as a spectroscopic method is suitable for the analysis of complex mixtures (McKenzie, Donarski, Wilson, & Charlton, 2011). NMR is based upon the measurement of absorption of radiofrequency radiation by atomic nuclei with non-zero spins occurring in a strong magnetic field. The absorption of atomic nuclei is affected by the surrounding atoms, which cause small local modifications of the external magnetic field. In this way, detailed information about the molecular structure of a food sample can be obtained. Among

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nuclei with a non-zero spin, the isotopes of hydrogen-1 (spin = 1/2) and carbon-13 (spin = 1/2) are the most used in NMR, although other isotopes, such as nitrogen-15 (spin = 1/2), oxygen-17 (spin = 5/2), fluorine-19 (spin = 1/2), or phosphorous-31 (spin = 1/2) are also frequently employed (Luykx & van Ruth, 2008). A NMR technique has been developed to directly quantify anti-microbial MGO in honey (Donarski, Jones, Harrison, Driffield, & Charlton, 2010a), which can also be measured using liquid chromatography–infrared spectrometry (LC-IR) after derivatization. Irudayaraj, Xu, & Tewari (2003) and again Tewari and Irudayaraj (2004) applied micro-attenuated total reflectance to quantify saccharides in a number of floral honeys. In the same way, i.e. based on the phenolic fraction, it was possible for Sergiel, Pohl, Biesaga, & Mironczyk (2014) to discriminate between honey samples of different floral origin using a three-dimensional synchronous fluorescence spectroscopy.

2.1.3 DNA-based techniques

DNA is a macromolecule that contains all genetic information of an organism which makes it is an excellent target for food analysis. However, a single DNA molecule contains coding information for a high number of genes which makes it practically impossible to analyze the entire molecule. For that reason, only sample specific genes are examined using what called polymerase chain reaction (PCR) (Lee et al., 2017).Rahmati, Nurhidayatullaili, Yehye, & Basirun (2016) published the review on the identification of meat origin in food products. They highlighted the methods most extensively used for meat-producing species' identification based on the DNA hybridization techniques. They reported LODs spanning from 0.1 % to less than 0.01 %, depending on the type of meat. The polymerase chain reaction amplifying a fragment of the mitochondrial DNA D-loop region was developed by De et al. (2011) for the species-specific detection of cattle and buffalo milk. The sensitivity was excellent, allowing for the detection of 0.1 % - adulteration of cow and buffalo milk or cheese. Liao, Liu, Ku, Liu, & Huang (2017) optimized a novel DNA extraction method in order to identify milk powder based on the PCR analysis. The results showed that a sufficient amount and quality of DNA can be isolated from milk powder and analysed by both PCR and real-time PCR, allowing for the detection of cow milk components in goat milk powder. Another, also DNA-based technique wasfirst described by Cunha et al. (2016). It is called the Randomly Amplified Polymorphic DNA (RAPD) - Sequence Characterized Amplified Region (SCAR) technique and seems to be useful in the detection of origin of milk present in dairy products, which is important for the efficient detection of adulterant breeds in

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milk mixtures used for the fraudulent production of the Serra da Estrela cheese registered as a product of the Protected Designation of Origin (PDO).

Recently, Seçkin, Yilmaz, & Tosun (2017) used real-time PCR to determine the amount and origin of milk used in cheese production. The objective was to identify animal species (such as cow, sheep and goat) in a total of 90 different cheeses of 30 brands coming from various sources. The results showed that only 36.67 % of samples were produced from 100 % cow milk, while 16.67 % of goat cheese samples turned out to be produced from 100 % sheep milk. The authors also observed the absence of linear relationship between chemical composition, fatty acid ratios and the amount and origin of cheeses analysed ($P < 0.05$).

2.2. Determination of geographical origin

Geographic authentication of food of animal origin is less obvious because it needs to consider the composition of food but also the environmental conditions that have influenced this composition. Spectroscopic techniques are good candidates for that issue as they provide global information about the food sample. Besides, isotopes analyses have also been revealed very useful as the distribution of certain stable isotopes chemical elements are affected by biological-environmental interactions in addition to hydrological- and climate-based variations (http://stableisotopes.com).

2.2.1 Spectroscopic techniques

NIR spectroscopy has been successfully applied for a rapid determination of floral origin of honeys (Chen et al., 2012; Escuredo, González-Martín, Rodríguez-Flores, & Seijo, 2015). It has been shown that NIR spectroscopy can be applied in honey analysis, especiallyin order to predict its geographical origin (Woodcock, Downey, Kelly, & O'Donnell, 2007). Within the frame of the EU-funded TRACE project, oneof the project tasks was to confirm that the honey solabelled actually originates from Corsica. Several analytical techniques were employed to investigate the authenticity of the Corsican honey and to differentiate it from non-Corsican ones. To the latter end, NIR and the Raman spectroscopy were employed. Through a PLS model anda variable selection procedure, Woodcock, Downey, & O'Donnell (2009) managed to correctly classify 90.4% of Corsican and 86.3% of non-Corsican honey samples.These samples were also analysed by Fernández Pierna, Abbas, Dardenne, & Baeten (2011) using the FT-Raman spectroscopy. The authors succeed in discrimination of honey origin, with the correct classification rateo f 85% to

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90%, obtained with the Partial Least Squares-Discriminant Analysis (PLS-DA) or the Support Vector Machines (SVM).

Infrared techniques have been revealed as useful for the classification of geographical and floral honey origin (Ruoff et al., 2006; Tewari & Irudayaraj, 2005; Bertelli, Plessi, Sabatini, Lolli, & Grillenzoni, 2007). Gok, Severcan, Goormaghtigh, Kandemir, & Severcan (2015) pointed towards significant variations in spectral profile of honey samples of different botanical origin, mainly based on the differences inthe water, carbohydrate and protein contents. All samples were tested via unsupervised pattern recognition procedures like hierarchical clustering and the Principal Component Analysis (PCA). The results showed a successful discrimination of honey samples over the spectral range of 1,800–750 cm^{-1}. The traceability of honey has been widely investigated during years through various analytical techniques. Corvucci, Nobili, Melucci, & Grillenzoni (2015) proposed an innovative approach capable of improving the successfulness of honey discrimination based on its botanical/geographical origin, which couples melissopalynology and micro-FT-Raman spectroscopy techniques with multivariate analysis.

MIR spectroscopy has also been widely used for the authentication of geographical origin (Gallardo-Velázquez, Osorio-Revilla, Zuñiga-de Loa, & Rivera-Espinoza, 2009; Kelly, Downey, & Fouratier, 2004; Kelly, Petisco, & Downey, 2006a; Irudayaraj, Xu, & Tewari, 2003). Bertelli, Plessi, Sabatini, Lolli, & Grillenzoni (2007) analysed a total of 82 (robinia, chestnut, citrus, and poly-floral) honey samples. Spectral data processing by virtue of general discriminant and classification tree analysis yielded the classification accuracy of nearly 100%. Ottavian et al. (2012) showed that NIRS can be a reliable tool for the real-time authentication of the Asiagod'Allevo cheese. They developed the PLS models subsequently used to classify the studied samples according to the location(lowland or alpine) and management of the cheese-making factory the ripening age, the altitude of milk production, and the period of the cheese production year. They also used the variable importance in projection index (Andersen & Bro, 2010) to identify the most significant discriminating variables. NIR spectral data offer the same discrimination capacity as the traditional chemical analysis. Some years ago, Karoui, Mazerolles, Bosset, de Baerdemaeker, & Dufour (2007) used MIR spectroscopy to determine the geographical origin of the Gruye`re and Etivaz Swiss cheeses. The application of FDA (factorial discriminant analysis) to the first 10 principal components (PCs) of the PCA applied in different spectral regions shows the best classification rates over the regions of 3,000–2,800 cm^{-1} and 1,500–900 cm^{-1} (90.5% and 90.9%, respectively). These spectral regions can be considered as a valuable discrimination tool. In order to develop a fast and non-invasive method for the quality control of

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grated cheese, ATR-FTIR spectroscopy in combination with chemometrics (PCA and LDA (Linear discriminant analysis) (Gori, Maggio, Cerretani, Nocetti, & Caboni, 2012) was applied to classify the grated Parmigiano-Reggiano cheese from other grana-type cheeses coming from Italy, central and northern Europe. During the cross-validation procedure, the LDA-FTIR analysis showed a predictive classification ability of around 100%, making this technique suitable for the classification of PDO (Protected Designation of Origin) grated cheeses.

Lamb meat has become very popular among consumers because of its high quality resulting from a higher protein and lower fat content, evoking also a great interest in rapid and effective methods capable of authenticating its origin. To that effect, the use of NIR spectroscopy was proposed by Sun, Guo, Wei, & Fan (2012) as an effective method to predict $\delta^{13}C$ and $\delta^{15}N$ values considered to be good indicators of the relationship between biological products and their growth environment. The Partial Least Square regression PLS-R models were established, having the determination coefficient (R^2) of 0.76 and 0.87, respectively.

NMR stable isotope (2H, 1^3C, ^{15}N, ^{18}O, ^{34}S and ^{87}Sr) analyses are generally considered as excellent tools for assessing the food origin. These analyses can be used to determine the geographical origin of meat, as well as the feeding regime of animals the food was produced from (Sentandreu & Sentandreu, 2014). The basis for discrimination lies in the fact that the ratios of elements such as $^2H/^1H$, $^{13}C/^{12}C$ can vary depending on the geographical origin of soil, water and feed used on a farm. The study by Perini, Camin, Sanchez Del Pulgar Rico, & Piasentier (2013) used stable isotope ratios to authenticate Italian PDO hams. Two main techniques used for the determination of isotope ratios are isotope ratio mass spectrometry (IRMS) and site-specific natural isotope fractionation from nuclear magnetic resonance (SNIF-NMR).

SNIF-NMR was used for the determination of geographical origin of food based on the isotopic ratio of a given nucleus found in a constituent of the analysed food (Ibañez & Cifuentes, 2001). This can be explained by the fact that the specific proportions of hydrogen and oxygen isotopes present in molecules are mainly dependent on climatic conditions and geographical surroundings (Reid, O'Donnell, & Downey, 2006).

2.2.2 Isotope and elemental techniques

By virtue of more sophisticated applications of multi-element stable isotope analysis, the geographic origin (rearing location) of animals used in meat production can be determined (Heaton, Kelly, Hoogewerff, & Woolfe, 2008). This approach can be applied to any agricultural product whose provenance adds value. The use of stable isotope analysis to the effect of verifying

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the provenance of premium food products is underpinned by systematic global variations in the distribution of hydrogen and oxygen isotopes in precipitation and ground water. The measurement of stable isotope ratios was also used in the study by Heaton, Kelly, Hoogewerff, & Woolfe (2008), carried out in order to distinguish Brazilian from UK and Irish beef.

IR-MS technique was used in the study by Zhao et al. (2013), who undertook to classify beef coming from different Chinese regions. By measuring C and N stable isotope composition in addition to 23 trace elements in defatted beef, the authors were able to correctly determine the origin of 60-80 % of samples. The authors stated that, to obtain high accuracy, it is important to collect samples in the same period of year so as to avoid seasonal variations. Another study aiming to detect the origin of beef was conducted by Liu, Guo, Wei, Shi, & Sun (2013), where authors used cattle tail hair to measure stable isotope ratios.

Determination of geographical origin of milk is also an important requirement that can be met by studying the elemental composition of milk. Zain, Behkami, Bakirdere, & Koki (2016) analysed 24 essential and trace elements in raw and factory cow milk samples. The following elements - Ca, Na, Fe, Zn, Mn, K, Ba and Mg - seem to be the most discriminative when it comes to the application of chemometrics tools, helping therefore in geographical origin clustering.

2.3. Substitution of ingredients

As for the food of animal origin authentication via the determination of specific ingredients, its accomplishment via the identification of ingredient substitution needs techniques as chromatographic, spectroscopic and DNA-based techniques. The latter are extensively studied in the literature.

2.3.1 Chromatographic techniques

Some organic or synthetic compounds may also be added into meat products so as to act as colorants, aromas, preservatives, stabilizers, etc. (Nakyinsige, Bin Che Man, & Sazili, 2012). Colorants can be used to improve fresh meat appearance. Smoke aroma, for example, can be fraudulently used instead of natural meat smoking. The choice of analytical technique is determined by the nature of the target chemical compound; for the detection of organic compounds added to meat products, both HPLC and GC may be appropriate (Ballin, 2010). In case of the determination of soy in highly processed meat mixtures (Castro, García, Rodríguez, Rodríguez, & Marina, 2007), HPLC has shown potential. Liquid chromatography–electrospray–tandem mass spectrometry with a proteomic-like sample preparation was used for the detection of

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milk adulteration with cheese whey (Campos Motta et al., 2014). Caseinomacropeptide (CMP) is a peptide released by chymosinduring cheese production; it remains in whey and can be used as a biomarker of fluid milk adulteration.The method shows a satisfactory precision (< 11%), with the detection limit of 1.0 μg mL^{-1}. Earlier, a liquid chromatography–mass spectrometry has been proposed for the detection of fraudulent addition of cow milk to water buffalo milk and mozzarella. Czerwenka, Műller, & Lindner (2010) used the whey protein β-lactoglobulin as a marker of adulteration.

2.3.2 Spectroscopic techniques

One of the most common practices of meat adulteration is the addition of water to increase size, weight and, consequently, the final price of a product (Sentandreu & Sentandreu, 2014). The standard method of extraneous water determination in meat involves the determination of the water/protein ratio using differences in mass established before and after meat drying. A too high water/protein ratio serves as an indicator of water addition. However, protein and phosphate can be added to meat products thus leaving the water/protein ratio close to the natural one. In this case, the detection of a foreign protein or phosphate might provide proof of a fraudulent practice (Ballin, 2010). A more powerful technique capable of determining a fraudulent addition of water to meat is NMR, as shown by Bertram & Andersen (2007), and Sentandreu & Sentandreu (2014).

Another case is honey. Even though the adulteration of honey does not pose a health risk, it has an unfavourable impact on market growth and consumer confidence. A large-scale honey adulteration was witnessed on the world market in the 1970s, mirroring in the presence of fructose-rich corn syrup (HFCS) introduced by the industry (Mehryar, 2011). Therefore, the need for an easy-to-use, rapid, non-destructive and low-cost analytical methods such as NIR spectroscopy capable of detecting and quantifying adulteration on a commercial scale has emerged.

NIR transflectance spectroscopy was already used by Downey, Fouratier, and Kelly (2003) to detect adulteration of the Irish artisanal honey with fructose and glucose. Kelly, Petisco, & Downey (2006) developed qualitative and quantitative models to detect honey adulteration with sugar-beet invert syrup and HFCS. Later on, Zhu et al. (2010) worked on the detection of adulterants such as sweeteners (fructose/glucose mixtures) in honey using near-infrared spectroscopy and chemometrics. They applied different classification models, the best being the least square support vector machine (LS-SVM) leading to the total accuracy of 95.1%. On the other hand, wavelet transformation (WT) for data compression proved itself as a highly effective

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variable selection tool. The authors concluded that the WT-LS-SVM can be used as a rapid screening technique for the detection of this type of honey adulteration, with a good accuracy and a better generalization.

In 2011, Chen et al. were able to classify unadulterated and adulterated Chinese honeys collected from apiaries and purchased in local groceries using a FT-NIR spectrometer equipped with a fibre optic probe. The main discriminative bands were over the range of 6,000–10,000 cm^{-1}. A correct classification was attained in 100 % of unadulterated and 95 % of adulterated honey samples. Li, Shan, Zhu, Zhang, & Ling (2012) reported the use of the Raman spectroscopy as a rapid and efficient tool in the detection of HFCS-induced honey adulteration. Bázár et al. (2016) developed a NIR model for screening the unifloral Robinia honey using a fibre optic probe. The most accurate detection of adulteration was obtained over the spectral range of 1,300–1,800 nm, rich in absorption bands of both water and carbohydrates. The combination of NIR and aquaphotomics permitted a good description of differences in the water molecular structure of theRobinia honey and HFCS. Just recently, near-infrared spectroscopy (NIR) was successfully used for the qualitative and quantitative detection of honey adulterated with high-fructose corn syrup or maltose syrup (Li et al., 2017). Different chemometric tools such as the Competitive adaptive reweighted sampling (CARS) and the Partial least squares linear discriminant analysis (PLS-LDA) aid, both separately and combined, in adulteration detection and quantification. The R^2coefficients of prediction were higher than 0.9 regardless of the floral origin of a honey sample. Other less frequently used techniques have been tested to detect adulteration of honey, for instance the VIS-NIR hyper-spectral imaging system and data mining-based classifiers, which were investigated by Shafiee et al. (2016), or the determination of isotopic composition of honey in terms of δ13C and its proteins, used to assess honey adulteration with a fructose-rich corn syrup (HFCS) or other C4-adulterants. The latter technique was applied by Berriel & Perdomo (2016) to honey coming from different Uruguayan regions.

As for NIR, MIR spectroscopy combined with chemometrics has been selected by several researchers as a technique to determine honey adulteration (Sivakesava & Irudayaraj, 2001; Kelly, Petisco, & Downey 2006a; Rios-Corripio, Rojas-López, & Delgado-Macuil, 2012; Subari, Saleh, Shakaff, & Zakaria, 2012). A three-dimensional fluorescence spectroscopy has also been investigated for its adulteration-detecting potential, for example by Chen et al. (2014), who employed it in order to detect the concentration of rice syrup added as adulterant into a pure honey. Data processing by virtue of partial least squares (PLS) and back-propagation neural

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network (BP-ANN) algorithms has permitted the prediction of adulterants' concentrationsin honey (the optimum models thereby giving the RMSEP values of 0.0235 and the R values of 0.9787).

Milk is also a subject to unscrupulous producing practices in terms of adding adulterating substances in order to increase profits. The addition of sodium citrate, sodium hydroxide, sodium chloride, sucrose, phosphates, carbonates, bicarbonates and hydrogen peroxide so as to correct milk defects was reported by Hoorfar (2012). In 2013, another scandal broke out revealing the utilization of fertilizers containing urea and formaldehyde in order to mask the addition of water into milk (Talkhan, 2015). It is therefore important to establish screening and detection methods to ensure marketed milk safety. Botelho, Reis, Oliveira, & Sena (2015) proposed the use of attenuated total reflectance mid-infrared spectroscopy combined with multivariate supervised classification method to the end of detecting the presence of adulterants such as water, starch, sodium citrate, formaldehyde and sucrose in milk samples containing one to five of the above in the range of 0.5–10 % w/v. Nieuwoudt, Holroyd, McGoverin, Simpson, & Williams (2016) developed a rapid and sensitive method of liquid milk adulterants' analysis that makes use of a portable Raman spectrometer and a simple, optimized sample holder. The technique was successfully tested on adulterated milk and allowed for the limit of detection between 140 and 520 mg/L for 4 N-rich compounds and between 7,000 and 36,000 mg/L for sucrose. In the meantime, fatty acid ratios established by the Argentinean legislation were examined by Rebechi, Vélez, Vaira, & Perotti (2016) so as to detect adulterations of milk fat with animal fats. Regression models based on gas chromatography data have been proven suitable for the evaluation of these adulterations if present at levels higher than 10 % for tallow and 5 % for lard.

In the last decades, the meat industry in Europe has witnessed several scandals, one of them beingthe use of horsemeat as a new adulterant (FSA, 2013). Horsemeat is used instead of beef due to the lower breeding costs. Since an early identification of this adulterant became necessary, rapid analytical methods such as the Raman spectroscopy had to be employed. The technique was selected by Boyacı et al. in 2014 for its advantagesin fat analysis. The authors extracted pure fat from beef, horsemeat and beef adulterated with horsemeat in different concentrations (25%, 50 %, 75 % w/w). The Raman spectral data were processed using the principal component analysis. The developed model was good enough to differentiate between unadulterated beef samples and samples adulterated with horsemeat.

In recent years, many investigators applied various vibrational techniques for the detection of adulterants in bovine meat (Al-Jowder, Defernez, Kemsley, & Wilson, 1999; Ding & Xu, 2000; Meza-Márquez, Gallardo-Velázquez, & Osorio-Revilla, 2010). Alamprese, Casale, Sinelli,

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Lanteri, & Casiraghi (2013) explored the capabilities and performances of separated or combined UV-Vis, NIR and MIR spectroscopy coupled with chemometric techniques in detecting bovine meat adulteration with turkey meat (5-50 % (w/w)). They concluded that the best choice for the purpose would be the use of a fused UV-Vis-NIR-MIR data matrix. Another case of adulteration is adulteration of meatballs through the substitution of beefmeat with pork or rat meat, again undertaken to raise profits. Kurniawati, Rohman, & Triyana (2014) and Rahmania, Sudjadi, & Rohman (2015) applied FTIR spectroscopy in combination with chemometrics to quantitatively determine lard and rat meat in meatballs. The spectral regions of 1,200-1,000 cm^{-1} and 1,000-750 cm^{-1} have been revealed as very useful for the classification of lard/beef fat and rat/beef fat, respectively. Regression models used in both studies showed high-quality performances. The coefficients of determination (R^2) and the root mean square errors of calibration (RMSEC) were over 0.9 and less than 2% (v/v), respectively. Adulteration of meat with cheaper meat such as pork raises socio-religious, safety and consumer confidence issues.

In the last ten years, the suitability of FTIR in conjunction with multivariate analysis for the assessment of meat quality has been demonstrated. Mid-infrared spectroscopy combined with the SIMCA (Soft Independent Modelling Class Analogy) method was used to detect and quantify the adulteration of mincemeat with horse meat, fat beef trimmings, and textured soy protein. It permitted a 100 %-accuratediscrimination between adulterated and unadulterated samples on the basis of their protein, fat, water, and ash content (Meza-Márquez, Gallardo-Velázquez, & Osorio-Revilla, 2010). Recently, Nunes, Andrade, Santos Filho, Lasmar, & Sena (2016) analysed samples originating from criminal networks dismantled by the Brazilian Police. The scandal that brokeout in 2012 (DPF, 2012) involved the injection of solutions of non-meat ingredients (NaCl, phosphates, carrageenan, maltodextrin) into bovine meat in order to increase its water-holding capacity. The authors developed the PLS models based on ATR-FTIR spectra and physicochemical properties, the best of them allowing for a correct detection of 91% of the adulterated samples.

Rapid analytical methods continue to be developed in order to confront these issues and respond to them at different levels, going from laboratories to industries. FT-IR spectroscopy is a method that can meet the above requirements (Schmutzler, Beganovic, Böhler, & Huck, 2015). Meat and fat adulteration could be revealed down to the level of contamination of 10 % using laboratory, industrial fibre optics or on-site systems. Furthermore, it was even possible to measure directly through the polymer sample packaging, but the limits of detection varied in function of the measurement set-up. In the same way, identification and quantification of raw, frozen-thawed

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and cooked minced beef meat adulteration with turkey meat was investigated using FT-NIR spectroscopy (Alamprese, Amigo, Casiraghi, & Engelsen, 2016). The PLS regression models have been developed with R^2 higher than 0.88, while PLS-DA models have been applied to discriminate between the two sample classes (adulteration threshold = 20%) with sensitivity and specificity higher than 0.84 and 0.76, respectively. Another kind of fraud, the injection of solutions of non-meat ingredients (NaCl, phosphates, carrageenan, maltodextrin) into bovine meat in order to increase its water-holding capacity, has been investigated using data fusion (protein, ash, chloride, sodium, phosphate content) and ATR-FTIR spectroscopy (Nunes, Andrade, Santos Filho, Lasmar, & Sena, 2016). The best PLS-DA model correctly detected 91% of adulterated samples.

Finally, compositional differences between meat species can obviously be used for meat identification. Bilge, Velioglu, Sezer, Eseller, & Boyaci (2016) selected laser-induced breakdown spectroscopy (LIBS) for this purpose. The limits of detection (LOD) were found to be 4.4% for pork-adulterated beef and 2.0% for chicken-adulterated beef, respectively, which makes the LIBS a valuable technique for the routine meat quality control.

2.3.3 DNA-based techniques

Mislabelling of products has received special attention after recent events concerning the adulteration of meat products with non-declared species such as horse meat, where a range of supposedly beef products was found to contain horse meat (FSAI, 2013) in spite of the clear European Union (EU) regulations governing food traceability and labelling. This fraud was detected using DNA-based methodologies of species identification (Griffiths et al., 2014). Besides DNA-based methods that make use of electrophoresis techniques, protein-based methods including immunological techniques are the most used for the detection of this type of fraud in the meat industry (Ghovvati, Nassiri, Mirhoseini, Moussavi, & Javadmanesh, 2009). In comparison to proteins that become denaturated upon heating, DNA is a more thermo stable molecule so that, despite of possible degradation during processing, short fragments are generally recoverable and can be used as the rationale of authenticity tests carried out with processed foods. However, DNA can also be extensively degraded during processing, due to which its detectable amount can be markedly reduced. Many other factors can also influence the accuracy of the results should composite foods be analysed, such as the competitive PCR (polymerase chain reaction) and PCR inhibition arising on the grounds of matrix effects (Primrose, Woolfe, & Rollinson, 2010). With PCR detection, both genomic and mitochondrial genes can been targeted. However, the use of

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mitochondrial DNA was shown to be better for processed meat, since the chances of its survival under different processing conditions are greater (Ghovvati, Nassiri, Mirhoseini, Moussavi, & Javadmanesh, 2009).

DNA-based methods are related to PCR and its numerous modifications, as well as to electrophoresis in agarose or polyacrylamide gel (Butorac et al, 2013). The principle of PCR is based on the exponential amplification of specific DNA fragments, generating an amount of DNA fragments sufficient for further analysis. Amplified DNA fragments are mostly analysed using electrophoretic techniques (Reid, O'Donnell, & Downey, 2006). Among other, these methods can include the detection of single nucleotide polymorphisms (SNPs) (Sazaki et al., 2004), the restriction fragment length polymorphisms (RFLPs) (Russell et al., 2000), random amplified polymorphic DNA (RAPD) (Martinez & Yman, 1998), a single-strand conformation pattern (SSCP) (Rehbein, Kress, & Schmidt, 1997), real-time PCR, species-specific PCR and multiplex PCR (Che Man, Aida, Raha, & Son, 2007; Ghovvati, Nassiri, Mirhoseini, Moussavi, & Javadmanesh, 2009; Dalmasso et al, 2004).

PCR represents a highly sensitive test, which allows for the detection of an animal species in a rapid and reliable manner. During the horse meat scandal, PCR techniques were used to screen for horse DNA in processed food samples. DNA was extracted from the meat portion of products such as lasagne, and tested using a real-time PCR. The products found to contain horse meat and deemed adulterated were then tested for the anti-inflammatory drug phenylbutazone, banned for the treatment of animals entering the human food chain since it may, although rarely, cause blood disorders. Griffiths et al. (2014) showed that for most laboratories testing the seafood authenticity, mitochondrial DNA gets into the testing focus.

DNA methods are the most specific and the most sensitive and should therefore be chosen whenever two closely related species or breeds are to be identified and mutually distinguished. This is especially important when it comes to the authentication of meat content in traditional and regional meat products, since during the production of these products animal breeds typical of the producing geographical area are employed (Sentandreu & Sentandreu, 2014). For example, Sazaki et al. (2004) used the SNP genotyping to identify the Holstein and the Japanese Black cattle breeds, and the RAPD to differentiate between horse breeds (Martinez & Yman, 1998).

The RFLP analysis is used to identify the change in genetic sequence that occurs at the site where the restriction enzyme cuts. RFLPs can be used to trace inheritance patterns, identify specific mutations, as well as for other molecular genetic assay purposes. Russell et al. (2000) used the RFPL for the identification of 10 different salmon-like species. Rehbein, Kress, &

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Schmidt (1997) used PCR for the amplification of the conserved region of cyt b. The SSCP enables electrophoretic separation of single-strand nucleic acids in which a single nucleotide change can considerably affect the strand electrophoretic mobility by altering the intra-strand base paring and its resulting conformation. Genetic methods can also be used to identify the presence of offal coming from species that reduce the quality of meat products using the methylation pattern of inactive genes. Genes not used by certain tissues are inactivated by methylation. Detection of a gene in an unmethylated state in a tissue that does not express this gene indicates a different tissue origin. To this end, organ-specific genes not expressed in the muscle tissue, for example phosphatidylcholine, which is liver-specific, or copper amine oxidase, which is lungs-, kidney-, heart- and spleen-specific, are identified (Popping, 2002).

Preventing adulteration of meat and meat products with less desirable meat species is an important issue. For example, a new type of meat fraud witnessed in China is the use of murine meat to substitute formutton meat (Fang & Zhang, 2016). The adulteration was identified using the TaqMan© real-time PCR. The results show the limit of detection (LOD) of less than 1 pg of DNA per reaction, and of 0.1 %-murine contamination in meat mixtures. Yalçınkaya, Yumbul, Mozioğlu, & Akgoz (2017) worked in order to select a DNA extraction method easy to perform, inexpensive, environmentally friendly and of the highest yield. As the existing methods of meat speciation are sometimes still heavy to perform, Masiri et al. (2017) developed a highly specific lateral flow immunoassay that can rapidly identify raw and cooked horse meat in xenogeneic meat sources down to 0.01 %- and 1.0 %- contamination, respectively.The analysis takes about 35 min and evokes no false-positive signals.

2.3.4 Other techniques

Protein-based methods represent techniques that use proteins as specific markers. They include electrophoretic methods, such as isoelectric focussing (IEF), chromatography, immunological techniques like Western-Blotting and enzyme-linked immunosorbent assay (ELISA), and proteomics (Montowska & Pospiech, 2007; Vallejo-Cordoba, Rodriguez-Ramirez, Gonzalez-Cordova, 2010; Chou et al., 2007; Ashoor & Osman, 1988). Immunological tests are often used because of their specificity, sensitivity, easy implementation, and low costs as compared to other analytical techniques used within this area. All these characteristics make them suitable for routine use in food control laboratories (Butorac et al, 2013). The performance of an assay relies on the ability of antibodies to specifically detect the target protein characteristic of a particular animal species, tissue or meat adulterant. If antibodies are not highly specific for a

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particular species or tissue, problems associated with cross-reactions can appear and give false-positive results. This is especially the issue in cases of closely related species (Sentandreu & Sentandreu, 2014). To overcome the problem arising with highly-processed meat products' analysis, in which aggressive production conditions can lead to protein denaturation, antibodies against thermostable proteins are produced, for example, osteocalcin, i.e. a tissue-specific protein of the extracellular bone matrix.

Among different immunological assays, ELISA is probably the most widely used for food authentication purposes (Liu, Chen, Dorsey, & Hsieh, 2006). There exists a variety of commercial test kits that detect and identify the species content in raw or thermally processed meat and meat products. For fish species identification, no ELISA test kit has been developed yet (Asensio, Gonzalez, Garcıa, & Martın, 2008), perhaps due to the great variety of fish species commercialized; however, polyclonal antibodies against the muscular protein of sardineshave been produced (Taylor & Jones, 1992). Macedo-Silva, Shimokomaki, Vaz, Yamamoto, & Tenuta-Filho (2001) determined using ELISA soy proteins in hamburgers prepared from beef, chicken and swine meat. Indeed, non-meat proteins, most commonly soybean ones, are added into some meat products due to their nutritional and functional properties. Soy protein has also been detected using the ELISA immunochemical diagnostic kits that enable a rapid detection and quantification of these proteins in foods. The ELISA diagnostic kit that allows for the determination of risk material (brain and spinal cord) in extracts from meat products or sausages is available. This is important since the tissues originating from the central nervous system are sometimes mixed with meat products and could transfer BSE to humans (Asensio, Gonzalez, Garcıa, & Martın, 2008).

To detect specific tissue types, proteomics can also be used. The basis of these methodologies is the use of high-resolution mass spectrometry that identifies unique proteins or peptides and determines food components. It is often used to identify animal species represented in a food product: to that end, it couples protein sequence information with the analytical power of mass spectrometry (MS) and can also be used to differentiate species and varieties based on their specific protein patterns. The achieved discrimination power is comparable to that of DNA-based methods, since the peptide sequences used as biomarkers are specific for a given animal species. In the study performed by Sentandreu, Fraser, Halket, Patel, & Bramley (2010), upon the hydrolysis of thermostable myofibrillar proteins, species-specific peptide biomarkers were identified. The method was able to detect the presence of 0.5% of chicken in a mixture with pork meat. The method allows for the quantification that makes use of isotope-labelled marker peptides as internal standards (Primrose, Woolfe, & Rollinson, 2010). Proteomics was developed to

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determine the species origin of gelatine and similar highly-processed hydrolysed proteins coming from animal skin and bones and added to foods. Most of the gelatines are prepared from a collagen material found in pigs, cows and fish carcasses; for many consumers, religious and ethical issues regarding the types of gelatine they consume may arise. Gelatines manufactured from the skin and bone material and the pertaining collagen proteins are incorporated into many foods, confectioneries, beverages and pharmaceuticals in form of thickeners, gelling agents, clarifying agents and 'mouthfeel' enhancers. The analytical method employs a proprietary database built up over a number of years that contains gelatine peptide mass data from mass spectrometry experiments encompassing scores of species and phylogeny data used to match unique species-specific peptides to a species or a tissue (skin or bone). The method can determine the species provenance of a wide range of gelatines including pig, cow, horse, fish or poultry. The method was recently employed so as to investigate the suspected addition of hydrolysed protein to chicken fillet samples intended to serve as a water-binding agent. The fillets labelled as 'chicken only' or as 'Halal-slaughtered' were shown to be adulterated with hydrolysed proteins derived from a cow material.

Another kind of analysis should be evoked, the analysis of stable isotopes in food of animal origin can reveal economically motivated adulteration, such as the addition of cheap sugar syrups in order to extend honey and maple syrup shelf life; it also enables the verification of the fact that chicken had been 'corn-fed', as well as the differentiation between organic and conventional farming practices (Kelly, 2003; Kelly & Bateman, 2009).

Finally, taking the case of butter, it contains a large amount of milk fat, which is often replaced by cheaper animal fats or vegetable oils. Tomaszewska-Gras (2016) examined the adulteration with palm oil using the Differential Scanning Calorimetry (DSC) technique. The results indicated that the technique is applicable for quantitative, peak area- and peak height-based detection of palm oil in butterfat if present in the concentration range of 2 to 35%.

3. Challenges and Perspectives

Food safety is an issue continuously investigated by researchers for authentication and in order to counteract new types of adulteration. In fact, literature reported in this review and our own research experience indicate that several methods using chromatography, spectroscopy and DNA-based techniques but also others have been developed for the authentication of food products of animal origin. Theses methods seem quite successful. However, they have mostly focused on research work and rare is the information about their implementation in official food

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control laboratories or food industry for routine analysis. The method to be implemented has first to be validated in order guarantee reliability. The next step is to consider several requirements as the availability of technology, instrumentation, and cost in addition to training to carry out the method.

Targeted approaches as chromatography and DNA-based techniques, that seem to be more used for the determination of specific ingredients or substitution of ingredients, are long and expensive. But, benefits like high accuracy, sensitivity and selectivity make them the methods of choice in official food control laboratories. They are very useful for identification and complete characterization of food samples. Inversely, non-target approaches as spectroscopic techniques that seem to be largely used to geographic authentication, may be less sensitive comparing to chromatography or DNA-based technique but their cost is much lower. They are rapid, non-destructive and can be miniaturized or handled which fit to the industry constraints. Research needs to be encouraged to develop handled and portable instruments. Besides, spectroscopic techniques are potential solution for food screening at large scale which permits more and systematic control ensuring food safety chain.

Another reflection which starts to emerge is data fusion to answer the question if there is any advantage of coupling different techniques to resolve one problem. This kind of strategy involves use of advanced chemometric modelling to extract the significant information.

4. Conclusion

In recent years, food products of animal origin with an added value are more and more often a subject to fraud. In order to prevent food fraud, product's specificities have to be defined in an evidence-based manner; otherwise, food fraud scandals could destroy consumers' trust in a specific label and subsequently cause economic losses in an already sensitive sector of small traditional food producers. A wide variety of scientific techniques have been developed to protect consumers and screen for adulteration in the food chain. Target and non-target approaches have been followed regrouping a series of analytical techniques mainly chromatography, spectroscopy and DNA-based techniques even if other techniques as isotopes measurements or ELISA are emerging. These sophisticated tools should continue to be applied and continuing research efforts should be made so as to address the newly-emerging food quality issues and to ensure brand and consumer protection. Methods have to be validated and scaled up to meet routine analyses requirements. One has to conclude that it is also urgent for the authorities to address the adequacy

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of labelling and improve and extend the scope of official monitoring methods, equally as for the food industry to establish an accurate control over its products.

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Table 1. Application of analytical techniques in authentication of food of animal origin (adopted from (Cubero-Leon, Peñalver, & Maquet, 2014)

Food of animal origin	Purpose of analysis	Analytical technique	References
Meat, fish and seafood	Sea bass rearing system	NIR	Trocino et al. (2012)
	Wild and cultured sea bass	NMR	Mannina et al. (2008)
	Sea bass farming system	NMR	Savorani et al. (2010)
	Pork adulteration in beef meatball	FTIR	Rohman, Sismindari, Erwanto, & Che Man (2011)
	Beef production systems	NMR	Osorio, Moloney, Brennan, & Monahan (2012)
	Geographical origin of beef	NMR	Jung et al. (2010)
	Detection of mechanically recovered meat	GC–MS	Surowiec, Fraser, Patel, Halket, & Bramley (2011)
	Identification of animal species	ELISA	Céspedes et al. (1999); Rencova, Svoboda, & Necidova (2000)
		PCR	Calvo, Orca, & Zaragoza (2002); Dooly, Sage, Clark, Brown, & Garrett (2005)
Milk and dairy products	Milk adulteration	FTIR	Nicolaou, Xu, & Goodacre (2010)
		NIR	Hsieh, Hung, & Kuo (2011);
	Milk and cheese geographic origin	NMR	Consonni & Cagliani (2008a); Sacco et al. (2009)
	Milk species	ELISA	Hurley, Coleman, Ireland, & Williams (2004)
Honey	Geographical origin	NMR	Donarski, Jones, & Charlton (2008); Consonni, & Cagliani (2008b); Donarski, Roberts, & Charlton (2010b)
		GC–GC–TOF-MS	Cajka, Hajslova, Pudil, & Riddellova (2009); Stanimirova et al. (2010)

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	NIR	Woodcock, Downey, & O'Donnell (2009)
	Raman	Pierna, Abbas, Dardenne, & Baeten, 2011
Botanical origin	NIR	Liang, Li, & Wu (2013)
	FTIR	Etzold, & Lichtenberg-Kraag (2008)
	GC–MS	Aliferis, Tarantilis, Harizanis, & Alissandrakis (2010); Castro-Vázquez, Díaz-Maroto, González-Viñas, & Pérez-Coello (2009)
	NMR	Beretta, Caneva, Regazzoni, Bakhtyari, & Maffei Facino (2008); Lolli, Bertelli, Plessi, Sabatini, & Restani (2008); Schievano, Peggion, & Mammi, 2010; Schievano, Stocchero, Morelato, Facchin, & Mammi (2012)
Adulteration	FTIR	Hennessy, Downey, & O'Donnell (2008); Gallardo-Velázquez, Osorio-Revilla, Zuñiga-de Loa, Rivera-Espinoza (2009)
	NIR	Zhu et al., 2010; Chen et al., 2011
	Raman	Li, Shan, Zhu, Zhang, & Ling (2012)

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Highlights

Analytical approaches to authentication of food of animal origin

Determination of specific food ingredients

Determination of geographical origin

Substitution of ingredients

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