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Does Traceability Lead to Food Authentication? A Systematic Review from A European Perspective

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ABSTRACT

Food safety and quality concern increasingly both consumers and the entire food sector involved. The European Union has implemented specific legislation for food products in order to monitor food safety and quality, protecting each country's culture, history and local economy at the same time. Moreover, Europe's heritage is being enriched, by establishing quality schemes such as Protected Designation of Origin (PDO) and Protected Geographical Indication (PGI). Europe's interest and concern regarding food safety proved also by Framework Program 5 Horizon 2020 continuous funding.Food traceability as well as food authentication constitute inseparable parts of food safety and quality. This systematic review presents innovative research technologies which are used in Europe for food traceability and authentication and published within the period 2010–2018. Moreover, information about the method used, the country of origin, the food type and the level of food process, are provided.This paper offers a new possible approach regarding the combination of the existing definitions of food traceability and authentication, as far as food provenance is concerned.

KEYWORDS

Food traceability; food authentication; geographical origin; food safety; food fraud

Introduction

Food traceability is defined as the ability to provide information about the history and origin of a product through a supply chain. This way could contribute to documentation of a specific production chain. Also, through a traceability system, actions such as a recall, if a product is found to be unsafe to consume or not according to quality or safety standards, are easily performed[1]. Food traceability can offer a new aspect for consumer's protection by being a basic part of food safety, food quality, and essential for food supply chain. However, a food traceability system is related to food authentication because it is based on the creditability of originality. International and national research have revealed several cases of food adulteration. In Europe, during the period 2016–2018, many reports related to adulteration, counterfeiting, and mislabeling have been published. Wine, spirits, olive oil, fish, meat, cheese, honey, herbs and spices represent the most commonly reported adulterated foods (*European Commission, 2019*).

For these reasons, standardization organizations for controlling the origin of foodstuff and the production process have appeared all over the world e.g., the French 'Institut National des Appellations d'Origine (INAO), Italy's 'Denominazione di Origine Controllata', Spain's 'Denominación de Origen', South Africa's 'Wine of Origin' or the United States' 'American Viticultural Areas'. Moreover, the European Union (EU) introduced legislation presenting specific names for agricultural products, foods and beverages of a quality or reputation (*Council Regulation, EEC No 2081/92*) such as PDO, PGI and TSG. PDO (Protected Designation of Origin) identifies food products that are produced, processed, and prepared in

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a defined geographical area. PGI (Protected Geographical Indication) identifies agricultural products and foods linked to a geographical area where at minimum one production step occurred and TSG (Traditional Specialties Guaranteed) identify and protect traditional methods of production [2]. These EU schemes are established to protect the reputation of the regional foods, to help producers obtain premium prices for authentic products and to minimize the unfair and misleading competition from fraudulent products (*1151/2012 EU Regulation*). Food authentication is main concern for severe organizations including the scientific community, law enforcement, food producers, importers, exporters, and consumers. This field is in the phase of exponential growth and is attracting a high level of attention from authorities and media around the world due to the production of counterfeit food and illegal food trade.

Food safety and quality authorities are asking for an extended and updated list of the analytical techniques for confirmation of food authentication and to support law enforcement. Therefore, there is a growing need for reliable analytical methods that can give a decisive answer about the authenticity of foodstuffs. Food authentication methods must be scientifically proven, accurate and reliable so that food products can be protected and clearly distinguished from any illegal substitutes. Several studies have attempted to summarize different aspects of evaluating food authenticity, such as country of origin, or reactions to specific food fraud cases and public attitudes towards food safety associated with traceability .[3]

We conducted this systematic review to analyze the correlation between food traceability and authentication, record the lab-based methods used for food authentication or/and traceability, the countries and food types involved and the level of the food process (raw or processed). As far as we know, there is no similar systematic review published.

Methods

Protocol and registration

In the present study a protocol based on PRISMA statement was performed, followed in all steps: literature search, study selection and analysis process (Fig. 1 and 2, 3).

Eligibility criteria

All study designs were included in the first step, irrespectively of the date of their publication. The literature search was conducted without language limitations, on the condition that an abstract in English existed reporting the information of interest. The inclusion and exclusion criteria were set as follows:

Inclusion criteria:

- Lab-based techniques
- European studies
- Food type

At least one of the following information is referred: *food authentication, food traceability, geographical origin*

Exclusion criteria:

- Feed traceability/authentication
- Drugs/Medicine type
- Non Lab-based techniques
- Non EU-based study

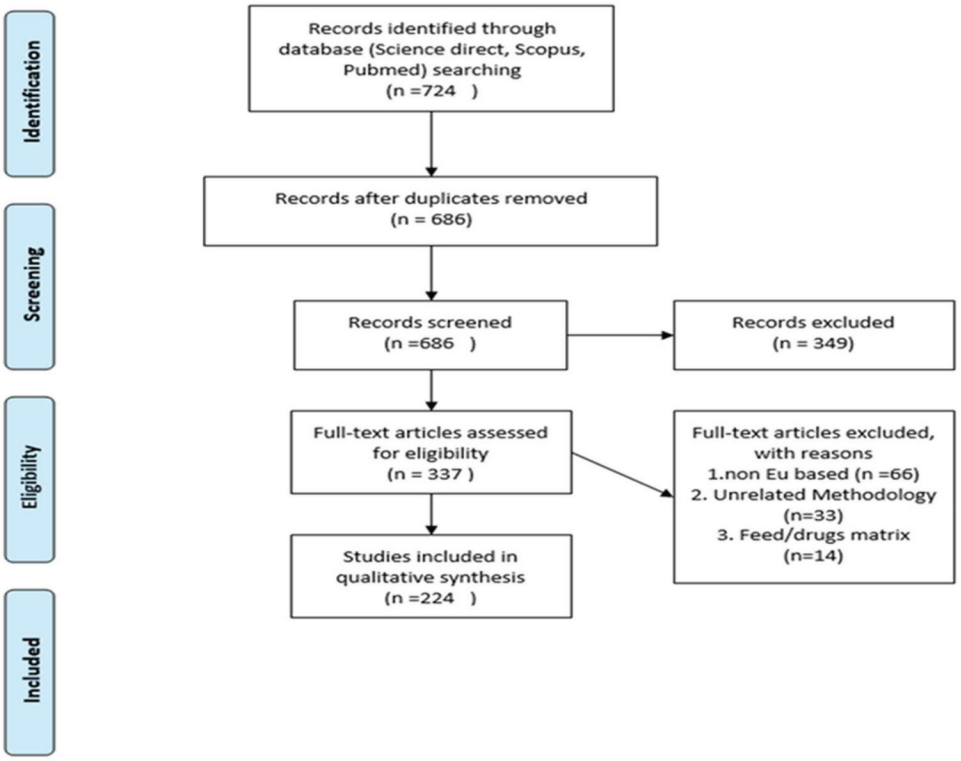

Figure 1. Prisma flow diagram.

Information sources and literature search

Scopus, Science direct and Pub Med were searched for empirical studies published over the past 8 years i.e. 2010 onwards: A comprehensive literature review was conducted using three online databases: Science Direct (Elsevier), Scopus (Elsevier) and PubMed (NCBI). We used the following search terms (adapted for each database): (Food) AND (traceability) AND (Food) AND (authentication) AND (geographical origin).

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Study selection

The selection of the studies to be included in the systematic review was evaluated by two reviewers independently: (M.D and A.V). Mendeley was used to identify duplicated publications and include each article only once. A first screening was performed by titles and abstracts, using the inclusion and exclusion criteria. The potentially relevant articles were passed on to the next step for further assessment. A second selection of the relevant studies was conducted by the full text of the included publications. The authors independently reviewed the potentially relevant studies according to the eligibility criteria to determine which studies would finally be included in the review.

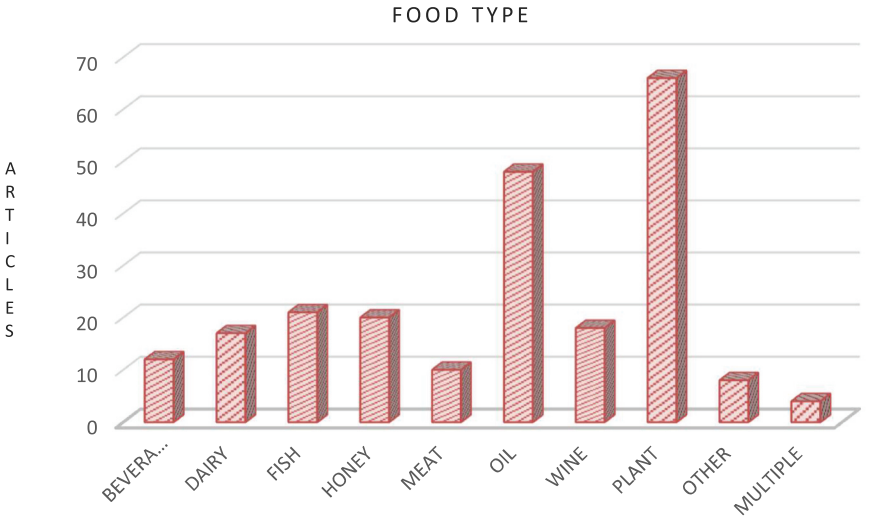

Figure 2. Number of publications according to the food type.

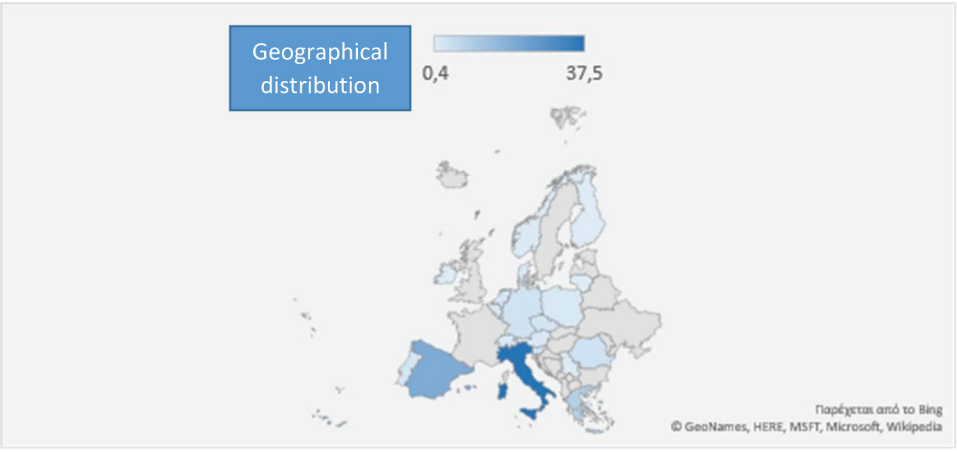

Figure 3. Origin countries of publications on food traceability and/or authentication methods.

Data collection process

Food traceability and/or authentication method, location of the study, food matrix and type and correlation with geographical origin were the information extracted from the full text articles selected to be included in the study.

Results

Study selection

Only original research papers published with at least a summary in English were included. Initially, a total of 724 articles were identified. Next the number of publications was reduced to 693 by removing duplicates. The next stage involved the elimination of articles in which titles and abstracts did not refer

to lab-based methods of traceability or authentication. Three Publications were excluded due to the lack of full access. After assessing the remaining papers (n = 337) relevant research papers in terms of traceability and authentication methods, were identified. From those 113 papers were discarded due to the fact that were non Europe study or the matrix was feed or drugs or the methodology was unrelated. Papers in the final set (n = 224)[4–227] mainly concerned food traceability and authentication lab-based methods in Europe. The systematic literature review approaches depicted in Fig. 1

Food type

Seafood, dairy, wines, beverages, plants, honey, meat and oils, were the most common food types used, in the included studies, in order to confirm the geographical origin or the identity of the product. The highest percentage, 29,4% (66 publications) presented techniques for identification of geographical or botanical origin of plants such as citrus, cherries, artichokes, garlic, tomato, wheat, rice etc . In addition, in this systematic review, 48 articles (21,4%) report methods for oils' authentication and traceability, considered that, olive oils' production has a great economic impact, resulting to a growing need for olive oil authentication. These specialty products such as plants or oils are sensitive to counterfeit admixture with cheaper varieties. Furthermore, the aim of 21 published articles (9,3%) was seafood authentication and traceability. Honey aldulteration has been recorded in 20 publications due to commercial and health aspects. Eighteen reports (8,9%) targeted wine geographical traceability or wine authenticity. The origin and identity of dairy products is studied in 17 publications, whereas beverages and spirits counterfiet insidents reported in 12 articles. It is also known that meat industry is falling behind of its' major competitors, in terms of traceability, authentication and quality assurance, fact that is indicated by a small number of publications concerning this matter. Indeed, only ten publications are included in this review, which analysed methods for distinguising meat species or for identifying meat geographical origin. Food types such as mushrooms, truffles, flours etc are analysed in 8 publication (Food type:Other). Last, but not least, 4 studies handled multiple food types.

Origin countries

Italy, during the period 2010–2018, emerged in the field of food traceability and authentication. This period, Italy was the first country in publications, with 37, 5% from the total included studies, potentially due to the high number the high number of PDO, PGI and TSG Italian food specialties, which are quality labeled and protected by the European Union. The fact that PDO, PGI and TSG products often command higher prices and have a worldwide economic impact, makes them subject to adulteration and attractive to fraudsters. Moreover, the global trade, the new distribution channels and the economic pressure, contribute to food fraud. Mediterranean European countries such as Spain and Greece, have also contributed with a large number of articles. Spain had 45 publications about food traceability and authentication, while Greece had 20. France, England, Germany and Romania were countries with approximately 10 publications each.

Level of food process

The reports described in this systematic review used either raw or processed material, while some of studies decided to use both, in order to explore whether the food process is a contributing factor. For instance, in the plants/crops' category, 42 articles are based on raw material, 14 articles on processed and 10 articles are based on both. Oils' traceability refers mostly to processed food, whereas 4 reports are based on raw food and 8 reports are based on both. Only a small number of studies focus on raw material, as a means for meat or dairy products' authentication. In the case of seafood, raw material and food of both processing levels are used in 9 articles respectively. Furthermore, in order to achieve honey traceability, authors in 9 publications used a processed source, in 8 publications raw material

and in 5 articles both. Wine or beverages' authentication and traceability generally appear to be based on raw material.

According to Fig. 4, almost half of the included studies (44, 6%) used processed material in order to conduct their research. This high percentage is explained by the implemented food categories such as wine, oils and beverages, which are only consumed after their process. Furthermore, 35, 3% of the articles aim to traceability and authentication through raw material, especially as far as crops are concerned. As far as studies concerning both level of processing, they represented the 18, 3% of the total number. These studies probably used different levels of food process in order to evaluate their impact on food traceability and authentication results. The remaining 1, 8% refer to publications which used many food sources of multiple levels of process.

Traceability/authentication methods

Numerous publications have focused on the food traceability and authentication methods. This is due to the increased interest and concern of producers, traders, consumers, quality and safety authorities on the definition of the geographical origin of products. Nuclear magnetic resonances (NMR), Infrared spectroscopy (IR), isotopic techniques, DNA-based methods, Mass spectrometry (MS), chromatographic (GC-LC) prove to be very interesting in the study of food traceability and authentication. A variety of combinations of these techniques can also be used according to included studies. Fig. 5 demonstrates the number of publications in relation with the method used. Typical examples of the above mentioned methodologies are hereby provided. Table 1 records the methods used for every different food type. It has to be mentioned that several publications include more than one food type.

Nuclear Magnetic Resonance (NMR)

Nuclear Magnetic Resonance (NMR) methodologies are already used for analysis of target food compounds, since they provide information about chemical and molecular food composition.[228] From the included studies, 11, 1% used NMR technologies for food traceability and authentication. An important necessity to obtain high resolution NMR spectra is an accurate molecular mobility. All liquid foods and beverages can be analyzed without any treatment because of the zero viscosity of

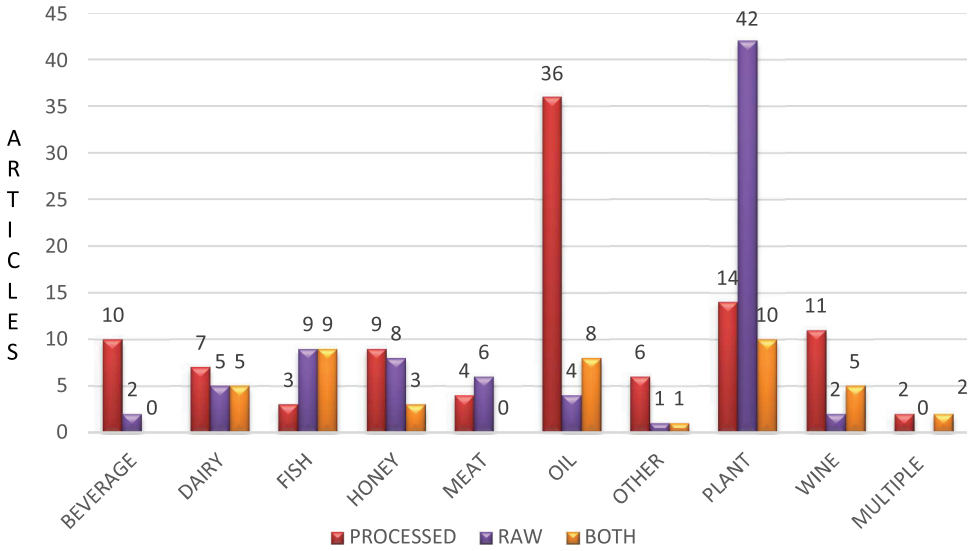

Figure 4. Articles according to the food matrix used in methodology.

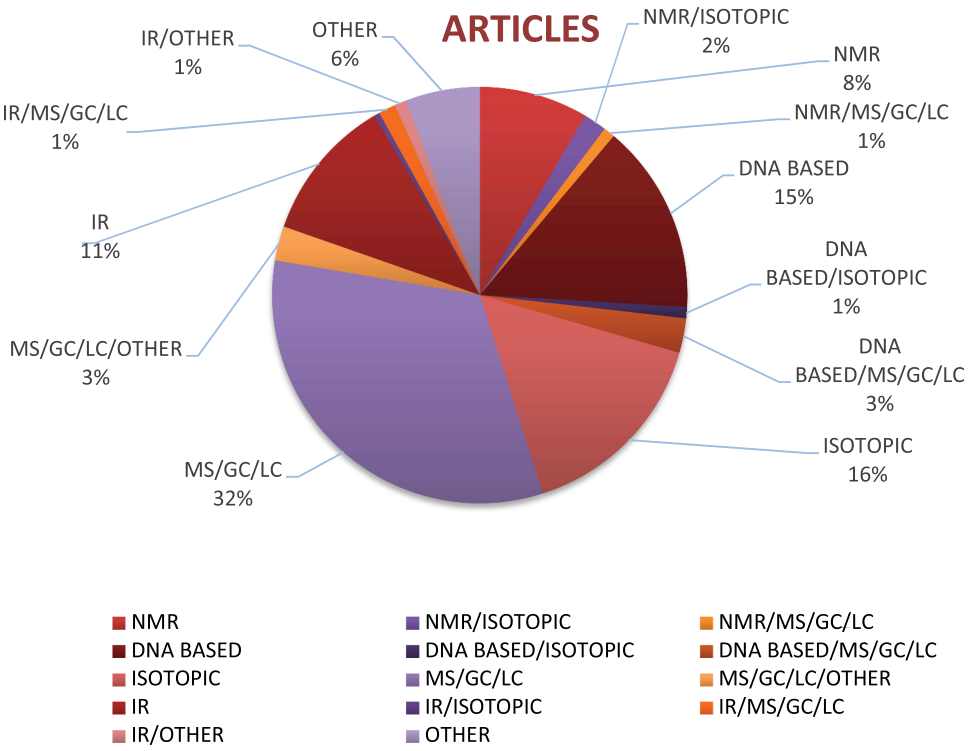

Figure 5. Number of publications according to methodology.

Table 1. Method used according to food type.

FOOD TYPE	NMR	IR	DNA BASED	ISOTOPIC	MS/GC/LC	OTHER
BEVERAGE	4	1	–	2	5	3
DAIRY	2	5	3	4	5	–
FISH	2	–	15	2	3	1
HONEY	2	4	2	–	12	2
MEAT	1	1	3	3	3	–
OIL	7	8	5	9	21	8
WINE	1	2	1	6	9	1
PLANT	6	12	13	14	30	6
OTHER	–	–	1	2	4	2
TOTAL	**25**	**33**	**43**	**42**	**92**	**23**
%	***9,68%***	***12,79%***	***16,66%***	***16,27%***	***35,60%***	***8,91%***

fluids.[229] Depending on the type of food sample and the NMR analytical method, different sample treatment protocols can be applied.[229] NMR-based metabolomics studies have also been applied to investigate the origin of several food products. NMR based metabolomics protocol implemented, for example, in untreated Greek grape marc distillates, accomplished their classification and revealed potent metabolic markers[220].[1]H NMR approaches were used for fingerprinting of the unsaponifiable fraction of virgin olive oil and for classification according to their geographical origin[90].[1] H NMR metabolomics has been referred as a tool for analyzing boreal varietal honeys and botanical origin of Finnish honeys[221]. Clear discrimination of different botanical origin of a product can also be achieved by NMR metabolomics.[13]C NMR phospholipid profiles have been analyzed for discrimination among lean gadoid fish species and stocks [222]. Characterization of metabolic profile of the "Tonda Gentile Trilobata " Italian hazelnuts, both raw and roasted by NMR approaches, as well as

differentiation between cultivars of other geographical origins, were also reported [20]. There were also studies which demonstrated NMR-based lipid fingerprints for food traceability. Thus, NMR technologies seem to be a valid method in assessing botanical and geographical origin of a product.

Infrared spectroscopy (IR)

Spectroscopic technologies have been applied worldwide as an express method for routine analysis of a variety of food products, such as dairy, meat, wines and grains and in this systematic review represent the 13.8%. The most common techniques for food analysis are IR spectroscopy with sub-division in NIR (near-infrared) and MIR (mid-infrared). NIR lies in region between 780 nm < ʎ < 2,500 nm (12,820–4,000 cm^{-1}) and MIR in region 3000 nm < ʎ < 50,000 nm.[230, 231] NIR is a rapid and reliable method for food authentication, including the determination of its' geographical origin. It has already been used as a reliable tool for real-time authentication of Asiago d'Allevo cheese, via the estimation of several chemical properties and also the classification of the dataset according to different farm management, ripening age, production period and production height[223]. It has also been reported, that NIR spectra provide information for authenticate genuine PGI-Galician honey samples[226]. NIR spectroscopy coupled with chemometrics classification methods were demonstrated for the authentication of the geographical origin of PDO pistachio samples from Bronte (Italy) [224]. Analysis of FT-NIR (Fourier Transform-Near Infrared Spectroscopy) fingerprints can provide also total species and even subspecies' identification. Analysis of a food sample using the MIR spectrum (4000–400 cm_1) or ATR-FTIR (Attenuated Total Reflection Fourier-Transform Infrared spectroscopy) spectra reveals information about the authentication of food origin. MIR spectra of 250 samples of sheep milk were studied in order to discriminate the geographical area where the sheep milk samples had been collected [45].Overall, IR spectroscopy is a powerful, fast and cheap method to trace food products and detect food fraud.

Mass Spectrometry – Gas Chromatography-Liquid chromatography (MS-GC-LC)

Mass profile of a food sample can be regarded as an analytical signature of the food product and thus can lead to an effective discrimination among food types. 40, 1% from studies included here, based on MS/GC/LC methodologies to access food traceability and authentication. For example, by monitoring the volatile compounds by SIFT-MS (Selected Ion Flow Tube-Mass Spectrometry) technique, discrimination and classification of a variety of food products can be achieved. A study which analyzed the chemical profiling (i.e. free acidity, peroxide value, spectrophotometric indices, fatty acid-, tocopherols- and sterol-composition) of extra virgin olive oils and applied three SIFT-MS spectra ($H3O^+$, NO^+ and $O2^+$ reagent ions) for different geographical origin determination is reported[96]. Prediction of virgin olives oils provenance was verified by analysis of MS-spectra of the volatile fractions[98]. Moreover, chemometric processing of mass spectrometry seem to be an easy, quick and untargeted approach for differentiation between authentic and adulterant food samples. Analytical strategies for food authentication based on non-targeted fingerprinting approach utilizing gas chromatography coupled to tandem high-resolution mass spectrometry (Q-TOF mass analyzer) were also reported during 2010–2018. Mass spectrometric techniques that used fusion approaches, represent a robust way for food authentication were presented too. Chromatographic techniques such as GC, LC, HPLC, and HTGC were also reported in order to establish models for predicting the provenance of food products. Chromatographic methods can create unique chemical food fingerprints that enable differentiation between products. Aim of a research article included in this systematic review was to investigate the applicability of metabolic fingerprinting using ultra high performance liquid chromatography coupled to high resolution tandem mass spectrometry (UHPLC–HRMS) in order to authenticate saffron (*Crocus sativus)* according to their geographical origin and/or harvest years[186]. Food traceability and authentication by chromatographic technologies are widely applied and implemented upon the identification of minimal analytical differences between patterns or the identification of unique marker compounds[2].

DNA based

New DNA-based technologies which allow species identification with reliability and sensitivity are gaining attention in the food traceability and authenticity field and consist of the 18,3% from the included studies. For example, PCR-Restriction Fragment Length Polymorphism (PCR-RFLP) and COI barcoding are used for confirming mislabeling fraud in raw and processed foods [10]. These methods have been already used for mislabeling fraud in processed anchovy products and for discrimination *E.enrasicolus* among species. In addition, DNA markers, such as SSR, enable genetic characterization and distinction between species [88]. Genetic traceability in the case of Shambucana sheep was proven efficient for certifying the origin and discriminate from other breeds[100]. Next Generation Sequencing (NGS) technologies are also a new reliable DNA-based method for species identification and food authentication. Real time-PCR, q-PCR, PCR-Denaturing Gel Gradient Electrophoresis (PCR-DGGE), High resolution melting (HRM), PCR-cloning, are some of the most popular and reliable molecular techniques for this purpose. Verification of Asian surimi traceability, using DNA-mini barcoding coupled with PCR-cloning for species authentication has been assessed[218]. Discrimination of organic from conventional fruits by molecular microbial approaches,such as PCR-DGGE, was also reported[71]. All these DNA-based techniques could be reference methods for species identification, for detecting accidental contamination and counterfeits or even verifying products geographical origin.

Isotopic

The stable isotopes ratios (SIR) of bio elements, which depend on botanical, geographical, agronomic and climatic factors, have been widely suggested for food authenticity and origin assessment. Severe isotopic analysis methods, according to our work (18, 75%) have been applied for both geographical traceability and authentication. Stable isotopes (C and O) can be compared between different geographical areas to achieve total discrimination of products. Stable isotope ratios of bio-elements [,2/,1,, 18/,16;,13/,12,,15/14N] combined with elemental profiles are also applied for determination of different food products. Use of δ13 C of glycerol in balsamic vinegars of Modena has presented as a suitable method for a deeper knowledge of this food source and for vinegars authentication [204]. Furthermore, Sr isotopes are already used to access the geographical origin of a food product.[87]Sr/[86]Sr isotopes of bioavailable fractions, which are related to the geological substratum of the vineyard, were demonstrated as a useful analytical tool to check the geographical provenance of fresh grape fruits[7]. Combination of[87]Sr/[86]Sr isotopes and light stable elements (C, N, O, S) with multi-elemental profiling for the authentication of geographical origin of European cereal samples was already conducted[150].

Methods linked with geographical origin

Table 2 depicts whether there is a correlation between food authentication and geographical origin according to the methodologies presented in this review. Among the 224 articles reviewed in total, 130 of them linked their methodology with the food production's geographical area. Studies examined the relationship between traceability and authentication. In 47 publications, authors used Mass spectrometry or Liquid/Gas chromatography and achieved the determination of the food products' geographical origin. Over the 35 articles concerning isotopic techniques, 26 have also resulted to proof about the geographical area. Less than half of the articles which used DNA-based traceability methods, associated the food product with its' geographical origin. On the other hand, more than half of the articles based on NMR or NIR/FTIR methods, linked food traceability to food provenance.

Discussion

Most of the approaches for food traceability and/or authentication presented in this review are very promising in fulfilling this role. The findings of this systematic review confirm the fact that methods

Table 2. Correlation of each methodology with geographical origin and authentication.

METHOD	GEOGRAPHICAL ORIGIN	AUTHENTICATION
NMR	14 (6,25%)	11 (4,91%)
DNA BASED	14 (6,25%)	27 (12,05%)
IR	19 (8,48%)	12 (5,35%)
ISOTOPIC	26 (11,6%)	9(4,01%)
MS/GC/LC	47 (20,9%)	32 (14,28%)
OTHER	10 (4,46%)	3 (1,33%)
TOTAL ARTICLES	***130 (58,03%)***	***94 (41,96%)***

used for food traceability could also provide information related to food authentication. From 224 articles searched, 130 of them concluded that food traceability is high related with food authentication and that an interdependent correlation between these terms exists.

Most of the publications included, target plant or oil traceability and/or authentication techniques. The level of food type process (raw, processed or both) used to conduct these methods is also presented. Italy seems to be the leader in food traceability/authentication publications with 84 articles out 224 articles included. The reason for this could be explained by the fact that Italy has numerous products with EU schemes labels such as PDO Lambrusco wines, extra virgin olive oils from Sabina PDO, PDO lard, PDO Pecorino Siciliano cheese etc.

According to this systematic review, the analytical techniques most commonly used to food traceability and/or authentication are MS/GC/LC based techniques. Mass Spectrometry can offer some important advantages, such as sensitivity, or the possibility to establish more robust couplings with separation techniques. Because of this method's high sensitivity, some compounds at low concentrations in a sample can be easily evaluated .[232] So far, these techniques are applied for food traceability and/or authentication by target and non-target approaches. The highest percentage of the publications involved, have conducted non-target approaches for food traceability and/or authentication. More specific, by the total metabolite screening of a sample, end up with a specific characteristic compound each product's [13] [40] [186]. Metabolite fingerprinting coupled with appropriate statistical data process, provide a mean of comparing the profiles of a group of samples, for determination, identification and discrimination quali- and quantitative metabolite differences. On the other hand, metabolite profiling is a targeted method, in which samples are compared on the basis of the quali-quantitative distribution of a selected number of known metabolites (markers) [164]. For non-target approaches is essential, result's validation by an appropriate model, but it is often found to be used inefficiently or unreliable. So, when the identification of metabolites cannot be determined, several chemometric tools can be employed. Although, target compound approaches are also referred such, as Arabica coffee's authenticity by 16-O-Methylcafestol [5] or cereal's authenticity by specific peptide markers [80].

In detail, in order to analyze metabolomic data of a food sample, several steps need to be executed such as peak detection, integration, and data alignment before multivariate statistical analysis. Principal components analysis (PCA) and Partial least square (PLS) are the most-used multivariate statistical techniques. PCA intends to group the data from the food samples among new variables called principal components in order to find correlations among the different samples and by PLS, samples differentiation achieved by reduction of dimensionality while maximizing correlation between variables.

In conclusion, non-targeted approaches are much more requiring compared to classical targeted approaches. In targeted analysis, results are usually estimated compound-by-compound using univariate statistics. By contrast, the data extracted for non-targeted approaches typically needs to be evaluated using multivariate statistical models .[233, 234, 235] Thus, it is essential need a target compound analysis for every food product to be found and validated.

Conclusions

During the period 2010–2018, the number of publication research articles which have focused on food traceability and/or authentication has been increased due to the fact that consumers are looking for high quality products. Consumers also tend to prefer food products with PDO, PGI or TSG status. As a consequence, there is a growing need to establish innovative and reliable techniques for food authentication and traceability.

In conclusion, apart from confirming the geographical origin of the product, these methods appear to represent a new pioneer tool for consumers and importers/exporters, who can be protected from food fraud and be reassured of the products' authenticity and geographical origin. Mass spectrometry, Gas and Liquid Chromatography technologies were explored in this systematic review and it was confirmed that these methods were able not only to lead to food traceability, but also to food authentication.

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