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Analytical Methods

An overview of analytical methods for determining the geographical origin of food products

Dion M.A.M. Luykx, Saskia M. van Ruth *

Department of Authenticity and Identity, RIKILT – Institute of Food Safety, Wageningen UR, P.O. Box 230, 6700 AE Wageningen, The Netherlands

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Abstract

There is an increasing interest by consumers for high quality food products with a clear geographical origin. These products are encouraged and suitable analytical techniques are needed for the quality control. This overview concerns an investigation of the current analytical techniques that are being used for the determination of the geographical origin of food products. The analytical approaches have been subdivided into four groups; mass spectrometry techniques, spectroscopic techniques, separation techniques, and other techniques. The principles of the techniques together with their advantages and drawbacks, and reported applications concerning geographical authenticity are discussed. A combination of methods analysing different types of food compounds seems to be the most promising approach to establish the geographical origin. Chemometric analysis of the data provided by the analytical instruments is needed for such a multifactorial approach.
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Keywords: Analytical methods; Food products; Geographical origin; Mass spectrometry; Separation techniques; Spectroscopy

Contents

1. Introduction . . . 898
2. Mass spectrometry techniques . . . 899
   2.1. Isotope ratio mass spectrometry (IRMS) . . . 899
   2.2. Inductively coupled plasma mass spectrometry (ICP-MS) . . . 900
   2.3. Proton transfer reaction mass spectrometry (PTR-MS) . . . 900
   2.4. Gas chromatography mass spectrometry (GC-MS) . . . 901
3. Spectroscopic techniques . . . 901
   3.1. Nuclear magnetic resonance spectroscopy (NMR) . . . 901
   3.2. Infrared spectroscopy (IR) . . . 902
   3.3. Fluorescence spectroscopy . . . 902
   3.4. Atomic spectroscopy . . . 903
4. Separation techniques . . . 904
   4.1. High performance liquid chromatography (HPLC) . . . 904
   4.2. Gas chromatography (GC) . . . 904
   4.3. Capillary electrophoresis (CE) . . . 905
5. Other techniques . . . 905
   5.1. Sensor technology . . . 905

* Corresponding author. Tel.: +31 317 475414; fax: +31 317 417717.
E-mail address: saskia.vanruth@wur.nl (S.M. van Ruth).

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5.2. DNA technology 906
5.3. Sensory analysis 906
6. Conclusions 907
References 907

1. Introduction

By nature, food products have a land-based, and therefore geographical origin. Historically, food consumption habits were shaped by socio-cultural factors and available local natural resources (Delamont, 1995). Such links between food and territory have disappeared over time by various means. These include changes in food production and transportation technologies, urbanisation, and consumer exposure to non-local experiences through travel and the media (Montanari, 1994). This has led to a similarity of lifestyles and habits across regions such that food consumption patterns within a region no longer necessarily reflect the production patterns of that region (Ritzer, 1996). However, the last ten years consumers have a renewed interest in foods strongly identified with a place of origin. A public enquiry by the UK Food Standards Agency revealed that labelling of food products with their country of origin is high on the consumers list of demands for change (Food Standard Agency, 2001). A Swiss evaluation pointed out that the origin of food is important for the purchase decision of 82% of the customers, with the origin of meat being a very relevant criterion for 71% (Franke, Gremaud, Hadorn, & Kreuzer, 2005). The reasons for this increasing interest by consumers vary from (1) specific culinary or organoleptic qualities associated with regional products, (2) health, (3) patriotism, (4) media attention, (5) decreased confidence in the quality and safety of products produced outside their local region, country or the EU or (6) concern about animal welfare and environmentally friendly production methods more often adopted by smaller regional producers (Gilg & Battershill, 1998; Ilbery & Kneafsey, 1998; Kelly, 2003). As a result fair trade, organic and slow food products have received more attention and accompanying organisations (e.g., fair trade federation, slow food organisation) were founded.

The EU has recognised and supported the potential of differentiating quality products on a regional basis (Dimara & Skuras, 2003). In 1992, EU regulation 2081/92 and 2082/92 introduced an integrated framework for the protection of geographical indications and designations of origin for agricultural products and foodstuffs. The regulation also seeks to achieve wider social and environmental objectives with respect to the rural economy. Furthermore, laws enforce labelling of the geographical origin of agricultural products in many countries due to consumer demands for more information on foods. Recently, EU regulation 2081/92 and 2082/92 have been replaced by regulation 510/2006 and 509/2006, respectively, and EU regulation 1898/2006 was added. The EU regulation allows the application of the following geographical indications to a food product: protected designation of origin (PDO), protected geographical indication (PGI) and traditional specialities guaranteed (TSG) (Mout, 2004). PDO is the term used to describe foodstuffs that are produced, processed and prepared in a given geographical area using recognised methods. Examples are: Olives noires de Nyons (table olive, France), Roquefort (cheese, France), Noord-Hollandse Gouda (cheese, The Netherlands) and Opperdoezer Ronde (potato, The Netherlands). For the PGI, the geographical link must occur in at least one of the stages of production, processing or preparation. Examples are: Borrega da Beira (fresh meat, Portugal), Coppia Ferrarese (bread, Italy) and Westlandse Druif (grape, The Netherlands). A TSG designation does not refer to the origin but highlights a traditional character, either in composition or means of production. Examples are: Mozzarella (cheese, Italy), Traditional Farmfresh Turkey (UK) and Panellets (Spain). These indications are particularly important in Europe, where there has been a long tradition of associating certain food products with particular regions. This is also demonstrated by the existence of a European Wine Databank on authentic European wines (as foreseen in EC Regulation No. 2729/2000) requiring isotopic and nuclear magnetic resonance determinations.

The use of geographical indications allows producers to obtain market recognition and often a premium price. False use of geographical indications by unauthorised parties is detrimental to consumers and legitimate producers. From this point of view, the development of new and increasingly sophisticated techniques for determining the geographical origin of agricultural products is highly desirable for consumers, agricultural farmers, retailers and administrative authorities. It is an analytically challenging problem that is currently the focus of much attention within Europe and the USA. Reports on analytical methods for determining the geographical origin of agricultural products have been increasing since the 1980s. The initial focus was on processed agricultural products such as wine, honey, teas, olive oil, and orange juice, while later studies examined fresh products such as potatoes, Welsh onions, pistachios, and garlic, chiefly because world-wide trade in fresh agricultural products has increased year by year and the law now enforces labelling of their geographical origin. Various techniques have been studied based on organic constituents, mineral contents or composition, light- or heavy-element isotope ratios, or combinations thereof. Chemometric analysis of the data provided by analytical instruments which have the ability to determine more than one component at a time in a sample can be a

Table 1
Overview of analytical techniques which can be used for determination of the geographical origin of food products, abbreviations in brackets

Principle	Main technique	Specific forms of the technique
Mass spectrometry	Isotope ratio mass spectrometry (IRMS)	Continuous flow IRMS (CF-IRMS)
	Inductively coupled plasma mass spectrometry (ICP-MS)	Dual inlet IRMS (DI-IRMS)
	Proton transfer reaction mass spectrometry (PTR-MS)	
	Gas chromatography mass spectrometry (GC-MS)	
Spectroscopy	Nuclear magnetic resonance spectroscopy (NMR)	Low resolution NMR
		High resolution NMR (e.g., site specific natural isotope fractionation (SNIF))
	Infrared spectroscopy (IR)	Fourier transform IR (FTIR)
		Mid-infrared IR (MIR)
		Near-infrared IR (NIR)
	Fluorescence spectroscopy	Front-face fluorescence spectroscopy
	Atomic spectroscopy	Atomic absorption spectroscopy (AAS)
		Atomic emission spectroscopy (AES)
Separation	High performance liquid chromatography (HPLC)	
	Gas chromatography (GC)	
	Capillary electrophoresis (CE)	
Others	Sensor technology	'Electronic nose'
	DNA technology	Polymerase chain reaction (PCR)
	Sensory analysis	

support to establish links to the food origin. If the components have sufficient discriminatory power, the set of their concentrations will form a characteristic pattern or 'fingerprint' relating to the geographical origin of the sample. Chemometrics provides the ability to detect these patterns, and is essentially helpful when the number of components necessary to differentiate samples from different geographical origins increases. In this review analytical approaches that have been developed so far for determining the geographical origin in combination with or without chemometrics are outlined and evaluated below. For this overview the analytical approaches have been subdivided into four groups; mass spectrometry techniques, spectroscopic techniques, separation techniques, and other techniques. All techniques and abbreviations are summarised in Table 1.

2. Mass spectrometry techniques

Mass spectrometry (MS) is a powerful analytical technique for measuring the mass-to-charge ratio of ions (Aebersold & Mann, 2003). In general, MS is applied to elucidate the composition of a sample by generating a mass spectrum representing the masses of the sample components. This is achieved by first ionising the sample, separating the ions of differing masses and recording their relative abundance by measuring intensities of the ion flux. MS is often combined with other techniques as will be described below.

2.1. Isotope ratio mass spectrometry (IRMS)

IRMS is a technique that can distinguish chemically identical compounds based on their isotope content (Brenna, Corso, Tobias, & Caimi, 1997). The ratio of the stable isotopes of the elements that constitute almost all biological material, $^{13}C/^{12}C$, $^{15}N/^{14}N$, $^{18}O/^{16}O$ and $^{2}H/^{1}H$ can be determined. In addition other elements like $^{34}S/^{32}S$ can be included to improve the discriminative power of the technique. In general, the isotopic composition of the constituents of agricultural products (proteins, carbohydrates, fats, minerals) depends on various factors. Some of those factors can be expected to be indicative for the geographical origin, others are more related to the production factors. These factors include the use of fertilisers, certain feedings stuffs in the diet of farm animals, seasonal variations and geological factors (e.g., soil composition, altitude, etc). Especially, these latter factors that affect the stable isotope ratio can be used in assigning the regional origin of agricultural products. For example, the $^{18}O/^{16}O$ ratio is highly dependent on the distance to the ocean and on the altitude above sea level of the production site. IRMS data are generally collected for several elements and interpreted using chemometric methods.

The two most common types of IRMS instruments are continuous flow (CF-IRMS) and dual inlet (DI-IRMS) (Benson, Lennard, Maynard, & Roux, 2006). With DI-IRMS, the samples for analysis are prepared (i.e. converted into simple gases) off-line. The off-line sample preparation procedure utilises a specially designed apparatus involving vacuum lines, compression pumps, concentrators, reaction furnaces, and micro-distillation equipment. This technique is time-consuming, usually requires larger samples, and contamination and isotopic fractionation can occur at each of the steps. The CF-IRMS sample introduction technique consists of a helium carrier gas that carries the analyte gas into the ion source of the IRMS. This technique is used to connect an IRMS to a range of automated sample preparation

devices. While dual inlet is generally the most precise method for stable isotope ratio measurements, continuous flow mass spectrometry offers on-line sample preparation, smaller sample size, faster and simplified analysis, increased cost effectiveness, and the possibility of interfacing with other preparation techniques, including elemental analysis, gas chromatography (GC), and more recently, liquid chromatography (LC).

In practice, IRMS combined with or without other techniques (e.g., elemental analysis, NMR, GC) and/or chemometric methods has been applied to determine the geographical origin of a variety of food products. These products range from dairy and animal products (Brescia et al., 2003; Pillonel et al., 2003; Renou et al., 2004; Rossmann et al., 2000; Schmidt et al., 2005) to vegetables (Georgi, Voerkelius, Rossmann, Grassmann, & Schnitzler, 2005), natural flavours, honey, wines (Martinelli et al., 2003; Ogrinc, Košir, Kocjančič, & Kidrič, 2001; Rossmann, 2001), coffee (Serra et al., 2005) and fruit (juices) (Perez, Smith, & Anderson, 2006; Rossmann, 2001). For example, stable isotope ratio determinations of the light elements (C, N, O, S; bio-elements) and the heavy-element, Sr, have been used for regional origin assignment of butter (Rossmann et al., 2000). Furthermore, for the determination of the geographical origin of wine, a combination of isotopic ratios of carbon (ethanol) and oxygen (wine water) have been used (Rossmann, 2001).

The instrumental progress in the application of multi-element stable isotope analyses will remarkably extend the scope of this technique for assignment of the regional origin of foods. A prerequisite is the availability of databases and the establishment of increased basic knowledge on stable isotope fractionation effects in nature for all elements in question. Although the financial cost of purchasing and operating high specification MS instruments is quite high and sometimes sample preparation before analysis is time-consuming (Franke et al., 2005), IRMS is applicable for the determination of the geographical origin of numerous food products.

2.2. Inductively coupled plasma mass spectrometry (ICP-MS)

ICP-MS is a powerful tool for the quantitative determination of a range of metals and non-metals (inorganic elements) in a wide variety of samples at trace (ppb–ppm) and ultra-trace (ppq–ppb) concentration levels (Lachas, Richaud, Herod, Dugwell, & Kandiyoti, 2000; Vanhoe, 1993). With ICP-MS, samples are decomposed to neutral elements in a high-temperature argon plasma and analysed based on their mass to charge ratios. ICP-MS encompasses four main processes, including sample introduction and aerosol generation, ionisation by an argon plasma source, mass discrimination, and the detection system. Even though it can broadly determine the same elements as atomic spectroscopic techniques (see also Section 3.4), ICP-MS has clear advantages in its multi-element characteristics, speed of analysis, detection limits, and isotopic capabilities. Next to this, unlike the atomic emission spectrometer, ICP-MS spectrometers can accept solid as well as liquid samples.

ICP-MS can screen the geographical origin of food products by the analysis of numerous inorganic elements and obtaining fingerprints of the element pattern (Ariyama & Yasui, 2006). In this way the technique has been successfully applied to vegetables (Ariyama et al., 2007; Ariyama, Nishida, Noda, Kadokura, & Yasui, 2006; Ariyama & Yasui, 2006), nuts (Gómez-Ariza, Arias-Borrego, & García-Barrera, 2006), tea (Moreda-Piñeiro, Fisher, & Hill, 2003) and wines (Coetzee et al., 2005). For example, the geographical origin of onions was determined by analysing twenty-six elements (Ariyama et al., 2006). Although the fertilisation conditions, crop year, onion variety, soil type, and production year caused concentration variations of several elements, these variations were smaller than the differences observed between production places if appropriate elements were selected. In these studies, chemometric techniques were used for statistical analyses of the obtained ICP-MS data (Ariyama, Horita, & Yasui, 2004). In a similar way wines were classified according to their geographical origin (Coetzee et al., 2005). Here, the method was based on the assumption that the provenance soil is an important contributor to the trace element composition of a wine. The combination of elements characterising wines from a particular region appeared to be different in each region. There are only a few studies determining the geographical origin of (processed) agricultural products by a composition of inorganic element ratios excluding isotope ratios. ICP-MS is often combined with IRMS, but also atomic absorption/emission spectrometry (see Section 3.4) for simultaneous elemental analysis, and LC (Gómez-Ariza et al., 2006). Despite the increasing interest in elemental speciation, most of the plasma-based techniques are nowadays used for total metal detection and the full capabilities of ICP techniques have yet to be realised (Meija, Mounicou, & Caruso, 2004).

2.3. Proton transfer reaction mass spectrometry (PTR-MS)

PTR-MS allows quantitative on-line monitoring of volatile organic compounds (VOC) (Hansel & Märk, 2004; Lindinger, Hansel, & Jordan, 1998). The fundamental difference between a conventional MS and PTR-MS is the soft ionisation method used to ionise the organic molecules. PTR-MS uses chemical ionisation, in which the VOC molecules (analyte) react with charged ions, in most cases hydroxonium ions (H_3O^+) produced in an external glow discharge ion source operating in pure water vapour. H_3O^+ ions transfer their proton exclusively to VOC molecules that have proton affinities higher than that of water, yielding protonated analyte VOCs. In PTR-MS is an electric field which accelerates the ions through the reaction chamber (Critchley et al., 2004). The presence of this field can lead to collision-induced dissociation of ions. After

scanning a mass range, fingerprints of the volatile compounds will be obtained.

The great advantage of PTR-MS, besides giving immediately absolute concentrations, is that fragmentation of the analyte molecule is very much reduced so the mass spectra produced are much easier to interpret and are more straightforward to quantify (Hansel & Märk, 2004; Lindinger et al., 1998). This rather novel technique enables a variety of organic species (such as alkenes, alcohols, aldehydes, aromatics, ketones, nitriles, sulphides and many others) in complex matrices to be monitored in real-time (within seconds), with detection limits as low as a few parts per trillion, volume (pptv). This is without any work up procedure. The PTR-MS instrument can be fully automated and no experienced operators are needed (Boscaini, Mikoviny, Wisthaler, von Hartungen, & Märk, 2004). Yet, PTR-MS is a one-dimensional technique that characterises compounds only via their mass, which is not sufficient for positive identification of the individual VOCs.

The often typical aroma, i.e. volatile compounds, of regional products can be measured by PTR-MS. At the authors' laboratory, PTR-MS has been explored for its capabilities in the field of geographical authentication of olive oils. Advanced multivariate statistics revealed that the country of origin of the olive oils was 90% classified correctly by their mass spectral data. Elsewhere, the origin of truffles, Grana cheese and wine has been assessed by PTR-MS (Aprea, Carlin, Versini, Märk, & Gasperi, 2007; Boscaini, van Ruth, Biasioli, Gasperi, & Märk, 2003; Spitaler et al., 2007). Although PTR-MS is a rather new technique that needs further research for improvements, PTR-MS has potential to be become a suitable technique for determining the geographical origin of food products more often.

2.4. Gas chromatography mass spectrometry (GC-MS)

GC-MS is an instrumental technique, comprising a gas chromatograph coupled to a mass spectrometer (Gohlke & McLafferty, 1993). GC (see also Section 4.2) separates the components of a mixture and MS characterises each of the components individually. In this way, one can both qualitatively and quantitatively analyse complex mixtures containing numerous compounds. In order for a compound to be analysed by GC-MS it must be sufficiently volatile and thermally stable. In GC-MS, the ions required for mass analysis are generally formed by electron impact ionisation. Gas molecules exiting the GC are bombarded by a high-energy electron beam (70 eV). As the MS detector is only designed to analyse clean materials careful sample preparation must be considered prior to injection in the GC-MS. GC-MS is one of the most widely used techniques and represents the method of choice for the analysis of food volatiles because of its high reproducibility (Pillonel et al., 2003). However, this technique is rather expensive and time-consuming.

GC-MS has also been applied for the determination of the geographical origin of food products. This concerns mainly dairy products. For example, Emmental cheese samples from different countries and regions were easily differentiated by using GC-MS measured compounds (Pillonel et al., 2003). Furthermore, GC-MS elucidated the relationship between the flavouring capabilities and geographical origin of natural whey cultures used for traditional water-buffalo Mozzarella cheese manufacture (Mauriello, Moio, Genovese, & Ercolini, 2003). GC-MS has also contributed to the detection of specific markers for tracing the geographical origin of food products (Fernandez, Astier, Rock, Coulon, & Berdagué, 2003). In this way the influence of pasture from a certain region on the volatile compounds in Ewes' dairy products (milk and cheese) was shown (Povolo, Contarini, Mele, & Secchiari, 2007). Specific markers were also found by GC-MS in honey (Radovic et al., 2001). These markers indicated if the honey was from Denmark, England, The Netherlands, Spain or Portugal.

3. Spectroscopic techniques

3.1. Nuclear magnetic resonance spectroscopy (NMR)

NMR is based upon the measurement of absorption of radiofrequency radiation by atomic nuclei with non-zero spins in a strong magnetic field (Ibañez & Cifuentes, 2001). The absorption of the atomic nuclei is affected by the surrounding atoms, which cause small local modifications to the external magnetic field. In this way detailed information about the molecular structure of a food sample can be obtained. Among nuclei with 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 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.

In food analysis two types of NMR are applied, low resolution NMR (LR-NMR) and high resolution NMR (HR-NMR) (Ibañez & Cifuentes, 2001). Nowadays, LR-NMR instruments (using frequencies of 10–40 MHz) are small, easy to use, and relatively inexpensive which make them suitable to perform rapid and reproducible measurements. However, LR-NMR requires reference methods to carry out quantitative analysis, and in many cases the precision of such reference method is a limiting factor. HR-NMR (using frequencies above 100 MHz) has been applied in many more food authenticity studies than LR-NMR (Reid, O'Donnel, & Downey, 2006). The advantage of HR-NMR over LR-NMR is that it is possible to obtain much more detailed information regarding the molecular structure of a food sample. The major disadvantage is that it is one of the most expensive analytical techniques to employ, both in terms of initial capital outlay and running costs. Furthermore, extraction procedures may be necessary to enrich the studied compound

as the sensitivity of HR-NMR is rather poor (Ibañez & Cifuentes, 2001).

The combination of ^{1}H NMR or ^{13}C NMR fingerprinting with advanced chemometric methods provides an original approach to study the profile of a food product in relation to its geographical origin. Although the use of ^{13}C NMR for this purpose is quite rare, ^{13}C NMR spectra of olive oils from different Italian regions were sufficiently different to permit their discrimination (Shaw et al., 1997). ^{1}H NMR has been applied more often to classify mediterranean olive oils according to their geographical origin (Mannina, Patumi, Proietti, Bassi, & Segre, 2001; Rezzi et al., 2005). For example, one approach concerned measuring ^{1}H NMR spectra of phenolic extracts of olive oils for discrimination (Sacco et al., 2000). Besides olive oils, Italian wines could be differentiated from Slovenian wines (Brescia, Kosir, Caldarola, Kidric, & Sacco, 2003) and the geographical origin of propolis samples could be determined by using ^{1}H NMR (Watson et al., 2006). Propolis is a complex resinous substance used by bees to seal their hives and is marketed by health food stores for its claimed beneficial effect on human health.

One of the major current applications of HR-NMR is site-specific natural isotope fractionation (SNIF)-NMR (Ibañez & Cifuentes, 2001). This technique is often used in food analysis and allows the determination of the geographical origin of foods based on the isotopic ratio of a given nucleus found in a constituent of the analysed food. This can be explained by the fact that the specific proportions of the particular isotopes of hydrogen and oxygen present in molecules are dependent mainly on climatic and geographical conditions (Reid et al., 2006). SNIF-NMR is often combined with IRMS (see also Section 2.1) and chemometric methods. The main drawback of SNIF-NMR is that it requires laborious sample preparation involving many purification and concentration steps (Ibañez & Cifuentes, 2001). SNIF-NMR has particularly been used for the geographical authentication of various wines (Martin et al., 1999; Ogrinc et al., 2001). By determining the natural abundance isotopic ratios of hydrogen, oxygen, and carbon, from water and ethanol extracted from the wine it is possible to distinguish between regions. Furthermore, SNIF-NMR has also been applied successfully to identify the geographical origin of natural mustard oils (Remaud, Martin, Martin, Naulet, & Martin, 1997).

3.2. Infrared spectroscopy (IR)

IR spectroscopy is the measurement of the wavelength and intensity of the absorption of infrared light by a sample (Putzig et al., 1994). In fact IR measures the vibrations of molecules. Each functional group, or structural characteristic, of a molecule has a unique vibrational frequency that can be used to determine what functional groups are present in a sample. When the effects of all the different functional groups are taken together, the result is a unique molecular "fingerprint" that can be used to confirm the identity of a sample. The infrared portion of the electromagnetic spectrum is divided into three regions; the near-, mid- and far-infrared, named for their relation to the visible spectrum. The far-infrared, (approx. 400–10 cm^{-1}) lying adjacent to the microwave region, has low energy and may be used for rotational spectroscopy. The mid-infrared (MIR) (approx. 4000–400 cm^{-1}) may be used to study the fundamental vibrations and associated rotational–vibrational structure, while the higher energy near-infrared (NIR) (14,000–4000 cm^{-1}) can excite overtone or harmonic vibrations. This means that NIR can provide more complex structural information than MIR.

IR spectroscopy is a non-invasive and non-destructive technique (Karoui et al., 2004a). The technique is rapid, relatively inexpensive and can be easily applied in fundamental research, in control laboratories, and on-line in the factory to analyse food products. The introduction of the Fourier transform technique in IR (FTIR) has increased the use of IR in food analysis (McKelvy et al., 1998). A FTIR spectrometer obtains infrared spectra by first collecting an interferogram of a sample signal with an interferometer, which measures all of infrared frequencies simultaneously. In this way the technique allows a very rapid screening and quantification of components and therefore a high throughput of samples. IR spectroscopy cannot eliminate the need for more detailed laboratory analyses, but it may help to screen samples that require further examination. Special care is necessary with regard to possible interference between components possessing similar IR spectral regions, which occurs very frequently when food products are analysed.

With respect to MIR, various wines (Picque, Cattenoz, Corrieu, & Berger, 2005), cheeses (Karoui et al., 2004a), olive oils (Tapp, Defernez, & Kemsley, 2003) and honey (Ruoff et al., 2006) have been differentiated on the basis of geographical origin. With NIR the geographical classification of grapes (Arana, Jaren, & Arazuri, 2005), wines (Liu, Cozzolino, Cynkar, Gishen, & Colby, 2006), rice (Kim, Rhyu, Kim, & Lee, 2003), soy sauce (Iizuka & Aishima, 1997) and olive oils (Downey, McIntyre, & Davies, 2003) have been accomplished. These results were obtained by combining the MIR and NIR data with chemometric methods.

3.3. Fluorescence spectroscopy

Molecular fluorescence is the optical emission from molecules that have been excited to higher energy levels by absorption of electromagnetic radiation. A typical fluorimeter contains an excitation source, a sample cell and fluorescence detector. Broad-band excitation light from a lamp passes through a monochromator, which passes only a selected wavelength. The fluorescence is dispersed by another monochromator and detected by a photomultiplier tube. Scanning the excitation monochromator gives the excitation spectrum and scanning the fluorescence monochromator gives the fluorescence spectrum. Fluorescence

spectroscopy can be used to identify and analyse fluorescent compounds at very low concentration levels (in the parts per billion range) while providing information about structure, formulation, and stability. Both solids and liquid samples can be analysed. The main advantage of fluorescence detection compared to absorption measurements (e.g., IR) is the greater sensitivity (100–1000 times) achievable because the fluorescence signal has in principle a zero background (Froehlich, 1989). Furthermore, fluorescence spectroscopy is a simple non-invasive and non-destructive technique, relatively inexpensive and allows rapid analysis.

With classical right-angle fluorescence spectroscopy, the measurements are carried out in dilute solutions where the absorbance is below 0.1. At a higher absorbance rate, a decrease of fluorescence intensity and a distortion of emission spectra are observed due to the inner filter effect. To overcome such problems, front-face fluorescence spectroscopy was developed where only the surface of the material is illuminated and examined (Ruoff et al., 2005). The emitted photons are collected at an angle of 56° to the surface of the sample, to minimize artifacts generated by the photons of excitation reflected from the sample. This technique allows a quantitative investigation of fluorophores in powders as well as in concentrated or even opaque samples.

The potential of using fluorescence in food research has increased during the last few years with the propagated application of chemometrics and with technical and optical developments of spectrofluorometers. Food products contain numerous intrinsic fluorophores and are therefore suitable for fluorescence spectroscopy investigations. Many foods contain proteins, peptides, and free amino acids which include tryptophan, tyrosine, and phenylalanine residues. Fluorescence spectroscopy provides information on the presence of these aromatic amino acids and their environment in biological samples (Luykx, Casteleijn, Jiskoot, Westdijk, & Jongen, 2004). In this way the protein structure and protein interactions in dairy products have been studied (Herbert et al., 2000). It has been shown that (front-face) fluorescence spectroscopy allows the determination of the geographical origin of various cheeses (Karoui, Bosset, Mazerolles, Kulmyrzaev, & Dufour, 2005; Karoui et al., 2004b), milks (Karoui, Martin, & Dufour, 2005) and olive oils (Dupuy et al., 2005). In one of these studies (protein) tryptophan and vitamine A (also a fluorophore) fluorescence spectra were recorded directly on Emmental cheese samples for geographical discrimination (Karoui et al., 2004b).

3.4. Atomic spectroscopy

Atomic spectroscopy can be used to analyse the vaporised atoms of metals and non-metals in a variety of samples. In atomic absorption spectroscopy (AAS) the absorption of light is used to measure the concentration of gas-phase atoms (Hook, Hosseini, & Elin, 1985; Hou et al., 2001). The atoms absorb ultraviolet or visible light and make transitions to higher electronic energy levels. The analyte concentration is determined from the amount of absorption. In general, AAS instruments can only analyse one or a few elements simultaneously. AAS requires that the analyte atoms are in the gas phase. Therefore, ions or atoms in a sample must undergo desolvation and vaporisation in a high-temperature source such as a flame or electrically heated graphite furnace. Graphite furnace AAS (GF-AAS) has several advantages over flame AAS (F-AAS). For example, F-AAS can only analyse solutions, while GF-AAS can accept solutions, slurries, and solid samples. Furthermore, the use of controlled temperature and heating time in GF-AAS result in higher sensitivity than achieved with F-AAS (Ibañez & Cifuentes, 2001). This is due to the lower dilution of elements and the longer times that they remain in the vapour phase and, therefore, in the optical path. Moreover, the ability to reproduce from sample to sample the time–temperature profile, now controlled by computer in new instrumentation, is fundamental for achieving a good accuracy and precision. On the other hand, the sample matrix has a much higher influence on the results obtained in GF-AAS than in any other atomic spectroscopic technique. Therefore, special care has to be put on the sample preparation, and different strategies have been developed to overcome this deficiency (e.g., standard addition, modifiers). In general, instrumental miniaturisation has to be considered for the atomic spectroscopy techniques in order to achieve less expensive equipments.

Atomic emission spectroscopy (AES) uses quantitative measurement of the optical emission from excited atoms to determine analyte concentration (Hook et al., 1985; Hou et al., 2001). Analyte atoms in solution are aspirated into the excitation region where they are desolvated, vaporised, and atomised by a flame, discharge, or plasma. The high-temperature atomisation sources provide sufficient energy to promote the atoms into high-energy levels. The atoms decay back to lower levels by emitting light. Since the transitions are between distinct atomic energy levels, the emission lines in the spectra are narrow. The spectra of multi-elemental samples can be very congested, and spectral separation of nearby atomic transitions requires a high resolution spectrometer. Since all atoms in a sample are excited simultaneously, they can be detected simultaneously, and is the major advantage of AES compared to AAS. Inductively coupled plasma AES (ICP-AES) provides higher reproducibility and quantitative linear range than the conventional methods (Ibañez & Cifuentes, 2001). Here, a very high-temperature (7000–8000 K) excitation source is used and molecular interferences are greatly reduced. However, analyses are more expensive and all samples have to be introduced in a dissolved form. Furthermore, in ICP-AES complex samples usually provide patterns of very difficult interpretation.

The inorganic elemental analysis by AAS and/or AES and often combined with chemometrics enabled the determination of the geographical origin of various food products. Via AAS it was possible to relate the selenium content of beef to the geographical region (Hintze, Lardy,

Marchello, & Finley, 2001), and geographically discriminate honeys (Paramás et al., 2000) and wines (Frías, Trujillo, Peña, & Conde, 2001) by measuring various mineral elements. A combination of AAS and AES allowed the determination of the geographical origin of orange juice, nuts (Schwartz & Hecking, 1991) and potatoes (Rivero, Hernández, Rodríguez, Martín, & Romero, 2003). Chemical profiling by ICP-AES differentiated the geographical growing origins of coffee (Anderson & Smith, 2002) and several fruits (Perez et al., 2006). Furthermore, ICP-AES and AAS have also been successfully combined with ICP-MS (see also Section 2.2) to classify onions (Ariyama et al., 2007; Ariyama et al., 2004) and tea (Moreda-Piñeiro et al., 2003) according to region of origin.

4. Separation techniques

4.1. High performance liquid chromatography (HPLC)

HPLC is a form of LC to separate, analyse and quantify compounds that are dissolved in solution. HPLC instruments consist of a reservoir of mobile phase, high pressure pump, injector, analytical column (temperature controlled), and detector. Compounds are separated by injecting the sample mixture (carried by mobile phase) onto the column. The different components in the mixture pass through the column at different rates due to differences in their partitioning behaviour between the mobile liquid phase and the stationary phase (column material). Dependent of the type of stationary phase, compounds can be separated based on their charge (weak/strong cation or anion exchange chromatography), molecular mass (size exclusion chromatography), hydrophobicity/polarity (reversed-phase HPLC, hydrophobic interaction chromatography), and specific characteristics (affinity chromatography). The most common detectors in HPLC are an ultraviolet–visible (UV–vis) light absorbance detector, a fluorescence detector, an electrochemical detector and a diffractometer.

HPLC has been used in food analysis for measuring numerous compounds, e.g. carbohydrates, vitamins, additives, mycotoxins, amino acids, proteins, tryglycerides in fats and oils, lipids, chiral compounds and pigments. HPLC is a straightforward, robust and reproducible technique and the price of an HPLC instrument is modest. Several sensitive selective detectors are available to utilise with HPLC depending on the compound to be analysed. Another advantage of HPLC is that both the stationary and mobile phases can be selected to achieve the necessary separation of sample components. Furthermore, separated compounds can be collected as they elute from the column. Moreover, the rather new monolithic HPLC columns provide higher efficiency and better permeability than the conventional HPLC columns, allowing a higher flow rate and therefore reducing total analysis time (Cabrera, 2004). One of the most important trends is the miniaturisation of the analytical systems; therefore micro- and nano-columns and microparticles have been developed for HPLC to obtain even faster separations (e.g., ultra high performance LC). Furthermore, two-dimensional LC makes HPLC an even more valuable and powerful analytical tool (Tranchida, Dugo, Dugo, & Mondello, 2004).

European wines from different geographical origin have been correctly classified on basis of the chromatography profiles obtained with HPLC in combination with a UV–vis and/or fluorescence detector. These HPLC studies encompassed the analysis and quantification of either phenolic compounds (Rodriquez-Delgado, Gonzalez-Hernandez, Conde-Gonzales, & Perez-Trujillo, 2002), amino acids and biogenic amines (Hérberger, Csomós, & Simon-Sarkadi, 2003), or contaminant ochratoxin A (Bellí, Marín, Sanchis, & Ramos, 2002). Apart from wines, HPLC has also been used to geographically discriminate honey (Tomás-Barberán, Ferreres, García-Viguera, & Tomás-Lorente, 1993), nuts (Gómez-Ariza et al., 2006), olive oil (Stefanoudaki, Kotsifaki, & Koutsaftakis, 1997) and cheese (Di Cagno et al., 2003) on basis of the HPLC profiles of flavonoids, metal-binding proteins, triglycerides and peptides, respectively. Most studies described above were employed in conjunction with chemometric methods.

4.2. Gas chromatography (GC)

GC is one of the most universal separation techniques used in food analysis, mainly in volatile and semi-volatile composition studies, aromas, and pesticides (Chang, Holm, Schwarz, & Rayas-Duarte, 1995). A mixture of compounds to be analysed is initially injected into the GC where the mixture is vaporised in a heated chamber. Subsequently, the gas mixture travels through a GC column, where the compounds become separated as they interact with the coating of the column. This column contains a liquid stationary phase which is adsorbed onto the surface of a thin fused-silica capillary tube. The sample is transported through the column by the flow of an inert carrier gas. Commonly used gases include nitrogen, helium and hydrogen. There are different GC detectors with various types of selectivity (roughly in order from most common to the least): the flame ionisation detector (FID), thermal conductivity detector, electron capture detector, photo-ionisation detector, flame photometric detector, thermionic detector, atomic emission detector and ozone- or fluorine-induced chemiluminescence detector (Grob, 2004). All these detectors produce an electrical signal that varies with the amount of analyte exiting the column. Another GC detector that is expensive but very powerful is the MS detector (see also Section 2.4). The detectability of the detectors varies from femtogram to nanogram. The FID is a useful general detector for the analysis of organic compounds. It has high sensitivity, a large linear response range, and low noise. It is also robust and easy to use, but unfortunately, the sample is no longer available after detection.

Some significant advances for GC have been the development of coupling GC to IRMS (Meier-Augenstein,

2002), two-dimensional GC to analyse very complex samples (Tranchida et al., 2004), and high speed GC (Eiceman, Hill, & Gardea-Torresdey, 1998). These latter developments allow real-time process monitoring and on-site analysis (portable GC). In comparison with HPLC, the mobile phase in GC has a very limited role in the separation process. Next to this, analysing polar and ionic molecules with GC and collecting components after GC separation for further analysis are difficult to do and are rarely done.

By analysing the GC profiles of various compounds (e.g., alkanes, aldehydes, alcohols, acids) present in wine it is possible to classify wines according to their geographical origin (Etièvant, Schlich, Cantagrel, Bertrand, & Bouvier, 2006; Shimoda, Shibamoto, & Noble, 1993). Determination of the fatty acid composition and corresponding concentrations by GC allowed the geographical discrimination of milk samples (Collomb, Bütikofer, Sieber, Jeangros, & Bosset, 2002) and olive oils (Ollivier, Artaud, Pinatel, Durbec, & Guérère, 2003). Furthermore, determination of the geographical origin of cocoa masses (Hernández & Rutledge, 1994) and orange juices (Ruiz del Castillo, Caja, Blanch, & Herriaz, 2003) were accomplished via GC analysis. It must be noted that the GC data were subjected to chemometric methods.

4.3. Capillary electrophoresis (CE)

CE can be used to analyse and characterise a wide variety of analytes ranging from simple inorganic ions, small organic molecules, peptides, proteins, nucleic acids to viruses, microbes, and particles (Kvasnička, 2005). CE comprises a family of electrokinetic separation techniques that separate compounds based upon differences in electrophoretic mobility. This mobility can be controlled by the charge/mass ratio of the analyte, isoelectric point, molecular size, or hydrophobicity depending on the separation conditions. The most employed modes of CE include capillary zone electrophoresis, capillary isoelectric focusing, capillary gel electrophoresis and capillary electrochromatography. The main components in CE instrumentation are a sample vial, source and destination vials, a capillary (internal diameter 10–100 μm), electrodes, a high-voltage power supply and a detector. The source vial, destination vial and capillary are filled with an electrolyte such as an aqueous buffer solution. The sample is normally introduced by placing the capillary inlet into the sample vial and applying pressure. After returning the capillary inlet into the source vial, the migration of analytes is initiated by applying an electric field between the source and destination vials. This electric field is supplied to the electrodes by the high-voltage power supply. All ions, positive or negative, are pulled through the capillary in the same direction by electroosmotic flow. The analytes separate as they migrate due to their electrophoretic mobility and are detected near the outlet end of the capillary. Separated chemical compounds appear as peaks with different retention times in an electropherogram. Typical CE detectors are based on spectrophotometry, fluorescence, conductivity or MS (Vallejo-Cordoba, González-Córdova, Mazorra-Manzano, & Rodríguez-Ramírez, 2005).

In general CE is simple, rapid and low cost, because it needs neither laborious treatment of the samples nor long times for analysis (Mato, Huidobro, Simal-Lozano, & Sancho, 2006). Moreover, high separation efficiency can be achieved. The main disadvantage of CE is its lower reproducibility when compared to other (chromatographic) methods. Furthermore, although the sample volume usually consumed per CE analysis is a few nanoliters, the sensitivity in terms of concentration is not very high, which precludes the use of CE for determination of trace compounds. However, extremely sensitive detection systems like a laser-induced fluorescence and MS detector enhance the sensitivity (Vallejo-Cordoba et al., 2005). The use of multi-capillary arrays and chip-based separations are very helpful to overcome throughput limitations (Frazier, 2001).

In the nineties CE has been proposed for the first time as a technique for studying the geographical origin of a food product (Delgado, Tomás-Barberán, Talou, & Gaset, 1994). This study concerned the determination of flavonoids which accumulate in different proportions in honey depending on their geographical origin. In a similar way CE has been applied recently to geographically discriminate Chinese fruit extracts (Peng, Liu, & Ye, 2006). CE has also been used to determine the content of six metals in a variety of Spanish red wines in a single analysis (Núñez, Peña, Herrero, & García-Martín, 2000). The results allowed the classification of the wines according to their geographical origin. Furthermore, CE-profiles were suitable to differentiate herb samples by geographical origin (Wang, Cao, Xing, & Ye, 2005).

5. Other techniques

5.1. Sensor technology

Sensor technology, sometimes referred to as 'electronic nose technology', is based on the detection by an array of semi-selective gas sensors of the volatile compounds present in the headspace of a food sample (Strike, Meijerink, & Koudelka-Hep, 1999). The technology includes a sample delivery system, detection system, and computing system. The sample delivery system enables the generation of the headspace (volatile compounds) of a sample. The system then injects this headspace into the detection system of the electronic nose which consists of a sensor set. Each sensor is sensitive to all volatile molecules but each in their specific way. Most electronic noses use sensor-arrays that react to volatile compounds on contact: the adsorption of volatile compounds on the sensor surface causes a physical change of the sensor. A specific response is recorded by the electronic interface transforming the signal into a digital value. Recorded data are then computed based on statistical models. The computing system works to combine the responses of all of the sensors, which represents the input

for the data treatment. This part of the instrument performs global fingerprint analysis and provides results and representations that can be easily interpreted. Moreover, the electronic nose results can be correlated to those obtained from other techniques (e.g., sensory panel).

Advantages of electronic nose technology include a relatively small amount of sample preparation, a simple procedure, and a fast and cheap analysis (Franke et al., 2005). A disadvantage of this technique is that the identification of the chemical compounds detected is not possible and that the detection limit is high compared to that of other methods (e.g., GC-MS). Furthermore, this technique is sensitive to water vapour and drifts of the recorded response occur during the day. Nevertheless, the electronic nose has been successfully applied for differentiation of olive oils (Guadarrama, Rodriguez-Mendez, Sanz, Rios, & de Saja, 2001), wines (Penza & Cassano, 2004), orange juice (Steine, Beaucousin, Siv, & Pfeiffer, 2001) and cheese (Pillonel et al., 2003) on the basis of geographical origin.

5.2. DNA technology

The polymerase chain reaction (PCR) is a biological technique allowing the detection of very low amounts of nucleic acid probes and the determination of their sequence via the amplification of DNA or RNA individual strains (Ibañez & Cifuentes, 2001). This technique is used extensively to identify the species of origin in foods. PCR is a multiple-process with consecutive cycles of three different temperatures, where the number of target sequences grows exponentially according to the number of cycles (Marmiroli, Peano, & Maestri, 2003). In the first step of the cycle the template, the DNA serving as master copy for the DNA polymerase is separated into single strands by heat denaturation at 95 °C. In the second step the reaction mix is cooled down to a temperature of 50–60 °C to allow the annealing of primers (short DNA sequences) to the target sequence. In the third step, the annealed primers are extended using a polymerase and nucleotides at the optimum temperature of 72 °C. With the elongation of the primers, a copy of the target sequence is generated. The cycle of these three different temperatures is then repeated from 20 to 50 times, depending on the amount of DNA present and the length of the amplified DNA fragment. Subsequently, the amplified DNA is analysed for size and purity by biological procedures, mainly by size fragment length polymorphism, and visualised via gel electrophoresis (De La Fuente & Juárez, 2005).

PCR has the advantage of high sensitivity and rapid performance with high sample numbers being automatically processed (De La Fuente & Juárez, 2005). Furthermore, PCR is based on resistant material (nucleic acids). In order to use PCR, the exact nucleotide sequences that flank both ends of the target DNA region must be known (Marmiroli et al., 2003). Any PCR-based detection strategy will thus depend on the selection of the oligonucleotide primers and the detailed knowledge of the molecular structure and DNA sequences used. The yield obtained by PCR depends on the DNA concentration and the quality and purity of DNA (Ibañez & Cifuentes, 2001). DNA fragments of similar species within a sample can bring about cross-reactions, and more fragmented DNA provides lower amplification. Therefore, special care has to be paid during the sample treatment prior to DNA amplification because it can modify to a large extent the final results.

It was shown by PCR fingerprinting that the genetic diversity of PDO water-buffalo Mozzarella's microflora can be exploited to geographically discriminate cheeses (Bonizzi, Feligini, Aleandri, & Enne, 2006). In a similar way, the microbial diversity of natural whey cultures proved to be closely related to the geographical origin (Mauriello et al., 2003). With respect to the geographical discrimination of grapes, determination of the grapes' number of yeasts and yeast species by PCR was a suitable approach (Raspor, Milek, Polanc, Možina, & Čadež, 2006). PCR should also allow the identification of the geographical origin of olive oil (Woolfe & Primrose, 2004). Here, it is mainly the identification of the olive cultivar used for the oil production which is of importance although the same variety may be used in different regions within Europe albeit under different names.

5.3. Sensory analysis

Sensory analysis (or sensory evaluation) is considered as an important technique to determine product quality. It comprises a set of techniques for accurate measurements of human responses to foods (Pérez Elortondo et al., 2007). Appearance, odour, flavour and texture properties are important characteristics determining the quality of food products. Sensory analysis requires panels of human assessors, on whom the products are tested, and recording the responses made by them. By applying statistical techniques to the results it is possible to make inferences and insights about the products under test. Sensory analysis requires extensive training of judges, and adequate replication and detailed statistical analysis of the observations. Numerous studies have been performed in attempts to find correlations between sensory qualities and objective instrumental measurements (Karoui & De Baerdemaeker, 2007).

Principal component analysis of sensory data permitted differentiation between wines from different geographical areas (Schlosser, Reynolds, King, & Cliff, 2005; Vilanova & Vilariño, 2006). In these studies usually multiple descriptors are identified by the wine tasters in the first phase. Subsequently, the descriptors are reduced in number using statistical methods. In the second phase, the wine tasters identify and rate the intensity of the remaining descriptors for each wine evaluated. After that characteristic descriptors may be found in wines from each particular geographical area. It must be noted that often grape cultivar and wine production area are linked. Normally, a better geographically classification of wines is achieved based on chemical data instead of sensory data (Sivertsen, Holen,

Table 2
Overview of several characteristics of the analytical techniques which can be used for determination of the geographical origin of food products

Techniques	Sensitivity	Simplicity	Time analysis	Costs	Reported applications	Compounds	Identification/profiling
MS							
IRMS	+	+/−	+/−	−	+	Various	i + p
ICP-MS	+	+/−	+	−	+	Elements	i + p
PTR-MS	+	+	+	−	−	Volatile	p
GC-MS	+	+	+/−	−	+	(Semi) volatile	i + p
Spectroscopy							
NMR	−	+/−	+/−	−	−	Various	i + p
IR	+/−	+	+	+	+	Various	p
Fluorescence	+	+	+	+	−	Various	p
Atomic	+/−	+/−	+/−	+/−	+	Elements	i + p
Separation							
HPLC	+/−	+	+/−	+	+	Various	p
GC	+	+	+/−	+	+	(Semi) volatile	p
CE	−	+	+/−	+	−	Various	p
Other							
Sensor technol.	−	+	+	+/−	+/−	Volatile	p
PCR	+	+/−	+/−	+	−	DNA	i + p
Sensory analys.	+/−	+/−	−	−	−	Various	p

The characteristics include the sensitivity and simplicity of the analytical tool, time of the analysis, instrumental costs, number of reported applications for determining the geographical origin, types of compounds which are analysed and possibility of identifying (i) and/or profiling (p) (including chemometrics) these compounds. Favourable (+), moderate (+/−), unfavourable (−).[a]

[a] For abbreviations of the techniques, see Table 1.

Nicolaysen, & Risvik, 1999). Sensory analyses have also been applied to geographically discriminate a spirit drink (absinthe) (Lachenmeier, 2007), cheeses (Pillonel et al., 2002) and olive oils (Stefanoudaki, Kotsifaki, & Koutsaftakis, 2000).

6. Conclusions

The interest of consumers in high quality foods with a clear geographical identity has grown rapidly. Therefore, participation to protected food names (PDO, PGI, TSG) is encouraged in the EU. This means that suitable techniques for determining the geographical origin of food products are highly desirable. In the last 15 years analytical tools for studying food have become more sensitive, reliable and faster. Computer software and hardware have made the analytical instruments more user-friendly so that they can be used routinely by workers. The instruments are also more operated in conjunction with computer networks to store information about a certain food or constituent in databases. The application of chemometrics is often required to handle the amount of accumulated data and to detect subtle differences that frequently exist between food samples. All these progressions in instrumental analysis have led to the development of several suitable analytical methods for the determination of the geographical origin of food products. These methods are capable of analysing specific characteristics of a product which are influenced by geographically specific factors. An overview of these analytical methods with their pros and cons is presented in Table 2. It is clear that all described techniques have advantages and limitations. For example, IRMS is a successful tool for determining the geographical authenticity of numerous food products, even without the use of chemometric analysis. However, the instrumental costs are quite high and the speed of the analysis is moderate. An accurate determination of the geographical origin of a food product seems feasible when various parameters are measured in a food product (e.g., identification and quantification of inorganic elements, and profiling volatile compounds). Likewise, a combination of different analytical techniques (e.g., ICP-MS and PTR-MS) could be more useful than relying on one single method. Other methods (e.g., determination of animal genotype, gross chemical composition, sensory properties) could be supportive as they help to determine related indicators such as species and feeding conditions but not directly the geographical origin. By way of such a multifactorial approach, all data must be carefully interpreted and cross-validated with chemometric tools.

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