Review paper

HONEY TRACEABILITY AND AUTHENTICITY. REVIEW OF CURRENT METHODS MOST USED TO FACE THIS PROBLEM

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Abstract

In this review, we have focused on analytical methods for characterizing the geographical and botanical origin of honey. We evaluated works concerning the methods most used in the analysis of the geographical and botanical origin of honey, paying particular attention to what has been used in the last decade. We investigated starting from the most common methods used at the beginning of this century, such as melissopalynological analysis, the analysis of the mineral profile or chemico-physical parameters, to the current state-of-the-art technologies and methods including the metabolomic and genomic approaches, the blockchain or Internet of Things. For each methodology, several examples have been given, and advantages and limitations are explained. Whereas currently only one approach is rarely used, many methods are used in combination because a combined approach usually leads to greater accuracy, also thanks to the current possibilities that multivariate statistical methods or chemometric methods offers us.

Keywords: analytical methods, geographical origin, honey, "omic" sciences, spectroscopy

INTRODUCTION

Honey is a natural viscous, aromatic and sweet substance from various types of secretions of different plants (Codex Alimentarius Commission, 2001; Trifković et al., 2017) and produced by the honey bees, such as the western honey bee (*Apis mellifera* L.) (Engel, 1999). Honey contains over 180 different organic and inorganic compounds of which the sugars, in particular fructose and glucose, accounts for the main percentage (Alvarez-Suarez et al., 2010). Honey is reported to have a positive effect on the human health, due to its antioxidant, antimicrobial, anti-inflammatory, anti-atherogenic, antithrombotic, immunomodulatory and analgesic properties (Gomes et al., 2010; Trifković et al., 2017; Conti et al., 2018; Moein et al., 2022). In particular, among the antioxidants, phenolics are a chemical heterogeneous group well represented in honey, but their quantity is based on the botanical composition and geographic origin and also on the processing and conservation methods (Al-Mamary et al., 2002; Beretta et al., 2005; Andersen & Markham, 2006; Bertoncelj et al., 2007). Honey also contains minerals ranging from 0.04 to 0.20% depending on the type (Bogdanov et al., 2007). The mineral content of honey can encompass some undesired trace elements, called "heavy metals" such as lead, which are relevant markers for assessing the honey quality under the food safety standpoint (Hernández et al., 2005; Solayman et al., 2016). Toxic elements in honey can come from such pollution sources as industrial plants, environmental disasters, urbanization, anthropic areas or pesticides usage (Bogdanov, 2006; Solayman et al., 2016; Perna et al., 2021; Mititelu et al., 2022).

Notwithstanding, the expectations for the honey are usually high, and the authenticity of honey can depend on (Bogdanov & Martin, 2002) honey production (geographical and botanical origins) and labelling (*e.g.*, natural, organic, raw and unheated). The botanical origin of floral honeys are divided into two broad groups, the cheaper multifloral honeys and the more expensive monofloral honeys, which

are appreciated by the market and subject to adulteration (Everstine et al., 2013; Ballabio et al., 2018). In Europe more than one hundred botanical species can potentially give monofloral honeys (Persano Oddo & Piro, 2004). Different methods for the determination of the botanical and geographical origins of honey have been developed, applied and evaluated over the last decades. There is often a strong relationship between botanical and geographical origins, and some nectareous plant species are strictly related to a specific geographic area, *e.g.*, the Mediterranean basin (Trifković et al., 2017). Composition, physical and organoleptic properties of honey depend on the types of flowers, plant secretion or sucker insect secretions gathered by the honey bees. In principle, types of honey having different chemical profiles should be rather easy to distinguish and to identify, but in some circumstances, the chemical profiles of different monofloral honeys are similar, making discrimination difficult (Rodríguez-Flores et al., 2019).

Identifying its botanical and/or geographic origin of requires the history of the honey to be traced of from the source, *i.e.*, the hive, to the consumer. For the traceability of agricultural products, a large volume of data must be collected across the supply chain. This approach implies risks, such as misregistration of information, and to avoid such a problem, in recent years the study of honey traceability has evolved in the search for increasingly reliable, accurate, economic and fast methods suitable for standardization (Laube et al., 2010; Soares et al., 2015). Parallel to the development of these traceability approaches, research is also moving towards the study of the authenticity of honey controllable with official and/or standardized methods (Sobolev et al., 2017). This review is aimed at summarizing the methods for characterizing the origin (botanical and/or geographical) of honeys or, in other words, their traceability and authenticity. In particular, this review takes a retrospective investigation about what has been the evolution of these studies.

METHODOLOGY OF REVIEWING

In this review, a detailed computer-based search of the methods and strategies used for the identification of honey was performed through some publications databases (*e.g.,* Google Scholar, Scopus and Web of Science). Keywords included "honey origin", "mineral profile", "antioxidant activity", "physical parameters", "chemical parameters", "genomic", "metabolomic", "hyperspectral imaging", "botanical origins", "spectrometry", "spectroscopy", "adulteration", "mineral profile", "blockchain", "mellissopalynology" and "isotopic ratio". Methods that had feedback on more than one article were examined with the aim to verify the reliability of the methods and their application to different geographical areas and/or honey types. The main outcomes of the literature search were grouped according either to the main methodological approach for describing the authenticity of monofloral honeys or their geographic/botanic origin.

MELISSOPALYNOLOGICAL ANALYSIS

Melissopalynology, one of the most affirmed methods for honey identification, is based on the analysis of pollen grain naturally occurring in honey. A microscopic analysis identifies all the components of the pollen spectrum, which can be compared with pollen spectra already known (Anklam, 1998). This method unequivocally provides important information about the nectar source and evidence of the botanical origin of a honey sample (Von Der Ohe et al., 2004; Corvucci et al., 2015). In addition, this method is a valid choice for the geographical fingerprint of honeys in case of areas with site-specific melliferous flora (Anklam, 1998). Through the melissopalynological analysis, honey are classified as monofloral if the characteristic pollen (*i.e.,* the pollen granules referable to a specific botanical species) exceeds a specific threshold, usually expressed as percentage of the total pollen amount (Louveaux et al., 1978).

However, some types of pollen (*e.g.*, from *Robinia pseudoacacia*, *Citrus* spp., *Tilia* spp.)

are underrepresented or overrepresented (*e.g.*, *Castanea sativa*, *Eucaliptus* spp.). For example, *Citrus* and lavender honey must have 10-20% of the related pollen, but rosemary honey over 20% of *Salvia rosmarinus* pollens (Louveaux & Maurizio, 1970; Louveaux et al., 1978; Mendes et al., 1998; Soria et al., 2004). However, in the majority of circumstances such pollen percentages can be considered as a reference, because the true pollen quota of a given plant species occurring in monofloral honey varies according to its geographic origin. Melissopalynological analysis is usually used in combination with other analyses, for example sensory and physico-chemical analysis (Persano Oddo & Bogdanov, 2004; Rodriguez et al., 2010). Though a well-recognized method for honey typing, melissopalynology has such disadvantages as (Molan, 1998; Von Der Ohe et al., 2004) 1) the considerable amount of experience needed by the analyst, 2) seasonal variability of the amount of pollen grains, 3) insufficient amount of pollen grains due to filtration processes, and 4) addition of pollens fraudulently or involuntarily by bees themselves (pollen of flowers that the bee has not visited directly, but which accidentally end up on its body). In addition, the air where the honey is managed might be contaminated (Corvucci et al., 2015). Currently, new artificial pollen classification methods based on automated systems for counting and classification have helped researchers to have faster and more accurate palynological analysis. Such approaches are based on a combination of image processing of light microscope or SEM images, with some statistical or machine learning-based classifier (Ronneberger et al., 2002; Holt et al., 2011; France et al., 2014). As an example, Classyfinder (Lagerstrom et al., 2015) combines microscopy, robotics, pattern recognition, image processing and data science to form a complete, stand-alone automated pollen analysis system which can be applied in any palynology lab where conventional slide-based pollen analysis is performed. Sevillano et al. (2020) used Classifynder for the classification of forty-six different types of pollen grains. An image analysis and machine learning model with a classification accuracy over 97% allowed pairs of pollen types to be correctly distinguished which were considered difficult to distinguish virtually (*e.g.*, *Leptospermum scoparium* and *Kunzea ericoides*) by palynologists.

Physico-chemical parameters

Each physico-chemical trait has a different discriminatory value for each group of floral honeys. In general, the minimum/maximum values are used as diagnostic tools rather than the average values. For example, the honeys of *Robinia* sp., *Hedysarum* spp., *Rhododendron* spp., and *Citrus* spp. are discriminated on the base of their light colour and low electrical conductivity. In addition, *Citrus* honeys are also characterized by low diastase levels (Persano Oddo et al., 1995). Kavanagh et al. (2019) studied the power of colour, pH and total phenolic content (TPC) to discriminate 131 Irish honeys (124 multifloral, 6 monofloral) from eight monofloral non-Irish. Kavanagh et al. (2019) highlighted that the physico-chemical characteristics of unifloral honey samples are useful for the discrimination of their botanical and geographical origin and suggested the pH as one of the most discriminatory parameters. However, as several chemical and physical parameters are jointly analysed with the aim of enhancing the discriminating power of the approach, it can be useful to determine the minimum set to be studied to get a satisfying discrimination. Devillers et al. (2004) tried to determine the minimum number of physico-chemical traits necessary to obtain the best classification of botanical origin of 469 samples of monofloral honeys (fir, cinder heather, chestnut, lavender, acacia, rape and sunflower) from different geographic areas of France (not specified). Conductivity, pH, free acidity, and the percentage of fructose, sucrose and raffinose out of thirteen traits allowed maximum 100% accuracy of classification. Uršulin-Trstenjak et al.

(2017) measured in honey samples from three different regions of Croatia the water content, the electrical conductivity, the free acidity, the diastase activity, the reducing sugars, the content of pollen and of twelve minerals (K, Ca, Na, Mg, Zn, Al, Fe, Cu, Mn, Ni, Pb, Cd). Through Cluster Analysis and Principal Component Analysis, the East Croatia honey samples were shown to have the highest concentrations of water, HMF, and all the elements except for Al. In comparison, the Istria samples were richer in reducing sugars and had higher free acidity, diastase activity, conductivity, content of *R. pseudoacacia* pollen grains, while lower concentrations of most elements. Kadar et al. (2011) discriminated orange from lemon honeys by integrating physical-chemical parameters (including electrical conductivity, colour, diastase activity, flavonoids and phenolics acids), melyssopalinological analysis and thirty-seven volatile compounds (acetic acid, 8 alcohols, 10 aldehides, 3 hydrocarbons, 7 ketones, 3 furanes, 1 ester and 1 sulphur compound, extracted and desorbed thermally and subsequently analyzed by gas chromatography - mass spectrometry). The success rate of the classification was higher than 96% combining physico-chemical parameters and volatile compounds.

Bioactive compounds content

Honey is a natural antioxidant food with many beneficial properties (Aljadi & Kamaruddin, 2004; Chua et al., 2013), but the different geographical and botanical origin of honey leads to variable content of phenolic compounds and different antioxidant properties (Dżugan et al., 2018). Overall, the botanical origin of the honey most influences its antioxidant activity, while processing, handling and storage only slightly affect honey antioxidant activity (Beretta et al., 2005; Bertoncelj et al., 2007). In a study carried out by Gül and Pehlivan (2018) in Turkey, *Rhododendron* sp. and *Petroselinum* sp. honeys showed a greater antioxidant activity assayed as total phenolic content, 2,2-diphenyl-1-picrylhydrazyl (DPPH), iron reduction power (FRAP) and by the β-carotene-linoleic acid emulsion method in comparison to black locust and *Citrus* honeys. Interestingly from the geographic traceability standpoint, honeys of the same variety but of different origins can have variations in their antioxidant activity (using DPPH test, FRAP test and PLC to determine water, and fat-soluble antioxidant fractions) as shown by Tomczyk et al. (2019) who compared different types of honey (multifloral, linden, black locust and forest) coming from Poland or from the Czech Republic. However, Tomczyk et al. (2019) failed to discriminate rapeseed honey that showed more homogenous characteristics regardless the geographical provenance.

Mineral profile

Minerals in honey are divided (Solayman et al., 2016) into main elements (>50 mg/g), trace elements (<50 mg/g) and ultra-trace elements (<1 μg/g). Some trace minerals (*e.g.*, Se) are useful for consumers' health, especially if they come from an organic source (Solayman et al., 2016), but others (*e.g.*, Pb or Cd) can exhibit toxic activity (Leita et al., 1996). The content of main minerals and trace elements in honey samples above the background concentrations also provides information about environmental pollution (Rashed & Soltan, 2004; Perna et al., 2014; Solayman et al., 2016; Conti et al., 2018; Oliveira et al., 2019). Different honeys are characterized by different mineral profiles that in conjunction with another type of analysis (*e.g.*, physico-chemical) can be potentially used to discriminate them at the botanical and/or at the geographic origin level (Rizelio et al. 2012; Jovetić et al. 2017; Bergamo et al. 2018). Fernández-Torres et al. (2005) in a study on discriminating the botanical origin of four different Spanish honeys (*Eucalyptus*, orange, rosemary and heather) established a methodology based on the mineral profiling (analysed with Inductively coupled plasma atomic emission spectroscopy, ICP-AES) with wide margins of usability. The concentrations of elements Zn, Mn, Mg and Na were dependent on honey botanical origin and revealed these honeys' discriminatory power.

Furthermore, Patrignani et al. (2015) used a combined analytical approach encompassing total phenolic content (with Folin-Ciocalteu method), colour, ash content and mineral profile (by AAS for Fe, Ca, Cu, Zn and Mg, and AES for K and Na) for the geographic traceability of blossom honeys from seven geographical regions of the Buenos Aires province.

The results of this approach allowed a right-classification as high as 98% (Patrignani et al., 2015). Karabagias et al. (2018) observed that the content of B, Ca, Si, Fe, P, Mn, and Mg in honey samples from Greece, Egypt and Cyprus gave less reliable results (79.4% right-classification success) than by using moisture content, free acidity, electrical conductivity, total dissolved solids, salinity, colour and pH (91.2% of right-classification success). These differences were attributed to honeys' botanical origin, similarities in soil and vegetation conditions of production areas, harvesting year, honey processing and extraction methods. Louppis et al. (2017) used a combined methodology of mineral profiling (Tab. 1) and the analysis of physico-chemical traits (pH, moisture content, electrical conductivity, free acidity, lactonic acidity, total acidity and ash content) to discriminate the botanical origin of 207 honey samples. The combined approach resulted in satisfactory discriminating performances reaching a 96.1% right botanical classification. Oroian et al. (2015) carried out a study (Tab. 1) in the Suceava, Botoșani and Vaslui regions of Romania and highlighted the mineral profiling as a good approach for the botanical classification of honey (80.8% of right-classified samples) but judged it as unsuitable for the geographical origin (only 21.2% of the samples were rightly classified on the basis of the area of production). Such poor performance was explained by the diffuse occurrence of Fe and Zn that biased subtle differences among the regions under study. Tab. 1 lists other experimental works aimed at studying the mineral profile of honey for botanical anf/or geographical classification.

Chemico-physical properties evaluable through spectroscopy/spectrometry

Spectroscopy techniques can be used with excellent results to determine the botanical and geographical origin of honey (Noviyanto & Abdulla, 2020). The advantage of using some spectroscopic techniques on honey samples is their low invasiveness and suitability to work with non-manipulated samples (Minaei et al., 2017). The VIS-NIR hyperspectral imaging is a promising approach in this field that involves both spectroscopy and digital imaging (Schaepman, 2007; Minaei et al., 2017). Based on NIR hyperspectral imaging data, Noviyanto and Abdulla (2020) built a classification model using fifty-eight samples of New Zeland honey, that gave an overall balanced accuracy of classification of the botanical origin (90%). Lastra-Mejías et al. (2018) experimented the fluorescence spectroscopy technique on honey (Tab. 2), a economical, easy and fast technique at the same time, which has proved its efficiency for the characterization of a group of forty-four samples of honey with nine different botanical origins and rice syrups (adulterant of honey), with a 95% prediction accuracy.

The rapid evaporative ionization mass spectrometry analysis (REIMS) is a form of ambient mass spectrometry, which was initially used for *in situ* real-time analysis of tissue samples for medical and surgical purposes (Balog et al., 2010). REIMS has more recently been used in food analysis applications and also for food safety purposes (Connolly et al., 2016; Wang et al., 2019). Wang et al. (2019) used REIMS to accurately identify the botanical origin and adulteration of 127 samples of honey (unfloral and multifloral), fifteen corn syrup and four rice syrup samples. By this way, the botanical origin of the honey samples was classified very fast and accurately (up to 99.7%).

Another way to obtain reliable information on the origins of honey samples is measuring their stable isotopic abundance with the Isotope Ratio Mass Spectroscopy (IRMS). The first application of this techniques was for the identification of honey adulteration

Table 1.

Studies in which analyses of the mineral profile of honey were carried out to evaluate its geographical (GO) and botanical origin (BO)

Reference	Type of honey	Number of samples	From	Analytical technique	Aim	Elements
Conti et al. (2007)	*Acacia*, Multifloral, Honeydew	69	IT	F-AAS, GF-AAS	BO	Na, K, Ca, Mg, Fe, Cu, Mn
Vanhanen et al. (2011)	*Leptosermum*, *Weinmannia*, Honeydrew, *Thymus*, *Metrosideros*, *Echium*, *Ixerba*, *Knighita*, *Carduus*, *Trifolium*		NZ	ICP-OES	GO	Al, As, B, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, P, Pb, S, Zn
Oroian et al. (2015)	*Acacia*, *Tilia*, *Sunflower*, Multifloral	52	RO	ICP-MS	GO and BO	As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Zn
Ahmed et al. (2016)	*Jujube*, Multifloral, *Acacia*	16	PK, DE, FR	AAS	GO	Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, ONi, Pb, Zn
Louppis et al. (2017)	*Citrus*, *Pinus*, *Abies*, *Thymus*	207	GR	ICP-AES	BO	Ag, Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, Hg, Mg, Mn, Mo, Ni, Pb, Sb, Se, Si, Ti, Tl, V and Zn
Bontempo et al. (2017)	*Acacia*, Multifloral, Honeydew, *Rhododendron*, Chestnut, *Eucalyptus*	265	IT	ICP-OES	GO and BO	Al, B, Ba, Ca, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Rb, Sr, Zn
Bilandžić et al. (2017)	Multifloral, *Erica*, *Arbutus unedo*, *Canulla*, Honeydew, *Citrus*	26	HR	ICP-MS	BO	Al, CA, Cu, Fe, K, Mg, Mn, Na, Zn, Ag, As, Ba, Be, Cd, Co, Cr, Mo, Ni, Se, Sb, V, U, Th
Di Rosa et al. (2019)	*Eucalyptus*, *Castanea*, *Hedysarum*, *Citrus*	28	IT	ICP-MS	BO	Na, K, Ca, Mg, Fe, Mn, Zn
Lanjwani & Channa (2019)	Multifloral	15	PK	F-AAS	GO	Fe, Zn, Co, Cu, Mn, Cr, Ni, Pb, Cd, Na, K, Ca, Mg
Squadrone et al. (2020)	Multifloral	75	BALKANS, KZ, SOUTH AMERICA, TZ	ICP-MS	GO and BO	Ag, Al, As, Be, Bi, Cd, Co, Cr, Cu, Fe, Ga, In, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sn, Tl, U, V, Zn

F-AAS: Flame Atomic Absorption Spectroscopy; GF-AAS: Graphite Furnace Atomic Absorption Spectrometry; ICP-OES: inductively coupled plasma optical emission spectrometer; ICP-MS: Inductively coupled plasma mass spectrometry; AAS: Atomic Absorption Spectroscopy: ICP-AES Inductively Coupled Plasma Atomic Emission Spectroscopy

The ISO 3166-1 alpha-2 standard code was used to indicate the provenance of the samples

Table 2.

Studies in which spectroscopic techniques are used for the analysis of honey in order to verify its correct botanical (BO) and geographical (GO) origin and any fraudulent additions

Reference	Type of honey	Number of samples	From	Technique	Statistic appr.	Aim of the work	Results
Schellen-berg et al. (2010)	Honeydew, unfloral	516	IT, PT, DE, ES, FR, GR, IS, IE, PL, GB, AT, DK.	IRMS		GO	70% of correct classified samples
Zheng et al. (2016)	Unfloral, multifloral	75	CN	1H NMR	PCA, OPLS-DA	GO and BO	Discrimination of sample variations by analysing saccharides and all types of amino acids and organic carboxylic acids
Minaei et al. (2017)	8 *Robinia pseudoacacia* L., 10 *Fagopyrum esculentum* Moench, 9 *Calluna vulgaris* L., 15 *Tilia* spp., 10 *Brassica napus* L.	52	PL	VIS-NIR (range 400-1000 nm)	PCA	BO	Three machine learning algorithms (radial basis function, support vector machine and random forest of which the best achieved 94% accuracy
Lastra-Mejías et al. (2018)	Unfloral, multifloral	10	ES, MX.	Fluorescence spectroscopy	PLS-2	BO	Simple approach, 95% accuracy
Chen et al. (2019)	Longan, non-longan	163	TH, TW	IRMS	LSD	Anti -fraud	7% adulterated samples, Differences between Taiwan and Thailand longan honeys
Suhandy & Yulia (2021)	Durian, multifloral	400	ID	UV-VIS spectrometer	PCA	BO	100% correct discrimination
Stefas et al. (2021)	Unfloral, multifloral	45		LIBS	LDA	Anti -fraud	2 predictive models with classification accuracy greater than 90%

IRMS: isotope ratio mass spectrometry; ^{1}H NMR: Proton nuclear magnetic resonance; VIS-NIR: visibly-near-infrared spectroscopy; UV-VIS: visibly-ultraviolet spectroscopy; LIBS: Laser Induced Breakdown Spectroscopy

The ISO 3166-1 alpha-2 standard code was used to indicate the provenance of the samples

by exogenous sugar sources (Zhou et al., 2018; Magdas et al., 2021). Monocotyledonous (some C4 plants) and dicotyledonous (C3) plants have distinctive ^{13}C to ^{12}C carbon isotope ratios that are the result of different photosynthetic cycles. Since most melliferous plants are C3 plants, and both cane and corn, the two main sources of industrial sugar syrups, are C4 plants, authentic honeys are expected to have the characteristic properties of C3 plants rather than those of C4 plants (Souza-Kruliski et al., 2010; Chesson et al., 2013; Soares et al., 2017). Some limitations of this technique are that not all adulterations are easy to identify. In fact, sometimes honey is adulterated using C3 sugars derived from plants such as sugar beet, and in other cases the adulterants in honey are difficult to detect due to the development of new more sophisticated practices (Soares et al., 2017; Tosun, 2013; Wu et al., 2017; Zhou et al., 2018).

Notwithstanding, some authors have proven that IRMS can be applied to the investigation of the honey's geographical and botanical origins. In fact, some isotopic ratios, especially those of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$), are highly influenced by the botanical origins of honey (Dinca et al., 2015; Bontempo et al., 2017) but also by the length of the growing season of the plants, the sunny days and the mean temperature (Schellenberg et al., 2010). These factors inevitably lead to difference between the isotopic ratios found in a temperate climate zone and the Mediterranean one, hence allowing for geographical discrimination. Bontempo et al. (2017) reported that the botanical origin of honey affects isotopic characteristics by using the IRMS in synergy with the Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES), for the mineral profiling, analysing 265 samples (112 multifloral, 60 acacia, 37 chestnut, 18 citrus, 15 rhododendron, 13 eucalyptus and 10 honeydew). Berriel et al. (2019) studied carbon isotopic signature ($\delta^{13}C$) in the protein fraction of forty-seven honey samples from pastures and forty-two from eucalyptus. A logistic regression model was thus developed through which 90% of the samples were correctly assigned, using as variables the $\delta^{13}C$ data of protein fraction and the isotopic index of honey. She et al. (2019) evaluated the geographical origin of seventy-one Chinese acacia honey from six different regions by analyzing both the chemico-physical parameters of oligosaccharides using gas chromatography-mass spectrometry, polyphenols with HPLC-MS and carbon isotope using Liquid Chromatography Isotope Ratio Mass Spectrometry. The combination of the thirty-one parameters analysed made it possible to obtain a correct classification rate of 94.12%. Dong et al. (2017) analysed forty-five commercial honey samples of fifteen different botanical origins from seven Chinese regions with the use of IRMS together with element analyzer (EA) and liquid chromatography (LC). Based on the analysis of these parameters, more than 30% of the analyzed samples were considered pure.

Omics approaches

In the last decade, studies have been carried out on the omics sciences, with particular regard to the metabolomic and genomic approaches. Metabolomic methods concern the analysis of metabolites, targeted and untargeted, quantitatively or qualitatively (Cevallos-Cevallos et al., 2009; Patti et al., 2012; Cubero-Leon et al., 2014; Cajka et al., 2016; Koulis et al., 2021), and their usefulness has been proven in food traceability and against frauds. The analysis of the amino acid (AA) profile of honey is one of the techniques, and in fact the variation of the amino acid profile could be used to determine a honey's geographical and botanical source (Anklam, 1998). Hermosín et al. (2003) analysed the amino acids occuring in forty-eight Spanish honey samples of different botanical origins (lavender, eucalyptus, orange blossom, rosemary, thyme, heather, holm oak, forest, oak, multifloral), and the results showed that some unifloral honeys were effectively differentiable, and lavender had the highest tyrosine concentration. Cotte et al. (2004) analysed by HPLC the AA profiles of 280 samples of French honeys from seven different botanical origins (acacia, chestnut, rape, lavender, fir, linden, and

Table 3.

Studies in which genomic techniques have been adopted for the analysis of honey, with the respective results obtained

Reference	Type of honey	Provenance	Sequencing	Number of samples	Advantages	Problems
Bruni et al. (2015)	Multifloral	IT	RbcL and trnH-psbA plastid	4	high performances, possibility of replacing mellissopalinological analysis in the future	Need improving plant database
Soares et al. (2015)	Unifloral/ multifloral	PT	-	4	Solidity, sensitivity, specificity and less margin of human subjectivity in the analysis of the results	-
Laha et al. (2017)	Multifloral	IN	NGS	20		Need improving plant database
Prosser & Hebert (2017)	Mixed	CA/MX	Ion Torrent	7	Successful in 5/7 of the samples	Risk of being acted upon by small fraudulent additions of pollen
Utzeri et al. (2018)	Unifloral/ multifloral/ honeydrew	MIXED	TrnL-UAA plastid	9	Low cost in prospective and more efficacy	-
Saravanan et al. (2019)	-	IN	3730XL automated DNA Sequencer	29	Quicker, easier than mellissopalinological analysis	Need improving plant barcode database

RbcL and trnH-psbA: plastid regions as barcode markers; NGS: Next generation sequencing; TrnL-UAA plastid region; 3730XL: DNA ANALYZER (THERMO FISHER)

The ISO 3166-1 alpha-2 standard code was used to indicate the provenance of the samples

sunflower), but the obtained data discriminated only the lavender samples for the selected amino acids histidine, threonine, phenylalanine, proline, and their total content. Janiszewska et al. (2012) investigated the composition of free amino acids, with HPLC, in eighteen Polish honeys of buckwheat, raspberry, acacia, heather, goldenrod and honeydew. Although they identified the honeys using the amino acid profile alone, the main differences were noted in the high amount of proline in the heather honey (387.88 mg/kg) and of aspartic acid in the raspberry and buckwheat honey samples (22.41 mg/kg and 30.07 mg/kg, respectively).

The Nuclear Magnetic Resonance Spectroscopy (NMR) and its metabolomic applications allows for the identification, even simultaneous, of chemical compounds occurring in food (Siddiqui et al., 2017; Consonni & Cagliani, 2019). Schievano et al. (2019) applied NMR with honey organic extracts to distinguish the geographical differences from 234 samples of acacia honey

from Italy, eastern Europe and Hungary, which resulted in 100% accurate classification rate. Donarski et al. (2008) used NMR and multivariate data analysis to characterise 182 Corsican and non-Corsican honeys: strawberry, chestnut, autumn scrubland, honeydew scrubland, spring scrubland, summer scrubland, orange clementine and multifloral. In particular, they studied four biomarkers specific for authentic Corsican honey and were able to identify trigonelline ($C_7H_7NO_2$) as a selective biomarker for non-Corsican honeys. Trigonelline, which it may be indicative for dry and saline habitats. The overall classification rates for the discrimination of Corsican and non-Corsican honeys were 75.8%, 94.5%, 96.2%, respectively by using Partial Least Squares-Linear Discriminant Analysis (PLS-LDA), two-stage Gaussian Process, and Partial Least Squares Gaussian Process (PLS-GP). Zhou et al. (2014) tested a method of liquid chromatography-diode array detection-tandem mass spectrometry (HPLC-DAD-MS/MS) for tracing the botanical origin of honeys. Kaempferol ($C_{15}H_{10}O_6$), morin ($C_{15}H_{10}O_7$) and ferulic acid ($C_{10}H_{10}O_4$) were used as floral markers to distinguish chaste and rapeseed honey. A total of 187 honey samples were identified and separated using the chromatographic fingerprint in use with the similarity analysis (Similarity Evaluation System for Chromatographic Fingerprinting of Traditional Chinese Medicine, Version 2004A). DNA markers from pollens have also been used in recent years to identify accurately plant nuclear material found in honey.

However, the low amounts of pollen DNA in honey and substances interfering with PCR amplification (*e.g.*, organic acids, polyphenols, pigments and enzymes) require effective DNA-extraction protocols including sample preparation and elimination of interfering substances (Soares et al., 2015). This technique's greatest advantage at the control level is that, differently from the melissopalynological approach, even though pollen grains can be filtered away the pollen-free DNA in the liquid phase remains detectable as a tracer of the botanical origin of the honey sample (Haynes et al., 2019). This methodology was used by Utzeri et al. (2018) (Tab. 3) in the botanical analysis of nine honeys of intercontinental origin. The method was based on DNA sequencing and resulted effective with the use of the plastid trnL-UAA universal marker for barcoding identification of plant DNA occurring in honey, and the discriminatory power of this fragment was enough to mark the botanical signature of all analysed honey.

This method is also compatible with other methods (*e.g.*, mellissopalynology). Laha et al. (2017) (Tab. 3) used Next Generation Sequencing (NGS) with a single target locus. This method proved to be less laborious but at the same time less accurate with respect to the classification power than the melissopalynological analysis. However, a palynological and plant database of the study area was considered as necessary without which it is not possible to compare and make reliable the results obtained by NGS (Laha et al., 2017; Liu et al., 2022). To increase the efficiency of the NGS technique, it will be necessary to use a multi-loci apporach and to improve the knowledge of of plants and pollens species occurring in the area of interest (Laha et al., 2017; Liu et al., 2022). Bruni et al., 2015 (Tab. 3) studied the potential utilization of the DNA barcoding on four honey samples. They underlined the necessity of having a detailed list of plants of the study area that have to be characterized at a molecular level, against which comparing the data collected with the analysis of a given sample.

IoT and blockchain for honey traceability

Agricultural traceability requires a large volume of data that must be collected across the supply chain. Early tracking and tracing systems used workers to record information in the field and then manually transferred it to forms or a computer system with the risk of information loss or errors. In recent decades, rapid development in automated processes, as well as in communication technologies, has given rise to the so-called Internet of Things (IoT) paradigm. This rapid evolution of IoT and

sensor technology favours the data collection procedure by offering such fast and reliable methods as barcodes, QR codes, RFID and wireless sensor networks (WSN) (Lin et al., 2018; Demestichas et al., 2020). Among them, blockchain technology offers the possibility to create a smarter and more secure supply chain. Traceability information on product origin and possible allergens or added substances, which must be stored in a safe and immutable way, can be established and shared through a collaborative blockchain network among farmers, producers and distributors.

While scientific research on blockchain and traceability in agricultural supply chains is growing rapidly, the same is not reflected in the availability of large-scale commercial applications (Caro et al., 2018). One of the most famous relevant applications is the IBM Food Trust, that was tested with Walmart, a collaboration to trace the origin of Chinese mango and pork products (Kamath, 2018). As a further example, identifying the origin of the mango could take seven days without the Food Trust system, while it takes about 2.2 s with the Food Trust (Demestichas et al., 2020; Kamath, 2018). Also, AgriOpenData aims to trace in a transparent, safe and public manner, the entire agri-food chain and the processing of agricultural products, allowing them to certify their quality (Demestichas et al., 2020). Blockchains increase the transparency of the food supply chain as well as consumer confidence but carry a huge energy and financial cost. It is mandatory for companies to invest much money and time to train all the personnel involved and also to obtain the required equipment, so a cost-benefit analysis is essential (Galvez et al., 2018; Behnke & Janssen, 2020). Concerning honey and bee products, Rünzel et al. (2021) have hypothesized that the blockchain could be used for an open traceability system that includes predictive and verification models on honey yield, an intelligent distribution system based on blockchain technology, the verification of the pollen signature with machine learning algorithms and an information portal accessible to the final customer, all designed for sustainable development and be built and used all over the world. Srivastava & Dashora (2022) explained that the use of this technology is organized for the traceability of honey starting from production, processing, packaging, transport and consumption. Oracle works with IoT technology via the World Hive Network for the fraudulent honey market, and the technology helps to define which beehives were involved in the production of a single honey lot. The information is collected by the Oracle Blockchain platform (Mary Hall, n.d.).

DISCUSSION

To date, the melissopalynological analysis remains the method more commonly used for investigating the geographical and botanical origin of honey, (Persano Oddo & Bogdanov, 2004; Von Der Ohe et al., 2004). Melissopalynological analysis has also been standardized and is performed according to standard protocols (*e.g.*, the ISO method N. TC34/SC19). Furthermore, it is used as a reference method for most of the trials presented in this review (Persano Oddo & Bogdanov, 2004; Von Der Ohe et al., 2004; Corvucci et al., 2015; Reyes et al., 2019). However, in the last decade, as preconized by Anklam (1998), the melissopalynological analysis has been progressively integrated by other methods. In fact, most articles have not analyzed only one aspect of honey and instead at least two aspects, because on the basis of the bibliographic research the single parameter determined in honey is evidently insufficient for tracking its origin.

This is why a series of methods are used in combination with one another, and the solutions with the most reliable results combine data deriving from different measurements (multicomponent analysis), especially with current statistical data evaluation techniques (Anklam, 1998; Schiassi et al., 2021). The new frontier concerns those methodologies that aim to reduce analysis times, experience and staff, automate processes and have more accurate results (Consonni & Cagliani, 2019). Genomics as well as the others

"omics" sciences seem to have interesting perspectives, but preliminary digitization and cataloguing of the plants are necessary. Currently, these methodologies appear to be limited and dependent on other verification methods including melissopalynology, losing the advantages of speed, accuracy, simplicity and cost. In the future, in terms of economic, time and cost savings, analysis will be needed that deals with the control of the adulteration of honey, its origin, authenticity and typicity. Finally, the standardization of the main analysis methods would lead to data homogeneity, more effective product traceability and simplicity in data interpretation.

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