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Chapter

Gas Chromatography in Food Authentication

Kristian Pastor, Marijana Ačanski and Djura Vujić

Abstract

Authentication of food products and food fraud detection are of great importance in the modern society. The application of sophisticated instrumentation, such as gas chromatography (GC), with this aim helps to improve the protection of consumers. Gas chromatography mostly combined with the most powerful detector, a mass spectrometer (MS), and various multivariate data processing tools is in the last few decades being increasingly applied in authenticity and traceability of a wide spectra of food products. These include animal and plant products, beverages and honey. This chapter gives an overview of the most recent applications of gas chromatography technique in determining food authenticity, described in scientific literature.

Keywords: food products, authenticity, food fraud, consumer protection, gas chromatography

1. Introduction

The adulteration practices on food product market are known since ancient times [1, 2]. It was found that, during the nineteenth century, gypsum and alum were added to bakery flour to increase weight, strychnine was added to beer to increase bitterness, and salts of copper, lead, and mercury were added to sweets in order to get a beautiful color and gloss [3–5]. Consumer interest in safety, authenticity and quality of food products is constantly increasing [6]. Authenticity is related to truthfulness, so a food product can be said to be authentic if it was not subject to any fraud [7]. European and global food policies require food on the market to be authentic. This means that the label on the product must match its actual composition, origin (geographical, botanical and genetic) and the process of production (conventional, organic and traditional) [2, 8, 9]. With globalization, market development and rapid distribution systems, as well as expanding the range of food items, counterfeiting and contamination of food products, are becoming international in character, and the possible consequences are far-reaching [2, 4, 9–11]. The most common type of adulteration—economically motivated food adulteration—is defined as *a misleading and deliberate substitution or addition of certain ingredients to a food product in order to increase the apparent value of the product or reduce the cost of its production, with the consequence of a certain economic gain* [4, 5]. Depending on the nature of an added substituent, the obtained adulterated products may pose a potential danger to the health of the consumer. In this way, the determination of authenticity in the food industry is gaining health and safety aspects, in addition to the economic one [6, 8, 12]. With all this in mind, global

policies require strict monitoring and quality control of food. Therefore, there is a clear tendency toward the development of new techniques and analytical methods that would enable this goal to be achieved. Traditional and standard methods of analysis are still very commonly used. Due to lower costs and/or faster analytical protocols, there is an urge for new authentication methodologies that would be complementary or even replace existing ones [8, 9]. This trend is stimulated by consumers, regulatory bodies and the food industry itself. Contemporary authentication analysis is based on the detection and measurement of various chemical parameters that would have the potential of discrimination factors of the investigated food samples [2, 9]. According to Danezis et al. [2], the first 10 countries in the world that are most intensively engaged with food authentication, in addition to the United States and China, are members of the European Union. These countries actively subsidize and encourage the development of this scientific area [2]. The European Commission regulations and directives testify about the rights of consumers to get the genuine information about food products that they buy [13–15]. These regulations aim to prevent (i) fraud and misleading actions, (ii) adulteration of food products and (iii) any other fraudulent procedures. An example of a very frequent way of food adulteration is the substitution of some ingredient in a food item with a similar and cheaper one, so that the consumer cannot recognize this procedure [1, 6, 8, 16]. According to the literature data, food products mostly subjected to adulterations include cereal and bakery products, edible oils and fats, milk and dairy products, meat and fish, fruit and fruit juices, honey, coffee, tea, wine, organic products and many others [9, 11]. Basically, there are three analytical approaches to determine the authenticity of food products: (i) chemical approach, determination of the composition and content of various chemical components in food; (ii) biomolecular approach, analysis of DNA and proteins; and (iii) isotopic approach, determination of the composition of stable isotopes of certain atoms [7]. Chromatographic techniques are the most common choice in the analysis of the authenticity of most food items [2, 9]. This is partly because techniques, such as chromatography, can be applied both for the purpose of detecting adulterations and for the purpose of determining authenticity [7]. In addition, the analytical capability of mass spectrometry, often used in conjunction with chromatographic techniques, allows the characterization of a wide range of components in very complex systems [17]. Some authors believe that the future of determining food authenticity is reflected in the synergistic fusion of various complementary instrumental techniques and the processing of such a complex block of enormous amounts of data using modern techniques of multivariate analysis [6]. Since 2001, a large number of scientific articles have appeared, relating to food authentication using new or existing analytical techniques in combination with multivariate data analysis. However, it has to be noted that the adulteration practices are also very contemporary and in constant development, with constant interest in surpassing the power of the established analytical methods of their discovery [14].

This chapter represents a thorough overview of the analytical methods employing a GC technique that are dealing with authentication and adulteration detection of various types of foodstuffs. The methods described are published in scientific literature in the last two decades.

2. Authentication and adulteration detection in various food products

2.1 Olive oil and other edible vegetable oils

Edible vegetable oils represent a matrix which is usually analyzed with the application of GC. That is why there are a large number of papers dealing with

authentication and adulteration detection in this type of food, using GC. Among them, extra virgin and virgin olive oils are definitely the most investigated. The suggested analytical methods are focused on the determination of constituents in oil mixtures of high prices and quality, the discrimination of extra virgin olive oils from defected oils, the possibilities of the authentication of various edible oils and fats and the determination of geographical origin. Triacylglycerol composition, fatty acid composition, $^{13}C/^{12}C$ and $^{2}H/^{1}H$ ratios and enantiomeric distributions of certain compounds, and just in some cases volatile organics and phenolic compounds, are usually considered as discrimination factors. Considering that this kind of analysis provides a large amount of data, the recently published papers are almost exclusively coupling GC with various unsupervised and supervised techniques of multivariate chemometric data analysis. Among unsupervised principal component analysis is definitely the mostly used, and among supervised techniques and machine learning algorithms, there are many different described: LDA and SLDA, PLS-DA, OPLS-DA, SIMCA, ANN-MLP, R-SVM and OC-SVM and some other. **Table 1** lists chronological literature data on authentication and adulteration detection procedures of the most commonly investigated olive oil, and also edible oils of other plant species, and some examples of animal fats.

2.2 Honey and other bee products

The authenticity of honey and other bee products has two aspects. Authenticity in respect of production, i.e., to prevent adulteration by the addition of other food ingredients (various types of sugar syrups), and authenticity of botanical and geographical origin. The GC method for determining the addition of sugar syrups relies on carbohydrate profiling in combination with classical statistical procedures for data processing. However, methods for authentication of geographical and botanical origin of honey samples usually employ more complex sample preparations, such as solid-phase microextraction in a headspace mode, and more sophisticated instrumentation, such as multidimensional GC. These methods mostly rely on the analysis of volatile organic compounds and also usually involve the application of multivariate chemometric tools for data analysis—unsupervised and supervised pattern recognition techniques. Unsupervised techniques, PCA and HCA, are more commonly used, but some studies also report the application of supervised tools: LDA and SLDA, OPLS-DA, SIMCA and ANN-MLP. **Table 2** lists examples from literature data on authentication and adulteration detection procedures of honey and other bee products, such as beeswax, propolis and royal jelly.

2.3 Milk and dairy products

Authenticity of milk and dairy products, such as cheese and fermented milk, using GC, is usually based on the determination of fat content of samples: triacylglycerols and fatty acids. Therefore, it is usually enough to combine GC with FID, to perform a successful analysis. In some particular cases, MS or olfactometry is used (if the analytical method is based on determining volatile profiles of the samples). Methods described in the literature rarely use chemometric data analysis, in some cases PCA, LDA and PLS-DA, but rather rely on the application of classical statistics. Papers describing the authentication of milk and dairy products usually deal with discriminating organic from conventionally produced ones, discriminating samples according to geographical origin and according to the animal breed they are produced of. **Table 3** shows literature examples of authentication and adulteration detection practices in milk and dairy products, such as cheese.

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Olive oil			
Discrimination of "Ligurian" from "non-Ligurian" olive oils	HS-SPME/GC-ITMS	LDA, ANN-MLP	[18]
Differentiation of monovarietal olive oils according to olive variety	HS-SPME/ GC × GC-TOF-MS HT-GC-ITMS HS-SPME/GC-MS GC-TOF-MS	PCA PCA, HCA, PLS-DA — PLS-DA	[19] [20] [21] [22]
Differentiation of extra virgin and virgin olive oils according to geographical origin (various regions in Spain, Italy)	HS-SPME/GC-MS HS-SPME/GC-MS GC-C/P-IRMS	SLDA — —	[23] [21] [24]
Discrimination of extra virgin olive oils from defected oils	HS-SPME/ GC × GC-MS	PCA, PLS-DA	[25]
Detection of extra virgin olive oil, virgin olive oil and olive oil adulteration (with various types of edible oils)	LC-GC-ITMS LC-chiral-GC-ITMS GC-MS SPME/ GC × GC-MS GC-MS SPME/GC-MS GC-FID GC-MS GC-MS	— — PCA — SIMCA, kNN, PLSR — PCA, PLS PCA, TFA, SIMCA, PLS OC-SVM	[26] [27] [28] [29] [30] [31] [32] [33] [34]
Other edible oils			
Discrimination of various vegetable oils according to botanical origin (sunflower, corn, sesame, soybean, olive, rapeseed, camellia, peanut, canola, palm, rice bran, coconut, grapeseed, hazelnut, walnut, apricot seed, red pepper seed, prikachberry, pumpkin)	GC-C-IRMS GC-C-IRMS HT-GC-FID GC-FID GC-FID HT-GC-MS GC-MS GC-MS GC-MS GC-MS	CDA — PCA PCA, KNN, CNN PCA, PLS SIMCA, PLS, GA-PLS PCA, PLS-DA, OPLS-DA PCA, HCA, RF LDA, GA-SVM	[35] [36] [37] [38] [32] [39] [40] [41] [42] [43]
Differentiation of almond oils according to almond variety	HS-SPME/GC-MS	SLDA	[44]
Detection of corn oil adulteration	GC-C-IRMS	—	[45]
Detection of flaxseed oil adulteration	GC-MS	PCA, R-SVM	[46]
Detection of sesame oil adulteration	GC-FID GC-FID GC-MS	— SVM OC-SVM	[47] [48] [49]
Authenticity and geographical origin of pumpkin seed oil	GC-FID GC-C-IRMS	PCA, RDA	[50]
Fats			
Authenticity of cocoa butter	LC-GC-MS	—	[51]
Discrimination of various edible oils and fats (pig, mutton, beef and chicken)	GC-MS	PCA, PLS-DA, OPLS-DA	[40]

Table 1.
Literature examples of authentication and adulteration detection procedures of olive oil and other edible oils and fats.

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Honey			
Detection of the addition of sugar syrups to honey (high-fructose corn syrup and inverted syrup)	GC-FID GC-FID/MS GC-FID	PCA — —	[52] [53] [54]
Differentiation of four types of multifloral Portuguese honeys (produced in Madeira Island)	HS-SPME/GC-MS	PCA, SLDA	[55]
Authenticity of "Corsica" honey	HS-SPME/GC × GC-TOF-MS HS-SPME/GC × GC-TOF-MS	PCA, ANN-MPL LDA, SIMCA, SVM, DPLS	[56] [57]
Detection of honey adulteration with high-fructose inulin syrups	GC-MS	—	[58]
Authenticity of thistle honey	HD-SPME/GC-MS	—	[59]
Authenticity of botanical origin of unifloral chestnut (*Castanea sativa* L.) and eucalyptus (*Eucalyptus globulus* Labill.) honeys	GC-MS	—	[60]
Differentiation between lemon blossom honey (*Citrus limon*) and orange blossom honey (*Citrus* spp.)	GC-MS	PCA	[61]
Geographical origin identification of honey (samples from various regions of Greece; samples from various Mediterranean countries: Egypt, Greece, Morocco, Spain)	HS-GC-MS HS-SPME/GC-MS HS-GC-MS HS-SPME/GC-Q-TOF-MS HS-SPME/GC-MS SPME-GC/MS	— HCA, SLDA, kNN OPLS-DA, SIMCA, OPLS-HCA PCA LDA LDA	[62] [63] [64] [65] [66] [67]
Differentiation of honeys according to botanical origin: heather, raspberry, rape, alder buckthorn, lime, rosemary, chestnut, sunflower, acacia, thyme, orange, linden, amaranth, honeydew, citrus, *Gossypium*, rhododendron, alfalfa, white clover, carob, calden	HS-GC-MS SPME/GC-MS HS-GC-MS SPME/chiral-GC × GC-MS SPME/GC-MS/O SPME-GC/MS	— LDA OPLS-DA, SIMCA, OPLS-HCA — — AHC, CA PCA, HCA	[62] [68] [64] [69] [70] [71]
Establishment of orange honey authenticity	SPME/GC-MS	—	[72]
Other bee products			
Authenticity of royal jelly; detection of the addition of sugar syrups	HR-GC	—	[73]
Characterization of traditional plant syrups from Spain, namely, palm honey (miel de palma), must syrup (arrope) and sugarcane honey (miel de caña)	GC-MS	—	[74]
Detection of adulterated beeswax from *Apis mellifera* L.	HT-GC-FID/MS HT-GC-FID/MS	— HCA, PCA, LDA	[75] [76]
Geographical origin identification of propolis	HS/GC-MS/O	PCA	[77]
Establishment of sugarcane honey authenticity	HS-SPME/GC-MS	PCA, LDA	[78]

Table 2.
Literature examples of authentication and adulteration detection procedures of honey and other bee products.

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Milk			
Authenticity of goat milk	GC-FID	—	[79]
Differentiation between cow milk produced in the lowlands, mountains and highlands of Switzerland	HR-GC-FID	—	[80]
Authenticity of milk fat: detection of foreign fat in milk fat (such as pork lard, bovine tallow, fish oil, peanut oil, corn oil, olive oil, soy oil, sunflower oil, coconut fat)	GC-FID UFM-GC-FID GC-FID GC-FID GC-FID	— — LDA — —	[81] [82] [83] [84] [85]
Differentiation of milk produced under conventional and organic management	GC-FID GC-FID GC-MS	— — —	[86] [87] [88]
Differentiation of cow, goat, sheep, water buffalo, donkey, horse and camel milk	GC-FID	PCA	[89]
Determining the origin of milk samples: hay milk vs. conventional (silage) milk	GC-FID	PCA, PLS-DA	[90]
Dairy products			
Geographic origin of Emmental cheese	GC-FID HS-GC-FID/MS	— PCA	[91] [92]
Differentiation of Grana Padano, Parmigiano-Reggiano and Grana Trentino cheeses	GC-O	PCA	[93]
Examining foreign fat origin in cheese from cow milk fat	GC-FID	—	[94]
Differentiation between certified organic and conventional probiotic fermented milks	GC-FID	—	[95]
Quality control for Parmigiano-Reggiano cheese	GC-MS	PCA	[96]

Table 3.
Literature examples of authentication and adulteration detection procedures of milk and various dairy products.

2.4 Fruits and fruit-made beverages

Most of the papers dealing with fruit authenticity testing using GC are focused on determining discriminating factors that will enable discrimination of varieties of certain fruit species. These factors are mostly constituted of free and bound volatile compounds belonging to different chemical groups, namely, linear and branched esters, terpenes, alcohols and others. The paper published by Kurz et al. [97] is an exception, which is dealing with the analysis of neutral sugars of cell wall polysaccharide profiles of apricots, peaches and pumpkins using GC-FID. In some cases GC was also combined with other analytical techniques, thus enabling the wider spectra of chemical species to be included in the analysis, such as LC, in order to include nonvolatile carbohydrates, fatty acids and organic acids. The obtained data were mainly processed using multivariate data analysis techniques, such as HCA, PCA, PLS-DA, LDA and OPLS-DA. Older investigations usually do not include multivariate data analysis. Schmarr and Bernhardt [98] used image processing techniques in order to process the data obtained after comprehensive two-dimensional GC

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Fruits			
Differentiation of blackcurrant (*Ribes nigrum* L.) berries	SPME/GC-FID	—	[97]
Differentiation of apricots (*Prunus armeniaca* L.), peaches (*Prunus persica* L.) and pumpkins (*Cucurbita* sp.)	GC-FID	—	[99]
Differentiation between *Passiflora* fruit species	HS-SPME/GC-MS	PCA	[100]
Discrimination of red grape varieties of southern Italy (Aglianico, Uva di Troia, Negroamaro, Primitivo)	GC-MS	PCA	[101]
Differentiation of apples, pears and quince fruit	HS-SPME/ GC × GC	—	[98]
Classification of apple varieties (Golden Delicious, Granny Smith, Pinova and Stark Delicious)	HS-SPME/GC-TOF-MS	PCA, PLS-DA	[102]
Differentiation between grape varieties: *Vitis vinifera*, *Vitis cinerea* and interspecific crosses	GC-MS	HCA	[103]
Differentiation of Chinese bayberry cultivars (*Myrica rubra*)	HS-SPME/GC-MS HS-SPME/GC-MS/O	PCA —	[104] [105]
Discrimination of nine passion fruits: yellow, purple, lemon, orange, pineapple, peach, melon, banana and tomato	HS-SPME/GC-MS	PCA, PLS-DA	[106]
Differentiation between Tanzanian grown fruits: mango, pineapple, jackfruit, baobab and tamarind	GC-MS	HCA, PCA	[107]
Discrimination of *Eugenia uniflora* L. biotypes	HS-SPME/GC-MS	PCA, HCA	[108]
Differentiation between date palm fruit (*Phoenix dactylifera* L.) varieties from Egypt	SPME/GC-MS	PCA, HCA, OPLS-DA	[109]
Characterization of organic oranges (*Citrus sinensis* L. Osbeck)	HS-SPM/GC-MS	PLS-DA	[110]
Differentiation of sun-dried raisins made from different grape varieties	HS-SPME/GC-TOF-MS	—	[111]
Differentiation between citrus species: mandarin, sweet orange, sour orange, papeda, pummelo, lemon, *Fortunella* Swingle	GC-MS	HCA	[112]
Differentiation of apple cultivars from geographical origin and growing conditions (organic and conventional)	HS-SPME/GC-MS	PLS-DA	[113]
Differentiation of Chinese Jujube varieties	HS-SPME/GC-MS	HCA	[114]
Fruit beverages			
Detecting adulteration of blackcurrant juice	GC-FID	—	[115]
Authentication of apple and orange juice	GC-FID	—	[116]
Detecting the addition of aromas to fruit beverages	SPME/chiral-GC-MS	—	[117]
Citrus juice classification (lemon, grapefruit, mandarin, orange, lime)	HS-SPME/GC-MS	LDA	[118]
Assessment of premium organic orange juices authenticity	HS-SPME/GC-MS	PLS-DA	[119]

Table 4.
Literature examples of authentication and adulteration detection procedures of various fruits and fruit juices.

analysis. **Table 4** chronologically lists some literature examples on authentication and adulteration procedures of various fruit species and fruit-made juices.

2.5 Cereals and bakery products

Cereals, pseudocereals, flours and bread, as mostly used bakery products in human nutrition, are usually differentiated according to varietal, botanical or geographical origin by combining GC analysis with chemometric processing of the obtained data. The chemical compounds that have the role of discriminating factors usually involve small molecules, such as simple soluble sugars and free fatty acids. Chemometric methods involve most often exploratory data analysis techniques, such as PCA, PCO and HCA, but in some cases also classification methods of LDA and QDA were applied to measure the classification and prediction abilities. **Table 5** chronologically lists some literature examples of authentication and adulteration detection practices of cereals, flour and the most commonly used bakery product in human nutrition-bread.

2.6 Meat, fish and seafood

The studies of authenticity of seafood and meat products using a GC technique usually focus on the determination of freshness of a seafood or meat product. Chemometric techniques, such as PCA, were able to successfully discriminate between fresh samples, deteriorated samples and gradually decaying samples of seafood, and ANN were employed in order to classify samples of fresh meat, frozen-thawed meat and spoiled meat. The PCA of gas chemometric fingerprints was able to show separation not only between oyster species but also between oysters originating from different cultivation areas, as well as oysters harvested at

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Cereals			
Differentiation between *Triticum durum* and *Triticum aestivum*	GC-FID	PCA, LDA, QDA	[120]
Differentiation between hexaploid (*T. aestivum*, *T. spelta*) and tetraploid (*T. durum*, *T. dicoccon*) wheats	GC-MS	—	[121]
Classifications of cereals (wheat and corn) used in DDGS material by geographical and botanical origin	GC-FID	PLS-DA	[122]
Flour			
Differentiation of corn and small grain flour (wheat, rye, triticale, barley, oats)	GC-MS	HCA, PCO HCA, PCA	[123] [124]
Differentiation of corn and oat flour, from other small grains (wheat, barley, triticale, rye)	GC-MS	HCA, PCO	[125]
Differentiation of flours of corn, spelt, buckwheat, amaranth and small grains (wheat, rye, triticale, oats, barley)	GC-MS	HCA, PCA	[126]
Bakery products			
The content of buckwheat flour in wheat bread	GC-MS	HCA HCA, PCA	[127] [128]

Table 5.
Literature examples of authentication and adulteration detection procedures of cereals, flour and bakery products.

different time intervals. There was only one paper found in the literature that deals with differentiation of meat according to the breed origin. The PCA was successfully applied to discriminate between samples of pork, chicken, beef and mutton meat. **Table 6** represents a chronological list of examples of authentication and adulteration detection procedures of various types of meat, fish and seafood.

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Fish and seafood			
Differentiation between fresh and deteriorated oyster *Crassostrea gigas*	HS-SPME/GC-MS	PCA	[129]
Differentiation between fresh and frozen-thawed cultured gilthead sea bream fish (*Sparus aurata*)	SPME/GC-MS	—	[130]
Razor clam (*Sinonovacula constricta* Lamarck), redspot swimming crab (*Portunus sanguinolentus* Herbst) and prawn (*Penaeus japonicus* (Bate; Kuruma prawn)) freshness determination	HS-SPME/GC-MS	PCA	[131]
Differentiation of European flat oyster (*Ostrea edulis*) and Pacific cupped oyster (*Crassostrea gigas*): species, different cultivation areas, different time intervals of harvest	GC-FID GC-MS	PCA	[132]
Meat			
Halal authentication of pork meat	HS/GC-MS	PCA	[133]
Differentiation of fresh and frozen pork	UFGC	PCA, ANN	[134]

Table 6.
Literature examples of authentication and adulteration detection procedures of meat products and seafood.

Purpose of the study	Analytical technique	Chemometric technique	Ref.
Coffee			
Differentiation between arabica (*Coffea arabica* Linn.) and robusta (*Coffea canephora* Pierre ex Froehner var. *robusta*) coffees, either in green or in roasted stage	HR-GC-FID	HCA, CVA, DA	[135]
Determining the geographical origin of coffee samples	HS-SPME/GC-TOF-MS	PCA	[136]
Tea			
Differentiation of *Echinacea* species (*E. angustifolia*, *E. pallida*, *E. purpurea*)	GC-MS	HCA, PCA, LDA	[137]
Discrimination of oolong tea (*Camellia sinensis*) varieties	HS-SPME/GC-MS	PCA, HCA, SLDA	[138]
Discrimination of two roselle (*Hibiscus sabdariffa*) flower cultivars	SPME/GC-MS	PCA, HCA, OPLS-DA	[139]
Discrimination of different teas (*Camellia sinensis*)	HS-SPME/chiral-GC-MS	HCA, PLS-DA	[140]
Discrimination of American ginseng (*Panax quinquefolius* L.) and Asian ginseng (*Panax ginseng* Meyer)	GC-MS	PCA, PLS	[141]

Table 7.
Literature examples of authentication and adulteration detection procedures of coffee and tea.

2.7 Coffee and tea

Differentiation of coffee samples is based mostly on fatty acid profiles and volatile and semi-volatile compounds (organic acids, sugars, terpenoids). Differentiations of various tea plants were based exclusively on volatile components. In order to enable differentiations and classifications of investigated samples of beverages, the data obtained after GC analysis were combined with various chemometric techniques: HCA, PCA, SLDA and OPLS-DA. **Table 7** represents chronological literature data on the authentication and adulteration detection procedures of coffee and tea from various plant species.

3. Conclusions

Gas chromatograph, as a common instrument in most analytical laboratories worldwide, can be successfully applied in authentication and fraud detection procedures of various food and beverage products, such as olive oil and other edible vegetable oils, honey and other bee products, milk and dairy products, cereals and bakery products, meat, fish and seafood, as well as coffee and tea. In this manner, gas chromatograph is coupled to flame ionization detector or single/tandem mass spectrometers. It can be concluded that utilization of a GC device in further development of authentication methodologies could provide us with meaningful results, thus representing a significant contribution to this emerging field in the future.

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Conflict of interest

The authors declare that they have no conflict of interest.

Acronyms and abbreviations

AHC	agglomerative hierarchical clustering
ANN	artificial neural networks
C-IRMS	combustion isotope ratio mass spectrometry
CA	correspondence analysis
CDA	canonical discriminant analysis
CNN	counterpropagation neural network
CVA	canonical variates analysis
DA	discriminant analysis
DPLS	discriminant partial least squares
FID	flame ionization detector
GA	genetic algorithm
GC	gas chromatography
GC-chiral GC	fast multiple heart-cut enantioselective multidimensional gas chromatography

chiral GC × GC	enantioselective comprehensive two-dimensional gas chromatography
HCA	hierarchical cluster analysis
HR	high resolution
HS	headspace
HT	high temperature
IR	isotope ratio
IT	ion-trap
kNN	k-nearest neighbors
KNN	Kohonen neural network
LC	liquid chromatography
LDA	linear discriminant analysis
MLP	multilayer perceptron
MCOCPLS	Monte Carlo one-class partial least squares
MS	mass spectrometry
O	olfactometry
OC-SVM	one-class support vector machine
OPLS-DA	orthogonal projections to latent structures discriminant analysis
P-IRMS	pyrolysis isotope ratio mass spectrometry
PCA	principal component analysis
PCoA	principal coordinate analysis
PLS	principal least squares regression
Q-TOF-MS	quadrupole accurate mass time-of-flight mass spectrometry
QDA	quadratic discriminant analysis
R-SVM	recursive support vector machine
RDA	regularized discriminant analysis
RF	random forests
SIMCA	soft independent modeling of class analogy
SLDA	stepwise linear discriminant analysis
SPME	solid-phase microextraction
SVM	support vector machine
TOF	time-of-flight
UF	ultrafast module

Author details

Kristian Pastor*, Marijana Ačanski and Djura Vujić
Faculty of Technology Novi Sad, University of Novi Sad, Novi Sad, Serbia

*Address all correspondence to: pastor@tf.uns.ac.rs

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