# Alternative data mining/machine learning methods for the analytical evaluation of food quality and authenticity – A review

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## Abstract

In recent years, the variety and volume of data acquired by modern analytical instruments in order to conduct a better authentication of food has dramatically increased. Several pattern recognition tools have been developed to deal with the large volume and complexity of available trial data. The most widely used methods are principal component analysis (PCA), partial least squares-discriminant analysis (PLS-DA), soft independent modelling by class analogy (SIMCA), k-nearest neighbours (kNN), parallel factor analysis (PARAFAC), and multivariate curve resolution-alternating least squares (MCR-ALS). Nevertheless, there are alternative data treatment methods, such as support vector machine (SVM), classification and regression tree (CART) and random forest (RF), that show a great potential and more advantages compared to conventional ones. In this paper, we explain the background of these methods and review and discuss the reported studies in which these three methods have been applied in the area of food quality and authenticity. In addition, we clarify the technical terminology used in this particular area of research.

## Keywords

Data mining; random forest; CART; decision tree; food analysis

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## 1. Introduction

The assurance of food authenticity is the main concern of many consumers and manufacturers of high-quality products as well as official bodies and authorities in response to the need to protect consumers by detecting potential food fraud. Food authenticity is necessarily linked to the compliance; hence an authentic foodstuff is a product which strictly complies with genetic identity, natural composition, geographical and typological origin, ingredients, production technology, implicit quality features and explicit claims stated in the label. Overall, the food fraud entails a deception about the origin, quality or quantity of a foodstuff aimed of making an illicit profit. Globalization and free trade agreements have fostered an increased exchange of and access to food around the world. However, this has also led to an increase in problems associated with food fraud. There are three kind of main food frauds: non-conformity, adulteration and contamination. Non-conformity occurs when a food product does not fulfil the features which are stated in the label; it is identified by counterfeiting or imitation. Adulteration involves a deliberate and non-stated alteration of the intrinsic composition of the original food product. At least, contamination involves an unintended or accidental presence of extrinsic substances.

The serious nature of food fraud depends on the kind of fraud carried out. For instance, a food adulteration might be the substitution of the original ingredients by ingredients cheaper [Spink, Hegarty, Fortin, Elliot, & Moyer, 2019], as in the case of the olive oil that could be adulterated with cheaper vegetable oils. In this case, the consumer is paying more for a food product of inferior quality, but it does not involve any health risk. However, there are other types of food frauds which might affect to the human health. For example, the use of contaminated commodities, ingredients or allergens. In this sense, it is important that the food chain and the possible food fraud are extremely controlled by official bodies [Manning, 2016]. On the other hand, it is also important to ensure the authenticity of the product in term of geographical origin in order to control the replacements of genuine food products [Huch, Pezzei, & Huck-Pezzei, 2016; Medina, Perestrelo, Silva, Pereira, & Câmara, 2019]. Analytics involves several activities such as the analytical determination of specific physico-chemical characteristics, the qualification/quantitation of adulterants and/or contaminants and residues, and the verification of quality-differentiated technical requirements.

In this context, multivariate data analysis and pattern recognition techniques are powerful tools to conduct quality control and food authentication [Zielinski et al., 2014; Bevilacqua at al., 2017; Brereton et al., 2017; Callao & Ruisánchez, 2018; Efenberger-Szmechk, Nowak, & Kregiel, 2018; Granato et al., 2018].

The main purpose of multivariate pattern recognition methods is to perform the most appropriate data treatment in order to model and characterize a set of objects or samples that exhibit a particular feature or behaviour. To this end, significant and non-evident information is extracted to establish relationships between the objects/samples of the set, or between the set of objects/samples and one or several characteristics, according to the similarity of their spectra, chromatograms, elementary analysis, images, and so on. These tools must also be able to classify new samples into a certain group and reliably predict the value of a specific property in a fast and objective way [Brereton, 2015].

Pattern recognition methods are divided into two main groups: unsupervised methods, whose main tools are principal component analysis (PCA) and hierarchical cluster analysis (HCA); and supervised methods of analysis, such as k-nearest neighbours (kNN) [Steinbach & Tan, 2009], partial least squares-discriminant analysis (PLS-DA) [Ballabio & Consonni, 2013], and soft independent modelling by class analogy (SIMCA) [Oliveri & Smilde, 2012], among others. Likewise, in machine learning field are known as unsupervised and supervised learning techniques [Kavakiotis et al., 2017]. Figure 1 shows a straightforward flowchart of conventional pattern recognition methods.

Figure 1

An exploratory analysis is generally used to scout the data structure and determine whether there are trends in the data set. Principal component analysis (PCA) is a valuable statistical tool whose goal is to maximize the information of the variance in the data and show it visually in as few components as possible. It is mainly used to provide information on natural groupings of objects/samples and to reduce the number of variables necessary to represent the system, providing a new set of latent variables known as 'principal components' [Bro & Smilde, 2014]. Nevertheless, sometimes PCA has been erroneously applied and is used in some studies as a classification method to develop and validate classification models [Hakki, 2014; Chung, Kim, Lee, & Kim, 2015; Sun, Lin, Li, Shen, & Luo, 2015]; this is a serious mistake that unfortunately still happens. Cluster analysis is based on the intrinsic similarity between groups of objects/samples. The results of the hierarchical clusters analysis are presented as a dendrogram where the objects/samples are distributed in a ramified tree where the data are organised in categories and subcategories (branches) and the nodes represents the clusters according to their similarity [Drab & Daszykowski, 2014].

Supervised methods of analysis are divided into two groups: (i) classification or qualification methods and (ii) calibration or quantitation methods. Multivariate classification/qualification methods have been defined as chemometric techniques designed to find mathematical models that can recognize which class each object/sample belongs to base on a particular data set; they involve the use of various chemometric algorithms with two main statistical backgrounds related to discrimination and class-modelling approaches [Marini, 2010; Belvilacqua et al., 2014].

There are many classification methods but the most common ones are kNN, PLS-DA and SIMCA. Multivariate calibration/quantitation methods are in fact multivariate regression methods aimed at determining the functional relationships between the analytical signal acquired from a set of samples and a characteristic feature of such samples such as their composition. The most widely used algorithm is partial least squares (PLS) regression [Mehmood & Ahmed, 2016]. It should be noted that, although the classification is intrinsically a qualitative process, the assignment of the objects or samples to a specific class can have a qualitative basis (as by kNN or SIMCA) or a quantitative basis (as by PLS-DA). Indeed, the PLS-DA method involves performing a multivariate regression and placing a numeric value to each object/sample first, and then classifying them into a specific class [Brereton & Lloyd, 2014]. In addition, there are other kinds the multivariate methods that are applied when working with second order data. That means that a matrix of data is obtained for each sample rather than a vector of data (first order data). In this case, the most common methods are parallel factor analysis (PARAFAC) and multivariate curve resolution – alternating least squares (MCR-ALS).

The development of a pattern recognition supervised model involves two stages. The first stage is to build the model using a set of objects or samples whose class or particular features are known (i.e., training set or calibration set). In this stage, an internal validation or cross-validation could be applied in order to assess the goodness of fit of the model from the samples/objects of the training set. However, cross-validation by its own designs purpose, never able to achieve all the necessary objectives of a right validation [Esbesden & Geladi, 2010]. The second stage is to evaluate and externally validate the performance of the model built in the previous stage; this is done using additional objects or samples (i.e., test set or validation set) that fulfil the same requirements but were not part of the original training set [Szymanska et al., 2015; Westad & Marini, 2015]. In these methods, it is assumed that there are enough reference objects/samples that act as analytical standards because the outcomes of interest (i.e., the qualitative class or the value of one or more quantitative features) are formerly known or have been accurately measured. There are not definitive rules on the minimum number of samples/objects which are necessary for model development as this depends on the particular problem; it would however be desirable to devote the 40-50% of the reference samples/objects for the validation set.

The assessment of the quality of the classification models is evaluated through several performance features. These are estimated using the contingency table which records the number of both correct and incorrect assignations for each class in which samples of the validation set are arranged [Cuadros Rodríguez, Pérez Castaño, & Ruiz Samblas, 2016; Ballabio, Grisoni, & Todeschini, 2018]. In the same way, specific figures of merit have been proposed to assess the multivariate calibration models [Olivieri et al., 2006; Oliieri, 2014]. However, the assessment of the multivariate models is not enough and the whole analytical method should also be properly validated [Van der Veer, Van Ruth, & Akkermans, 2011; Alewijn, Van der Voet, & Van Ruth, 2016].

As regards the effective use of classification methods, some authors argue that it is better to use class-modelling methods such as SIMCA to perform an adequate food authentication [Rodionova & Titova, 2016]. This is because class-modelling methods operate, in the training stage, by defining a well-delimited acceptance region that contains all the objects/samples of the target class; consequently, only new objects/samples located in the acceptance region are assigned as belonging to the target class.

In recent years, the applications of new pattern recognition algorithms are growing in the area of food, due to their advantages and potential to solve complex problems related to food authenticity. The most widely used ones are support vector machine (SVM), classification and regression tree (CART), and random forest (RF), which can be used in both classification and calibration models. Surprisingly, their application is still scarce in the area of food quality and authenticity, although they are widely used in other areas such as metabolomics. Some authors have even reported their advantages compared to conventional techniques. For example, it has been stated that [Gromski et al., 2015] "… compared to PLS-DA, SVM is not influenced by the distribution of the different sample classes but rather focuses on which side of the support vectors particular test samples fall on". Similarly, the advantages of the RF algorithm have been reported in the area of ecology [Cutler et al., 2007].

As stated above the supervised multivariate methods are split in two groups (i) qualification or classification methods and (ii) quantification methods. In turn, classification methods are conventionally divided in discriminant analysis methods and class modelling methods depending on how the model is built. Discriminant analysis, as PLS-DA, works by establishing the boundaries between the different classes defined by the training objects while the class modelling methods, as SIMCA, define successive enclosed space domain which contain the objects of each class. Nevertheless, the models generated by decision trees methods (DT), as CART or RF, do not establish the separation of data in different classes as way above but the samples are divided into subsets (or classes) based on the value of certain variables, and this process is repeated on each derived subset of samples [Ai et al., 2014]. Consequently, the classifications are based on a set of concatenated decisions, similar to artificial neural networks (ANN). Figure 2 (a) shows a straightforward flowchart of the most common data mining/ chemometrics methods used for the analytical evaluation of food quality and authenticity, and figure 2(b) shows schematically how these methods operate. Table 1 assembles some of the advantages and disadvantages of them.

Figure 2

Table 1

All the methods cited above perform the classification using a threshold that is automatically established by the typical software of treatment multivariate data as PLS_Toolbox (under Matlab) (Eigenvector Research, WA, USA), SOLO (Eigenvector Research, WA, USA), SIMCA (Umetrics, Sweden) Unscrambler, (CAMO, Norway), Pirouette (Infometrix, WA, USA) or perClass Toolbox (under Matlab) (perClass BV, The Netherlands), to list only the most known. However, practitioners can decide on the classification threshold to conduct a more reliable classification [Vitale, Marini, & Ruckebush, 2018]. Table 2 collects a summary of the most common data mining methods which can be applied with the different software of multivariate data analysis.

Table 2

This paper reviews and describes the use of these alternative data mining/machine learning methods (i.e., SVM, CART, and RF) in the area of food analysis. Examples are provided to demonstrate the potential of these techniques in this area of study.

## 2. Background

### 2.1 Some basic terms

The automation and computerization of analytical laboratories have resulted in numerous changes; one of them is the acquisition of a high volume of data, giving rise to a new scientific discipline known as 'big data science', which has had a strong impact on many scientific disciplines. In chemistry, the term 'big data' refers to large and complex data sets that contain useful and non-evident chemistry-related information that must be extracted using complex data analysis tools [Parastar & Tauler, 2018]. Nevertheless, having a large amount of data does not mean that adequate answers can be provided unless the right data processing tools are applied. Collecting data is not synonymous with possessing information; data must be treated and interpreted to convert them into useful information for the user or the analyst. This subject, the right use of big data, and how it could satisfy the ISO/IEC 17025 requirements in the accreditation of laboratories has been already described [Ghernaout, Aichouni, & Alghamin, 2018].

The nomenclature used to refer to this kind of tools depends on the area of study. Analytical chemistry is the area with the greatest variability of terms. Some authors use the terms 'pattern recognition methods' or 'multivariable analysis methods', but the most commonly-used term is 'chemometric tools' to refer to the methods applied to the treatment of chemistry-related data. At its inception, chemometrics were defined as *an approach to analytical and measurement science that uses mathematical, statistical and other methods of formal logic to determine (often by indirect means) the properties of substances that otherwise would be very difficult to measure directly* [Lavine, 2000]. Currently, the International Union of Pure and Applied Chemistry (IUPAC) considers chemometrics as *the science of relating measurements made on a chemical system or process to the state of the system via application of mathematical or statistical methods* [Hibbert, 2016].In the field of engineering, these types of techniques for the processing of signals or images are often referred to as 'computational intelligence' or 'artificial intelligence' tools. The IUPAC has defined artificial intelligence as *the capability of a machine to perform human-like intelligence functions such as learning, adapting, reasoning and self-correction. The main areas of application are currently in expert systems, computer vision, natural language processing, robotics, and speech synthesis and recognition* [Kingston & Kingston, 1994]. Other authors define this term as *the interaction of several kinds of disciplines, such as computer science, cybernetics, information theory, psychology, linguistics, and neurophysiology. Artificial intelligence is a branch of computer science involved in the research, design and application of intelligent computers* [Lu, Chen, & Zheng, 2012]. Artificial neural networks (ANN) are the most widely used algorithm in this area. They are based on a series of 'nodes' or 'artificial neurons' that are interconnected with each other in a network that attempts to simulate the network of neurons in the human brain [Hibbert, 2016]. ANN are not explained in this study due to their different applications, although it is also usually classified as an alternative data treatment method [Yu, Low, & Zhou, 2018; Ropoli, Panagou, & Nychas, 2016; Marini, 2009].

In the areas of health care and biology (e.g., medicine, pharmacy, biology and biotechnology) the term 'bioinformatics' is routinely used and defined as the *discipline encompassing the development and utilization of computational facilities to store, analyse, and interpret biological data* [Duffus, Nordberg, & Templeton, 2007].

## 2.2 Data mining *vs.* machine learning

'Data mining' is a general term that encompasses all these tools regardless of the area of study in which they are used. This term appeared in the 1960s, but only became consolidated in the 1980s with the concept of 'knowledge discovery in databases' (KDD) [Mikut & Resichl, 2011; Han, Kamber, & Pei, 2012]. The term 'machine learning' is also commonly used for the same purpose [Zheng, Fue, & Ying, 2014]. Both terms are often used interchangeably to refer to all these processing data techniques although, strictly speaking, some differences can be observed between them.

Data mining can be used for descriptive purposes (i.e., showing similarities between the elements of a data set), or predictive purposes (i.e., predicting specific features of new data based on models that have previously been built and validated). It is based on the collection, storage, and treatment of a large amount of data in order to make the best decisions about a particular problem. It is an interdisciplinary field with the overall objective of revealing relationships in data from whatever source or origin. To this end, complex data treatment tools are used to detect and identify hidden patterns, associations, and structures that are proper of the raw big data set, or to select and filter useful information from big databases [Mitra & Acharya, 2003]. The concept of machine learning is also known as *the techniques involved in dealing with vast data in the most intelligent fashion (by developing algorithms) to derive actionable insights. In these techniques, we expect the algorithms to learn by them without being explicitly programmed* [https://www.analyticsvidhya.com]. Consequently, data mining refers to the area in general and machine learning refers exclusively to the algorithms used and it is linked to pattern recognition.

The IEEE International Conference on Data Mining, held in Hong Kong 2006, identified the top 10 data mining algorithms which were among the most influential data mining algorithms in the research community: C4.5, k-Means, SVM, Apriori, EM, PageRank, AdaBoost, kNN, Naïve Bayes, and CART. A survey paper was published describing the basis of each one [Wu et al., 2006]. Two of these, SVM and CART, are considered in this review.

In addition, data mining methods have been classified in four machine learning categories: (i) information-based learning, (ii) similarity-based learning, (iii) probability-based learning, and (iv) error-based learning. In general, DT methods (e.g., CART and RF) fall into the category of information-based learning and SVM belongs to the category of error-based learning [Keller, Name, & D'arcy, 2015]. We consider this classification to be very appropriate, since the methods are sorted according to how they build the different regions for each object/sample class of the classification model.

## 2.3 Data mining in food analysis

In recent years, data mining has been used more frequently in the area of food analysis, leaving the concepts of pattern recognition techniques or methods and chemometric tools to refer to the algorithms used to process the data. Both data mining and chemometrics represent very similar concepts. In fact, the only difference is that chemometrics has been used in reference to the application of machine learning techniques in order to obtain information of material system from data of mainly chemical or physical-chemical nature, while data mining is extensively used in many other areas such as security, facial recognition, customised marketing, medical diagnosis, air navigation, etc.

Researchers have reviewed some of the data mining methods that are increasingly used in chemometrics, that is, exploratory data analysis, artificial neural networks, pattern recognition, and digital image processing [Mutihac & Mutihac, 2008; Kumar, Bansal, Sarma, & Rawal, 2014; Messai, Farman, Sarraj-Laabidi, Hammami-Semmar, & Semmar, 2016]. Data mining methods are widely used in the area of food quality to verify compliance with regulations and quality-differenced requirements in order to ensure the authenticity of food. Besides this, consumers increasingly demand more information and knowledge about foodstuffs from producers.

The food sector is highly competitive and global, so food producers seek to become consolidated in emerging domestic and international markets and to make a difference with their products. Product differentiation is key to take a leading position in the global market of the sector. For example, a good strategy to outcompete competitors is to take advantage of the difference in the chemical composition or organoleptic characteristics of food. As a result, a current trend in analytical chemistry is to develop quick and reliable analytical methods to authenticate food products. This has led to the development of more powerful analytical instruments and the use of new methodologies to obtain more and better information about the objects/samples of study. An example of this is the development of more advanced sensors that can monitor food with a high level of detail, collecting a large volume of data. Thus, alternative methods to conventional data processing techniques are required.

Traditionally, chemometrics has been used in the area of food analytical chemistry to refer to the use of well-known conventional methods such as PCA, kNN, SIMCA, PLS-DA, and algorithms applied to second-order data such as parallel factor analysis (PARAFAC) and multivariate curve resolution-alternating least squares (MCR-ALS) [Zielinski et al., 2014; Callao & Ruisánchez, 2018; Rodopi, Panagou, & Nychas, 2016; Dai, Sun, Xiong, Cheng, & Zeng, 2014]. Nevertheless, as explained in the Introduction section, it is becoming more frequent to use the most up-to-date data processing methods, in food analysis, since they exhibit advantages and greater power than the conventional methods previously cited.

Figure 3 shows a plot of the trend in publications on food chemistry that have applied data mining methods in recent years. As can be seen, the SVM method is the most widely used method; however, in the last years the RF algorithm, which was used scarcely in food chemistry, has become more widespread and in 2018 the number of papers applying RF has tripled to the ones using SVM. In addition, the figure 4 shows the increase in the use of the 'data mining' term in the papers published in the area of food analytical chemistry in recent years.

Figure 3

Figure 4

## 3. Alternative methods

In most cases, the reported studies apply conventional multivariate pattern recognition methods with the main purpose of analysing the similarity between signals for food identification, classifying food according to various criteria (e.g., botanical or animal species, geographical origin), detecting adulterations and other non-conformities, and predicting properties related to food quality, such as antioxidant capacity [Jiang, Zheng, & Lu, 2015; Cuberon-Leon, Peñalver, & Maquet, 2016; Popescu et al., 2015; Pisano, Silva, & Olivieri, 2015] and stability. Several authors have reviewed the published studies about the use of these methods [Bosque-Sendra, Cuadros-Rodríguez, Ruiz-Samblás, & de la Mata, 2012; Berrueta, Alonso-Salces, & Héberger, 2007; Olivieri & Downey, 2012; Khakimov, Gürderniz, & Engelsen, 2015; Olivieri, 2012; Ortiz & Sarabia, 2007].

Recently, a comprehensive and valuable review has provided an overview of all the stages of the analysis of large analytical chemical datasets [Szymanska, 2018]. Nevertheless, it only considered conventional data processing methods and SVM, leaving out new data mining methods such as CART, RF, and others. Similarly, other recent reviews have focused on traditional chemometric methods and do not include any reference to these alternative methods [Callao & Ruisánchez, 2018, Cocchi, 2017]. This demonstrates that the inclusion of data mining methods in the area of analytical chemistry and specifically in food chemistry is relatively recent.

Considering only SVM, CART, and RF methods, the first method is the most widely used in food analytical chemistry and has been explored in a greater number of studies than the others (see Figure 2) [Mutihac & Mutihac, 2008; Brereton & Lloyd, 2010; Luts et al., 2010]. The goal of SVM is to find the best hyperplane in space that differentiates between the classes of the objects/samples by applying a maximization method. The aim is to maximize the 'margin', which is based on the sum of the distances from the hyperplane to the closest samples, that is, those correctly classified into their corresponding classes; SVM penalizes the number of misclassified samples. SVM algorithm uses a set of mathematical functions that are defined as the kernel. The kernel functions transform the original data into the required format. If the hyperplane is built in the original space the SVM model applies a linear kernel (it works similarly to the PLS-DA algorithm); if it is built in a different space (e.g., a higher dimension space), the SVM model is non-linear and alternative kernel functions must be used as the radial basis function (RBF) [Xu, Zomer & Brereton, 2006].

The main advantage of SVM over PLS-DA is that it creates a separation between the regions of the different classes when these are not sufficiently evident. Nevertheless, it is easier and faster to conduct a classification using PLS-DA than using SVM, since PLS-DA only performs a regression by partial least squares on the original data whereas SVM takes into account the transformation of the data in a higher dimension space. SVM is used for different purposes in the area of food analytical chemistry: (i) to classify food according to its geographic origin, (ii) to conduct a sensory evaluation, (iii) to detect adulterations, (iv) to quantify compounds, and (v) to conduct quality control.

DT is one of the most popular classification machine learning methods and is also widely used in the selection of features to determine food quality [Debska & Guzowska-Swider, 2011]. DT are sequential models which logically combine a sequence of simple comparisons between a numeric value of an input variable against a threshold value or a nominal attribute against a set of possible values [Kotsiantis, 2013]. DT divides the variable space into rectangular regions and predict the label associated with an particular instance by traveling from a root node of a tree to a leaf, where each label corresponds to one class. Each of these results creates additional nodes that branch out into other possibilities. This creates a structure that resembles a tree. There are three types of nodes: probability nodes, decision nodes, and terminal nodes [Witten & Frank, 2005]. DT can be translated into a set of rules by creating a separate rule for each path from the root to a leaf in the tree. Thus, the classification of a new sample begins in the root node of the tree and follows the branch that is appropriate to its outcome. The most known DT method is CART which is a single tree that shows many branches where the data set is split according to the selected decision, and the procedure is repeated as often as necessary. Figuratively, CART implies building a tree by growing and pruning it [Kucheryavskly, 2018].

DT models can be combined into ensembles by using boosting or bagging for yielding better predictive results than any of their constituent models when used separately [Kotsiantis, 2013; Kucheryavskly, 2018]. Boosting and bagging imply a sequential improving of a single tree by using random subsets from the whole dataset to build a set of small trees. These merged DT are called 'ensemble methods' and sometimes 'decision forest'. The main idea behind this is to combine several individual classifiers to obtain a classifier that outperforms every one of them [Rokach, 2010; Ruiz-Samblás, Cadenas, Pelta, & Cuadros-Rodríguez, 2014]. The main difference between both ensemble methods is the iteration. Boosting works iteratively weighting the individual instances in each run and learning successive models from the miss-classified examples while bagging generates independent models, each one from a different data-subset. One of the most known methods for bagging the trees is RF. Figure 5 shows the differences between the ensemble processes of boosting, bagging and random forest [Yang, Hwa-Yang, Zhou & Zomaya, 2010].

Figure 5

RF involves a set of stochastically different trees, each built from its own bootstrap samples [Mitchell, 2014], i.e., a combination of decision trees that is built using different sets of randomly selected input) variables [Gromski et al., 2015; Granitto, Gasperi, Biasioli, Trainotti, & Furlanello, 2007; Kucheryavskly, 2018]. Figure 6 graphically shows the operation of the RF method [Mitchell, 2014]: six decision trees forming a (very small) Random Forest for classification; trees A, B and E assign to the red class, however trees C and D assign to green class and tree F assigns to yellow class, so that the Random Forest will classify the object as red by a majority. An additional advantage of RF, compared to other classification methods such as PLS-DA or SVM, is the ability to directly discriminate in a single process between a set of samples/objects into a number of class higher than two (i.e. a multiclass classification problem).

Figure 6

## 4. Applications in food authentication

Table 3 reviews the most recent papers (i.e., published since 2010) in which SVM, DT, CART, and RF are applied in food analytical chemistry, among other more conventional chemometric methods. As mentioned in the Introduction section, RF has been scarcely used in food analytical chemistry. Yet, in recent years several papers have shown its potential in this area. Moreover, new software has been developed to apply RF using spectroscopic techniques in food chemistry [Smith, Baker, & Palmer, 2018].

Table 3

One of the most important aspects in order to assure the reliability of a pattern recognition supervised model is the validation step. This is fundamental for the assessment of the quality of the classification/quantification rate obtained in the multivariate models. The samples/objects used in this stage, which constitute the validation set or the test set, should be other than those used in the training stage. Thus the recommended is to use an external validation set. Nevertheless, sometimes the total number of samples of study is very limited and it is not possible to generate an external validation, and therefore it is carried out an internal cross-validation. Consequently, the quality performance features of the different multivariate models are calculated from the results obtained in cross-validation step. In this sense, the papers collected in table 1 there are 34 which applying internal cross-validation and 41 external validation.

Next, a comprehensive description on the gathered papers is carried out. For this, two blocks have been considered: support vector machine methods and decision trees methods. In addition, the meaning of all the abbreviations or acronyms is stated at the foot of the table3.

### 4.1 Support vector machine methods

#### 4.1.1 Fruits and juices

The studies carried out for the fruits and juices authentication are focused on the determination of some compounds as pesticides and additives, on the detection of adulterations and on the classification according to the geographical origin [Fan, Lai, Rasco, & Huang, 2015; Khanmohammadi et al., 2014; Naderi-Boldaji et al., 2015; Hong & Wong, 2014]. For example Guo et al. [Guo, Ni, & Kokot, 2016] developed different models for the quality control of jujube (Z. jujube Mill.) applying LDA, LS-SVM and BPANN to build classification models in order to classify the samples from four geographical regions. Moreover, PLS, LS-SVM and BPANN were used to quantify the content of total sugars, total phenols, and total acids, and the total antioxidant activity and concluded that the LS-SVM prediction models produced best results compared to the conventional chemometrics.

### 4.1.2 Honey and sugar

Honey is a sweet substance produced by bees from the nectar of flowers. Most studies, in which SVM is used, are based on the classification of commercial honeys from different geographical regions applying mainly spectroscopic and chromatographic analytical techniques. In this regard, El Alami et al. [El Alami et al., 2018] developed a model to discriminate between honeys from France and Morocco, classified correctly all the samples. On the other hand, Wei et al. [Wei, Wang & Wang, 2010] honey samples from different regions from China. In addition, the floral origin of the honey samples was predicted using rheometric features. Concerning the authentication of sugar, Ramírez-Morales et al. [Ramírez-Morales, Rivero, Fernández-Blanco, & Pazos, 2016], applying NIR spectroscopy and support vector regression (SVR) to perform the quality control of ºBrix and sucrose parameters of sugar industry.

### 4.1.3 Liquors and spirit beverages

Most of the papers are focused on performing a quality control of the different beverages in order to detect adulterations with other fake drinks [Pérez-Caballero et al., 2017; Andrade, Ballabio, Gómez-Carracero, & Pérez Caballero, 2017; Contreras et al., 2010; Ceballos-Magaña et al., 2012]. It should be highlighted the study published by Cheng et al. [Cheng, Fan & Yan, 2013] in which the authors tested two ways of data reduction using PCA and PLS prior to the application of SVM to classify different kinds of liquors from China. Overall, the authors concluded that the reduction of data by PLS was the best.

### 4.1.4 Meat

Regarding the authentication of meat, the reported works are focused on the analysis of volatile compounds collected using GC-MS or electronic nose to carry out a quality control in order to differentiate between fresh and refrigerated meat [Papadopolou, Panagou, Mohareb, & Nychas, 2013; Arredondo et al., 2014; Moharabeb, Papadopolou, Panagou, & Nychas, 2016].

### 4.1.5 Milk and dairy products

Majcher et al. [Majcher, Kaczmarek, Klenporf-Pawlik, Pikul, & Jelén, 2015] developed a rapid method for the authentication of cheeses protected under a 'Denomination of Origin' using SPME-MS as analytical measuring technique. The classification methods used were LDA, SIMCA and SVM. The highest classification accuracy (97.9%) for the test set was obtained using SVM.

### 4.1.6 Plant products

This subsection collects the studies related to the authentication of pepper, tea, cocoa, coffee and rice using spectrometric (UV-Vis, FTIR, NIR, Raman and ICP), voltammetric and chromatographic (HPLC) analytical techniques [Li, Sun, Pu, & Jayas, 2017; Liu et al., 2014; Zheng et al., 2009; [Gonçalvez et al., 2016]; Wood, Allaway, Boult, & Scott, 2010; Teye & Huang, 2015; Barbosa et al., 2014; Bona et al., 2017; Barbosa et al., 2016; Maione, Lemos Batista, Campiglia, Barbosa, & Barbosa, 2016b; Kyu et al., 2017; Feng, Zhang, Cong, & Zhu, 2013]. In these works SVM was applied for the purpose of performing the quality control and authenticity evaluation of the foodstuffs.

One of the most significant works was carried out by Teye et al. [Teye, Huang, Han, & Botchway, 2014], in which FDA, kNN and SVM classification models to distinguish between cocoa bean samples were developed. The results revealed that SVM was better than kNN and FDA since 100% of the samples were correctly classified.

The tea authenticity studies are focussed on distinguishing the botanical or geographical origin. Among the studies is outstanding the one published by Liu et al. [Liu et al., 2014] that applied the voltammetry as analytical technique from which the whole analytical signal was used to build the SVM classification model.

### 4.1.7 Vegetable oils

It is noteworthy that for the authentication of vegetable oils is mostly applied the chromatographic techniques in contrast to the rest of food, in which is more common the use of the spectroscopic techniques. Moreover, high performance liquid chromatography (HPLC) coupled to different detection system is more usual than gas chromatography (GC) for the authentication of the olive oil.

All the reported studies are focused on; (i) the detection of adulterations; (ii) the verification of the geographical origin; and (iii) the discrimination of different kinds of edible oils [Dong, Zhang, Zhang, & Wang, 2013; Devos, Downey, & Duponchel, 2014; Sayago, González-Domínguez, Beltrán, & Fernández-Recamales, 2018; Ordukaya & Karlik, 2017; Jiménez-Carvelo, Pérez-Castaño, González-Casado, & Cuadros-Rodríguez, 2017a; Jiménez-Carvelo, González-Casado, Pérez-Castaño, & Cuadros-Rodríguez, 2017b]. Furthermore, the quantification of olive oil in blends with other vegetable oils and the classification according to the cultivar are reported [Dong, Zhang, Zhang, & Wang, 2012; Jiménez-Carvelo, Osorio, Koidis, González-Casado, & Cuadros-Rodríguez, 2017c; Jiménez-Carvelo, González-Casado, & Cuadros-Rodríguez, 2017d; Jiménez-Carvelo, Cruz, Olivieri, González-Casado, & Cuadros-Rodríguez, 2019].

#### 4.1.8 Wine

Another relevant aspect of food authenticity is the varietal authentication. This is often the case of the studies published about the analysis of the wine, since the chemical composition it is greatly influenced by the kind of grape, besides of the agronomic conditions.

An attractive strategy to take a prominent position over the competitors is to take advantage of the difference in chemical composition, bearing a recognised quality-differentiated food seal as the 'Protected Designation Origin' (PDO) or 'Protected Geographical Indication' (PGI). Thus, it is important to develop rapid methods to authenticate such protected foodstuffs. In this sense, Costa et al. [Costa, García Llobodanin, Alves Castro, & Barbosa, 2018] reported a study based on the analysis of different parameters of the wine and ~~the~~ then SVM was applied to discriminate wine from Brazil of wine from Uruguay; the classification model achieved an accuracy rate of 79.97%. Martelo-Vidal et al. [Martelo-Vidal, & Vazquez] developed LDA, SIMCA and SVM classification models based on the polyphenolic profile to differentiate between wines from Spanish PDO 'Rias Baixas' and 'Ribeira Sacra'.

#### 4.1.9 Others

In this category are included the studies carried out to ensure the quality and authenticity of tofu and vinegar.

Xu et al. [Xu et al., 2012] applied FTIR spectroscopy to analyse the shelf-life of the tofu. The samples were measured with different age (from 29 to 161 days). In the subsequent statistical analysis different pre-processing methods were tested before to the development of the PLS and SVM multivariate models. The models were evaluated and SVM was the best option, since it was obtained the lowest root mean squared error of prediction (RMSEP). Although the difference with PLS was such a small that the authors concluded that PLS should be used since it is low complexity.

Bao et al. [Bao et al., 2014] developed an analytical method for the quality control of the ºbrix and pH of the white vinegar. What is remarkable about this study was the application of PLS prior to the use of LS-SVM in order to select the latent variables (LVs), which were used as the inputs of the LS-SVM to develop the calibration model. The predictive capability of the models was evaluated estimating the correlation coefficient (r), the root mean square error of calibration and prediction (RMSEC & RMSEP), and the residual predictive deviation (RPD).

### 4.2 Decision tree methods: CART and RF

#### 4.2.1 Fruits and juices

Organic foods are appreciated by customers increasing their sales in the last years. The published scientific paper devoted to authentication of fruits and juices are focused on the differentiation of ecologic products from non-ecologic ones and the detection of additives.

Maione et al. [Maione et al., 2016a] developed an analytical method using ICP-MS to discriminate organic from conventional grape juice. In addition, they carried out the comparison between different data mining methods (SVM, CART and MLP) and the results obtained showed that all the methods provided good results for the intended purpose.

#### 4.2.2 Honey

The quality of honey varies depending on the floral and geographical origin. The conventional methods are based on the measure of several physicochemical parameters what are time-consuming and involve a high consumption of solvents. For this reason, Popek et al. [Popek, Halagarda, & Jursa, 2017] and Chuddzinks et al. [Chudzinksa & baralkiewicz, 2011] proposed different analytical methods combined with CART in order to reduce the time and the complexity of the analysis.

#### 4.2.3 Spirit beverages

The study carried out by Martínez-Jarquín et al. [Matínez-Jarquín, Moreno-Pedraza, Cázarez-García, & Winkler, 2017] was based on the application of the mass spectrometry along with PCA and RF to discriminate agave tequilas from traditionally processed mezcal. The most noticeable of this work was the application of RF to select the number of the variables, which were used in the new PCA model. Surprisingly PCA was applied as a classification method when it is an unsupervised pattern recognition method which only should be used to explore the variability of the samples in the dataset and/or to screening the inherent sample grouping when the dimensionality of the data is reduced. However RF, which is in itself is a multivariate classification method, is not applied to this end.

#### 4.2.4 Milk and dairy products

Fabris et al. [Fabris et al., 2010] developed different multivariate classification methods with the data acquired using PTR-TOF-MS in order to perform the quality control of Trentingrana cheese, when it is produced with milk stores in different conditions. Four binary classification models were built using PDA, DPLS, SVM and RF in order to select the best data mining method. Finally, they concluded that all the methods provided similar performance.

#### 4.2.5 Rice

Rice is a staple food in many developing and least developed countries. There are a lot of countries producers of rice, being Chine the world's largest producer; therefore the differentiation in the global market is important for the producers. In this sense the reported studies are focused on classifying the rice according to the geographical origin [Kyu et al, 2017; Mahdavi, Farimani, Fathi, & Chassempour, 2015; Weng et al., 2018].

Maione et al. [Maione, Lemos Batista, Campiglia, Barbosa, & Barbosa, 2016b] developed different classification methods to discriminate rice samples according to their geographical origin. For this purpose, the authors applied SVM, RF and MLP methods. They evaluated these multivariate classification methods using the following performance metrics: accuracy, sensitivity, specificity, and area under the receiving operating curve (AUC); in all cases, SVM and RF yielded better results than MLP.

#### 4.2.6 Tea

RF was applied to classify the tea according to the botanical and geographical origin and to discriminate between different varieties of tea [Gonçalvez et al., 2016; Zheng et al., 2009; Wang, Huang, Fan, & Lu, 2015].

Ni et al. [Ni, et al., 2018] built several classification models to distinguish between green tea from different regions in China. They compared the results using LDA, PLS-DA, and DT. The best results were obtained when DT was applied.

4.2.7 Vegetable oils

Edible vegetable oils are globally a kind of important food, a lot of them are present in several diets of the different regions of the world, such as olive oil in Mediterranean diet, which is characteristic of Spain, Italy and Greece or seeds oils in Asian. The process of obtaining of some high-price vegetable oils is expensive and consequently these are subject to possible adulterations in order to reduce the production cost. For this reason, ensuring the authenticity of the vegetable oils is currently required in order to detect such adulterations.

In this regard the published papers are focused on the detection of adulterations, the discrimination of edible oils according to the botanical and geographical origin [Zhang et al., 2014; Hu et al., 2014; Ruiz-Samblás, Cadenas, Pelta & Cuadros-Rodríguez, 2014; Nasibov, Kantarci, Vahaplar, & Kinay, 2016; Sayago, González-Domínguez, Beltrán, & Fernández-Recamales, 2018; Jiménez-Carvelo, Cruz, Olivieri, González-Casado, & Cuadros-Rodríguez, 2019]. Although there is one study which stands out since the RF method was used in an unconventional way, Ai et al. [Ai et al., 2014] analysed the fatty acid composition of six different kinds of vegetable oils (tea, olive, rapeseed, corn, sunflower and sesame oil) and they applied RF as unsupervised technique to carry out a cluster analysis in order to test if there were natural grouping of the oils samples.

4.2.8 Wine

Within the wine industry the authenticity evaluation in terms of geographical and brand origin influence in the choice of the consumers. Such is the case of Loannou-Papayianni et al. [Loannou-Papayianni, Kokkinfta, & Theocharis, 2011] who developed an analytical method using FTIR and CART to authenticate Cypriot traditional wine and to differentiate it from its competitors. Gómez-Meire et al. [Gómez-Meire, Falqué, Díaz & Fdez-Riverola, 2014] appled GC-MS combined with RF and MLP to ensure and to classify wine elaborated in Galicia (a region of the Nord of Spain); they concluded that the application of machine learning methods allows ensuring the authenticity of different white wines elaborated from several grape varieties and origins.

## 4. Final remarks

SVM, CART, and RF are an alternative group of pattern recognition methods that are yielding promising results in the area food quality and authenticity. Considering only these three methods, SVM is by far the most widely used and, in most cases, it is stated that SVM has an improved performance compared to other better-known conventional methods such as PLS-DA.

In addition, CART and RF are alternative pattern recognition methods that are currently used in the area of food. In other related areas such as metabolomics, some authors have already highlighted the advantages of these machine learning methods as compared to conventional techniques. However, there are still very few reported studies in which CART and RF are used in studies on food analytical chemistry even though their value has been widely proven and they have yielded outstanding results.

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**Table 1.** Advantages and disadvantages of some data mining/chemometric methods

| Method | Advantages | Disadvantages |
|---|---|---|
| PCA | This displays quickly and easily similarities and differences between the samples and the variables relationships. | It does not allow classifying and assigning a class to each sample. |
| kNN | User-friendly method of applying. | If there are more samples of one class than other class (skewed distribution of classes), it could cause a wrong classification of the samples since the dominant class controls the classification. |
| SIMCA | It is able to develop a binary classification model training only with the target-class since it defines an acceptance region that contains all the objects/samples of the target class. | In models trained with two classes or more might give rise to overlapping between acceptance regions which contain the samples of the different classes. Thus, some samples might be classified in one or more classes. |
| PLS-DA | The classification model is built quickly and easily, and the results use to be very successful. | If the separation between the regions of the different classes are not sufficiently evident could give rise to classification errors. |
| SVM | This can circumvent the technical difficulty when the separation between the regions of the different classes of the samples are not sufficiently evident. | For non-lineal SVM models, alternative kernel functions must be used. Thus, the development of the model is difficult, and a lot of Informatics resources are necessary. |
| DT / CART | This is not affected by outliers or non-linear relationships. The models are presented in a simplified and are easy-to-interpret way. | It performs poorly when training set is small in comparison with the number of classes, especially for continuous data. |
| Boosted DT | This achieves more accurate classification by decreasing bias. | The model could be overfitted, therefore might fail to fit new samples and predict them incorrectly. |
| Bagged DT / RF | This is very suitable for unstable models or for class imbalance problems since the variance is decreased. The | Understanding results is complex since the classification is not displayed as a graphical tree. |

| | | |
|---|---|---|
| | risk of overfitting is minimised. | |

**Acronyms:** PCA (principal component analysis), SIMCA (soft independent modelling by class analogy), kNN (k-nearest neighbours), PLS-DA (partial least squares – discriminant analysis), SVM (support vector machine), DT (decision trees), CART (classification and regression tree), RF (random forest).

**Table 2.** Pattern recognition methods implemented in some of the more used software in multivariate data analysis for food quality and authenticity data

| Software | Exploratory analysis | | Class-modelling | Discriminant analysis | | | Decision rules | | | Variable reduction | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | HCA | PCA | SIMCA | kNN | SVM | PLS-DA | ANN | CART | RF | MCR-ALS | PARAFAC |
| PLS_Toolbox | • | • | • | • | • | • | • | | | • | • |
| SOLO | • | • | • | • | • | • | • | | | • | • |
| SIMCA | • | • | • | | | • | | | | | |
| Unscrambler | • | • | | | • | • | | | | | |
| Pirouette | • | • | • | • | | | | | | | |
| PerClass | • | • | • | • | • | • | • | • | • | | |

**Acronyms:** PCA (principal component analysis), HCA (hierarchy cluster analysis), SIMCA (soft independent modelling by class analogy), kNN (k-nearest neighbours), PLS-DA (partial least squares – discriminant analysis), SVM (support vector machine), ANN (artificial neural network), CART (classification and regression tree), RF (random forest), PARAFAC (parallel factor analysis), MCR-ALS (multivariate curve resolution – alternating least squares).

**Table3.** Compilation of papers on food chemistry in which SVM, DT, CART, and RF have been used

| **Food** | **Purpose** | **Analytical technique** | **Tool** | **Ref.** |
|---|---|---|---|---|
| Apple | Determination of pesticides | Raman spectrometry | PLS, **SVM** | [Fan, Lai, Rasco, & Huang, 2015] |
| Beef | Evaluation of sensory quality | Electronic nose | PCA, DFA, **SVM** | [Papadopolou, Panagou, Mohareb, & Nychas, 2013] |
| | Evaluation of sensory quality | GC-MS | LDA, SIMCA, PLS-DA, **SVM** | [Arredondo et al., 2014] |
| | Evaluation of sensory quality | Electronic nose | **SVM** | [Mohareb, Papadopolou, Panagou, & Nychas, 2016] |
| Beer | Selection of the optimal number of parameters describing beer qualities | *Several chemical parameters* | **DT** | [Debska & Guzowska-Swider, 2011] |
| Cheese | PDO authenticity | SPME-MS | PCA, LDA, SIMCA, **SVM** | [Majcher, Kaczmarek, Pawlik, Pikul, & Jelén, 2015] |
| | Quality control | PTR-TOF-MS | **RF**, **SVM**, PDA, DPLS | [Fabris, et al., 2010] |
| Cocoa | Evaluation of sensory quality | Sensory tasting | PLS, **SVM**, MLR | [Wood, Allaway, Boult, & Scott, 2010] |
| | Quantification of the total fat content | FT-NIR spectrometry | PLS, **SVM** | [Teye & Huang, 2015] |
| | Classification according to their geographical origin | Electronic tongue | FDA, PCA, kNN, **SVM** | [Teye, Huang, Han, & Botchway, 2014] |
| Coffee | Evaluation of authenticity according to | ICP-MS | MLP, **SVM**, NB | [Barbosa et al., 2014] |

| | | | | |
|---|---|---|---|---|
| | the trace element | | | |
| | Classification according to geographical origin | NIR and FTIR spectrometry | **SVM** | [Bona et al., 2017] |
| Fruit | Classification of persimmons according to geographical origin | FT-NIR spectrometry | HCA, PCA, **SVM** | [Khanmohammadi et al., 2014] |
| | Classification of jujube fruit according to geographical origin | NIR spectrometry | PCA, LDA, **SVM**, ANN | [Guo, Ni, & Kokot, 2016]] |
| Grape | Quality control | Imaging spectrometry | **SVM** | [Liu, & Whitty, 2015] |
| | Detection of adulteration | Dielectric sensor | PCA, HCA, LDA, **SVM** | [Naderi-Boldaji et al., 2018] |
| Ginseng | Classification according to geographical origin | FT-MIR, NIR | **RF** | [Li, Zhang, & Wang, 2018] |
| Honey | Classification according to floral and geographical origin | Rheometry | PCA, PLS, PCR, **SVM** | [Wei, Wang, & Wang, 2010] |
| | Classification according to geographical origin | GC x GC-TOF | SIMCA, DPLS, LDA, **SVM** | [Stanimora et al., 2010] |
| | Classification according to geographical origin | Electronic tongue | PCA, HCA, PLS, **SVM** | [El Alami et al., 2018] |
| | Classification according to phenolic composition | HPLC-UV | PLS-DA, **SVM** | [Kemal, de B Harrington, Sahin, Demir, & Gunes, 2017] |
| | Classification according to botanical origin | viscosimetry, UV-Vis spectrometry, HPLC-IR, HPLC-UV | **CART** | [Popek, Halagarda, & Jursa, 2017] |
| | Classification according to botanical and geographical origin | ICP-MS | LDA, **CART** | [Chudzinska & Baralkiewicz, 2011] |

| | | | | |
|---|---|---|---|---|
| Juices | Detection of adulteration of tomato juices | Electronic nose and tongue | CDA, **SVM**, PCR | [Hong & Wang, 2014] |
| | Discrimination of organic grape juice from conventional grape juice | ICP-MS | **SVM**, **CART,** MPL | [Maione et al., 2016a] |
| | Detection of additives | Electronic nose | PLS, **SVM**, **RF** | [Qiu & Wang, 2017] |
| Licors | Quality control | HS-SPME-MS | PLS, **SVM** | [Cheng, Fan & Yan, 2013] |
| Olive oils | Classification according to geographical origin | NIR and MIR spectrometry | **SVM** | [Devos, Downey, & Duponchel, 2014] |
| | Detection and quantification of adulteration of extra virgin olive oil with other edible vegetable oils | Raman spectrometry | PLS, **SVM** | [Dong, Zhang, Zhang, & Wang, 2012] |
| | Classification of oil blends according to the vegetable oil used for blending and prediction of the proportion of olive oil used in each blend | GC-MS | **CART**, **RF** | [Ruiz-Samblás, Cadenas, Pelta, & Cuadros-Rodríguez, 2014] |
| | Discrimination of olive oil from other edible vegetable oils | HPLC-CAD | PCA, kNN, PLS-DA, OCPLS, SIMCA, **SVM** | [Jiménez-Carvelo, Pérez-Castaño, González-Casado, & Cuadros-Rodríguez, 2017a] |
| | Discrimination of olive oil from other edible vegetable oils | HPLC-CAD | PCA, PLS-DA, OCPLS, kNN, SIMCA, **SVM** | [Jiménez-Carvelo, González-Casado, Pérez-Castaño, & Cuadros-Rodríguez, 2017b] |
| | Discrimination of olive oil from other edible vegetable oils and quantification of the proportion of olive oil in blends with other vegetable oils. | FTIR and Raman spectrometry | PCA, PLS-DA, OCPLS, kNN, SIMCA, **SVM** | [Jiménez-Carvelo, Osorio, Koidis, González-Casado, Cuadros-Rodríguez, 2017c] |

| | | | | |
|---|---|---|---|---|
| | Quantification of olive oils in blends with other vegetable oils | HPLC-CAD | PLS, **SVM** | [Jiménez-Carvelo, González-Casado, & Cuadros-Rodríguez, 2017d] |
| | Classification according to geographical origin | HPLC-IR, GC-FID | PCA, **DT** | [Nasibov, Kantarci, Vahaplar, & Kinay, 2016] |
| | Classification according to geographical origin | ICP-MS | PLS-DA, **SVM**, **RF** | [Sayago, González-Domínguez, Beltrán, & Fernández-Recamales, 2018] |
| | Identification and classification of Turkish olive oils according to geographical origin | Electronic Nose | NB, kNN, LDA, ANN, **SVM** | [Ordukaya & Karlik, 2017] |
| | Quality control | Synchronous spectrofluorimetry | LDA, QDA, RDA, kNN, **SVM, RF** | [Dankowska & Kowalewski, 2018] |
| | Classification of extra virgin olive oils according to their cultivars | HPLC-DAD | PCA, MCR, PLS-DA, NPLS-DA, **RF** | [Jiménez-Carvelo, Cruz, Olivieri, González-Casado, & Cuadros-Rodríguez, 2019] |
| Pepper | Determination of pesticides | Raman spectrometry | **SVM** | [Li, Sun, Pu & Jayas, 2017] |
| Rice | Discrimination of organic rice from conventional rice | ICP-MS | **SVM** | [Barbosa, et al., 2016] |
| | Classification according to geographical origin | ICP-MS | **SVM**, **RF**, ANN | [Maione, Lemos Batista, Campiglia, Barbosa, & Barbosa, 2016b] |
| | Detection of adulterations | MS | **SVM**, **RF**, kNN | [Kyu et al., 2017] |
| | Classification according to geographical origin | Raman spectrometry | kNN, SIMCA, PLS-DA, **SVM** | [Feng, Zhang, Cong, & Zhu, 2013] |

| | | | | |
|---|---|---|---|---|
| | Quality control | GC-MS | **RF** | [Mahdavi, Farimani, Fathi, & Chassempour, 2015] |
| | Quantification of Ediphenphos | Raman spectrometry | PCA, PLS, **RF** | [Weng et al., 2018] |
| Sugar | Quality control | NIR spectrometry | **SVM** | [Ramírez-Morales, Rivero, Fernández-Blanco, & Pazos, 2016] |
| | Authenticity evaluation | ICP-MS | NB, **RF** | [Barbosa et al., 2015] |
| Tea | Discrimination of green and black tea | Voltammetry | PCA, **SVM** | [Liu et al., 2014] |
| | Classification between different kinds of tea | HPLC-UV | PCA, **SVM**, **RF** | [Zheng et al., 2009] |
| | Discrimination of five varieties of green tea and quantification of polyphenolic compounds | NIR and UV-Vis spectrometry | PCA, PLS, **RF** | [Wang, Huang, Fan, & Lu, 2015] |
| | Classification according to geographical origin | ICP-AES | LDA, PLS-DA, **DT** | [Ni, et al., 2018] |
| | Classification according to botanical and geographical origin | UV-Vis spectrometry | kNN, SIMCA, PLS-DA, PCA-LDA, **CART** | [Gonçalvez et al., 2016] |
| Tequila | Discrimination of tequila from traditionally processed mescal | (DIESI)LTP-MS | PCA, **RF** | [Martínez-Jarquín, Moreno-Pedraza, Cázarez-García, & Winkler, 2017] |
| | Differentiation of different kinds of tequila | UV-Vis spectrometry | kNN, SIMCA, PCA, PLS, **CART**, **RF**, **SVM** | [Pérez-Caballero et al., 2017] |
| | Differentiation of different kinds of tequila | UV-Vis spectrometry | QDA, PLS-DA, PLS-KERNEL, **SVM,** CPANN | [Andrade, Ballabio, Gómez-Carracedo, & Pérez-Caballero,, 2017] |

| | | | | |
|---|---|---|---|---|
| | Detection of adulterations | UV-Vis spectrometry | PCA, LDA, **SVM** | [Contreras, et al., 2010] |
| | Classification according to the geographical origin | ICP-AES | PCA, LDA, **SVM** | [Ceballos-Magaña, *et al.*, 2012] |
| Tofu | Study of its shelf-life | FTIR spectrometry | PLS, **SVM** | [Xu et al., 2012] |
| Vegetable oils | Quantification of fatty acid compounds | Raman spectrometry | **SVM** | [Dong, Zhang, Zhang, & Wang, 2013] |
| | Detection of adulteration | GC-MS | PCA, **RF** | [Zhang et al., 2014] |
| | Discrimination of vegetable oils according to their quality | GC-MS | PCA, HCA, **RF** | [Ai et al., 2014] |
| | Detection of adulteration | GCxGC-TOF | PCA, HCA, **RF** | [Hu et al., 2014] |
| Vinegar | Authenticity evaluation | Electronic tongue | **SVM, RF,** BPANN | [Liu, Wang, Wang, & Li, 2013] |
| | PDO authenticity | Spectrofluorimetry | PARAFAC, PLS-DA, **SVM** | [Ríos-Reina et al., 2017] |
| | Quality control | NIR spectrometry | **LS-SVM,** BPANN, PLS | [Ji-yong et al., 2013] |
| | Quality control | Vis/NIR spectrometry | PLS, **LS-SVM** | [Bao et al., 2014] |
| Wine | Authentication based on the grape variety | GC-MS | **DAG tree**, OPLS-DA, SIMCA | [Springer, 2019] |
| | Authenticity evaluation | FTIR spectrometry | PCA, HCA, LDA, **CART** | [Loannou-Papayianni, Kokkinfta, & Theocharis, 2011] |
| | Assurance of the authenticity according to the grape variety and different family compounds | GC-FID; GC-FPD; GC-MS | **SVM**, **RF**, MLP, kNN, NB | [Gómez-Meire, Campos, Falqué, Díaz & Fdez-Riverola, 2014] |
| | Classification according to the geographical origin | HPLC-DAD | **SVM** | [da Costa, Castro, & Barbosa, 2016] |

| | | | |
|---|---|---|---|
| Classification according to the geographical origin | HPLC-DAD; HPLC-DAD-MS | **SVM** | [Costa, García Llobodanin, Alves Castro, & Barbosa, 2018] |
| Classification according to geographical origin | UV/Vis/NIR spectrometry | LDA, SIMCA, **SVM** | [Martelo-Vidal, Dominguez-Agis, & Vázquez, 2013] |
| Classification of white wine from different brands according to their elemental profile | ICP-MS | **SVM** | [Jurado, Alcázar, Palacios-Morillo, & de Pablos, 2012] |
| Classification according to their grape variety | NIR spectrometry | RBFNN, **SVM** | [Yu, Zhan, & Huang, 2017] |
| Evaluation of sensory quality | GC-MS | **RF** | [Vigneau, Coureoux, Symoneaux, Guérin, & Villière, 2018] |
| PDO authenticity | HPLC-DAD | LDA, SIMCA, **SVM** | [Martelo-Vidal & Vázquez, 2016] |
| Quality Control | UV-Vis spectrometry | PCA, **SVM** | [Liu, Pan & Zhang, 2018] |

**Acronyms**: BPANN (back propagation artificial neural network), CAD (charged aerosol detector), CART (classification and regression tree), CDA (canonical discriminant analysis), CPANN (counter propagation artificial neural networks), DAD (diode array detector), DAG (directed acyclic graph), DIESI (direct-injection electrospray ionisation), DFA (discriminant function analysis), DPLS (discriminant partial least squares), DT (decision tree), FID (flame ionization detector), FDA (Fisher's discriminant analysis), FPD (photometric flame detection), FTIR-HATR (Fourier transform infrared spectroscopy - horizontal attenuated total reflectance), FT-NIR (Fourier transform-near infrared), GC (gas chromatography), HCA (hierarchical cluster analysis), HPLC (high performance liquid chromatography), HS (head space), ICP (inductively coupled plasma), IR (refractive Index), kNN (k-nearest neighbour), LDA (linear discriminant analysis), LTP ( low-temperature plasma), LS-SVM (least-squares support vector machine), MLP (multilayer perceptron), MS (mass spectrometry), NB (naïve Bayes), OCPLS (one class partial least squares), PARAFAC (parallel factor analysis), PCA (principal component analysis), PCR (principal component regression), PDA (penalised discriminant analysis), PLS (partial least squares), PLS-DA (partial least squares-discriminant analysis), PTR (proton-transfer-reaction), QDA (quadratic discriminant analysis), RDF (regularized discriminant analysis), RF (random forest), SIMCA (soft independent modelling by class analogy), SPME (solid-phase microextraction), SVM (support vector machine), TOF (time of flight), UV (ultra violet), Vis (visible).

**Figure caption**

**Figure 1**. General overview of conventional chemometric pattern recognition methods.

**Figure 2**. (a) Diagram showing the most–usual data mining chemometric methods used in food quality and authenticity; (b) Simple graphical description of how some of the most-usual data mining/chemometric methods carry out the classification.

**Figure 3**. Trends in publications in the area of food chemistry in which SVM, RF, DT, and CART have been used.

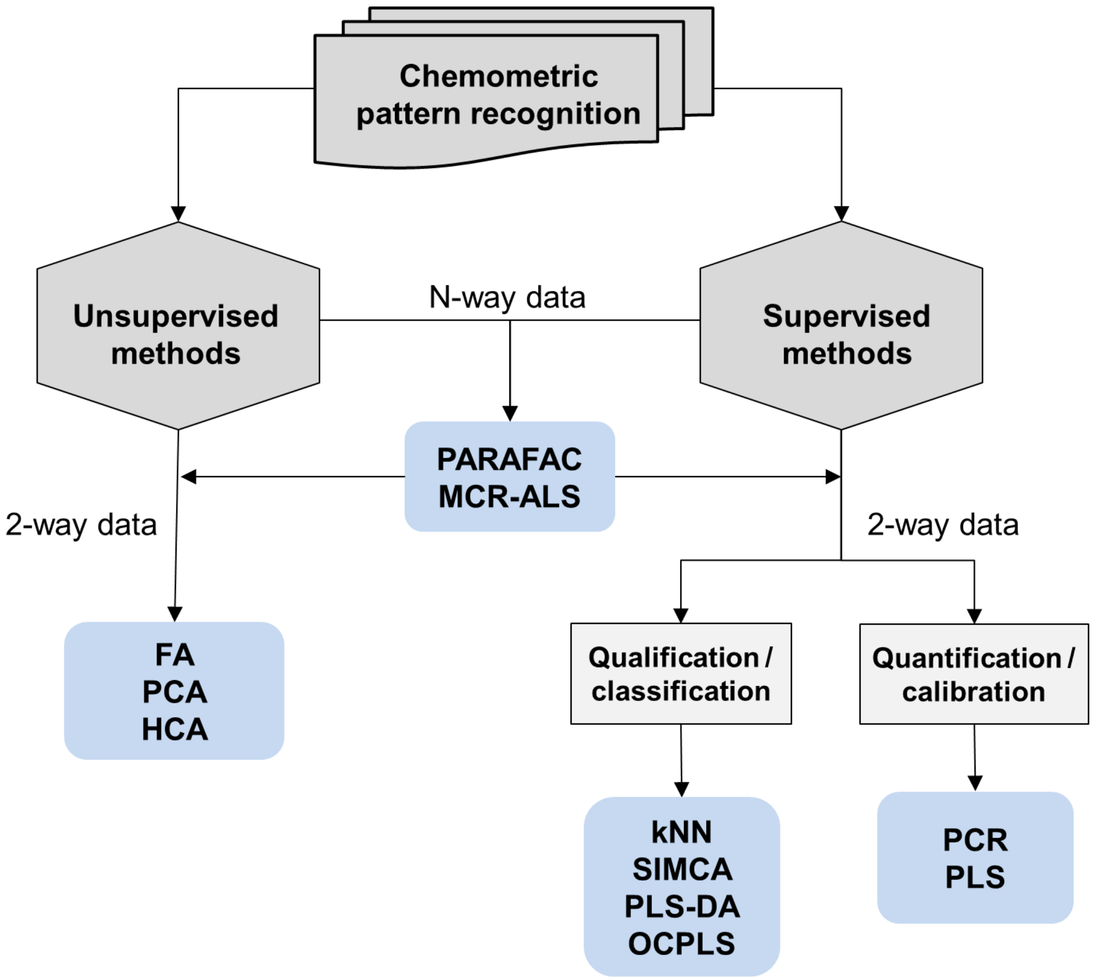
**Figure 4**. Use of the concepts of data mining and machine learning terms in the last 10 years.

**Figure 5**. Schematic illustration of the ensemble methods (D: data-set; sD: data-subset). The light cyan colour shows the decision taken by the classifier tree. In this example the target class is represented by a rectangle in red colour.

**Figure 6**. Graphical description of how RF performs the classification. A probability node (i.e., root node) is represented by a circle and shows the probability of certain results; a decision node is represented by a square and shows a decision to be made; finally, a terminal node shows the result of a decision path (see text for additional explanations on the operation of RF classification).

681 **<Figure 1>**

682

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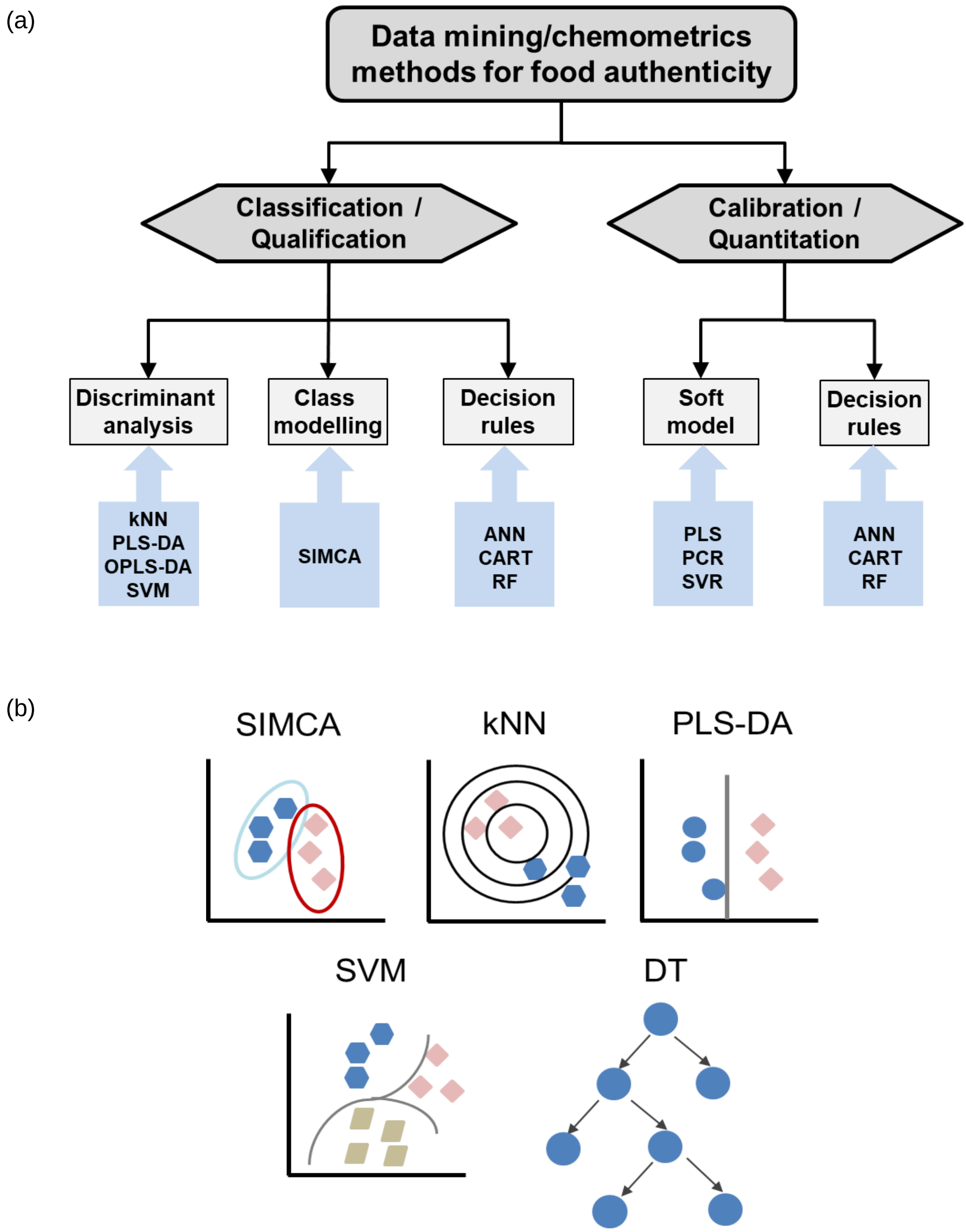
684

**<Figure 2>**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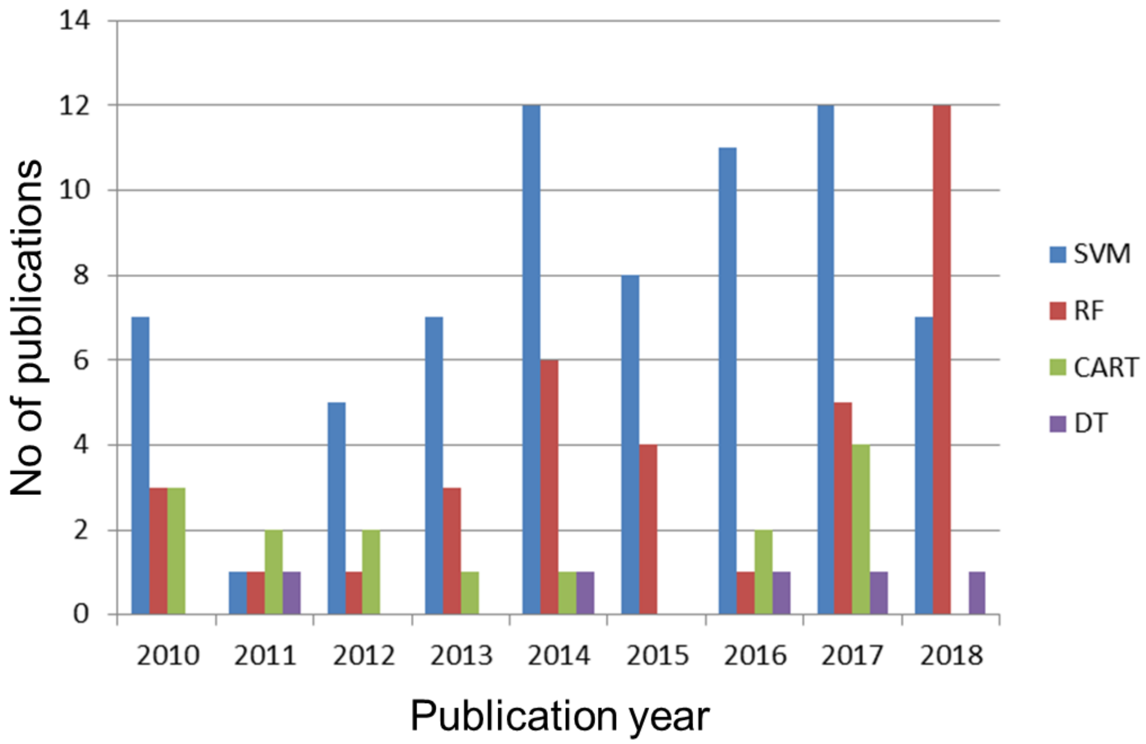
Data mining/chemometrics methods for food authenticity
Classification / Qualification
Calibration / Quantitation
Discriminant analysis
Class modelling
Decision rules
Soft model
Decision rules
kNN
PLS-DA
OPLS-DA
SVM
SIMCA
ANN
CART
RF
PLS
PCR
SVR
ANN
CART
RF
(b)
SIMCA
kNN
PLS-DA
SVM
DT

**<Figure 3>**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2
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2010
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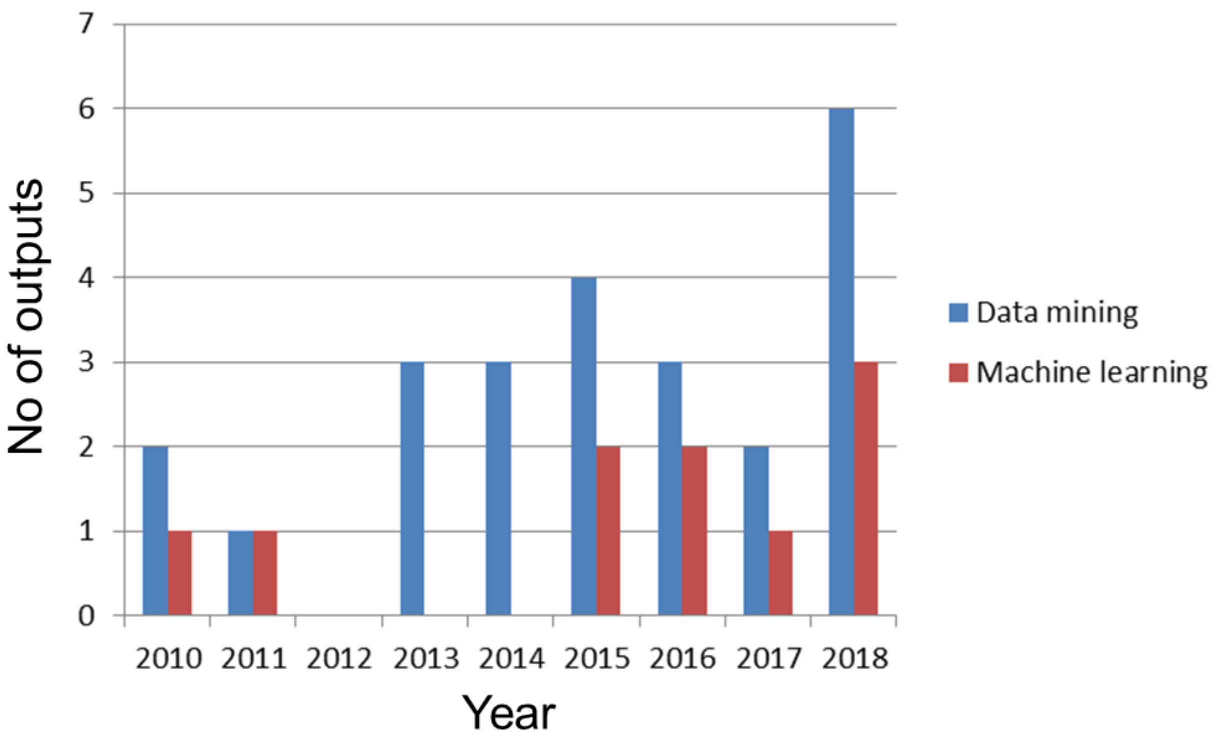
SVM
RF
CART
DT

693 **<Figure 4>**

694

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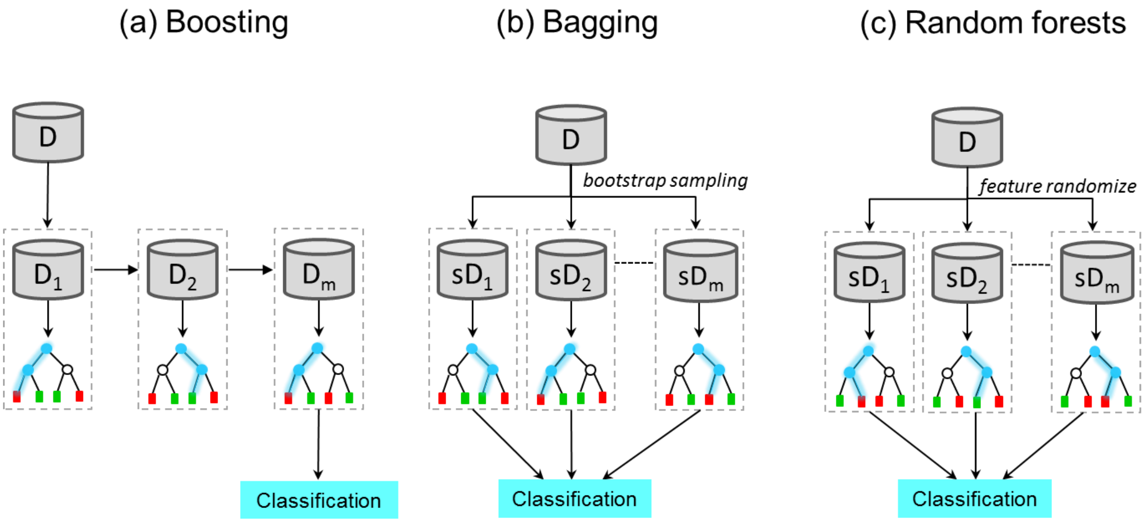
**<Figure 5>**


(a) Boosting
(b) Bagging
(c) Random forests
D
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Classification
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Classification

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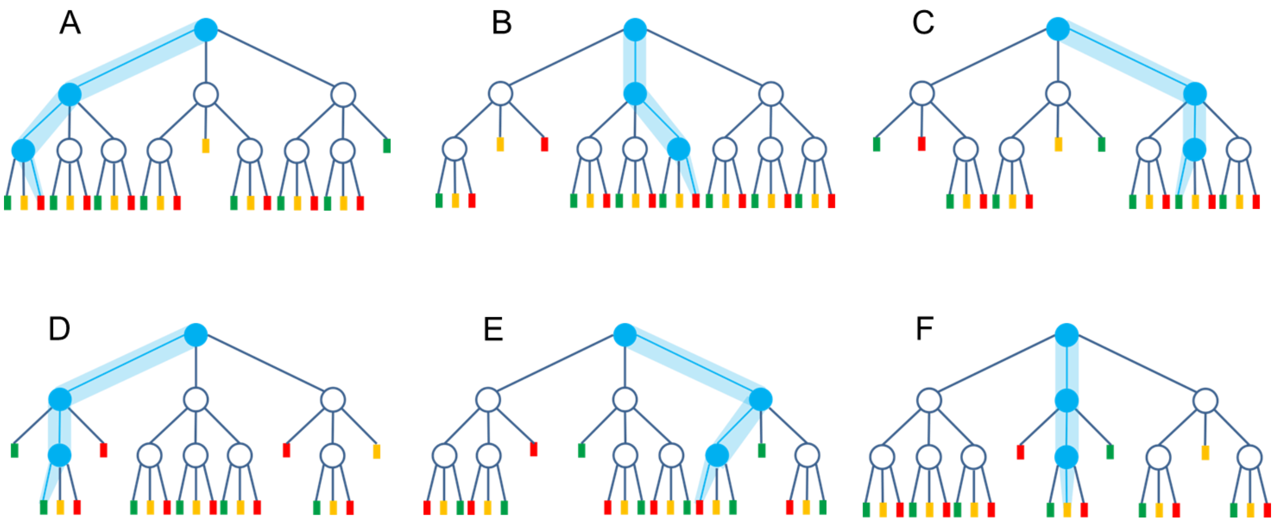

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