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Convenient and accurate method for the identification of Chinese teas by an electronic nose

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RESEARCH ARTICLE

Abstract

A convenient, accurate, and effective approach for the identification of Chinese teas and their production area has been developed. For this, Chinese tea samples from different regions were collected and their odours were analysed by an electronic nose (E-nose). An unambiguous identification of the Chinese teas could not be achieved by means of traditional principal component analysis or linear discriminant analysis methods. Thus, multiple logistic regression (MLR), support vector machines (SVM), and random forests (RF) were employed as alternative to build identification models. The experimental results show that the method aiming within scope based on the RF performs very well, with prediction accuracies and computation times being superior to the two others (MLR and SVM). The results were demonstrated that E-nose could be used in the classification of Chinese teas, when an optimal pattern recognition algorithm is selected. The present study provides a critical outlook on the developments of Chinese teas identification, authenticity control and against adulteration in the Chinese circulation market.

Keywords: Chinese teas, electronic nose, identification, random forest

1. Introduction

Tea is one of the most popular beverages in the world. In particular, Chinese tea has been used since ancient times, during thousands of years of human history and culture. Tea not only has a long history of producing, with special shapes, tastes and fragrance, but has also been attributed to have a beneficial effect to human health (Liao *et al.*, 2001). In 2016, the amount consumed was 2.41 million tons, and the market for Chinese tea was about 44.2 billion US dollars (China Tea Marketing Association, http://www.ctma.com.cn/index.html). Chinese Tea contains substantial amounts of polyphenols, caffeine, volatile oils, vitamins, aroma-forming substances and other compounds that have unique biological activities and health benefits (Sereshti *et al.*, 2013). The main varieties of Chinese tea are classified into six groups (green tea, black tea, oolong tea, white tea, yellow tea, and dark tea). Chinese teas are produced mainly in the Southwest, South, Jiangnan and Jiangbei tea-area's. Geographical origin is an important quality parameter for Chinese tea because its chemical composition varies with climate, water, soil, cultivation method and production process. The aroma information of Chinese tea is one of the main characteristics for tea identification. Chinese teas labelled with false aroma information not only harm the interests of consumers but also damage the reputation of the producers. The aroma of tea is determined by various factors, such as types of tea, its production area, tea making techniques, etc. As tea is traded all over the world, some trade disputes as regard to the types of teas sometimes occur. For example, many tea-producing countries or areas, such as Japan, Taiwan, and Korea, tax imported teas differently, based on the fermentation degree (Wang *et al.*, 2008). However, no internationally recognised standard method for tea classification exists.

Generally, aroma identification is performed by professionals, and the outcomes depend mainly on the acute olfaction and gustation of the tester. This approach is tedious and may be affected by subjective factors or even bias. Other methods of analysis are very helpful, such as gas chromatography, mass spectrometry and liquid chromatography, etc., but they are time-consuming and labour-intensive (Brudzewski *et al.*, 2012). So, there is a call for cost-effective, easy-to-build and convenient detection systems for Chinese teas.

Electronic nose (E-nose), an apparatus designed to mimic the human olfactory perception, can be an innovative measurement system in this area (Qiu and Wang, 2017). The principle of E-nose detection is that the sensor array defines a smell composed of a large amount of different volatiles in a sample's headspace, and then provides an output that represents a 'fingerprint' of all the components for the sample (Hartyáni *et al.*, 2013). The 'fingerprint' described by E-nose sensors is employed to mine potential information about samples based on appropriate algorithm. In recent years, as an objective automated non-destructive technique, E-nose is effective in dealing with odour analysis problems (Ciosek and Wróblewski, 2006; Liu *et al.*, 2013; Sohn *et al.*, 2008), and has been introduced to many fields, such as disease diagnosis (Chapman *et al.*, 2012; Green *et al.*, 2011; Jia *et al.*, 2014, 2016), food engineering (Dai *et al.*, 2015; Gobbi *et al.*, 2015; Loutfi *et al.*, 2015; Roy *et al.*, 2016, 2018), environmental control (Cesare *et al.*, 2008; Romain and Nicolas, 2009), explosive detection (Brudzewski *et al.*, 2012; Ling *et al.*, 2007; Norman *et al.*, 2003), spaceflight applications (Young *et al.*, 2003) and so on.

As we know, each of the sensors in the array in the E-nose, responds to different set of volatile organic compounds in tested substances. Because the differing responses the response of the array is unique for each test sample. The sensor responses are digitised and from these, relevant features are extracted. The appropriate algorithm is one of the key factors of the E-nose application.

In this study, an E-nose was employed as a convenient, automated and alternative technique to identify the different information (category, origin) of Chinese tea based on multinomial logistic regression (MLR), support vector machines (SVM), and random forests (RF). The main aims of this study were: (1) to characterise the 'fingerprints' of Chinese tea using an E-nose; and (2) to identify an algorithm with the best performance to discriminate category and origin of Chinese teas.

2. Materials and methods

Instruments and equipment

In this work, a PEN3 E-nose (Portable Electronic Nose, Airsense Analytics GmbH, Hagenover, Schwerin, Germany) was used. This E-nose has an array of 10 different metal oxide sensors (MOS) positioned inside a small chamber (1.8 ml). Each MOS sensor detects a different set of volatile molecules during the process, resulting in a change of the conductivity of sensors. Therefore, a unique set of response curves of the sensor array can be obtained for each distinct sample. The nomenclature and characteristics of the sensors used are listed in Table 1.

Experimental samples

Six kinds of Chinese tea (Table 2) were tested by means of the PEN3 E-nose. All the experimental samples were purchased from an official tea market (Beijing) authorised by the China Tea Marketing Association.

Experiments and data acquisition

All the experiments were carried out in the author's lab, the whole samples were measured at a temperature of 25±1 °C and air relative humidity of 39±2%. As shown in Figure 1, the experiments were carried out on six groups of samples (class 1-6). For each group, 20 samples were prepared. Thus, a total of 120 tea samples were used in the experiments. For each experiment, 4 g of the respective tea was put into a vial (50 ml) and was allowed to equilibrate with the air in the vial for 90 min. Five separate experiments were carried out every day. The experiments lasted for 4 days. As shown in Figure 2, before measurement, the filtered air was suctioned in reverse through the E-nose to flush the sensor array and the gas line with the valve 2 open. This flushing lasted for 100 seconds. For the measurement process, valve 2 was closed and valve 1 was opened, to pump the headspace gas of the sample into the sensor chamber at a constant rate of 10 ml/sec via a Teflon-tubing connected to a needle. The measurement time was 100 s, and data were acquired by the WinMuster software (version 1.6.2.18, Airsense Analytics GmbH) every second. For the whole 120 samples, only

Table 1. The standard sensor array in a PEN3 E-nose.[1]

No. Sensor	Sensor	Object substances for sensing
MOS1	W1C	aromatic compounds
MOS2	W5S	sensitive to nitric oxides
MOS3	W3C	ammonia, aromatic compounds
MOS4	W6S	hydrogen
MOS5	W5C	alkane, aromatic compounds
MOS6	W1S	sensitive to methane
MOS7	W1W	sensitive to sulphide
MOS8	W2S	sensitive to alcohol
MOS9	W2W	aromatic compounds, organic sulphur compounds
MOS10	W3S	sensitive to alkane

[1] MOS = metal oxide sensors.

Table 2. Details of the teas in the study.

No.	Samples	Year of production	Place of origin	Price ($/50 g)	Category
1	Mudan White	2017	Fuding City (Fujian Province)	5.9	White tea
2	Biluochun	2017	Dongting Hill (Jiangsu Province)	8.5	Green tea
3	Lapsang Souchong	2017	Wuyishan City (Fujian Province)	9.8	Black tea
4	Huoshan Huangya	2017	Huoshan Country (Anhui Province)	9.9	Yellow tea
5	Fermented Pu-erh	2017	Puer City (Yunnan Province)	4.4	Dark tea
6	Tieguanyin	2017	Anxi Country (Fujian Province)	7.9	Oolong tea

the data acquired in steady phase (10 data points) was kept for the later analysing, a 1,200×10 matrix formed the dataset (dataset A). Furthermore, a new testing samples set was used to verify the generalisation of those three built regression models. Note that all the samples were produced in 2017 and purchased from the same manufacturer and each kind of teas has 10 samples. After all samples had been tested by the PEN3 e-nose, similar to the process of building dataset A, a 600×10 matrix formed the new dataset (dataset B).

3. Results

Response curves and feature

To describe the sensor response to a given tea sample, the relative change of the sensors' conductance during the measurement was calculated by using the formula R=G0/G, where R is the response, G0 is the conductance of the sensor in reference air, and G is the conductance of the sensor when exposed to the sample vapor. Figure 3 shows the typical response signals of the sensor array to the six tea samples during 100 s of measurement, respectively. Each response curve represents the variation in conductivity of each sensor with time when the tea volatiles reached the measurement chamber. In the initial period, the R value of each sensor was low, then increased quickly, typically reaching a peak value before a slow decrease over the next seconds. After about 76 s, steady signals could be obtained, with the exception of the MOS7 sensor responding to the Tieguanyin sample (Figure 3F).

Of the ten sensors, five (MOS7, MOS9, MOS2, MOS6 and MOS8) responded to the volatiles in the teas and their response sensitivity levels were distinct for each kind of sample. Thus, six fingerprints could be obtained, showing the difference between the six teas (Figure 4).

Generally, several kinds of feature (mean-differential coefficient value (Yun *et al.*, 2007) and response area value (Wei *et al.*, 2013)), extracted from E-nose signals, were used in pattern recognition algorithms. Here, we used a simpler feature parameter, i.e. the stable value. Since the detection lasted 100 s, and the response value of each sensor stabilised after about 75 s, the value after 75th second of each sensor was regarded as the stable value. So, 10 data points (from 85th to 94th seconds; the black dotted-line area in Figure 3A) were used as input features for classification in our study.

Principal component analysis classification

Principal component analysis (PCA) is a linear, unsupervised and pattern recognition technique used for analysing, classifying and reducing the dimensionality of numerical datasets with a minimum loss of information (Fluky, 2012) in a multivariate problem (Chen *et al.*, 2013).

In the study, PCA was used first for classification of the same batch experimental data. PCA was operated with the raw data imported into the WinMuster software. The accumulated variance contribution rate of the first two principal components (PC) included sufficient information about the samples, which was of 99.96%. And the variance of PC1 and PC2 accounted for 99.56 and 0.40%, respectively. Thus, PC1 and PC2 were utilised to make a score plot with standardised scores.

As shown in Figure 5A, the six tea samples of which data were acquired from same experiment are distinguished correctly though a small overlap exists. However, in Figure 5B, where we tried to classify using all the experimental data that was collected on the first day (exp. 1-5), the PCA was not qualified anymore because during the process of dimension reduction, some information was lost and ambiguity could arise.

Linear discriminant analysis classification

Linear discriminant analysis (LDA) is a generalisation of Fisher's linear discriminant and has been widely used in statistics, pattern recognition and machine learning to find a linear combination of features that characterises or separates two or more classes of objects or events. It projects high-dimensional data onto a low dimensional space where the data achieves maximum class separability and considers the information related to both the within-

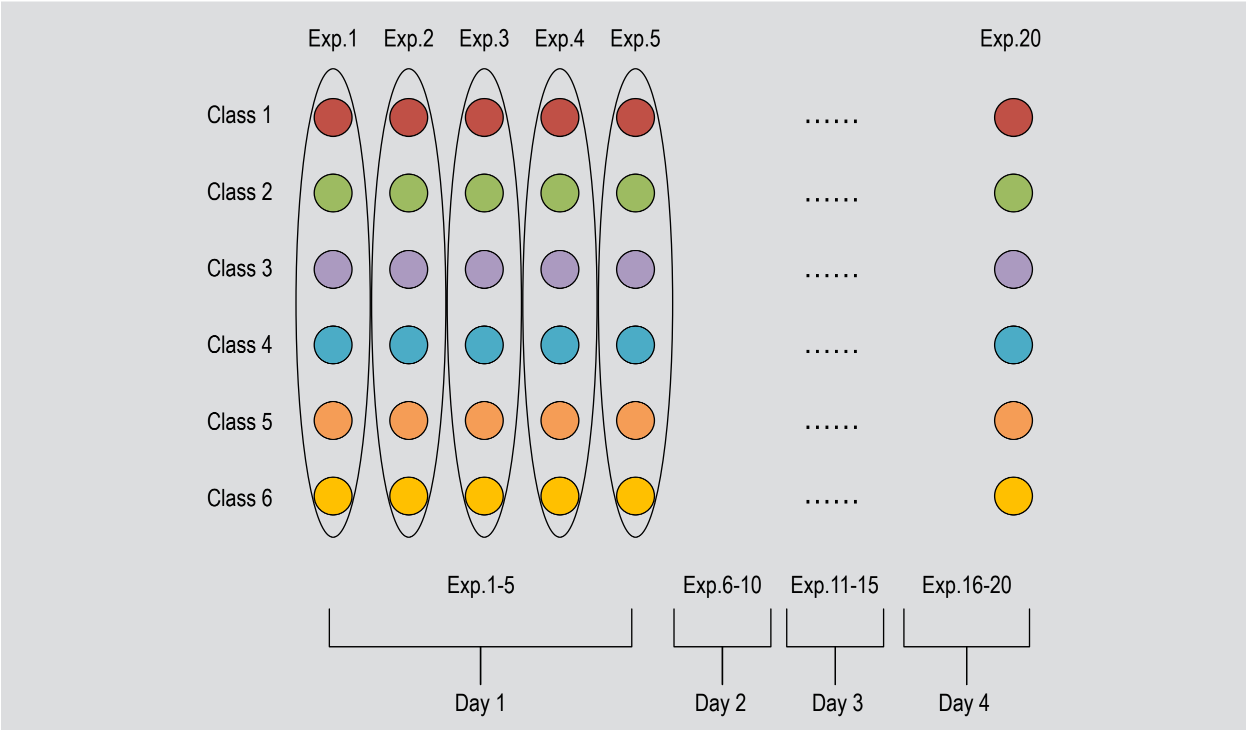

Figure 1. Illustration of the experimental program. Class codes: 1 = MuDan White tea; 2 = Biluochun tea; 3 = Lapsang Souchong tea; 4 = Huoshan Huangya tea; 5 = Fermented Pu-erh tea; 6 = Tieguanyin tea.

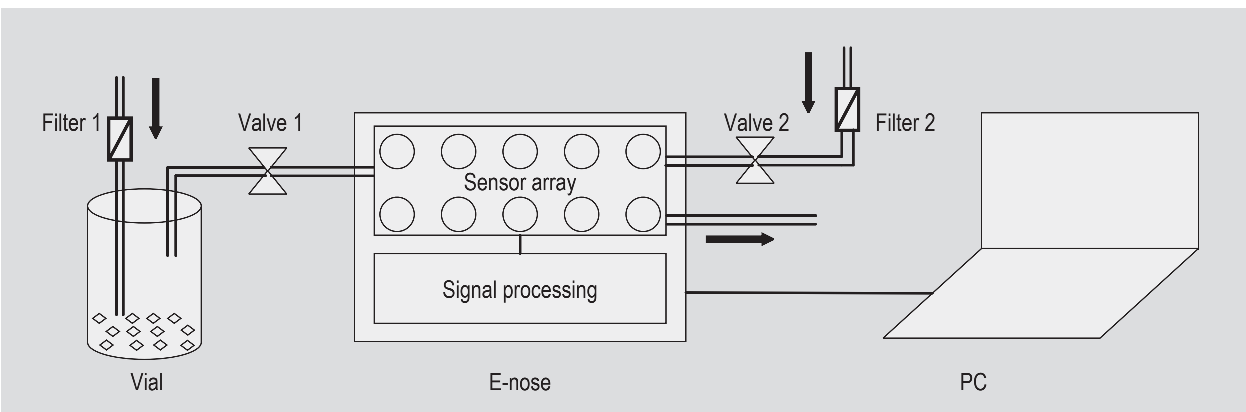

Figure 2. Illustration of the test for a given sample.

class as well as the between-class distribution (Banerjee *et al.*, 2012).

As shown in Figure 6A, for the same experiment, the cumulative variance of the first two canonical linear discriminant functions (LD) reached 98.79% (greater than 85%), a score plot was depicted with LD1 and LD2, whose variances were of 69.39 and 29.40%, respectively. The figure suggests that the tested samples of tea could still be distinguished without effort. As shown in Figure 6B, LDA is poor for the distinction of all the samples, except for Tieguanyin tea that was tested on the first day.

Pattern recognition

Multiple logistic regression

In statistics, logistic regression or logit model is a regression model where the dependent variable is categorical (Barrett, 2009). Binomial or binary logistic regression deals with situations in which the observed outcome for a dependent variable can have only two possible types, '0' and '1' (which may represent, for example, 'yes' vs 'no' or 'win' vs 'loss') by means of logistic function (sigmoid function). MLR is frequently the method of choice when the response is a

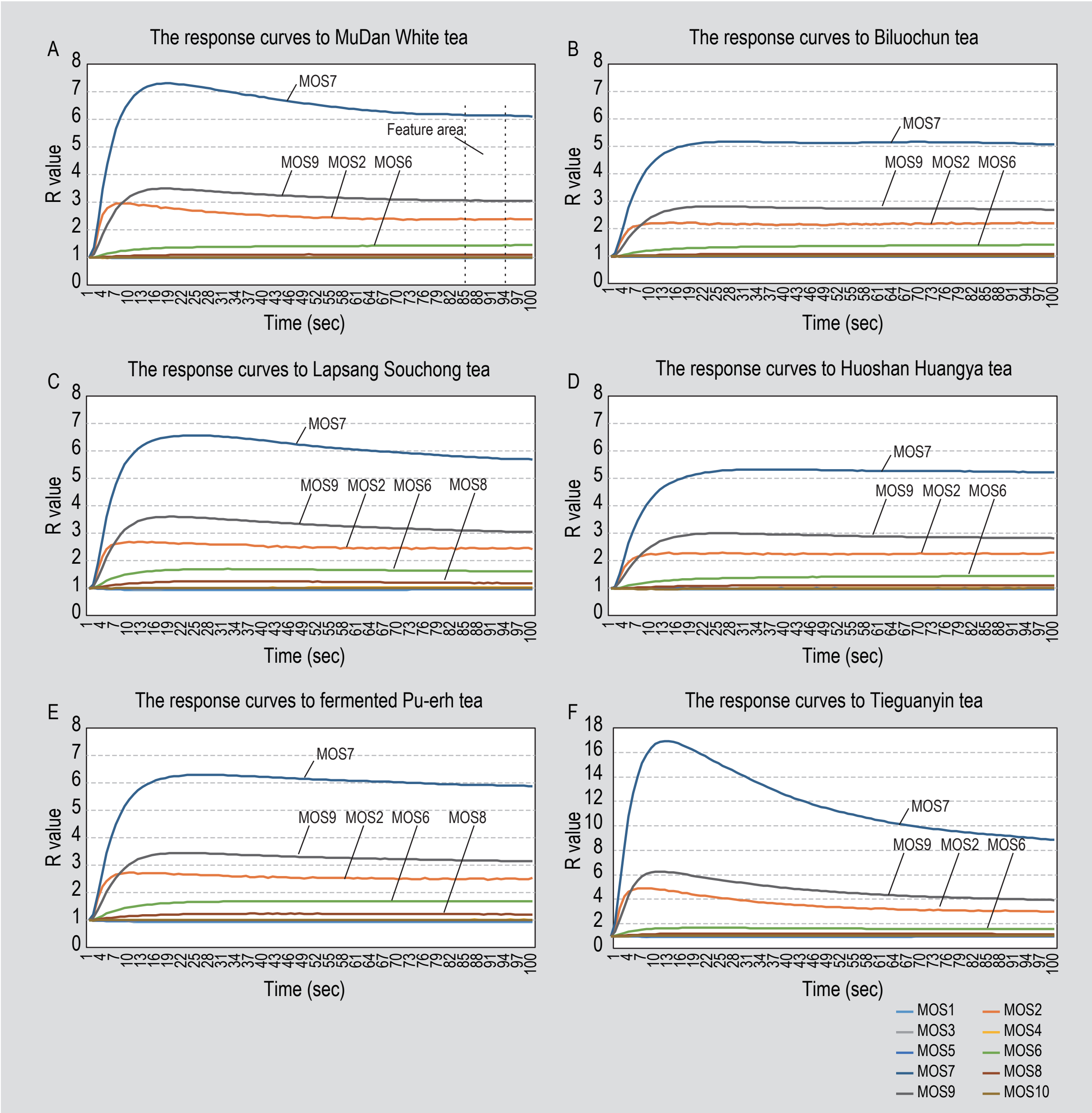

Figure 3. Response curves of the ten sensors in the E-nose to six kinds of tea samples: (A) MuDan White tea; (B) Biluochun tea; (C) Lapsang Souchong tea; (D) Huoshan Huangya tea; (E) Fermented Pu-erh tea; (F) Tieguanyin tea; metal oxide sensors (MOS) codes are explained in Table 1.

qualitative variable, with two or more mutually exclusive unordered response categories (Castilla *et al.*, 2017).

As shown in Figure 7A and B, two structures of the MLR are available when it is employed for multi-classification (n≥3; n is the number of categories) tasks. For one-vs-rest structure, n classifiers are needed, to train a logistic regression classifier (one versus rest) for each class samples. And for many-vs-many structure, n(n-1)/2 (n is the number of categories) classifiers are needed, to train n-1 logistic regression classifiers (many versus many) for each class samples. Both structure A and B, on a new input, to make a prediction, pick the maximum probability vin output of all the classifiers, the class associated with the model is considered to be the final result. It can be deduced from the above description that the one-vs-rest structure is faster, while the many-vs-many structure is more accurate. In our study, we have used the second one with higher accuracy.

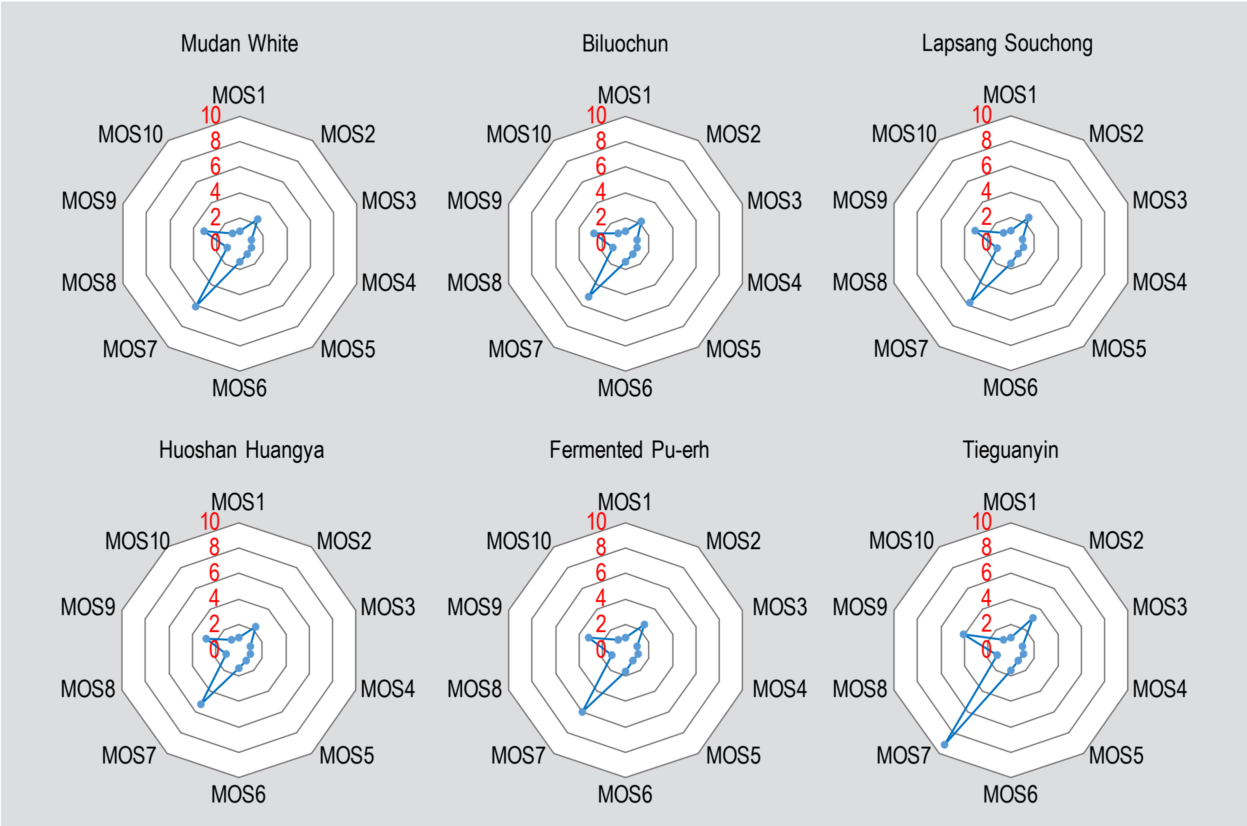

Figure 4. The fingerprints to six kinds of tea samples at the 90th second; metal oxide sensors (MOS) codes are explained in Table 1.

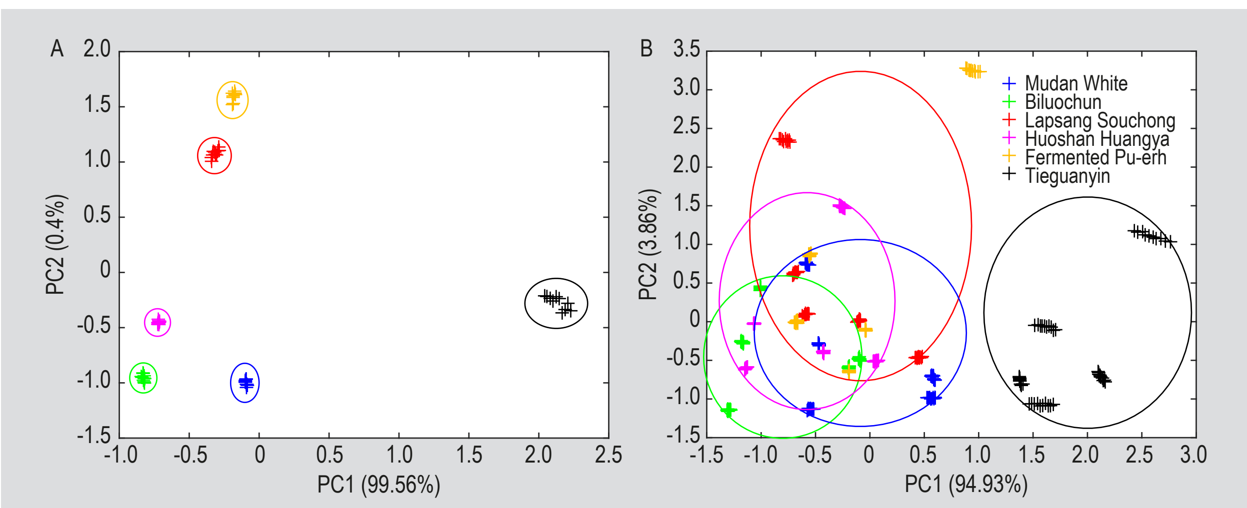

Figure 5. Principal component analysis (PCA) score plot responses to samples with PC1, and PC2. (A) The same ergodic experimental data; (B) The whole experimental data collected on the first day.

Support vector machines

SVM are supervised learning models with associated learning algorithms that analyse data used for classification and regression analysis (Cortes and Vapnik, 1995). Operation of the SVM algorithm is based on finding the hyperplane that gives the largest margin to the training examples. Therefore, the optimal separating hyperplane maximises the margin of the training data.

As shown in Figure 8A, for a given training set:

$$D=\{(x_1,y_1),(x_2,y_2), \ldots , (x_m,y_m)\}, y_i \in \{-1,+1\}$$

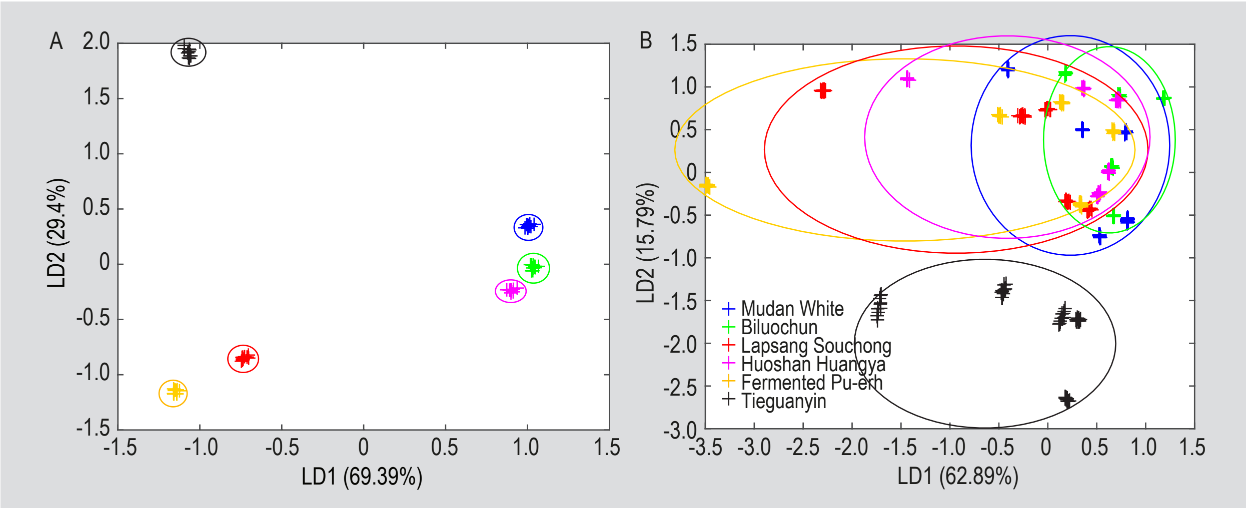

Figure 6. Linear discriminant analysis (LDA) score plot responses to samples with LD1, and LD2. (A) The same ergodic experimental data; (B) The whole experimental data collected on the first day.

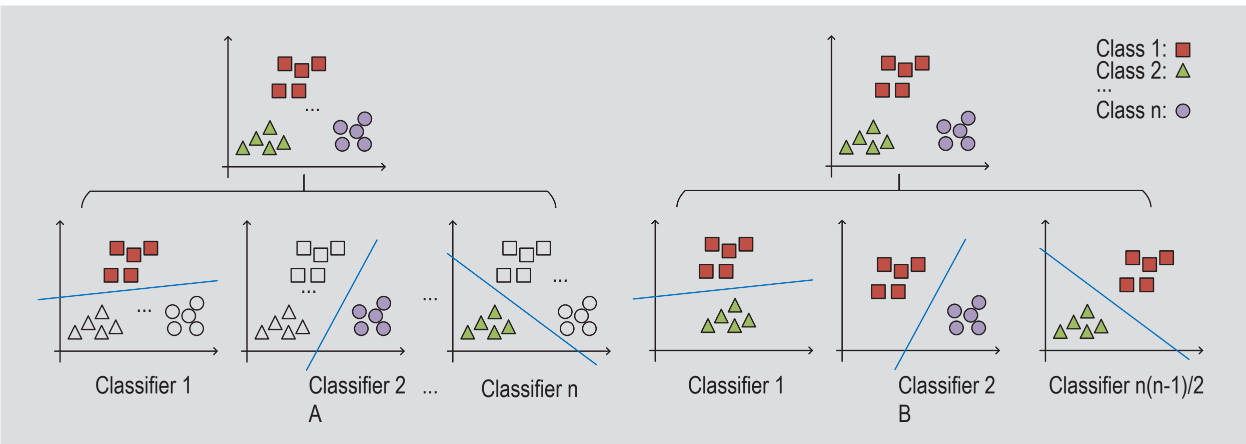

Figure 7. Illustration of the multiple logistic regression (MLR) for multi-classification task. (A) One-vs-rest structure; (B) Many-vs-many structure.

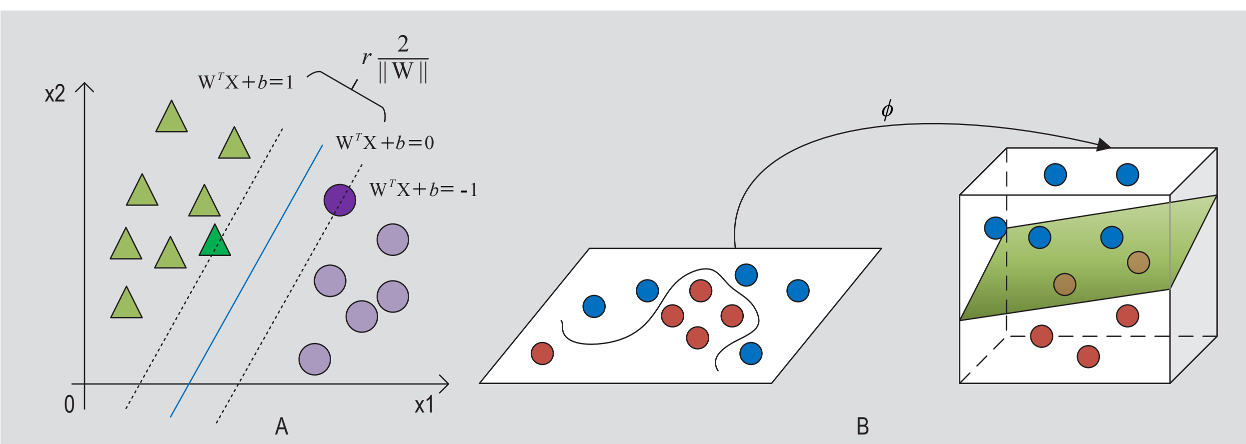

Figure 8. Illustration of support vector machines (SVM) basic principle. (A) SVM basic structure; (B) Feature mapping

Obviously, when the selected hyperplane is $w^Tx+b=0$ it has the best robustness and generalisation ability. The basic idea of SVM is to use linear models to implement nonlinear class boundaries through some nonlinear mapping of the input vector into the high-dimensional feature space (Wang *et al.*, 2009).

For nonlinear separable classification problems, the SVM applies a kernel function K (v_i, v_j) to transform the original space to a higher-dimensional space, and a hyper plane is constructed in the higher-dimensional space to solve problems of nonlinear separable classification in the original low-dimensional space as shown in Figure 8B. The four most known kernels are commonly used: linear, polynomial, radial basis function, and sigmoid (Li *et al.*, 2017a). In this work, we applied an SVM algorithm (Pardo and Sberveglieri, 2005) for classification of teas. A radial basis function was chosen as the kernel function. To optimise penalty parameter (C) and kernel parameter gamma (c) in the SVM model, a grid search method with exponentially growing sequences of C and c were applied.

Random forests

RF is a classification method consisting of independent classification trees. The prediction of the classification is obtained by the majority voting of the classification trees that have been formed (Kuswanto *et al.*, 2017). In this work, RF was employed in classification of the Chinese tea samples. The illustration of the algorithm is shown in Figure 9.

First, for a given training set, some bootstrap samples (the amount depends on the number of classification and regression trees) were obtained by bootstrapping. Secondly, it was crucial for RF algorithm to growing classification and regression trees (CARTs). Every CART is built by using random vectors. The general approach used to insert random vectors in the formation of the tree is to choose the number of variables (N_F) in the random subset at each node, as N_F attributes input to be split at each node in the CART to be formed. Experimentally, the N_F can be determined by using the formula (Li *et al.*, 2017b):

$$N_F=\log_2(M+1)$$

where M is the total number of features. Finally, a RF classifier was built by growing CARTs and training was supervised, and it determined the final classification results based on the CARTs' voting (majority rule).

Comparison of classification results

The ultimate performance of the above three classifier on testing samples is shown in Figure 10. Although MLR-based method achieved accuracy rate of 97.2% and consumed less time, its robustness is not good. In repeated experiments, the MLR method often has to re-adjust the parameters in order to achieve a good result. Both SVM-based and RF-based methods achieved an accuracy rate of 100%, but the training time of SVM is five times the time of RF.

In summary, in the case of the current sample size, the RF-based classification method is an ideal choice for the

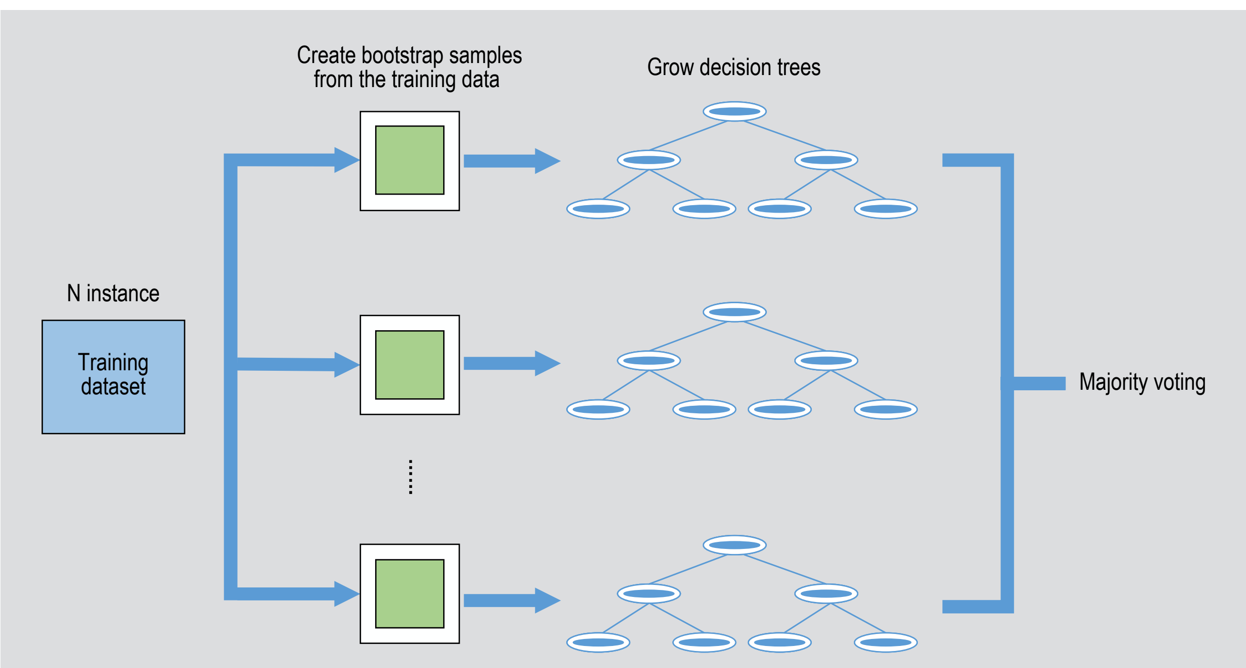

Figure 9. Illustration of the random forests algorithm.

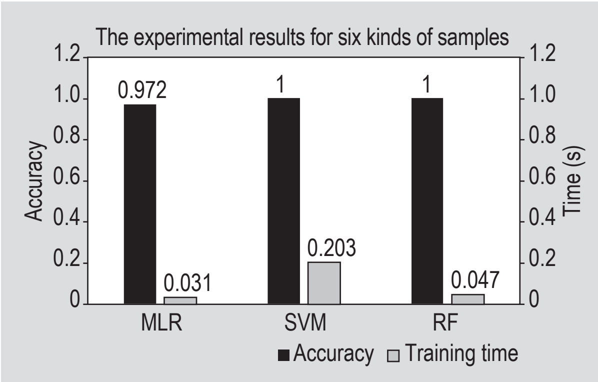

Figure 10. Three methods to identify the accuracy and training time of the comparison results. MLR = multiple logistic regression; SVM = support vector machines; RF = random forests.

distinction of Chinese tea. So, for a real application, we can conclude that training the classifier with data collected at different time (different dates) should be enough to palliate the negative effect of deviation during the measuring, and improve the overall classification performance.

4. Conclusions

The discrimination result of RF is better than that of PCA, LDA, MLR and SVM. The essence of PCA and LDA is dimension reduction of data, by which the classification result relies on human observation. Especially, PCA is useful to identify the tea type clusters but it only fits to a handful of samples at a time. If the number of tea samples to be investigated increases, the identification of the tea types clusters in a crowded PCA plot would become difficult. The performance of LDA in multiple experiments is far less than that of a single experiment. Also, the performance of MLR-based algorithm indicates that the MLR model suffers from underfitting because it is not complex enough to capture the pattern in the training data well. Both SVM and RF have achieved good experimental results, however according to our experiments, the training time of SVM model is five times of that of the RF model.

In conclusion, the results are encouraging, and it was demonstrated that E-nose, a convenient, accurate, and non-destructive practical means, could be used in the classification of Chinese tea, when an optimum pattern recognition algorithm is selected. Further results have revealed that E-nose technology has excellent sensitivity and selectivity to the teas with different aroma. The present study provides a critical outlook on the developments of Chinese tea identification, authenticity control and against adulteration in the Chinese circulation market.

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