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Quality grade classification of China commercial moxa floss using electronic nose

A supervised learning approach

Min Yee Lim, PhD[a], Jian Huang, MD, PhD[b,*], Fu-rong He, MD[c], Bai-xiao Zhao, MD, PhD[d,*], Hui-qin Zou, PhD[e], Yong-hong Yan, PhD[f], Hui Hu, MD, PhD[b], Dong-sheng Qiu, MD[g], Jun-jie Xie, MD[g]

Abstract

Moxa floss is the primary material used in moxibustion, an important traditional Chinese medicine therapy that uses ignited moxa floss to apply heat to the body for disease treatment. Till date, there is no available data regarding quality control of different grades of moxa floss. The objectives of this study were to explore the probative value of the electronic nose (e-nose) in differentiating different quality grades of commercial moxa floss sold in China, and to investigate if data mining techniques could be used to optimize the sensor array while retaining classification accuracy of the samples. The e-nose with 12 metal oxide semiconductor type sensors was used to analyze the odor profiles of 15 commercial moxa floss samples of different quality grades. Feature selection algorithms using principal component analysis (PCA) and BestFirst (BC) coupled with correlation-based feature subset selection (CfsSubsetEval) method were used to obtain the most efficient feature subsets. Results for the BC feature selection method identified 3 optimized sensors (S2, S6, and S11), suggesting that aromatic compounds relate more to the identification of the samples. Radial basis function (RBF), multilayer perceptron (MLP), and random forests (RF) performed well in discriminating the samples, retaining prediction accuracies above 85%, which achieved cost-effectiveness and operational simplicity, while retaining prediction accuracy. The e-nose could be a rapid and nondestructive method for objective preliminary classification of quality grades of moxa floss and may be used for future studies related to moxa products safety and quality.

Abbreviations: ANN = artificial neural network, BC = BestFirst, CfsSubsetEval = correlation-based feature subset selection, E-nose = electronic nose, MLP = multilayer perceptron, PCA = principal component analysis, RBF = radial basis function, RF = random forests.

Keywords: electronic nose, moxa floss, moxibustion, pattern recognition algorithms

1. Introduction

Moxibustion is an integral therapy of Chinese medicine that uses ignited moxa floss as the primary material to apply heat to certain points or body areas for disease treatment. It is a treatment method that is used extensively both in hospitals and homes in countries such as China, Japan, and South Korea, and has gained more acceptance worldwide due to its reported effects in disease treatment and prophylactic capabilities.[1–3]

Moxa floss has a distinctive odor that contributes to one of the most important attributes associated with moxa floss quality. The odor of moxa floss is characterized by its volatile compounds which are highly dependent on the species, geographical origin,

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JH and B-xZ should be considered as co-corresponding authors.

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[a] International School, [b] Acupuncture Department, Dongfang Hospital, Second Affiliated Hospital of Beijing University of Chinese Medicine, [c] School of Acupuncture-Moxisbution and Tuina, Fujian University of Traditional Chinese Medicine, Fujian, [d] Chinese Medicine, [e] Library, [f] School of Chinese Materia Medica, Beijing University of Chinese Medicine, Beijing, [g] Acupuncture Department, Xiamen Hospital, Affiliated Hospital of Beijing University of Chinese Medicine, Xiamen, China.

[*] Correspondence: Jian Huang, Acupuncture Department, Dongfang Hospital, Second Affiliated Hospital of Beijing University of Chinese Medicine, Beijing 100078, China (e-mail: xiamenhuangjian@sina.com); Bai-xiao Zhao, Chinese Medicine, Beijing University of Chinese Medicine, Beijing 100029, China.

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and collection time of its starting material mugwort leaves.[4–6] The production of moxa floss also comprises multiple steps including drying, pulverization, and sifting of mugwort leaves, which can change the volatile profile of moxa floss. Studies using gas chromatography–mass spectrometry (GC–MS) have reported different classes of volatile compounds, including aldehydes, ketones, alcohols, esters, terpenoids, and acids in the chemical pattern of mugwort leaves.[4] However, analysis by GC–MS requires long and complicated pretreatment preparation steps that is both expensive and requires an effective extraction method to ensure the completeness and representativeness of the experimental odor profile. All of these drawbacks have limited the use of GC–MS as an easy-to-operate method in the analysis of the volatile fraction of mugwort leaves and moxa floss.

The electronic nose (e-nose) is a technology that has been developed over the past decade mimicking the human olfactory perception to perform odor evaluation on a continuous basis. It responds to the mixture of volatile constituents as a whole without having to detect individual chemical constituents and allows for non-destructive assessment in a rapid and reproducible manner.[7,8] The cross-reactive sensor array in the e-nose interacts with the volatile compounds of the sample and generates multivariate responses that can be used as an "electronic fingerprint" to discriminate the odors through pattern recognition methods.[9] The e-nose technology has shown to be a promising method for the identification and quality control of Chinese medicinal material such as Radix Angelica Sinensis,[10] ginseng,[11] musk,[12] and Asari Radix et Rhizoma.[13]

There is very little information available in the literature to date investigating the properties of moxa floss, which is the primary material used in moxibustion and would invariably affect the safety and quality of this treatment. We hypothesize that using the e-nose combined with data mining techniques can provide a rapid, nondestructive, and cost-effective analysis for the preliminary classification of moxa floss according to its quality grades. Therefore, the main objectives of this research were: to explore the probative value of the e-nose in differentiating different quality grades of commercial moxa floss sold in China, and to investigate if data mining techniques could be used to optimize the sensor array while retaining classification accuracy of the samples.

2. Materials and methods

2.1. Experimental samples

Fifteen commercial moxa floss samples of 3 different quality grades as specified on the product market labels were randomly purchased in Beijing. The samples were verified by one of the authors (MYL). Specific details of the products are listed in Table 1. Ethical approval was not necessary as this study did not involve human participants.

Table 1

Details of China commercial moxa floss samples.

No.	Place of manufacture	Quality grade as printed on the product label
1	Suzhou, China	Normal grade
2	Henan, China	
3	Henan, China	
4	Henan, China	
5	Beijing, China	
6	Henan, China	3-years grade
7	Shanghai, China	
8	Hubei, China	
9	Henan, China	
10	Beijing, China	
11	Henan, China	Top grade
12	Henan, China	
13	Henan, China	
14	Henan, China	
15	Beijing, China	

2.2. Electronic nose analysis

All samples were analyzed on an α-FOX3000 (Alpha MOS, Toulouse, France) combined with a headspace auto-sampler (Fig. 1). The instrument consists of a sampling apparatus, a detector unit containing an array of sensors, air generator equipment, HS-100 auto-sampler, and pattern recognition software (Alpha Soft V11) for data recording. The sensor array is composed of 12 metal oxide semiconductor (MOS) type chemical sensors divided into chambers as 3 types: T, P, and LY. Table 2 shows a list of the sensors used and their main applications.

2.3. Experiment procedure: optimization of headspace temperature and headspace time

0.20 g of sample was accurately weighed and placed in a 10 mL glass jar, sealed and loaded into the auto-sampler tray. To optimize the main parameters, different headspace temperatures (35, 50, and 80 °C) and headspace times (300, 480, and 600 seconds) were investigated. Based on the response intensities and relative standard deviation of values from 6 parallel tests, the headspace temperatures and times were selected to be 50 °C and 480 seconds, respectively.

2.4. Sampling procedure

Forty-five samples (3 samples for each sampling point) were used in the experiment. The experiment started when the resistance of the gas sensors remained stable. After headspace equilibration time of 480 seconds at 50 °C under agitation (500 rpm), 2500 μL of headspace gas was injected into the testing chamber and the measurement began. The acquisition time was 200 seconds to allow sufficient time for the sensors to obtain a stable value. The maximum response points were automatically recorded for each of the 12 sensors and used to generate the electronic nose response curves. After sample analysis, the clean phase was activated and the system was purged for 400 seconds with processed pure air prior to the next sample injection to return the sensors to their baseline values. Each sample was measured 3 times and the average of the results was used for subsequent statistical analysis.

2.5. Data analysis: feature selection

Principal component analysis (PCA) is an unsupervised method that reduces multidimensional data with a minimum loss of information into orthogonal coordinates based on maximum variance by linear projection.[13] In this study, PCA was used to derive the first 2 principal components to allow the visualization

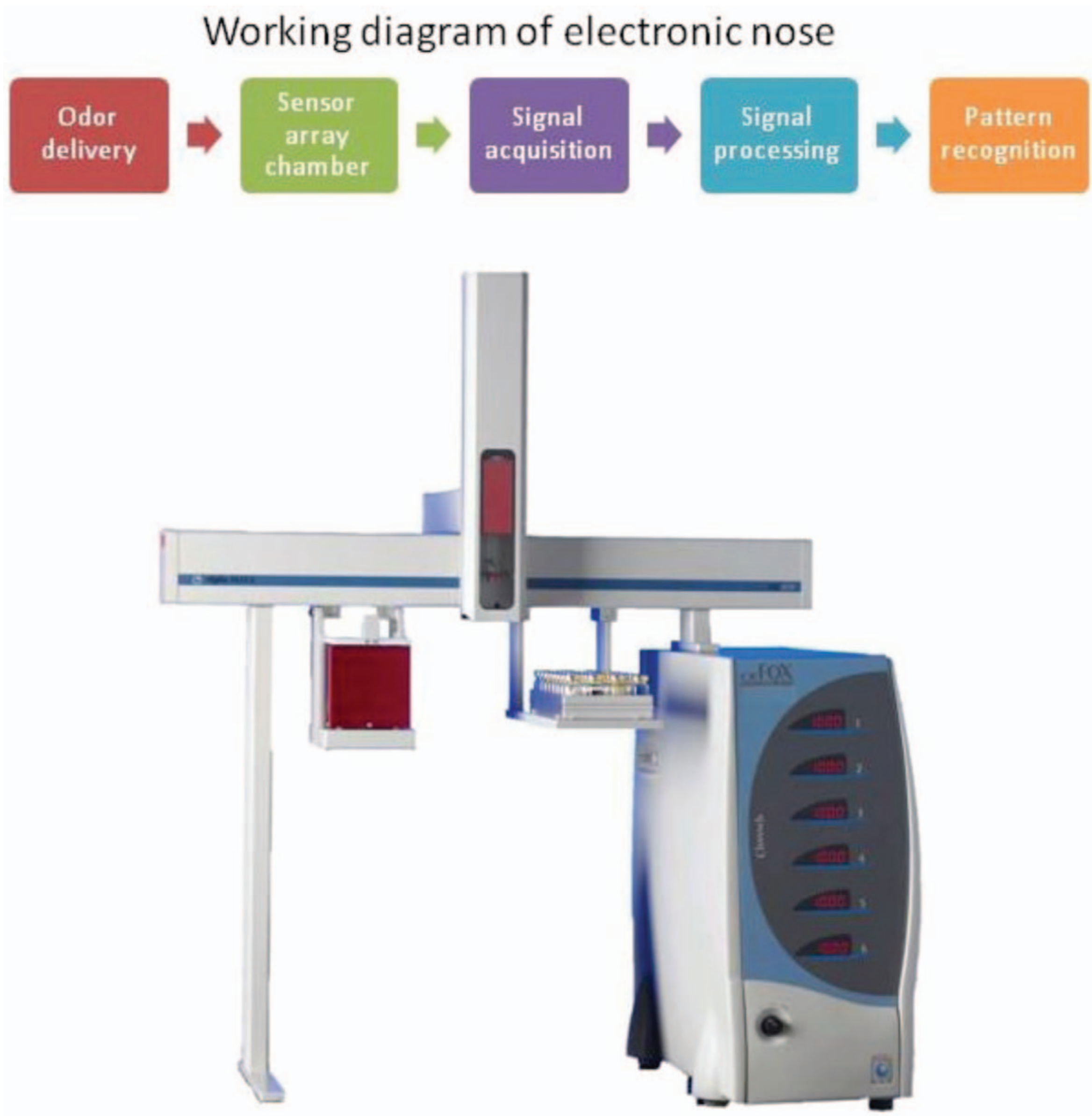

Figure 1. Electronic nose α-FOX3000 with working diagram.

Table 2

Sensors used and their main applications in electronic nose.

Number in array	Name	Main application
S1	LY2/LG	Oxidizing gas
S2	LY2/G	Ammonia, carbon monoxide
S3	LY2/AA	Ethanol
S4	LY2/GH	Ammonia/organic amine
S5	LY2/gCTL	Hydrogen sulfide
S6	LY2/gCT	Propane/butane
S7	T30/1	Organic solvents
S8	P10/1	Hydrocarbons
S9	P10/2	Methane
S10	P40/1	Fluorine
S11	T70/2	Aromatic compounds
S12	PA/2	Ethanol, ammonia/organic amine

of the information into clusters and outliers to provide an exploratory overview.

This study also employed the data mining software, WEKA 3.6.11 (New Zealand), and used the BestFirst (BC) coupled with Correlation-based Feature Subset Selection (CfsSubsetEval) method to prevent over-fitting and reduce the possibility of chance effects. BC is a type of feature extraction method, which can screen out the characteristic parameter vectors with high relevance to each other, thereby generating an optimum set of MOS sensors for final identification. CfsSubsetEval evaluates the worth of a feature subset by considering the individual predictive ability of each feature along with the degree of redundancy between them, which is acceptable with respect to both speed and accuracy. Ten-fold cross-validation method was applied to verify the classification accuracy. Classification accuracy above 80% is considered acceptable.

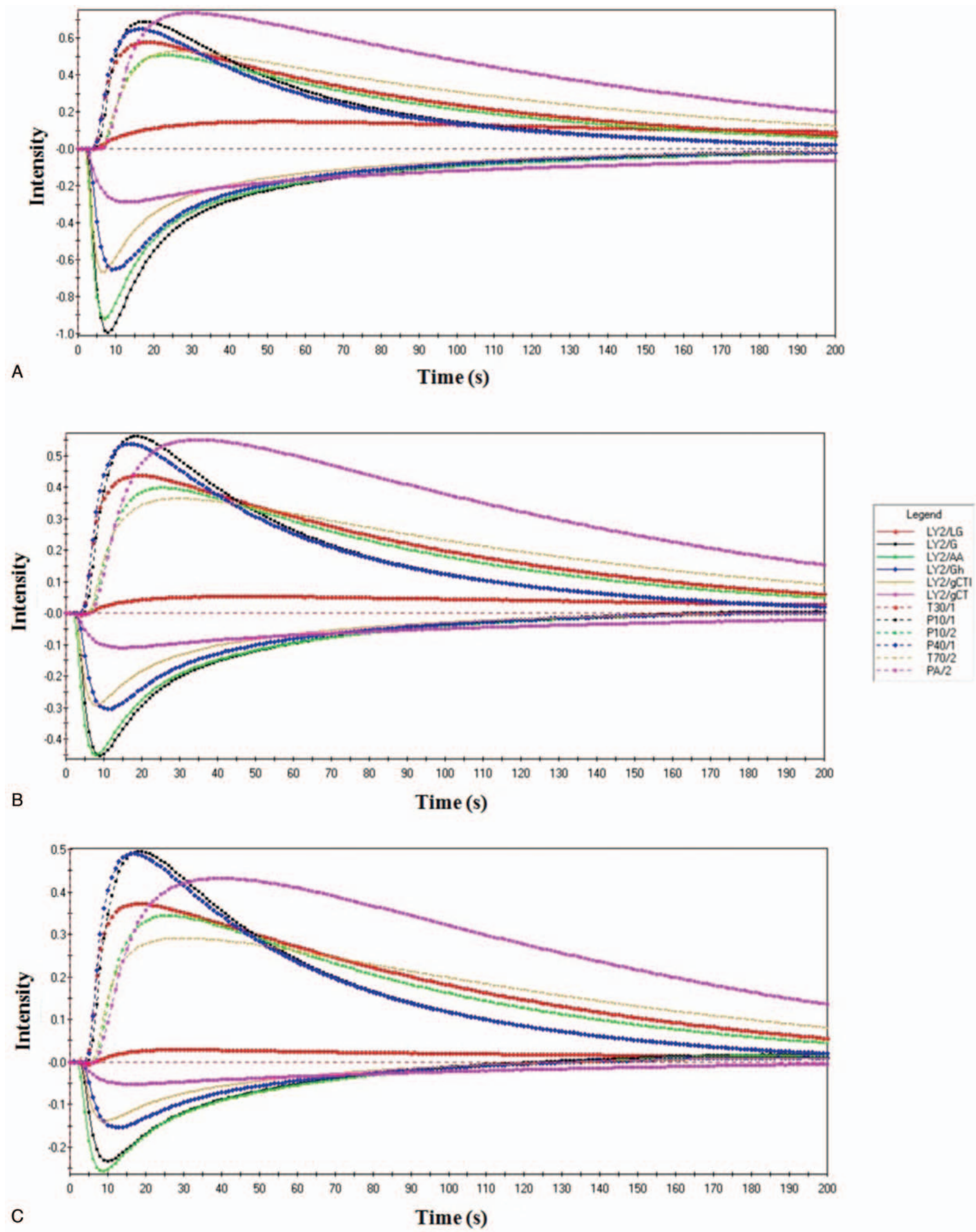

Figure 2. Typical sensor response of the moxa floss samples of (A) normal grade; (B) 3-years grade; and (C) top grade.

3. Results

3.1. E-nose response to volatile components of moxa floss samples

The response of E-nose sensors to odorants is generally regarded as a first order time response and the odor intensity is calculated using the following expression $\Delta R=(R-R_0)/R_0,$ where R is the response of the system to the sample gas, and R_0 is the baseline reading of the sensor response.[14]Figure 2 shows the typical responses of the 12 MOS sensors during the 200 seconds acquisition time for the samples. Each curve represents a different sensor transient and reflects the conductivity of each sensor

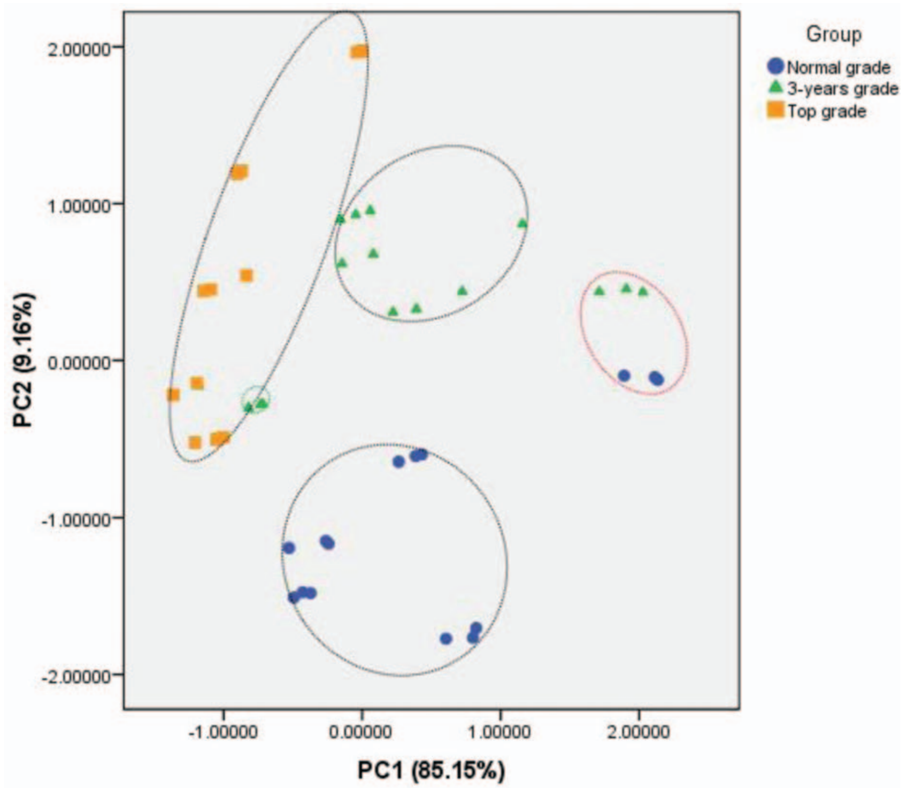

Figure 3. PCA plot of China commercial moxa floss samples. PCA=principal component analysis.

against time due to the electro-valve action when the volatiles reached the measurement chamber.[15] During the initial period, the conductivity of each sensor was low and subsequently increased to reach a steady-state after about 35 seconds. In this paper, the maximum response values of each sensor were extracted and analyzed individually. The repeatability of the established method was evaluated with 6 parallel tests of the samples. The values of the relative standard deviation (RSD, n = 6) obtained from the 12 sensors were <5%, indicating a high repeatability of the e-nose response.

3.2. PCA analysis

While feature plots (e.g., responses of single sensors over time) and descriptive statistics (e.g., calibration tables) may be sufficient for the analysis of small, low dimensional sensor data, proper pattern recognition methods are needed to evaluate the performance of e-nose sensor systems in practical tasks.[16]

To further determine the relationship between sample aroma characteristics and e-nose sensors, a biplot of PCA was conducted. As shown in Fig. 3, the first principal component (PC1) and second principal component (PC2) captured 94.31% of data variance (PC1 and PC2 were 85.15% and 9.16%, respectively), indicating that 2 principal components were sufficient to explain the total variance in the dataset (Fig. 3). It was noted that the normal grade and 3-years grade samples were better grouped and largely located in the region of positive PC1. The top grade samples were more dispersed and were distributed in the region of negative PC1. This indicated that the commercial samples marketed as "top grade" on the product labels had more dissimilar volatile profiles as compared with the other 2 groups, suggesting a certain level of disarray in the market production of moxa floss, particularly for those labeled as "top grade." The normal and 3-years grade samples from Beijing (circled in red) were also seen to be clustered together, and 1 group of 3-years grade sample from Henan (circled in green) was classified nearer to the top grade group as compared with the 3-years group. Overall, the PCA results indicated that the e-nose data were stable, repeatable, and well separated.

3.3. Neural network analysis and optimization of classifiers

Artificial neural networks (ANNs) are an extended approach to modeling complicated real-world problems by imitating the structure and function of the human brain. Radial basis function (RBF), multilayer perceptron (MLP), and random forests (RF) are some of the most prolific neural networks used for classification, consisting of 3 layers: input, hidden, and output. These depend on the number of input features, the complexity of the classification problem, and the number of output classes. The activation functions used were softmax and hyperbolic tangent at the hidden layer for the analysis. The prediction accuracies for the moxa floss classification using the 3 different pattern recognition algorithms were all above 85%, as shown in Table 3.

4. Discussion

The e-nose feature plots of the samples showed overall similar response patterns but with varying intensities for the sensors as seen from Fig. 2. The sensors gave different signals due to

Table 3

Comparison of result of the 3 different pattern recognition algorithms for moxa floss prediction.

Discriminative model	Before BC select attributes (%)	After BC select attributes (%)
Radial basis function (RBF)	91.23	87.72
Multilayer perceptron (MLP)	94.74	92.98
Random forest (RF)	92.98	89.47

BC = BestFirst.

different headspace composition of the samples, illustrating discrimination capabilities of the array. The MOS sensors work on the interactive principle that the higher the concentration, the greater the intensity of the sensor responses. It could be concluded that moxa floss samples of different quality grades had similar odor profiles but different concentrations of volatile compounds. For samples of normal grade, the sensors PA/2 and LY2/G displayed the maximum positive and negative intensities, respectively (Fig. 2A). However, for the other 2 groups of samples (3-years and top grade), the highest positive and negative readings were from the sensors P10/1 and LY2/AA (Fig. 2B and C). Combining the sensor characteristics with its main application as shown in Table 2, higher concentrations of organic amines might be found in moxa floss samples of normal grade, while 3-years and top grade samples might have higher concentrations of hydrocarbons and alcohol. This is in agreement with other studies investigating the volatile compounds of mugwort leaves using GC–MS, which detected 85 chemical components, largely comprising of aromatic hydrocarbons, terpenes, and long-chain aliphatic hydrocarbons.[17,18] The e-nose results gave an output that represented a “fingerprint” of all the components for the samples and showed good capacity in differentiating commercial moxa floss products of different quality grades. However, it was difficult to know what are specific for these samples. GC–MS analysis could have been utilized to analyze the samples to gain a better understanding of the volatile compounds contributing to the odor of the samples.

Aroma is often regarded as a valuable characteristic in assessing the quality and internal properties of agricultural and food products. The properties of moxa floss can be affected by a series of factors such as genotype, processing conditions, harvest season, and storage conditions.[19,20] Moxa floss harvested in different regions also differ in cultivation and have different growing environments due to differences in factors such as temperature, light, and rainfall. These factors control the quantitative and qualitative composition of the volatile profile, which in many cases determine the perceived quality of moxa products. As observed from the PCA results, the “top grade” products were more dispersed as compared with the normal and 3-years grade products. One possible reason could be that the top grade products are usually stored for a longer period of time and undergo more rounds of processing. Different manufactures might also have different post harvest and storage conditions, thus resulting in greater dissimilarity in the volatile profile of the top grade samples. This further justifies the need for some form of control mechanism or production guidelines to ensure a certain level of quality control of moxa products.

Based on BC feature selection method, 3 MOS sensors (S2, S6, and S11) contributing the most valuable information for identifying the moxa floss samples were screened out. Combining the sensor characteristics as listed in Table 2, sensors S2, S6, and S11 are sensitive to aromatic compounds. It could be assumed that aromatic compounds probably relate more to the moxa floss samples identification. Table 3 shows a comparison of the prediction accuracies of the 3 different classifier methods built on the e-nose data set before and after optimization by 10-fold cross validation. Using 3 MOS sensors, RBF, MLP, and RF analysis retained prediction accuracies above 85%, which indicated that the improved classification models with lower dimension data had similar performance as in the original models with higher-dimension data. This is significant as it meant that supervised learning approach helped to improve the classification model by reducing the dimension data while retaining prediction accuracy. The main objectives of this study were to explore the probative value of combining the e-nose and data mining techniques in differentiating moxa floss of different quality grades. With the help of good performance of sensor arrays along with advances in pattern recognition algorithms, e-nose devices have already been established as effective gas-sensing tools for the quality evaluation of various food and agricultural products.[21] The findings from this study indicated that on the basis of e-nose technology, data refinement for different aroma volatile compounds of moxa floss could be achieved for better discrimination performance and data clustering which is useful in reducing cost and time in practical application.

5. Conclusion

The odor characteristics of commercial moxa floss samples of different quality grades were analyzed using e-nose. The present study confirmed that data mining techniques possess satisfactory accuracy for preliminary classification of moxa floss according to quality grades. This approach of combining the e-nose with data mining techniques showed well potentiality in providing a rapid and nondestructive method for differentiating moxa floss of different quality grades. Being a fast detection technique, the e-nose could help to contribute to future studies related to moxa products, which have a direct goal towards safety and quality improvement.

Author contributions

Conceptualization: Min Yee Lim, Jian Huang, Bai-xiao Zhao.
Data curation: Min Yee Lim, Jian Huang, Fu-rong He.
Formal analysis: Min Yee Lim, Jian Huang, Hui-qin Zou.
Investigation: Min Yee Lim, Jian Huang, Fu-rong He, Dongsheng Qiu, Jun-jie Xie.
Methodology: Min Yee Lim, Jian Huang, Hui-qin Zou.
Project administration: Bai-xiao Zhao, Yong-hong Yan, Hui Hu.
Supervision: Bai-xiao Zhao, Yong-hong Yan, Hui Hu.
Writing – original draft: Min Yee Lim, Jian Huang, Fu-rong He
Writing – review & editing: Min Yee Lim, Jian Huang, Bai-xiao Zhao

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