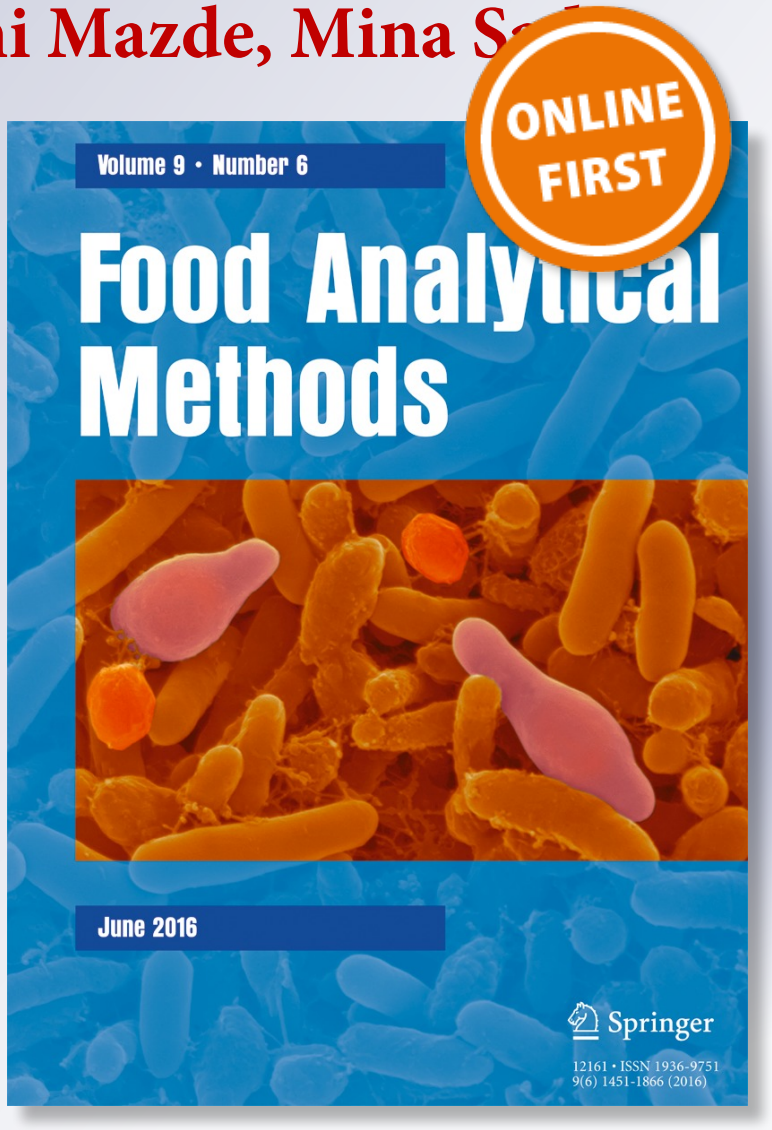
Application of Organic Acid Based Artificial Neural Network Modeling for Assessment of Commercial Vinegar Authenticity

Mannan Hajimahmoodi, Mahnaz Khanavi, Omid Sadeghpour, Mohammad Reza Shams Ardekani, Fatemeh Zamani Mazde, Mina S

Food Analytical Methods

ISSN 1936-9751

Food Anal. Methods
DOI 10.1007/s12161-016-0510-x



Springer

Food Anal. Methods
DOI 10.1007/s12161-016-0510-x

Application of Organic Acid Based Artificial Neural Network Modeling for Assessment of Commercial Vinegar Authenticity

Mannan Hajimahmoodi[1,2] · Mahnaz Khanavi[3] · Omid Sadeghpour[4] · Mohammad Reza Shams Ardekani[3] · Fatemeh Zamani Mazde[2] · Mina Sadat Khoddami[5] · Sheida Afzalifard[6] · Ali Mohammad Ranjbar[4,5]

Received: 26 November 2015 / Accepted: 11 April 2016

Abstract Vinegar as a nutraceutical substance is classified to various types related to the different substances applied in production process. Therefore, identity of the source and authenticity of the samples would be inevitable. The present study addresses determination of organic acid composition of 47 vinegar samples categorized to four types including distilled, apple, grape, and pomegranate and uncategorized vinegars (5, 12, 15, 3, and 12 samples, respectively). High-performance liquid chromatography (HPLC) method was performed according to ICH guidelines using simple sample preparation for determination of eight organic acids including oxalic, formic, ascorbic, lactic, acetic, malic, citric, and propionic acids. Findings were treated by a nonlinear computational analysis called artificial neural network (ANN) utilizing a back propagation method for training the multilayer feed-forward neural network to determine vinegar type. HPLC method resulted in suitable separation where limit of detection (LOD) and limit of quantification (LOQ) were ranged from 0.11 to 0.89 ppm and 0.34 to 2.69 in malic acid and oxalic acid, respectively. The recovery process was also ranged from 97.1 to 106.4 for oxalic acid in apple vinegar and lactic acid in grape vinegar, respectively. ANN modeling indicated a comparative model to recognize sample origin where accuracy estimation was 88.6 %. The obtained model was applied to determine the probable origin of some uncategorized commercial vinegars. It was concluded that ANN model along with analytical methods such as HPLC could be established for evaluation of commercial samples in food control laboratories.

Keywords Vinegar · HPLC · Organic acid · ANN modeling

✉ Ali Mohammad Ranjbar
amranjbar@alumnus.tums.ac.ir

[1] Department of Drug and Food Control, Faculty of Pharmacy, Tehran University of Medical Sciences, Tehran, Iran

[2] Food and Drug Administration, Tehran University of Medical Sciences, Tehran, Iran

[3] Department of Pharmacognosy, Faculty of Pharmacy, Tehran University of Medical Sciences, Tehran, Iran

[4] Herbal Medicine Department, Research Institute for Islamic & Complementary Medicine, Iran University of Medical Sciences, Tehran, Iran

[5] Department of Pharmacognosy, Faculty of Pharmacy, Shahid Sadoughi University of Medical Sciences, Yazd, Iran

[6] Department of Electrical and Computer Engineering, Yazd University, Yazd, Iran

Introduction

Vinegar is a common solution condiment used in many cuisines and drinks such as oxymel (Ranjbar et al. 2015) including distilled, wine, or brew and artificial vinegar. It has been traditionally applied as a food element and preservative (Sina 1978; Aqili Khorassani 1991) as well as control for infections, burns, oral, and dermal problems simply or in combination with herbal ingredients in traditional Iranian medicine (Sina 1978). Vinegar plays an important role in daily life due to the remarkable nutritional and therapeutic values such as antioxidant and antibacterial properties (Dávalos et al. 2005; Verzelloni et al. 2007; Sakanaka & Ishihara 2008), reducing blood pressure (Honsho et al. 2005; Tanaka et al. 2009), triglyceride, cholesterol, and glycemic index (Leeman et al. 2005; Johnston & Gaas 2006; Kondo et al. 2009), food appetizing (Darzi et al. 2010), and anti-inflammation (O'Keefe et al. 2008; Lee et al. 2011) relevant to the phytochemicals such as organic acids, amino acids, and phenolic compounds. Vinegar is produced

Published online: 19 May 2016

through a fermentation process by yeast and bacteria activities through ethanol fermentation which produces its key ingredient of organic acids (Vinegar 1997). The organoleptic and biologic properties of vinegar (Sáiz-Abajo et al. 2005; Valentão et al. 2005) such as protection against oxidative stress-related diseases (Verzelloni et al. 2010) and chelating metals and multivalent cations can be affected by organic acids (Oliveira et al. 2008). Some physicochemical methods have been introduced for determination of vinegar quality and authenticity such as total acid content, total solid content, caramel test, and the content of sugar and alcohol (Latimer 2012). These tests are not able to determine the source of vinegar and can be faked simply. Some equipment such as electronic nose has been also applied to detect vinegar characteristics according to the volatile component combined by pattern recognition techniques, analogous to human smelling power (Wilson & Baietto 2009). The composition of organic acids which will be produced during vinegar fermentation could be a valid consequence to detect vinegar authenticity, and although high-performance liquid chromatography (HPLC) method is not as easy to access as physicochemical methods, it could be applied for determination of the mentioned composition due to the good resolution, sensitivity, and selectivity (Zong et al. 2015). Then, the results can be treated by chemometric techniques in order to attain the final objective (Cordella et al. 2002). Different multivariate methods such as multiple regression analysis, principle component analysis, and partial least square analysis have been used to classify wine, juice, and some other foods (Arvanitoyannis et al. 1999; Movagharnejad & Nikzad 2007). Artificial neural network (ANN) which is applied in classification and pattern identification is a technique that processes the parallel-distributed information inspired by biological nervous system (Satish & Setty 2005). It fundamentally consists of simple processing nodes or units where their function is based on human neurons. Briefly, its ability is reserved in inter-node connection weights which would be adopted by a process of learning relevant to input-output pattern and the training rules refine the memory of the neural network (Hussain et al. 2002). It has been applied in classification of some foods such as Idaho-labeled potato based on elemental analysis combined with neural network techniques (Anderson et al. 1999), European Emmental cheeses (Pillonel et al. 2005), and yogurt according to the pH, color, and hardness of commercial yogurt to low and reduced fat categories (Da Cruz et al. 2009). It also has been applied to differentiate wine production location according to the metal composition (Díaz et al. 2003).

The present study focused on the development of an ANN model to distinguish the origin of vinegar based on its organic acid contents obtained by HPLC as a way of evaluating vinegar authenticity.

Material and Methods

Reagents

All reagents and standards were analytical grade that were purchased from Merck (Darmstadt, Germany). Organic acids including ascorbic, acetic, formic, lactic, citric, malic, oxalic, and propionic acid were stored at 4 °C in a dark place.

Samples

Out of 47 vinegar samples purchased during 2013 to 2014, 15 samples were factory made and purchased from Iran market. The remains were traditionally prepared through alcoholic fermentation by healers from Tehran, Yazd, Qom, and Isfahan provinces, Iran. The mentioned 47 samples were distilled, apple, pomegranate, grape, and uncategorized types which were categorized according to the primary materials applied in vinegar preparation (Table 1). It is common to fulfill the earthen pot with the seed of pomegranate, pieces of apple, and crashed grape, daub outside of the pot with the oil, and puddle its valve during the traditional preparation of relevant vinegar types. The samples were checked for residue of ethanol according to the Richard method (Duggins 1979) and stored in airtight glass container at room temperature and dark place.

High-Performance Liquid Chromatography

Chromatographic procedure was carried out using Agilent 1200 series liquid chromatography which was equipped with a pump, 20-μL loop injector, vacuum membrane degasser, and UV visible detector along with 150 mm × 4.6 mm, 5-μm Eclipse-XDB C18 column (Agilent, CA, USA). The mobile phase was H_2SO_4 (0.01 M) in double-distilled water with 0.65 mL/min flow rate. The detection was conducted by a UV detector in which the wavelength was set at 210 nm.

Table 1 Number of sample types according to their healer's region

City	Distilled	Grape	Apple	Pomegranate	Uncategorized
Tehran	3	5	4	0	4
Yazd	2	5	3	3	4
Qom	0	3	3	0	2
Isfahan	0	2	2	0	2

Sample Preparation

One milliliter of each sample was diluted to 250 mL by double-distilled water in a volumetric flask. Prior to injection, they were filtered through a 0.45-μm PVDF filter (Millipore, Ireland).

Method Validation

The method was validated according to the ICH guidelines (Walfish 2006) where selectivity, linearity, precision, accuracy, limit of detection (LOD), and limit of quantification (LOQ) were calculated. The selectivity was determined by spiking structurally related compounds such as alcohols, esters, and glycerols in the samples inspecting any interference in the related chromatograms. Calibration curves were constructed for the abovementioned organic acids over the range of 6.25 to 100 mg/kg, and the linearity as well as correlation coefficients was assessed. The accuracy of the method was evaluated by sample recovery performed by spiking known amounts of the studied organic acids. To evaluate the intraday (RSD_r) and inter-day (RSD_R) precision, each sample was analyzed three times on the same day and in three different days. The following equations were used to calculate LODs and LOQs of each organic acid.

$$LOD = 3.3\ \sigma/s$$

$$LOQ = 10\ \sigma/s$$

where σ is the standard deviation of *y*-intercept and *S* is the slope of the calibration curve.

Data Analyzing

The obtained data from HPLC was applied to a multilayer feed-forward neural network model that performed with back propagation training algorithm which has one hidden layer with four neurons. A total of eight input neurons were existed as feature in the network that represented organic acid constitution of vinegar, and four output classes were set corresponding to the group of vinegar. In the mentioned process, 35 samples were divided to the test data (15 %), validation data (15 %), and train data (70 %). Then 12 uncategorized samples were checked with obtained model to identify sample type. The outcomes originally were calculated from the following equation which operates as a sigmoidal function.

$$y_j = f\left(\sum_{i=1}^{n} w_{ji} x_i\right)$$

where y_j is the activation of the prepared node, and *f* is the activation function; *i* is weights and outputs from the previous layer; w_{ij} indicates the relevant weights connecting layer *i* with

Table 2 Method validation parameters of eight organic acids

Organic acid	Calibration equation	R^2	LOD (mg kg^{-1})	LOQ (mg kg^{-1})	Recovery (%)				Precision (%)	
					Grape vinegar	Apple vinegar	Pomegranate vinegar	Distilled vinegar	RSD_r	RSD_R
Oxalic acid	$y = 20/894x + 10/253$	0.9997	0.89	2.69	103.6	97.1	99.8	99.9	0.78	2.28
Formic acid	$y = 2/5494x + 0/8346$	0.9997	0.17	0.51	104.5	98.6	101.3	100.4	1.13	3.56
Lactic acid	$y = 1/7246x - 0/592$	0.9998	0.40	1.24	106.4	101.2	103.0	100.2	0.41	1.24
Acetic acid	$y = 1/3088x - 1/8638$	0.9987	0.21	0.61	101.3	97.8	100.7	100.5	1.09	3.93
Malic acid	$y = 205/21x + 1/5319$	0.9999	0.11	0.34	102.2	99.1	101.4	100.6	0.96	1.45
Citric acid	$y = 2/5095x - 0/2124$	0.9999	0.19	0.58	102.3	98.5	99.4	100.1	0.25	0.64
Propionic acid	$y = 1/2026x - 1/315$	0.9983	0.12	0.38	98.8	97.3	98.9	99.7	0.81	1.16
Ascorbic acid	$y = 30/826x - 0/5355$	0.9980	0.67	2.0	99.7	98.0	99.1	99.8	1.07	2.23

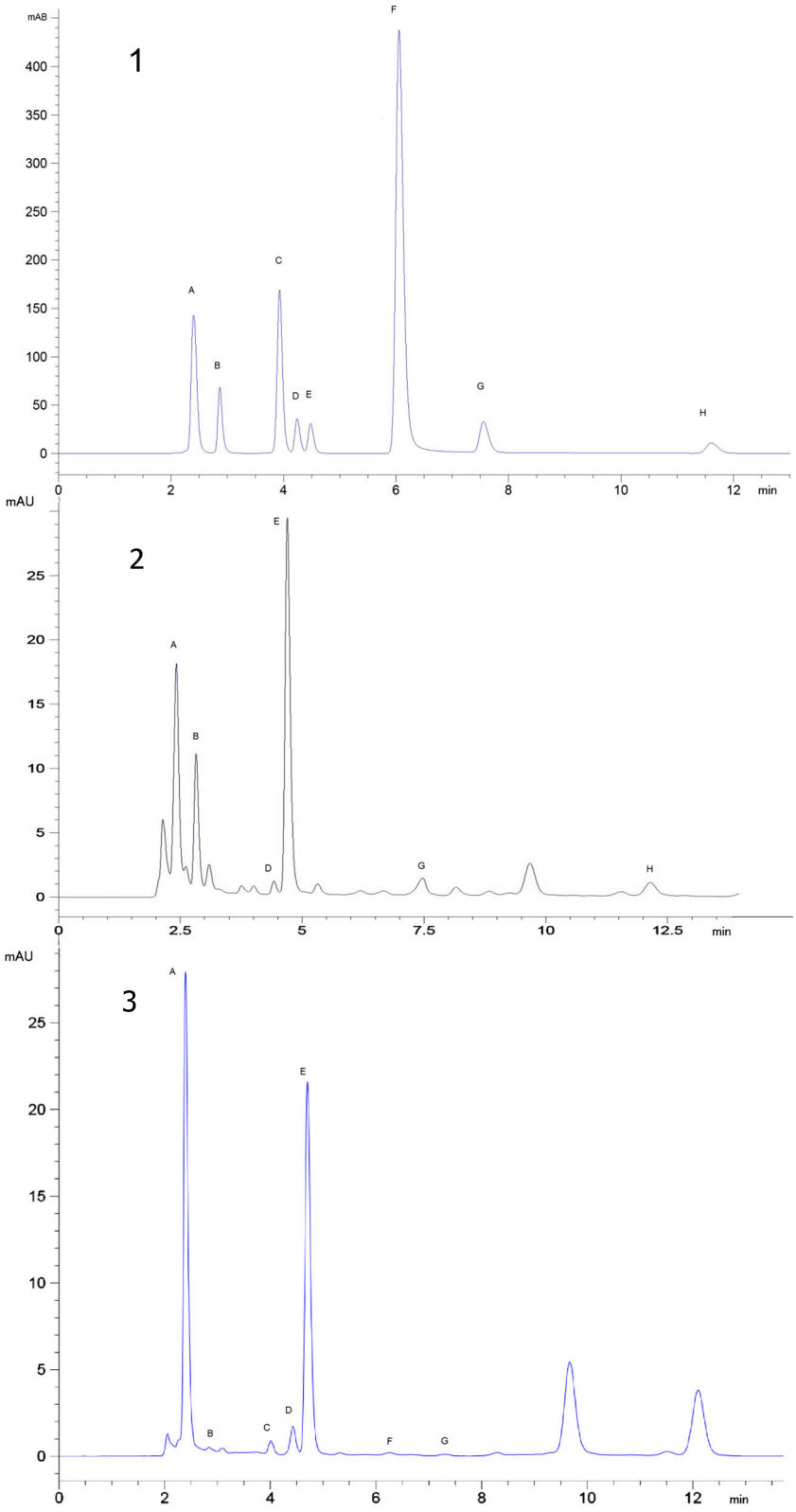


Fig. 1 Chromatogram of organic acid standard mixture (*1*) and two real vinegar sample chromatograms (*2* & *3*). *A* oxalic acid, *B* formic acid, *C* ascorbic acid, *D* lactic acid, *E* acetic acid, *F* malic acid, *G* citric acid, *H* propionic acid

layer j, and x_i indicates the activations of the nodes in the previous layer i. Back propagation is commonly the first-order gradient method utilized for training neural network to correlate between variables. It includes two phases of forward and backward pass including the information process from input to output, and then the output layer conflicts return to

Table 3 Organic acid concentration of different types of real vinegar samples including distilled (D), grape (G), apple (A), pomegranate (P), and uncategorized (U)

Vinegar type	Organic acid concentration mg/kg							
	Oxalic acid	Formic acid	Ascorbic acid	Lactic acid	Acetic acid	Malic acid	Citric acid	Propionic acid
D1	0.00	0.31	0.00	0.00	75.11	0.00	0.00	0.00
D2	0.00	0.00	0.00	0.00	87.86	0.00	0.00	0.00
D3	1.66	0.00	0.00	0.00	73.71	0.00	0.00	0.00
D4	1.48	0.44	0.00	0.27	67.55	0.00	0.00	0.00
D5	1.02	0.30	0.00	0.00	71.07	0.00	0.00	0.00
G1	2.79	0.84	0.00	2.15	49.11	0.00	0.00	0.00
G2	0.00	0.91	0.00	2.15	47.34	0.00	1.95	0.00
G3	0.00	0.44	0.00	0.15	72.93	0.14	0.29	0.00
G4	3.20	2.49	0.01	0.97	47.25	0.00	0.00	0.00
G5	2.68	1.90	0.00	0.52	55.76	0.00	0.00	0.00
G6	0.00	2.41	0.10	1.11	47.79	0.20	0.95	0.00
G7	0.00	0.00	0.00	0.00	44.32	0.28	1.32	0.00
G8	2.23	2.38	0.46	3.10	21.68	0.00	0.00	0.00
G9	1.13	0.10	0.68	0.50	38.03	0.00	5.31	0.00
G10	0.00	0.12	0.00	0.44	37.05	0.17	1.53	0.00
G11	0.00	6.07	0.00	2.10	36.82	0.00	0.00	0.00
G12	2.14	0.00	0.64	1.59	15.24	0.00	0.00	3.75
G13	2.79	0.00	0.00	0.85	25.88	0.41	0.00	0.00
G14	2.43	1.71	0.00	4.05	31.79	0.03	0.64	0.00
G15	0.00	0.00	0.00	0.30	74.92	0.00	0.52	0.00
A1	2.58	0.61	0.22	0.00	52.63	0.00	0.00	0.00
A2	0.00	0.00	0.71	4.30	74.54	0.00	0.82	0.00
A3	0.00	0.15	0.37	4.46	57.63	0.38	0.00	0.00
A4	0.00	0.35	0.00	4.65	68.93	0.00	0.00	0.00
A5	2.83	2.13	0.00	16.75	24.62	0.00	0.00	0.00
A6	0.00	0.00	0.00	16.75	27.41	0.11	20.68	0.00
A7	0.00	0.00	0.81	10.23	52.05	0.18	2.70	0.76
A8	0.00	1.76	0.40	6.32	56.37	0.30	1.34	0.76
A9	4.19	0.32	1.01	2.43	31.00	0.15	0.33	0.00
A10	0.00	0.00	0.00	2.79	46.49	0.00	0.00	0.00
A11	0.00	0.35	0.00	1.70	68.89	0.00	0.42	0.00
A12	0.00	0.00	0.00	4.54	60.77	0.00	2.28	0.00
P1	2.49	3.58	0.00	2.39	44.44	0.00	1.93	1.65
P2	0.00	0.00	0.13	0.23	0.99	0.00	0.08	0.63
P3	0.00	4.09	0.13	7.10	23.47	2.96	1.29	0.00
U1	0.00	0.39	0.00	0.36	73.50	0.00	0.00	0.00
U2	0.00	3.03	0.00	0.18	72.52	0.00	0.00	0.00
U3	5.26	1.31	0.00	11.16	10.81	0.00	0.00	0.00
U4	0.00	0.53	0.00	22.72	31.39	0.00	0.00	0.00
U5	8.62	0.00	0.00	0.18	24.56	0.00	0.16	0.00
U6	0.00	0.00	0.00	21.44	6.13	0.00	0.00	0.00
U7	0.00	0.00	15.06	0.00	16.69	0.30	0.00	0.00
U8	0.00	0.87	0.13	0.14	1.27	2.75	0.99	0.00
U9	0.00	2.55	1.01	10.30	7.96	0.79	0.34	0.00
U10	2.51	0.28	0.00	0.94	61.56	0.00	0.00	0.55
U11	0.00	0.00	0.00	8.04	28.73	0.00	0.49	0.00
U12	0.00	0.00	0.00	0.68	66.34	0.00	4.24	1.72

the input layer to modify the network weights. Receiver operating characteristic (ROC) and confusion matrix were revealed to evaluate method. Vertical columns of the matrix show producer's accuracy since it is a measure of the omission error, and horizontal columns represent user's accuracy as there is a measure of the commission error. The mentioned parameters are calculated as described below.

$$\text{producer}'\text{s accuracy}(\text{for class } k) = \frac{N_{jj}}{\sum N_{ij}}$$

$$\text{user}'\text{saccuracy}(\text{for class } k) = \frac{N_{ii}}{\sum_i N_{ji}}$$

where N is the total number of nodes, and N_{ij} is the number of nodes in cell state i ($i = 1, 2,\ldots,k$) in simulation and cell state j ($j = 1, 2,\ldots, k$) within the set of data.

Results and Discussion

System Optimization

The method was designed for the determination of acetic, citric, formic, lactic, ascorbic, malic, propionic, and oxalic acid in vinegar (Latimer 2012). Ionized form of the mentioned organic acids in water leads to short retention time. Therefore, acidic mobile phase, H_2SO_4 (0.01 M), was applied to change ionized organic acids to their non-ionized forms that resulted in increase in retention time. Acceptable separation and retention times were achieved using 0.65 mL/min flow rate, and chromatograms were detected by UV detector at 210 nm wavelength.

HPLC Method Validation

No interference was observed in the corresponding organic acid retention times following the injection of samples spiked by structurally related substances such as alcohol, ester, and glycerol which can be concluded as method selectivity. The results of method validation are presented in Table 2 where RSD_r and RSD_R precisions were ranged from 0.25 % in citric acid to 1.13 % in formic acid and 0.64 % in citric acid to 3.93 % in acetic acid, respectively, which are in accepted range as mentioned by AOAC (8 and 16 %, respectively) (Latimer 2012). Accuracy of the method as recovery percentage was determined according to the standard addition method where it was ranged from 98.8 to 106.4 % for propionic acid and lactic acid in grape vinegar and 97.1 to 101.2 % for oxalic acid and lactic acid in apple vinegar, respectively. It was ranged from 98.9 to 103.0 % for propionic acid and lactic acid and 99.7 to 100.6 % for propionic acid and malic acid in the case of pomegranate vinegar and distilled vinegar, respectively. The calculated LOD and LOQ were ranged from 0.89 to 2.69 mg kg^{-1} in oxalic acid to 0.11 and 0.34 mg kg^{-1} in malic acid, respectively, which follow the acceptable range of the LOD (0.10 to 10.0 μg/mL) and LOQ (0.30 to 30.0 μg/mL) (Zong et al. 2015).

Real Sample Detection

The chromatographic method was applied in 47 vinegar samples in which the residue of ethanol was not exceeded form 0.5 % as set by Iranian National Standard Organization (Institute of Standards and Industrial Research of Iran 2016) showing end point of vinegar fermentation. Figure 1 shows the organic acids actually were found in two randomly selected vinegar samples and in the mixture of organic acid standards. As it can be seen in Table 3, acetic acid was the most prevalent organic acid presented in studied vinegar samples while propionic and malic acid were infrequent in the studied samples. The results obtained by Castro et al. (Castro et al. 2002) are in agreement with the current research since the propionic and malic acid were not detected in the most of Brazilian vinegars. Yang and Choong (Yang & Choong 2001) reported acetic, isovaleric, lauric, capric, lactic, and levulinic acids in the vinegar samples where acetic and lactic acid were the most predominant organic acids as well as in presented study. Acetic acid was the most prevalent acid (40–56 g L^{-1}) followed by lactic acid (0.2–2.2 g L^{-1}) and was reported by Castro et al. on the commercially available vinegar samples from red wine, white wine, apple, and rice (Castro et al. 2002). Out of ten apple vinegar samples from 12 analyzed ones, lactic acid was the second acid in quantity that was detected at present study. The amount of organic acids in pomegranate, blackberry, mulberry, cherry, blueberry, red ginseng, and cactus vinegars which are reported by Kim et al.'s

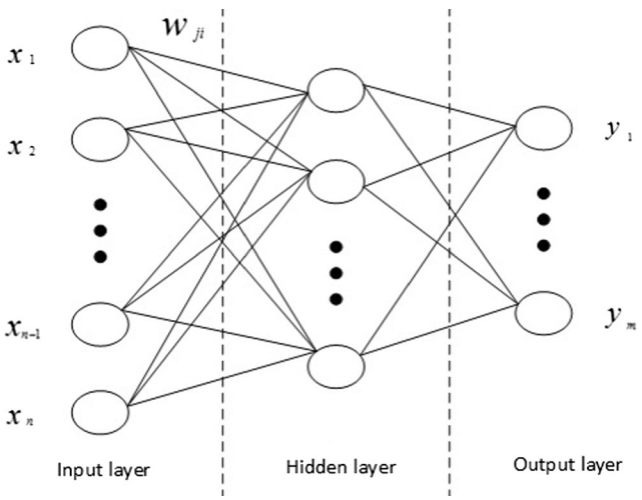


Fig. 2 A simple model of neural network layers. Inputs are indicated by x, weights are identified by w, and y presents outputs

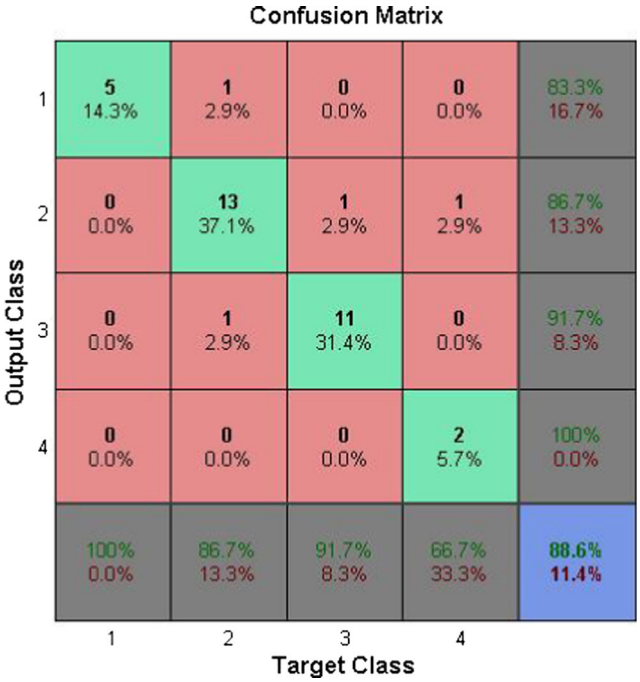


Fig. 3 Confusion matrix of the neural network model

(Kim et al. 2012) research is significantly lower than the presented work excluding malic acid content. Therefore, it could be claimed that distilled, apple, and grape vinegars are well qualified due to the organic acid influence on sourness and thereby had a major impact on overall quality (Ha & Kim 2000). According to the Soyer et al. study, the organic acid concentration of 11 different white grape cultivars and grape juices was determined where tartaric acid was the major acid in all analyzed grape varieties but due to detartration process, grape juices had lower tartaric acid content than grapes (Soyer et al. 2003). Additionally, oxalic, malic, and citric acid in Moroccan apricot were 28.15, 6.44, and 16.14 g kg^{-1} in Hasib et al.'s study (Hasib et al. 2002).

Time of fermentation is one of the most significant factors in the physicochemical properties of vinegar. In Oguntoyinbo et al.'s study which revealed that vinegar could be produced from sweet orange peels, fermentation gave the highest yield (75 % *v/v*) after 14 days (Oguntoyinbo et al. 2011). This achievement may be able to describe differences between the physicochemical properties of various studies and also between the different samples of the same group in this study.

ANN Analysis

ANN shows better results in comparison with other models such as partial least square (PLS) and multilinear regression (MLR) as it does not follow any previous conception or mathematical model and usually apply for data sets that indicate nonlinear conjunction. Indeed, it recognizes patterns of input and output data of train set (Zhang 2000). According to the electronic nose equipment, only the volatile components have

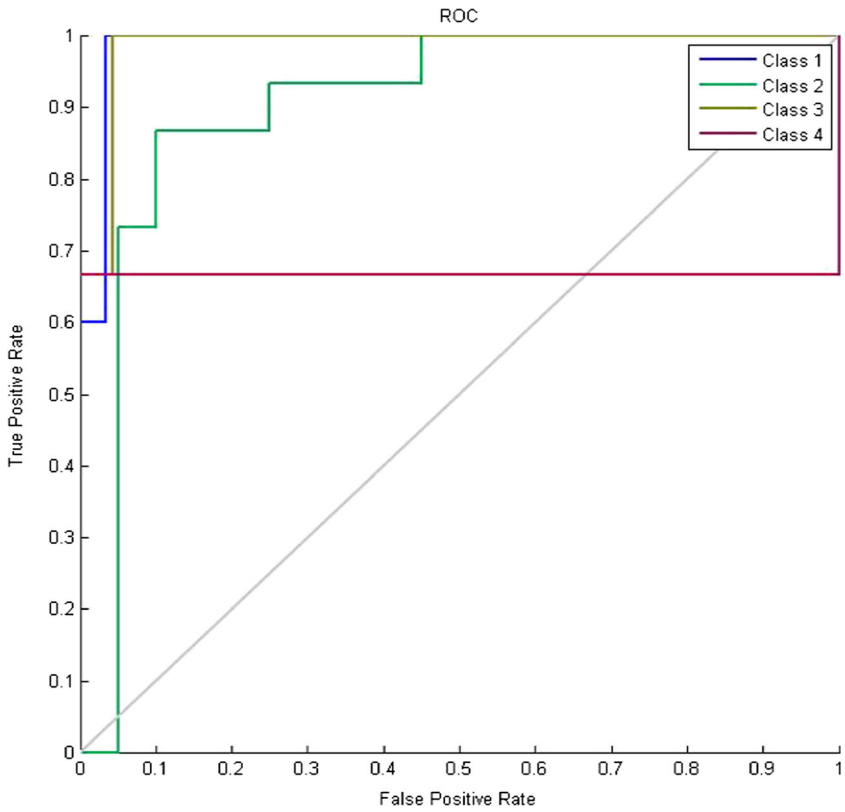


Fig. 4 ROC curve of 35 samples modeled by neural network

been applied in vinegar quality control where nonvolatile components are mentioned to have an important role in the mentioned quality. Therefore, the efficiency of this method depends on the correlation of the pattern of volatile components with nonvolatile components which participate in quality of vinegar (Röck et al. 2008). In addition, humidity and temperature would influence on electronic nose result which claimed to be decreased by virtue of new sensors and combination of it with gas chromatography or spectroscopy methods (Ampuero & Bosset 2003; Prieto et al. 2012; Cheng et al. 2013). Although the components analyzed at present study are important in the quality of vinegar, classification of vinegar according to the source of primary materials was noticed here and it is suggested to consider other components that are related to quality control of vinegar such as flavonoids.

Multilayer feed-forward neural network which was used at present study is depicted as a simple model in Fig. 2. Overall accuracy that acquired due to the classification was 88.6 % as depicted in the last cell of confusion matrix presented in Fig. 3. Each row of the matrix describes the individual truth groups, and each column shows a network selection. Correct responses represent the diagonal of the matrix and off diagonals indicate errors. Based on the confusion matrix, identification accuracies were 83.3, 86.7, 91.7, and 100 % for classes 1 to 4, respectively, which were distilled, grape, apple, and pomegranate in turn. There were not adequate samples in the case of class 4 (pomegranate), and as a result, the obtained model would rarely be able to predict that a sample belongs to this group. In comparison with other studies, the authenticity of Idaho-labeled potato and different location sources of wine were determined by approximately 100 % of accuracy (Anderson et al. 1999). Classification of European Emmental cheeses in regard to their origin also resulted in 91 % of accuracy (Pillonel et al. 2005). The performance of the neural network has been detected by ROC curve where output groups were presented with colored line and the best specificity and sensitivity were noted on the upper left regions which is showed in Fig. 4 (Kalyan et al. 2014). The true positive rate against the false positive rate is shown in the threshold for whole classes (Meistrell 1990). Obviously, this ratio can vary between 0 and 1. Thus, the higher is the accuracy, the nearer to 1 is the ROC. The obtained model was applied for 12 uncategorized samples where the result indicated U1, U5, and U10 as distilled vinegar by 83.3 % accuracy, U2, U3, U8, U11, and U12 as grape vinegar by 86.7 % accuracy, and U4, U6, U7, and U9 as apple vinegar by 97.1 % accuracy. It should be noted that the presented model can predict the origin of unknown samples based on the similarity to the known groups that have been trained, and as a result, it could not be able to recognize a sample out of the train group set. On the other hand, it would trap the unknown sample in the most similar group based on its composition by less accuracy.

Conclusion

The vinegar types applied at present study are the most commercially important kind of vinegar in Iran's market. The employed HPLC method was conducted without complicated sample preparations, serving the time to analyze a variety of samples. Although ANN represents a suitable accuracy, more sample analysis produced in different regions can lead to a more applicable model in a variety of control laboratory.

Acknowledgments This study is a part of a Ph.D. thesis, and a project has been funded and supported by Tehran University of Medical Sciences (TUMS), Grant No: 91-01-86-13394.

Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

Ethical Approval This article does not contain any studies with human participants or animal performed by any of the authors.

Informed Consent This article does not contain any human participants.

References

Ranjbar AM, Sadeghpour O, Khanavi M, Ardekani MRS, Moloudian H, Hajimahmoodi M (2015) Effects of the deslagging process on some physicochemical parameters of honey. Iran J Pharm Res 14(2):657

Sina I (1978) Ghanoon Dar Teb [The canon of medicine], Bulaq Edition. Sharafkandi A, trans Tehran: Univ of Tehran Pr

Aqili Khorassani M (1991) Makhzan al adviah. Tehran: Safa publication, 361

Dávalos A, Bartolomé B, Gómez-Cordovés C (2005) Antioxidant properties of commercial grape juices and vinegars. Food Chem 93(2):325–30

Verzelloni E, Tagliazucchi D, Conte A (2007) Relationship between the antioxidant properties and the phenolic and flavonoid content in traditional balsamic vinegar. Food Chem 105(2):564–71

Sakanaka S, Ishihara Y (2008) Comparison of antioxidant properties of persimmon vinegar and some other commercial vinegars in radical-scavenging assays and on lipid oxidation in tuna homogenates. Food Chem 107(2):739–44

Honsho S, Sugiyama A, Takahara A, Satoh Y, Nakamura Y, Hashimoto K (2005) A red wine vinegar beverage can inhibit the renin-angiotensin system: experimental evidence in vivo. Biol Pharm Bull 28(7):1208–10

Tanaka H, Watanabe K, Ma M, Hirayama M, Kobayashi T, Oyama H et al (2009) The effects of γ-aminobutyric acid, vinegar, and dried bonito on blood pressure in normotensive and mildly or moderately hypertensive volunteers. J Clin Biochem Nutr 45(1):93

Leeman M, Östman E, Björck I (2005) Vinegar dressing and cold storage of potatoes lowers postprandial glycaemic and insulinaemic responses in healthy subjects. Eur J Clin Nutr 59(11):1266–71

Johnston CS, Gaas CA (2006) Vinegar: medicinal uses and antiglycemic effect. Medscape Gen Med 8(2):61

Kondo T, Kishi M, Fushimi T, Ugajin S, Kaga T (2009) Vinegar intake reduces body weight, body fat mass, and serum triglyceride

levels in obese Japanese subjects. Biosci Biotechnol Biochem 73(8):1837–43

Darzi J, Frost G, Cooke C, Johnson L, Martin V, Pickard J et al (2010) Can vinegar supplementation acutely influence appetite and glycaemic response? Proc Nutr Soc 69(OCE1):E92

O'Keefe JH, Gheewala NM, O'Keefe JO (2008) Dietary strategies for improving post-prandial glucose, lipids, inflammation, and cardiovascular health. J Am Coll Cardiol 51(3):249–55

Lee CS, Yi EH, Kim H-R, Huh S-R, Sung S-H, Chung M-H et al (2011) Anti-dermatitis effects of oak wood vinegar on the DNCB-induced contact hypersensitivity via STAT3 suppression. J Ethnopharmacol 135(3):747–53

Vinegar AM (1997) Microbiology of fermented foods: Springer, p. 1–44

Sáiz-Abajo M, González-Sáiz J, Pizarro C (2005) Multi-objective optimisation strategy based on desirability functions used for chromatographic separation and quantification of l-proline and organic acids in vinegar. Anal Chim Acta 528(1):63–76

Valentão P, Andrade PB, Rangel J, Ribeiro B, Silva BM, Baptista P et al (2005) Effect of the conservation procedure on the contents of phenolic compounds and organic acids in chanterelle (Cantharellus cibarius) mushroom. J Agric Food Chem 53(12):4925–31

Verzelloni E, Tagliazucchi D, Conte A (2010) From balsamic to healthy: traditional balsamic vinegar melanoidins inhibit lipid peroxidation during simulated gastric digestion of meat. Food Chem Toxicol 48(8):2097–102

Oliveira AP, Pereira JA, Andrade PB, Valentão P, Seabra RM, Silva BM (2008) Organic acids composition of Cydonia oblonga Miller leaf. Food Chem 111(2):393–9

Latimer GW (2012) Official methods of analysis of AOAC International: AOAC international

Wilson AD, Baietto M (2009) Applications and advances in electronic-nose technologies. Sensors 9(7):5099–148

Zong Y, Lin J, Xu H, Jia Z, Yang X, Choi MM (2015) Optimization and validation of an HPLC-photodiode array detector method for determination of organic acids in vinegar. J AOAC Int 98(2):422–30

Cordella C, Moussa I, Martel A-C, Sbirrazzuoli N, Lizzani-Cuvelier L (2002) Recent developments in food characterization and adulteration detection: technique-oriented perspectives. J Agric Food Chem 50(7):1751–64

Arvanitoyannis I, Katsota M, Psarra E, Soufleros E, Kallithraka S (1999) Application of quality control methods for assessing wine authenticity: use of multivariate analysis (chemometrics). Trends Food Sci Technol 10(10):321–36

Movagharnejad K, Nikzad M (2007) Modeling of tomato drying using artificial neural network. Comput Electron Agric 59(1):78–85

Satish S, Setty YP (2005) Modeling of a continuous fluidized bed dryer using artificial neural networks. Int Commun Heat Mass Tran 32(3):539–47

Hussain M, Rahman MS, Ng C (2002) Prediction of pores formation (porosity) in foods during drying: generic models by the use of hybrid neural network. J Food Eng 51(3):239–48

Anderson KA, Magnuson BA, Tschirgi ML, Smith B (1999) Determining the geographic origin of potatoes with trace metal analysis using statistical and neural network classifiers. J Agric Food Chem 47(4):1568–75

Pillonel L, Bütikofer U, Schlichtherle-Cerny H, Tabacchi R, Bosset J-O (2005) Geographic origin of European Emmental. Use of discriminant analysis and artificial neural network for classification purposes. Int Dairy J 15(6):557–62

Da Cruz A, Walter E, Cadena R, Faria J, Bolini H, Fileti AF (2009) Monitoring the authenticity of low-fat yogurts by an artificial neural network. J Dairy Sci 92(10):4797–804

Díaz C, Conde JE, Estévez D, Pérez Olivero SJ, Pérez Trujillo JP (2003) Application of multivariate analysis and artificial neural networks for the differentiation of red wines from the Canary Islands according to the island of origin. J Agric Food Chem 51(15):4303–7

Duggins RS (1979) Means and method for aging liquor. Google Patents

Walfish S (2006) Analytical methods: a statistical perspective on the ICH Q2A and Q2B guidelines for validation of analytical methods. Biopharm Int 19(12)

Vinegar - Specifications [Internet]. Institute of standards and industrial research of Iran. Available from: http://www.isiri.org/portal/file/?15095/ICS:67.220.10

Castro R, Moreno M, Natera R, García-Rowe F, Hernández M, Barroso C (2002) Comparative analysis of the organic acid content of vinegar by capillary electrophoresis and ion-exclusion chromatography with conductimetric detection. Chromatographia 56(1–2):57–61

Yang M-H, Choong Y-M (2001) A rapid gas chromatographic method for direct determination of short-chain (C < sub > 2</ sub > −C < sub > 12</sub>) volatile organic acids in foods. Food Chem 75(1):101–8

Kim S-H, Cho H-K, Shin H-S (2012) Physicochemical properties and antioxidant activities of commercial vinegar drinks in Korea. Food Sci Biotechnol 21(6):1729–34

Ha Y, Kim K (2000) Civilization history of vinegar. Food Ind Nutr 5:1–6

Soyer Y, Koca N, Karadeniz F (2003) Organic acid profile of Turkish white grapes and grape juices. J Food Compos Anal 16(5):629–36

Hasib A, Jaouad A, Mahrouz M, Khouili M (2002) HPLC determination of organic acids in Moroccan apricot determinación por HPLC de Ácidos Orgánicos en Albaricoque Marroquí determinación por HPLC de ácidos orgánicos en Albaricoque Marroquí. CYTA-J Food 3(4):207–11

Oguntoyinbo SI, Babajide JM, Adenekan MK, Ajayi JO, Kareem SO, Ayelaagbe IOO, Atanda OO, Bodunde G (2011) Chemical properties of vinegar produced from sweet orange peels (Citrus sinensis). J Agric Vet Sci 3:51–61

Zhang GP (2000) Neural networks for classification: a survey. IEEE Trans Syst Man Cybern Part C Appl Rev 30(4):451–62

Röck F, Barsan N, Weimar U (2008) Electronic nose: current status and future trends. Chem Rev 108(2):705–25

Ampuero S, Bosset J (2003) The electronic nose applied to dairy products: a review. Sensors Actuators B Chem 94(1):1–12

Prieto N, Rodriguez-Méndez M, Leardi R, Oliveri P, Hernando-Esquisabel D, Iñiguez-Crespo M et al (2012) Application of multi-way analysis to UV–visible spectroscopy, gas chromatography and electronic nose data for wine ageing evaluation. Anal Chim Acta 719:43–51

Cheng H, Qin Z, Guo X, Hu X, Wu J (2013) Geographical origin identification of propolis using GC–MS and electronic nose combined with principal component analysis. Food Res Int 51(2):813–22

Kalyan K, Jakhia B, Lele RD, Joshi M, Chowdhary A (2014) Artificial neural network application in the diagnosis of disease conditions with liver ultrasound images. Adv Bioinforma

Meistrell ML (1990) Evaluation of neural network performance by receiver operating characteristic (ROC) analysis: examples from the biotechnology domain. Comput Methods Prog Biomed 32(1):73–80