Multivariate analysis to discriminate the origin of sesame seeds by multi-element analysis inductively coupled plasma-mass spectrometry

Young Hee Choi^{1,2}, Chae Kyu Hong¹, Misun Kim¹, Sun Oak Jung¹, Juseong Park¹, Young Hee Oh¹, and Joong-Ho Kwon^{2,*}

¹Special Inspection Team, Seoul Metropolitan Government Research Institute of Public Health and Environment, Seoul 13818, Korea

²School of Food Science & Biotechnology, Kyungpook National University, Daegu 41566, Korea

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*Corresponding Author
Tel: +82-53-950-5775
Fax: +82-53-950-6772
E-mail: jhkwn@knu.ac.kr

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Abstract In this study, inductively coupled plasma-mass spectrometry (ICP-MS) was used to determine the concentration of 15 elements (Mg, Al, K, Ca, Cr, Mn, Co, Ni, Cu, Zn, Rb, Sr, Cd, Ba, and Pb) of sesame seeds. Multivariate analysis was then performed to discriminate the origin of sesame seeds. Korean (48), Chinese (44), and Indian (21) samples were used to develop the calibration model. Another 10 samples were used to validate this model. All elements were significantly different ($p < 0.05$) among the samples from three countries, and all elements were subjected to both principal component analysis (PCA) and discriminant analysis. The concentrations of multi-element showed a trend of clustering according to the origin of samples based on PCA. They showed a discrimination rate of 92.0% in the discriminant analysis. The results demonstrated that a combination of ICP-MS multi-element determination and multivariate analysis could be used to discriminate the sesame seed origin.

Keywords: discriminant analysis, geographical origin, inductively coupled plasma-mass spectrometry, principal component analysis, sesame seed

Introduction

Sesame seed is mainly produced in India, China, Sudan, and Myanmar. Approximately 70% of the sesame seeds in the world are cultivated in these countries, accounting for 60% of the total production of sesame seeds (International Union of Food and Agriculture Organization, statistic database, FAOSTAT, 2012). Each year, declining trends are observed in both cultivation area and annual production of sesame seeds. Currently, most sesame seeds consumed domestically in Korea are imported from China and India (Korea's Ministry of Food and Drug Safety) (1). Primarily, the major reason for importing sesame seeds from the above mentioned countries is the increased prices of sesame seed cultivars of Korean origin as compared to those of Chinese and Indian origin. Owing to economic benefits, some traders have committed fraud, which sometimes includes false labeling of the sesame seed origins. To prevent this problem, analytical methods are required to distinguish domestic sesame seeds from imported sesame seeds.

Several studies have reported that the contents of the main components in sesame seeds are different depending on the origin of production, cultivating environment, and varieties (2–5). Some studies have discriminated the origins of sesame seed and sesame oil

by combining the quality components with the statistical method according to the origin of sesame seeds (6–8).

In recent years, analytical application by means of inductively coupled plasma-mass spectrometry (ICP-MS) has proven to be a boon for the quantitative determination of elements in agricultural products. Diverse researches have already been conducted to establish the origins of food and plant products by ICP-MS multi-element analysis, including pistachios (9), cabbage (10), rice (11), wine (12), coffee bean (13), tea (14), pomelos (15), and fermented cocoa beans (16).

To develop a model to discriminate the origin of sesame seeds, pragmatic statistical approaches—principal component analysis (PCA) and discriminant analysis—were combined with ICP-MS multi-element content analysis in this study. First, PCA was employed to provide meaningful information regarding an unsupervised pattern recognition analysis. This was conducted without any obvious requirement of knowledge from previously employed dataset. PCA serves the purpose of reducing the dimensionality of the dataset of multivariate nature and renders meaningful information in terms of smaller components—also known as principal components—for the interpretation of variance indices. The suitability of PCA lies in its ability to find tendencies and patterns. However, the main limitation

of PCA is non-assigning class numbers to samples of unknown statuses (17). As far as the supervised pattern recognition is concerned, establishment of a class model was conducted on the basis of experimental data for the assignment of unknown samples of the class of a previously defined sample (based on recognized pattern). Supervised pattern recognition can establish a classification model based on experimental data in order to assign unknown samples to a previously defined sample class based on the pattern it recognizes. Discriminant analysis, which is based on discriminant functions, is the best studied supervised pattern recognition method (18).

In our previous study (19), the geographical origin of sesame seed has also been discriminated using X-ray fluorescence spectrometry equipped with energy dispersive capability and near-infrared spectrometry. The present study was conducted sequentially after the previous study to investigate the possibility of discrimination of sesame seeds with respect to their geographical origin using ICP-MS multi-element analysis data combined with PCA and discriminant analysis.

Materials and Methods

Sample preparation In total, 123 sesame seed samples of diverse origins (Korean origin, $n=52$; Chinese origin, $n=48$; and Indian origin, $n=23$) were used in this study. The procurement of these samples was performed from indigenous grocery stores and Korean manufacturers dealing in sesame seed oil in 2014. These 123 samples were divided (in a random manner) into two groups. The first group comprised the calibration group and second group was named as the validation group. To create the discriminant calibration models, 113 sesame seed samples were used, whereas 10 samples (Korea, $n=4$; China: $n=4$; and India, $n=2$) were used to validate these models.

All samples were ground into a powder before use. According to the Food Code for trace nutrients test (dry digestion of inorganic component), 0.3 g of sesame seeds were accurately weighed into a porcelain crucible, pre-burnt on a burner, and dried in a furnace (Fisher Scientific Co., Hampton, NH, USA) at 450°C until it became ash. The residue was dissolved in 0.5-M HNO₃, filtered (Whatman filter paper No. 5A) into a 50-mL volumetric flask, and filled with 0.5-M HNO₃. Each sample was analyzed in triplicate.

Multi-element determination by ICP-MS The prepared sample solutions were analyzed using ICP-MS for ²⁴Mg, ²⁷Al, ³⁹K, ⁴³Ca, ⁵²Cr, ⁵⁵Mn, ⁶³Cu, ⁵⁹Co, ⁶⁰Ni, ⁶⁶Zn, ⁸⁵Rb, ⁸⁸Sr, ¹¹¹Cd, ¹³⁷Ba, and ²⁰⁸Pb. Each sample was analyzed in triplicate and quantified. Multi-element calibration standard 2A (Agilent Technologies, Santa Clara, CA, USA) was used to create 6-point calibration curves. Good linearity ($R^2>0.99$) was observed from all calibration curves. The analysis was conducted using ICP-MS (Agilent 7700 Series; Agilent Technologies) attached to an octopole reaction system (ORS) to determine the contents of multiple elements in a sample. Prior to ICP-MS multi-

Table 1. Recovery rate (%) and standard deviation (SD) of elements added into sesame seeds, limit of detection (LOD), and limit of quantitation (LOQ) of elements analyzed using inductively coupled plasma-mass spectrometry (ICP-MS)

Element	High concentration	Low concentration	LOD (μg/kg)	LOQ (μg/kg)
	Mean±SD ⁴⁾	Mean±SD		
Mg ¹⁾	83.0±0.5	82.0±0.3	0.666	2.018
Ca ¹⁾	100.4±7.3	117.2±7.2	3.493	10.584
K ¹⁾	88.0±5.6	107.2±3.3	2.927	8.870
Al ²⁾	104.2±6.1	108.0±2.2	1.855	5.621
Mn ²⁾	87.5±4.8	109.2±4.4	0.709	2.148
Cu ²⁾	103.4±3.9	98.3±3.6	0.341	1.032
Zn ²⁾	99.0±7.8	86.3±4.4	0.880	2.666
Rb ²⁾	95.6±1.8	84.1±3.6	0.390	1.183
Sr ²⁾	96.2±1.5	86.3±3.7	0.312	0.946
Ba ²⁾	98.2±3.3	82.7±5.0	0.754	2.285
Cr ³⁾	102.4±1.9	112.3±3.5	0.455	1.378
Co ³⁾	99.9±5.2	97.0±6.1	0.331	1.003
Ni ³⁾	106.0±8.2	111.0±4.9	0.755	2.288
Cd ³⁾	100.4±4.8	112.0±5.9	0.429	1.300
Pb ³⁾	95.5±3.5	110.6±6.4	0.182	0.552

¹⁾High concentration: 5,000 mg/kg added; Low concentration: 1,000 mg/kg added.

²⁾High concentration: 50 mg/kg added; Low concentration: 10 mg/kg added.

³⁾High concentration: 1.0 mg/kg added; Low concentration: 0.2 mg/kg added.

⁴⁾Mean values obtained from three measurements.

element analysis, method validation was conducted under the same conditions using sesame seeds added with multi-element standards. The limit of detection (LOD) and limit of quantitation (LOQ) of the instrument were calculated using the following equations (20):

$$\text{LOD}=3.3 \sigma/S \text{ and } \text{LOQ}=10 \sigma/S,$$

where σ is the standard deviation and S is the individual slope. Multi-element recovery rates (%) in the sesame seeds added with standards, LOD, and LOQ of all elements are summarized in Table 1.

Statistical analysis All analyses were conducted in triplicate and mean values of multi-element concentrations of sesame seeds were used. Analysis of variance (ANOVA) technique was employed for the determination of differences in multi-element concentrations among sesame seeds originated from Korea, China, and India. When ANOVA F value was significant, the differences between the means of multi-element concentrations were determined using Duncan's multiple-range test. PCA and discriminant analysis were then performed using the concentrations of elements selected by the ANOVA test for 113 sesame seeds analyzed using ICP-MS. Following the stepwise method, discriminant analysis was used to generate two discriminant functions because there were three origins (18,21). All statistical analyses were performed using the software package IBM SPSS Statistics 20 (SPSS Inc., Chicago, IL, USA).

Results and Discussion

Elemental comparison of sesame seeds of different origins (Korean, Chinese, and Indian) The concentrations of 15 elements (^{24}Mg , ^{27}Al , ^{39}K , ^{43}Ca , ^{52}Cr , ^{55}Mn , ^{63}Cu , ^{59}Co , ^{60}Ni , ^{66}Zn , ^{85}Rb , ^{88}Sr , ^{111}Cd , ^{137}Ba , and ^{208}Pb) were compared among the sesame seeds of Korean, Chinese, and Indian origins. Their mean values and standard deviations are summarized in Table 2. Based on the ANOVA test, significant ($p<0.05$) differences were shown by all elements among the sesame samples from the three origins. Duncan's multiple comparison test was then performed to identify how the element contents were different among the three origins.

The Korean sesame seed samples contained 3,518, 20.4, 17.5, 53.1, and 0.0415 mg/kg of Mg, Al, Mn, Zn, and Pb, respectively, which were significantly ($p<0.05$) higher than the Indian or Chinese samples. The Indian sesame seed samples contained the highest levels of Ca, Cu, Rb, Sr, and Co (at 9,805, 17.6, 10.4, 56.6, and 0.2691 mg/kg, respectively). The contents of K, Cr, and Co in the Chinese sesame seeds were significantly lower than those in the Korean or Indian samples ($p<0.05$). The Indian samples could be distinguished from the Korean and Chinese samples because they had the lowest Ba content. The Rb and Sr contents showed significant differences among the sesame seeds of the three origins ($p<0.05$). There was the high degree of standard deviations in few elements of the samples from the same region. This reflects that agricultural products have great variability although they are cultivated in the same locality (22). However, the significant differences among the element concentrations of the three origins made it possible to distinguish them based on their origins, thus providing reliable results for further statistical analyses.

Principal component analysis (PCA) PCA has been frequently used to establish a classification model based on multi-element content differences among samples from different provenances (17,23-25). To evaluate the differences among sesame seeds originated from Korea, China, and India, the concentrations of 15 elements analyzed using ICP-MS with significance at $p<0.05$ were subjected to PCA. The PCA can transform originally measured variables to new principal components (PCs), also known as uncorrelated variables. Each PC is a linear combination of the 15 original measured variables (concentrations of 15 elements). When PCA was performed with these variables, the total accumulative variance from the first five PCs accounted for 77.8% of the total variance. Only the first five PCs were accepted to reduce the 15 original ones because their eigenvalues were over 1.0, which was the mean of all eigenvalues.

As shown in Fig. 1, three groups were clearly discriminated on the basis of their respective spatial positioning along the first two principal components, although the accumulative contribution rate from PC 1 and PC 2 to the total variance was 49.7% (The figure using five PCs could not be shown). PCA results showed that multi-element concentrations of sesame seeds could be used to obtain the cluster trend of sesame seeds according to their geographical origins, although a few plots were overlapped.

Discriminant analysis Discriminant analysis was performed based on the results of chemical analysis for 15 variables: ^{24}Mg , ^{27}Al , ^{39}K , ^{43}Ca , ^{52}Cr , ^{55}Mn , ^{63}Cu , ^{59}Co , ^{60}Ni , ^{66}Zn , ^{85}Rb , ^{88}Sr , ^{111}Cd , ^{137}Ba , and ^{208}Pb . All elements were subjected to discriminant analysis because they showed significant differences among sesame samples with the three origins in the ANOVA test. However, only eight variables (Mg, K, Ca, Mn, Co, Rb, Sr, and Ba) were applied in the discriminant

Table 2. Ranges, mean values, and standard deviation (SD) of elements in sesame seeds cultivated in Korea, China, and India based on ICP-MS measurement

Elements	Korean ($n=48$)		Chinese ($n=44$)		Indian ($n=21$)	
	Range	Mean ($\pm$ SD)	Range	Mean ($\pm$ SD)	Range	Mean ($\pm$ SD)
Mg ¹⁾	2,758–4,165	3,518 ^c $\pm$ 425	2,586–3,938	3,187 ^b $\pm$ 313	2,281–3,584	2,859 ^a $\pm$ 306
Al ¹⁾	4.4–81.6	20.4 ^b $\pm$ 15.1	4.4–42.9	14.1 ^a $\pm$ 8.6	7.0–25.4	14.0 ^a $\pm$ 5.2
K ¹⁾	3,843–6,955	5,363 ^b $\pm$ 776	3,641–5,990	4,589 ^a $\pm$ 502	4,078–6,576	5,067 ^b $\pm$ 580
Ca ¹⁾	2,775–9,395	7,133 ^a $\pm$ 1,466	6,855–11,279	8,900 ^b $\pm$ 1,152	7,571–12,730	9,805 ^a $\pm$ 1,409
Mn ¹⁾	12.9–33.0	17.5 ^b $\pm$ 3.5	9.7–20.1	13.8 ^a $\pm$ 2.3	12.0–18.3	14.8 ^a $\pm$ 1.6
Cu ¹⁾	5.9–18.7	13.9 ^a $\pm$ 2.6	12.4–19.6	15.7 ^b $\pm$ 1.4	14.4–21.6	17.6 ^a $\pm$ 1.5
Zn ¹⁾	35.2–71.5	53.1 ^b $\pm$ 8.4	36.1–54.7	45.7 ^a $\pm$ 4.9	39.9–59.4	48.7 ^a $\pm$ 4.4
Rb ¹⁾	1.5–8.5	4.35 ^a $\pm$ 1.8	3.9–10.6	6.62 ^b $\pm$ 2.0	8.1–13.7	10.4 ^c $\pm$ 1.5
Sr ¹⁾	3.54–46.4	20.6 ^a $\pm$ 8.4	12.5–54.9	26.4 ^b $\pm$ 10.8	33.9–86.5	56.6 ^c $\pm$ 13.9
Ba ¹⁾	3.82–45.1	16.1 ^b $\pm$ 7.4	9.0–42.7	16.5 ^b $\pm$ 5.3	3.12–20.0	4.95 ^a $\pm$ 3.6
Cr ²⁾	47–197	104.3 ^b $\pm$ 33.8	34–250	82.4 ^a $\pm$ 47.0	49–286	119.4 ^b $\pm$ 55.0
Co ²⁾	43–566	177.7 ^b $\pm$ 99.7	44–228	118.7 ^a $\pm$ 42.1	201–358	269.1 ^c $\pm$ 41.0
Ni ²⁾	507–3,043	1418 ^b $\pm$ 520	644–3,159	1,452 ^b $\pm$ 648	709–1,265	953 ^a $\pm$ 119
Cd ²⁾	31–352	90.4 ^b $\pm$ 48	2–212	86.6 ^b $\pm$ 52	21–53	36.3 ^a $\pm$ 7.3
Pb ²⁾	17–92	41.5 ^b $\pm$ 16.9	12–61	29.3 ^a $\pm$ 10.8	13–38	23.4 ^a $\pm$ 6.5

¹⁾The unit for the elemental concentration was mg/kg.

²⁾The unit for the elemental concentration was $\mu\text{g/kg}$.

^{a-c)}Mean values show differences at $p<0.05$ among origins based on ANOVA.

n =sample size.

Fig. 1. Score cluster plot with principal components (PC) 1 and 2 of the multi-element contents in sesame seeds of different origins analyzed using inductively coupled plasma-mass spectrometry.

Fig. 2. Plot of discriminant analytical values (function 1 and function 2) of the multi-element contents in sesame seeds cultivated in Korea, China, and India

analysis as a stepwise method of variables was used. The linear combination of the eight variables produced two discriminant functions because there were three groups of samples. Each discriminant function was evaluated for its statistical significance on the basis of Wilks' Lambda factor (the ratio of within-groups' sums of squares to the total sums of squares). The range of Wilks' Lambda factor is from 1.0, which donates zero discriminatory power, to 0.0, which demonstrates perfect discrimination (21,26). The discriminant function 1 (Wilks' Lambda=0.049, $p=0.000$, and canonical correlation=0.937) and discriminant function 2 (Wilks' Lambda=0.402, $p=0.000$, and canonical correlation=0.773) were found to be statistically significant. The discriminant function 1 explained 82.9% of the total variance, whereas the discriminant function 2 accounted for 17.1% of the total variance. The classification model was based on the

Table 3. Classification results of the canonical discriminant functions of multi-element contents analyzed using ICP-MS

	Predicted origin			Total	Correctly classified (%)
	Korea	China	India		
Korean sesame seeds	39	9	0	48	81.3
Chinese sesame seeds	0	44	0	44	100.0
Indian sesame seeds	0	0	21	21	100.0
Total				113	92.0

discriminant scores of each sample calculated in the two discriminant functions (Fig. 2).

When reclassification of the total number of sesame seeds (113) was conducted using canonical discriminant functions, 92.0% of the samples were assigned to the correct production site or origin (Table 3). Only 9 seeds out of 48 Korean sesame seeds were misclassified as Chinese sesame seeds. All Chinese and Indian sesame seeds were classified correctly. The discriminant score plot clearly distinguished the three groups of sesame seeds, although 9 Korean sesame seeds were misclassified.

For validation of the developed discriminant model, the concentrations of eight elements in the ten validation samples (4 Korean, 4 Chinese, and 2 Indian samples) were employed as variables for Fisher's linear discriminant functions. Determination of the geographical origin of an unknown sample was performed with respect to the largest value calculated in Fisher's linear discriminant functions of the three origins. All predictions were correct for all 10 samples in terms of their geographical origins.

Multi-element analysis using ICP-MS or inductively coupled plasma-atomic emission spectroscopy has been utilized in an effective manner for the discrimination of different agricultural products in terms of their geographical origins as the elemental patterns in agricultural products demonstrated resemblance with the patterns observed in the soil (27). For example, Ma *et al.* (22) applied ICP-MS to discriminate the geographical origin of Chinese green tea (*Dongting Biluochun*) with an accuracy of 96.4%. Pilgrim *et al.* (14) discriminated the origin of tea samples (*Camellia sinensis*) from Asia with an accuracy of 97.6%. Bertoldi *et al.* (28) discriminated the origin of cacao beans with 100% accuracy. Chinese wheat samples harvested in 2007 and 2008 have also been analyzed using ICP-MS to classify their origins with accuracies of 84.2 and 95.8%, respectively (26). Moreover, the origins of paprika, wine, and rice have been successfully discriminated using ICP-MS (29-31). In these studies, discrimination models between different provenances were developed with multivariate analysis for the statistical analyses of the obtained ICP-MS data. Although the present results showed a lower discrimination rate than previous studies, this study demonstrated that a combination of ICP-MS concentration determination for multiple elements and multivariate analysis could be used to discriminate the origin of sesame seeds.

Disclosure The authors declare no conflict of interest.

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