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RESEARCH NOTE

Classification of geographical origin of kimchi by volatile compounds analysis using an electronic nose

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Abstract Food authenticity is one of the largest concerns in recent days. As kimchi has been a global food, its production origin has been important issue, particularly due to the large import from China. Among the potential methods, electronic nose which can measure volatile compounds in foods is considered to be a powerful device for identifying country of production. This study is to classify 69 kinds of kimchi produced in South Korea (39) and China (30) by analyzing volatile compounds in kimchi using electronic nose-mass spectrometry. Two widely used multivariate analyses, discriminant function analysis and principal component analysis, were used. Results showed that both multivariate analyses can completely separate Korean and Chinese kimchi using 10 kinds of molecular weights among 10–160 amu. The results indicate that the volatile compounds in kimchi are a suitable target to determine the geographical origin of kimchi.

Keywords Food authenticity · Electronic nose · Kimchi · Discriminant function analysis · Principal component analysis

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Introduction

Kimchi is a traditional vegetable fermented food representing Korea, which is well known for its unique flavor and functional aspect. Kimchi is made with can be made with various vegetables, and the most general source is Chinese cabbage. Kimhi contains various nutrients, such as vitamin C, B_1, and B_2, β-carotene, chlorophyll and minerals, and functional ingredients, e.g., β-sitosterol, unsaturated fatty acids, capsaicin, glucosinolate, isothiocyanate, indole, and allyl compounds (Cheigh et al., 1994; Park, 1995). Also, it has many organic acids, lactic acid bacteria, and dietary fiber (Choi et al., 2013). For this reason, it has been found to have an effect on dieting and skin health (Park et al., 2014) as well as constipation, colon cancer prevention (Park et al., 2006), anti-cancer action (Park, 1995), anti-aging action (Kim et al., 2011), antioxidative action (Kwon et al., 1998), and immunity enhancement (Park et al., 2014).

Kimchi has been globally recognized as one of healthy foods, a few famous national foods including Spanish olive oil, Greek yogurt, Indian lentils and Japanese soybean. As globalization of kimchi increases the export and import of kimchi various foods, the need for protection of producers and consumers is emphasized, and kimchi is no exception. In particular, the Chinese kimchi problem (detection of lead and parasite eggs in kimchi) that occurred in 2005 is an example of the adverse effects of food exports and imports, thereby increasing the importance for indicating origin of kimchi production. Currently, significant amount of kimchi produced in China with unqualified Chinese cabbage and low labor cost are imported, but it is difficult to properly inspect the quality. For this reason, identification of production origin of kimchi is required for safe kimchi consumption and for globalization of kimchi (Hwang, 2006).

The electronic nose is capable of analyzing aromatic components in a short time using the gas sensor and pattern recognition software (Röck et al., 2008). Due to this characteristic, the electronic nose can qualitatively analyze the fragrance and volatile components in foods, and has been used for evaluating the quality of various foods whose fragrance is important but sensory evaluation is limited (Ampuero and Bosset, 2003; Di Natale et al., 1997; Kim et al., 2004; Wilson, 2013). In recent years, some researches have been carried out to discriminate geographic origin of food sources by using electronic nose because it can analyze the sample without a specific preprocessing process. A research group tested the discrimination of origin of pine mushroom by using electronic nose and classify them by domestic and imported pine mushroom with 95.79% accuracy (Lee et al., 2006). Also, an electronic nose was used for classifying garlic cultivars with combing multivariate analysis, resulting that Thai garlic cultivars were consistently characterized by three groups (Trirongjitmoah et al., 2015). For other food product, volatile compounds in Emmental cheese sampled from various regions in Switzerland, and Finland were measured by mass spectrometry-based electronic nose, and analyzed using principal component analysis for classifying its geographic origin (Pillonel et al., 2003). The electronic nose also used to evaluating animal product, applying for measuring beef odor to estimate freshness (Kim et al., 2004). Also, Lim et al. (2008) used electronic nose to determine the country of origin of beef produced in South Korea, Australian, and New Zealand, showing the possibility of electronic nose application into livestock products (Lim et al., 2008), but the unevenness of fat content caused according to production area and beef breed limited the accuracy (Blixt and Borch, 1999). In the case of kimchi, because of its unique flavor the electronic nose is expected device for effectively analyzing the flavor-related components which is largely affected by seasonings and aging. Shin et al., (2005) analyzed the aroma pattern and acidity of Chinese cabbage kimchi, which changes with storage time, using electronic nose (Shin et al., 2005). Another study classified the origin of Chinese cabbage cultivated either in South Korea or China based on volatile compounds measured by electronic nose with multivariate analysis, showing geographic origin of Chinses cabbage could be discriminated through volatile compounds when adequate statistical analysis was applied (Lee et al., 2017). Also, electronic noses have been used to examine the possibility of grading the spicy taste of red pepper powder, an important ingredient of kimchi (Kang et al., 2010).

Despite of current emphasis on production area of kimchi, only its ingredients was attempted to classify by their geographic origins, and the area of production of kimchi has never been tested. This study, hence, is to determine area of production of kimchi based on multivariate analysis of volatile compounds in kimchi measured by electronic nose. Specifically, kimchi produced either in South Korea or China was analyzed by electronic nose to measure volatile compounds, and then 2 types of multivariate analyses were performed to classify them by the area of production.

Materials and methods

Samples

Total 69 kinds of kimchi, including 30 kinds of kimchi produced at kimchi factories in northeastern China, 19 kinds of kimchi made directly from Korean cabbages, and 20 kinds of Korean kimchi produced at small factories and sold at large supermarkets were used for the experiments. With helps from Korea Central Customs Laboratory, kimchi produced in China were regionally collected from Weihai (5 types), Lushan (4 types), Bunsi (2 types), Angu (1 type), Yantai (1 type), Mundeung (1 type), and Qingdao (16 species), where many kimchi factories for exporting to South Korea are located. Commercial kimchi produced in Korea (20 types) was purchased at a domestic supermarket chain, and more kimchi was prepared by ourselves using cabbages from Jeonnam (7), Gangwon (6), and Chunnam province (6). All kimchi samples were put in polybags and stored at −20 °C for the experiments.

Sample analysis

About 1 kg of kimchi samples were sliced, mixed, then homogenized using house-hold blender at 4°C. The volatile compounds of the samples were analyzed using Smart Nose® system (Smart Nose SA, Marin-Epagnier, Switzerland). The analysis was conducted as previously described by Lee et al., (2017) without any modification. Briefly, 4 g of each sample was used and heated at 90 °C for 10 min. The detection range for the mass analysis was 10 to 160 amu. Three replicates were analyzed separately for each sample.

Multivariate analysis

Multivariate analysis is a statistical method for the interpretation of multidimensional interdependence or dependency of variables as previous study successfully discriminate Chinese cabbage cultivated either in South Korea and China (Lee et al., 2017). There are a few types of multivariate analyses: principal component analysis

(PCA), discriminant function analysis (DFA), factor analysis, cluster analysis and canonical correlation analysis (Afifi et al., 2019). Among them, PCA and DFA are commonly used in the analysis of agricultural products and food data (Choi et al., 2012; Yang et al., 2015; Yang and Irudayaraj, 2000), deciding to use the PCA and DFA in this study.

Before applying the DFA, we initially removed molecular weights (MW) which were not significant for classifying the origin, resulting in removing 29 MWs. Then, all the significant MWs were used in DFA to calculate DFA scores and for constructing the linear combination of them (called DFA function) which could be used to deciding the dividing points of two groups (Afifi et al., 2019). From this analysis, we additionally listed the MWs according to the determinant of coefficient assigned for each MW, and selected ten MWs which showed the determinant of coefficient larger than 0.5; thus, they would be more effective in classifying the origin with the DFA. The best MWs were used for further DFA application and compared the result with the DFA with all the significant MWs to consider the number of MWs necessary for classifying the origin. Finally, the classification performance was calculated based on the number of correctly and incorrectly classified kimchi by their origins. A plain composed by the first and second DFA functions were then constructed to project DFA scores. PCA was used as a comparable method that confirmed the classification ability in the kimchi origin. Because PCA is unsupervised method, all the MWs were implemented to produce principal components (PCs) by linearly combining MWs based on covariance matrix because of unit consistency among MWs. After calculating the PCs, we selected the first two PCs (PC1 and PC2) which cumulatively explained approximately 80% of total variance in MWs (Afifi et al., 2019; Shim et al., 2018). The selected PC1 and PC2 were then composed a plain to project eigenvectors assigned to each MWs and to display classification ability.

Statistical analysis

Multivariate analysis was used to determine the area of production of kimchi based on data measured volatile compounds by electronic nose. All statistical analyses were performed using SAS statistical software package (Version 9.4, SAS Institute Inc., Cary, NC, USA) and statistical significance was $p < 0.05$.

Results and discussion

Nowadays, food authenticity is very important for protecting customer's rights and domestic industries as well as food safety concerns. In 2020, more than 300,000 metric ton of kimchi was imported and mostly from China. South Korean customers do not favor on imported kimchi from China, so 80% of imported kimchi is distributed to restaurants. It is mandatory that origins of foods should be notified in all types of restaurants in South Korea. However, food adultration often happens especially on origin of kimchi production due to the difficulties of discrimination by sensory evaluations.

Volatile compounds measured in this study had MWs between 10 and 160. Based on the data of volatile compounds measurement, DFA showed that volatile compound with molecular weight of 120 was the most significant factor in classifying the area of production either in South Korea or in China (Table 1). Besides it, a few volatile compounds less than MW 100 amu were key factors in the discrimination, in general. This is a bit different with the previous study which demonstrated that volatile compounds of MW larger than 130 amu were crucial for classifying Chinese cabbage cultivated in South Korea and China (Lee et al., 2017). This difference in key classification compounds may be due to the effect of seasoning of kimchi added to Chinses cabbage (Ku et al., 2005; Park and Lee, 2005; Ryu et al., 1984). For example, the characteristics of red pepper powder, a main ingredient of kimchi seasoning, affects a flavor of kimchi (Ku et al., 2001; Lim et al., 2014). In particular, geographic origin of red pepper powder produce in South Korea and China was classified by near infrared reflectance spectroscopic analysis, suggesting that the volatile compounds originated from red pepper powder was important for this classification (Kwon

Table 1 Key molecular weight of volatile compounds for classifying kimchi by country of production

MW (amu)	R^2	F-value	*P*-value
120	0.61	106.79	< .0001
48	0.59	94.56	< .0001
73	0.57	89.11	< .0001
45	0.57	87.33	< .0001
61	0.55	82.60	< .0001
94	0.55	81.19	< .0001
46	0.54	78.08	< .0001
23	0.53	74.78	< .0001
121	0.50	67.22	< .0001
36	0.50	66.36	< .0001

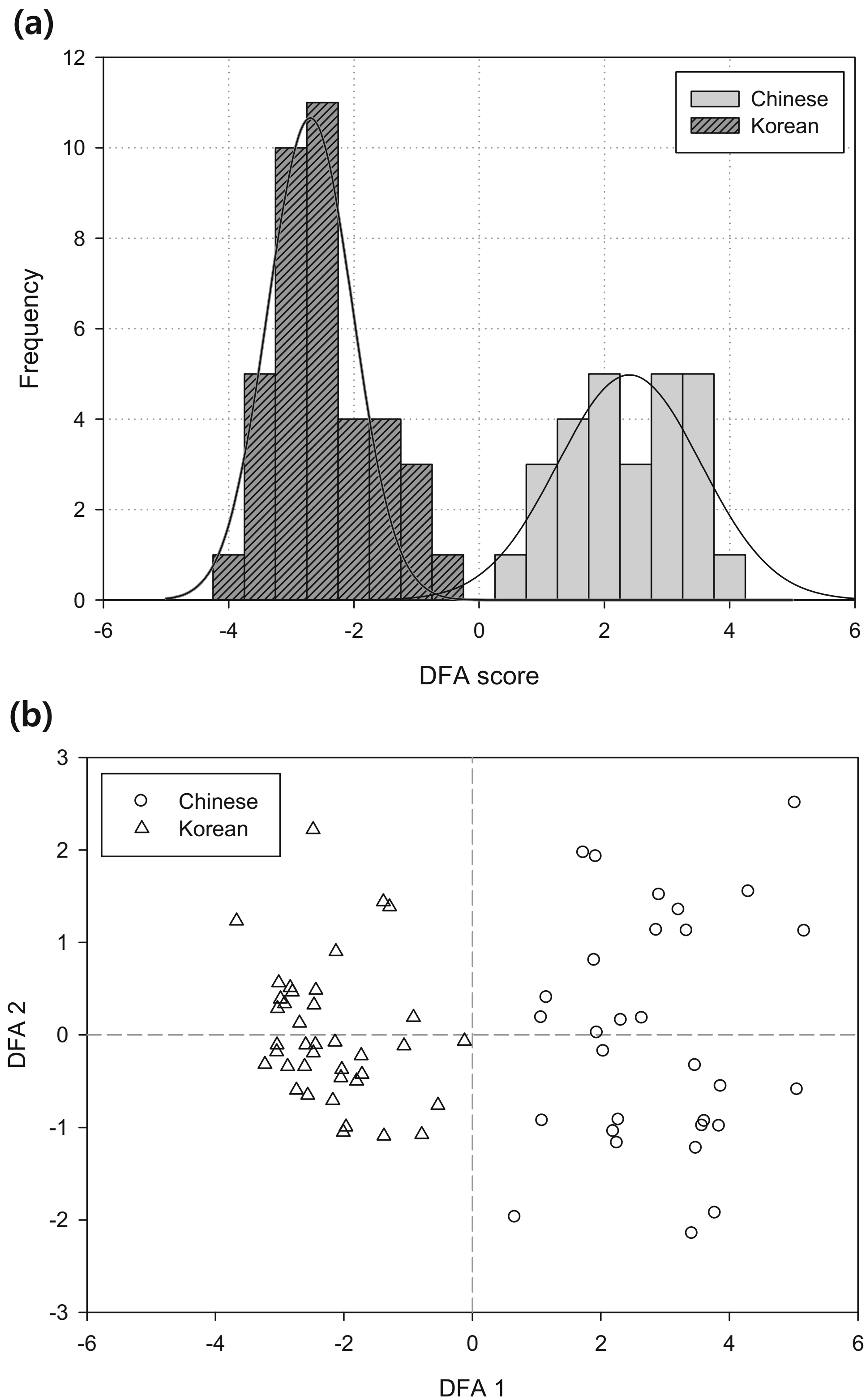

Fig. 1 Results of classification illustrated by (a) frequency distribution of DFA score and (b) projection on the plane constructed by DFA function 1 and 2. Area of overlapping region in **(A)** indicates the risk of incorrect classification; thus, the area is small in this figure, suggesting the high classification ability of electronic nose with DFA, while **(B)** shows complete separation of Korean and Chinese kimchi

et al., 1999). However, it may be necessary to identify the ingredient matching to the key MWs of volatile compounds by using further analyzing device.

With only one variable (MW 120), DFA mistakenly identified 7 of Korea-produced kimchi (76.7% accuracy), and 1 of China-produced kimchi (97.4% accuracy). The lower accuracy in classifying Korean kimchi may be due to

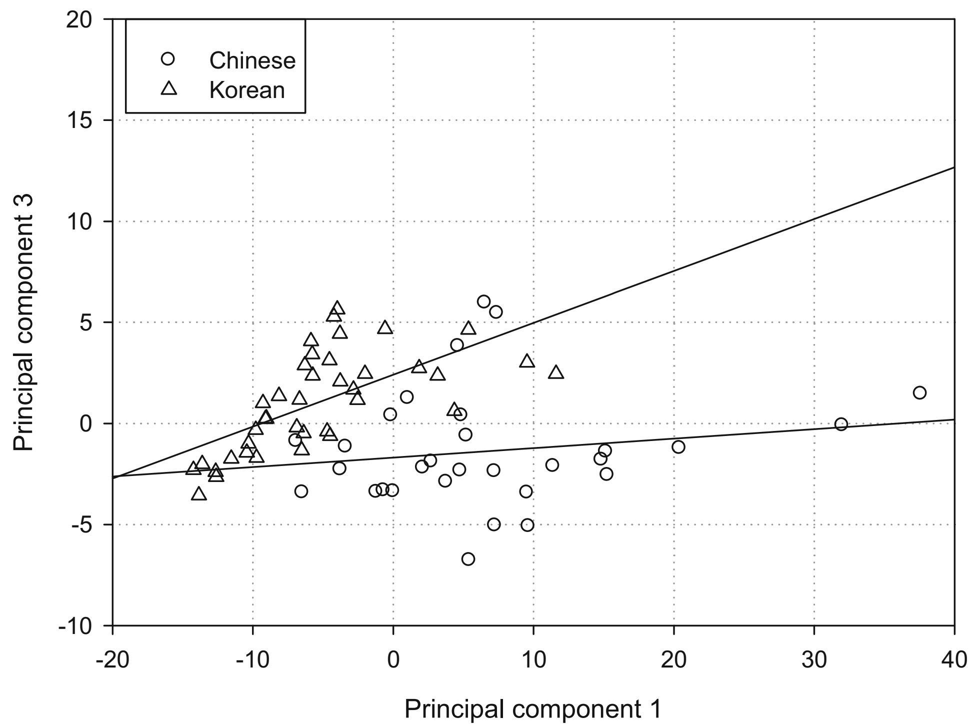

Fig. 2 Statistical fitting of Korean and Chinese kimchi using two main PCs. Two clearly different slopes suggest that Korean and Chinese kimchi are separable by electronic nose

various local characteristics of kimchi in South Korea as the origin of it (Ku et al., 2005; Lee et al., 2004; Ryu et al., 1984). In addition, quality characteristics of red pepper were affected by cultivation area in South Korea, suggesting that red pepper powder might increase the variations in kimchi characteristics and consequently reduce the classification accuracy (Hwang et al., 2011). This result suggested that the accuracy of discrimination of origin by only one volatile compound was not reliable and more components were required. To increase the classification accuracy, we gradually added the number of MW of volatile compounds selected based on statistical significance until the discrimination accuracy reached 100% (Table 1). Finally, kimchi produced either in South Korea or in China was perfectly separated with 10 MWs of volatile compounds. This indicated that only 10 key MWs of volatile compounds among 10 to160 amu (6.25% of total MW) can fully classified kimchi into Korean and Chinese kimchi.

Figure 1 shows the discrimination ability of DFA through the distribution of DFA score calculated based on the electronic nose data. In the (Fig. 1(A)), the region where the two normal distribution curves overlapped was small enough not to misclassify the area of production, suggesting that classification of Korean and Chinses kimchi correctly achieved. (Fig. 1(B)) shows the distribution of electronic nose data of Korean and Chinese kimchi distributed by the major DFA functions which are developed by the linear combination of 10 key MWs of volatile compounds (Afifi et al., 2019). It was observable that two data were completely divided based on DFA 1 axis at point 0 and indicated that the 10 key MWs could completely classify the kimchi produced either in South Korea or China.

Because the classification procedure for agricultural products is usually carried out without any prior information, unsupervised statistical analysis is often necessary. For this reason, we used PCA for the electronic nose data of volatile compounds measured from either Korean or Chinese kimchi whether it provided the same classification result as DFA, a supervised method. (Fig. 2), a plane composed by two principal components, showed that regression lines fitting the data of Korean and Chinese kimchi had a distinctly different slope (0.257 for Korean and 0.047 for Chinese). This suggested that the two kimchi were completely separable, and demonstrated the consistent results with DFA. From two consistent results by DFA and PCA, it is conclusive that the electronic nose is applicable for classifying area of kimchi production with adequate multi-variate analysis. In particularly, the result of PCA further indicates that the electronic nose may be very useful for classifying large samples without prior information. As previously suggested, the reason for possible classification of Korean and Chinese cabbage may be because of geographical origin of Chinese cabbage, a main source of kimchi. Even though the key MWs of volatile compounds were completely different with the study for detecting origin of Chinese cabbage (Lee et al., 2017), the interaction between Chinese cabbage and other ingredients mainly caused by fermentation would affect the key volatile compounds (Ryu et al., 1984). Otherwise, some ingredient such as red pepper powders directly influences the classification of kimchi as it contains strong flavor that is generally from volatile compounds (Hwang et al., 2011; Ku et al., 2005).

Nowadays, kimchi becomes a popular food in many country and is well recognized as a typical Korean traditional food. Therefore, geographical origin of kimchi is more important than ever, because 'made in Korea' labels provide positive price-premium on the products. In this context, developing a practical methodology to identify the geographical origin of kimchi is of great interest.

This study demonstrated the use of multivariate analysis (DFA and PCA) on data obtained from Korean and Chinese kimchi using electronic nose in order to analyze the origin of kimchi production. The multivariate analysis showed that 100% classification was possible by means of electronic nose-based measurement of volatile compounds only with a small amount of molecular weights. This was achieved by using relatively simple instruments with short analysis time. Academically, however, the current method did not account for which ingredient in kimchi is crucial for its classification. To identify the important ingredients of classifying kimchi by its origin of production, further analysis able to figure out the corresponding MWs may be necessary.

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