



Discrimination of wine from grape cultivated in Japan, imported wine, and others by multi-elemental analysis

Hideaki Shimizu,* Fumikazu Akamatsu, Aya Kamada, Kazuya Koyama, Masaki Okuda, Hisashi Fukuda, Kazuhiro Iwashita, and Nami Goto-Yamamoto

National Research Institute of Brewing, 3-7-1 Kagamiyama, Higashi-Hiroshima, Hiroshima, 739-0046, Japan

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Differences in mineral concentrations were examined among three types of wine in the Japanese market place: Japan wine, imported wine, and domestically produced wine mainly from foreign ingredients (DWF), where Japan wine has been recently defined by the National Tax Agency as domestically produced wine from grapes cultivated in Japan. The main objective of this study was to examine the possibility of controlling the authenticity of Japan wine. The concentrations of 18 minerals (Li, B, Na, Mg, Si, P, S, K, Ca, Mn, Co, Ni, Ga, Rb, Sr, Mo, Ba, and Pb) in 214 wine samples were determined by inductively coupled-plasma mass spectrometry (ICP-MS) and ICP-atomic emission spectrometry (ICP-AES). In general, Japan wine had a higher concentration of potassium and lower concentrations of eight elements (Li, B, Na, Si, S, Co, Sr, and Pb) as compared with the other two groups of wine. Linear discriminant analysis (LDA) models based on concentrations of the 18 minerals facilitated the identification of three wine groups: Japan wine, imported wine, and DWF with a 91.1% classification score and 87.9% prediction score. In addition, an LDA model for discrimination of wine from four domestic geographic origins (Yamanashi, Nagano, Hokkaido, and Yamagata Prefectures) using 18 elements gave a classification score of 93.1% and a prediction score of 76.4%. In summary, we have shown that an LDA model based on mineral concentrations is useful for distinguishing Japan wine from other wine groups, and can contribute to classification of the four main domestic wine-producing regions of Japan.

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[Key words: Elemental composition; Geographical origin; Inductively coupled-plasma mass spectrometry; Inductively coupled plasma-atomic emission spectroscopy; Metrology; Multi-element analysis; Wine]

Official rules for the labeling of Japan wine were established on October 30, 2015, and will be enforced in October 2018 to improve the brand value of Japan wine, which is defined as wine that is domestically made from grapes cultivated in Japan only (1). Currently, there are two types of domestic wine in the Japanese wine market: the above-mentioned Japan wine, and domestically produced wine mainly from foreign ingredients (DWF), mostly including imported concentrated grape juice. Until now, these two types of wine (i.e., Japan wine and DWF) have been categorized in the same group as wine made in Japan. Thus, it is not easy for consumers to distinguish Japan wine from DWF in the marketplace. Against this background, from the perspectives of easy-to-understand labeling for consumers and improving the brand value of Japan wines, the National Tax Agency, Japan established “Labeling Standards for Manufacturing Process and Quality of Wine, etc.” pursuant to law (1). As a result, there is a need to develop scientific analytical methods to verify the authenticity of Japan wine.

Several analytical techniques have been reported to discriminate the geographic origin of wine, including strontium stable isotopes by inductively coupled-plasma mass spectrometry (ICP-MS) (2); (D/H)₁, (D/H)₂ in wine ethanol by site-specific natural

isotope fractionation coupled with nuclear magnetic resonance (3); and stable isotopes of oxygen in wine water by isotope ratio mass spectrometry (3). One of the mostly applied techniques is mineral composition determined by ICP-MS and inductively coupled-plasma atomic emission spectrometry (ICP-AES).

The mineral composition of wine is thought to be primarily influenced by that of the viticulture soil: some of the minerals in soil are absorbed into grapes through the vine roots, and minerals in grape are subsequently transferred to wine through the wine-making process. Although many other factors, including viticulture (fertilization, pesticides, and fungicides), heavy-metal pollution, and the wine-making process (fermentation process, winery equipment, clarification with bentonites, and filtration practices), influence the mineral composition of wine (4,5), harvesting region is thought to be a key factor. Therefore, many studies based on mineral composition by ICP-MS and ICP-AES have been conducted in wine regions and countries worldwide to distinguish the geographic origin of wines. For example, Kwan et al. (6) reported the relationship between 17 elements analyzed by ICP-AES and geographic origin in a study of 40 wines of *Vitis vinifera* cv. Pinot Noir from France and the United States. Furthermore, classification of the geographic origin of seven countries (Bulgaria, Chile, France, Hungary, Italy, Spain, and USA) by analysis of 23 elements using ICP-MS was examined by Serapinas et al. (7). In addition, Gómez et al. (8) and Thiel et al. (9) attempted to classify and predict

* Corresponding author. Tel.: +81 82 420 0812; fax: +81 82 420 0850.
E-mail address: h.shimizu@nrib.go.jp (H. Shimizu).

German wines according to the analysis of several elements by ICP-MS. Other similar studies have been carried out in Italy (10,11), Spain (12,13), Portugal (14), United States (5), Canada (15,16), Australia (17), New Zealand (18), South Africa (19,20), Czech Republic (21,22), Romania (23), and China (24,25).

In terms of Japan wine, a previous study examined the discrimination of domestic wine from imported wine by ICP-AES, which determined the concentrations of 21 major minerals in 99 wines from Japan and four other countries (26). The authors developed a linear discriminant analysis (LDA) model using five elements and identified the regional origin of domestic wines, which were actually Japan wine by the new definition (1), with 91% accuracy (26).

To our knowledge, however, there are no reports of the discrimination of Japan wine from DWF or the discrimination of geographic origin within Japan. Here, therefore, we have analyzed the mineral composition of 214 wine samples by ICP-MS and ICP-AES, and carried out LDA analyses both for the discrimination of three wine groups (Japan wine, imported wine, and DWF) and for the discrimination of four main domestic wine-producing regions (Yamanashi, Nagano, Hokkaido, and Yamagata Prefectures).

MATERIALS AND METHODS

Wine samples Overall, 214 different bottled wines purchased from the Japanese market were sampled: Japan wine ($n = 82$), imported wine ($n = 99$), and DWF ($n = 33$). The 82 Japan wines were from four principal wine-producing regions, Yamanashi ($n = 23$), Nagano ($n = 18$), Hokkaido ($n = 16$), and Yamagata ($n = 15$), as well as other regions ($n = 10$). The 99 imported wines were from Europe ($n = 49$; France, $n = 21$, Italy, $n = 18$; and other, $n = 10$), the United States ($n = 13$), South America ($n = 16$; Chile, $n = 12$; and Argentina, $n = 4$), Oceania ($n = 14$; Australia, $n = 11$; and New Zealand, $n = 3$), and South Africa ($n = 7$). The 33 DWFs comprised wine made from imported concentrated grape juice only ($n = 17$), wine made from imported concentrated grape juice and blended with imported wine ($n = 13$), and wine that further used apple and kiwi ($n = 1$) and domestic grapes ($n = 2$), in addition to the above two ingredients.

Of the total 214 wines, 109 samples were red, 104 samples were white, and 1 sample was rosé. The 214 samples represented different grape varieties, including Chardonnay ($n = 29$), Cabernet Sauvignon ($n = 21$), Merlot ($n = 14$), Pinot Noir ($n = 10$), Koshu ($n = 8$), Muscat Bailey A ($n = 8$), and other ($n = 124$), and were vintages from 2010 to 2014 ($n = 176$) or 2002 ($n = 1$), or were non-vintage ($n = 37$; mainly DWF).

Sample preparation For ICP-MS and ICP-AES analysis, 2 mL of wine was mixed with 5 mL of 61% nitric acid (Kantokagaku, Tokyo, Japan) in a digestion vessel (SCP Science, Montreal, Canada) (27–29). The sample was left at room temperature for 60 min and then diluted by adding ultra-pure water (>18.1 M Ω) obtained via a MilliQ system (Millipore, MA, USA) to a total volume of 50 mL (final concentration 6% nitric acid).

Elemental analysis The mineral concentrations of all wine samples were analyzed by ICP-MS (7700x; Agilent Technologies, CA, USA) and ICP-AES (ICPE-9000; Shimadzu, Kyoto, Japan). Argon gas (purify higher than 99.99%) was used as the plasmogen and carrier gas.

The analytical conditions for ICP-MS were as follows: RF power, 1.55 kW; RF matching, 1.8 V; sampling position, 8.0 mm; carrier gas flow, 1.07 L/min; extract2, -160 V; gas mode, non-gas and He; integration time, 0.99 s. Be, Y, In and Tl (SPEX CertiPrep, NJ, USA) were included as an internal standard for ICP-MS at a concentration of 100 $\mu\text{g/L}$. For ICP-MS, the quantified concentrations of mineral elements were performed based on ten-point linear normalization using external standards of a calibration mixture standard solution including 34 elements (Ag, Al, As, B, Ba, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, Ge, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, Sb, Se, Si, Sn, Sr, Ti, V, W, Zn, and Zr) (SPEX CertiPrep) in the concentration range of 0, 1, 5, 10, 50, 100, 500, 1000, 5000, and 10,000 ng/L. The calibration standards solution was 6% nitric acid.

The analytical conditions for ICP-AES were as follows: RF power, 1.20 kW; plasma gas, 10.0 L/min; auxiliary gas, 0.6 L/min; carrier gas, 0.7 L/min; exposure time, 15, and detection wavelength: B, 249.773 nm; K, 769.896 nm; Na, 589.592 nm; P, 178.287 nm; S, 180.731 nm; Si, 251.611 nm; Ca, 422.673 nm; Mg, 285.213 nm. Y (Kanto Chemical, Tokyo, Japan) was included as an internal standard for ICP-AES at a concentration of 1 mg/L. For ICP-AES, the quantified concentrations of mineral elements were performed based on six-point linear normalization using external standards: B and Na (Merck KGaA, Darmstadt, Germany), 0, 50, 100, 500, 1000, and 5000 $\mu\text{g/L}$; P (Kanto Chemical), Si (Wako Pure Chemical Industries, Osaka, Japan) and S (Sigma-Aldrich, Co. MO, USA), 0, 100, 500, 1000, 2000, and 4000 $\mu\text{g/L}$; K (Kanto

Chemical), 0, 10, 30, 50, 100, and 150 mg/L. The calibration standards solution was 6% nitric acid.

For minerals present at less than 10 $\mu\text{g/L}$ in the sample, the concentration was determined by ICP-MS, whereas for those present at 10 $\mu\text{g/L}$ or higher, the concentration was determined by ICP-AES.

The concentrations of mineral elements were determined from triplicate measurements by using Mass Hunter Workstation software (B.01.01 G7201B) for ICP-MS analysis and ICPsolution (ver. 1.20) for ICP-AES analysis. The whole analysis was carried out at least twice, including the above-described sample preparation with nitric acid. Linear calibration regressions consisting of ten standard levels with correlation coefficients of >0.995 were established before sample analysis. The limit of detection (LOD) was determined as a 3-fold standard deviation of 10 replicate measurements of the blank solution.

Selection of elements for statistical analysis Among 40 mineral elements determined by ICP-MS and AES, 22 were eliminated for the following reasons: their concentrations were below LOD in many samples or the relative standard deviation of their measurements was $>10\%$. Thus, the remaining 18 elements (Li, B, Na, Mg, Si, P, S, K, Ca, Mn, Co, Ni, Ga, Rb, Sr, Mo, Ba, and Pb) were used for statistical significance testing in the discriminant analysis procedures. Among these, 7 (Li, Co, Ni, Ga, Mo, Ba, and Pb) and 3 (Mn, Rb, and Sr) elements were simultaneously determined by ICP-MS using sample dilutions of 1:25 and 1:250, respectively; and 6 (B, Na, Si, P, S, and K) and 2 (Mg and Ca) elements were simultaneously determined by ICP-AES using sample dilutions of 1:25 and 1:125, respectively. Only 2 of the 214 samples had a concentration of an element (both Li) below the LOD; in these cases, a concentration of LOD/10 was assigned for statistical significance testing.

Recovery rates of the 18 mineral elements were determined by spiking a wine sample with known amounts of each element according to Okuda et al. (29). Recovery rates from the spike recovery tests ranged from 82% (Ni) to 103% (Sr) by ICP-MS, and 93% (S) to 108% (Na) by ICP-AES.

Statistical analysis All statistical tests were performed by using JMP 12.2 (SAS Institute Inc., NC, USA). The differences in elemental concentrations among the three wine groups (Japan wine, imported wine, and DWF) and among the four main domestic wine-producing regions were independently tested by one-way analysis of variance (ANOVA). When ANOVA showed significant differences, the means were separated by using Tukey's honestly significant difference (HSD) test. In all tests, a p value of 0.05 was taken to indicate statistical significance.

Linear discriminant analysis (LDA) was carried out to discriminate the wine samples according to geographic origin. We established one model for three-group discrimination (Japan wine, imported wine, and DWF) and one for four-region discrimination (Yamanashi, Nagano, Hokkaido, and Yamagata). We performed 10-fold cross-validations to evaluate classification performance. In this approach, the overall set of samples was randomly divided into 10 approximately equal subsets. One of these subsets was then excluded from the main set and used as a test set, and the remaining samples were used as a training set. This calculation was repeated over the 10 subsets, and the discrimination model was evaluated by summing the accuracies of all predictions (26,30).

RESULTS AND DISCUSSION

Elemental profiling of three wine groups Among the three groups of wine (Japan wine, imported wine, and DWF), Japan wine showed characteristic features regarding mineral concentration: the concentration of potassium was higher, whereas those of 8 elements (Li, B, Na, Si, S, Co, Sr, and Pb) were lower as compared with the other two groups (Table 1). In particular, the lithium concentration of Japan wine was 20 times lower than that of DWF. As mentioned above, the potassium concentration was higher in Japan wine than in the other two groups; a high concentration of potassium in Japanese wine has also been reported by Horii et al. (26).

DWF also showed specific features (Table 1): the concentrations of five elements (Li, Na, Si, Ca, and Mo) were higher, whereas those of six elements (Mg, K, Mn, Ga, Rb, and Ba) were lower as compared with the other two groups.

Regarding imported wine, the concentrations of four elements (Mn, Ga, Rb, and Ba) were similar to those in Japan wine, while the concentrations of five elements (B, S, Co, Sr, and Pb) were similar to those in DWF.

Discrimination of three wine groups by LDA analysis An LDA model was created to discriminate the three wine groups (Japan wine, imported wine, and DWF) using 18 elements (Li, B, Na, Mg, Si, P, S, K, Ca, Mn, Co, Ni, Ga, Rb, Sr, Mo, Ba, and Pb). In the

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