

Typification of Cider Brandy on the Basis of Cider Used in Its Manufacture

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A study of typification of cider brandies on the basis of the origin of the raw material used in their manufacture was conducted using chemometric techniques (principal component analysis, linear discriminant analysis, and Bayesian analysis) together with their composition in volatile compounds, as analyzed by gas chromatography with flame ionization to detect the major volatiles and by mass spectrometric to detect the minor ones. Significant principal components computed by a double cross-validation procedure allowed the structure of the database to be visualized as a function of the raw material, that is, cider made from fresh apple juice versus cider made from apple juice concentrate. Feasible and robust discriminant rules were computed and validated by a cross-validation procedure that allowed the authors to classify fresh and concentrate cider brandies, obtaining classification hits of >92%. The most discriminating variables for typifying cider brandies according to their raw material were 1-butanol and ethyl hexanoate.

KEYWORDS: Typification; cider brandy; raw material; volatile

INTRODUCTION

Cider brandy is a popular drink elaborated by distilling fermented apple juice in alembics and is usually matured in oak woods barrels to improve its sensorial qualities. However, several technological factors, namely, raw material type, distillation technology, wood type, or maturation time, influence the chemical composition of the final product and, thus, the organoleptic characteristics of cider brandy.

The raw material is a decisive factor involved in the quality of distilled beverages. In this respect, the presence of acrolein (2-propenal) and allyl alcohol (2-propen-1-ol), considered to be unpleasant flavors, is related to the use of poor quality ciders (1). Likewise, the use of selected yeast strains could guarantee the production of an adequate brandy wine base (2). On the other hand, Mangas et al. (3) observed significant differences in the chemical composition of cider brandy depending on whether the cider was made from apple juice concentrate or fresh apple juice. Cider brandy obtained from fresh apple juice presented a higher content of 1-butanol, 1-hexanol, and ethyl acetate and a lower content of furfural.

A great number of reactions take place in the batch during distillation, such as esterification, acetalization, and dehydration. The type of distillation apparatus, the material employed in its construction, and the source of heat are well-known factors involved in the composition of distilled beverages. Rodríguez et al. (4) showed that Charentais alembics generate higher levels of methanol compared to a rectification column, whereas the latter system generates more fusel alcohols. Guymon and

Crowell (5) and Rodríguez et al. (6) observed that Charentais alembics produce more furfural than either a continuous still or a rectification column, and Cardoso et al. (7) detected higher methanol contents when a column was packed with aluminum. On the other hand, using a vacuum distillation system can simplify the operation of the equipment and lead to an overall reduction in the heating required, although there are problems of losses of desirable aromatic compounds (8).

The subsequent aging of spirits will change their chemical composition. During this stage, the wood can transfer a large number of compounds (phenolic and furanic compounds, sugars, oak lactones, and so on) to the distillate that improve its quality. These substances increase their concentration in cider brandy irrespective of the fresh distillate used; however, their presence in the spirit is related to the type of wood (American or French) and the aging time (6, 9, 10). Furthermore, additional changes in the fresh spirit take place that also affect the final quality of the final product. The pungent character due to acetaldehyde and acrolein is reduced as a consequence of their acetalization (4); ethanoic acid esters, responsible for fruity and floral notes, decrease as a result of transesterification with ethanol (4, 11, 12).

Phenolic and volatile compounds have been successfully employed in several characterization and typification studies of distilled beverages. However, both the number of compounds and the variety of factors that influence their levels in the final product necessitated the use of multivariate techniques. In this respect, pattern recognition techniques, such as cluster analysis, principal component analysis (PCA), linear discriminant analysis (LDA), Bayesian analysis, and soft independent modeling of class analogy, have generally been used. For instance, Lisle et

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al. (13) used cluster analysis to distinguish between brandies, whiskeys, and rums; Miyashita et al. (14) and Mazzerolles et al. (15) classified brandies according to their geographic origins; and Versini et al. (16) carried out the characterization of grappa from different Italian regions. More recently, our work group (17) used chemical composition to characterize cider brandy on the basis of aging time.

The significant influence of raw material on cider brandy quality and its commercial value made it necessary to carry out studies of characterization of cider brandy on the basis of the raw material employed in its manufacture. To accomplish this aim, accurate instrumental analysis techniques, such as GC-MS for volatile analysis, and multivariate techniques were employed.

MATERIALS AND METHODS

Standards and Samples. All standards were of analytical quality. Ethyl esters were supplied by Fluka (Buchs, Switzerland), acetaldehyde by Merck (Darmstadt, Germany), fatty acids by Sigma (Madrid, Spain), and alcohols, ethanoate esters, and acetaldehyde diethyl acetal by Aldrich (Madrid, Spain). Standard solutions of major volatiles were prepared in ethanol/water (65:35) and filtered through a 0.45 μm poly(vinylidene difluoride) (PVDF) membrane. 3-Pentanol was used as internal standard at 0.5 g/L. After the addition of the internal standard, samples were filtered and directly injected into the GC system. Standard solutions of minor volatiles were prepared in ethanol/water (30:70). The standard and samples were subjected to microextraction with Freon 113 according to the method optimized by Ferreira et al. (18) and modified and validated by us for cider brandy analysis (12). Ten milliliters of cider brandy, previously diluted to 30% (v/v) in ethanol, was jointly introduced into the extraction centrifuge tubes with 150 μL of Freon 113, 2 g of ammonium sulfate, and 5 μL of a solution of 2-ethyl-1-hexanol (13 g/L in ethanol at 30%, v/v, internal standard). The tubes were shaken by means of a water shaking bath at 20 °C for 60 min and centrifuged (12000g, for 10 min). One microliter of the organic phase was recovered and injected into the chromatographic system. Characteristics of the samples analyzed are summarized in **Table 1**, according to the raw material, distillation technology, and wood type.

Chromatographic Equipment and Conditions. Major Volatiles. The following equipment and column were used for GC analysis of the major volatiles: a Hewlett-Packard 5890 gas chromatograph fitted with a flame ionization detector and a 5 m \times 1/8 in. stainless steel column packed with Carbowax 1,500 on Carbowax C 8/100 to 0.5%. Chromatographic conditions were as follows: initial temperature, 45 °C for 10 min; program rate, 3.5 °C/min; final temperature, 125 °C; isothermal to 125 °C for 25 min; injector and detector temperature, 170 °C; carrier gas, He at 60 psi; injection volume, 1 μL . All analyses were carried out in duplicate.

Minor Volatiles. The following equipment and column were used for GC analysis of the minor volatiles: a Hewlett-Packard 5890 gas chromatograph interfaced with a Hewlett-Packard 5972 mass selective detector (MSD), fitted with a column (FFAP 50 m \times 0.22 mm i.d.; phase thickness, 0.33 μm) and inserted into a split/splitless injector (splitless mode, 1 min). Chromatographic conditions were as follows: initial temperature, 40 °C for 5 min; program rate, 3.0 °C/min; final temperature, 220 °C; isothermal to 220 °C for 60 min; injector and detector temperature, 250 °C; carrier gas, He at 1 mL/min; injection volume, 1 μL . Analysis was performed in the electron impact (EI) mode, the ionization voltage was fixed at 70 eV, and the quantification was carried out in selected ion monitored (SIM) mode. All analyses were carried out in triplicate.

Statistical Analysis. Multivariate analysis was performed with the PARVUS statistical package (19). A data matrix was structured with 56 rows representing cider brandies and 27 columns corresponding to the concentration of chemical variables (major volatiles, acetaldehyde, methanol, ethyl ethanoate, 1-propanol, 2-methyl-1-propanol, 1-butanol, acetal, 2-methyl-1-butanol, 3-methyl-1-butanol; minor volatiles, ethyl 3-methylbutyrate, 3-methyl-1-butyl ethanoate, ethyl hexanoate, hexyl ethanoate, 1-hexanol, ethyl octanoate, 1-octanol, ethyl decanoate, diethyl

Table 1. Characteristics of Samples Included in the Database

sample	raw material ^a	distillation ^b	maturation		origin ^c
			in wood		
1	C	Charentais	no		Asturias
2	C	Charentais	yes		Asturias
3	C	Charentais	yes		Asturias
4	C	column	no		Asturias
5	C	column	yes		Asturias
6	C	column	yes		Asturias
7	C	column	yes		Asturias
8	C	column	yes		Asturias
9	C	column	yes		Asturias
10	C	column	yes		Asturias
11	C	column	yes		Asturias
12	C	column	yes		Asturias
13	C	column	yes		Asturias
14	C	Charentais	yes		Asturias
15	C	Charentais	yes		Asturias
16	C	Charentais	yes		Asturias
17	C	Charentais	yes		Asturias
18	C	Charentais	yes		Asturias
19	C	Charentais	yes		Asturias
20	C	column	yes		Asturias
21	C	column	yes		Asturias
22	C	column	yes		Asturias
23	C	column	yes		Asturias
24	C	column	yes		Asturias
25	C	column	yes		Asturias
26	C	Charentais	no		Asturias
27	F	Charentais	yes		Calvados
28	F	Charentais	yes		Calvados
29	F	Charentais	yes		Calvados
30	F	Charentais	yes		Calvados
31	F	Charentais	yes		Calvados
32	F	Charentais	yes		Calvados
33	F	Charentais	no		Asturias
34	F	column	yes		Asturias
35	F	column	yes		Asturias
36	F	column	yes		Asturias
37	F	Charentais	no		Asturias
38	F	column	no		Asturias
39	F	column	yes		Asturias
40	F	column	yes		Asturias
41	F	column	yes		Asturias
42	F	column	yes		Asturias
43	F	column	yes		Asturias
44	F	column	yes		Asturias
45	F	Charentais	yes		Herefordshire
46	F	alquitara	no		Asturias
47	F	alquitara	no		Asturias
48	F	Charentais	yes		Asturias
49	F	Charentais	yes		Calvados
50	F	Charentais	no		Asturias
51	F	alquitara	no		Asturias
52	F	unknown	yes		Asturias
53	F	Charentais	yes		Asturias
54	F	Charentais	yes		Asturias
55	F	Charentais	no		Asturias
56	F	Charentais	no		Asturias

^a C, spirit elaborated from cider obtained from apple juice concentrate; F, spirit elaborated from cider obtained from fresh apple juice. ^b Alquitara is a very old distillation system in which the condensation process is carried out in a small swan's neck cooled with water. ^c Asturias (Spain); Calvados (Normandy, France); Herefordshire (U.K.).

butanedioate, 2-phenylethyl ethanoate, ethyl dodecanoate, ethyl tetradecanoate, octanoic acid, ethyl hexadecanoate, decanoic acid, dodecanoic acid, ethyl 9,12-octadecadienoate, and tetradecanoic acid). Samples were included in the training set and were initially categorized as spirits made from cider elaborated from fresh apple juice (F, 30) and spirits elaborated from cider made from apple juice concentrate (C, 26). The chemical variables were autoscaled before statistical treatment. PCA was employed to visualize the data structure, and the LDA and Bayesian analysis were used for classification purposes. The descriptive statistics are shown in **Table 2**.

Table 2. Descriptive Statistics of Database by Categories (in Milligrams per 100 mL of Pure Ethanol)

compound	category C ^a (n = 26)				category F ^a (n = 30)			
	mean	max	min	SD	mean	max	min	SD
acetaldehyde	39.44	74.17	6.02	20.02	24.15	51.73	1.60	11.83
methanol	158.16	259.60	94.21	59.79	86.50	222.96	7.68	40.83
ethyl ethanoate	55.32	86.76	18.81	20.57	201.08	1288.93	9.55	239.21
1-propanol	19.96	37.11	16.38	4.34	54.67	285.15	12.75	54.70
2-methyl-1-propanol	34.47	54.92	20.24	13.17	44.79	100.65	14.45	24.77
1-butanol	1.90	9.68	0.45	2.05	12.31	21.15	0.83	3.83
acetal	27.30	58.52	4.67	15.78	12.94	29.94	1.28	8.49
2-methyl-1-butanol	23.43	36.39	17.67	4.15	39.69	72.45	6.67	13.95
3-methyl-1-butanol	116.42	183.71	92.89	24.82	169.97	295.00	29.84	69.71
ethyl 3-methylbutyrate	0.02	0.10	0.00	0.02	0.04	0.16	0.00	0.05
3-methyl-1-butyl ethanoate	1.13	2.81	0.06	0.70	0.60	1.15	0.00	0.31
ethyl hexanoate	0.76	1.01	0.03	0.22	0.35	0.72	0.09	0.18
hexyl ethanoate	0.02	0.18	0.00	0.04	0.05	0.15	0.00	0.05
1-hexanol	0.85	5.75	0.05	1.35	8.49	17.39	0.63	3.98
ethyl octanoate	2.03	3.93	0.19	1.31	0.94	2.14	0.02	0.68
1-octanol	0.03	0.07	0.02	0.01	0.22	1.14	0.02	0.27
ethyl decanoate	1.90	6.22	0.27	1.50	1.58	6.11	0.00	1.43
diethyl butanedioate	0.07	0.42	0.02	0.09	0.21	1.57	0.00	0.31
2-phenylethyl ethanoate	0.20	1.32	0.00	0.25	0.70	3.30	0.00	1.00
ethyl dodecanoate	0.58	3.19	0.06	0.77	0.65	2.99	0.00	0.67
ethyl tetradecanoate	0.29	2.39	0.02	0.50	0.27	1.12	0.00	0.26
octanoic acid	5.29	11.17	1.44	2.74	3.75	9.48	0.2	2.11
ethyl hexadecanoate	0.38	1.74	0.01	0.39	0.34	1.43	0.03	0.31
decanoic acid	4.60	11.10	1.35	2.59	4.68	12.63	0.32	2.71
dodecanoic acid	0.70	4.87	0.11	0.95	1.16	2.88	0.08	0.66
ethyl 9,12-octadecadienoate	0.69	2.64	0.05	0.77	0.64	3.18	0.00	0.98
tetradecanoic acid	0.33	1.41	0.06	0.35	0.48	1.22	0.04	0.31

^a C, spirit elaborated from cider obtained from apple juice concentrate; F, spirit elaborated from cider obtained from fresh apple juice; n, number of observations; max, maximum value; min, minimum value; SD, standard deviation.

RESULTS AND DISCUSSION

Univariate and Correlation Analysis. To classify cider brandies, a univariate analysis was carried out to ascertain whether any variable by itself would allow us to distinguish between fresh and concentrate cider brandies. To do so, we used the Fisher classification technique. Fisher weights (FW) were computed for classifying cider brandies, with 1-butanol (FW, 5.95), 1-hexanol (FW, 3.41), ethyl hexanoate (FW, 2.20), 2-methyl-1-butanol (FW, 1.29), and methanol (FW, 1.02) being found to be the variables with FW values > 1.00. However, the use of these variables did not allow us to correctly differentiate fresh and concentrate cider brandies, so a multivariate treatment was necessary. Likewise, before the multivariate techniques were applied, an analysis of correlation was carried out to detect the most correlated variables, using a confidence level of 90%. The highest correlations detected were the following: acetaldehyde/acetal (0.84), 1-butanol/1-hexanol (0.89), 2-methyl-1-butanol/3-methyl-1-butanol (0.90), ethyl dodecanoate/ethyl tetradecanoate (0.88), ethyl dodecanoate/dodecanoic acid (0.81), ethyl tetradecanoate/dodecanoic acid (0.87), and dodecanoic/tetradecanoic acids (0.90). Consequently, the following five variables were removed: acetal, ethyl tetradecanoate, dodecanoic acid, 1-hexanol, and 2-methyl-1-butanol. Thus, the new data matrix dimension was made up of 56 objects × 22 chemical variables.

PCA. Five significant factors, validated according to full double-cross-validation (five groups in cancellation matrix), which accounted for 76% of the variance, were chosen. The best visualization of the data structure is achieved by means of the first and third factors. **Figure 1** displays the projections of the original variables and cider brandies belonging to the training set on the factorial plane formed by these factors. As can be seen, projection of samples onto the factorial plane allows visualization of two groups of samples: distillates made from cider elaborated from fresh apple juice (F) and distillates

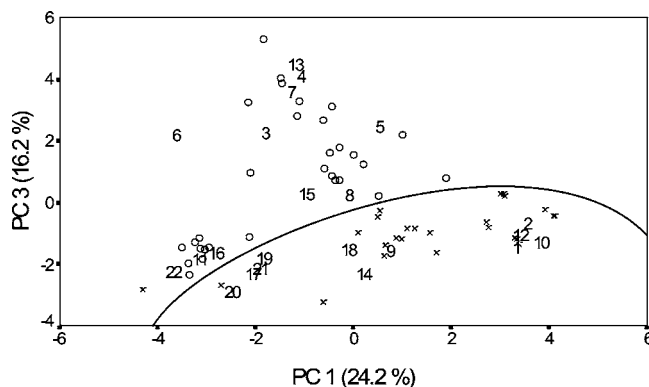


Figure 1. Projection of original variables and cider brandies on the factorial plane formed by the first and third significant factors: 1, acetaldehyde; 2, methanol; 3, ethyl ethanoate; 4, 1-propanol; 5, 2-methyl-1-propanol; 6, 1-butanol; 7, 3-methyl-1-butanol; 8, ethyl 3-methylbutyrate; 9, 3-methyl-1-butyl ethanoate; 10, ethyl hexanoate; 11, hexyl ethanoate; 12, ethyl octanoate; 13, 1-octanol; 14, ethyl decanoate; 15, diethyl butanedioate; 16, 2-phenylethyl ethanoate; 17, ethyl dodecanoate; 18, octanoic acid; 19, ethyl hexadecanoate; 20, decanoic acid; 21, ethyl 9,12-octadecadienoate; 22, tetradecanoic acid; (○) F category (fresh apple juice); (×) C category (apple juice concentrate).

elaborated from cider made from apple juice concentrate (C), with the majority of C samples placed in the lower right side of the factorial plane (see borderline in **Figure 1**). Acetaldehyde, methanol, ethyl esters of medium-chain fatty acids, and 3-methyl-1-butyl ethanoate were the chemical variables most strongly linked to the cider brandies made from apple juice concentrate, and 1-butanol, ethyl, hexyl and 2-phenylethyl ethanoates, 1-octanol, 1-propanol, 3-methyl-1-butanol, ethyl 3-methylbutyrate, and tetradecanoic acid were linked to cider brandies made from fresh apple juice. It is well-known that the apple juice

clarification and aroma recovery processes carried out during apple juice concentration produce an increase in the level of methanol (20) (an alcohol linked to pectic substances) and a decrease in the concentration of the varietal and prefermentative aromas such as 1-butanol and 1-hexanol. On the other hand, acetate esters, except 3-methyl-1-butyl ethanoate, were associated with cider brandies made from fresh apple juice, whereas ethyl esters of the middle aroma fraction were associated with cider brandies made from apple juice concentrate. This fact might be explained by taking into account the fact that fermentation of fresh apple juice is generally carried out in the presence of a complex group of yeasts that includes both non-*Saccharomyces* and *Saccharomyces* yeasts, whereas fermentation of apple juice concentrate is, generally, conducted by selected *Saccharomyces* yeasts. It is well-known that non-*Saccharomyces* yeasts produce acetate esters (21), particularly under aerobic prefermentative conditions, whereas *Saccharomyces* yeasts accumulate ethyl esters of fatty acids under anaerobic fermentation conditions.

LDA. The computed discriminant rule was validated by means of an internal cross-validation procedure, where objects are randomly divided into a selected number of groups (in this case four groups for cancellation) and each group is successively used as a prediction set for validating the discriminant rule. Classification hits were 100%, and prediction hits were >96%. Taking into account these results, we may consider the computed discriminant rule to be sufficiently feasible and robust for the purposes of classifying cider brandies according to their raw material (fresh apple juice versus apple juice concentrate). At the same time, a stepwise LDA, using Wilks' λ criterion, was carried out to identify the most discriminating variables. Values of *F*-to-enter and *F*-to-remove of 3.84 and 2.71 (confidence level of 90%), respectively, were used. The computed Wilks' λ was 0.048; thus, 95% of total variance is explained by within-group differences. The most important variables selected were 1-butanol and ethyl hexanoate, which is in accordance with the results obtained in PCA. As can be seen in **Figure 1**, these compounds are placed in the discriminant direction of the factorial plane that allows the differentiation of both categories.

Bayesian Analysis. The use of modeling techniques allows the computation of classification rules to classify unknown samples. The Bayesian technique constructs a hyperellipsoid for each category characterized by the points in space placed at a critical Mahalanobis distance, where a probability of 95% of the multivariate normal distribution is accumulated. The construction of robust models from Bayesian analysis requires a number of observations for each category that should be, at least, twice the number of variables. Bearing this in mind, we used the 5 principal components as variables instead of the 22 chemical variables. Bayesian models (fresh, F; concentrate, C) were computed from the new data matrix (56 observations \times 5 variables) for each category. The first and second class errors (α and β) related to model sensitivity ($1-\alpha$) and specificity ($1-\beta$) were estimated. As can be seen in **Figure 2**, where Coomans' diagram is displayed, the sensitivities of models, concentrate and fresh, were 92.3 and 90.0%, respectively; thus, fewer than 10% of samples belonging to each model are rejected from their own model. The specificity of both models was >96%; thus, fewer than 4% of samples are accepted by a model to which they do not belong. Correct classifications were >98%, whereas correct predictions, evaluated by cross-validation using three groups for cancellation, were 93%. We can therefore assume that the constructed models are sufficiently feasible and robust for classifying cider brandies according to the origin of raw

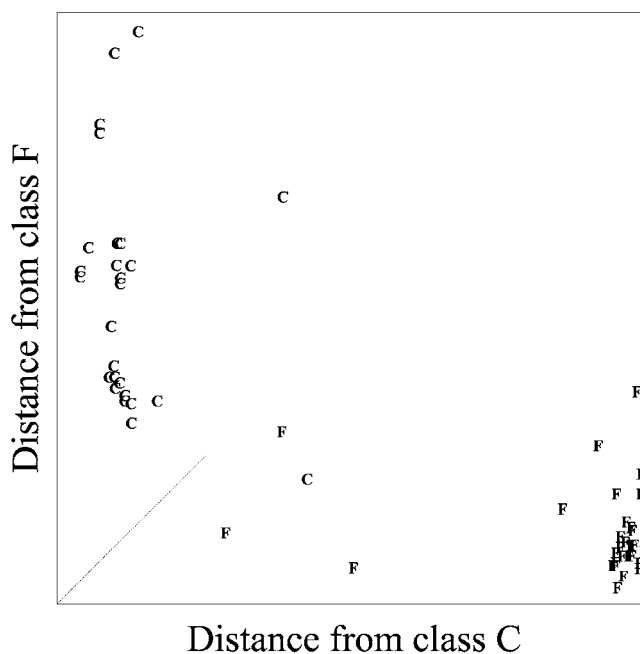


Figure 2. Coomans' diagram for the Bayesian model: F, distillates made from cider elaborated from fresh apple juice; C, distillates made from cider elaborated from apple juice concentrate.

material, although the classification and prediction hits obtained by Bayesian analysis were lower than those computed by LDA.

In conclusion, the use of the chemical composition of cider brandies together with chemometric techniques (PCA, LDA, and Bayesian analysis) allowed us to typify cider brandies on the basis of the cider used in their manufacture. The most discriminating variables were 1-butanol and ethyl hexanoate, which were associated with spirits made from fresh apple juice and apple juice concentrate, respectively. The LDA rule and Bayesian model computed allowed us to correctly classify >92% of cider brandies. We thus have an analytical tool for differentiating, within a reasonable degree of error (<8%), cider brandies on the basis of the raw material used in their manufacture that could be useful in detecting fraudulent practices.

LITERATURE CITED

- Drilleau, J. F. *Pommes et cidres matières premières du Calvados*. In *Élaboration et Connaissance des Spiritueux*; Cantagrel, R., Ed.; Lavoissier-Tec & Doc: Paris, France, 1992; pp 193–200.
- Steger, C. L. C.; Lambrechts, M. G. The selection of yeast strains for the production of premium quality South African brandy base products. *J. Ind. Microbiol. Biotechnol.* **2000**, *24*, 431–440.
- Mangas, J. J.; Rodríguez, R.; Moreno, J.; Blanco, D. Changes in the major volatile compounds of cider distillates during maturation. *Lebensm.-Wiss. Technol.* **1996**, *29*, 357–364.
- Rodríguez, R.; Blanco, D.; Mangas, J. J. Influence of distillation system, oak wood type, and aging time on volatile compounds of cider brandy. *J. Agric. Food Chem.* **2003**, *51*, 5709–5714.
- Guymon, J. F.; Crowell, E. A. GC-Separated brandy components derived from French and American oaks. *Am. J. Enol. Vitic.* **1972**, *23*, 114–120.
- Rodríguez, R.; Blanco, D.; Mangas, J. J. Influence of distillation system, oak wood type, and aging time on composition of cider brandy in phenolic and furanic compounds. *J. Agric. Food Chem.* **2003**, *51*, 7969–7973.
- Cardoso, D. R.; Lima-Neto, B. S.; Franco, D. W. Influencia do material do destilador na composição das aguardentes de cana. Parte II. *Quim. Nova* **2003**, *26*, 165–169.

- (8) Da Porto, C.; Longo, M.; Sensodini, A. Effects of low pressure and a rectification column on the volatile composition of fermented grape distillate. *Int. J. Food Sci. Technol.* **1996**, *31*, 403–410.
- (9) Mangas, J. J.; Rodríguez, R.; Moreno, J.; Suárez, B.; Blanco, D. Evolution of aromatic and furanic congeners in the maturation of cider brandy. A contribution to its characterization. *J. Agric. Food Chem.* **1996**, *44*, 3306–3307.
- (10) Blanco Gomis, D.; Muro Tamayo, D.; Mangas Alonso, J. J. Evolution of sugars in cider brandy aged in oak barrels: A contribution to its characterization. *J. Agric. Food Chem.* **2003**, *51*, 923–926.
- (11) Onishi, M.; Guymon, J. F.; Crowell, E. A. Changes in some volatile constituents of brandy during aging. *Am. J. Enol. Vitic.* **1977**, *28*, 152–158.
- (12) Mangas, J. J.; Rodríguez, R.; Moreno, J.; Blanco, D. Volatiles in distillates of cider aged in American oak wood. *J. Agric. Food Chem.* **1996**, *44*, 268–273.
- (13) Lisle, D. B.; Richards, C. P.; Wardleworth, D. F. The identification of distilled alcoholic beverages. *J. Inst. Brew.* **1978**, *84*, 93–96.
- (14) Miyashita, Y.; Ishikawa, M.; Sasaki, S. Classification of brandies by pattern recognition of chemical data. *J. Sci. Food Agric.* **1989**, *49*, 325–333.
- (15) Mazerolles, G.; Vidal, J. P.; Cantagrel, R.; Gaschet, J.; Faure, A.; Lambert, F. Application des méthodes d'analyse de données à la caractérisation analytique des cognacs. In *Élaboration et Connaissance des Spiritueux*; Cantagrel, R., Ed.; Lavoissier-Tec & Doc: Paris, France, 1992; pp 410–416.
- (16) Versini, G.; Monetti, A.; Dalla Serra, A.; Inama, S. Analytical and statistical characterization of grappa from different Italian regions. Presented at the First Symposium International sur les Eaux-de-vie Traditionnelles d'Origen Viticole, Bourdeaux, France, 1990; pp 137–150.
- (17) Rodríguez, R.; Blanco, D.; Mangas, J. J. Characterization of cider brandy on the basis of aging time. *J. Food Sci.* **2003**, *68*, 1958–1961.
- (18) Ferreira, V.; Rapp, A.; Cacho, J. F.; Hastrich, H.; Yavas, I. Fast and quantitative determination of wine flavor compounds using microextraction with Freon 113. *J. Agric. Food Chem.* **1993**, *41*, 1413–1420.
- (19) Forina, M.; Leardi, R.; Armanino, C.; Lanteri, S. In *PARVUS. An Extendable Package of Programs for Data Exploration, Classification and Correlation*; Elsevier: Amsterdam, The Netherlands, 1988.
- (20) Kilara, A.; Van Buren, J. P. Clarification of apple juice. In *Processed Apple Products*; Downing, D. L., Ed.; Van Nostrand Reinhold: New York, 1989; pp 83–96.
- (21) Cabranes, C.; Mangas, J. J. Selection and biochemical characterization of *Saccharomyces cerevisiae* and *Kloeckera apiculata* strains isolated from Spanish cider. *J. Inst. Brew.* **1997**, *103*, 165–169.

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