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Influence of Distillation System, Oak Wood Type, and Aging Time on Composition of Cider Brandy in Phenolic and Furanic Compounds

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A control of phenolic and furanic compounds in cider brandy was carried out during maturation in oak casks, studying three technological factors: distillation (rectification column vs double distillation), oak wood type (French vs American), and aging time (32 months). Gallic acid and benzoic and cinnamic aldehydes significantly increased during maturation of cider brandies, the highest level of these phenolics being obtained when aging was conducted in French oak casks. Benzoic acids increased during aging, though furanic compounds were not influenced by the time factor. Distillation and wood factors significantly influenced furanic concentration; 5-hydroxymethylfurfural not was detected in fresh spirits and was extracted in the highest proportion in French oak. Volatile furanics, such as 5-methylfurfural, furfural, and 2-furylmethyl ketone, were influenced by the distillation factor, with the use of the double distillation system producing a higher level of these compounds. Scopoletin was the majority coumarin detected in cider brandies, the highest yield of which was obtained with the use of American oak.

KEYWORDS: Phenolic; furanic; distillation; oak; aging; cider brandy

INTRODUCTION

Cider brandy is a popular alcoholic beverage elaborated in Asturias (North of Spain) by distilling cider in pot stills or rectifying stills and maturing the fresh distillate in oak wood barrels for several years. This spirit is included in the Council Regulation (CEE) 1576/89, related to the definition, description, and presentation of spirit drinks.

A combination of chemical interaction and extraction takes place between the spirit and wood during the aging process that markedly changes the taste and color of cider brandy due to the presence of a large number of phenolic and furanic compounds incorporated from the wood. Distillates are usually matured in European or American oak casks; the components of the most widely employed species (*Quercus robur*, *Quercus petraea*, and *Quercus alba*) are similar, differing basically in the concentration of extracted substances.

Oak wood is composed of certain polymers that play an important role in the aging phenomena, namely cellulose, hemicelluloses, lignin, and tannins. Acid ethanolysis of lignin produces aromatic aldehydes, and the hydrolysis in acid medium of tannins and hemicelluloses produces phenolic acids and monosaccharides, respectively. Other pigments detected in aged spirits, such as flavonols, are directly extracted from the wood. At the same time, the action of oxidant agents (oxygen and peroxides) on analytes extracted from oak wood allows the synthesis of new aromatic compounds such as quinones, aldehydes, methyl ketones, etc.

Tannins are responsible for astringency, or a bitter or harsh character (*1*). Ellagitannins are quickly solubilized during the first year of aging and are simultaneously degraded into ellagic acid (*2*). Moreover, the ellagitannin content of the stave wood used in cooperage is strongly influenced by the position of the stave wood in the tree (*3*) and the species employed (*4*). Another important factor influencing the tannin level in wood is the drying stage; the content of these substances decreases during natural seasoning (*5*), but is not so pronounced in kiln-drying (*6*).

Aldehydes and aromatic acids are an important group of substances found in spirits matured in oak (*7*−*15*) and chestnut (*16*) wood. They derive from lignin through hydro alcoholysis (*17*, *18*). Levels of benzoic and cinnamic acids and aldehydes increase with seasoning and toasting (*5*, *19*, *20*) and are influenced by geographical origin, thus enabling the classification of American and French oaks (*21*).

Charring and toasting also destroy carbohydrates. As a consequence, furanic compounds such as furfural, 5-methylfurfural, and 5-hydroxymethylfurfural are generated in staves

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and extracted into the distillate. Cutzach et al. (*22*, *23*) identified several new molecules of furanic compounds and pyranones in toasted wood with the characteristic "toasty-caramel" aroma. Quesada et al. (*24*) established that the concentration of furanic aldehydes is related to the age of the cask, the charring process, the wood type and the presence or absence of caramel. Thus, these substances cannot be employed as aging markers. On the other hand, the most volatile furanics may be incorporated into fresh spirits during distillation (*25*) by degradation of residual sugars present in the fermented raw material.

Likewise, a coumarine, scopoletin, has been proposed as a signal of degree of aging (*26*−*28*). Recently, however, Fernández Izquierdo et al. (*29*) found scopoletin and other coumarins in hydro alcoholic extracts and caramel coloring employed in the industry, which means that coumarins cannot be used as aging markers. Gómez-Cordovés (*30*) suggested a new compound, 2,3-dihydroxy-1-guaiacylpropan-1-one, related to the genuine aging of alcoholic beverages in oak.

The aim of this work was to study the influence of the distillation system and oak type on the concentration of phenolic and furanic compounds in cider brandy during the maturation process.

MATERIALS AND METHODS

Reagents. All standards were of analytical quality. Gallic acid, vanillic acid, and scopoletin were supplied by Sigma (Madrid, Spain); syringic acid, syringaldehyde, ferulic acid, coniferaldehyde, and sinapaldehyde by Aldrich (Madrid, Spain); vanillin by Merck (Darmstadt, Germany), and furanic compounds by Fluka (Busch, Switzerland).

Standards. Standards were prepared in a solution of 2% acetic acid and 0.02 M sodium acetate (pH 3.2), and samples were previously subjected to an ethanol removal process under vacuum at 30−35 °C (conc 2:1), and then filtered through a 0.45-μm PVDF (poly(vinylidene difluoride)) membrane.

HPLC Equipment and Conditions. Experimental data were obtained from a high-performance liquid chromatography (HPLC) system (Waters Associates) equipped with a 712 automatic injector, two M510 pumps, a Millenium v. 2.0 software data module, and a 996 photodiode array detector. Separation of analytes was carried out on a Spherisorb ODS-2 (250 × 4.6 mm, 3 μm) at 40 °C using 2% acetic acid and 0.02 M sodium acetate (pH, 3.2; solvent A) and methanol (solvent B) as the mobile phase. Elution conditions were as follows: starting, 97% A; isocratic for 4 min; linear increase of solvent B in solvent A to 30% solvent B for 21 min; isocratic for 25 min; flow rate was 1 mL/min; and injection volume, 10 μL. Gallic acid, vanillic acid, syringic acid, furfural, 5-hydroxymethylfurfural, 5-methylfurfural, 2-furylmethyl ketone, vanillin, and syringaldehyde were detected at 280 nm, while scopoletin, ferulic acid, coniferaldehyde, and sinapaldehyde were detected at 320 nm. This analytical method was optimized and described elsewhere (*15*), the recovery levels for the analytes studied ranged between 88 and 109%, and the relative standard deviations were less than 5%.

Raw Material. Cider (20 000 L) was made from apple juice concentrate (AJC). The AJC was diluted with water (density of reconstituted apple juice, 1056 g/L), and fermented by a starter of *Saccharomyces cerevisiae* belonging to the SERIDA microorganism collection. Chemical composition of cider is shown in **Table 1**. The cider was distilled by two methods: double distillation and rectification column. In the double distillation system, a first distillation of cider is made to obtain three fractions, namely heads, hearts, and tails. Heads and tails are redistilled in the next batch of cider. In the second distillation, the hearts (28−30% alcoholic strength) are distilled, obtaining again heads, hearts (fresh cider brandy, 69.8% alcoholic strength), and tails. For distillation with the rectification column system, we have used a still with a sixteen-plate-rectification column. In this distillation technology, cut points were as follows: heads (0.5% of the distilled cider volume), hearts (6.5% of the distilled cider volume; alcoholic strength of fresh cider brandy, 67.3%), and tails (1.5% of the distilled cider volume). Fresh distillates were matured in wood casks of French (*Quercus sessilis*) (three barrels for each fresh distillate; total, six distillates matured in French wood) and American oak (*Quercus alba*) (three barrels for each fresh distillate; total, six distillates matured in American wood) for 32 months. American oak came from Ohio, and French oak came from the Allier forest. The wood, once brushed and cut, was subjected to a light toasting (20 min at 180 °C), using gas as energy source. The thickness of the staves and cask capacities were 28 mm and 120 L, respectively.

Table 1. Chemical Composition of Cider

parameter	conc
total acidity[a]	4.7
volatile acidity[b]	0.2
alcoholic strength[c]	7.2
acetate esters[d]	27.5
higher alcohol[d]	146.8
acetaldehyde[d]	37.8
methanol[d]	147.8

[a] In g/L sulfuric acid. [b] In g/L acetic acid. [c] In %, v/v. [d] In mg/L.

Experimental Design. A factorial design (2 × 2) with three replicates was used (*31*). The factors or independent variables studied were the following: distillation system (two levels, double distillation and rectification column), wood (two levels, French oak and American oak), and aging time (21 samplings for 32 months). Response variables were as follows: gallic acid, 5-hydroxymethylfurfural, furfural, 5-methylfurfural, 2-furylmethyl ketone, vanillic acid, syringic acid, vanillin, syringaldehyde, ferulic acid, coniferaldehyde, sinapaldehyde, and scopoletin. Statistical treatment (*32*) consisted of a two-factor ANOVA with two levels. ANOVA for the time factor was carried out for each combination (distillation × wood). Duncan's multiple range test was carried out for pairwise comparisons among means. Differences were considered significant at 5%. Results of ANOVA are displayed in **Table 2**.

RESULTS AND DISCUSSION

Alcoholic strength of fresh distillates obtained from the two distillation technologies studied were very similar (69.8%, double distillation vs 67.3%, rectification column). However, fresh distillate produced from rectification column was more acidic (pH: 2.8) than fresh distillate elaborated by double distillation system (pH 3.5), which can influence the equilibria of the esterification and transesterification processes (data not shown).

Gallic acid is a phenolic compound derived from the hydrolysis of hydrolyzable tannins in oak wood. Aging time and wood factors (**Table 2**) had a significant effect on the concentration of this phenolic acid. As can be seen in **Figure 1**, gallic acid increased during aging, a greater amount of this phenolic being extracted when French oak wood was employed. This result was in agreement with works previously reported. Thus, Puech (*18*) showed the low tannin content of American oak in relation to French oak, and Miller et al. (*33*) detected higher gallic content in *Q. robur* than in *Q. alba*. The same profile was detected, in the case of benzoic aldehydes (vanillin and syringaldehyde) (**Figures 2** and **3**), which derive from hydroalcoholysis of lignin, according to the mechanism proposed by Puech (*18*). However, cinnamic aldehydes (coniferaldehyde and sinapaldehyde) did not present a significant evolution with the time factor (**Figures 4** and **5**). This might be explained by bearing in mind the fact that these aldehydes directly derive from lignin transformation and are precursors of other phenolics such as cinnamic acids and benzoic aldehydes and acids. Likewise, a significant effect of oak wood type (**Table 2**) was detected for cinnamic aldehydes (**Figures 4** and **5**). Once again,

Table 2. ANOVA Results[a]

	error source		distillation			wood			distillation × wood		
compound	SS	MS	SS	*F*	*p*	SS	F	*p*	SS	*F*	*p*
gallic acid	6.56	0.82	0.59	0.72	0.421	6.04	7.37	0.026	0.10	0.12	0.735
5-hydroxymethylfurfural	0.42	0.05	0.13	2.39	0.160	1.33	25.36	0.001	0.05	0.87	0.379
furfural	7.27	0.91	53085	58442	<0.001	13.58	14.95	0.005	0.74	0.82	0.393
5-methylfurfural	0.15	0.02	4.93	268.84	<0.001	0.15	7.94	0.023	0.01	0.32	0.586
2-furylmethyl ketone	<0.01	<0.01	0.64	7476.1	<0.001	<0.01	1.09	0.327	<0.01	1.09	0.327
vanillic acid	0.04	<0.01	0.03	5.46	0.048	0.01	1.66	0.234	<0.01	0.60	0.462
syringic acid	0.12	0.02	0.04	2.92	0.126	0.05	3.53	0.097	0.01	0.69	0.431
vanillin	0.44	0.05	0.18	3.33	0.106	0.70	12.9	0.007	0.04	0.67	0.437
syringaldehyde	3.30	0.41	1.22	2.96	0.124	3.00	7.28	0.027	0.20	0.49	0.505
ferulic acid	<0.01	<0.01	<0.01	0.06	0.815	<0.01	0.06	0.815	<0.01	1.62	0.239
scopoletin	<0.01	<0.01	<0.01	0.39	0.549	0.47	1114.0	<0.001	<0.01	4.04	0.080
coniferaldehyde	649.36	81.17	208.28	2.57	0.148	1105.5	13.62	0.006	55.20	0.68	0.433
sinapaldehyde	26.74	3.34	8.29	2.48	0.154	43.11	12.9	0.007	2.97	0.89	0.373

[a] SS, sum of squares; MS, mean square; *F*, *F* statistic; *p*, probability; degrees of freedom for error = 8; degrees of freedom for each factor = 1.

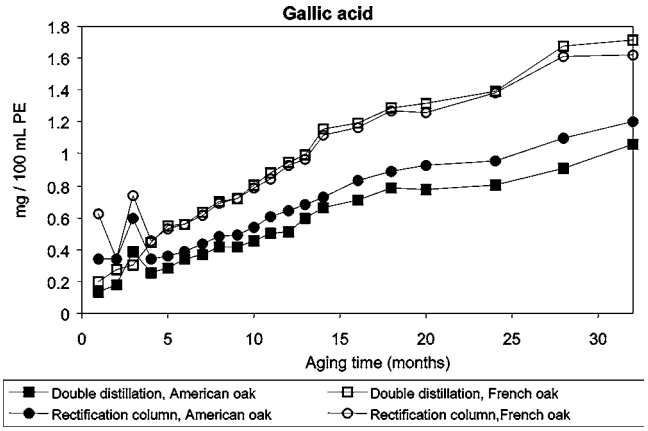

Figure 1. Changes in gallic acid content during aging. P. E., Pure ethanol.

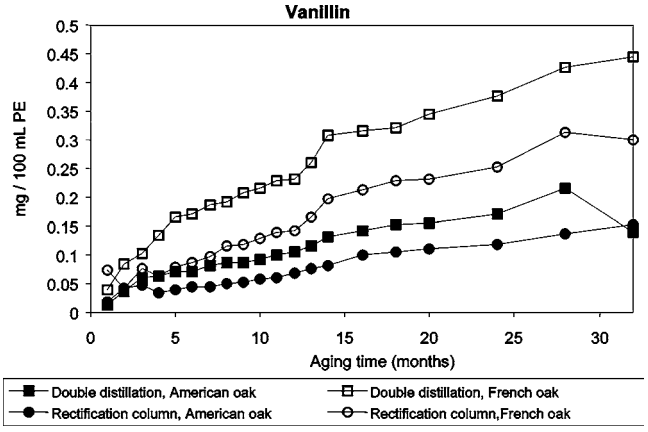

Figure 2. Changes in vanillin content during aging. P. E., Pure ethanol.

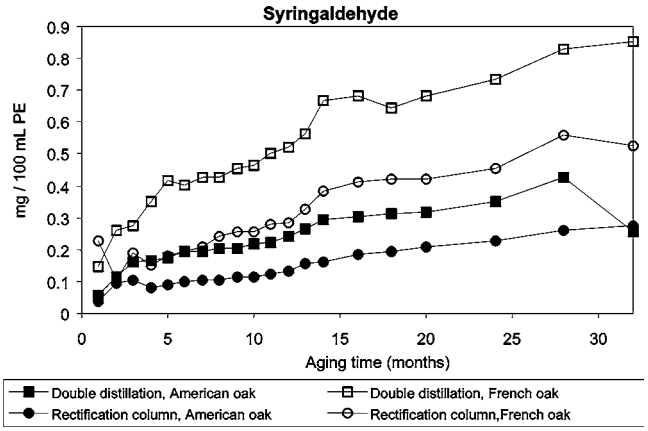

Figure 3. Changes in syringaldehyde content during aging. P. E., Pure ethanol.

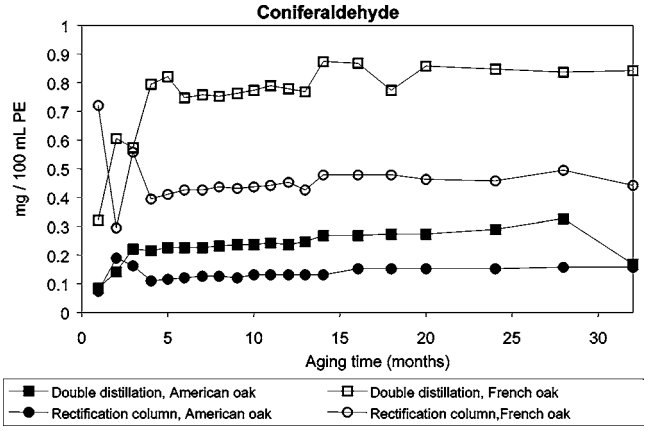

Figure 4. Changes in coniferaldehyde content during aging. P. E., Pure ethanol.

spirits aged in French oak presented a higher concentration of cinnamic aldehydes than those aged in American oak.

Benzoic acids (vanillic and syringic) are extracted from the cask wood or synthesized in the spirit from benzoic aldehydes. Their concentration increased throughout the maturation process (**Figures 6** and **7**), the highest concentration of these acids being detected in the spirits elaborated from a double distillation system. It should be noted that no significant effect of the factors studied was detected with respect to ferulic acid (data not shown).

The furanic compounds controlled during the aging process (furfural, 5-hydroxymethylfurfural, 5-methylfurfural, and 2-furylmethyl ketone) were not influenced by aging time ($p > 0.05$). These results show that furanic compounds may not be used as aging markers in accordance with the results obtained by Quesada-Granados et al. (*24*). However, other factors studied, such as the distillation technology and wood type, influenced the concentration of these aromas. 5-Hydroxymethylfurfural was not detected in fresh spirits, as a consequence of its high boiling point. This furanic was extracted in a higher proportion when the maturation process was conducted in French oak wood (**Tables 2** and **3**). The same effect due to the wood factor was also observed in the case of furfural and 5-methylfurfural (**Tables 2** and **3**). The distillation technology influenced the furanic compounds with lower boiling points, furfural, 5-methylfurfural, and 2-furylmethyl ketone (**Table 2**). When the

Table 3. Average Furanic Compounds Concentration (mg/L Pure Ethanol)[a]

	furanics			
factor combination (distillation × wood)	furfural	5-hidroxymethylfurfural	2-furylmethyl ketone	5-methylfurfural
double distillation × American oak	33.52	0.05	0.10	0.30
double distillation × French oak	34.11	0.23	0.10	0.36
column system × American oak	3.89	0.03	n.d.	0.02
column system × French oak	4.25	0.15	n.d.	0.06

[a] n.d., not detected.

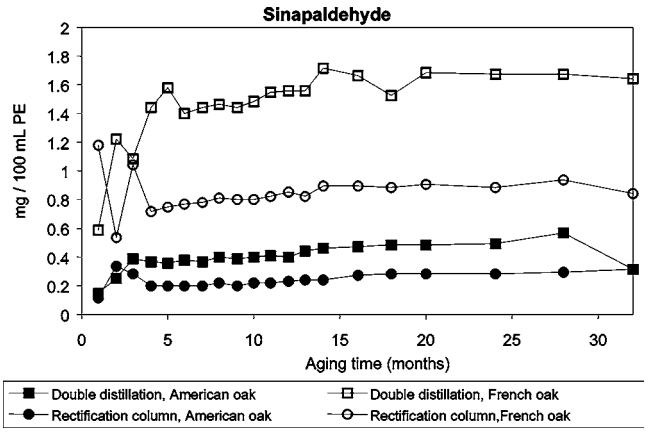

Figure 5. Changes in sinapaldehyde content during aging. P. E., Pure ethanol.

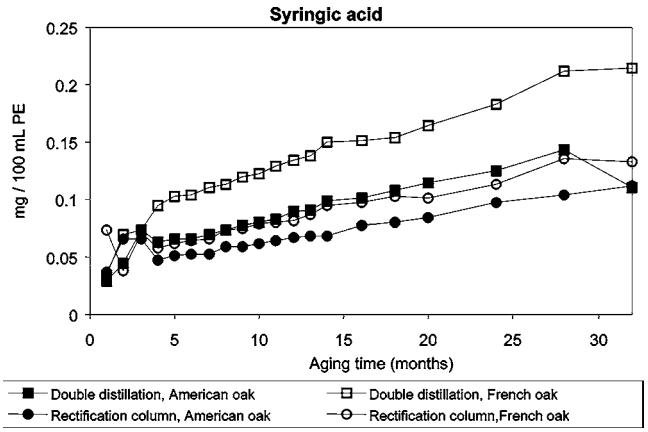

Figure 7. Changes in syringic acid content during aging. P. E., Pure ethanol.

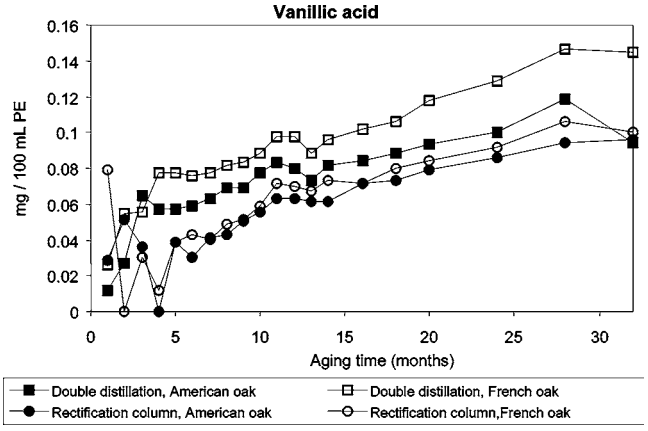

Figure 6. Changes in vanillic acid content during aging. P. E., Pure ethanol.

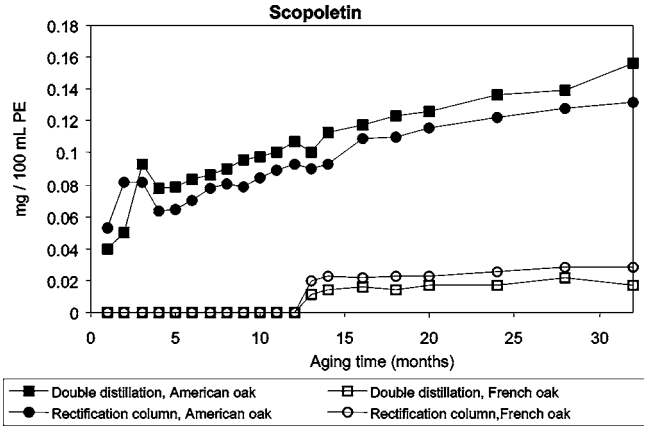

Figure 8. Changes in scopoletin content during aging. P. E., Pure ethanol.

double distillation system was used, a higher proportion of these analytes was obtained (**Table 3**). As is well known, the distillation time in the Charente technology (double distillation) is more extended, which facilitates the synthesis of the furanic compounds from carbohydrate degradation. In general, the detected concentration of these aromas in the spirits distilled by means of the rectification column system was low, and 2-furylmethyl ketone was not detected in the distillates obtained from this distillation technology (**Table 3**). In any case, it is worth noting that the high concentration of furfural found in the samples analyzed could be due to the use of cider elaborated from apple juice concentrate, containing high levels of furfural as a consequence of the heating process involved during the concentrate making (data not shown).

Coumarins are lactones derived from *o*-hydroxycinnamic acids by cyclizing between *o*-hydroxy and carboxy chemical groups. Scopoletin was the major coumarin detected in cider brandies; cooperage practices and oak wood type can modify the quantity extracted from the casks during the aging process. In fact, it has been reported that American oaks yield higher quantities of scopoletin than Spanish and French oaks (*34*). These results have been confirmed in our case, because, as can be seen in **Figure 8**, maturation in American oak barrels produced a higher concentration of scopoletin than when the aging process was conducted in French oak casks, with an increase in its concentration being observed throughout the maturation process (**Figure 8**). As can be seen in **Figure 8**, the effect of the wood factor on the scopoletine level was more relevant than that of the aging process. Thus, this phenolic should not be used as aging marker as was already suggested by Fernández-Izquierdo et al. (*29*).

In conclusion, French oak provides higher quantities of gallic acid, benzoic and cinnamic aldehydes, 5-hydroxymethylfurfural, furfural, and 5-methylfurfural than American oak, which means that distillates aged in French oak may have a greater aromatic complexity. Phenolic compounds increase during aging, except for cinnamic aldehydes, while furanic compounds are not influenced by the maturation process in wood. Thus, these analytes cannot be used as maturation markers of cider brandies.

Volatile furanics, such as furfural, 5-methylfurfural, and 2-furylmethyl ketone, are recovered in a higher proportion with the use of the double distillation system, as a consequence of the long distillation time employed in this distillation technology. Scopoletin is extracted in a higher proportion in distillates aged in American oak barrels. This analyte could thus be used for distinguishing cider brandies on the basis of the wood type employed in their maturation process.

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