

Monitoring Toasting Intensity of Barrels by Chromatographic Analysis of Volatile Compounds from Toasted Oak Wood

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Toasting changes both the quantity and the quality of the extractable substances in the oak wood of barrels used for the aging of fine wines and spirits. Mastery and repeatability of toasting are vital in the production of quality barrels to be used for the aging of wines and spirits. In this study, we show that it is possible for a given cooperage to differentiate barrel toasting levels by analyzing a certain number of volatile and semivolatile compounds resulting from the thermal degradation of oak. Toasted wood components, which can be extracted by the wines or spirits during the aging process, are normally analyzed after the wood has soaked in standard alcoholic solutions and can be used to control the intensity of the wood toasting. The results of the comparative analysis presented in this work show that headspace analysis with a microextractive method using a stationary polydimethylsiloxane type phase is a promising technique for analyzing toasted oak wood from barrels. It is easier to use than the traditional maceration and extraction method and provides similar information.

Keywords: Oak wood; toasting; volatile compounds; SPME

INTRODUCTION

Several studies have described changes in the composition of oak as it is toasted over a wood fire (Singleton, 1974; Marche et al., 1975; Ribereau-Gayon et al., 1976; Nishimura et al., 1983; Dubois, 1989; Nomdedeu, 1987; Chatonnet et al., 1989; Puech and Visockis, 1986; Puech, 1987; Chatonnet, 1995; Cutzach et al., 1997) or by means of infrared rays (Chatonnet et al., 1993). Research devoted to determining the intensity of toasting a posteriori is, however, by no means as common and has rarely reached any firm conclusions. As shown by a general model, changes produced by heating oak increase the concentrations of certain compounds up to a certain level of toasting. If toasting continues beyond this point, concentrations tend to decrease. It is thus difficult to determine the exact toasting level exclusively by chemical analysis. Furthermore, the toasting level corresponding to the maximum chemical concentration level depends on the chemical nature of the compounds (Chatonnet et al., 1989). Phenyl ketones, resulting from heat breakdown of lignans, are the only compounds that increase steadily with the toasting level. However, these compounds appear in significant quantities only above 200 °C, higher than the average toasting temperature. It is thus impossible to measure toasting temperatures of <200 °C with any accuracy. As has already been pointed out, each cooperage has its own toasting methods (Chatonnet and Boidron, 1989; Chatonnet et al., 1993). Wood composition can thus vary greatly despite toasting temperatures that are theoretically the same.

In this study, we will show that it is possible for a given cooperage to differentiate barrel toasting levels by analyzing a certain number of volatile and semivolatile compounds resulting from the thermal degradation of oak. We will also show that a microextractive method with a stationary polydimethylsiloxane type phase (HSSPME; Belardi and Pawliszyn, 1989; Arthur and Pawliszyn, 1990) is a promising technique for analyzing the headspace of a sample taken from a toasted barrel. It is easier to use than the traditional maceration and extraction method and provides similar information.

MATERIALS AND METHODS

Toasted Oak Samples. The oak used for this experiment came from French forests of the center of France (haute futaille), had the thin and regular grain typical of sessile oak (*Quercus petraea*), and had been air-seasoned during 24 months. The toasted oak samples were taken from the inner sides of barrels (stave of 27 mm thickness) toasted over a wood fire. Wood shavings (0–6 mm) were from four barrels toasted at three levels [light (L) surface temperature of the inner face of the staves = 115–125 °C; medium (M) = 200–215 °C; heavy (F) = 220–230 °C] according to standards defined by the coopers on 10 staves (over a total of ~30) for each barrel. Heat intensity corresponded to the length of time the barrels spent over a variety of braziers. Intensity levels are listed as described by shops with different working methods (Seguin-Moreau Cooperage, Merpins, France).

Preparation and Extraction of Samples for Analysis. *Extracting Compounds in a Hydroalcoholic Model Solution.* Twenty grams of wood shavings was extracted after soaking for 2 weeks in a liter of dilute alcohol solution (12% ethanol by volume, 5 g/L tartaric acid, NaOH, N qsp pH 3.5). The mixture was shaken daily and then filtered through glass wool prior to analysis.

Extraction of Volatile Compounds by Headspace Solid-Phase Microextraction (HSSPME). One gram of dry wood (12–14%

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Table 1. Analysis of Compounds in Toasted Wood Extractable by Maceration in a Standard Dilute Alcohol Solution (20 g/L Wood, 2 Weeks of Maceration, Assay after Extraction by Solvent)

compound ^a	$\mu\text{g/g}$ of dried wood					
	light toast (L)		medium toast (M)		high toast (F)	
	av	SD	av	SD	av	SD
volatile phenols						
guaiacol	0.45	0.324	1.26	0.170	0.40	0.238
4-methylguaiacol	0.80	0.223	1.72	0.401	0.74	0.150
phenol	0.47	0.482	0.83	0.139	0.41	0.416
4-ethylguaiacol	0.04	0.022	0.14	0.032	0.04	0.015
eugenol	1.87	1.577	1.27	0.744	1.65	1.234
4-ethylphenol	0.30	0.239	0.29	0.039	0.26	0.188
4-vinylguaiacol	0.13	0.055	0.17	0.113	0.12	0.044
isoeugenol	0.29	0.466	0.89	0.388	0.24	0.371
syringol	0.94	0.692	3.66	0.382	0.83	0.508
4-methylsyringol	0.94	0.419	4.07	0.726	0.85	0.283
4-allylsyringol	1.13	0.932	3.88	0.558	0.10	0.700
4-ethylvanillate	0.12	0.112	0.07	0.023	0.10	0.088
vanillin	27.17	4.993	49.81	23.573	25.45	3.065
syringaldehyde	162.08	49.175	524.61	141.201	149.82	28.819
furanones—pyranones						
cycloten	0.27	0.190	1.82	0.372	1.69	0.711
dihydromaltol*	0.00	0.000	1.84	0.939	1.16	0.779
<i>trans</i> -methyloctalactone	1.61	2.310	5.24	8.938	5.86	5.756
<i>cis</i> -methyloctalactone	0.49	0.491	0.45	0.742	0.83	1.304
maltol	1.61	0.472	9.75	1.103	6.93	2.121
furaneol	0.29	0.186	1.33	0.082	1.21	0.714
dihydrodihydroxydimethylpyranone	0.00	0.000	0.01	0.007	0.01	0.011
isomaltol*	2815.09	2926.251	866.50	1335.187	3947.05	4479.461
hydroxymaltol*	20.24	26.286	219.93	380.72	141.59	147.390
furfural	336.05	119.191	795.90	131.499	899.98	89.662
5-methylfurfural	44.68	17.558	112.30	72.518	145.53	70.145
5-hydroxymethylfurfural	1212.93	241.121	5077.42	862.511	5242.15	1891.670

^a The asterisk indicates the compounds are given in maltol equivalents.

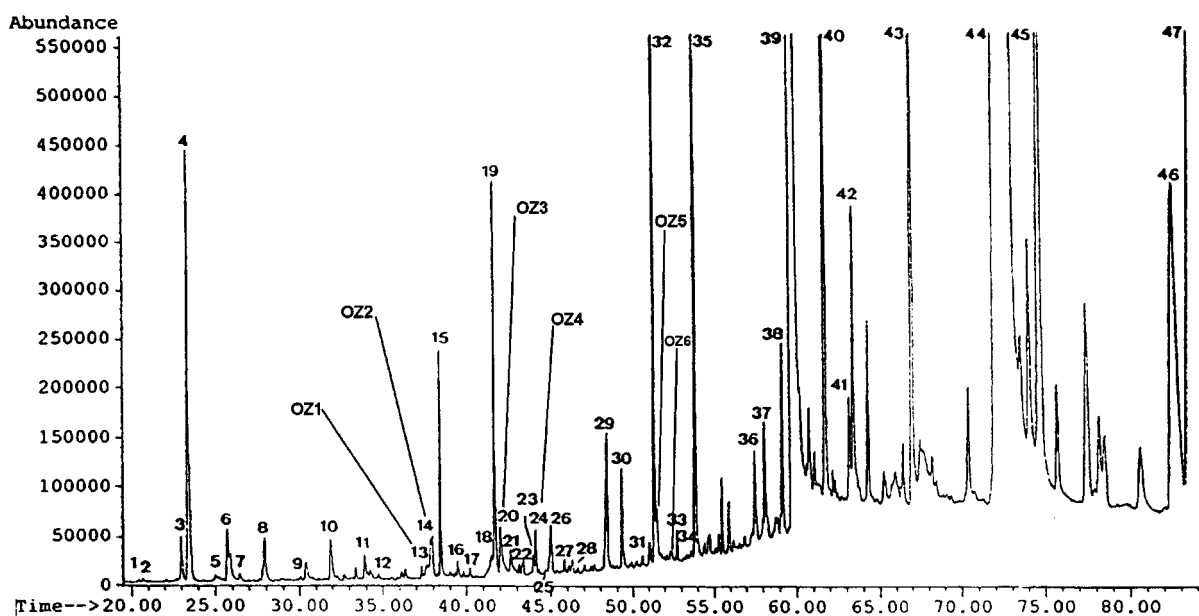


Figure 1. Chromatogram of volatile compounds in toasted wood extractable by soaking (15 days) in a model hydroalcoholic solution (12% volume, pH 3.5) analyzed by GC/MS (scan mode detection): 1, 2,5-dimethylpyrazine; 2, 2,6-dimethylpyrazine; 3, acetic acid; 4, furfural; 5, furanyl-1-ethanone; 6, benzaldehyde; 7, propionic acid; 8, 5-methylfurfural; 9, butyrolactone; 10, hydroxybenzaldehyde; 11, 3,4-dimethylfuranone-2(5*H*); 12, 2(3*H*)-furanone; 13, cycloten; 14, hexanoic acid; 15, guaiacol; 16, *trans*-methyloctalactone; 17, 2-phenylethanol; 18, benzothiazole; 19, *cis*-methyloctalactone + 4-methylguaiacol; 20, maltol; 21, 2,5-diformylfuran; 22, *o*-cresol; 23, phenol; 24, 4-ethylguaiacol; 25, 1*H*-pyrrolcarboxaldehyde; 26, octanoic acid; 27, *p*-cresol; 28, *m*-cresol; 29, eugenol; 30, isomaltol; 31, 4-vinylguaiacol; 32, syringol; 33, decanoic acid; 34, isoeugenol; 35, 4-methylsyringol; 36, dodecanoic acid; 37, 5-hydroxymethylfurfural; 38, 4-allylsyringol; 39, vanillin; 40, acetovanillon; 41, tetradecanoic acid; 42, propiovanillon; 43, butyrovaniillon; 45, acetosyringol; 46, propiosyringol; 47, coniferaldehyde.

relative humidity), ground for 2 min in a rotary grinder, was placed in a 30 mL glass vial with an elastomer stopper having a Teflon-coated bottom. An aluminum capsule was crimped on to ensure that the beaker was airtight. The vial was heated for 30 min at 80 °C in a ventilated oven to accelerate the

volatilization of the wood's volatile chemicals in the gas phase. The headspace was then sampled by inserting a fiber (SPME, Supelco) used for solid-phase microextraction (Arthur and Pawliszyn, 1990; Chai and Pawliszyn, 1995) into the headspace in the vial at room temperature (22 ± 2 °C). Preliminary tests

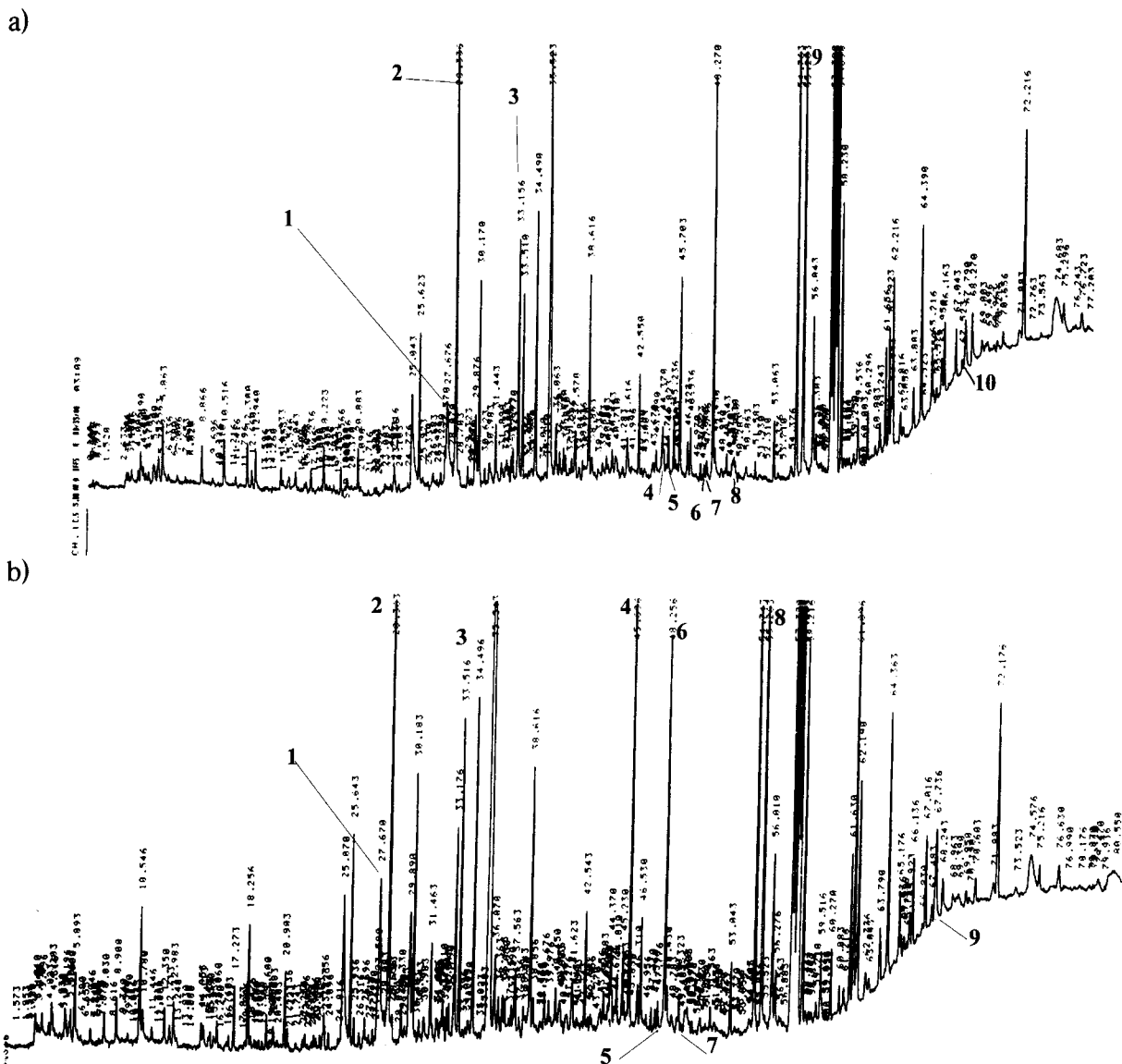


Figure 2. HSPME chromatogram of medium (a) or heavily (b) toasted oak wood using flame ionization (FID): 1, acetic acid; 2, 2-furfural; 3, 5-methyl-2-furfural; 4, guaiacol; 5, *trans*-methyloctalactone; 6, 4-methylguaiacol; 7, *cis*-methyloctalactone; 8, phenol; 9, eugenol + X; 10, vanillin.

Table 2. HSPME Analysis of Toasted Wood at Different Toasting Intensities (Area of Chromatographic Peaks in Arbitrary Units, Average of Three Analyses)

compound	light toast (L)		medium toast (M)		high toast (F)	
	av	SD	av	SD	av	SD
furfural	3342.0	1065.8	10434.5	1116.4	6333.5	1678.0
5-methylfurfural	1298.0	557.0	3360.0	394.5	1797.3	819.4
guaiacol	1861.3	810.8	3470.8	1272.8	772.3	187.5
<i>trans</i> -methyloctalactone	2129.0	1542.4	4037.3	2456.1	1560.0	1562.3
<i>cis</i> -methyloctalactone	2316.3	1875.0	2367.5	1538.5	504.8	334.1
4-methylguaiacol	1574.7	1199.5	1682.3	580.2	636.5	242.0
phenol	2749.7	2497.8	1715.3	1086.4	826.0	294.4
vanillin	1851.3	731.6	5715.3	4173.4	1426.3	124.4

(not presented) were carried out to select the most suitable stationary phase for compounds found in the toasted wood. Interesting results were obtained for a large range of compounds using a silica fiber impregnated with 100 μm of polydimethylsiloxanes (PDMS). A 7 μm fiber, on the other hand, did not trap enough of the most volatile compounds. Prior to use, the fiber was heated to 260 $^{\circ}\text{C}$ with a jet of U hydrogen during 60 min in a gas chromatograph injector in open split mode.

The fiber was exposed to the flask atmosphere during 30 min at room temperature (23 ± 2 $^{\circ}\text{C}$). The absorption balance

was quickly reached (5 min) for medium and high compounds. After several tests, we found that the allotted time was a good compromise for studying both volatile and semivolatile substances in toasted wood at the same time with interesting recuperation.

Extraction and Assays of Volatile Wood Compounds in a Model Hydroalcoholic Solution. *Volatile Phenols.* Volatile phenols were assayed using an adapted form of the method described by Chatonnet and Boidron (1988). After addition of an internal standard (100 μL of dimethyl-3,4-phenol to 20 mg/L in ethanol) and adjustment of the sample's ionic

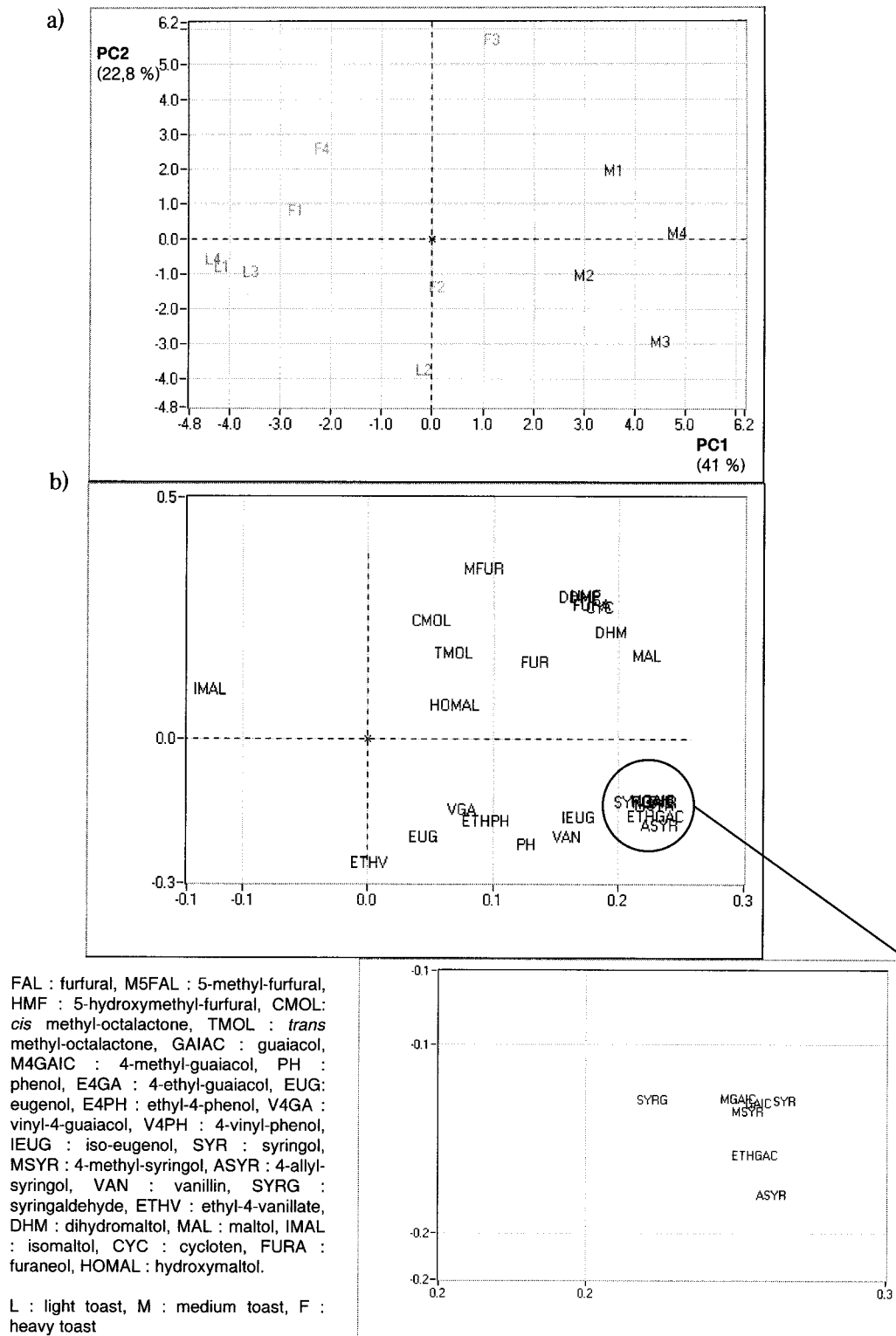


Figure 3. PCA on the data obtained from the analysis of wood soaking in a model hydroalcoholic solution: samples (a) and variables (b).

strength by dissolving 7.5 g of ammonium sulfate in 50 mL of wine, the model solution was extracted three times with dichloromethane (10.5 and 5 mL). The resulting organic phase was washed twice in 20 mL of sodium bicarbonate solution (5% dissolved in water) before slowly being concentrated to 0.5 mL under a nitrogen flux. The final extract was analyzed by gas-phase chromatography (Varian 3400 chromatograph; injection = 1 μ L; 230° splitless injection; valves closed, 0.5 min, split ratio = 30; BP20 capillary column, 50 m \times 0.22 mm, phase 0.25 μ m thick; carrier gas, 18 psi hydrogen; programming from 45 to 230 °C at a rate of 3°/min with a 25 min final

isotherm). Detection was by flame ionization (sensitivity = 1, range = 10^{-11}). Concentrations were measured by comparison with a standard range of pure phenols analyzed under the same conditions.

Furan Derivatives, γ -Lactones, and Compounds with an Enolone Structure. Extraction of 100 mL of the model solution was carried out after the addition of 100 μ L of 2-cyclopentene-2-one at 2 g/L (internal standard) in ethanol and 15 g of ammonium sulfate was dissolved in 20, 10, and 5 mL of dichloromethane. The organic phases were separated by plain sedimentation and then recombined; the resulting extract was

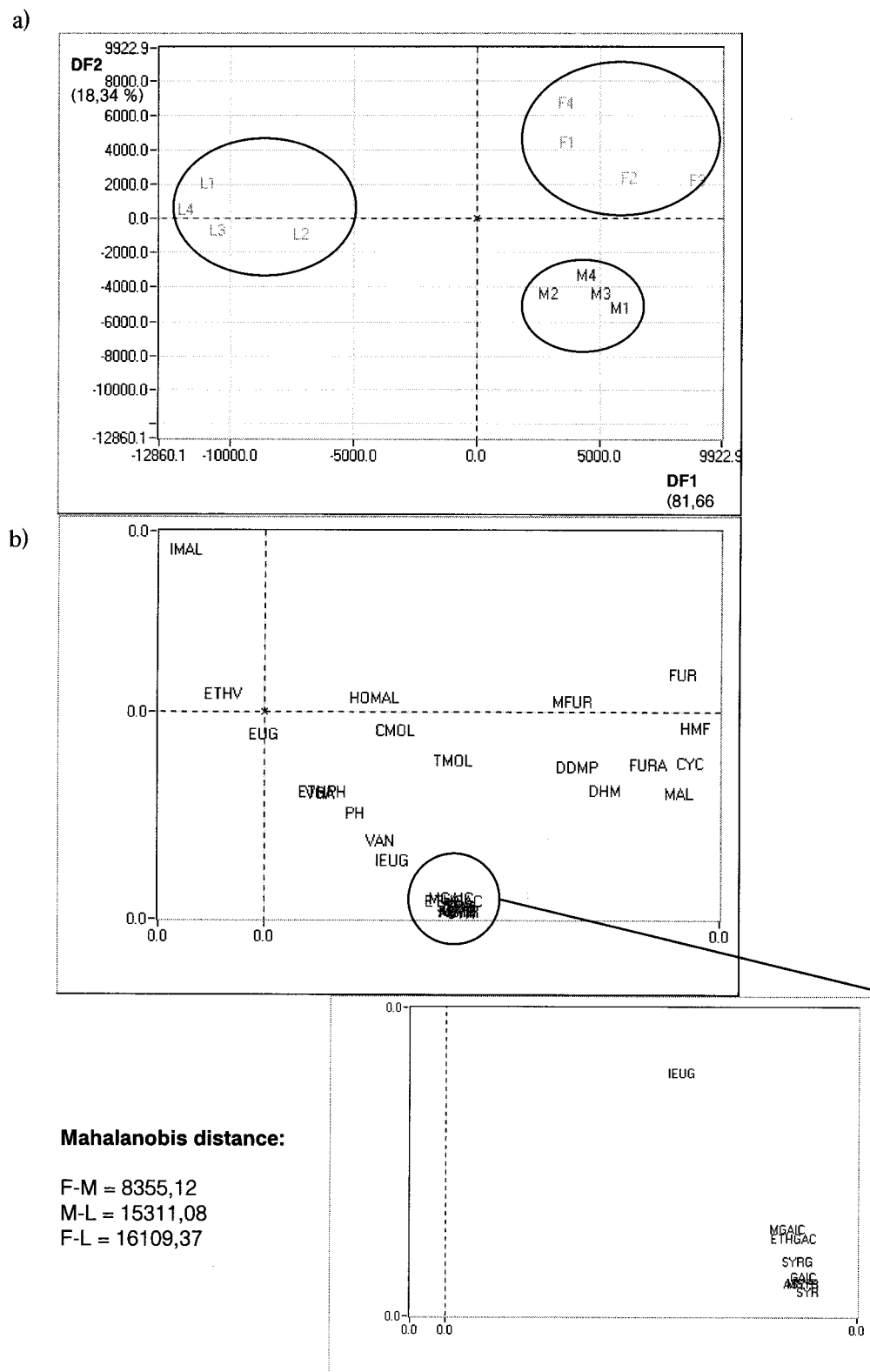


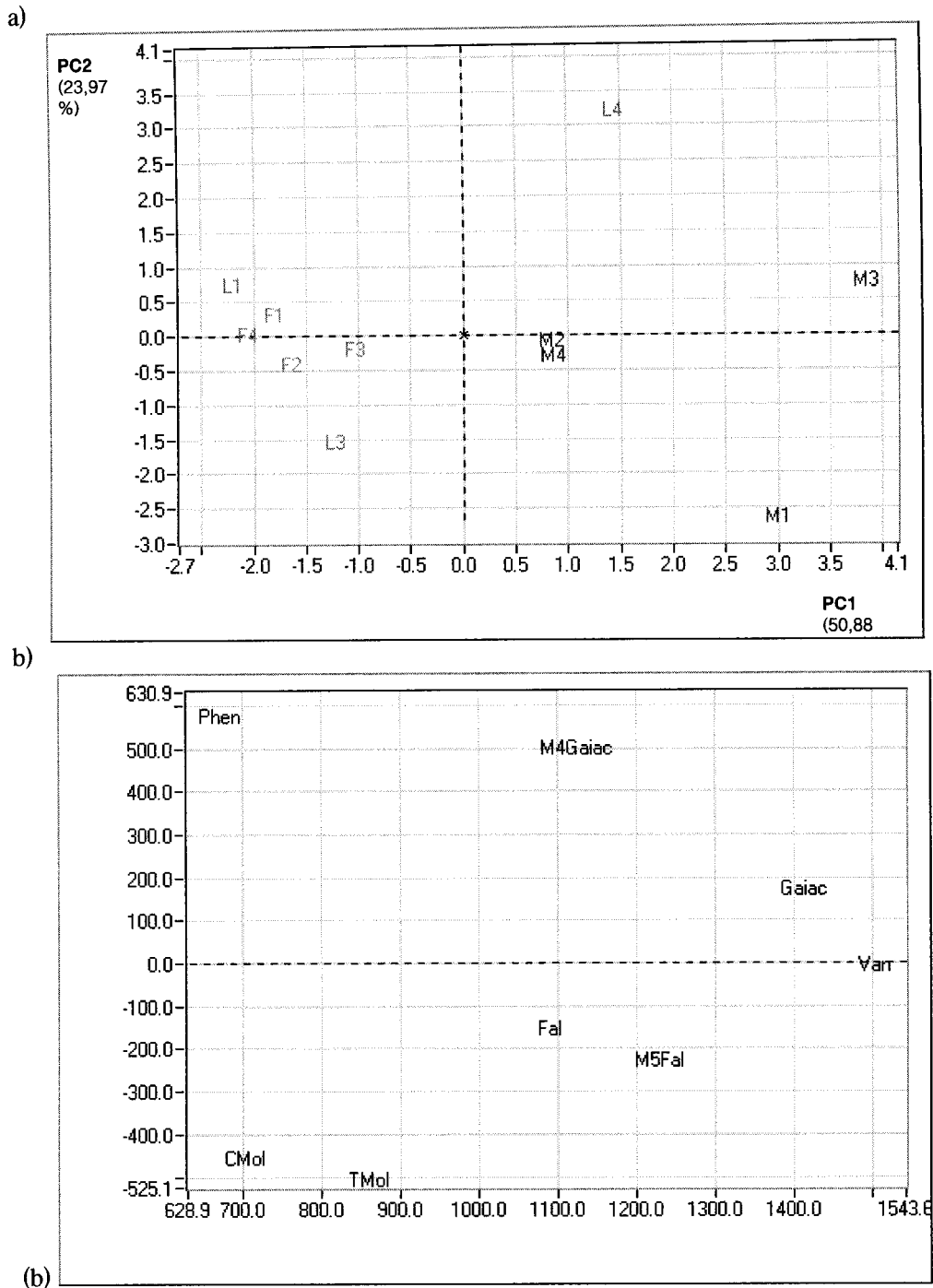
Figure 4. DFA to differentiate toasting intensities by analysis following soaking of toasted wood in a model hydroalcoholic solution: samples (a) and variables (b) (L, light toast; M, medium toast; F, heavy toast).

dried on sodium sulfate and slowly concentrated to 0.5 mL in N-flux.

The extract (2 μ L) were injected in splitless mode (temperature = 230 °C; closed time = 0.5 min; split ratio = 25; carrier gas, 18 psi N55 helium) on an HP-5890 chromatograph with a capillary column, as previously described (BP20, SGE), directly connected to an HP-5970 b working with electronic impact mass (70 keV, 250 °C) and selective ion recording (Table 1). Concentrations of each substance were measured

with calibrations made with pure reference compounds analyzed in the same conditions. When the latter were not available, concentrations were estimated by using the available molecules with the nearest chemical structure (maltol for hydroxy-, dihydro-, and isomaltol forms).

Desorption and Analysis of Volatile Compounds Analyzed by HSSPME. The SPME fiber was thermally desorbed at 250 °C for 5 min in a splitless injector (closed valve, insert SPME 0.75 mm) mounted on a Varian 3400 chromato-



Guaiac : gaïacol, Van : vanillin, M5FAL : 5-methyl-furfural, Fal, furfural, CMol : *cis* methyl-octalactone, TMol : *trans* methyl-octalactone, Phen : phenol, M4Gaiac : 4-methyl-gaïacol.

Figure 5. DFA to differentiate between toasting intensities on the basis of HSSPME analysis data: samples (a) and variables (b) (L, light toast; M, medium toast; F, heavy toast).

graph equipped with a fused silica capillary column impregnated with a polar phase (polyethylene glycol, Carbowax 20 M, 50 m × 0.25 mm, 0.25 μm, BP-20, SGE, Melbourne).

The carrier gas was U hydrogen (15 psi); the temperature was maintained at 40 °C for 5 min and then programmed to reach 230 °C at a rate of 3 °C/min, with a final isotherm of 25 min. A high-sensitivity FID (sensitivity = 1, range = 10⁻¹²) was used for detection. The signal was recorded and integrated by means of a D2500 Merck-Hitachi integrator.

Compounds were identified by coupling gas chromatography with a quadrupole mass spectrometer HP-5970 (70 keV, 250 °C). Chromatographic conditions were as previously described.

Statistical Analysis of Results. General descriptive statistics were processed in Microsoft Excel 5.3 spreadsheets, and the multivariate statistics (PCA and DFA) were processed using a special program developed by Laboratoire Excell (Merignac, France) after centering of the data.

RESULTS AND DISCUSSION

Volatile Compounds in Model Hydroalcoholic Solution. Figure 1 presents the chromatogram of volatile compounds extracted by soaking toasted oak wood in a model hydroalcoholic solution (12% volume)

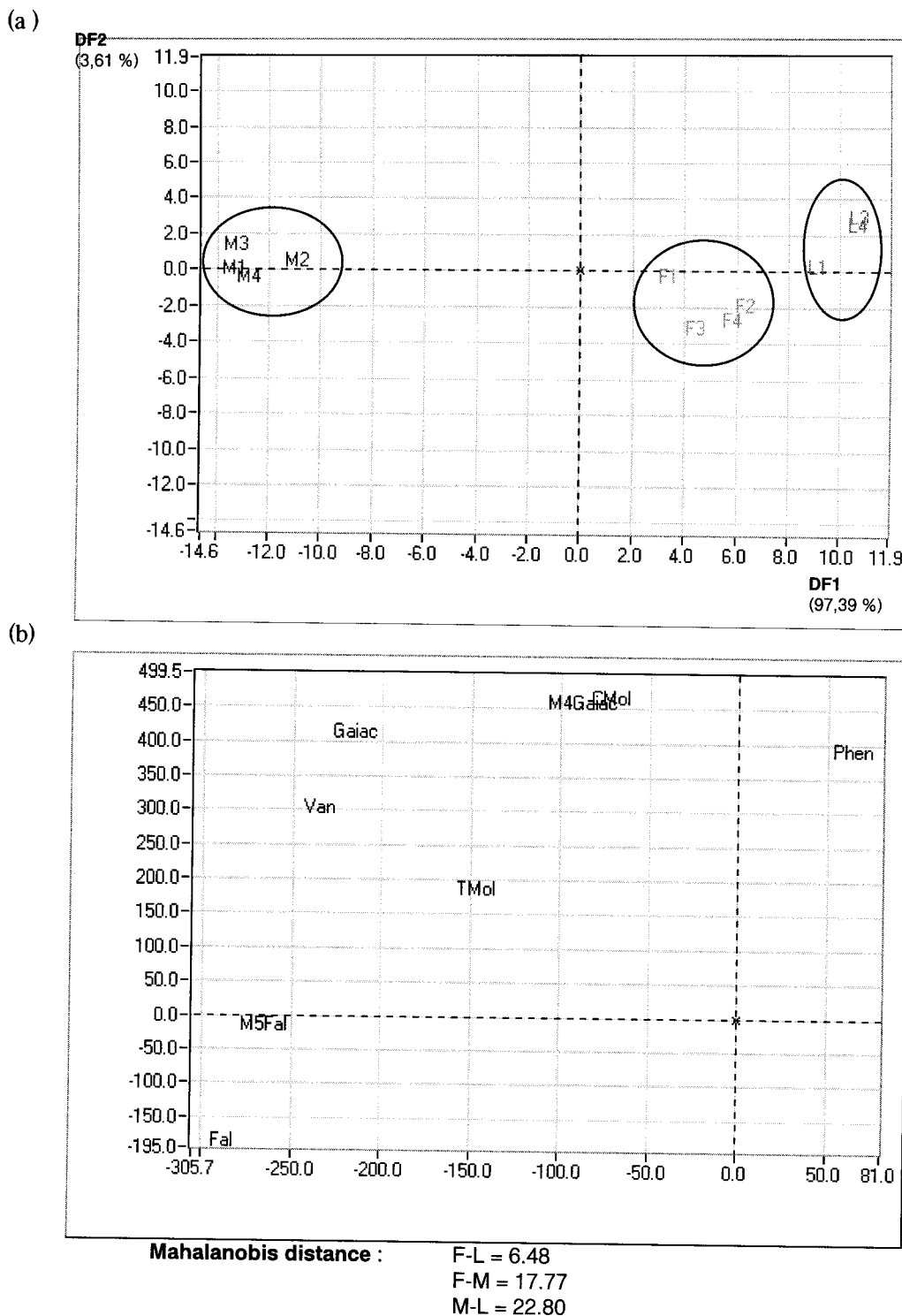


Figure 6. DFA to differentiate between toasting intensities on the basis of HSSPME analysis data: samples (a) and variables (b) (L, light toast; M, medium toast; F, heavy toast).

analyzed by GC/MS in scan mode. Quantification of various compounds is presented in Table 1.

HSSPME Analysis of Volatile Compounds. Figure 2 presents the chromatogram of volatile compounds of toasted oak wood with a medium or heavy toast analyzed by HSSPME and FID. Table 2 presents the results from triplicate injections. The numbers shown correspond to the surface of the peaks (arbitrary area units) integrated for each molecule positively identified by GC/MS.

Statistical Processing of Analysis Data. *Analysis of Extractable Compounds in Dilute Alcohol Solution.*

Figure 3 shows principal component analysis (PCA) on the data obtained from volatile compounds extracted by soaking. The intensities of lightly toasted (L) and heavily toasted (H) wood sharply contrasted with that of medium toasted wood (M) for PC1. PC2 showed a distinction between L and H toasting. The same differentiation was observed in the case of volatile phenols, produced by heat breakdown of lignin, and the furanic derivatives and enolone structures, produced by the breakdown of wood polysaccharides.

The three wood toasting intensity levels were easily identified by factorial discriminant analysis (DFA =

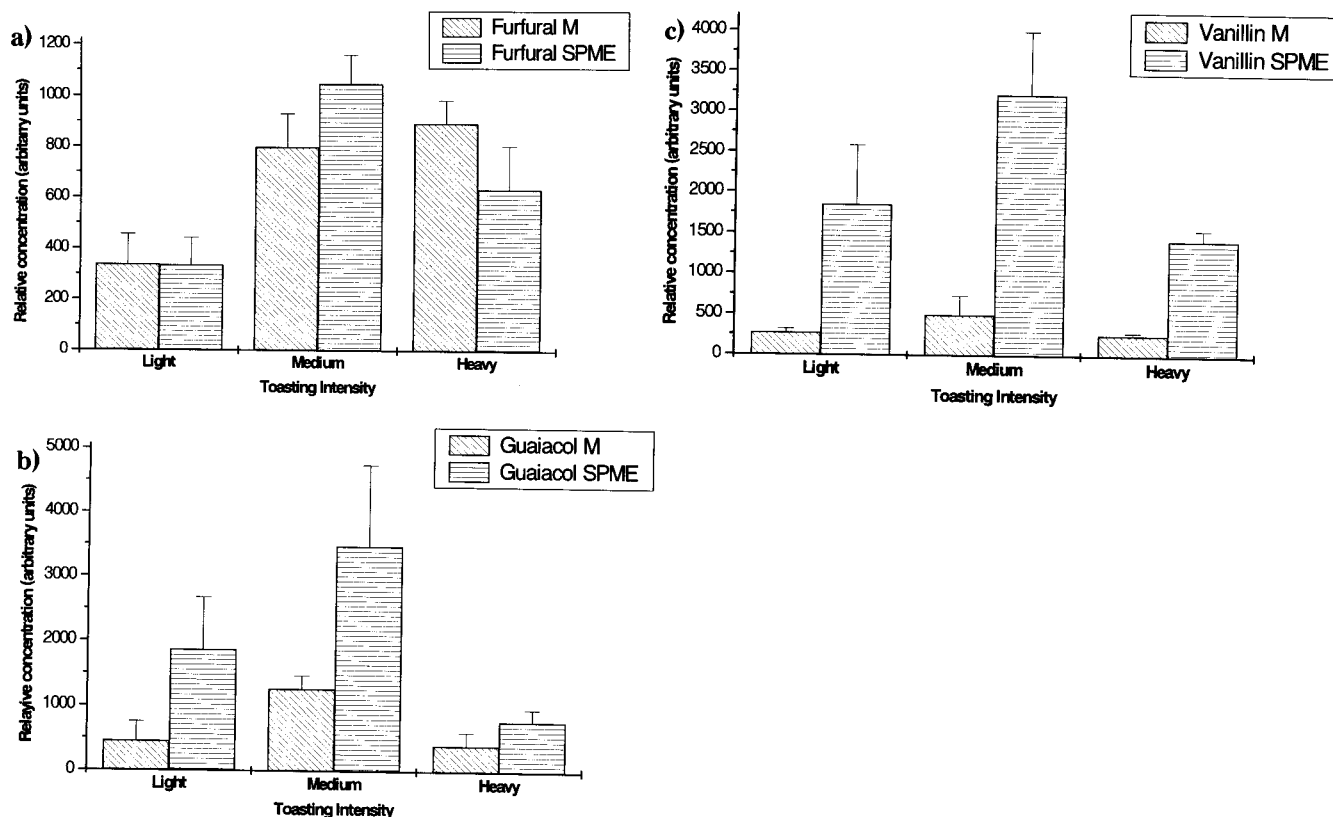


Figure 7. Comparison of the profiles of various compounds in toasted wood measured either by analyzing the wood macerations (M) or by HSSPME (SPME): (a) furfural; (b) guaiacol; (c) vanillin.

Table 3. Discriminating Potential of the Various Parameters Used for the DFA of Analytical Data from Dilute Alcohol Soakings (Coding cf. Figure 3)

variable	discriminant power ^a	variable	discriminant power ^a
MSYR	44.78	DDMP	19.84
SYR	42.67	VAN	19.26
MAL	42.60	IEUG	17.21
FUR	40.82	MFUR	16.75
SYRG	39.82	PH	9.85
ETHGAC	38.99	IMAL	6.88
ASYR	38.03	HOMAL	5.39
MGAIC	35.97	TMOL	4.09
GAIC	35.32	VGA	3.11
HMF	35.18	ETHV	2.61
CYC	34.16	EUG	1.95
FURA	26.94	CMOL	1.68
DHM	26.87	ETHPH	0.34

^a Square of the coefficient determinant.

100% of the variance; Figure 4) on these same data. The M and H intensities were not easily distinguished by physicochemical analysis of extractives; with PCA the most efficient variables for differentiating toasting intensity levels (Table 3, discriminating power > 10%) were the dimethoxy phenol and aldehyde derivatives, furans resulting from the breakdown of hemicelluloses, and all of the products of Maillard reactions (furanones and pyranones).

Analysis of Volatile Compounds in Gas Phase by HSSPME. PCA of the headspace assay data relating to several clearly identified molecules in toasted wood (Figure 5) did not clearly distinguish between lightly (L) or heavily (H) toasted wood. The furan variables, methyl-5-furfural, and the methyloctalactone isomers in component 2 distinctly contrast with the volatile phenols derived from the lignin.

Table 4. Discriminating Power of the Various Parameters Used for the DFA of the Data from HSSPME Analysis (Coding cf. Figure 5)

variable	discriminant power	variable	discriminant power
Fal	40.97	M4Gaiac	15.68
M5Fal	33.31	CMol	14.83
Gaiac	31.56	TMol	12.21
Van	31.27	Phen	10.61

The factorial discriminant analysis (Figure 7, DFA = 100% of the variance) makes a clear distinction among the three toasting levels possible, but the Mahalanobis intergroup distances were rather weak. The furanic aldehydes, both volatile phenols, and vanillin variables were the greatest contributors in distinguishing the levels of toasting (Table 4).

These results may be interpreted in relation to the curve of total compounds formed during toasting over time, which is bell-shaped. Formation of these substances increases at first and then tails off when the toasting intensity increases (Table 2). Under these conditions, it was not surprising to find that the intensity values of toasting byproducts at L and H were rather close.

Comparison of the Development of the Various Volatile Compounds by Measuring Their Concentrations by Soaking or HSSPME. Figure 7 shows the development of the various compounds in toasted wood, measured either by analysis of the maceration solutions or directly by HSSPME. Note that, with the exception of methyloctalactone, both analytical techniques show similar development profiles: concentrations increase during light to medium toasting and then decrease as toasting intensifies toward "heavy".

This result confirms previous observations (Chatonnet, 1991). In the case of isomers and methyloctalac-

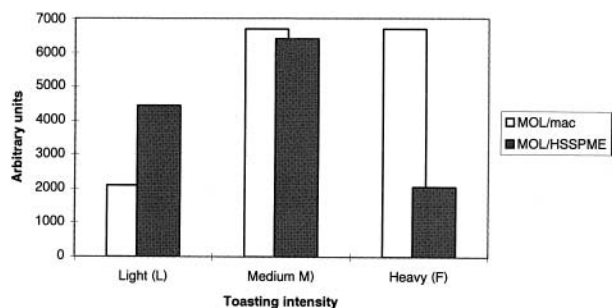


Figure 8. Comparison of the methyloctalactone *cis* + *trans* (MOL) quantified in wood macerations (mac) or by HSSPME.

tones, it is likely that soaking the wood releases lactones from deeper in the wood or those present in precursor form that cannot be measured in gas phase during HSSPME analysis (Figure 8).

DISCUSSION AND CONCLUSIONS

The assay of various compounds resulting from wood toasting extractable in a dilute alcohol medium provides a wealth of information. In PCA, the lightly (L) toasted samples were negatively correlated with the characteristic volatile compounds resulting from heating the wood. Multivariate statistical analysis clearly shows the contrast among compounds produced by the pyrolysis of polysaccharides (furanic aldehydes) or Maillard reactions (furanones and pyranones) and those formed from lignin (phenolic aldehydes and volatile phenols). PCA and DFA provide clear visual data for separating samples according to toasting intensity. There is always, however, a rather wide dispersion of the heavily toasted (H) samples.

Maceration analysis provides insight into the behavior of wine during wood-aging. The HSSPME analysis tested here was easier and quicker to implement than the previous technique. Even though, to date, fewer compounds were accessible through HSSPME analysis, current work on compound identification will soon improve the situation considerably. The complexity of the chromatograms obtained indicates that, once this technique has been perfected, it will be possible to analyze a great number of volatile and highly volatile wood compounds more rapidly. Sample preparation requires only minutes versus days for the maceration technique. Compared to the analysis of liquid-liquid extraction of wood with an internal standard, calibration of HSSPME for quantitative assays will always be more difficult. In most cases, the shapes of the development curves for volatile compounds according to toasting intensity analyzed by HSSPME were similar to those obtained using the maceration technique. The methyloctalactones were the only compounds to present a different development profile.

PCA of the HSSPME test results, and especially DFA, provided a clear differentiation among toasting levels. Given the smaller number of parameters used, the distance measured between levels was shorter, but nonetheless significant.

In conclusion, it is possible to identify barrel toasting levels accurately using the analytical data both from extractives in dilute alcohol solution and, much more simply, from volatile compounds emitted in gas phase analyzed by HSSPME. Initial results are quite promis-

ing. It is possible to envisage direct application of this technique to barrels, without having to take wood samples. Objective assessment of barrel toasting levels should be quick and easy and would not require destructive sampling.

Research in progress aims to perfect new, even faster, and nondestructive analysis techniques. It is possible to envisage in the near future the monitoring of barrel toasting levels during production, or on finished barrels, with the aim of improving the quality and regularity of finished products.

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