Application of Chemometric Techniques in Obtaining Macerates with Phenolic Compound Content Similar to That of Wines from the Jerez-Shery Region Subjected to Oxidative Aging

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This paper studies and quantifies the final concentration of phenolic aldehydes and acids (determined by HPLC) in a series of sobretablas wine macerates prepared with American oak shavings subjected to an accelerated aging system developed by our research group and based on thermal processes traditionally used in cooperage. This experiment aims to considerably reduce and control the oxidative aging period of oloroso wines from the Jerez-Sherry region as occurs in the dynamic system of soleras and criaderas, with the consequent economic benefits. To standardize the process by controlling the production technique of the macerates, the results were subjected to surface response methodology as a means of optimizing the experiment. The proposed model was found to be suitable after evaluation of the factors affecting the final concentration. Of the factors studied, it was found to be essential to control the charring time and/or the interactions between temperature and charring time for 10 of the 11 phenolic compounds studied.

Keywords: *Phenolic compounds; oxidative aging; Jerez-Sherry; accelerated aging*

INTRODUCTION

Oxidative aging is that applied to Andalusian fortified wines from Jerez-Sherry (oloroso wines). It is carried out in oak or chestnut barrels, and after fortification to 18 or 19% (v/v) alcohol, the wine ceases to be protected by the velo de flor (Cano, 1990).

For an average period of eight years, the wine is decanted several times from one barrel to another in what is known as the criaderas and soleras system, which is characteristic of this region. This technique guarantees the production of uniform wines that do not vary in taste, color, or bouquet and have the same organoleptic characteristics. The main disadvantage of the method is its poor economic viability, because only 20% of the wine in storage is annually available for sale. Together with the cost of the enormous number of barrels required, the interest on immobilized capital, and loss by evaporation, which can reach up to 7% per year, this means that any reduction in price of these wines is quite out of the question (Gomez and Carrascal, 1996).

As in other recognized wine-growing countries, this situation has led to attempts to speed up aging processes and reduce the time spent in the barrel to reduce production costs.

Alternative maturing or aging methods are based on a number of techniques, such as changes of temperature by application of hot, cold, or even freezing temperatures, although they appear to result in unpleasant tastes and smells (Lafont, 1971). Hydrogenation removes most of the unpleasant smells and tastes (especially roughness) from young wines, whereas oxygenation reduces aging times by subjecting the wine to a continuous flow of O_2 in the barrel (Krolenko et al., 1981). Other authors such as Semenenko et al. (1982) or Mndzhoyan et al. (1989) have proposed the simultaneous use of heat and oxygen, whereby oxidative processes are encouraged and at the same time extrac-

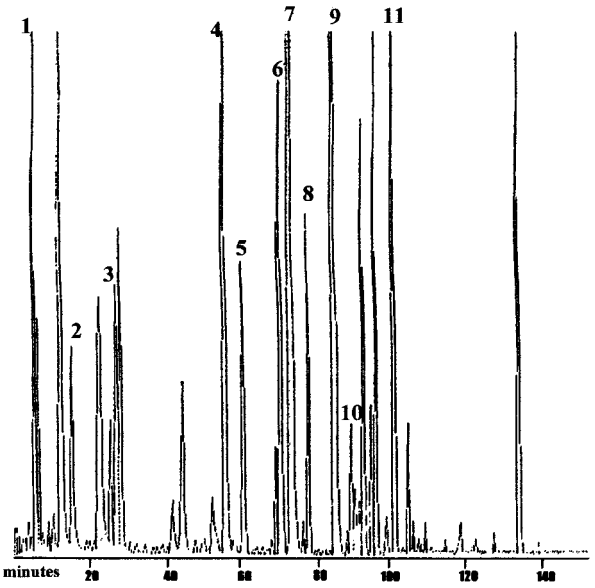

Figure 1. Chromatogram of phenolic compounds in one of the samples analyzed [following the analytic technique developed by Monedero et al. (1998)]. Peaks: gallic acid (1); protocatechuic acid (2); *p*-hydroxybenzoic acid (3); vanillic acid (4); caffeic acid (5); syringic acid (6); vanillin (7); *p*-coumaric acid (8); syringaldehyde (9); ferulic acid (10); sinapaldehyde (11).

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Table 1. Statistical Summary of Concentrations (Milligrams per Liter) Detected in Commercial Oloroso Wines following Monedero et al. (1998)

	n	mean	SD	CV (%)	range
Benzoic Acids					
gallic acid	18	1.691	1.039	61.460	3.330
protocatechuic acid	18	1.286	0.381	29.640	1.563
p-hydroxybenzoic acid	18	1.541	0.287	18.650	0.975
syringic acid	18	1.092	0.318	29.190	1.052
vanillic acid	18	0.624	0.165	26.470	0.613
Cinnamic Acids					
caffeic acid	18	1.473	0.268	18.250	0.888
p-coumaric acid	18	1.214	0.147	12.110	0.622
ferulic acid	18	0.546	0.111	20.370	0.398
Phenolic Aldehydes					
syringaldehyde	18	2.492	0.589	23.640	2.117
vanillin	18	1.349	0.392	29.100	1.438
sinapaldehyde	18	3.467	0.707	20.390	2.341

tion of components is increased. Other physical methods have also been suggested, such as the application of magnetic fields, acoustic waves, or ultrasounds (Kwasnik and Ziobrowsky, 1984; Lafont, 1971).

Nonetheless, despite the range of methods available, the most widely used acceleration technique is the use of oak wood extracts (Puech, 1987; Puech and Moutounet, 1992) obtained by decoction, infusion, leaching, and maceration (Mangas et al., 1996; Puech and Sarni, 1990), with or without prior subjection of the wood to physical treatments, such as ultrasounds, high pressure, or charring (Tanahashi et al., 1989), or chemical treatments using acids or alkalis (Fenner and Lephardt, 1981; Bredenberg et al., 1987; Skorikhin and Irazikhanov, 1986).

With the aim of considerably reducing the oxidative aging period and thus providing significant economic benefits for the wine-making industry, we have attempted to perfect a rapid aging technique by producing sobretablas wine macerates using oak shavings subjected to thermal treatment. This process is based on the traditional techniques used in cooperage and represents an alternative to traditional aging (Chatonnet et al., 1993; Artajona, 1991; Sarni et al., 1990).

Table 2. Phenolic Compounds Concentrations (Milligrams per Liter) Detected in Each of the Macerates Studied

charring time (h)	maceration time (months)	gallic acid	protocatechuic acid	*p*-hydroxybenzoic acid	syringic acid	vanillic acid	caffeic acid	*p*-coumaric acid	ferulic acid	syringaldehyde	vanillin	sinapaldehyde
(A) Charring Temperature of Shavings: 180 °C												
sobretablas wine		0.78	0.90	0.92	1.08	0.35	0.31	0.75	0.42	0.25	0.46	0.13
3	1	2.57	0.98	1.02	1.50	0.82	1.64	0.75	0.51	4.43	1.70	3.80
6	1	2.71	1.15	1.10	1.53	0.83	1.86	0.74	0.59	4.61	1.73	4.50
15	1	3.30	1.47	1.36	1.62	0.89	1.93	0.81	0.58	4.85	1.79	6.30
3	2	3.13	1.35	1.01	1.52	0.92	1.99	0.90	0.61	4.72	1.68	7.00
6	2	3.05	1.06	1.24	1.40	0.99	1.97	0.95	0.70	4.00	1.60	8.25
15	2	2.14	0.80	1.04	3.30	1.24	1.43	0.82	0.62	6.81	2.80	4.58
3	3	2.15	0.86	1.11	3.20	1.34	1.74	0.81	0.63	6.96	2.60	7.19
6	3	2.89	1.38	1.50	3.39	1.41	1.70	0.85	0.74	7.09	2.98	10.90
15	3	2.78	1.30	1.19	4.03	2.05	1.93	0.91	0.79	10.38	3.96	12.25
3	4	2.62	1.50	1.26	5.21	2.53	1.99	1.08	0.80	11.41	4.89	14.73
6	4	2.13	1.27	1.04	7.93	3.85	1.50	0.757	0.52	20.23	6.87	12.78
15	4	1.99	1.15	1.12	8.84	4.68	1.40	0.78	0.54	22.51	7.96	13.22
3	10	2.65	1.45	1.43	10.03	5.79	1.83	0.73	0.55	22.35	8.76	14.2
6	10	2.54	1.34	1.33	10.21	5.98	1.92	0.71	0.55	23.45	8.99	14.30
15	10	2.42	1.45	1.35	10.13	6.09	1.98	0.80	0.51	24.05	9.03	15.00
(B) Charring Temperature of Shavings: 195 °C												
3	1	2.00	1.27	1.16	1.90	0.99	1.67	0.76	0.61	6.23	2.10	11.40
6	1	2.12	1.12	1.14	2.70	1.31	1.87	0.80	0.84	6.45	2.88	14.40
15	1	2.34	1.15	1.23	2.01	1.33	1.83	0.81	0.83	6.42	2.20	18.60
3	2	2.23	1.16	1.16	2.83	1.15	1.70	0.82	0.84	6.22	2.98	11.50
6	2	2.35	1.23	1.09	4.10	1.24	1.63	1.05	0.85	10.62	4.37	12.80
15	2	1.78	1.06	0.92	3.76	2.68	1.44	0.75	0.50	8.43	3.01	14.80
3	3	1.99	0.78	1.12	3.40	3.23	1.46	1.00	0.58	9.08	2.98	14.90
6	3	2.21	1.38	1.12	4.35	3.40	1.87	0.94	0.65	13.35	4.10	17.60
15	3	2.26	1.63	1.20	5.53	2.80	1.89	0.96	0.81	15.76	4.96	19.90
3	4	2.29	1.60	1.23	6.34	2.64	1.81	1.05	1.12	19.69	5.92	23.90
6	4	1.70	0.93	1.25	9.84	5.69	1.57	1.02	0.62	21.48	8.15	13.90
15	4	1.80	0.92	1.37	10.30	5.83	1.9	1.15	0.73	23.33	8.20	14.20
3	10	1.99	1.15	1.39	10.80	6.1	1.88	1.21	0.71	25.83	8.60	14.40
6	10	2.03	1.342	1.19	12.30	6.02	1.95	1.04	0.73	28.37	10.20	13.30
15	10	2.08	1.51	1.36	13.10	6.91	1.99	1.39	0.74	31.84	11.1	12.90
(C) Charring Temperature of Shavings: 215 °C												
3	1	1.87	1.12	1.08	2.47	1.21	1.81	1.03	0.72	8.79	3.01	17.60
6	1	1.80	1.46	1.07	2.59	1.22	1.81	0.94	0.76	5.15	3.11	16.80
15	1	1.98	1.59	1.21	2.72	1.58	1.83	1.11	0.73	11.15	3.20	18.30
3	2	1.99	1.62	1.22	2.93	1.63	1.78	1.10	0.74	10.89	3.46	17.40
6	2	1.96	1.63	1.36	4.52	1.92	1.84	1.40	0.89	18.40	4.70	17.30
15	2	1.60	1.23	0.99	3.08	1.22	1.87	0.96	0.81	9.79	3.03	10.60
3	3	1.65	1.23	0.95	2.99	1.39	1.87	0.79	0.86	8.60	2.60	11.20
6	3	1.86	1.48	1.13	3.21	1.76	1.89	0.84	0.89	12.24	3.04	12.10
15	3	1.91	1.38	1.20	4.32	1.93	1.91	0.91	0.85	12.96	3.48	12.40
3	4	1.94	1.73	1.46	5.89	2.06	1.84	1.33	1.15	20.16	5.43	13.01
6	4	1.51	0.73	1.02	4.20	1.56	1.51	0.90	0.62	10.55	3.63	4.59
15	4	1.59	0.99	1.20	4.90	1.83	1.61	1.03	0.71	12.82	3.98	5.20
3	10	1.64	1.14	1.45	5.60	2.03	1.65	1.20	0.83	14.20	4.60	6.90
6	10	1.51	1.01	1.11	5.30	1.98	1.50	1.14	0.82	13.02	4.10	5.90
15	10	1.71	1.83	1.20	5.70	2.20	1.51	1.12	0.95	15.858	4.62	4.40

Table 3. Variance Analysis for Design of Effects

effect	sum of squares	DF	mean sq	*F* ratio	*p* value
ANOVA for Gallic Acid					
A, temp	4.09340981	1	4.0934098	85.39	0.0000
B, time	0.91154061	1	0.9115406	19.02	0.0001
C, mac time	0.41285757	1	0.4128576	8.61	0.0056
AB	0.04930914	1	0.0493091	1.03	0.3169
AC	0.01958680	1	0.0195868	0.41	0.5333
BC	0.00059976	1	0.0005998	0.01	0.9127
total error	1.82153047	38	0.0479350		
total (corr)	8.52926244	44			
$R^2 = 0.786438$	R^2 (adj for df)	=	0.762067		
A, temp	5.06851864	1	5.0685186	109.89	0.0000
B, time	1.12244882	1	1.1224488	24.34	0.0000
C, mac time	0.44726882	1	0.4472688	9.70	0.0034
total error	1.89102617	41	0.0461226		
total (corr)	8.52926244		44		
$R^2 = 0.77829$	R^2 (adj for df)	=	0.762067		
ANOVA for Syringaldehyde					
A, temp	0.64191	1	0.6419	0.07	0.7920
B, time	1006.67884	1	1006.6788	113.78	0.0000
C, mac time	243.81964	1	243.8196	27.56	0.0000
AB	380.75867	1	380.7587	43.04	0.0000
AC	32.40120	1	32.4012	3.66	0.0632
BC	0.73539	1	0.7354	0.08	0.7777
total error	336.20665	38	8.8475		
total (corr)	2346.34695	44			
$R^2 = 0.856711$	R^2 (adj for df)	=	0.834086		
B, time	1264.05733	1	1264.0573	135.73	0.0000
C, mac time	248.86210	1	248.8621	26.72	0.0000
AB	368.32551	1	368.3255	39.55	0.0000
total error	381.82757	41	9.3129		
total (corr)	2346.34695	44			
$R^2 = 0.837267$	R^2 (adj for df)	=	0.82536		

In this paper we describe how we measured a total of 11 phenolic compounds including benzoic acids, cinnamic acids, and phenolic aldehydes using HPLC after concentration of the sample and extraction using organic solvent (Laurichesse and Tricher, 1988; Fernández de Simón et al., 1996).

We should also point out the importance of this study given the role that these substances play in the organoleptic (oxide reduction, astringency, color, olfactory perception), rheological (colloidal processes), conservation (browning), and other characteristics of alcoholic beverages (Boidron et al., 1988; Glories, 1987; Alonso, 1988).

Finally, the Statgraphics statistics program was used to carry out a surface model study, to determine the influence of the factors chosen for the experiment on the phenolic compounds concentration, and also to quantify this influence by an equation.

MATERIALS AND METHODS

Samples. Preparation of Macerates. The chosen macerates were prepared with shavings of American oak (*Quercus alba*) at 2% wood in sobretablas wine provided by a winemaker of the region. The shavings were 3−5 mm in size, because studies by Giménez et al. (1996) showed that this was the best size for maximum extraction of furanic and phenolic aldehydes from the wood by alcohol spirit. The shavings were sieved through an appropriately sized mesh and heated to 180, 195, and 215 °C with a thermostatically controlled oven for 3, 6, and 15 h.

When the macerates had been prepared according to the procedure described abov3, they were deposited for aging in a room with humidity and temperature similar to those of a wine cellar (bodega). They were kept in these conditions of static maceration in the wine sobretablas for 1, 2, 3, 4, and 10 months. The samples were finally shaken for 24 h nonstop.

Analytic Determinations. Individual Phenolic Compounds. These determinations were carried out using HPLC, under the following conditions:

(a) Equipment. A Perkin-Elmer liquid chromatograph with a Waters model 717 plus automatic injector and a diode array 235 type UV−vis detector equipped with a Penelson 1020 integrator was used. The column used was a CH-18 reverse phase Spherisorb (25 × 0.46 cm) with 5 μm internal particle size.

(b) Standards. Reference compounds were purchased from Fluka (protocatechuic, *p*-hydroxybenzoic, syringic, *p*-coumaric, and ferulic acids and synapaldehyde), Sigma (vanillic acid), and Merck (gallic, vanillic, and caffeic acids and syringaldehyde).

(c) Chromatographic Conditions. Mobile phase A consisted of water/glacial acetic acid (98:2) and mobile phase B of methanol/water/glacial acetic acid (60:38:2) (García et al., 1985; Pinedo et al., 1994). Absorption wavelength was 280/340 nm.

(d) Sample Preparation. We followed the conventional (discontinuous) extraction method, taking 100 mL of sample and concentrating it in a vacuum (T < 40 °C) to 25 mL. Ethyl ether was used as organic extraction solvent (four extractions), and anhydrous sodium sulfate was used as desiccant when necessary. The resulting residue was dissolved in 1 mL of a methanol/water (1:1) mixture and filtered through a 0.45 μm membrane (Diez and Gómez Cordobés, 1980).

(e) Identification, Measurement, and Validation of the Method. Chromatographic peaks were identified by comparing their retention time and UV spectrum with the reference standards. Given the complexity of the sample, quantification was carried out by suing the external standard method. Three determinations were made on each wine sample. Figure 1 provides a graphic representation of the chromatogram of one of the samples analyzed. The accuracy and precision of the method are described in Monedero et al. (1998).

Statistical Analysis. To optimize the experiment, the results were analyzed using the experiment design section of the Statgraphics program, which offers surface response methodology by evaluation of the factors affecting the variable response, suitability of the model, its graphic representation, and the analysis of mathematical models (Montgomery, 1993; Davies, 1993; Miller and Miller, 1993).

RESULTS AND DISCUSSION

In a previous study carried out by our research group we determined the levels of phenolic compounds in samples of commercial wines from the main producers of oloroso wines (subjected to oxidative aging) in the Jerez district. The results of this study (Monedero et al., 1998) were taken into account for reference purposes (Table 1).

On the basis of these data, together with previous studies carried out by our group on most suitable conditions (temperature and time settings, etc.) (Gimenez et al., 1996; Quesada et al., 1996), we designed the experiment with three variable factors on which the response depended: charring temperature, charring time, and maceration time. In this case, the first two factors had three different values (180, 195, and 215 °C for charring temperature and 3, 6, and 15 h for charring time), whereas maceration time had five (1, 2, 3, 4, and

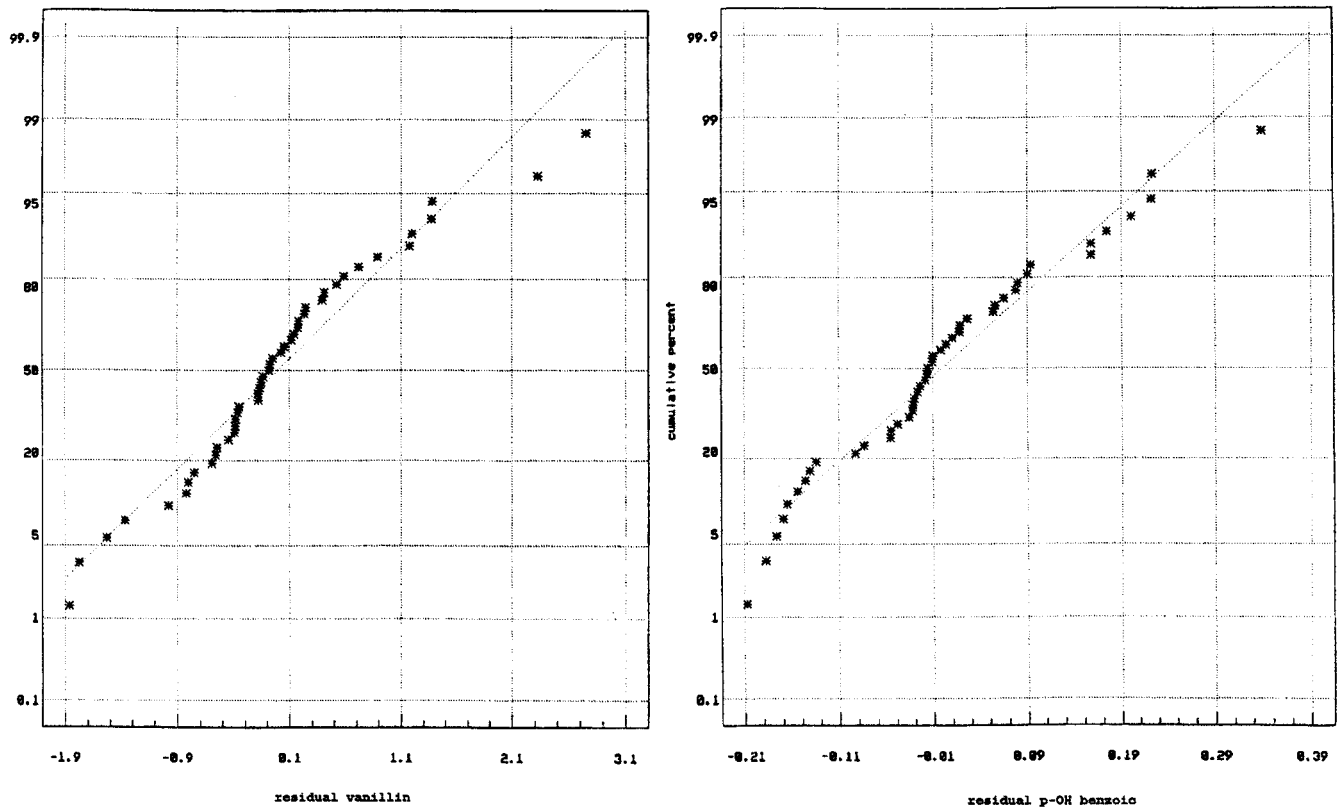

Figure 2. Graphic representation of the normal probability of residual values in the vanillin and *p*-hydroxybenzoic variables.

10 months) and one variable response: concentration of the phenolic compounds under study.

Because of the interaction between factors affecting the variable response, the model corresponds to a factorial design, having a development matrix in which two factors remain constant while the third is modified, thus leading to a total of 45 experiments.

The phenolic compound concentration (milligrams per liter) for each of the macerates studied is represented in Table 2.

An experiment design study was carried out using the Statgraphics program to examine the influence on the phenolic compound concentration of the factors chosen for the experiment, as well as the possibility of quantifying this influence by an equation.

The statistical treatment of the diagnosis was carried out by variance analysis, which, in this case, was multiway because there were three factors.

Total error was always taken into account. In all cases interactions higher than first order were discounted and when this model was unsatisfactory.

Table 3 shows the variance analyses (before and after elimination of nonsignificant effects) for gallic acid and syringaldehyde. In the first case, it can be seen that only the principal factors are significant, whereas, in the second, both the principal factors and the first-order interaction (charring temperature/charring time) are significant.

In this type of analysis, adequacy of fit is determined either by the values of the R^2 determination coefficient (regression quality indicator) or by graphic methods, such as the representation of residuals in comparison with the different parameters (either in the order in which the results were obtained or for each of the separate variables) or by the most common method in experiments with no replicas, which is the case here in view of the size, which is by using the graph of normal probability of residuals. Figure 2 shows the graphic representation of two other polyphenols, vanillin ($R^2 = 0.86744$), as an example of a significant pattern, and *p*-hydroxybenzoic ($R^2 = 0.25765$), as an example of a nonsignificant pattern.

Once the significant effects have been determined, their quantitative ordering can be observed in the pareto graph. If this graph is standardized, it also serves to test the significance of the factor in question. For example, Figure 3 represents the paretos, before and after elimination of nonsignificant factors, for the two phenolic compounds mentioned above (gallic acid and syringaldehyde).

Possible interactions are shown by the representations and criteria of Figure 4. For example, we can observe the interactions between charring temperature and charring time in the model obtained for syringaldehyde, which shows that for lower temperatures, higher responses can be obtained by increasing the charring time.

Our study also takes into account the first- or second-order coefficients of the model that allow the responses to be related to the separate variables (factors) and the fit of which with the experimental model we had previously determined.

The equations with the correlation coefficients for the phenolic compounds studied are shown in Table 4. It can be seen that application of the study to the 11 phenolic compounds described above leads to highly acceptable models/patterns ($R^2 > 0.8$) for syringic acid, syringaldehyde, and vanillin. We also found acceptable patterns ($R^2 > 0.7$) for gallic acid, synapaldehyde, and vanillic acid, patterns below the acceptable limit ($R^2 >$

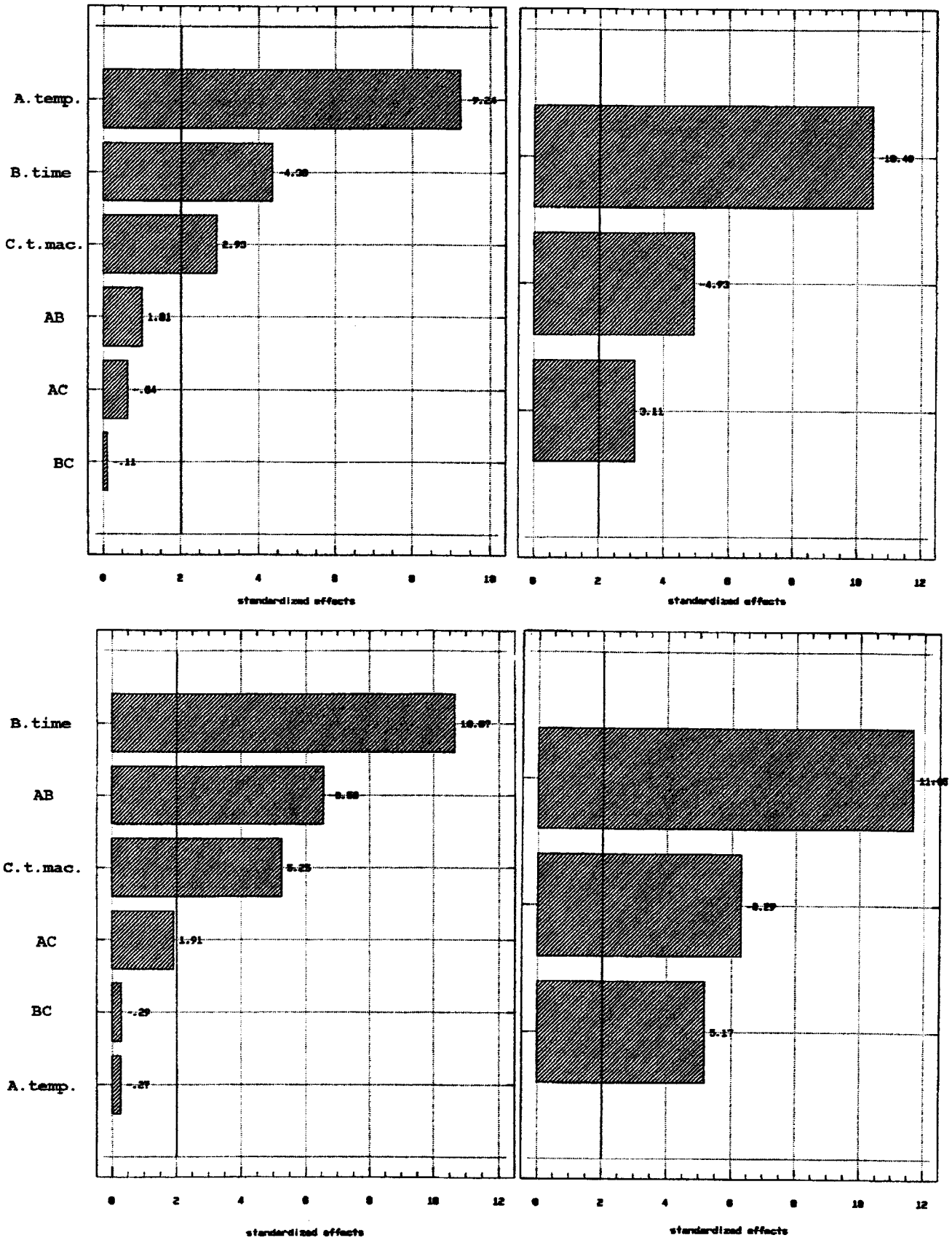

Figure 3. Representation of paretos before and after elimination of the factors having effects that are not significant for syringaldehyde and gallic acid.

0.5) for *p*-coumaric acid and protocatechuic acid, and unacceptable patterns for ferulic acid, *p*-hydroxybenzoic acid, and caffeic acid.

Table 5 shows that, in the significant patterns, charring time is the most influential factor, except in the case of gallic acid, for which charring temperature

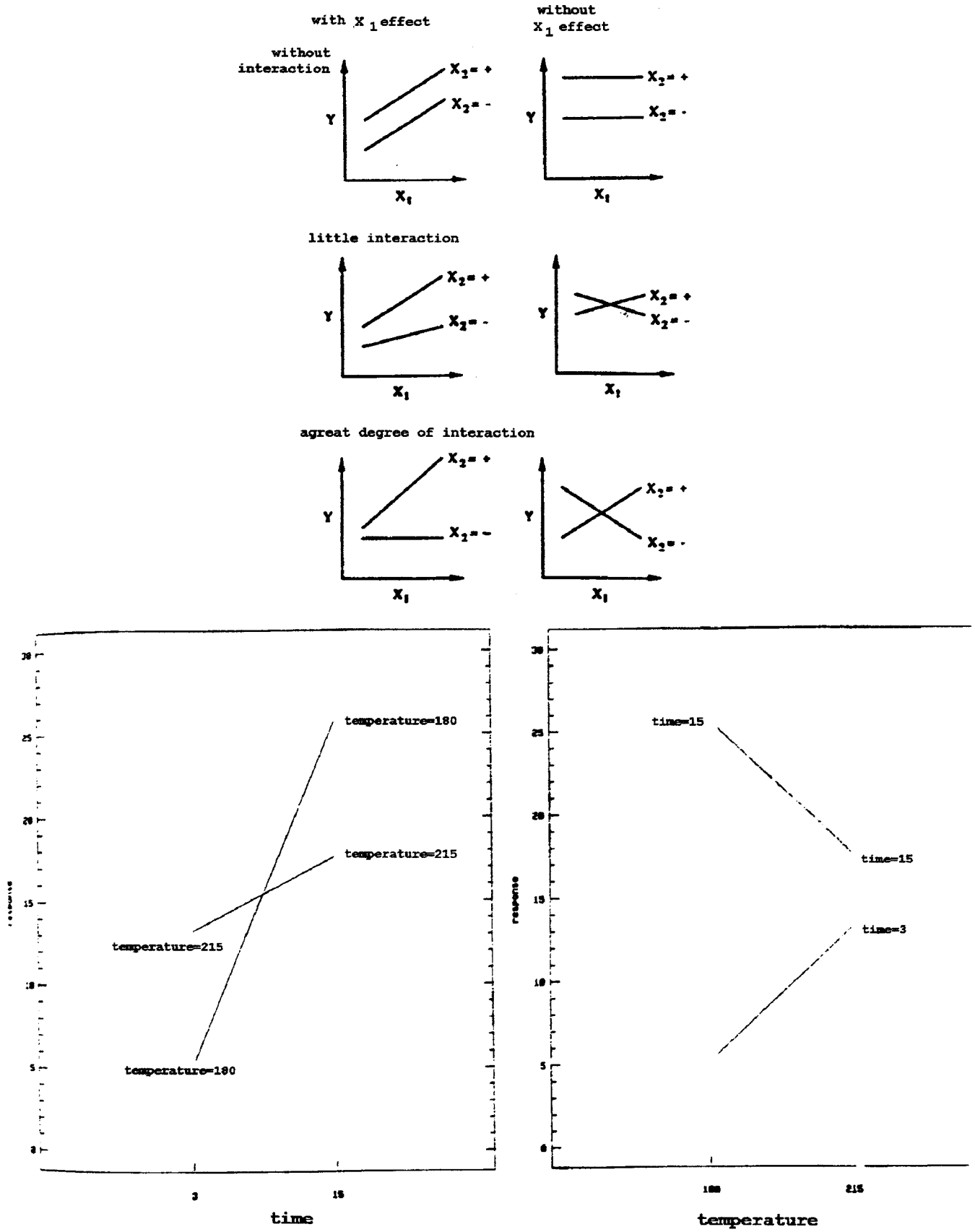

Figure 4. Interpretative criteria of possible interactions between the different factors and their application in the charring temperature/charring time interaction for the syringaldehyde variable.

is most influential. Similarly, caffeic acid is the only one of the 11 phenolic compounds studied on which neither of the factors mentioned has any effect. Maceration time is only significant in those models in which no acceptable relation was found between the response variable and the process (practically constant concentration).

Table 4. Equations with Correlation Coefficients for the Different Phenolic Compounds Studied

	A	*B*	*C*	*AB*	*AC*	Cte	R^2	R^2 (adj)
gallic acid	−0.0234	−0.0310	0.0315			6.876	0.7783	0.7621
protocatechuic acid			0.0197	-1.069×10^{-4}		1.241	0.5351	0.5011
p-hydroxybenzoic acid		7.846×10^{-3}	0.0186		3.537×10^{-3}	1.058	0.2576	0.2223
syringic acid	0.0867	3.3821	0.2217	−0.0146		−16.968	0.8657	0.8522
vanillic acid	0.0517	2.2679		−0.0102		−9.723	0.7944	0.7794
caffeic acid			0.0191			1.690	0.1207	0.0990
p-coumaric acid	6.361×10^{-3}		0.0291			−0.411	0.5361	0.5140
ferulic acid	5.835×10^{-3}		0.0237			−0.514	0.3546	0.3239
syringaldehyde		3.2926	0.7436	−0.0113		1.688	0.8373	0.8254
vanillin	0.0907	3.2292	0.2177	−0.0145		−17.194	0.8674	0.8542
sinapaldehyde		2.1526		−0.0104		13.452	0.7348	0.7224

Table 5. Summary of Significance of the Qualitative Effects of the Three Factors and Their Interactions on the Variable Response

	factor[a]								
	temp (*A*)	charring time (*B*)	mac time (*C*)	*AA*	*AB*	*AC*	*BB*	*BC*	*CC*
gallic acid	XXX ↑	XX	X						
protocatechuic acid	≈		XXX ↑		XX	≈	XX		
p-hydroxybenzoic acid		XX	XXX						
syringic acid	X	XXX ↑↑	X		XX				
vanillic acid	X	XXX ↑	≈		XX				
caffeic acid			XXX						
p-coumaric acid	XXX		XXX						
ferulic acid	XXX	≈	XX ↑						
syringaldehyde		XXX	XXX		XXX				
vanillin	X	XXX ↑	XX		XX				
sinapaldehyde	-	XXX	≈		XXX				

[a] X, degree of influence of the factor over the variable response; ≈, almost significant; ↑, it stands out in a highly significant way in relation to other factors.

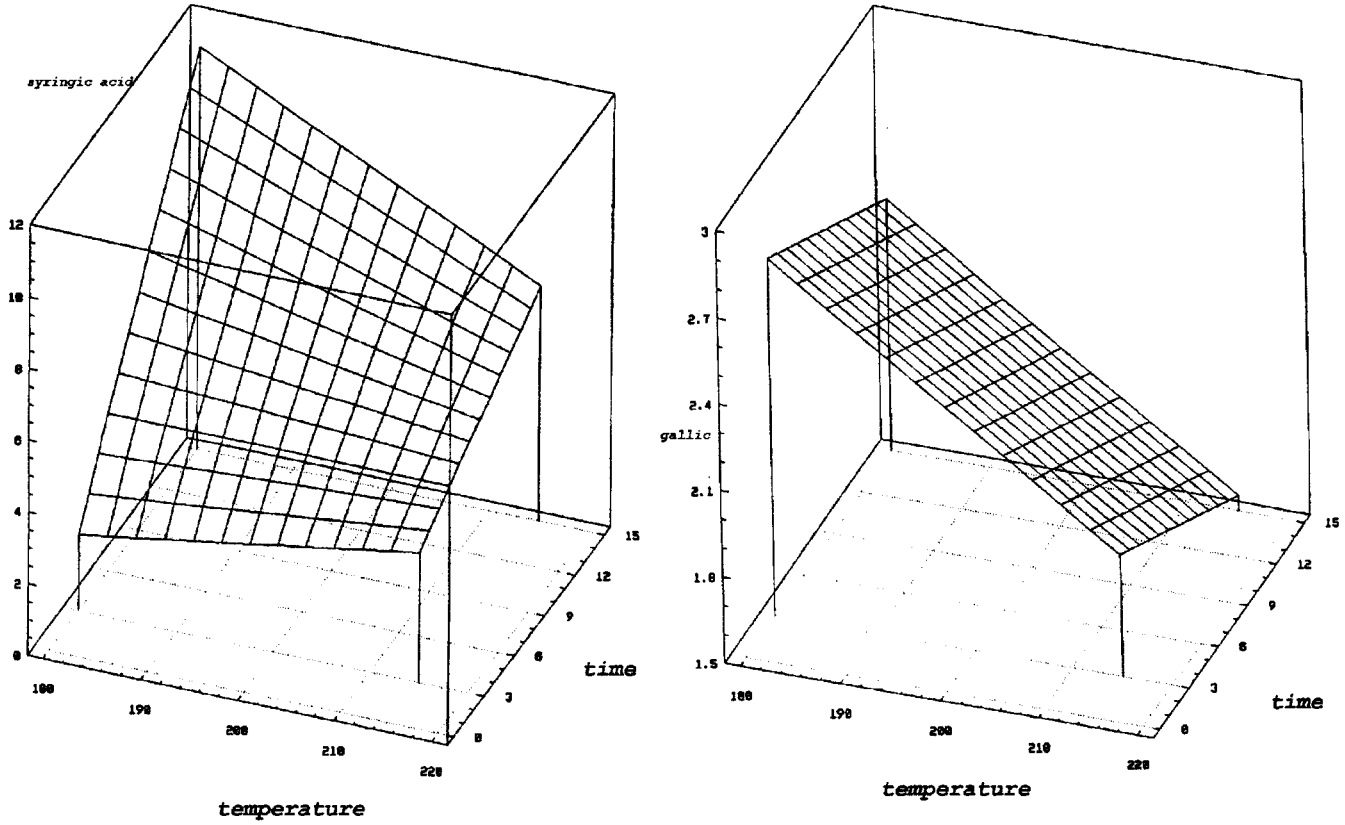

Figure 5. Tridimensional representation of the variable response (syringic acid and gallic acid) according to charring temperature, charring time, and maceration time.

Similarly, analysis of the coefficients (Table 4) determining the response surface equation shows clearly higher coefficients for charring time than for charring temperature—3.38/0.08 for syringic acid and 3.22/0.090 for vanillin. The exception is gallic acid, for which the two factors are approximately of the same order. These

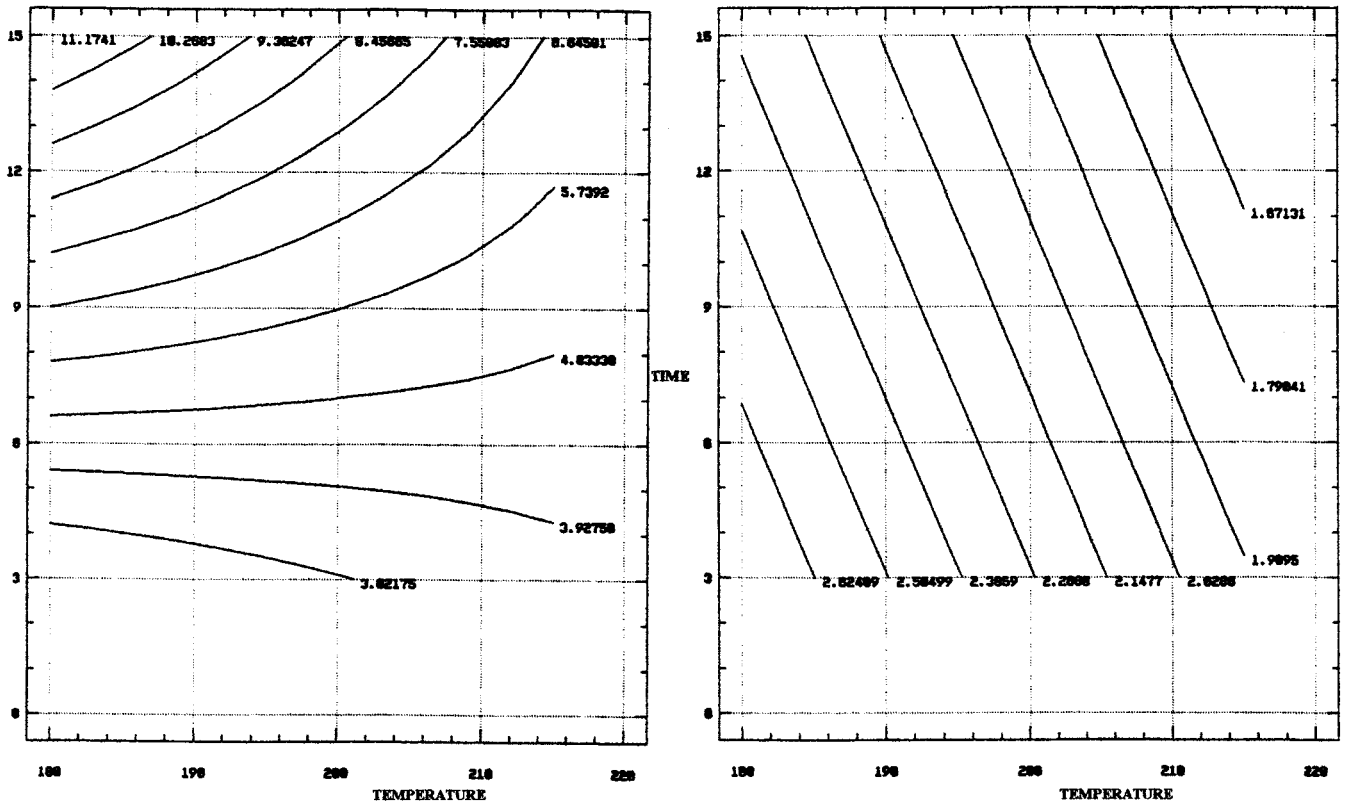

Figure 6. Contour diagrams for the gallic and syringic acid variables.

coefficients are positive, again with the exception of gallic acid for which they are negative (inversely proportional).

The most frequent interaction between factors is *AB* (t^a/charring time), which is negative in all of the significant models. The highest coefficient of all stands out as being syringaldehyde in Table 4, whereas the other interactions have hardly any influence, except in the case of protocatechuic acid in Table 5.

An interesting scheme can be achieved by tridimensional representation of the response variable according to the three factors studied. By way of example, Figure 5 shows the representation for gallic acid. These graphs show the variations in concentration of gallic acid and syringic acid when one variable is kept constant and the other two increase or decrease. The variations can clearly be observed throughout the tridimensional space.

As was seen in the analysis of coefficients, in general we can see that increase of charring temperature and time causes an increase in the concentration of the various phenolic compounds, with the exception of gallic acid, which reacts conversely. This graph also allows us to examine the quantitative variations and interactions between factors (syringic acid).

The responses to this model in the study zone are partially reflected by the surface response diagrams (Figure 6).

In summary, and in order to control the production of macerates, it is vitally important to gauge charring time and/or the interactions of charring time/temperature, with the exception of gallic acid, which is susceptible to charring temperature only. Apart from the latter exception, no well-fitting models were obtained for the phenolic compounds in which charring temperature was the predominant factor.

Finally, if we compare these results with the concentrations obtained in commercial oloroso wines (Table 1), it can be seen that the macerates aged for three months and prepared with shavings charred at 180 °C for 3 h are those that present phenolic compound concentrations most similar to those found in commercial samples. However, some compounds, such as gallic acid, syringaldehyde, and sinapaldehyde, are found in slightly higher concentrations in these macerates.

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