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Kinetics of browning onset in white wines: influence of principal redox-active polyphenols and impact on the reducing capacity

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Abstract

The browning capacity of white wines was studied, employing an accelerated test and samples produced and stored under identical conditions. Browning was approached from a kinetic point of view and efforts were focussed on the investigation of plausible correlations with major redox-active polyphenols, including substances with an *o*-diphenol feature, such as gallic acid, caftaric acid, 2-*S*-glutathionylcaftaric acid (GRP), caffeic acid, catechin, and epicatechin. Over a period of ten days, browning development was shown to obey zero-order kinetics from the third day of the treatment, and browning rate constants (k) varied from 15.3 to 74.5×10^{-3} day^{-1}. Regression analysis between k values and concentration of individual phenolics provided strong evidence that epicatechin is the principal browning agent ($r^2 = 0.8033$, $P < 0.01$). Furthermore, the monitoring of the reducing power (P_R), throughout treatments, indicated that increases in browning are accompanied by a commensurate decline in the reducing ability, raising concerns about the impact of browning reactions on the in vitro antioxidant properties of white wines.

Keywords: Antioxidant properties; Browning; Kinetics; *o*-Diphenols; Polyphenols; Reducing power; White wines

1. Introduction

There are two discrete processes of wine oxidation, the enzymic and the non-enzymic, and it is generally accepted that both ways proceed to some extent identically, irrespective of the initiation step. In any case, wine is capable of reacting with a considerable amount of oxygen, red rather more so than white, as the principal initial reactants are polyphenols. A certain degree of oxidation may be desirable in red wines as it can contribute towards a more stable colour, but quality of white wines is in many instances impaired by exposure to air. However, the phenomenon of browning is a rather more complicated procedure, since many other redox-active constituents may participate in the relevant reactions, including SO_2, ascorbic acid (if added), and transition metal ions (Danilewicz, 2003).

Abbreviations: AAE, ascorbic acid equivalents; CT, catechin; ECT, epicatechin; GRP, grape reaction product (2-*S*-glutathionylcaftaric acid); k, browning rate constant; P_R, reducing power; TRE, Trolox equivalents.

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The cascade of oxidative processes is initiated by the oxidation of catechol (*o*-diphenol) derivatives, such as gallic acid, caffeic acid and catechin, which are among the most readily oxidisable substrates in white wines. However, it has long been recognised (Rossi & Singleton, 1966) that not all phenolics yield derivatives of the same brownness upon oxidation and, despite the effort that has been expended on the investigation of caftaric acid involvement in browning reactions, since it is the most abundant white wine *o*-diphenol, there has been substantial evidence that oxidation of flavanols (catechins) may play a very prominent role in the development of brownish shades (Baron, Mayen, Merida, & Medina, 2000; Guyot, Vercauteren, & Cheynier, 1996; Oszmianski, Cheynier, & Moutounet, 1996).

In spite of the sensory attributes, which could be impacted significantly as a result of excessive browning onset, oxidation of principal polyphenolic compounds would presumably afford changes in the antioxidant status, as a consequence of changes in the redox equilibrium. Normally, one should expect oxidation of antioxidants to yield lower antioxidant capacity, but because oxidation of polyphenols may bring about formation of novel antioxidants (Makris & Rossiter, 2001), it would appear rather impossible to predict the antioxidant properties of wines which have developed browning. On the basis of this concept, the issue concerned with browning in white wines should be addressed as being essential, not only because of its influence on the organoleptic characters, but also due to its importance pertaining to the antioxidant potential. The present study was undertaken, first to examine browning development from a kinetic point of view and identify possible relevance to specific phenolics and, second, to evaluate browning in relation to its result on the reducing properties of white wines.

2. Materials and methods

2.1. Wines

Seven white, experimental wines were assayed, which were vinified in 2002. The cultivars used were Hellenic native *V. vinifera* species, whose origin may be seen in Table 1. Briefly, grapes were harvested at technological maturity, based on indices of sugar content and acidity established by the Vine and Wine Institute, destemmed, crushed and pressed. Musts were inoculated with a selected yeast strain (*S. cerevisiae* var. *cerevisiae*) and fermented at a controlled temperature (16–21 °C) to dryness (reducing sugar content $< 4\ g\,l^{-1}$). Following this, wines were bottled and stored at 15 ± 2 °C, in the dark.

Table 1
Origin of grapes used for the production of the experimental wines assayed

Sample No.	Variety	Origin	Location
S1	Chardonnay	Kavala	Eastern Macedonia (N)
S2	Athiri	Rhodes	Aegean Isles (S)
S3	Savvatiano	Attica	Sterea Ellada (C)
S4	Asyrtiko	Thessaloniki	Macedonia (N)
S5	Skiadopoulo	Zakynthos	Ionian Isles (S)
S6	Roditis	Magnesia	Peloponnese (S)
S7	Savvatiano	Attica	Sterea Ellada (C)

Letters N, C, and S denote Northern, Central and Southern Greece, respectively.

2.2. Chemicals

Catechin, epicatechin, caffeic acid and gallic acid were from Sigma Chemical Co. (St. Louis, MO, USA.). Citric acid and ferrous ammonium sulphate [$FeNH_4(SO_4)_2$] were from Merck (Darmstadt, Germany). Trichloroacetic acid (TCA) and ascorbic acid were from Panreac (Quimica S.A., Barcelona). 2,2′-Dipyridyl was from Acros Organics (Geel, Belgium).

2.3. Accelerated browning test and browning measurements

The model used to assess browning development was a modification of that described by Singleton and Kramling (1976). Wine lots of 20 ml were filtered through pharmaceutical cotton, placed in a 30 ml, screw-cap, glass vial and treated with 0.1 ml of trichloroacetic acid (TCA) solution to avoid microbial spoilage. The final concentration of TCA in the sample was $1\ mg\,l^{-1}$ (6.12 μM). Glass vials were of 7.5 cm length and 2.1 cm internal diameter, and the headspace over wine was of 2.7 cm length. Samples were subjected to heating at a constant temperature of 55.0 ± 0.2 °C in a water bath, in the dark. Aliquots were withdrawn at 24 h intervals over a period of ten days and browning (A_{420}) was measured against 12% EtOH in McIlvine buffer (pH 3.0). Samples were then immediately returned back in the vials, to maintain the initial headspace volume.

2.4. Determination of reducing power (P_R)

The reducing power of wines was estimated using the dipyridyl method, according to Psarra, Makris, Kallithraka, and Kefalas (2002), but with ascorbic acid as the calibration standard instead of quercetin (Arnous, Makris & Kefalas, 2001, 2002; Gil, Tomás-Barberán, Hess-Pierce, & Kader, 2002). Results were expressed as ascorbic acid equivalents (mM AAE). All samples were tested undiluted.

2.5. Determination of the analytical polyphenolic composition

The individual polyphenolic constituents were determined by HPLC. The chromatography apparatus used was an HP 1090, coupled with an Agilent 1100 diode array detector, and controlled by Agilent ChemStation software. The column was a Spherisorb ODS 2 (AnalyzenTechnik, MZ, Germany), 250 × 4 mm, 5 μm, protected by a guard column packed with the same material. Both columns were thermostatically controlled at 20 °C. The chromatographic conditions, as well as detection, identification and quantification of peaks were carried out as described previously (Makris, Psarra, Kallithraka, & Kefalas, 2003).

2.6. Statistics

All analyses were performed in triplicate ($n = 3$) and values were averaged and given along with the standard deviation (±S.D.), except for the accelerated browning test, which was carried out in duplicate. Browning rate constants (k) were determined from the slope of the regression line after plotting A_{420} as a function of time (days), and expressed as days^{-1}. Correlation between k values and concentrations of individual polyphenols was established by employing regression analysis at 99.9% significance level.

3. Results

The study of Singleton and Kramling (1976) on browning phenomena in white wines established a simple and reliable test for assessing browning capacity. The conditions employed permitted the examination of samples within a reasonable period of time, although the end-point was chosen arbitrarily, but it was suggested that the test period might be extended up to the point where maximum browning is attained. For reasons of obtaining an insight into the course of browning, increases in A_{420} were monitored continuously on a 24 h interval basis. It was observed, after an initial period of three days, where there was an increase, followed by a rapid decline, browning exhibited a constant onset up to ten days (Fig. 1). Therefore, in order to keep the examination period to a suitable minimum, day 10 was chosen as the end-point of the test.

The first notable observation was that browning actually exhibited a biphasic course and, after an period of three days, which was characterised by a rise and fall of browning (assigned as a in Fig. 1), A_{420} displayed a linear increase with time (assigned as b in Fig. 1), with the exception of sample S2, whose correlation coefficient was not satisfactory (Table 2), evidencing a deviation from linear course. A zero-order reaction model produced a good fit of the data:

$$A_{420} = A^0_{420} + kt,$$

where A_{420} may be considered as the concentration of brown product(s), A^0_{420} the initial concentration of brown product(s) (at the beginning of the linear increase, day 3), k the reaction rate constant (days^{-1}), and t time. The rate constants (k) were calculated graphically from the slope of the regression lines (Fig. 2) and given analytically in Table 2.

In order to identify any possible relevance of the reaction rates to important compositional factors, six prevalent, readily oxidisable phenolics were determined (Fig. 3), and correlation of the individual constituents with k values was performed, employing regression analysis. From the results (Table 3), it was shown that correlations with major hydroxycinnamates, including caftaric acid, 2-*S*-glutathionylcaftaric acid (GRP) and caffeic acid, but also gallic acid and catechin, were very low and statistically insignificant. Contrary to that, however, epicatechin appeared to be greatly involved in browning reactions, as indicated by the particularly high correlation of its concentration with k values ($r^2 = 0.8033$,

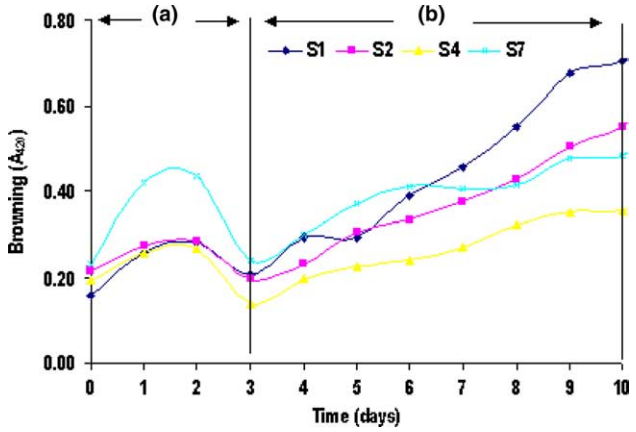

Fig. 1. Wine samples (S1, S2, S4, and S7) exhibiting typical browning course during incubation in a water bath at 55 °C, in the dark. Phase a is characterized by an increase, followed by a decline within the first three days. Phase b represents linear onset in A_{420} as a function of time for the seven remaining days of the treatment.

Table 2
Browning rate constants (k) calculated from the slope of the regression lines, obtained after plotting A_{420} as a function of time

Sample No.	k (day^{-1}) × 10^{-3}	r^2	Equation
S1	74.5	0.9735	$y = 0.0745x–0.0378$
S2	17.5	0.7762	$y = 0.0175x–0.0138$
S3	15.3	0.9627	$y = 0.0153x–0.1005$
S4	71.0	0.9715	$y = 0.0710x–0.0755$
S5	39.7	0.9674	$y = 0.0397x–0.2156$
S6	45.3	0.9406	$y = 0.0453x–0.0880$
S7	28.9	0.9593	$y = 0.0289x–0.0050$

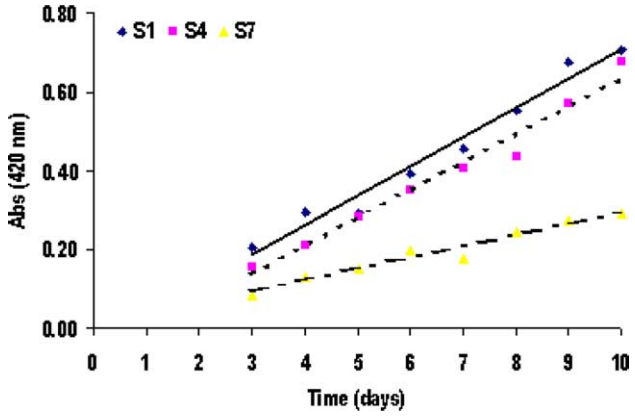

Fig. 2. Regression lines (samples S1, S4, and S7) calculated after plotting browning values (A_{420}) against time (days). The slope of the lines represents the zero-order browning rate constant (k).

$P < 0.01$). In order to obtain a deeper insight into the role of redox-active phytochemicals in browning reactions, the ferric-reducing ability of wines was monitored throughout treatments. The course of reducing power (P_R) gave a "mirror" image in relation to browning (A_{420}) (Fig. 4), suggesting that increases in browning are accompanied by commensurate decreases in the reducing status, but also that, upon browning diminution, the reducing capacity is restored.

4. Discussion

The browning capacity of white wines depends largely on the nature of white wine polyphenols, most of them being rather readily oxidised in regular winemaking processes, due to their catechol (*o*-diphenol) features in their structures. However, despite the fact that a considerable amount of work has been published on that issue, there are still many aspects to be investigated and quite many points to be clarified. Most of the information on the brown derivatives that may arise from oxidation of hydroxycinnamates, such as caffeic acid and flavanols, such as catechin, has been provided by the

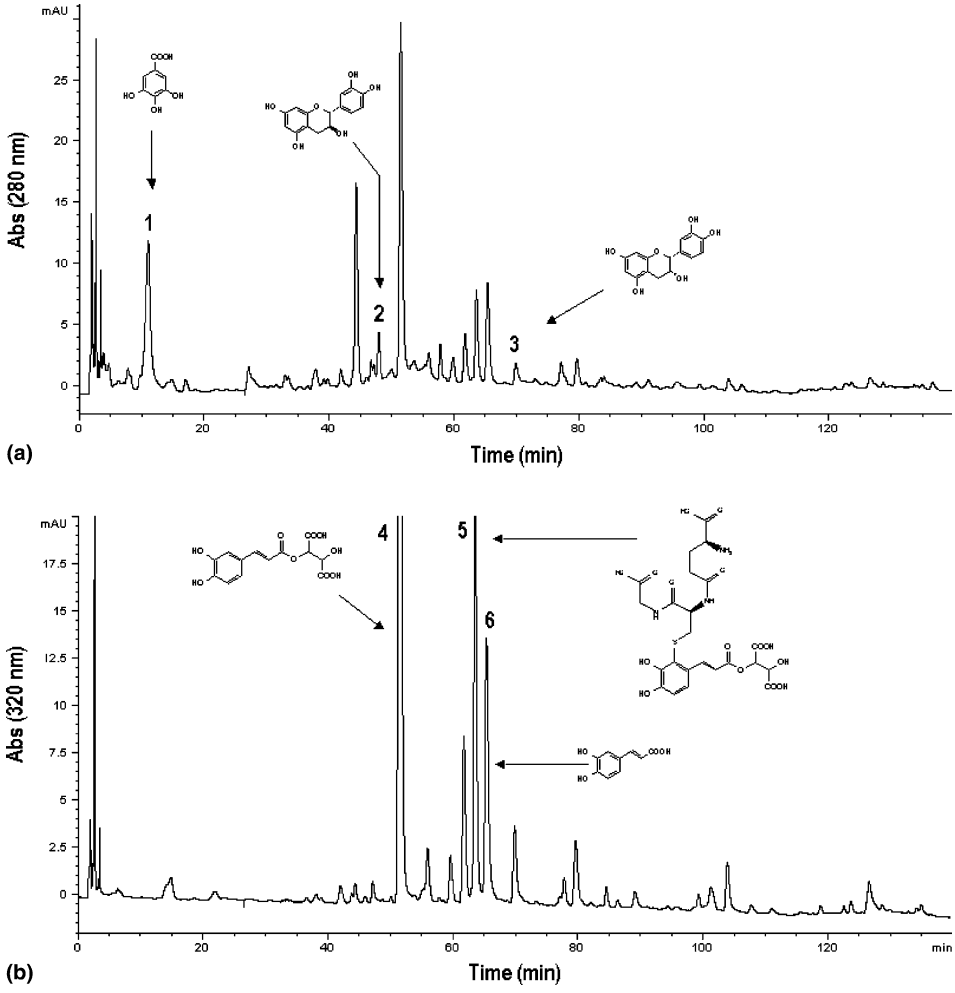

Fig. 3. Characteristic HPLC traces of the wines analysed, illustrating the composition of principal redox-active polyphenolic constituents (*o*-diphenols). Peak assignment: 1, gallic acid; 2, catechin; 3, epicatechin; 4, *trans*-caftaric acid; 5, 2-*S*-glutathionylcaftaric acid; and 6, caffeic acid.

Table 3
Statistical parameters calculated after regression analysis of browning rate constants (k) as a function of concentration of individual o-diphenols

Compound	r^2	P
CftA	0.0945	0.5024
GRP	0.0143	0.7982
CA	0.1439	0.4013
GA	0.1812	0.3409
CT	0.2874	0.2148
ECT	0.8033	0.0062*
Total o-Dp	0.0856	0.5244

* Value statistically significant ($P < 0.01$).

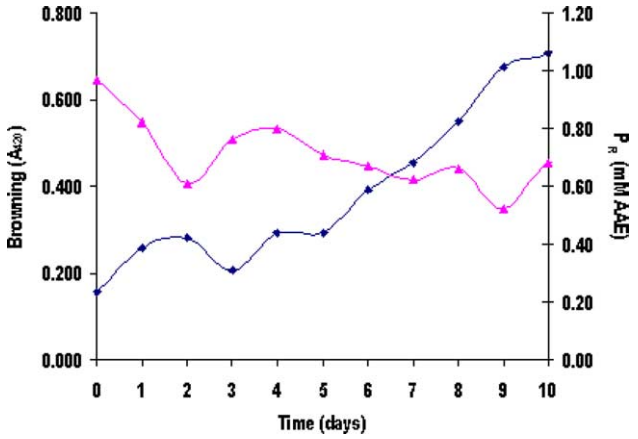

Fig. 4. Diagram illustrating the "mirror" course of reducing power (—▲—) in relation to the browning onset (—■—) (sample S1). Reducing power is expressed as ascorbic acid equivalents (mM AAE).

examination of model systems (Cilliers & Singleton, 1989; Clark, Prenzler, & Scollary, 2003; Oszmianski et al., 1996), whereas the nature of brownish substances in white wines is rather obscure. Furthermore, although important efforts have been made to achievie a reliable description of browning onset in terms of kinetics, there have been no conclusive data on this subject. In addition, the browning of white wines has been dealt with, so far, as a problem related to the aesthetic perception of this commodity, but its impact on the in vitro antioxidant capacity has been disregarded.

In the study presented herein, a well-established model system of accelerated browning was adopted to examine both kinetics of browning in wines produced and stored identically, the plausible involvement of principal redox-active polyphenols, and the consequences of such a process on the functional (antioxidant) properties of white wines. Because the whole investigation was based on an accelerated test, it should be emphasised that the increased temperature employed (55 °C) only reduces the time during which measurable browning changes may occur (Fernández-Zurbano et al., 1995), presumably by increasing the rate of the reaction(s) involved. Similar results for comparison of natural and electrochemically-induced browning have been reported (Palma & Barroso, 2002; Palma, Barroso, & Pérez-Bustamante, 2000). For this reason, the methodology proposed originally by Singleton and Kramling (1976) was modified, in that wines underwent no treatment with bentonite for protein removal, to maintain their original composition. Possible interactions of proteins with remaining sugars were shown to be without importance under similar conditions (Gonzales Cartagena, Pérez-Zúñiga, & Abad, 1994).

The kinetic browning behaviour was divided into two discrete phases (Fig. 1); phase A was characterised by a rapid increase and a consequent decrease within three days, and phase B by a fairly linear increase up to the 10th day. A similar trend for white wines was reported in an earlier study (Pérez-Zúñiga, Abad, & Cartagena, 2000), but a similar phenomenon was also seen during metal-catalysed, thermal degradation of rutin (Makris & Rossiter, 2000) and quercetin (Makris & Rossiter, 2002) in aqueous model systems. A hypothesis that may lie behind this observation is that the initial increase is due to quinone formation, following oxidation of caftaric acid, the most abundant oxidisable substrate, which could then be reduced either by SO_2 (Saucier & Waterhouse, 1999) or by coupled reactions with flavanols (Cheynier, Basire, & Rigaud, 1989; Cheynier & Ricardo da Silva, 1991), resulting in a temporary browning decline. Subsequent polymerisation of flavanol quinones could be responsible for the augmentation in browning observed thereafter (Guyot et al., 1996).

The increase of browning after the initial 3-day period could very efficiently be described in terms of zero-order kinetics, as A_{420} was found to increase linearly in relation to time (Fig. 2). Because browning is associated with polyphenol oxidation, it was conspicuous that oxidisable substrates, such as o-diphenols, could be implicated to a certain degree in the relevant mechanism(s). Indeed, by plotting the concentration of individual o-diphenols against browning rate constants, it was revealed that ECT is linked with browning development in a manner that cannot be overlooked:

$$k = 15.744 \times [\mathrm{ECT}] + 20.309 (r^2 = 0.8033, P < 0.01).$$

However, correlation of browning with other major parameters, in terms of concentration, o-diphenols were statistically insignificant (Table 3), evidencing possible relationships with specific structural features. In fact, the finding that ECT correlation with browning rate (k) was particularly significant could be explained theoretically by its higher oxidisability. Compared with CT, ECT exhibits lower oxidation potential (Yang, Kotani, Arai, & Kusu, 2001a), and it would normally be expected that, under identical conditions, ECT could be oxidised at a higher rate than CT. This claim is further corroborated by a number of studies on the

antioxidant potency of these two flavanols (Nakao, Takio, & Ono, 1998; Saint-Cricq de Gaulejac, Provost, & Vivas, 1999; Salah et al., 1995; Yang, Kotani, Arai, & Kusu, 2001b), which clearly demonstrate ECT to be a more efficient antioxidant in a wide spectrum of model systems. In support of this theory are the findings that demonstrated a negligible correlation of browning with hydroxycinnamates, but an important association with CT and ECT (Fernández-Zurbano et al., 1995).

In a similar fashion, ECT was shown to be highly associated with reducing power in white wines (Makris et al., 2003), despite its considerably lower concentration, as opposed to hydroxycinnamates, which were weakly associated with iron-reducing efficiency. ECT disappearance, following oxidation, would be very likely to afford a drop in the reducing effect and therefore the decrease in the reducing power that is observed throughout treatments (Fig. 4) could reflect ECT disappearance through oxidation. Presumably, excessive oxidation of other constituents with antioxidant potency could bring about further disturbance in the redox equilibrium, with rather negative effects on the antioxidant ability of white wines, and this issue merits a more profound examination.

5. Conclusions

The most important findings of the present study may be summarised as follows:

- Under the experimental conditions employed, after an initial period of three days, where browning exhibits fluctuations, there is a linear increase for at least ten days.
- The linear increase was found to obey zero-order kinetics.
- The browning rate constants calculated were significantly correlated with epicatechin concentration.
- Browning development affects the reducing ability of white wines, raising concerns for negative consequences on their antioxidant status.

References

Arnous, A., Makris, D. P., & Kefalas, P. (2001). Effect of principal polyphenolic components in relation to antioxidant characteristics of aged red wines. *Journal of Agricultural and Food Chemistry, 49*(12), 5736–5742.

Arnous, A., Makris, D. P., & Kefalas, P. (2002). Correlation of pigment and flavonol content with antioxidant properties in selected aged regional wines from Greece. *Journal of Food and Composition and Analysis, 15*(6), 655–665.

Baron, R., Mayen, M., Merida, J., & Medina, M. (2000). Comparative study of browning and flavan-3-ols during the storage of white sherry wines treated with different fining agents. *Journal of the Science of Food and Agriculture, 80*, 226–230.

Cheynier, V., Basire, N., & Rigaud, J. (1989). Mechanism of *trans*-caffeoyltartaric acid and catechin oxidation in model solutions containing grape polyphenoloxidase. *Journal of Agricultural and Food Chemistry, 37*, 1069–1071.

Cheynier, V., & Ricardo da Silva, J. M. (1991). Oxidation of grape procyanidins in model solutions containing *trans*-caffeoylcaftaric acid and polyphenol oxidase. *Journal of Agricultural and Food Chemistry, 39*, 1047–1049.

Cilliers, J. J. L., & Singleton, V. L. (1989). Non-enzymic autoxidative phenolic browning reactions in a caffeic acid model system. *Journal of Agricultural and Food Chemistry, 37*, 890–896.

Clark, A. C., Prenzler, P. D., & Scollary, G. R. (2003). The role of copper(II) in the bridging reactions of (+)-catechin by glyoxylic acid in a model white wine. *Journal of Agricultural and Food Chemistry, 51*, 6204–6210.

Danilewicz, J. C. (2003). Review of reaction mechanism of oxygen and proposed intermediate reduction products in wine: central role of iron and copper. *American Journal of Enology and Viticulture, 54*, 73–85.

Fernández-Zurbano, P., Ferreira, V., Peña, C., Escudero, A., Serrano, F., & Cacho, J. (1995). Prediction of oxidative browning in white wines as a function of their chemical composition. *Journal of Agricultural and Food Chemistry, 43*, 2813–2817.

Gil, M. I., Tomás-Barberán, F. A., Hess-Pierce, B., & Kader, A. A. (2002). Antioxidant capacities, phenolic compounds, carotenoids, and vitamin C contents of nectarine, peach, and plum cultivars from California. *Journal of Agricultural and Food Chemistry, 50*, 4952–4976.

Gonzales Cartagena, L., Pérez-Zúñiga, F. J., & Abad, F. B. (1994). Interactions of some environmental and chemical parameters affecting the color attributes of wine. *American Journal of Enology and Viticulture, 45*, 43–48.

Guyot, S., Vercauteren, J., & Cheynier, V. (1996). Structural determination of colourless and yellow dimers resulting from (+)-catechin coupling catalysed by grape polyphenoloxidase. *Phytochemistry, 42*, 1279–1288.

Makris, D. P., Psarra, E., Kallithraka, S., & Kefalas, P. (2003). The effect of polyphenolic composition as related to antioxidant capacity in white wines. *Food Research International, 36*, 805–814.

Makris, D. P., & Rossiter, J. T. (2000). Heat-induced, metal-catalyzed oxidative degradation of quercetin and rutin (quercetin 3-*O*-rhamnosylglucoside) in aqueous model systems. *Journal of Agricultural and Food Chemistry, 48*, 3830–3838.

Makris, D. P., & Rossiter, J. T. (2001). A comparison of quercetin and a non-orthohydroxy flavonol as antioxidants by competing in vitro oxidation reactions. *Journal of Agricultural and Food Chemistry, 49*, 3370–3377.

Makris, D. P., & Rossiter, J. T. (2002). Effect of natural antioxidants on heat-induced, copper(II)-catalysed, oxidative degradation of quercetin and rutin (quercetin 3-*O*-rutinoside) in aqueous model systems. *Journal of the Science of Food and Agriculture, 82*, 1147–1153.

Nakao, M., Takio, S., & Ono, K. (1998). Alkyl peroxyl radical-scavenging activity of catechins. *Phytochemistry, 49*, 2379–2382.

Oszmianski, J., Cheynier, V., & Moutounet, M. (1996). Iron-catalyzed oxidation of catechin in model systems. *Journal of Agricultural and Food Chemistry, 44*, 1712–1715.

Palma, M., & Barroso, C. G. (2002). Application of a new analytical method to determine the susceptibility of wine to browning. *European Food Research and Technology, 214*, 441–443.

Palma, M., Barroso, C. G., & Pérez-Bustamante, J. A. (2000). Evaluation of similarities between natural and accelerated browning of fino sherry wines by chemometric techniques. *The Analyst, 125*, 1151–1154.

Pérez-Zúñiga, F. J., Abad, F. B., & Cartagena, L. G. (2000). Kinetics of non-enzymatic oxidation reactions: browning in white wines. *European Food Research and Technology, 211*, 252–256.

Psarra, E., Makris, D. P., Kallithraka, S., & Kefalas, P. (2002). Evaluation of the antiradical and reducing properties of selected Greek white wines: correlation with polyphenolic composition. *Journal of the Science of Food and Agriculture, 82*, 1014–1020.

Rossi, J. A., & Singleton, V. L. (1966). Contributions of grape phenols to oxygen absorption and browning of wines. *American Journal of Enology and Viticulture, 17*, 231–239.

Saint-Cricq de Gaulejac, N., Provost, C., & Vivas, N. (1999). Comparative study of polyphenol scavenging activities assessed by different methods. *Journal of Agricultural and Food Chemistry, 47*, 425–431.

Salah, N., Miller, N. J., Paganga, G., Tijburg, L., Bolwell, G. P., & Rice-Evans, C. A. (1995). Polyphenolic flavanols as scavengers of aqueous phase radicals and as chain-breaking antioxidants. *Archives of Biochemistry and Biophysics*, 339–346.

Saucier, C. T., & Waterhouse, A. L. (1999). Synergistic activity of catechin and other antioxidants. *Journal of Agricultural and Food Chemistry, 47*, 4491–4494.

Singleton, V. L., & Kramling, T. E. (1976). Browning of white wines and an accelerated test for browning capacity. *American Journal of Enology and Viticulture, 27*, 157–160.

Yang, B., Kotani, A., Arai, K., & Kusu, F. (2001a). Estimation of the antioxidant activities of flavonoids from their oxidation potentials. *Analytical Sciences, 17*, 599–604.

Yang, B., Kotani, A., Arai, K., & Kusu, F. (2001b). Relationship of electrochemical oxidation of catechin on their antioxidant activity in microsomal lipid peroxidation. *Chemical and Pharmaceutical Bulletin, 49*, 747–751.