# Impact of the contact time of different oak wood chips on red wine phenolic composition evolution after bottling

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**Abstract.** The main object of the present work was to evaluate the potential influence of the oak wood chips-wines contact time (30 and 60 days) on the evolution of the red wine phenolic composition during storage in bottle. Thus, global phenolic composition, color parameters, and individual anthocyanins of bottled red wines that had previously been in contact with oak wood chips during different times were analyzed. The results obtained demonstrates that in general, after 6 months of bottle storage, red wines with a previous oak wood chips contact time showed a more evident decrease on anthocyanin content, independently of the oak wood chips species used and toasting level. This tendency was also confirmed by the decrease in the values obtained for color intensity and $a^*$ (redness) CIELab coordinate value. However, a positive impact of oak wood chips contact time on wine hue color and $b^*$ (yellowness) CIELab coordinate values, was detected. Thus, after 6 months of bottle storage, red wines that were in a previous contact with oak wood chips (particularly during 60 aging days), exhibited lower color hue and $b^*$ values compared with control wine (without any oak wood chips contact).

## 1. Introduction

The European Union authorized the use of oak wood pieces in winemaking since 2006 and now the EEC Regulation No. 606/2009 of 10 July 2009 (appendix 9), modified previous rules by regulating the use of pieces of wood solely from *Quercus* generous in wine production. Thus, the addition of oak chips has been generalized in order to obtain wines with wood characteristics similar to wines aged in barrels for several months. Currently, a great variety of oak wood pieces can be found on the market, with different toasting levels, particle size, forms and produced particularly from three main oak wood species: *Q. alba* (from USA), *Q. robur* and *Q. petraea*. These last two oak wood species are traditionally obtained from French forests. However, there are other oak wood species, such as, *Q. pyrenaica* and *Q. pubescens*, that are found in other European countries.

The majority of the published works studied the oak wood composition, particularly volatile [1] and phenolic composition [2], and also the impact of oak wood chips on red wine phenolic composition by conducting analysis immediately after removal of oak wood chips from wine [3–5]. However, less attention has been directed to the potential influence of the chips-wines contact on the further phenolic composition and color properties evolution during red wine storage in bottle. In addition, it is important to note that phenolic compounds are very important for wine quality, since they are responsible for most of the red wine sensory characteristics, particularly color, bitterness and astringency.

In this context, in order to deepen the knowledge about this topic, the present study aims to evaluate several global phenolic parameters, color characteristics, and individual anthocyanins from bottled red wines that had previously been in contact with two oak wood chip species (*Q. petraea* and *Q. pubescens*) during two different contact times (30 and 60 aging days).

## 2. Material and methods

### 2.1. Red wine, oak wood chips and experimental conditions

The red wine used in this experiment was elaborated from a Portuguese *Vitis vinifiera* grape variety "Touriga Nacional" cultivated in *Dão* wine region and produced during the vintage of 2017. The initial main general physico-chemical characteristics of the red wine used were the following: alcohol degree 13%, v/v 20 °C; pH 3.58; total acidity 4.0 g/L tartaric acid; volatile acidity 0.42 g/L acetic acid; sugar 1.9 g/L and malic acid 1.0 g/L.

The experimental work was performed at laboratory scale (10 liters for each essay) using oak wood chips from two different oak species: *Quercus petraea* from France and *Quercus pubescens* from Serbia. The oak wood chips

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**Table 1.** Experimental aging conditions and identifying wine codes used in the work.

| Wine codes | Experimental aging conditions |
|---|---|
| C | Control wine (without any oak wood chips contact) |
| OFL | Wine + *Q. petraea* wood chips with light toasting |
| OSL | Wine + *Q. pubescens* wood chips with light toasting |
| OFM | Wine + *Q. petraea* wood chips with medium toasting |
| OSM | Wine + *Q. pubescens* wood chips with medium toasting |
| OFF | Wine + *Q. petraea* wood chips with strong toasting |
| OSF | wine + *Q. pubescens* wood chips with strong toasting |

used showed also three different totasting levels: light, medium and strong. The oak wood chips concentration used was 2 g/L and with an average particle size of 8 mm. For the experimental essay two different contact times between oak wood chips and red wine were considered: 30 and 60 days. After these two different oak wood chips contact times, the wines were analysed, after which it were bottled and characterized after 6 months of botlle storage. All wines were storaged at cellar temperature (between 14–15 °C). Finnally, all essays were made in duplicate. The different experimental aging conditions are listed in Table 1.

### 2.2. Methods

Total polyphenolic content was determined according to Ribéreau-Gayon et al. [6], while flavonoid and non flavonoid phenols were determined using the method described by Kramling and Singleton [7]. For these parameters, the results were expressed as gallic acid equivalents by means of calibration curves. Total anthocyanins, and also total and polymeric pigments were quantified according to the method described by Somers and Evans [8]. Color parameters using the CIELab method, color intensity and color hue were also analyzed [9]. Finally, individual monomeric anthocyanins by HPLC-DAD were quantified using the methodology developed previously by Dallas and Laureano [10]. For total and individual anthocyanins the results were expressed as malvidin-3-monoglucoside equivalents by means of calibration curves established previously. All laboratory measurements were performed in triplicate.

### 2.3. Statistical analysis

The data are presented as mean ± standard deviation. To determine whether there is a statistically significant difference between the data obtained for the diverse parameters quantified an analysis of variance (ANOVA, one-way) and comparison of treatment means were carried out using the Tukey test ($p < 0.05$). This analysis was performed using SPSS software (version 25.0).

## 3. Results and discussion

### 3.1. General phenolic composition

Figure 1 shows the results obtained for global phenolic composition (total phenols, flavonoid and non flavonoid phenols) quantified in the red wines aged in contact with different oak wood chips during 30 and 60 days and after 6 months of bottle storage.

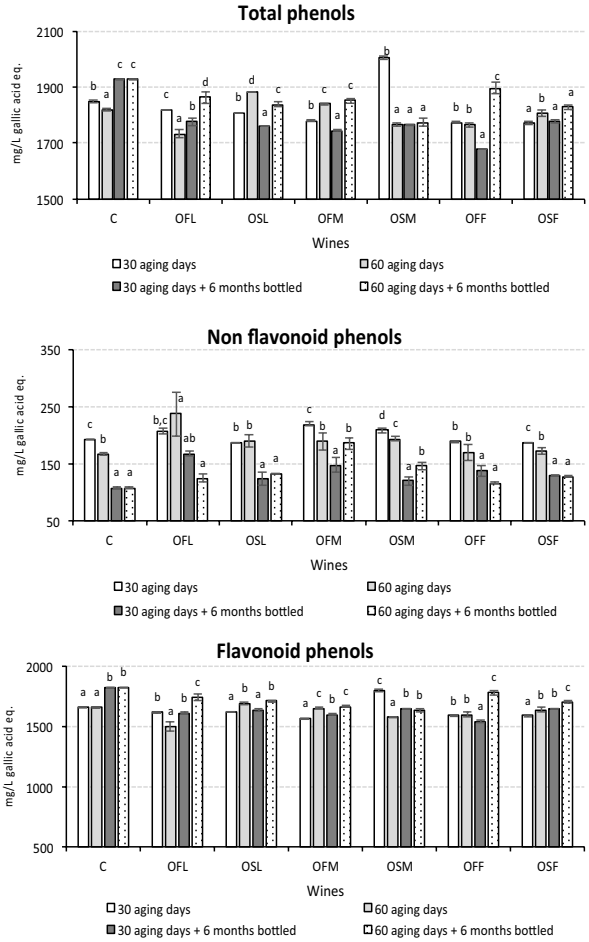


**Figure 1.** Global phenolic composition from red wines aged in contact with different oak wood chips (30 and 60 days) and after 6 months of bottle storage.

Legend: 30 or 60 aging days – wines aged during 30 or 60 days in contact with oak wood chips (except for control wine); wines codes *see* Table 1. Data points derived for each red wine with different aging time showing the same letter are not significantly different ($p < 0.05$).

The results concerning the effect of the contact time between red wine and different oak wood chip species with diverse toasting levels on the total phenolic content were not totally evident for the majority of red wines studied, particularly after 6 months of bottle storage. However, it was clear that control wine (without oak wood chips contact) showed the highest total phenol content, after 6 months of bottle storage. In addition, the red wines aged in contact with oak wood chips during 60 aging days, showed a slight tendency for a higher total phenols after 6 months of bottle storage, compared to wines aged previously in shorter contact with oak wood chips (30 aging days). It is also important to note that the oak wood chips concentration used in our study (2 g/L), although commonly used by winemakers, is much lower than the majority of the published works, where oak chips were used. In general, the concentrations used varied from 4 to 40 g/L [3,11,12]. In addition, a more extent bottle storage time could improve our understanding about the potential impact of the different oak wood chips and contact time on future evolution of total phenolic content of red wines.

For non flavonoid phenols an evident decrease of the values was detected in all red wines. This decrease was

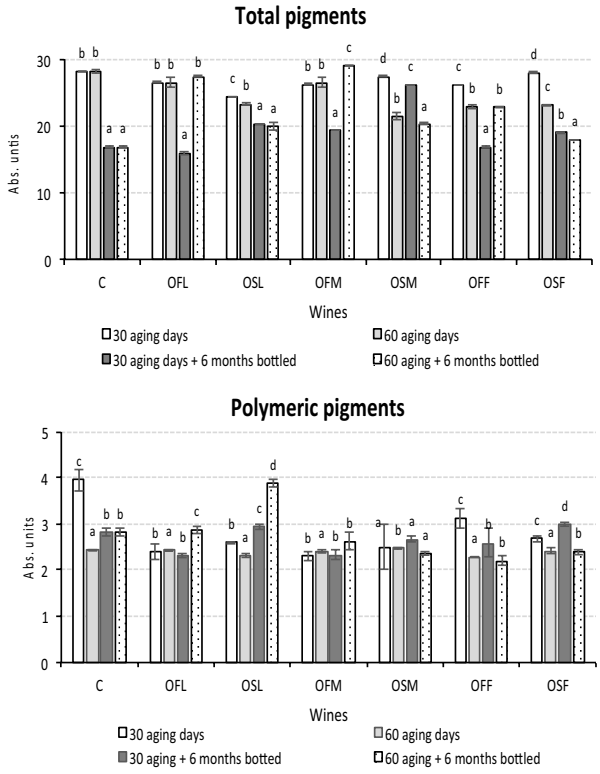


**Figure 2.** Total and polymeric pigments from red wines aged in contact with different oak wood chips (30 and 60 days) and after 6 months of bottle storage.

Legend: 30 or 60 aging days – wines aged during 30 or 60 days in contact with oak wood chips (except for control wine); wines codes *see* Table 1. Data points derived for each red wine with different aging time showing the same letter are not significantly different ($p < 0.05$).

more evident for the control wine, where no oak wood chips contact occurred before bottling. Furthermore, red wines with oak wood chips contact showed highest non flavonoid phenols values in all data points considered. These results could be explained by the fact that higher potential extraction of several individual non flavonoid compounds from oak wood chips to the wine occurred, namely, gallic, protocatechuic, vanillic, caffeic, syringic and *p*-coumaric acids, ellagitannins and ellagic acid. However, it was not totally evident that the contact time with the different oak wood chips had influenced the contents of these compounds found in wines after 6 months of bottle storage.

Finally, with respect to the evolution of flavonoid phenols, in general, it was evident that for the same previous contact time between the wines and the different oak wood chips, the wines maintained similar values after 6 months of bottle storage. In addition, control wine without any oak wood chips contact, showed highest values compared with the wines aged with oak wood chips.

For total and polymeric pigments evolution, the results are shown in Fig. 2. The results obtained for total pigments showed in general, a tendency for a decrease of the values in all red wines, including control wine. However, an exception occurred for the wines previously aged in contact with French oak wood chips during 60 aging days, where after 6 months of bottle storage, there was a tendency for an increase (OFM wine) or stabilization

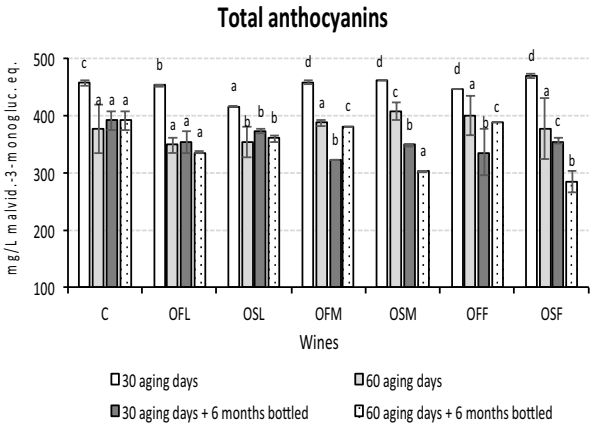


**Figure 3.** Total anthocyanins from red wines aged in contact with different oak wood chips (30 and 60 aging days) and after 6 months of bottle storage.

Legend: 30 or 60 aging days – wines aged during 30 or 60 days in contact with oak wood chips (except for control wine); wines codes *see* Table 1. Data points derived for each red wine with different aging time showing the same letter are not significantly different ($p < 0.05$).

(OFL and OFF wines) of total pigments values. Polymeric pigments evolution was characterized for the majority of the red wines by an increase of the values after 6 months of bottle storage, particularly for the wines with a previous oak wood chips contact during 60 aging days (OSL, OFL and OFM wines) or 30 aging days (OSF wine). In this case the results obtained allow us to consider that the previous oak wood chips contact could induced a potential faster evolution of wine phenolic compounds and consequently a development of derived polymeric compounds. This trend is in agreement with the results reported previously by other authors for red wines [5,13]. However, for two red wines (OFF and OSM wines) a clear tendency in polymeric pigments evolution was not evident. Probably the use of a more extended bottle storage time may make clearer the evolution of polymeric pigment values in wines.

### 3.2. Anthocyanin content and color parameters

The results obtained for total anthocyanins evolution from the different red wines are shows in Fig. 3. Thus, during the aging time considered in contact with different oak wood chips and bottle storage, a tendency for a decrease of total anthocyanin content in all red wines (including for control wine) was possible to detect. The total anthocyanin decrease detected was probably due to anthocyanin condensation and polymerization reactions, and the precipitation of these compounds during the aging process [3]. However, this decrease was more evident for all red wines that had a previous contact with oak wood chips before bottling. In addition, it was not evident that the wine contact time with the different oak wood chips species (30 or 60 aging days) has decisively determined the total anthocyanin content found in red wines after 6 months of bottle storage.

After 6 months of bottle storage, control wine (C wine code), wines aged previously in contact during 60 days with French oak wood chips with medium and strong toasting (OFM and OFF wines code) showed the highest average values of total anthocyanins (391.1, 379.4

**Table 2.** Individual monomeric anthocyanins content from red wines after 6 months of bottle storage and previously aged during 60 days in contact with different oak wood chips.

| Individual monomeric anthocyanins (mg/L)[(1)] | Wines[(2)] | | | | | | |
|---|---|---|---|---|---|---|---|
| | C | OFL | OSL | OFM | OSM | OFF | OSF |
| Del-3-monoglucoside | $3.59 \pm 0.02^{a}$ | $4.10 \pm 0.05^{b}$ | $3.54 \pm 0.22^{a}$ | $3.21 \pm 0.40^{a}$ | $2.43 \pm 0.34^{c}$ | $4.44 \pm 0.04^{b}$ | $3.96 \pm 0.07^{d}$ |
| Cya-3-monoglucoside | $8.46 \pm 0.09^{a}$ | $7.82 \pm 0.17^{b}$ | $6.65 \pm 0.12^{c}$ | $8.02 \pm 0.27^{a}$ | $5.26 \pm 0.39^{d}$ | $9.53 \pm 0.38^{e}$ | $7.24 \pm 0.78^{b}$ |
| Pet-3-monoglucoside | $14.88 \pm 0.15^{a}$ | $13.01 \pm 0.30^{b}$ | $11.88 \pm 0.39^{c}$ | $13.86 \pm 1.27^{b}$ | $8.7 \pm 0.23^{d}$ | $15.86 \pm 0.09^{e}$ | $11.92 \pm 0.03^{c}$ |
| Peo-3-monoglucoside | $7.5 \pm 0.14^{a}$ | $6.36 \pm 0.11^{cd}$ | $6.33 \pm 0.51^{d}$ | $6.57 \pm 0.85^{d}$ | $4.68 \pm 0.03^{b}$ | $8.17 \pm 0.09^{e}$ | $6.03 \pm 0.39^{d}$ |
| Malv-3-monoglucoside | $265.3 \pm 5.0^{a}$ | $230.3 \pm 1.7^{b}$ | $218.8 \pm 1.1^{c}$ | $241.8 \pm 5.2^{d}$ | $187.3 \pm 2.4^{e}$ | $255.2 \pm 6.1^{d}$ | $190.5 \pm 2.2^{e}$ |
| Cya-3- acetylglucoside | $9.41 \pm 0.69^{ab}$ | $9.82 \pm 0.60^{ab}$ | $9.35 \pm 0.13^{ab}$ | $10.39 \pm 0.12^{b}$ | $9.66 \pm 0.31^{b}$ | $10.58 \pm 0.03^{ab}$ | $10.41 \pm 0.47^{a}$ |
| Pet-3-acetylglucoside | $1.57 \pm 0.09^{a}$ | $1.94 \pm 0.04^{b}$ | $1.62 \pm 0.09^{a}$ | $1.86 \pm 0.04^{b}$ | $1.81 \pm 0.03^{b}$ | $1.92 \pm 0.24^{b}$ | $1.93 \pm 0.06^{b}$ |
| Peo-3-acetylglucoside | $4.48 \pm 0.04^{a}$ | $4.54 \pm 0.02^{a}$ | $4.42 \pm 0.02^{b}$ | $4.86 \pm 0.03^{c}$ | $0.91 \pm 0.06^{d}$ | $5.07 \pm 0.10^{e}$ | $4.46 \pm 0.02^{ab}$ |
| Del-3-acetylglucoside | $1.31 \pm 0.07^{a}$ | $1.09 \pm 0.02^{b}$ | $1.08 \pm 0.05^{b}$ | $1.29 \pm 0.02^{a}$ | $3.53 \pm 1.20^{c}$ | $1.46 \pm 0.07^{d}$ | $1.04 \pm 0.01^{b}$ |
| Malv-3- acetylglucoside | $22.65 \pm 0.49^{a}$ | $20.96 \pm 0.20^{d}$ | $18.58 \pm 0.06^{cd}$ | $22.42 \pm 0.21^{cd}$ | $13.16 \pm 0.03^{bc}$ | $25.06 \pm 0.10^{b}$ | $18.49 \pm 0.44^{a}$ |
| Pet-3-*p*-coumarylglucoside | $1.12 \pm 0.03^{a}$ | $0.92 \pm 0.04^{b}$ | $0.98 \pm 0.31^{b}$ | $1.05 \pm 0.03^{c}$ | $0.77 \pm 0.21^{d}$ | $1.23 \pm 0.07^{a}$ | $0.92 \pm 0.04^{b}$ |
| Peo-3-*p*-coumarylglucoside | $1.75 \pm 0.10^{a}$ | $1.47 \pm 0.01^{a}$ | $1.39 \pm 0.04^{a}$ | $1.64 \pm 0.02^{a}$ | $0.99 \pm 0.01^{a}$ | $1.93 \pm 0.04^{a}$ | $1.40 \pm 0.02^{a}$ |
| Malv-3-*p*-coumarylglucoside | $14.0 \pm 0.05^{a}$ | $12.13 \pm 0.14^{c}$ | $10.30 \pm 0.22^{c}$ | $13.15 \pm 0.02^{c}$ | $7.38 \pm 0.39^{b}$ | $15.29 \pm 0.10^{a}$ | $10.66 \pm 0.01^{c}$ |

[(1)] Individual monomeric anthocyanins expressed as malvidin-3-monoglucoside equivalents; [(2)] wines codes *see* Table 1; data points derived for each individual monomeric anthocyanin from different wines showing the same letter are not significantly different (Tukey, $p < 0.05$); ± standard deviation.

and 386.7 mg/L of malvidin-3-monoglucoside equivalents, respectively), while the lowest value (284.9 mg/L of malvidin-3-monoglucoside equivalents) was quantified in red wine aged previously in contact during 60 days with Serbian oak wood chips with strong toasting (OSF wine code). Several authors reported a more evident decrease of total anthocyanins in red wines aged in contact with oak wood chips [3,5]. However, the majority of these published works determined total anthocyanins during the red wine aging in contact with oak wood chips and not after several months of bottle storage. Thus, according to the results obtained in this research for total anthocyanins, the impact of the use of oak wood chips on wine aging may also have a potential impact on the decrease of these pigments even after removal of the chips and during wine bottle storage. In addition, it was also evident that this decrease was particularly evident for red wines previously aged in contact with oak wood chips with medium or strong toasting levels. However, the results were independent of the oak wood chip species used and wood chips-wine contact.

Table 2 shows the individual monomeric anthocyanins quantified in all red wines after 6 months of bottle storage and previously aged during 60 days in contact with different oak wood chips. As expected, monoglucosides derivatives forms were the major quantitiative anthocyanin group detected in all wines. This anthocyanin group are stable molecules and their presence gives stability to the red wines, because these compounds are relatively resistant to oxidant process.

In general, the results for individual monomeric anthocyanins are in line with the results obtained for the levels of total anthocyanin concentration, where the higest values were detected for control wine, where no oak wood chips contact occured.

Several authors reported that wines aged without oak wood contact showed less promounced changes in anthocyanin content than wines aged in contact with oak chips or in oak barrels as a consequence of the lower level

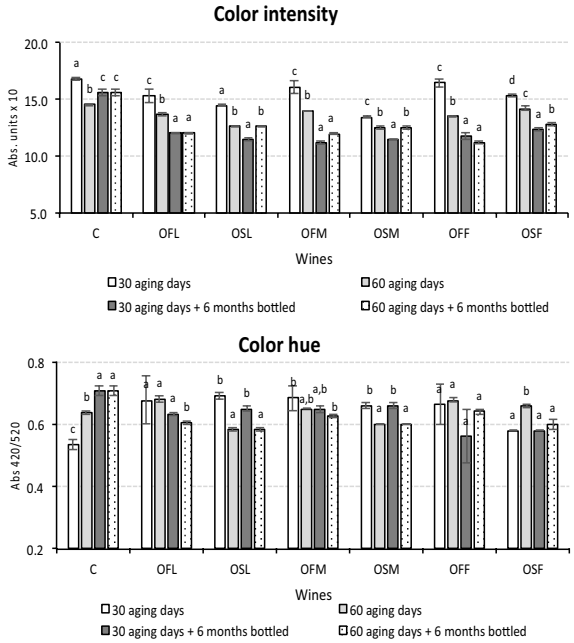


**Figure 4.** Color intensity and hue from red wines aged in contact with different oak wood chips (30 and 60 aging days) and after 6 months of bottle storage.

Legend: 30 or 60 aging days – wines aged during 30 or 60 days in contact with oak wood chips (except for control wine); wines codes *see* Table 1. Data points derived for each red wine with different aging time showing the same letter are not significantly different ($p < 0.05$).

of tannin condensation [14,15]. Jordão et al. [16] reported in model wine solutions that individual anthocyanins, namely malvidin-3-monoglucoside, declined more quickly in presence of oak wood extracts than in their absence, with a subsequent decrease in red color.

The effects of oak wood chips contact time on color intensity and color hue evolution is shown in Fig. 4. Thus, for color intensity it can be observed that during the

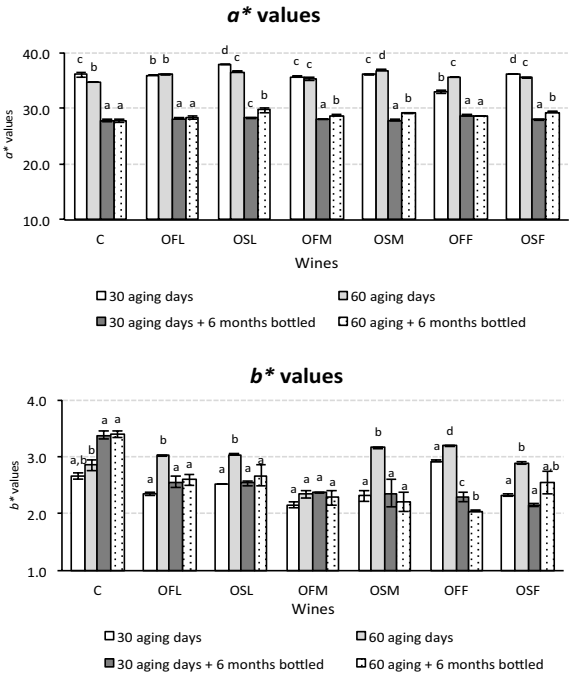


**Figure 5.** Chromatic characteristics by $a^*$ (redness) and $b^*$ (yellowness) coordinates from red wines aged in contact with different oak wood chips (30 and 60 aging days) and after 6 months of bottle storage.

Legend: 30 or 60 aging days – wines aged during 30 or 60 days in contact with oak wood chips (except for control wine); wines codes *see* Table 1. Data points derived for each red wine with different aging time showing the same letter are not significantly different ($p < 0.05$).

aging time considered, a tendency for a decrease of the values in all red wines was detected. However, the color intensity decrease was much more evident for all red wines with a previous oak wood chips contacts, independently of oak wood chips species and wood chips wine contact time. After 6 months of bottled storage, red wines with a previous oak wood chips contact showed the lowest color intensity values, particularly for the red wines with a previous oak wood chips contact during 30 aging days (except for OFL and OFF wines).

For chromatic characteristics using the $a^*$ (redness) and $b^*$ (yellowness) CIELab coordinates, the results are shown in Fig. 5. In general, regarding to $a^*$ values a significantly decrease of the values was detected as a consequence of the oak wood chips contact during the aging process. After 6 months of bottle storage, these results were independently of the oak wood chips contact time used and corresponding to a general decline of the red color that was also observed for color intensity values. In addition, similar $a^*$ values decrease was also detected for control wine. Finally for $b^*$ values (yellowness), a previous larger oak wood chips contact time (60 aging days) induced an increase of the $b^*$ values. However, after 6 months of bottle storage all red wines with a previous oak wood chips contact showed a decrease of the $b^*$ values, which corresponding to a decrease of the yellow color. This tendency is in line with the results obtained for color hue (Fig. 4), where after 6 months of bottled storage, a general decrease of the values was detected in all red wines previously aged in contact with oak wood chips. Finally, as was observed for the color hue, the control wine (C wine code) showed a tendency for a continuous increase of the $b^*$ values, which is corresponding to an undesirable increase of yellow color.

It is important to note that oak wood its an important source of hydrolyzable tannins, especially ellagitannins. These compounds can function as oxidation regulators quickly reacting dissolved oxygen and facilitating the hydroperoxidation of wine constituents. In addition, they limit also the oxidation of wine phenolic compounds, preventing the development of yellow color during the bottle storage. Thus, this may explain the reduction of yellow tones in red wines after 6 months of bottle storage, when a previous aok wood chips contact occurred.

## 4. Final remarks

This study identified the potential impact of the contact time of different oak wood chips on red wine phenolic composition after a bottle storage period.

In general, red wines aged with a previous oak wood chips contact showed after 6 months of bottle storage, a decrease of anthocyanins content and color intensity (corresponding to a red color decrease), but a higher resistance to the color degradation as a consequence of lower color hue and $b^*$ values (yellowness) compared with the control wine. In addition, the oak wood chips contact also induced a slight increment of total pigments quantified in all red wines after 6 months of bottle storage. We hope that further studies will be able to elucidate more specific impacts of the oak wood chips on red wine characteristics after bottling and try to explain also the potential impact on red wine sensory properties.

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