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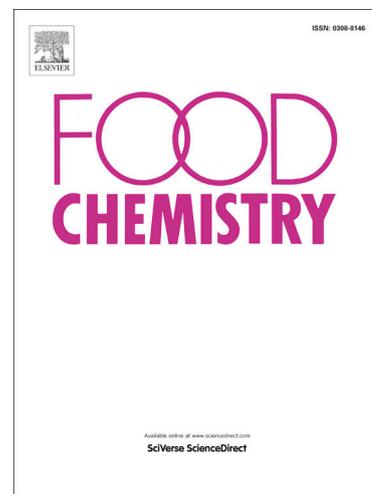
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## **Oxygen-induced faults in bottled white wine: A review of technological and chemical characteristics**

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**Abstract**

Several changes can take place in wine after blotting. Some of them lead to the desired evolution of wine being more complex, round and pleasant. However, unexpected changes can also occur ascribable to the premature wine oxidation (PremOx) arising when a wine, presumably with aging potential, results oxidized and often undrinkable. The complexity of PremOx, where aromas are also involved, makes difficult to identify all the oxidation products, and to predict its occurrence in wines. Despite most studies have been focused on the effect of time after wine bottling on PremOx as well as pinking phenomena, identification of pinking markers, reliable methods for their detection in wine, and correlations between markers and the wine-bottle-closure system are still unknown. This review aimed to highlight aspects PremOx-related, including wine-bottle-closure system, color change, with particular emphasis on pinking, and aroma decay based on the current knowledge becoming the bases for future perspectives.

Keywords: premature wine oxidation; markers; bottle-closure system; color; aroma compounds; decay.

## 1. Introduction

The phenomenon of premature oxidation (PremOx) occurs when a wine presumably with aging potential is expected to be in good condition, yet it is found to be oxidized and often undrinkable (Romanini, Colangelo, Lucini, & Lambri, 2019) with oxidative aroma degradation that has impact especially in young dry white wines (Silva Ferreira, Hogg, & Guedes de Pinho, 2003). The sensory characterization of PremOx is varied and controversial, including a premature loss of varietal aroma (Escudero, Cacho, & Ferreira, 2000) combined with an increase in off-flavors (Pons et al., 2015; Freitas et al., 2018). In the within of PremOx phenomena, the pinking phenomenon may be observed after bottling and storage of white wines and explained in terms of susceptibility of reductively produced wines to oxygen (Andrea-Silva et al., 2014). The oxygen trapped in the headspace of the bottle, in addition to the amount of oxygen dissolved in the wine and to the oxygen that permeates through the closure combined with temperature and light, can modify the oxidative status of the wine during storage (Godden et al., 2001). A step-by-step investigation of the mechanisms involved in the chemical oxidation processes, using simple model systems, has been underway for at least three decades, with emphasis on phenolic compounds, especially flavonoids, their subsequent polymers (Karbowski et al., 2010), and products from the conversion of flavenes that can cause pinking (Andrea-Silva et al., 2014). Such phenomena are complex and involve the aroma compounds leading to several oxidation products difficult to identify as a whole. As a consequence, the occurrence of PremOx and Pinking is hardly predictable in wine, especially in the white wines (Romanini et al., 2019). Since conditions such as oxidation-reduction reactions, temperature, oxygen, pH and light act as a whole on wine oxidation rate, this paper has been focused on certain issues which play a significant role along the wine supply chain and comprise (i) wine-bottle-closure system, (ii) modification of color and (iii) aroma decay.

## 2. Wine-bottle-closure system

The Wine-Bottle-Closure system includes all the chemical variables of the wine which are involved in the related oxidation phenomena as sulfur dioxide, volatile acidity, acetaldehyde, color, phenolics, and aroma, as well as stopper's properties, headspace and bottleneck, along with bottling modalities (Riboulet & Alegoet, 1986).

In this within, bottling operation could lead to large amounts of oxygen entering wine. Besides bottling, other operations being both static (ageing in tanks, in barrels or in bottle) or dynamic (pumping, filtering and cold stabilization) can increase the concentration of oxygen dissolved in wine. Focusing the attention of bottling that is a source of oxygen enrichment, the degree of oxygen addition has to be controlled. The exact amount of oxygen added appears to be dependent on the operating system of the bottling line. Indeed, at the beginning and the end of the bottling process, higher amount of oxygen is added to a wine containing less than 0.1 mg/L initial dissolved oxygen. The oxygen content in the final bottles could also be due to turbulence and contact with air at the end of the bottling cycle. In both cases, the use of an inert gas, such as nitrogen, could lead to a considerable decrease of oxygen addition during bottling, especially for bottling small wine volumes. As bottling is a source of oxygen enrichment, the degree of oxygen addition has to be controlled. The empty bottle itself contains 750 mL of air, or over 200 mg oxygen, which is mixed with wine during bottling under turbulent conditions, unless the bottle has previously been flushed with an inert gas such as nitrogen. Depending on bottling procedure, the approximately 3.5 mL of headspace in a cork closed bottle may contain from 0.2% to 7% of oxygen, which could potentially increase the dissolved oxygen in a 750-mL bottle of wine by 0.47 mg/L (Reeves, 2009). The headspace oxygen is then mixed with wine during bottling under turbulent conditions, unless the bottle has previously been flushed with an inert gas, such as nitrogen. During the corking operation itself, the closure goes down into the bottleneck, compressing the headspace atmosphere between the closure and the liquid. If no vacuum is drawn during

the corking operation, this displacement creates a potential increase of oxygen dissolution into wine and could lead to a four-fold increase of oxygen uptake (Karbowski et al., 2010). Once closed, the bottle contains oxygen from the original bulk wine, the headspace oxygen remaining after any gas flushing or vacuum application at the time of closure application, and any oxygen within a cellular structured closure if such was used. This latter oxygen will move with time, some escaping from the bottle to its surroundings and some passing into the interior. Over time steady state ingress may be established (Lopes, Saucier, Teissedre & Glories, 2006; Lopes et al., 2009; Karbowski et al., 2019).

Since oxygen, sulfur dioxide (SO<sub>2</sub>) and carbon dioxide transmission rates of the package including closures impact on the shelf life of both the packed food and the bottled beverage i.e., promoting or limiting wine oxidation (Godden et al., 2005; Lopes et al., 2006; Lopes et al., 2009; Karbowski et al., 2010), the permeability of materials to gases plays a fundamental role and is commonly used as a mean to assess the barrier. The flow of oxygen able to pass through a package (the closure of a bottle of wine, in our case) is referred to as OTR (oxygen transmission rate in 24 hours), a parameter that depends on both the thickness of the material and the partial pressure gradient between the atmosphere of the external environment and the headspace of the bottle (Crouvisier-Urien, Bellata, Gougeon, & Karbowski, 2018; Reeves et al., 2009). An indirect non-destructive method for determining oxygen permeability through closures during the post-bottling period is based on the oxygen-induced color change of an aqueous solution of indigo carmine (E132) (Crouvisier-Urien et al., 2018; Lopes, Saucier & Glories, 2005).

Much prior research has demonstrated that white wines closed with synthetic stoppers, screw caps, or cork closures (Godden et al., 2001; Marin, Jorgensen, Silva, Lambri, & Faveri, 2003; Kennedy, & Ferrier, 2007; Lopes et al., 2009; Lambri, Silva, & De Faveri, 2012), or packaged in bags within boxes (Fu, Lim, & McNicholas, 2009) exhibit different behaviors

during their shelf-life. For natural cork closures, the wine oxygen consumption depends on the length of the storage with a decrease in rate over time (Lopes et al., 2005; 2006; 2009). It could therefore be postulated the major role played by the rapid diffusion of the oxygen initially contained in the closure than by oxygen permeation through the cap. Indeed, as also observed by Ugliano (2013), the internal cavities of cylindrical closures contain air, which will be released into the bottle when the closure is compressed into the bottleneck, a phenomenon often referred to as "degassing". Therefore, the oxygen capable of being transmitted by a closure could be defined as the whole of that in it contained and of the one exchangeable based on its permeability (OTR). Further, the oxygen diffusion coefficient for a stopper inserted in a bottleneck is higher than for a stopper with interface glued, either uncompressed or compressed (Keenan et al., 1999). This indicated that the interface between the glass bottleneck and the stopper could represent a preferential pathway for gases, a key point which has been recently outlined (Crouvisier-Urien et al., 2018; Karbowski et al., 2019).

The bottle position during storage is another critical parameter to consider as horizontal or vertical storage leads to mass transfer in the gaseous or liquid state. Crouvisier-Urien et al. (2018) reported that when direct contact occurs there is potential for both interaction at the liquid-solid interface and transfer into the gaseous state within the stopper itself. Differently, when no direct contact exists, volatile organic compounds can move more probably than non-volatile compounds (Azevedo et al. 2014) through the liquid-gas interface into the headspace and reach the gas-solid interface between headspace and cork. Controversies in the research outlined for cork closures no significant variation in the wine oxidation between bottles stored in a horizontal and vertical position even for 36 months of storage (Lopes et al., 2006) and over (Skouroumounis et al., 2005), while for other closing materials the evolution of wine differed depending on the bottle position (Mas et al., 2002; Silva et al., 2003; Venturi et al., 2017). Finally, on red wine oxidation, as measured by SO<sub>2</sub> analysis,

Puech, Vidal, Pegaz, Riou, & Vuchot (2006) showed that temperature seemed to be the most significant parameter influencing oxidation when compared to bottle position and light.

### **3. The modification of color: from yellow to brown or (even) pink**

The oxidation level of a white wine in bottle is commonly estimated by color, extent of browning at 420 nm, a parameter that often correlates linearly with the decrease of SO<sub>2</sub> concentration in the wine (Waters, Peng, Pocock, & Williams, 1996; Godden et al., 2005; Lambri et al., 2012). It remains, however, an overall measurement of specific consequences of oxidation reactions occurring in wine. In addition to the reaction between SO<sub>2</sub> and oxygen in the liquid phase, the loss of total SO<sub>2</sub> also involves different parallel reactions or transfer schemes; these include loss as vapor through the closure, the formation of strongly bound compounds, such as with aldehydes, quinones, or keto acids, and the slow oxidation of SO<sub>2</sub> by previously oxidized phenols (Danilewicz et al., 2016). Thus, in addition to the absorbance at 420 nm, the total SO<sub>2</sub> level in white wines acts as an additional measurement indicating the progress of oxidation. Brajkovich et al. (2005) and Lambri et al. (2012) reported a similar tendency looking at the decrease of total and free SO<sub>2</sub> of wine over time as a function of the closure type.

Despite during the bottle storage, the oxidation of phenolic compounds can take place leading to the formation of *o*-quinones with different degrees of polymerization and finally causing the color turn to yellow-brownish hue (du Toit, Marais, Pretorius, & du Toit, 2006), these oxidative reactions can occur either with or without oxygen. When *o*-diphenols are oxidized to *o*-quinones free radicals may be produced, while oxygen is reduced to hydrogen peroxide (Singleton, 1987; Danilewicz, 2003). Due to the instability of *o*-quinones further reactions can happen and brown pigments can be formed (Li, Guo, & Wang, 2008). Hydrogen peroxide is a potent oxidative molecule and it can also oxidize ethanol to

acetaldehyde in the presence of transition metals (Guo, Kontoudakis, Scollary, & Clark, 2017). Acetaldehyde takes part into the formation of dimers via ethyl bridge leading to polymers at the end (Li et al., 2008). The transition metals are also responsible to the oxidation of other wine components, such as tartaric acid, by means of the Fenton reaction for iron in particular. The resulting glyoxylic acid, generated from the oxidation of tartaric acid, and other compounds deriving from the oxidation of organic acids (i.e. dihydroxyfumaric acid) can react with wine phenolics (i.e. catechin); as a consequence, xanthylum derivatives are obtained (Clark, 2008; Li et al., 2008). The formation of xanthylum ions can rise depending on both bottle color and storage temperature. The use of clear glass can make the wine susceptible to the light-induced changes which can lead to its browning (Maury, Clark, & Scollary, 2010). For high temperature (i.e. 45°C), an increase of browning phenomena was described (Dias, Smith, Ghiggino, & Scollary, 2012). This could not be ascribed to the xanthylum derivatives that resulted more easily degraded for higher storage temperature because they can be simultaneously formed and degraded (Bührle, Gohl, & Weber, 2017). However, Bührle et al. (2017) recently showed that the xanthylum derivatives are present in wine at concentrations lower than their perception threshold and they do not might directly impact the white wine color, but might play a role in color formation as intermediate products in polymerization and browning. It is expected that the reaction rates of the pathways above reported is affected by both storage temperature and light exposure. In such conditions, the wine decay occurs in a shorter time that makes necessary a deeper study of the overall characteristics to evaluate the mechanisms majorly affecting the quality of white wine during the storage. Beside the well-established oxidations responsible for the browning of white wine, little is known another color modification that can happen over the storage of white wine, the Pinking. This is a phenomenon (PP) leading to the formation of salmon-red blush color that may take place in white wine produced exclusively with white grape varieties (Simpson, 1977). This color alteration has been

associated to the presence of anthocyanins, mainly malvidin-3-O-glucoside at concentration of about 0.3 mg/L detected in white wines produced with Siria grape variety under reducing conditions (Cosme et al., 2019). The polymerization of anthocyanins under oxidative condition could favor the pink color formation (Andre-Silva et al., 2014). However, this color modification has been not fully understood. Jones, (1989) suggested that reactions or combinations between more than ten different monomers and polymeric compounds could lead to pink wine. Van Wyk, Louw, & Rabie (1996) indicated that a derivative from 2-S-glutathionyl-caftaric acid could be the pink chromophore responsible for the color change. The presence in grape of anthocyanins, even at a very low concentration level, and their polymerizations seems to be most convincing reason to explain PP of white wine. Considering the wine production, the hyperoxygenation of white must limits the formation of salmon-red hue that is contrarily increased by inhibiting the oxidative enzymes (i.e. polyphenol oxidase) (Lamuela-Raventós, Huix-Blanquera, & Waterhouse, 2001). In case of must, the pinking can appear just after grape crushing and it can disappear by the addition of SO<sub>2</sub> due to its bleaching effect (Singleton, Trousdale, & Zaya, 1979; Simpson, Miller, & Orr, 1982; Simpson, Bennett, & Miller, 1983). The winemaking treatments preventing PP of white wine include the use of polyvilylpyrrolidone (Lamuela-Roventós et al., 2001). Recently, the use of certain adjuvants, such as ascorbic acid and catechinic tannins, has been showed to effectively prevent the formation of pinking (Cojocarú & Antoceá, 2019). Promising results have been also reported in case the treatment of wine with UV light is applied (Cojocarú & Antoceá, 2019). During the storage, the pink color may be due to the anthocyanin polymerization and it does not result sensitive to pH and bleaching effect of SO<sub>2</sub> (Simpons, 1977). Further investigation maybe necessary in order to clarify the mechanisms behind PP and possible markers for its prediction in order to prevent its formation and preserve the quality of wine, especially after bottling and over the storage because only limited and no corrections can be carried out in this winemaking stage.

#### 4. Aroma decay

Wine longevity is mainly related to the occurrence of pleasant flavoring notes arising from grape properties, fermenting yeasts, possible flavoring compounds released from barrels and evolution of these compounds during aging and storage. The loss of many olfactory notes over the storage time is a natural phenomenon that limits wine longevity. Many flavors like terpenes and esters are quite unstable and degrade due to the acidity of wine, especially when stored at high temperature (Lambropoulos & Roussis, 2007). However, some physical and chemical factors can shorten wine longevity to a few months or even weeks (Oliveira, Silva Ferreira, De Freitas, & Silva, 2011). When the olfactory faults quickly arise from oxidative phenomena they are referred as untypical ageing (UTA) or PremOx (Pons, Lavigne, Darriet, & Dubourdieu, 2013). Actually, a range of diverse olfactory deviations and causes are included in these terms, but all of them are related to the excessive exposure of wine to air. The olfactory notes usually reported to describe these alterations are honey, cheese, hay, cooked potatoes (Oliveira et al., 2011). The wide range of expression used arise from the number of compounds that can be involved in the oxidative processes. Acetaldehyde is the most represented aldehyde in wine, nonetheless it has not a direct role in the PremOx odor (Escudero, Asensio, Cacho, & Ferreira, 2002), but it can condensate with  $\alpha$ - or  $\beta$ -diol compounds like glycerol or butanediol, so producing dioxanes and dioxolans whose role is still to be proved in wines not obtained from long oxidative ageing (Cutzach, Chatonnet & Dubourdieu, 1999; Silva Ferreira, Guedes de Pinho, Rodrigues, & Hogg, 2002; Câmara, Marques, Alves, & Silva Ferreira, 2003). More importantly, acetaldehyde is essential to the formation of sotolon (3-hydroxy-4,5-dimethyl-2[5H]-furanone). It is a powerful flavoring compound with an intense spicy/curry odor (Girardon, Sauvaire, Baccou, & Bessiere, 1986) which contributes to the characteristic sensory impression of several

foods (Pons, Lavigne, Landais, Darriet, & Dubourdieu, 2010). Its flavor in wine is usually described as curry, fenugreek, and old honey, and it is sought in oxidized aged wines such as Porto (5-958  $\mu\text{g/L}$ ) (Silvia Ferreira, Barbe, & Bertrand, 2003), Vin Jauen (120–268  $\mu\text{g/L}$ ) (Pham, Guichard, Schlich, & Charpentier, 1995), Sherry (0-500  $\mu\text{g/L}$ ) (Martin, Etievant, Le Quere, & Schlich, 1992), Madeira (0-2000  $\mu\text{g/L}$ ) (Câmara, Marques, Alves, & Silvia Ferreira, 2004), botrytised (or noble rot) wines (5-20  $\mu\text{g/L}$ ) (Masuda, Okawa, Nishimura, & Yunome, 1984), Vins Doux Naturels (0-26  $\mu\text{g/L}$ ) (Schneider, Baumes, Bayonove, & Razungles, 1998), and in barrel-aged white wines (0–140  $\mu\text{g/L}$ ) (Lavigne, Pons, Darriet, & Dubourdieu, 2008). Despite being pointed out as a key odorant of other fortified wines, sotolon is considered a defect in dry white wine, as it can dramatically depress the intensity of the fruity and flowery notes as well as the wine freshness character (Kobayashi, 1989). This chiral, thermolabile and polar lactone is highly stable in hydroalcoholic solution (14% ethanol, (v/v) at the pH of wine (Martin, Etibvant, & Henry, 1990). The olfactory perception threshold (OPT) of sotolon was set to 2  $\mu\text{g/L}$  in model wine and 8  $\mu\text{g/L}$  in dry white wine (Pons, Lavigne, Landais, Darriet, & Dubourdieu, 2008). Its aroma characteristics change from caramel-like at low concentrations to curry-like at high concentrations (Kobayashi, 1989). More recently, several authors (Pons et al., 2010; Freitas et al., 2018) determined the contribution of sotolon to the aromas of PremOx dry white wines. Concentrations of sotolon in PremOx dry white wines are generally lower than 10  $\mu\text{g/L}$  (Lavigne et al., 2008). As shown in Figure 1, sotolon develops in winemaking and ageing from a number of chemical pathways involving the Maillard reaction (Pons et al., 2010; Pereira, Albuquerque, Ferreira, Cacho, & Marques, 2011). Reducing sugars (Câmara et al., 2004; Oliveira e Silva, et al., 2008; Oliveira et al., 2011; Pereira, Santos, Cacho & Marques, 2017) as well as ascorbic acid (Barril, Rutledge, Scollary, & Clark, 2016),  $\alpha$ -ketobutyric acid, acetaldehyde and ethanol (Pons et al., 2010) are reported to be involved in the formation of sotolon. It is thermally produced from intermediates generated from the Maillard reaction (i.e. pyruvic,  $\alpha$ -ketoglutaric acids) by

chemical or enzymatic deamination of threonine followed by the aldol condensation of  $\alpha$ -ketobutyric acid and ethanal (Takahashi, 1976; Cutzach et al., 1998). In media containing ascorbic acid the same reaction can occur between acetaldehyde and  $\alpha$ -ketobutyric produced from oxidative degradation of ascorbic acid in the presence of ethanol (Konig et al., 1999; Scholtes et al., 2015). Moreover, sotolon is also produced by a strict oxidative mechanism, based on the peroxidation of acetaldehyde (Pisarnitzkyk et al., 1987), may be responsible for the high levels of sotolon detected in oxidative wines (i.e. Sherry and Madeira). However, the sotolon formation in wine is highly dependent upon oxygen, storage time (Cutzach et al., 1999; Silva Ferreira, Hogg et al., 2003; Câmara, Alves, & Marques, 2006; Lavigne et al., 2008; Jacobson, Monforte, & Silva Ferreira, 2013; Freitas et al., 2018) and temperature (Chatonnet, & Dubourdieu, 2000; Cutzach, Oliveira et al., 2011; Martins, Monforte, & Ferreira, 2013; Fracassetti, Gabrielli, Costa, Tomas-Barberan, & Tirelli, 2016; Coetzee, Van Wyngaard, Šuklje, Silva Ferreira, & du Toit, 2016). Furthermore, it has also been reported a synergistic effect between these physical-chemical parameters that could significantly increase the sotolon concentration in wines (Silva Ferreira, Oliveira, Hogg, & Guedes de Pinho, 2003; Lavigne et al., 2008; Martins, et al., 2013; Monforte, Jacobson, & Silva Ferreira, 2015).

On the other hand, the rational use of antioxidant compounds (i.e. sulfur dioxide and glutathione) and the wine aging on lees mitigate the formation of sotolon, due to their ability to reduce the dissolved oxygen or to bind the carbonyl group of the sotolon precursors in wine (Dubourdieu, & Lavigne, 2004; Lavigne et al., 2008; Pons et al., 2010; Kritzinger, Bauer, & du Toit, 2013; Badea, & Antoce, 2015; Pons et al., 2015). Furthermore, the variability of this aromatic deterioration is strongly affected by differences in permeability to oxygen among the different closures (Silva, Jourdes & Teissedre, 2011; Mayr et al., 2015). Due to the number of chemical pathways and physical factors that influence the sotolon

formation in wine, this compound has been suggested as a chemical marker of PremOx in white wine shelf-life (Pons et al., 2008; Pons et al., 2015; Mayr et al., 2015).

Like acetaldehyde also other aldehydes can be produced by the Fenton reaction that generates hydroxyl radical from decomposition of hydrogen peroxide, the latter formed from via coupled oxidation of diphenols to quinones. Hydroxyl radical, in the absence of antioxidants (i.e. sulfur dioxide, ascorbic acid, glutathione), oxidizes a large number of substrates, in particular alcohols to carbonyls. This is the mechanism responsible for the occurrence of methional from methionol oxidation (Escudero et al., 2000) or to 2-phenylethanal from 2-phenylethanol. Methional has an OPT in the range 0.25-0.75  $\mu\text{g/L}$  in model wine and shows olfactory notes of cooked potato or cooked vegetables without reminding to reduced or sulfide notes. 2-Phenylethanal has olfactory notes described as honey with OPT close to 1  $\mu\text{g/L}$  though values above 20  $\mu\text{g/L}$  might be needed for some white wines (Silva Ferreira et al., 2002; Coetzee et al., 2015). Both methional and 2-phenylethanal are effective in masking the odor of volatile varietal thiols (Coetzee & du Toit, 2015). The Fenton chemical pathway is not the only formation way of these amino acidic derivatives. Even though oxidative conditions are always needed to promote the PremOx odor, the Strecker degradation can have a major role in degrading methionine where oxygen can help the formation of the  $\alpha$ -dicarbonyl compounds needed to bind methionine and for producing the aldehyde (Coetzee & du Toit, 2015). Notably, the human olfactory sensitivity to methional is highly variable among people, as some of them cannot perceive its odor even at concentration approaching 50  $\mu\text{g/L}$  (Escudero et al., 2000). The formation of aldehydes by the Strecker degradation in dry white wine is easier to occur than in dry red wine due to the far higher content of sugars, as the malolactic activity is usually missing. However, it usually takes long time to occur unless wine is exposed to high temperature, like it come about under unsuitable ageing or transport conditions (Silva Ferreira et al., 2002). Also, certain aliphatic unsaturated aldehydes ([E]-2-alkenals) containing 6-9 carbons

are produced under oxidative conditions. Among them, [E]-2-nonenal can easily exceed its OPT (0.6 µg/L in model wine), and olfactory activity values as high as 6 have been reported. However, the lower homologues (C6-C8) proved to have additive olfactory effect, and are able to give olfactory notes of oxidized, dusty, earthy, rancid, brandy even when none of them exceeds the OPT on its own (Cullerea, Cacho, & Ferreira, 2007). Aliphatic carbonyl compounds are likely favorable to give oxidized perception to wine. 3-Methyl-2,4-nonanedione is a  $\alpha$ -diketon having olfactory notes of prune and rancid (OPT 16 ng/L) capable of reaching olfactory activity values of up to 21 in oxidized red wine, thus masking their typical fruity notes (Pons et al., 2013). 3-Methyl-2,4-nonanedione likely has a major role on the human olfactory perception of oxidized, as an olfactory receptor in the olfactory epithelium (OR1A1) specific to this  $\alpha$ -dicarbonyl was identified (Geithe, Noe, Kreissl, & Krautwurst, 2017). Moreover, in spite of its carbonyl structure, 3-methyl-2,4-nonanedione is not able to notably bind sulfur dioxide, which is thus unable to mask its odor (Pons et al., 2013). Instead, sulfur dioxide can be effective in preventing the olfactory faults related to the carbonyl compounds described above. Beyond the well-known affinity with acetaldehyde, sulfite can bind either the aliphatic or branched aldehydes or can act as oxidant scavenger in wine, thus preventing the formation of aldehydes (Danilewicz, 2016). However, such a behavior is ineffective for preventing the formation of aldehydes by the Strecker degradation, but sulfite can ease the formation of 2-aminoacetophenone (2AAP). This compound has been detected in PremOx white wine to whom it confers olfactory notes of wet wool, naphthalene, floor polish (Hoenicke et al., 2002). 2AAP arises from the oxidative degradation of tryptophan and is a powerful flavoring compound as its OPT is in the range 0.7-1 µg/L. Also, *Saccharomyces* can produce 2AAP but it is hardly capable to release it at concentrations exceeding the OPT. As white wines contain poor levels of phenols, sulfur dioxide oxidation and the following formation of oxygenated radicals is enhanced

(Danilewicz, 2012). Consequently, the oxidative degradation of tryptophan to 2AAP can occur (Hoenicke, Borchert, Gruning, & Simat, 2002).

Even the peptide-bound tryptophan can be involved in the oxidation process which can be triggered also by the heat- or light-excited riboflavin (Horlacher & Schwack, 2014). 2AAP can be produced also from the oxidative degradation of the indole-3-acetic acid naturally occurring in the grape as hormone. Early grape vintages, dry seasons or poor nitrogen availability to grape can increase the formation of 2AAP and develop the PremOx olfactory note (Hoenicke et al., 2002).

#### 4.1 *The light-induced changes*

Tryptophan is not the only amino acid acting as electron donor to reduce light-excited riboflavin. When bottled wine is exposed to light, the reaction involving methionine and riboflavin can occur generating sulfur-containing compounds, namely methanethiol and dimethyl disulfide. As a consequence, unpleasant cabbage, onion and garlic odors-like can appear that are associated to the light-struck taste (Maujean, & Seguin, 1983). When riboflavin is exposed to light, it reaches the singlet state that is converted to the triplet state with an intercrossing system. Riboflavin is reduced by the acquisition of two electrons from a donor compound, namely methionine (pathway Type I) that is oxidized to methional. In case oxygen is present, singlet oxygen is generated, a strong oxidant causing non-radical reactions (pathway Type II) (Figure 2). This fault is detrimental for the quality of wine as once it comes up, no corrective strategy can be applied on bottled wine. The proper winemaking management in terms of choice of fermenting yeast, depletion of riboflavin (Fracassetti et al., 2017), protection of wine against the light exposure (Clark et al., 2011), and the possible use of hydrolysable tannins (Fracassetti, Limbo, Pellegrino, & Tirelli 2019) can prevent the formation of light-struck taste. The latter could exert a combined effect as they can be able to compete with methionine into Type I mechanisms, act as scavenger of

singlet oxygen (Fracassetti, Tirelli, Limbo, Mastro, Pellegrino, & Ragg 2020) and bind the sulfur compounds deriving from the oxidation of methionine (Fracassetti et al., 2019).

## 5. Conclusion

Determining the amount of oxygen needed by a given wine type would represent a huge step towards improving wine quality, and would involve finding a specific, measurable molecular parameter in wine which can be correlated to sensory scale ranging from reductive to oxidative. Overall, wine closures must be considered a unique type of food packaging, and it may be of interest to investigate, in a more detailed way, the relationship of a closure's properties and the chemical reactions occurring in wine. The deep investigation of the reaction mechanisms taking place in white wine once it is bottled allows to clarify the actors of responsible for the shortening of wine shelf-life. Studies on conjunction between type of closure, wine composition and storage conditions, in particular light and temperature, will prevent the shortening of wine shelf-life ensuring the maintenance of quality characteristics.

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**Figures' captions**

Figure 1. Potential formation pathways of sotolon in wine. The formation pathways of sotolon have been classified as: Maillard reaction or Strecker degradation (red arrow), oxidative pathway (blue arrow) e aldol condensation (green arrow). References: (a) Takahashi, (1976); (b) Kobayashi, (1989); (c) Pham et al., (1995); (d) Cutzach, Chatonnet, & Dubourdieu, (1998); (e) Guerra & Yaylayan, (2011); (f) Blank, Lin, Fay, & Fumeaux, (1995); (g) Blank, Lin, Fumeaux, Welte, & Fay, (1996); (h) Hofmann & Schieberle, (1995); (i) Hofmann & Schieberle, (1997); (j) Schieberle & Hofmann, (1997); (k) Scholtes, Nizet, & Collin, (2015); (l) Hofmann & Schieberle, (1996); (m) Ferreira, Ávila, & de Pinho, (2005); (n) Danilewicz, (2003a); (o) König et al., (1999); (p) Schwab et al., (2001); (q) Shin & Feather, (1990); (r) Pisarnitsky, Bezzubov, & Egorov, (1987); (s) Pons et al., (2010).

Figure 2: Photo-degradative reactions involving riboflavin, methionine and phenolics (Fracassetti et al., 2019).



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### Highlights

- Premature wine oxidation causes a quick decay of bottled wine with aging potential
- Changes in the oxidative status are affected by both oxygen and storage conditions
- Wine-bottle-closure system plays a consistent effect
- Alteration of color to yellow-brownish or even pinkish hues can occur
- Aroma losses and off-flavor formation are associated to this phenomenon