Effect of Antioxidants on the Flavor Characteristics and the Gas Chromatography/Olfactometry Profiles of Champagne Extracts

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This paper aims to investigate the possibility of oxidation of the flavor active constituents of champagne wines during the critical steps of the extraction for analytical purposes and to estimate its consequences on the extract's aroma characteristics. The investigation uses conventional sensory and GC/O analyses. Triangle and similarity sensory tests carried out on champagne samples and on their extracts show that the absence of antioxidants implies a deterioration of the aroma quality. Conversely, the use of 2-*tert*-butyl-4-methoxyphenol (BHA) as antioxidant gives an extract with an odor judged to be more similar to the original odor of the champagne. The alteration in the odor of the unprotected extracts is explained, from GC/O and GC/MS analyses, as three types of uncontrolled reactions. These are, first, the chemical oxidation of the fusel alcohols and the amino acids; second, the oxidation of sensitive flavor compounds such as enolic lactones and monoterpenic alcohols; and, third, the hydrolysis of fatty acid esters and saturated γ-lactones. A systematic use of BHA during flavor extraction is therefore advisable, in particular when oxidation phenomena such as wine aging are under study.

Keywords: *Antioxidant; BHA; champagne; flavor extract; gas chromatography/olfactometry*

INTRODUCTION

Oxidation is a process that is very often uncontrolled. In food production, it can sometimes be noticed because of undesirable sensory effects. Oxidation may take place during processing, distribution, and home preparation of food, with possible negative consequences on consumer acceptability.

To control the oxidation phenomenon, it is therefore important to know more about the associated flavor alteration. However, because such studies involve an extraction of the volatile constituents from the food matrix, uncontrolled oxidation can also take place during the different steps of the analyses (Dubois, 1994) with a risk of masking the original oxidation phenomenon under study. In wines, the presence of natural polyphenolics (Ghiselli et al., 1998) or added SO_2 (Somers and Wescombe, 1982) prevents flavor oxidation. However, during the extraction and distillation (Priser et al., 1997)(where the volatile components are in the alcoholic phase), these are not protected by these antioxidants and oxidation may occur.

As a precaution, solvent distillation is often performed at reduced pressure, thus limiting the amount of available oxygen and so reducing the kinetics of the oxidation reactions, which depend mainly on temperature. However, oxidation may still take place (Dubois, 1989), mainly because it is impossible to avoid contact of the extract with air during decanting and storage, during sampling before injection, and also during injection in hot liners.

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When oxidation has occurred, it can sometimes be reversed with a reducing agent, that is, by placing the extracts in a solution of ascorbic acid and SO_2. The oxidized flavor of the extract then disappears, and the original flavor is perceived again (Dubois, 1989).

The use of antioxidants and evaluation of their efficiency to protect the wines and their extracts were therefore considered to be of great interest.

Antioxidants can interfere with the oxidation processes as radical scavengers (e.g., hydroxy radical acceptor), singlet oxygen quenchers, and enzyme inhibitors and in synergy with other agents (metal chelating agent plus a reducing agent) (Namiki, 1990).

Phenolic antioxidants are very efficient for the prevention of autoxidation. They are sometimes metal chelators, free radical terminators, and excellent hydrogen or electron donors (their radical intermediates are stabilized by resonance delocalization) (Nawar, 1985).

2-*tert*-Butyl-4-methoxyphenol (BHA) and 2,6-di-*tert*-butyl-4-methylphenol (BHT), the most widely used synthetic phenolic antioxidants, show a high efficiency in various food systems along with high stability and low cost. They are important and useful additives in packaging materials because due to their volatility they can migrate from the polymer into the food matrix (Shahidi et al., 1992). This property is important for this study, in which we want to protect the flavor constituents during extraction and distillation. BHT is more effective in suppressing the oxidation of animal fats than that of vegetable oils, whereas BHA is particularly useful in protecting the flavor and color of essential oils (Stuckey, 1972). BHA is also particularly effective in controlling the oxidation of short-chain fatty acids involved in the development of undesirable flavor active alcohols and aldehydes (Shahidi et al., 1992).

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Natural antioxidants, such as tocopherols or β-carotene, are multifunctional (free radical terminators and singlet oxygen quenchers; Shahidi et al., 1992). Recently some natural flavor compounds, 4-hydroxy-2,5-dimethyl-3(2*H*)-furanone and 4-hydroxy-2(or 5)-ethyl-5(or 2)-methyl-3(2*H*)-furanone, have been characterized as potent antioxidants because of their activity in lipid peroxidations (Koga et al., 1998). Other substances such as tannins, phenolic acids, and flavonoids are also known as free radical terminators and chelators of metal ions capable of catalyzing oxidation reactions (Shahidi et al., 1992; Namiki, 1990). These substances are present in white wines in lower amounts than in red wines, explaining the higher oxidation sensitivity of white wine.

Oxidation is a major problem during the elaboration of champagne wines and during their aging. Given the availability of different antioxidants and the fact that oxidation mechanisms in champagne wines, and their extracts, are not well-known, we chose to test BHA as a good synthetic antioxidant of flavors.

The purpose of this paper is to test the effects of this antioxidant on the flavor characteristics of the champagne extracts compared to those of the wine itself, using common sensory and instrumental flavor analysis methods, that is, triangle and similarity tests and gas chromatography coupled with olfactometry (GC/O).

MATERIALS AND METHODS

Champagne Wines. Two Moët & Chandon champagne wines, one vintage from 1996 (Ch2) and one from 1994 (Ch1), were stored on yeast in a cellar until analysis. Ch1 and Ch2 were bleedings of three wine varieties (Pinot Noir, Pinot Menier, and Chardonnay). One was a vintage (Ch1) and both were dry (brut). The alcoholic content of the two wines was 12.5% (v/v).

Extraction of Aroma Components. *Demixtion and Distillation*. One hundred seventy-one grams of $(NH_4)_2SO_4$, 41.85 g of $H_2NaPO_4 \cdot H_2O$, and a magnetic rod were successively added to a specially designed dry 500 mL flask, as described by Moio et al. (1995). The flask was then purged of oxygen by alternative connection to a vacuum pump and to a nitrogen supply (95%) using a three-way high-vacuum glass tap. In the experiment with antioxidant, 1 mL of a BHA solution (2 g L^{-1} in ethanol) was sucked into the flask under slight vacuum. In the control experiment (without antioxidant) the same volume of pure ethanol was similarly introduced. The champagne sample (375 g, estimated by weighing) was then directly introduced from the bottle into the flask using an aphrometer connected to the extraction flask with Teflon tubing ($^1/_8$ in. external diameter) tightened with Swagelock ferrules and knots, to eliminate contact with oxygen. The wine sample was mixed with the salts, and the BHA solution was until completely dissolved. The flask was then inverted for 1 h to permit phase separation. Air was then introduced into the flask, the aqueous phase was decanted and discarded, and the organic phase was transferred into a small vial and stored at −20 °C.

A two-stage distillation of these demixtion extracts was then performed to remove the nonvolatile constituents (mainly pigments and salts). First, 12 mL of extract was distilled for 1.5 h under a pressure of 0.2 Pa as described by Dumont and Adda (1970). Second, the residue was further distilled on a coldfinger kept at the temperature of liquid nitrogen for 1 h at 0.001 Pa, as described by Richard and Etiévant (1997). The condensate was rinsed from the coldfinger with the distillate obtained from the first distillation, and this final extract was stored at −20 °C.

The two extracts obtained from champagne Ch1 (with BHA and without BHA) were submitted to comparison tests.

All four extracts were analyzed by GC/O.

Solvent Extraction. The concentration factor of the volatile compounds in the final extracts (from 417 to 12 mL) was not high enough to identify them by GC/MS. To facilitate these identifications, an aliquot of each extract was re-extracted with dichloromethane. Five milliliters of each extract in a 25 mL screw-capped flask was diluted with 20 mL of the saline solution (263 g L^{-1} $(NH_4)_2SO_4$ at H_2O Milli-Q) and stirred with 0.5 mL of CH_2Cl_2 for 30 min. After separation of the two phases, the organic layer was taken off with a syringe and transferred into a small vial, which was stored at −20 °C. A 1 μL aliquot of these extracts (Ch1 and Ch2 with and without BHA) was analyzed by GC/MS equipment.

Gas Chromatography. *GC/O Analyses*. The analyses were carried out using a Hewlett-Packard 5890 chromatograph equipped with an on-column injector (J&W Scientific Inc.), a flame ionization detector (FID), a sniffing port, and a DB-Wax fused silica capillary column (30 m, 0.32 mm i.d., film thickness = 0.5μm, J&W Scientific Inc.) connected to the injector with a 5 m deactivated polar precolumn (0.53 mm i.d., J&W Scientific Inc.). The column effluent was split equally between the detector and the sniffing port, and humid air (100 mL min^{-1}) was added to the sniffing port effluent. The hydrogen carrier gas velocity was 50 cm s^{-1}, and the temperatures of the injector and detector were settled at 250 °C. The oven temperature was programmed from 67 to 240 °C at 5 °C min^{-1}.

A 5 μL aliquot of each extract was injected. During the analysis, FID and olfactometry signals were simultaneously recorded, using the hardware and the software developed by Mielle and Almanza (1993). The linear retention indices (LRI) of the FID and the olfactometry peaks were determined according to the method of Van den Dool and Kratz (1963), using a C_{10}−C_{28} *n*-alkane solution analyzed once a day.

GC/MS Analyses. GC/MS analysis was carried out on an HP 5970 quadrupole mass spectrometer directly coupled to an HP 5890 gas chromatograph. The same column and precolumn were coupled directly to the ion source (temperature = 150 °C). The carrier gas was helium, and the other conditions were identical to those described above. Electron impact mass spectra were produced with an ion source energy of 70 eV and recorded with an HP-UX Chemstation. The identifications made by GC/MS were systematically confirmed with the retention indices of the pure references, determined in the same analysis conditions.

Sensory Analyses. *Comparison Tests*. The internal panel consisted of 24 flavor chemists (10 women, 14 men; average age, 30).

An aliquot of the wine extracts (50 μL) was adsorbed onto 55 × 27 mm pieces of absorbent paper (OSI, type P110) and placed after 2 min (time necessary for solvent evaporation) in 60 mL dark glass, screw-capped flasks. For the champagne, 700 μL was adsorbed onto the same support and placed in the same flasks. For sensory evaluation, the panelists had to open the different coded flasks and sniff the samples.

The impact of the BHA on the aroma of the extracts and the odor of the BHA itself at the concentration used in the extracts (150 ppm) were assessed by triangle tests (AFNOR, 1983).

A similarity test was performed to compare the odor of the extracts (with and without BHA) with the odor of Ch1. The champagne wine was presented as the reference sample, and the extracts were presented in random order. The panel members were instructed to sniff and memorize the aroma of the reference sample and then to sniff the first coded flask (containing one of the two extracts) and determine the similarity of their odors. A 100 mm unstructured scale was used for this, anchored with "identical to the reference sample" on the left and "different from the reference sample" on the right. The panelists were then asked to repeat the evaluation using the same reference sample and the second coded flask.

GC/O Evaluation. The evaluations were made by nine assessors (five women, four men; average age, 33). These people were recruited from the population of Dijon, and they were paid. Seven of them had previous experience in this type of evaluation. They were asked to sit in front of the sniffing port during the analysis (30 min) and to press the space bar

Table 1. Characteristics of the 14 Odors Selected by Two McNemar Tests

LRI[a]	odor description	*p* values sample Ch1	*p* values sample Ch2	MS identification
901	sweet, strawberry, cherry	0.03	>0.1	ethyl ester or acetate coeluted with the solvent
1107	leek, acid apple, licorice	0.08	>0.1	2-methyl-1-propanol
1422	fresh cream, cut grass, sweet	>0.1	0.05	1-hexanol
1549	potato, flower, fruit, aldehyde	0.05	>0.1	benzaldehyde
1705	flower, sweat, potato	0.08	>0.1	diethyl succinate
1715	flower, anise, mint, fruit	>0.1	0.05	α-terpineol
1733	yeast, mercaptan, potato, cut grass	>0.1	0.08	3-(methylthio)-1-propanol
2031	caramel, toasted sugar	0.08	>0.1	Furaneol
2067	toasted wood, toasted bread	>0.1	0.08	*m*-cresol
2127	white flowers, sweet fruit chocolate	>0.1	0.05	ethyl cinnamate
2190	curry, celery, spices	0.08	>0.1	sotolon
2221	flower, leather, coffee	>0.1	0.03	not identified
2235	acid, toast	0.08	>0.1	decanoic acid
2238	flower, sweet, vanilla	0.08	0.08	γ-undecalactone

[a] LRI, linear retention indices.

on a computer key board when, and as long as, an odor was detected. Furthermore, they were asked to describe the quality of the odor detected, which was automatically recorded.

Data Analyses. *GC/O Data*. The data treatment was based on the detection frequency observed for each retention time (Pollien et al., 1997). The nine individual aromagrams, produced for each extract, were first adjusted for slight retention index shifts, then pooled into one aromagram by simple summation, and converted into a contingency table. A McNemar test was performed on each champagne sample to evaluate the antioxidant effect, using SigmaStat 2.0 software. This test allowed a selection of those odors that were evaluated differently for the extracts with or without antioxidant at $p < 0.08$.

A factorial correspondence analysis (FCA with no rotation, StatBox 2.1 software, Grimmer logiciels, Paris) was then performed on the selected odors using the same data.

Comparison Test Data. In the triangle tests, the numbers of total and correct answers were used to test, with a binomial law table settled for one-third probability, if there were differences of odors between the samples.

In the similarity test, the marks on the unstructured scales were read as distances in millimeters from the left anchor. A univariate analysis of the variance was carried out on these distances for the sample effect, to test the significance of the difference among the panel's mean answers.

RESULTS AND DISCUSSION

The purpose of this paper was to investigate the possibility of an oxidation of the flavor active constituents of champagne wines during the critical step of the extraction and to estimate its consequences on the aroma characteristics of the flavor extracts thus obtained. Among the numerous possible extraction techniques described to isolate the flavor constituents of wines, we chose the demixtion technique followed by a distillation, because this method was proven to give an extract descriptive odor profile for champagne which is closer to that of the original wine than the profiles provided by other techniques (Priser et al., 1997). The two wines chosen were relatively young (<3 years from bottling) to avoid aging-induced oxidation.

In a first experiment, a fixed amount of BHA (see Materials and Methods) was added to Ch1, and the overall aromas of these extracts were then compared, using triangle tests, to evaluate its effects.

The test demonstrated an odor difference due to BHA ($p = 0.003$) with 15 correct answers versus 24 total answers. BHA was therefore submitted to further evaluations.

To check if the difference found was due to BHA's own odor or to a real antioxidant effect, a new triangle test was performed. For this test, the same fixed amount of BHA was added to a synthetic solution similar to standard distilled champagne demixtion extract (ethanolic content = 57%). The results showed that, at the concentration used in the experiment, the odor of BHA was not detected by the panel (10 correct answers versus 24 total answers, $p = 0.25$). Therefore, the difference found previously in the wine extracts can be attributed to oxidation during the demixtion or the distillation.

To test the oxidation hypothesis, a similarity test was performed using the same champagne as the reference. This similarity test demonstrated that the mean estimated distances between the odor of the two extracts and the odor of the wine were not equal ($p < 0.05$). On the 100 mm scale, the extract with BHA was situated at 47.4 mm ($\sigma_{n-1} = 33.6$) and the extract without BHA at 62.3 mm ($\sigma_{n-1} = 28.5$) from the champagne. It can be therefore concluded that the BHA not only protected the flavor of the extract against oxidation but also preserved the original flavor characteristics of the wine.

The active flavor components and their potential oxidation during the extraction were further investigated by a GC/O analysis. Four extracts were made from the two champagnes, and BHA was added to each of them. The panel analysis, limited to 30 min, was as described by Acree (1993). The analysis of the four final aromagrams obtained by summation of the individual sniffings showed that 88 different odors were detected by the panel.

To determine the differences due to oxidation between the aromagrams, two McNemar tests (χ^2 analysis of the contingency table; Agresti, 1996) were applied. As a result, 14 odors were detected with a significant frequency difference ($p < 0.08$) between the protected and the unprotected extracts (Table 1).

The odor descriptors of these 14 selected compounds are also given in the same table, with their retention indices and their identifications. Although our panelists were not trained to describe odors, the sensory terms of Table 1 correspond roughly to the description expected for the compounds that we identified. These compounds belong to different chemical classes: esters, acids, phenols, alcohols, aldehydes, monoterpenes, thiols, and lactones. These identifications were successfully achieved both by GC/MS and with the pure substances for most of the odors detected, except for the odor at

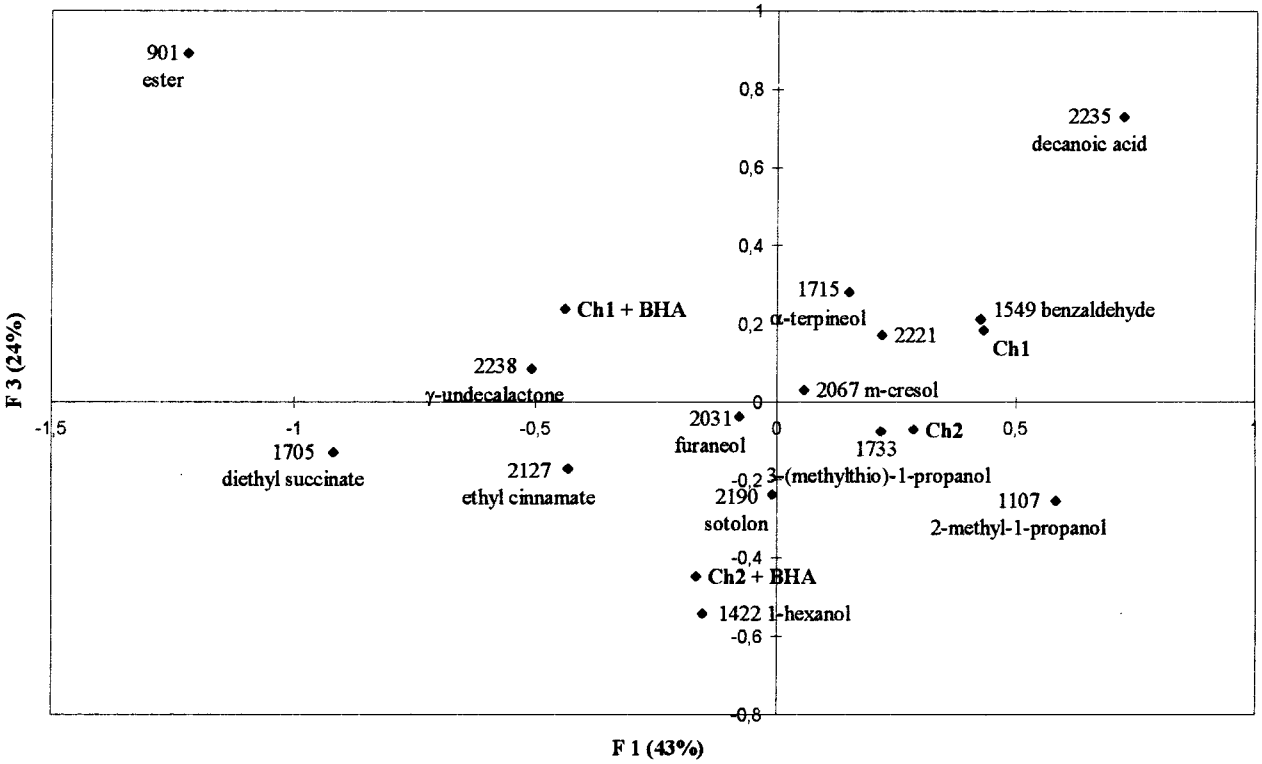

Figure 1. Factorial correspondence analysis: relative positions of 4 samples and 14 odors in the plane formed by the first and third axes. The odors are plotted with their LRI and their identification (see Table 1).

retention index 2221, for which no mass spectrum could be obtained.

To obtain a general view of the GC/O odors that characterized the four extracts, a factorial correspondence analysis was made using the detection frequency of the 14 GC/O odors selected above. The most interesting information given by this analysis is seen on the plane formed by the first and third axes as shown in Figure 1. The first axis (43% of the information) mainly distinguishes the extracts on the basis of BHA presence during their preparation, and the third axis (24% of the information) separates the extracts obtained from the two champagnes (Ch1 and Ch2). The second axis (33% of the information) is ignored in the interpretation because it separates the extracts on a basis independent of the sample used for extraction or of the presence or absence of antioxidant.

The difference between the two champagnes chosen was related to a greater detection in the Ch1 extracts of decanoic acid and an ester-like compound at LRI 901; there were also a lower detection of α-terpineol and a greater detection of hexanol in the Ch2 extracts (Figure 1). As seen from the same figure, the extracts containing BHA are characterized by a greater detection of diethyl succinate, ethyl cinnamate, and γ-undecalactone and by a lower detection of decanoic acid, benzaldehyde, and 2-methyl-1-propanol.

The detection frequency and thus the concentration (Pollien et al., 1997) of three esters (LRI 901, diethyl succinate and ethyl cinnamate) were clearly higher when BHA was added to the wines. In addition, it was observed that the extract containing BHA had a pH of 3.7, whereas that without BHA gave pH 4.2. This difference was the cause of the variation observed in the concentrations found of these three esters, given the dependence of hydrolysis equilibria and esterification on pH (Chisholm et al., 1995; Rapp and Mandery, 1986; Garofolo and Piracci, 1994).

Because γ-undecalactone (LRI 2238) is a natural cyclic ester (Dufossé et al., 1994), it was also detected more often in the extracts containing BHA; the same explanation can be invoked again. Conversely to the esters, decanoic acid (LRI 2235) was less abundant in extract Ch1 with added BHA. This observation seems logical because fatty acids are readily produced from the oxidation of alcohols and aldehydes (Nykänen, 1986). GC/MS was used to check that the fatty acids present in the samples were more abundant in the extracts without BHA. This affirmation could not be made using GC/O due to the saturation of the system's signal for these aromas.

The previously published hypothesis concerning the oxidation of alcohols into acids (Litchev, 1989; Profumo et al., 1988) when no BHA is added to the extracts is in accordance with the lower detection of hexanol in extract Ch2 containing no antioxidant. This has also been observed in deliberately oxidized wines (Ferreira et al., 1997). 2-Methyl-1-propanol was more abundant in the extracts without BHA, probably due to the oxidative deamination of the corresponding amino acids (Etiévant, 1991).

α-Terpineol (LRI 1715) was clearly much more abundant in the extracts without antioxidant. This observation is not surprising since Usseglio-Tomasset and Di Stefano (1980) showed that α-terpineol is formed from the oxidation of other terpenols such as linalol, geraniol, nerol, and citronellol.

As expected, the odor of benzaldehyde (LRI 1549) was detected more often in the extracts without antioxidant. The possible precursors of benzaldehyde are benzylic alcohol (Delfini et al., 1991) or phenylalanine (Wildenradt and Singleton, 1974).

Although methionol (3-methylthio-1-propanol) is known to disappear during wine oxidation (Ferreira et al., 1997), its more frequent detection in extracts without BHA (LRI 1733) can nevertheless be explained by an

oxidation involving quinones (Saijo and Takeo, 1970) or by photodegradation (Dozon and Noble, 1989) from a methionine precursor, another common amino acid in wine.

Sotolon (LRI 2190) is a powerful flavor compound that is found in champagnes aged on lees (Loyaux et al., 1981). It was detected more often by GC/O in the protected extract Ch1. Sotolon has been shown to be an unstable compound sensitive to oxygen, heat, and UV radiations (Martin et al., 1990), and a probable explanation for its more frequent detection in the extracts containing BHA is that the BHA gives a better protection against degradation.

Furaneol is also a very unstable compound with an enolic dihydrofurandione skeleton similar to that of sotolon (Girardon et al., 1986). It is therefore not surprising that its odor (LRI 2031) was detected more frequently in the BHA-protected extract.

m-Cresol (LRI 2067) is a phenolic compound that has been shown to undergo chemical oxidation during wine aging, in common with other phenols (Ferreira et al., 1997). However, its central position on the FCA analysis (Figure 1) was due to different behaviors in the two champagnes (increasing detection with BHA in sample Ch1 and decreasing in Ch2); this meant that no clear interpretation was possible.

Finally, the odor at LRI 2221 was detected more often in the extracts without antioxidant. This compound should arise from oxidation, but this hypothesis could not be checked because its identification was not possible.

CONCLUSIONS

The olfactory alteration detected by the panel in the extracts without antioxidant was conclusively attributed, using GC/O, to three types of reactions; first, the chemical oxidation of fusel alcohols and of amino acids; second, the oxidation of sensitive flavor compounds such as enolic lactones and monoterpenic alcohols; and, third, the hydrolysis of fatty acid esters and saturated γ-lactones. The protection of flavor constituents is therefore necessary to obtain reliable results on the flavor impact of volatile constituents of wines, in particular when the effects of oxidation are being studied.

ABBREVIATIONS USED

BHA, 2-*tert*-butyl-4-methoxyphenol; BHT, 2,6-di-*tert*-butyl-4-methylphenol; FCA, factorial correspondence analysis; FID, flame ionization detector; GC/MS, gas chromatography coupled with mass spectrometry; GC/O, gas chromatography coupled with olfactometry; H_2O Milli-Q, water obtained from a Milli-Q purification system (Millipore, Bedford, MA); LRI, linear retention indices.

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