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Dry vs soaked wood: Modulating the volatile extractible fraction of oak wood by heat treatments

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ABSTRACT

The aim of this study was to analyze the impact of the water content of wood on the concentrations of volatile compounds which can be extracted after heat treatments. Head Space-Solid Phase Micro Extraction Gas Chromatography coupled to Mass Spectrometry (HS-SPME GC–MS) has been used to compare the concentrations of six aroma compounds (vanillin, furfural, eugenol, guaïacol and *cis*- and *trans*-whisky lactones) in hydroalcoholic extracts of heated oak wood samples either previously soaked in hot water or not. Except for eugenol, concentrations of extracted aromas appeared to be lower in soaked woods than in dry woods for temperatures up to 200 °C. If a delaying effect of water could explain such overall lower extracted concentrations from soaked woods, a PCA analysis revealed that for the longer duration (25 min of heat treatment), the adsorbed water could promote a higher impact of furfural, eugenol and both whisky lactones on the composition of hydroalcoholic extracts, suggesting that alternative mechanisms of thermal modifications of the wood macromolecular network could exist at high temperatures in presence of adsorbed water.

1. Introduction

Initially intended as a wine suitable container, oak barrels are now considered to have an important effect on the aroma, the colour, and the stability of wines (Ho, Silva, & Hogg, 2001; Perez-Prieto, López-Roca, Martínez-Cutillas, Pardo-Mínguez, & Gómez-Plaza, 2003). During wine storage in oak barrels, various matter exchanges between wood and wine actually take place, which can lead either to a wine enrichment by wood aroma compounds or to the retention of wine aroma compounds by wood (Barrera-García, Gougeon, Karbowiak, Voilley, & Chassagne, 2008; Boidron, Chatonnet, & Pons, 1988; Cerdán, Goñi, & Azpilicueta, 2004; Garde-Cerdán & Ancín-Azpilicueta, 2006; Jarauta, Cacho, & Ferreira, 2005; Mosedale, Puech, & Feuillat, 1999; Perez-Prieto, López-Roca, Martínez-Cutillas, Pardo Mínguez, & Gómez-Plaza, 2002; Spillman, Pollnitz, Liacopoulos, Pardon, & Sefton, 1998; Waterhouse & Towey, 1994). Besides, physical processes such as micro-oxygenation thanks to the intrinsic porosity of barrels or spontaneous wine clarification (favoured by lowered temperatures) can also occur. Sensory properties acquired by the wine during barrel ageing are known to depend on many different factors including the initial wine composition, the ageing duration and above all the wood composition itself, which will actually govern its potential for aromatic and/or tannic impact (Garde-Cerdán & Ancín-Azpilicueta, 2006; Jarauta et al., 2005; Mosedale et al., 1999; Perez-Prieto et al., 2002; Spillman et al., 1998; Waterhouse & Towey, 1994). An extensive literature shows that the wood natural composition displays a great variability depending on the geographical origin (Ancín, Garde, Torrea, & Jimenez, 2004; Del Alamo Sanza, Nevares Dominguez, Carcel Carcel, & Navas Gracia, 2004; Mosedale et al., 1999; Perez-Prieto et al., 2002; Prida & Puech, 2006; Waterhouse & Towey, 1994), the species (Feuillat, Keller, Sauvageot, & Puech, 1999; Mosedale, Feuillat, Baumes, Dupouey, & Puech, 1998) and even the position in the tree itself (Doussot, Pardon, Dedier, & De Jéso, 2000; Prida & Puech, 2006).

This natural composition will be modulated throughout the barrel making process, which starts with an 18–36 months period of natural drying (or seasoning), during which the volatile compounds content may be either down- or up-modulated, whereas

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a decrease of the tannins content is generally observed (Cadahía, Fernández de Simón, & Jalocha, 2003; Cadahía, Muñoz, Fernández de Simón, & García-Vallejo, 2001; Doussot, De Jéso, Quideau, & Pardon, 2002).

The second step consists in two successive heat treatments, where the first one aims at bending the staves and the second – critical – one is designed to fine-tune the concentration of volatile compounds through a more-or-less controlled degradation of macromolecules. Three studies report genuine cooperage conditions (Hale, Mccafferty, Larmie, Newton, & Swan, 1999; Kim, Kim, Kim, & Yang, 2006; Nomdedeu et al., 1988) but with different combinations of temperature and duration for a same given "intensity". Furthermore, one can find as many results expressed in volatile mass/wood mass (mg/g) concentrations as results expressed in volatile mass/solvent volume (mg/L) concentrations, but tendencies can appear. Several studies have already focused on the impact of these heat treatments on the modulation of wood-related wine sensory properties, and it is now acknowledged that the degradation of celluloses and lignin, which already occurs during the bending process, is directly correlated to the levels of extractability of cooperage oak woods (Cadahía et al., 2001; Cadahía et al., 2003; Caldeira, Clímaco, de Sousa, & Belchior, 2006; Campbell, Sykes, Sefton, & Pollnitz, 2005; Canas, Belchior, & Falcao, 2007; Canas, Grazina, Belchior, Spranger, & de Sousa, 2000; Chatonnet, Boidron, & Pons, 1989; Sarni, Moutounet, Puech, & Rabier, 1990; Spillman, Sefton, & Gawell, 2004). However, if the evolution of the concentration of several volatile compounds as a function of heat treatment has been thoroughly studied, conditions used were not always comparable (real wine *vs* model wine extraction, wood chips *vs* staves...) and no general agreement can be found in the literature. For example the furfural concentration regularly increases with the toasting level according to Wielage, Lampke, Marx, Nestler, and Starke (1999), Gröndahl, Teleman, and Gatenholm (2003), (Marcovich & Villar, 2003), and (Kim et al., 2006) whereas Chatonnet et al. (1989) and (Hale et al., 1999) found highest levels in medium toasted oaks. Such discrepancies illustrate the fact that conditions of heat treatment are often described in the literature using a generic term such as "intensity" that can be actually associated with various conditions of temperature and duration as shown by Chatonnet and Boidron (1989)), Nomdedeu et al. (1988) and Sarni et al. (1990).

Despite such discrepancies, general trends exist for heat treatments of dry wood. It is acknowledged for instance that guaïacol, eugenol and vanillin result from the breakdown of lignin, and that even if low concentrations can be found in unheated wood (Jordão et al., 2006), their concentrations tend to increase with heating (Chatonnet, 1999). Similarly, furfural is known to be produced from the depolymerization of hemicelluloses catalysed by the acetic acid that is also formed during the thermal treatment. Therefore, its concentration generally increases with temperature. In contrast, both whisky lactones are initially present in unheated wood but may also be produced as a result of lipids degradation during heating (Chatonnet et al., 1989). The *cis* isomer is more abundant than the *trans* one, and its perception threshold being rather low, it is accepted that it plays an important role for the oak-related aroma of wines (Chatonnet et al., 1989). Oak lactones are certainly the compounds for which the literature shows greatest discrepancies in terms of the evolution of their concentrations with heat treatments. They have been shown to be constant (Caldeira et al., 2006), to decrease at high temperature (Campbell et al., 2005), to increase and then decrease (Chatonnet, Cutzach, Pons, & Dubourdieu, 1999; Chatonnet et al., 1989) or finally to decrease and then increase at high temperatures (Fernández de Simón, Cadahía, del Álamo, & Nevares, 2010). Furthermore, due to the diversity of values and units reported in the literature (μg/g of wood *vs* μg/L of wine) and whatever the compound, it is somewhat difficult to provide average concentrations for a given compound (as a function of heating temperature for instance).

A particular aspect of all of these studies so far is that the traditional heating process was only based on dry wood, that is with staves exhibiting an uncontrolled – though low – water content resulting from an equilibrium state with the relative humidity of the local environment. Actually, the water content impact has only been studied for wood moistening during treatment and results showed an interesting evolution of the cristallinity of cellulose (Shebani, van Reenen, & Meincken, 2008). According to Canas et al. (Canas et al., 2007), moistening only affects significantly the content of coniferaldehyde, which is a precursor of vanillin. Finally, Costa, Mendonça, and Figueiredo (2008) suggested that water could have a delaying effect on the formation of wood volatiles, explaining that if water is present, the wood temperature would stay around 100 °C.

In this model study and for the first time, Head Space-Solid Phase Micro Extraction Gas Chromatography coupled to Mass Spectrometry (HS-SPME GC–MS) has been used to compare the concentrations of six aroma compounds in hydroalcoholic extracts of oak wood samples – either previously soaked in hot water or not – which had then been heated at different temperatures and for two durations.

2. Material and methods

2.1. Wood samples

All wood samples come from a single 400-years old *Quercus petreae* tree from the "forêt des beaux Monts" (Oise, France), provided by Tonnellerie Seguin Moreau (Cognac, France). They were taken from staves after natural seasoning for two years.

2.2. Heat treatment

Wood pieces with uniform geometry (L, l, w = 70 mm, 25 mm, 3 mm) were used for heat treatments. A calcination oven initially stabilized overnight at desired temperatures was used to apply heat treatments. Heat treatments were done in triplicate, i.e. for a given condition (one temperature, one duration and one moisture condition), three distinct and randomly chosen pieces of wood were simultaneously heated, and this was done three times. Therefore, a given condition provided us with three sets of three distinct pieces of wood. Treatment conditions were defined according to temperatures observed in the industry (Prida, Drinkine-Magneux, Gougeon, & Chassagne, 2010). Fives temperatures were selected: 90, 120, 160, 200 and 240 °C; and two durations: 10 and 25 min. Before heat treatments, the wood was soaked in hot water (90 °C) for 20 min, or not.

2.3. Wood extraction

Each of the three sets of three distinct pieces of wood for a given condition of heat treatment were first cut into smaller shavings using a sharp crowbar and then grounded down to <345 μm powder with an electric coffee grinder, finally providing us with three sawdust samples for a given condition. For each set, 10 grinding cycles of 5 s separated by periods of 15 s were applied in order to avoid an overheating of the wood. The obtained sawdust samples were kept at 4 °C until the extraction.

Each of the three sawdust samples for a given condition were extracted once for volatiles using the following method. Sawdust samples were soaked in hydro alcoholic solution (13% ethanol, 5 g/L tartaric acid and pH adjust to 3.2 with sodium hydroxide) at a concentration of 30 g/L for 48 h at 25 °C in darkness. After