PRIMARY AROMATIC CHARACTER OF WINES

— review —

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Abstract: In this paper the current understanding of the accumulation of the flawours in grapes, mosts and wines is reviewed. The primary flavours are generally constituted by the terpenes and terpenoids, norisoprenoids, benzol derivatives, aliphatic and glycosidic substances, carotene substances.

Keywords: primary varietal flavour, grape, wines, musts

INTRODUCTION

The primary varietal flavours accumulate in the peel and grape through the specific metabolism processes, being determined by the genetic nature of the varieties, by the specific pedological and climatic factors (Tița, 2004) (Pop, 2008). They can pass unmodified from grape to wine, in the case of the Muscat wine, or by changing chemical form (from bonded to free), especially in the case of semi aromatic and red wine varieties. The terpenoides represent the main odour substances class of Vitis vinifera grapes. They are especially responsible for the characteristic flavour of grapes, musts and Muscat wines, but they can also be found in the semi flavoured varieties (in small quantities, sometimes even below the olfactory perception capacity). The terpenoides can be classified in isoprenoids and norisoprenoids. The isoprenoids class contains the terpenes and the terpenes compounds. The isoprenoids have methyl-butadiene at their basis, being able to generate monoterpenes, diterpenes, triterpenes, caroterpenoids, etc. They contain the polymers of the isoprenoids, and the terepenes and terpenes compounds are a part of them.

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THE TERPENES

The terpenes are part of a large and complex class; they are very wide spread in the vegetal reign, about 4000. Inside this class, the substances with an olfactory impact belong especially to the monoterpenes compound groups (substances with 10 carbon atoms built from 2 isoprenoids units) and sesquiterpenes (substances with 15 carbon atoms built from 3 isoprenoids units). There are about 20 terpenes in the grapes, must and wine, in general found in the form of monoterpenes, unsatured hidrocarcons like: mircen, ocimen, farnesen, cimen, limonene, terpinene, humulen, cadinen, terebenten, selinen, caren, cariofilen, copaen.

A team of researchers led, by Australian P.J. Williams, tried since 1981 to develop an adequate method to determine these aromas (Williams et al., 1981). The results appeared when the glycol glucose was analysed, succeeding through this method to establish the content of secondary metabolites of grapes, must and wine (Marais et al., 1983; Abbott et al., 1993).

The most important group of primary aromas are the terpenes and especially the monoterpenes. The monoterpenes are terpenoids compounds that result out of two isoprenes. According to Cordonnier and Bayonove (1974), Riebereau-Gayon et al. (1975), Williams et al. (1982) and Rapp et al. (1986), Mathotchina et al.(2012) the monoterpenes play a very important role in forming the aroma in the Muscat breed and other similar breeds.

According to Strauss et al. (1986), regarding the grapes in the Muscat breed, the total monoterpenes is of at least 6 mg / L while in the semi flavoured breeds, which have an aroma composed by independent monoterpenes, the total monoterpenes is less than 1mg/L.

The perception ridge of these compounds is situated at a fairly low level, between tens and hundreds micrograms in the case of the Muscat varieties (Muscat Ottonel, Muscat de Alexandria). Their concentration in these breeds is able to reach levels that are much higher than their perception ridge (Cordonnier et al., 1974).

They are also present in other varieties, especially in the German Gewürztraminer, Riesling, Müller-Thurgau, etc. or the French Auxerrois, Pinot gris, but they cannot explain certain olfactory nuance by themselves. Generally, the monoterpenes in the wine with simple aroma (Merlot, Cabernet Sauvignon or Franc) are below the perception ridges and thus cannot be detected by sensorial means (Terrier et al., 1972)(Wüst, 2003).

TERPENES COMPOUNDS

The terpenes compounds can be found in grapes, must and wine, free or combined, especially as glycoside, as precursors of aromas. The free terpenes compounds can be found under different forms as Figure 1 and 2 shows: aldehyde, (linalyl, geranial), alcoholic (linalool, geraniol), acid (linalic acid, geranic acid) or esters (linalil acetat). The most ordered monoterpenes are the monoterpenes alcohols, especially the linalool g/l. In this subclass, the most ordered substances are citronellol and linalool.

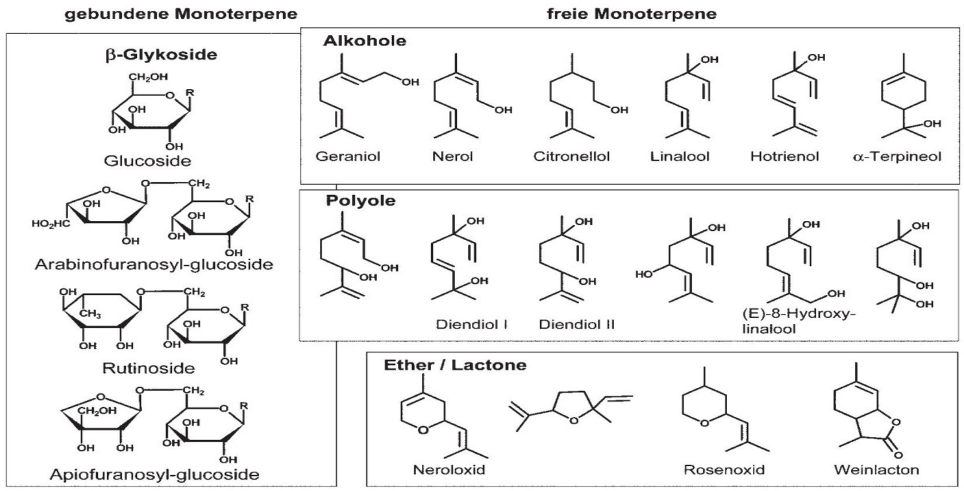


Figure 1. Free and bound monoterpenes (Wüst, 2003)

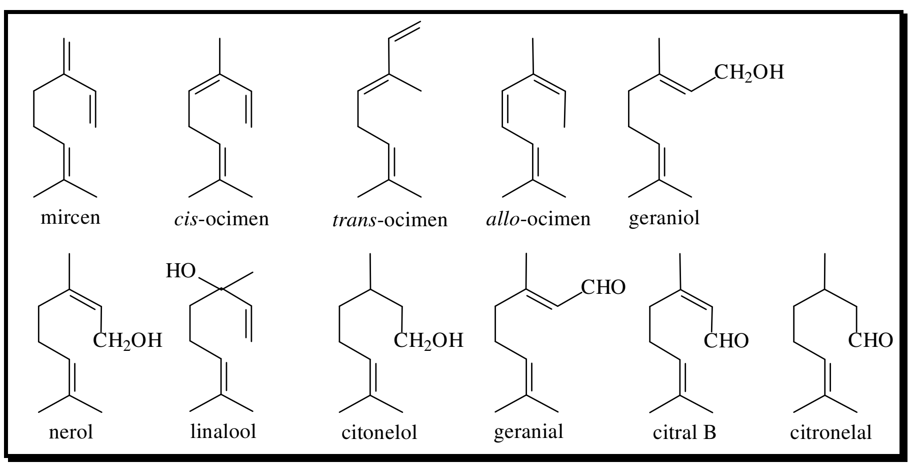


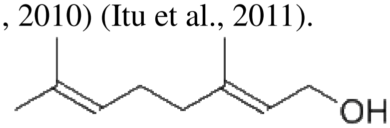
Figure 2. Acyclic monoterpenes (Georgescu, 2002)

The monoterpenes play an important role in the realization of the α–terpineol, nerol, geraniol, citronellol and hotrienol, which have a floral odour that reminds of roses. The free forms and the glycosides bound by linalool, nerol, geraniol, α – terpineol, cis-and trans-furan, linalool oxides, cis-and trans-pyran of linalool, have been determined out of the grape peel and must of Muscat de Alexandria and white Frontignac, determinations which concluded that in all the breeds the geraniol and nerol can be especially be associated to the peel of berries (Wilson et al., 1986).

TERPENES ALCOHOLS

This group contains a series of acyclic substances, monocyclic and bicyclical. The main terpenes alcohols that can be found in grapes are: geraniol, linalool, terpineol, nerol, citronellol, farnesol, but the first four present the highest values.

Geraniol is a colourless liquid, with a rose and citric floral perfume (Hoffman et al., 1973). It is found in aromatic breeds like Muscat Ottonel as high as 24,2% (Heroiu et al.,1994) and is concentrated in the peel. When fully mature it presents the full value, and then it slightly diminishes to around 10-12%. It presents a cis-trans isomeric structure. The distribution on the free monoterpenes and their glycolic bonds have been studied in the pulp and in the peel as well for some on the breeds of white grapes, the geraniol and other terpenic alcohols in the grapes leading to the creation of aromatic profiles for the aromatic breeds (Di Stefano et al., 1992) (Cotea et al., 2009) (Ţârdea et al., 2010) (Itu et al., 2011).

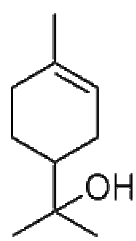


Geraniol

Linalool is a colourless liquid, with a fresh floral perfume, reminding of condiments and lemon. The linalool is transformed under the influence of acids in geraniol, nerol and α–terpineol. The linalool is joined by oxides in the grapes. The olfactory detection ridge for geraniol and linalool is about four to ten times smaller than for α-terpineol and nerol (Rapp et al., 1986). Having a very low perception ridge, it can be easily detected. The highest concentration of linalool is found in the flavoured and demy flavoured varieties especially in Muscat Ottonel, Tămâioasă Românească, Sauvignon or Fetească Albă, where it varies between 34%-54% (Heroiu, 1994). Under the action of the linalil-synthase enzyme, about one week after the grapes go into ripeness, the transformation of the geraniol-difosfat into S-linalool begins, this process being correlated with the free volatile linalool accumulation (De Billerbec et al., 2003).

Linalool

α-terpineol can be found in grapes, in smaller concentrations and it presents a very high olfactory perception ridge, and as a result it is very hard to distinguish. The α-terpineol emanates a perfume reminding of melon, sweet-floral, lilac like, spruce needle. It is formed out of monoterpen-glykoside in an acid environment (Strauss et al., 1986). According to Rapp and Mandery (1986) α-terpineol can be used as an ageing indicator, which grows proportionally to the growth of the exposure time. Due to the fact that every breed presents a personal configuration depending on the cultivation area, the pedological and climate conditions led to intense study in this direction. Recent studies have been conducted on the Sauvignon Blanc grapes, by Pinau et al., during 2011, in New Zealand, studies that cater for the grape maturation and the above mentioned factors, and which condition the sensorial proprieties of wine and the specificity degree.

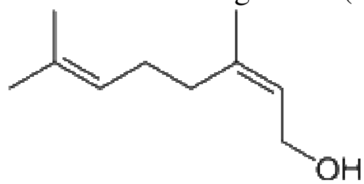


α-terpineol

The Gewürztraminer varieties have been submitted for identification and quantification of the main aromatic monoterpenes in grapes must and wine in 2008.

The utilized methods have been the solid micro-extraction SPME and the analysis on gas chromatography. The initial concentrations that have been obtained for the α-terpineol have been situated below 7 μg/L, but after using more pectolytic enzymes, the concentration rose to about 50% (Rusjan et.al., 2008).

Nerol is a colourless liquid with a fine perfume, reminding of a light plum miasma, roses, thyme and is accumulated in peels (Wüst, 2003) (Târdea, 2007). The chemical reactions of nerol correspond to the ones of geraniol. The geraniol and nerol are cis-trans isomers, with the same structure. The nerol goes through the cyclic process faster than geraniol and is transformed in α-terpineol. Both compounds are concentrated in the peels of grapes (Strauss et al., 1986). The nerol has a smell ridge of 400-500 mg / L, so it will take larger quantities for it to be distinguished (Wüst, 2003).



Nerol

OXIDES OF THE TERPENES ALCOHOLS

They accumulate in the peels of grapes, having a high perception ridge, without a major olfactory impact. In grapes, they can be found as cis-trans furanic of the linalool oxides, nerol oxides, piranic linalool oxides (Terrier et al., 1972) (Ţârdea, 2007).

GLYCOSIDES TERPENES

The glycosides terpenes results from combining the terpenes alcohols with carbohydrates from grapes. These carbohydrates can be glucose, arabinose, ramnose.

The first who studied and identified the terpenes and terpenoides in the linked form have been Di Stefano (1981) and Williams et al. (1981). This way, they established that the proprieties of the terpenes and linked terpenoides with a glucose form are not visible due to the glucose link. But through hydrolyse, a scission takes place, so the spread linked monoterpenes constitute a source of potential aroma.

The Muscat varieties contain the most important quantity of glycosides, although they can be found in all Vitis-vinifera samples. The glycosidase forms are in general more abundant than the free, ordered ones. The spread of free terpene from the glycosidase forms is realised under the action of the β-glycozidic enzymes (Wilson et al., 1986) (Buia, 2001) (Wüst, 2003).

The volatile compounds in the glycoside can be freed with help from the acids and by enzyme hydrolysis. The hydrolysis with acids of the glycoside terpenes can produce their transformation into monoterpenes, but they can also be transformed in other compounds.

The enzymatic hydrolysis is however recommended because the reaction provokes natural development of aromas in the wine (Gunata et al., 1994) (Ford et al., 1998) (Godden et al., 2005) (Guerin et al., 2009).

NORISOPRENOIDS

Norisoprenoids are a result of the oxidising decay of the carotene pigments which are themselves derivates of terpenes, with 40 carbon atoms. The β - damascenona , β-ionona and α-ionona are representative. During the ripe period, carotenes as β-carotenes, lutein can abruptly decrease while a norisoprenoid accumulation takes place as indicated in Figure 3 (Razungles et al., 1988) (Wüst, 2003).

β -damascenones present a flower and exotic fruit perfume, with a perception ridge at very low levels, β-ionone resembles viola, while 1,1,6-trimetil-1 ,2-dihydronaphtalena (TDN) determines a kerosene aroma (Strauss et al., 1987). Norisoprenoides have been identified in the aroma of Chardonnay, Sauvignon Blanc and la Semillon (Marais et al., 1992) (Wüst, 2003).

NONTERPENOIDES

Nonterpenoides are a very broad ordinate substance category that is composed of hydrocarbons, pyrazines, sulphur compounds, phenol compounds, aldehides, cetones, esters, ethers, acids, superior acids, acetamides (Tirdea, 2007).

DERIVATES OF THE BENZOIC ACID

The derivates of the benzoic acid come from the plant itself, being aromatic substances, from a chemical point of view. The following two types of benzoic links can be found in grapes: 2-phenylethanol and benzaldehyde .

2-phenylethanol is responsible for the rose aroma in the grapes, reaching values as high as 100mg/L. This can influence the aromatic character of the final wine (Fischer et al., 2001). Phenols, which belong to this group, cannot be considered aromatic substances, but they play a very important role in the formation of anthocyanins in the red varieties, which is responsible for grape color.With its catechetic form, it contributes at the formation of taste and astringent quality of the red wines (Papargyriou , 2003).

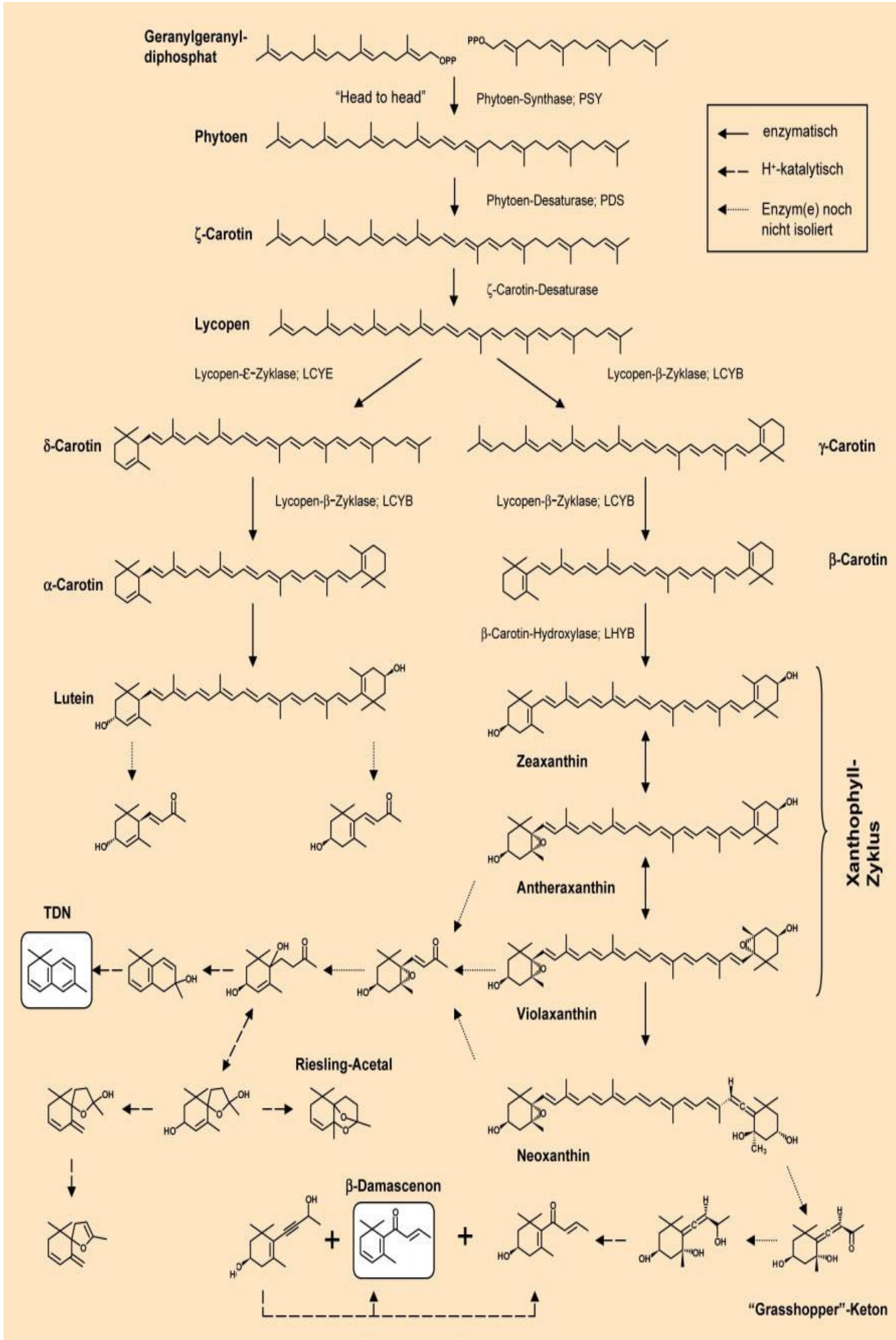


Figure 3. Biosynthesis of carotenoids and obtainment of important norisoprenoides for the flavours of grapes (Wüst, 2003)

PRECURSORS TO THE CYSTEINES OR S-CONJUGATES OF L-CYSTEINES

They are inodorous precursors to the compounds with functions of very aromatic thiols, and have recently been relieved and identified directly in the grapes. Due to the analytical difficulties, little quantitative data has been published regarding S-conjugates of cysteines. As Figure 4 shows the biosynthesis of cysteines takes place in the presence of the enzymes in the grapes during the alcoholic fermentation (Wüst, 2003).
In the grape, P4MMP (S-(4-metil-2-oxopent-4-il)-L-cysteine) is equally divided in the peel and pulp, while the P3MH (S-(1-hidroxihex-3-il)-L-cysteine) is mostly present in the peel (Tominaga et al., 1995) (Swiegers et al., 2007) (King et al., 2011).

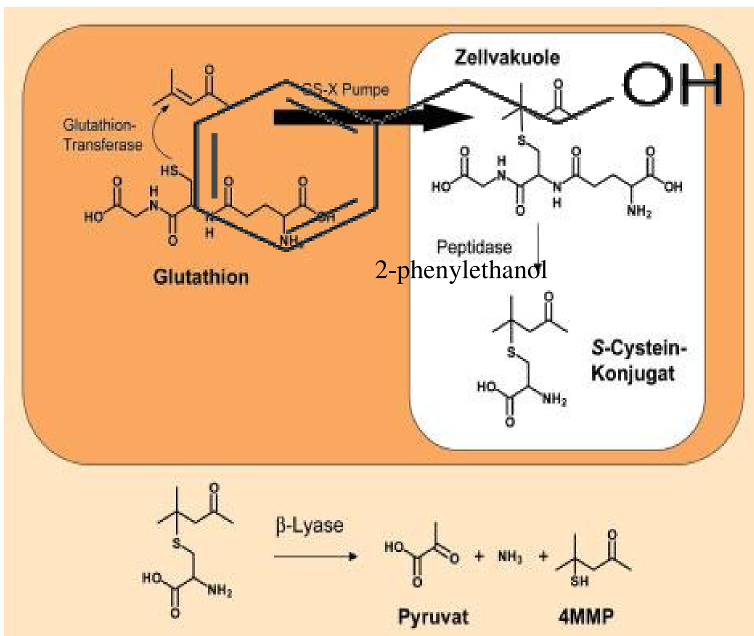


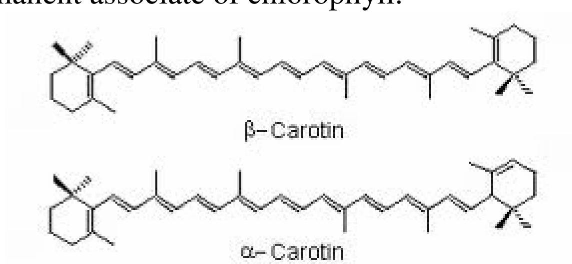
Figure 4. Biosynthesis of the cysteine in the presence of beta lyases (Wüst, 2003)

PRECURSORS TO DIMETHYL SULFUR

Precursors to dimethyl sulfur, which belong to the varietal aroma, have been recently identified in grapes, although the dimethyl sulfur (DMS) is known as an aromatic constituent of wine. It can be found in wines, in quantities which oscillate between 4-10 μg/L. Its aroma was highlighted during different stages of the vinification and wine storage process. DMS has a perception ridge of 27 μg/L in the red wine, olfactory note which was frequently mentioned in the reducing bouquet of high quality red wines and in the case of wines obtained from late harvested grapes (Eglinton et al., 1996) (Swiegers et al.,2005) (Bell et al., 2005) (Makhotkina et al., 2012).

CAROTENOIDES AROMATIC COMPOUNDS

Carotenoides are hydrocarbons with a poli-isometric structure $(C_5H_8)n$, the majority having molecules composed out of 40 carbon atoms.
The main carotenoides that are identified in grapes are: lutein, β- carotene, neoxantin, flavoxantin, violaxantin, luteoxantin and 5,6- epoxilutein, the more abundant being lutein and β- carotene. They are accumulated in the peel of the grape, in big quantities of 2,5 g/kg, depending on the breed and maturation degree of the grapes (Chkaiban et al. 2007) (Lashbrooke et al. 2010).
β-carotene is most the spread in nature, being present in all green organs of plants, as a permanent associate of chlorophyll.



Lutein appears in red and in yellow flowers, in fruit, algae and mushrooms.
During the ripeness period of grapes, when the colour of the grape turns pink-lavender, an important decrease of carotene begins (Valtaud et al. 2011). They are absent in the must due to their insolubility, their presence in the wine being excluded without the preferment maceration of the peels together with the must.

PHENOL AROMATIC COMPOUNDS

From the multitude of phenol compounds that are formed in grapes, the hydroxybenzoic and hydroxycinnamic phenol acids participate in the formation of wine aromas. The phenol acids are not ordered themselves, but through esterification with the alcohols in the wine, the specific aromatic compound will be formed, like ethyl and methyl vanillate, ethyl cinnamate (Methyl trans-3-phenylpropenoate).

The ethyl and methyl vanillates, with a pleasant vanilla flavour, are characteristic to wines obtained through carbonic maceration (Terrier et al. 1998) (Parley et al. 2001) (Du Toit et al. 2006) (Etaino et al., 2008).

The ethyl cinnamate has as precursor the ferulic acid and it imprints a fruity aroma to the wine. The olfactory perception ridge is very low, 0,048 mg/L.

The thyolic compounds like mercaptopentanon reacts with the oxidated phenol compounds (thyols). These kinds of molecules are formed in large quantities when the most is pressed, must which also contains large quantities of precursors to aromas.

AROMATIC PYRAZINES COMPOUNDS

Pyrazines is a heterocyclic substance with a role of odorant, actively participating in the formation of primary aroma. Their concentration varies very much depending on the breed, on the technology that is applied, the pedological and climatic conditions. The pyrazines is a heterocyclic substance with a cycle composed of 6 atoms, two of which are azoth. Their origin in grapes and plants is unknown; the possible precursors are amynic acids, (leucine, isoleucine, valine and glyoxalin). The odorant character of pyrazine results from their low basicity and the resistance to oxidation. The highest pyrazines content can be found in the uncured grapes (Allen et al., 1991) (Lacey et al., 1991) (Lawles et al., 2010). During maturation the content diminishes, the exposure of grapes to sun light limiting the pyrazines content. Methoxypyrazines are heterocyclics with azoth, derivates of pyrazines, and derivates by a metabolic transformation of some amino acids. These compounds present a green pepper, vegetal, with earth and herb nuance perfume, that can also be perceived in very small concentrations, as low as ng/l (Hartmann et al., 2002) (Howell et al., 2004) (Belancic et al., 2007) (Itu et al., 2011).

THIOLS

Compounds with sulphur are generally considered as being responsible for odour defects, but some substances with favourable effect on the bouquet of the wine can be found in this group. These compounds confer aromas of black berry, grapefruit, guava or other exotic fruit (Hernanz et al., 2009) (Mateo et al., 2010). The mercaptans are part of the compounds with thiol functions, two of them being frequently met in Vitis Vinifera: 3-mercaptopropionate of ethyl and 2- mercaptopropionate of ethyl (Tominaga et al., 1996) (Tominaga et al., 1998) (Tominaga et al., 2006) (Herbst et al., 2011).

CONCLUSIONS

The complexity of the compounds that offer grapes and wine the aroma has been subject to many studies, but these compounds have not been totally deciphered. Flavour compounds occur in low concentrations in raisin, most and wine. Non aromatic musts contain non-volatile aromas precursors that are transformated by enzyme action to odorous compounds with important roles as aroma precursors. Due to the different physical, chemical, agro-biological, microbiological or biochemical influences, each variety represents an enigma waiting to be deciphered.

Aknowledgements

This work was co financed from the European Social Fund through Sectoral Operational Programme Human Resources Development 2007-2013, project numbered 76851"Harmonization of Romanian academic valences to those of European Community coordinator Phd .eng. Ovidiu Tita

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