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AROMATIC COMPOUNDS IN WINES

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Abstract

The aroma of wines is represented by a complex of volatile compounds coming from several sources, as well as their interaction with the other chemical substances of the wine: water, ethyl alcohol and other alcohols, phenolic compounds, polysaccharides, fatty acids, etc. The main compounds that participate in the aroma of wine and which give, for the most part its typical flavour are the primary or varietal flavours, coming from grapes and which, besides the phenolic compound's secondary metabolites of vineyards. These compounds are in the free state or in the form of flavour precursors that are in a bonded state. During the processing of grapes and musts a multitude of volatile compounds are formed which together with the primary aromas give the aroma of the young wine, typical of the variety from which it originates, as well as terroir. The Saccharomyces cerevisiae yeasts have an essential role in the complex of fermentation flavors.Thus, in addition to the flavors formed in the fermentative process, other volatile compounds are formed by the action of the yeast on the primary flavors. In the case of matured wines, the flavour is complemented by compounds formed during the maturation period, forming the so-called wine bouquet.

Key words**:** *Varietal flavours, terpene, methoxypyrazines, volatile thiols, 3-mercapto-hexanol*

INTRODUCTION

The composition of wines in aromatic compounds is a basic component of their quality, the odorant compounds being responsible, for the most part, of the typical wines [26]. Thus, although they are secondary metabolites of grapevines, grape aromas are an essential component of the quality of the harvest and an important basis for the aromatic quality of wines [4], [14].

The aromatic substances are in the epicarp (in the cells of the hypodermis and epidermis), and in some varieties, called aromatic and semi aromatic varieties (*Muscat Ottonel, Traminer, Tămâioasa Românească, Busuioaca de Bohotin* - aromatic varieties, *Sauvignon* - semi aromatic variety) are also present in the grain pulp [13], [17]. Grapes of aromatic varieties contain, besides a high concentration of aromatics and specific molecules, called terpinols, which may constitute "key substances" or "signature" of the variety [8], [13]. Thus, depending on the aromatic variety potential *Vitis vinifera* varieties have been classified into two classes:

-simple flavour: Grapes are not flavoured, but the resulting wine contains flavouring substances derived from compounds called flavour precursors. These compounds are found in grapes in a bound form, being odourless.

Some examples of varieties belonging to this group: *Sarba, Cabernet Sauvignon, Pinot noir, Chardonnay, Sauvignon* (considered to be a semi aromatic variety).

-aromatic varieties, in which the grapes contain chemical odorants. Although they are found in very low concentrations, in the order of ng/L, these compounds are perceptible since most of them are in a free, odorous form.

From this variety class belong the varieties *Muscat (Muscat Ottonel, Muscat de Hamburg, Muscat d'Adda, Muscat de Alexandria), Tămâioasa Românească, Traminer*.

The aromatic potential of the grapes is changed during vinification. In addition to varietal aromas (also called primary flavours), other classes of aroma compounds, which are of major importance in the aromatic quality of wines, are found in wine [2]. Thus, they are: preferential flavours, which are born during the processing of grapes, enzymatically [6]; the aroma of fermentation (secondary flavours), which is formed during the fermentative process [5], through the metabolism of the yeast; the maturing aromas (wine bouquet)

that are formed post-fermenting during the maturing and aging process (Fig.1).

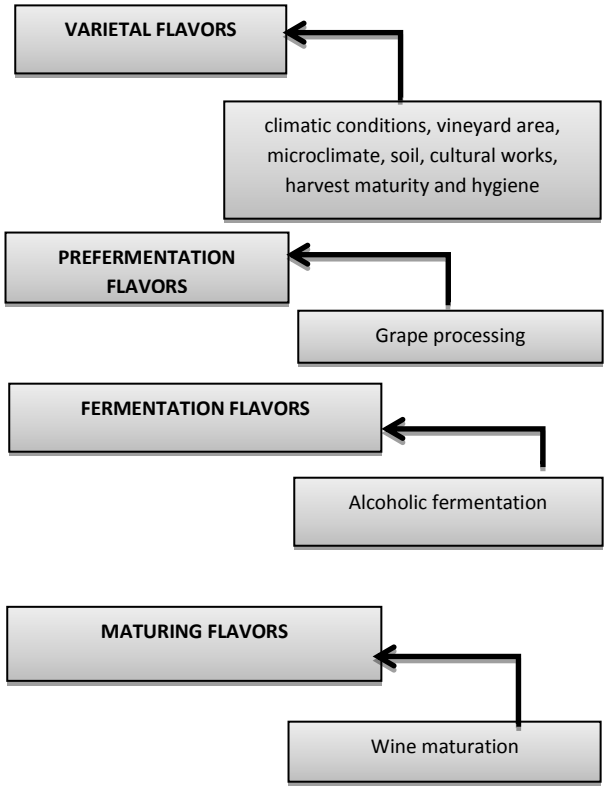

Fig. 1. Wine flavors
Source: Subileau M., Schneider R., 2008

During alcoholic fermentation, a very important class of flavours is formed which have an essential role for the aroma of wine. These compounds are born through a multitude of physicochemical and enzymatic processes due to levurian activity. In the fermentation process the yeast *Saccharomyces cerevisiae* acts on the terpenic compounds, by hydrolysis of glycosides and isomerization and reduction of free terpenic alcohols, leading to the production of aromatic compounds, compounds that give the typical wines [5].

Table 1. Classification of flavours according to the odour character imprinted on wines

Series	The flavor imprinted on wines
Animal series	musk, smell of meat
Balsamic series	amber, resin, conifer
Chemical series	mercaptan, phenol, sulfur
Wood series	bark, cedar, oak, pencil
The empireumatic series	cocoa, coffee, smoke, leather
Spicy series	anise, cinnamon, mint, pepper, sweet wood
The floral series	iris, peony, rose, violet
The fruity series	blueberries, cherries, pears, plums, strawberries, pineapples, bananas
Plant series	grass, hay, olives, pepper, tobacco, truffles

Source: http://biosol.free.fr.

Finally, the aroma of wines is a complex assimilation, due to a very large number of volatile compounds (over 1000 compounds), compounds that impart a specific odor character to wines [15]. From the point of view of this olfactory character, the aromas of the wines were classified in 9 flavoured series (Table 1).

MATERIALS AND METHODS

To highlight the complexity of wine aroma and the origin of volatile compounds, numerous bibliographic databases have been studied.

RESULTS AND DISCUSSIONS

I. Varietal flavours

These chemical compounds are born in grapes through the secondary metabolism of the grapevine [2], [3]. Primary flavouring compounds (varieties) are found in grape berries as free aromas and aroma precursors [14].

(i)Free aromas are made of olfactory perceptible compounds: terpenic compounds (terpene and terpenoids), methoxypyrazines and rotundones.

Terpenic compounds have tropical floral and tropical flavours and are found in aromatic varieties. Terpenes are found in grapes most often in the form of monoterpenes: α-terpineol, linalol, nerol, geraniol and citronelol [7], [16]. These terpenic compounds are attributed to various flavours: rose (geraniol, nerol), coriander, rosewood (linalol), field flowers (linalol oxide), citrus (citronelol) etc. and determine the floral flavour specific to the *Muscat* geraniol, nerol, linalol), *Tămâioasă* varieties (a-terpineol), *Traminer* and other [15].

The terpenic content varies depending on the variety, from very low content to simple varieties (*Feteasca, Cabernet*) of about 2 μg/L, at a higher concentration of approx. 2000 μg/L for *Muscat* varieties [7]. In the Table 2 are shows the terpenols identified and dosed in the *Muscat* white variety [31].

Methoxypyrazines are free aromas, with plant flavour, green pepper, capsicum, etc. The flavour of these compounds can have an undesirable effect on wines, with a negative impact on their quality or, on the contrary, they can cause a complex flavour, typical of the *Cabernet* sortgroup and the *Sauvignon* variety [1], [10].

The biosynthesis of methoxypyrazines in the vine is associated with the metabolism of the amino acids: leucine, isoleucine, valine and glyoxal [18], [24], [32].

Table 2. Terpenols content in grape wort *Muscat alb* variety (µg /L)

Terpenols	Free	Combine
trans-furaninaloloxid	33	44
cis-furaninaloloxid	18	33
Linalol	291	95
ho-trienol	19	19
α-terpineol	27	26
trans-piranlinaloxid	96	12
cis-piranlinaloxid	39	3
Nerol	25	160
Geraniol	45	247
3,7-dimetil-1,5-octadien-3,7-diolo	282	534

Source: Sârghi, C., Zironi, R., 1994.

Rotundone is another class of free flavours and is responsible for spicy flavour, white pepper and black pepper, specific to the *Syrah* variety. In fact, these flavourings have been identified in *Syrah* wines from Australia [25]. Subsequently, they were also identified in other wines: in the Italian red wines *Schioppettino, Vespolina* and *Groppello di Revò;* in the Austrian white wines of the Grüener Veltliner variety and in the French wines of the *Gamay* and *Pineau d'Aunis* varieties. Rotundones are very odorous molecules, their perception threshold being very low, 8 ng/L in water and 16 ng/L in wine. For example, *Syrah* wines, and those of *Schioppettino* and *Pineau d'Aunis* show concentrations of about 200 ng/L.

(ii)<u>The flavour precursors</u> are unnatural compounds in the bound form (compounds bound to other molecules). These compounds (fatty acids, glycosides, phenolic acids), during the processing of grapes and alcoholic fermentation are cleaved, thus obtaining volatile odorants [9]. The aroma precursors consist of chemically and organoleptically different substances, grouped into: glyosidic precursors, carotenoids, precursors of volatile thiols and dimethylsulfurite precursors [13].

The *glycosidic precursors* identified in grapes are: β-D-glucopyranoside; α-L-Ramnosyl: 6-O- (α-L-ramnopyranosyl) -β-D-glucopyranoside; α-L-Arabinofuranosyl: 6-O-α-L-arabinofuranosyl) -β-D-glucopyranoside; β-D-Apiosil: 6-O- (β-D-apofuranosyl) -β-D-glucopyranoside [30].

Carotenoids identified in grapes are the precursors of C13-norisoprenoids, the free forms of which are represented by a variety of compounds with a great aromatic diversity [3]: β-damascenone, with flower aroma, exotic fruits, plum jam, apples etc. identified in all wines); β-ionone, with violet flavour (compound perceived only in red wines); 1,1,6-Trimethyl-1,2-dihydronaphthalene (TDN) with kerosene flavour (compound to be found in Riesling's old wines); isotypes of vitisperine, with camphor aroma and eucalyptus; (E) -1- (2,3,6-trimethylphenyl) buta-1,3-diene with freshly cut grass aroma [16].

Precursors of volatile thiols. In grapes, thiols components are found only as precursors, non-volatile and odorous. Odorant character appears in wine during the fermentation process, thiol precursors being degraded by the yeasts of *Saccharomyces cerevisiae*, under the action of S-β-lyse enzymes [5]. Among the precursors of the volatile thiols identified in grapes, we mention: the precursors Cys-4MMP and Cys-3MH [39**], [**42], cysteine-linked molecules and a glutathione-containing 3-mercapto-hexanol thiol precursor identified in the *Sauvignon* variety (Table 3). In addition to these compounds, a *precursor of dimethyl sulfide*, a compound responsible for the typical *Syrah* [33**], [**34]. Segurel and others. shows that the most likely compound of DMS (dimethyl sulfide) in wine is S-methylthionine [13**], [**14**], [**33].

Table 3. Content in flavour precursors of *Sauvignon* variety (ng/L)

The Thiol precursors	**Contents**
4-mercapto-4-methylpentan-2-one	715
4-mercapto-4-methylpentan-2-ol	2059
3-mercapto-hexanol	14812

Source: Tominaga, T. et al., 1998; Deloire A., 2007.

II. Fermentation flavours

Fermentative aromas are also called secondary flavours and are born through the secondary metabolism of yeasts during alcoholic fermentation of grape must [19]. If the varietal flavours are very important for the aroma of wines, being responsible for their typical character, the fermentation aromas have an essential role, by imprinting the wine character of the wine, specific to this alcoholic beverage [5], [23]. Schneider shows the importance of leafy wines in wine making and the fact that a correct choice of spruce can interfere with the final flavour of wines. It also emphasizes that *Saccharomyces cerevisiae* not only leads to the formation of aromatic compounds derived from the secondary metabolism of yeast but can also interfere on varietal flavouring compounds [5], [35]. During the alcoholic fermentation process, higher alcohols, volatile fatty acids, aldehydes, esters, sulphur compounds, etc. - compounds with direct or indirect impact on the olfactory character of wine [20] are obtained by the enzymes produced by the yeasts. These compounds form the class of fermentation flavours [6].

1.<u>Group of superior alcohols</u>. Depending on the assimilable nitrogen level for the grape wort yeast

and the sugar concentration of the grape wort, higher alcohols are born through one of two metabolic pathways [26]:
-through the Ehrlich pathway, corresponding to the catabolism of the amino acids;
-from the anabolic pathway of amino acids, starting from sugars.
The concentration of wines in higher alcohols is quite low, 400-500 mg/L, most of them being found in concentrations lower than their sensory threshold of perception [6]. Except for 2-phenylethanol, which has rose floral aroma, the rest of the higher alcohols have a slightly aromatic flavour of wine.

Table 4. Superior alcohols present in wine

Higher alcohols	**Precursors**	**Aromatic notes**	**Content in wines**
D-amyl alcohol (2-methyl-1-butanol, Pentan-1-ol)	isoleucine	nail polish, solvent malt	8-150 mg/L
Isoamyl alcohol (3-methyl-1-butanol)	leucine	alcohol notes, nail varnish, solvent amilic notes, malt, fragipan, pungent, unpleasant smell	under the 0,2 g/L
Propanol		solvent odor, chemical	5-125 mg/L
Isobutanol or isobutyl alcohol (2-methyl-1-propanol)	valine	solvent, chemical alcoholic, malt notes pronounced wineosity note	9-174 mg/L
Butanol		alcoholic notes, alcoholics participates in fruity wine notes	traces - 8 mg/L
2-feniletanol	fenilalanine	floral, rose, honey notes, peach notes	4-197 mg/L
3-metiltiopropanol	metionine	cruel potatoes	
Thyrozol	from 2-phenyletha nol	bitter taste, honey odor	
Tryptophol	triptofan	alcoholic notes	

Source: Nykanen, L., 1986

Numerous authors believe that a low concentration of higher alcohols (below 300 mg/L) leads to a pleasant wine aroma; however, a concentration above 400 mg/L of higher alcohols is undesirable for its quality [20]. The main superior alcohols present in wine are shown in Table 4.
2.<u>The fatty acid group</u> comprises short chain fatty acids formed during yeast alcoholic fermentation by lipid metabolism. Volatile fatty acids result from acyl-S-CoA hydrolysis, either by the synthetic anabolic pathway (by which most fatty acids are formed) or by β-oxidation of the lipids. The production of volatile fatty acids by lipid oxidation occurs at the beginning of the fermentation process under the action of dissolved oxygen in the wort. Of the volatile compounds formed during the fermentation, higher alcohols and volatile fatty acids are linked to the "gross" character of the wine's flavour. However, there are authors who claim that volatile fatty acids have their importance in wine by helping to balance flavours.

Table 5. Volatile fatty acids and aldehydes present in wine

Fatty acids and aldehydes	Precursors	Aromatic notes
Volatile fatty acids		
acetic acid	asparagine, glycine, serine, alanine	sour smell, vinegar, pungent
propionic acid	threonine	disagreeable smell, sour
2-methylpropanoic acid (isobutyric acid)	Valine	apple rot, butter ran, sweat
2-methylbutanoic acid	isoleucine	fruity, sweat, sour
3-methylbutanoic acid (isovalerianic acid)	leucine	rotten fruits sweat
phenylacetic acid	phenylalanine	floral, green notes
hydroxyphenyl acid	tyrozine	floral, green notes
methylthiopropionic acid	methionine	
Aldehyde		
aldehidă acetică	threonine	green notes, ether, sour smell
2-methylpropanal	Valină	green notes, malt, animalic
2-methylbutanal (isovalerian aldehyde)	isoleucine	green notes, malt, animalic
3-metilbutanal	leucine	green notes, malt, animalic
Phenylacetaldehyde	phenylalanine	floral notes rose
benzaldehidă	phenylalanine	bitter, almond smell, sweet
3-metiltiopropanal	methionine	boiled potatoes
methylthioacetaldehyde	methionine	green notes, apples
Hexanal (caproic aldehyde)		Sweet, disgusting, floral, fruity
Heptanal		
Octanal (caprylic aldehyde)		Citrus, lemon, sweet, sour

Source: Lonvaud-Funel A.et al., 2010.

Fatty acids have unpleasant odors, but the content of wines in these compounds rarely exceeds their perceptual threshold, except for 2-methylbutanoic acid and 3-methylbutanoic acid (Table 5). As for branched aldehydes with pleasant smells of dried fruit, their contribution to the aroma of wines is positive [43].
3.<u>The group of esters</u>. Esters are volatile compounds essential to the aroma of young wines. Most of the esters have floral odours (rose, jasmine) and fruits (green apples, strawberries, pineapples) The formation of the esters is due to the metabolism of levurian and is achieved by enzymatic reactions involving acyl-S-CoA. Thus,

in grape wort found in fermented process, in which acetyl-coenzyme A and ethyl alcohol predominate, the yeasts generate fatty acids, but also their ethyl esters, as well as acetates of superior alcohols [36]. During the alcoholic fermentation three groups of esters are formed:
-ethyl esters of linear fatty acids; are formed through lipid metabolism;
ethyl esters of branched or hydroxylated fatty acids;
-acetates of superior alcohols; the latter two groups are associated with nitrogen metabolism. The main esters involved in wine flavour are presented in Table 6.

Table 6. Esters that participate in the aroma of wines

Esters	Aromatic note	Detection threshold µg/L
Ethyl butanoate	Pineapple, strawberries	20
Ethyl Hexanoate	green apples, strawberries, pineapples, blackberries	14
Ethyl Octanoate	floral, fruity, pear, soap	2
Ethyl Decanoat	floral, fruity, soap	200-500
Ethyl acetate	neplăcut, de dizolvant, fructat unpleasant, solvent, fruity	
Butyl acetate	Banana, floral, fruity	1 ppm
Ethyl Propanoate	cherries	
2-methylbutyl acetate	fruity	
3-methylbutyl acetate (isoamyl acetate)	bananas, ripe apples, candy	2000-3000
2-phenylethyl acetate	rose, fruity	2000
2--methylpropanoat of ethyl	strawberries, blackberries	
2-methylbutanoate of ethyl	apples, strawberries (chemical)	
3-methyl butanoate of ethyl	green fruits, blueberries, strawberries (chemical)	
Hexyl acetate	pear, plum, banana, currant	15 ppm
Linalool acetate	bergamot	
Benzyl acetate	apple, pear, jasmine	

Source: : Ribereau-Gayon J. et al., 1972

4.<u>Group of sulphur compounds</u>. The sulphur compounds present in the wine come from both grapes (varieties of sulphur, such as precursors of volatile thiols) and from the fermentation process (at the end of alcoholic fermentation) through sulphur and nitrogen metabolism [29], [36]. From the sulphur compounds, have importance the mercaptans (volatile thiols) and the sulphur fermentation compounds.
Mercaptans (volatile thiols) are part of the sulphur compound group and are of great importance in the aroma of wines, attended by exotic fruit aromas (pomelo, the fruit of passion). Volatile thiols were identified in different wines: *Sauvignon*, *Gewurztraminer*, *Riesling*, *Muscat of Alsace*, *Cabernet*, etc. [37; 38; 40; 41]. About *Sauvignon* wine, two volatile thiols participate in the flavour specific to this wine: 3-mercapto-hexanol and 3-mercaptohexyl acetate [41]. According to some authors, the flavour specific to *Sauvignon* wine is given by the thiol 4-mercapto-4-methylpentan-ol, which has been identified in concentrations between 5 and 50 ng/L ([11], [21], [22], [44]).
In the wines was identified 4 volatile thiols: 3-mercapto-hexanol-3MH; 3-mercapto-hexyl-A-3MH acetate; 4-mercapto-4-methylpentan-2-one-4MMP; 4-mercapto-4-methylpentan-2-ol-4MMPOH. These thiols are formed during alcoholic fermentation by enzymatic degradation (S-β-lyse type enzymes) of cysteine precursors, mainly Cys-4MMP and Cys-3MH [11], [12], [22], [36], [44].
The concentration of wines in volatile thiols is linked, on the one hand, to the content of grapes in precursors, and on the other hand to the levs involved in the alcoholic fermentation [29], [36].
Sulphur fermentation compounds. Although the concentration in wine does not exceed their threshold of olfactory perception, the contribution of these compounds to the fermentation of the wine depends on their molecular weight and their volatility. sulphur compounds may be classified, depending on the above-mentioned characteristics, in light and heavy or higher compounds [28].
The " lightweight " compounds are associated with the olfactory defects of the wines, they produce an unpleasant, repellent aroma ("reduced" smell, spoiled eggs, mushrooms, garlic etc.). However, under optimal fermentation conditions it does not distort the aroma of wine, being present in low concentration below the level of sensory perception.
During the alcoholic fermentation, most of these compounds are eliminated from the wine, being trained by CO_2 bubbles, and only a small part remains in wine and participates in the olfactory balance and the overall flavour of the wine. There are cases when alcoholic fermentation does not normally occur due to unfavourable conditions (temperature, speed, extreme anaerobiosis), certain strains of yew or because of the high content of grape wort in sulphates, sulphites, assimilable nitrogen [27]. In this case, the concentration of sulphur compounds increases and there are various olfactory defects that can compromise the wine.
"Heavy" or "superior" compounds with a complex aroma of wine are 2-methylthioethanol, 3-methylthiopropanol and their derivatives

(corresponding thioethers and disulphides). These compounds are actively participating in the aroma of young wines, the concentrations of which are around 0.1 mg/l. However, 3-methylthiopropanol and N-(3-methylthiopropyl) acetamide may exceed the value of 2-3 mg/L. The main sulphur fermentation compounds in wines are presented in Table 7 [2].

Table 7. Sulphur fermentation compounds present in wine

sulphur compound	Aromatic note
Thioethyl acetate	Burner, sulphurous
Thiomethyl acetate	Cheese, rotten plants
Acetyl 2-thiazoline	Rice, popcorn
Methyl-3-propionic acid	Grill, flavour of maderization
Benzothiazole	rubber
Dimethyl disulphide	Quince, asparagus
Ethan thiol	onion
Furan methane-2-thiol	Coffee
Hydrogen sulfide	Rotten egg
2-mercapto-ethanol	Burned rubber, barley smell
Methane thiol	Smell of low
Methyl thio-2-ethanol	Cauliflower
Methyl-2- -tetrahidrotiofenone	Gas
Dimethyl sulphide	Quince, asparagus, truffle

Source: Ségurel, M. A., 2005

CONCLUSIONS

The aromatic potential of wines involves a series of aromatic compounds that come from both grapes (the primary or varietal flavours) and their vinification, mainly formed during alcoholic fermentation (secondary or fermentation flavours). From the point of view of the varietal aroma, the varieties are classified in aromatic varieties, where the grapes contain odorant chemical compounds, perceptible due to the fact that most of them are in free, odorous form and varieties with a simple flavour, to which grapes are more or less odorless and do not contain terpenes the typical character of the wine being given by other volatile compounds formed during the fermentation process.

Primary or varietal flavours are represented of chemical compounds found in grapes in free form as well as in the form of flavour precursors.

Secondary or fermentation flavours are due to levurian metabolism, as well as its action on primary flavors; the secondary flavors are represented by several groups of volatile compounds: the group of higher alcohols; the group of volatile fatty acids and aldehydes (these groups form the "gross" character of the wine flavor), the group of esters; the group of sulphur compounds.

The latter group includes volatile mercaptans or volatile thiols and sulphur fermentation compounds, which in turn are "light" compounds associated with olfactory defects of wines and "heavy" or higher sulfur compounds that actively participate in the formation of the aroma of young wines.

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