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PHENOLIC COMPOUNDS OF RED WINES IN PODGORICA SUBREGION (MONTENEGRO)

SUMMARY

The paper presents the results of the investigation into polyphenol composition of wines Vranac, Kratosija and Cabernet Sauvignon produced in Podgorica subregion (Montenegro) in 2013. The wine was manufactured according to a uniform manner (traditional method) from grapes cultivated in different locations. The process was carried out in the winery of the Biotechnical Faculty, located at the experimental farm in Podgorica, at the vineyard location of Ljeskopolje. The analysis of basic chemical parameters was carried out and polyphenolic compounds in wine - total phenolics, total anthocyanins, low and high molecular proanthocyanidins were investigated in more detail by using spectrometry.

On average, the highest content of total phenolics (TP), low molecular weight (LMP) and high molecular proanthocyanidins (HMP) was found in the wine Cabernet Sauvignon, while the lowest content of low molecular proanthocyanidins (LMP) and the highest content of total anthocyanins (TA) was found in the wine Vranac. The results showed that the variety has a dominant impact on the content of polyphenols in wine.

Key words: red wine, Vranac, Kratosija, Cabernet Sauvignon, polyphenol composition

INTRODUCTION

The chemical composition of grape must has a significant impact on the quality of wine, and not only concerning the quantity of particular ingredients, but also the ratio of their quantities. (Pajovic *et al.*, 2013, Raicevic *et al.*, 2014). Sugar content, total acidity and pH values of must are important quality indicators determining the future quality of wine.

Polyphenolic compounds are a large and complex group responsible for the characteristics, colour and quality of wines, especially for red wines. The polyphenolic contents of wine consist of flavonoids and non-flavonoids and depend on the grape variety, vineyard location, cultivation system, climate, and soil type, vine cultivation practices, harvesting time, production process, and aging (Ribéreau-Gayon *et al.*, 2000, Hanlin *et al.*, 2011). Anthocyanins belong

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to the flavonoid group of polyphenols and are responsible for the red colour and colour stability in wine (Somers, 1971, Ribéreau-Gayon, 1982). Proanthocyanidins, as condensed tannins, are the compounds responsible for bitterness, astringency and structure of the wine (Chira *et al.*, 2009, Ivanova *et al.*, 2012).

Besides being technologically very important, polyphenols are also powerful antioxidants that play an important role in the maintenance of human health (Rodrigo *et al.*, 2011, Vichapong *et al.*, 2014).

Important factors that affect the quality of the grapes, and therefore the polyphenolic composition of wine are the grape variety and location, i.e. climatic and soil conditions (Ribereau- Gayon, 1982, Lee *et al.*, 2009, Mativi *et al.*, 2009, Roullier-Gall *et al.*, 2014).

The significant impact of the variety on the polyphenols in wine was shown in the research conducted by Pajovic *et al.* (2014a) in 2011 and 2012, whose aim was to characterise Vranac, Kratosija and Cabernet Sauvignon wines according to the polyphenol content. It was found that variety Kratosija has a lower polyphenol potential in comparison to Vranac and Cabernet Sauvignon. The highest total polyphenols, low molecular weight and high molecular proanthocyanidins were assessed in wine Cabernet Sauvignon, in both years of investigation, while Vranac had the highest content of anthocyanins in all studied locations.

Autochthonous grapevine variety Vranac is the most common variety in Montenegro (Ulicevic, 1966, Avramov, 1991, Bozinovic, 2005, Pejovic, 1987, Cindric, 2000, Popovic, 2013). Population of cultivar Kratosija is very heterogeneous, which explains the low presence in the vineyards of Montenegro and mostly in combination with Vranac variety (Maras *et al.*, 2004).

Vranac is the dominant grape among red grapes, whereas Kratosija is decreasing and Cabernet Sauvignon is increasing (Pajovic *et al.*, 2014a). Vranac variety is characterized by a high good phenolic content, as previous published for Vranac grape and wines grown and produced in Montenegro and Macedonia, (Avramov, 1991, Ivanova *et al.*, 2015, Pajovic *et al.*, 2014a, Raicevic *et al.*, 2014). It is characterized by a high concentration of anthocyanins in the skins, even in the pulp of the berry and wine (Avramov 1991; Cindric *et al.*, 2000, Pajović *et al.*, 2014b).

The contents of polyphenolic compounds is largely affected by the microclimate of vines, by the amount and distribution of rainfall in a particular location (Van Leeuwen *et al.*, 2004 and 2004b), as well as by the slope exposure and elevation. The average atmospheric temperature decreases with an increase in altitude, resulting in lower sugar content and increased content of total acids (Fregoni, 1973, Rankovic - Vasic *et al.*, 2011).

The aim of this study is to determine the chemical composition of grapes (must) and polyphenolic composition of Vranac, Kratosija and Cabernet Sauvignon wines produced from grapes sampled from representative locations in

Podgorica vineyard subregion in a vintage 2013, in order to verify the impact of the variety as determined by previous research.

MATERIAL AND METHODS

Samples and winemaking process

The wines Vranac, Kratosija and Cabernet Sauvignon were produced from grapes harvested at the time of their technological maturity (20–23 Brix), from 6 to 21 September 2013. Grape samples of Vranac ($n = 4$), Kratosija ($n = 3$) and Cabernet Sauvignon ($n = 3$) were (20 kg) sampled in a vineyard from 4 locations: Sipcanik (L1), Rogami (L2), Ljeskopolje (L3), Kokoti (L4), situated in Podgorica subregion, in the range from 25 and 50 meters above sea level. The average air temperature in 2013, in a vegetative period (from 1 April until the date of the harvest), amounted to 22.4 °C, while precipitation amount reached 130.6 L/m² (MONSTAT 2014).

Vinification of grapes is conducted in the winery of the Biotechnical Faculty, by applying the classical technique and by spontaneous fermentation. After crushing the grapes, the potassium metabisulfite was added to pomace (5g of SO₂ to 100 kg of grapes). The fermentation lasted for seven days at a temperature between 25 and 28°C. The cap was punched down manually, three times a day. After fermentation, the wine decantation was conducted, and the wine was stored at the room temperature. After the spontaneous malolactic fermentation, the wine was decanted again and necessary amount of SO₂ was added. Chemical and polyphenolic composition of the wine were analysed four months after fermentation.

Chemicals and reagents

Methanol, ethanol, hydrochloric acid, sodium hydroxide, sodium carbonate, Iron (II) sulfate were purchased from Sigma Aldrich (St. Louis, MO, USA) and Merck (Darmstadt, Germany). Ultra pure water was of Milli Q grade (Millipore Corporation, Billerica, MA, USA). The reagents Folin-Ciocalteu and vanillin were from Merck.

Chemical composition of must

The following parameters: total soluble solids (TSS), titratable acidity (TA) and the pH of must were analyzed according to the official methods of analysis of wines (OIV. 2011).

Spectrophotometric analyses

Spectrophotometric measurements were carried out with Varian Cary 100 spectrophotometer (Bio Tech, Maryland, USA), as described by Di Stefano and Guidoni (1989) and Di Stefano *et al.* (1989), under optimized conditions for red wine analysis (Rigo *et al.*, 2000).

Total polyphenols

Total polyphenols (TP) were determined by the reduction of Folin-Ciocalteu reagent to blue pigments caused by polyphenols in alkaline solution. A realistic estimation of total polyphenols can be obtained only after preliminary cleaning of samples from other compounds that interfere with the assay (Di

Stefano and Guidoni, 1989). When the absorbance was between 0.3 and 0.6 AU (the linear response range), the results were expressed against the corresponding blank as (+)-catechin = $186.5 \cdot A \cdot d$ in mg/L of wine; A = absorbance and d = sample dilution.

High-molecular mass proanthocyanidins

HMP were assessed by transformation into cyanidin (Di Stefano *et al.*, 1989). When the absorbance was between 0.3 and 0.6 AU the results were expressed against the corresponding blank as cyanidin chloride = $1162.5 \cdot DA \cdot d$ in mg/L of wine; DA = difference in absorbance between sample and blank, and d = sample dilution.

Low-molecular mass proanthocyanidins – index of vanillin

The optimised and controlled vanillin-HCl method of Broadhurst and Jones (1978), was used in order to analyze the catechins and proanthocyanidins reactive to vanillin, following the conditions described by Di Stefano *et al.* (1989).

The method provides an estimation of the free carbon 6 and carbon 8 of the A-ring of both catechins and proanthocyanidins. This index decreases with the increase in polymerisation, because mainly carbon 6 and carbon 8 are involved in polymerisation bonds. When the absorbance was between 0.2 and 0.4 AU, the LMP were evaluated as (+)-catechin = $290.8 \cdot DA \cdot d$ in mg/L of wine; DA = difference in absorbance between sample and blank and d = sample dilutions.

Total anthocyanins

Determination of total anthocyanins was performed by the method proposed by Di Stefano *et al.* (1989), on the basis of the maximum absorbance in the visible range (536 to 542 nm). When the absorbance was between 0.3 and 0.6 AU, the results were expressed against the corresponding blank as TA = $A \cdot 26.6 \cdot d$ in mg/L of wine; A = absorbance and d = sample dilution.

Statistical analysis

Data was processed by ANOVA (p indicated) and when significant, the means were separated using Tukey's honest significant difference (HSD) test ($p < 0.05$). Statistical analysis was performed using the Stat.Soft.Inc. (2003) Statistica.

RESULTS AND DISCUSSION

The chemical composition of grape must

The chemical composition of the grape must of the Vranac, Kratosija and Cabernet Sauvignon varieties, originated from different locations in Podgorica region in year 2013 are given in Table 1.

The table of the chemical content of the grape must shows that the average sugar content was the highest in the grape must of the Kratosija variety (22.1%), followed by a Cabernet Sauvignon (21.5%). The lowest content of sugar was found in Vranac variety (20.6%). Moreover, the average acidity was highest in the grape must of Kratosija variety (7.3 g/L), followed by a Cabernet Sauvignon (6.4 g/L), while the lowest acidity was assessed in Vranac variety (5.7

g/L). These results correlate with the ones obtained in the research Pajovic et al, 2014a. The variety has statistically high significant effect on the sugar content and a total acidity of must. The grape must of Vranac variety has a higher statistically significant pH value (3.35) compared to other two varieties, while there is no statistically significant differences between pH values of Kratosija (3.29) and Cabernet Sauvignon (3.31).

Table 1. Chemical characteristics (TSS - Total soluble solids, TA - Titratable acidity) of Vranac, Kratosija and Cabernet Sauvignon grape must at the time of harvest, from the 2013 vintage

Locality	TSS (°Brix)	TA (g/L tartaric acid)	pH
VRANAC			
L1	20.5	5.7	3.43
L2	20.2	5.3	3.30
L3	21.8	5.6	3.32
L4	20.1	6.20	3.33
Mean	20.6±0.79^A	5.7 ± 0.55^A	3.35± 0.06^A
KRATOSIJA			
L1	21.7	7.5	3.29
L2	21.9	7.2	3.29
L3	22.8	7.1	3.3
Mean	22.1±0.56^B	7.3±0.27^B	3.29±0.03^B
CABERNET SAUVIGNON			
L1	21.5	6.4	3.33
L2	21	6.6	3.31
L4	21.9	6.3	3.3
Mean	21.5±0.44^C	6.4±0.25^C	3.31±0.02^B

ANOVA was used to compare data (n.s. not significant, a,b,c $p \leq 0.05$, A,B,C $P \leq 0.001$). Different lower-case letters indicate significant differences of means between varieties using Tukey's HSD test($p \leq 0.05$).

It can be concluded that the sugar content and total acids in the grape must were typical for the studied grapevine cultivars. (Raicevic *et al.*, 2012, Pajovic *et al.*, 2013).

Looking at the vineyard locations, as regards to Vranac variety, the largest sugar content in grapes was found in Ljeskopolje (L3), then in Cemovsko field (L1), followed by the location of Rogami (L2), and Kokoti (L4). Grapes from the location 4 had the highest content of total acidity, followed by L4, L3, and the lowest acidity was assessed in the grapes from L2. The grape varieties Kratosija from L3 had the highest sugar content, and the lowest total acid content. It is followed by L2 and L1 location. In the case of Cabernet Sauvignon, the highest sugar content of the grapes, and the lowest total acid content was registered at L 4, then at L2, and finally at L3.

The polyphenol content in wines

Contents of total polyphenols, total anthocyanins, low- and high-molecular mass proanthocyanidins in four months old Vranac, Kratosija and Cabernet Sauvignon wines from different vineyard locations in Podgorica region in 2013 are shown in Table 2.

Table 2. Content of polyphenols (TP - Total polyphenols, TA - Total anthocyanins, LMP – low-molecular mass proanthocyanidins, HMP– high-molecular mass proanthocyanidins) in Vranac, Kratosija and Cabernet Sauvignon wines from the 2013 vintage.

Locality	TP (mg/L (+) catechin)	TA (mg/L)	LMP (mg/L (+) catechin)	HMP (mg/L cyaniding chloride)
Vranac				
L1	1130	696	306	1102
L2	907	605	149	602
L3	1504	952	380	1604
L4	807	644	144	699
mean	1087±279^A	725±141^A	244±106^A	1002±413^A
Kratosija				
L1	884	392	372	918
L2	765	310	263	512
L3	1429	600	473	1549
mean	1026±306^B	434±129^B	369±91^B	993±453^B
Cabernet Sauvignon				
L1	1296	549	486	1305
L2	1335	424	556	1436
L4	1243	580	399	1197
mean	1291±41^C	517±106^C	480±68^C	1313±104^C

ANOVA was used to compare data (n.s. not significant, a,b,c $p \leq 0.05$, A,B,C $P \leq 0.001$). Different lower-case letters indicate significant differences of means between varieties using Tukey's HSD test ($p \leq 0.05$).

Table 2 shows the graphical representation of the polyphenol composition of wines. It can be seen that the average contents of total polyphenols (1291

mg/L), low-molecular mass proanthocyanidins (480 mg/L) and high-molecular mass proanthocyanidins (1313 mg/L) was the highest in wine Cabernet Sauvignon. Pajovic *et al.* (2014a) reported similar results for the vintage years 2011 and 2012, showing the value of total polyphenols (1 301 and 1 393 mg/L in 2011 and 2012, respectively), low-molecular mass proanthocyanidins (483 and 787 mg/L) and high-molecular mass proanthocyanidins (1277 and 1564 mg/L).

The wine Vranac produced during 2013 proved to be highest in anthocyanin content (725 mg/L) which correlates with the results Pajovic *et al.* (2014a) for 2012 (832 mg/L), and for 2011 in the same locations as well as in research conducted during 2013.

The lowest content of low-molecular mass proanthocyanidins was recorded in the wine Vranac (244 mg/L). Kratosija wine showed the lowest content of total polyphenols (1026 mg/L), total anthocyanins (434 mg/L) and high-molecular mass proanthocyanidins (993 mg/L), which was also found by Pajovic *et al.* (2014a), who in two years of research got the average value of total polyphenols (1042 and 961 mg/L in 2011 and 2012, respectively), total anthocyanins (317 and 429 mg/L in 2011 and 2012, respectively) and high-molecular mass proanthocyanidins (1,024 and 846 mg / L in 2011 and 2012, respectively). It can be concluded that the variety Kratosija has a lower polyphenol potential compared to Vranac and Cabernet Sauvignon and that the impact of the variety has a high statistical significance on all the parameters, which is in line with research conducted by Pajovic *et al.* 2014a for the investigated varieties at the same locations.

Climatic conditions in 2013 (high rainfall before flowering followed by a strong hydric stress during development until berries ripening) allow us to assume that they have big impact on the phenol composition of wine which is consistent with previous research (Lorrain *et al.*, 2011)

With regards to all investigated locations, Vranac and Kratosija wines had the highest content of all the examined parameters at location Ljeskopolje (L 3), followed by Sipcanik L1.

Cabernet Sauvignon wine from the location Rogami (L2) had the highest content of total polyphenols (TP), LMP and HMP, and the lowest content of total anthocyanins (TA), while the one from the location Kokoti (L4) proved to be high in the content of total anthocyanins (TA).

In the Podgorica winegrowing subregion, vineyards at locations L1, L2, L3 and L4 lie between 25 and 50 m above sea level (m.a.s.l.). Catechin monomers, procyanidin dimers, trimer C1 and total extractable proanthocyanidins proved to be higher in both the skins and seeds of grapes growing at lower altitudes (Mateus *et al.*, 2001).

The impact of the vineyard location on the contents of the examined parameters can be explained by the fact that the total phenolic content is affected not only by the variety, but also by the ecological factors (air temperature, light, sunlight exposure, soil), well as by the maturity of grapes. A number of researchers support these claims (Revilla *et al.*, 1997, Puskas *et al.*, 2009).

The results obtained in this study confirm the high impact of the variety on the investigated parameters, which was also ascertained by previous studies (Pajovic *et al.*, 2014a, Ivanova *et al.*, 2015), of these varieties in Montenegrin and Macedonian region.

CONCLUSIONS

Study results of the chemical composition of grape must and polyphenol content of wine were analyzed at four vineyard locations in Podgorica subregion.

The highest average sugar content and total acidity in 2013 was found in grape variety Kratosija, then in Cabernet Sauvignon, while the lowest values were assessed in Vranac variety.

The high polyphenol content of all components: total phenolics, anthocyanins, low molecular weight and high molecular proanthocyanidins was recorded in wines from four different locations. The highest average content of total polyphenols (TP), low-molecular mass proanthocyanidins (LMP) and high-molecular mass proanthocyanidins (HMP) was assessed in Cabernet Sauvignon, while the highest content of total anthocyanins (TA) is registered in the wine Vranac. The lowest content of total polyphenols (TP), total anthocyanins (TA) and high-molecular mass proanthocyanidins (HMP) has Kratosija wine, while the lowest content of low-molecular mass proanthocyanidins (LMP) was observed in the wine Vranac. Kratosija variety showed a lower polyphenol potential in comparison to Vranac and Cabernet Sauvignon.

The grape variety proves to have a high statistically significant impact on all investigated parameters.

It is assumed that in addition to the environmental conditions of the grape growing area, the impact of the location on the investigated parameters may come from various winemaking practices.

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