

Wine adulteration and its detection based on the rate and the concentration of free amino acids

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Summary

Following the definition of wine adulteration, the authors briefly review its history from ancient times to present day and describe the methods that are applied for adulteration of wine from the historical point of view. More space is devoted to the Hungarian wine adulteration and to the detailed reports of the early methods, which attempted to discover fake wine. It describes in detail the current situation of wine adulteration and the fight against counterfeiting. The second half of the review article presents some examples of the analytical chemistry techniques with which fake wines can be detected. In doing so, priority is given to the discussion of high performance liquid chromatography and gas chromatography applications. The end of this paper describes the free amino acid content of wines, and the possibilities of using the results for detection of wine adulteration.

Keywords: wine adulteration, free amino acid content, wine analytics, ion exchange column chromatography, IEC, high performance liquid chromatography, HPLC, gaschromatography, GC

Introduction

The history of wine adulteration began when people discovered the technology of wine production. Wine is made only from grapes, but only in cases, when the production of the grapes was wrong, or when the phylloxera destroyed almost 50% of the grape plants, and in the case of greater wars, when the wine consumption demand increased at the military, the time has come for the adulteration of the wine. Wine imitations which were made form varieties of additives, sugars, colouring materials, grapes were not at all seen as fakes, caused significant competition for the honest winemakers, and made possible the quick enrichment of the dishonest (Csapó, 2016).

Definition of wine adulteration

Wine adulteration can be any technology or the use of different auxiliary materials, which are not authorised by the laws and legislations in wine-making. The 2004 annual „Wine law“ dispose on wine adulteration in Hungary, which states that „Such winery product, which have been produced or treated by a method not authorised by the legislation or using non authorised materials should not be qualified as grape wine.“

The law considers adulterated wine, if licensed material is used in wine-making, but in larger amounts than what is authorized, or unauthorized material, such as water, ethylene glycol, glycerol, artificial colours and flavourings are used during wine making. An example of the former, if you use sucrose to upgrade the sugar content of the must, but not in the maximum amount allowed (Csapó et al., 2016).

It can also be considered as counterfeiting if the wine is relabelled, rewrapped, such wine is also fake which is produced from the products unfit for human consumption, where the suitability for human consumption is masked by using, such as colour, taste or flavour materials, which is produced without the permission of a legally authorized person, which provide misleading information on the label and is placed on the market for the purpose of economic advantage or gain (Ebeler et al., 2011).

Historical overview of wine adulteration

Wine adulteration has always been banned throughout history, in fact the laws seriously punished it, and however, counterfeiters were always one step ahead of the law. Before Christ around 1800 the laws of Hammurabi punished strictly wine adulteration. In ancient Greece and Rome, the members of the poorer people class consumed much wine made from lot of additives, or even sometimes a mixture of vinegar and water which were added to the soldiers as wine. It was very common to upgrade the poor, undrinkable wine with resins and spices, but to the high-quality wines were flavoured by myrtle, honey, dried fruit and rose petals as well (Lees, 2000).

In the middle ages and even in modern times the tradition of wine adulteration has continued, and the very large wave started when at the end of the 19th century, with the development of chemistry and food science, the counterfeiters received the most toxic or nontoxic colouring matters, and with the help of these materials from white wine, or even just water, red wine reproduction has been able to be produced. After the cheap production of beet sugar and cane sugar low alcohol content wines were produced from grape marc, and often toxic dyes are also used during wine making. The wave of adulteration reached the world-famous Tokaj wines, which compromised the very good reputation it

established during the centuries. At the beginning of the 20th century, cheap, artificial wine were produced from water, alcohol, glycerol and various plant originated acids, which were relabelled, and the traders released on the market as high quality wines (Wartha, 1880).

The first wine law in the Monarchy was produced in 1893, (The Wine Law, XXIII law item of 1893), which was confirmed with an even stricter wine law in 1908. As an effect of the aggravation the adulteration of wine receded, however, in Hungary and around the world wine is still adulterated. As an effect of an unfolding the complete wine region could lose its reputation, just as it happened in our country in the Great Hungarian Plain Wine Region, which recently produced many times more wine than the amount of grapes would have made it possible.

Contemporary methods for the exposure of wine adulteration

For the exposure of the counterfeit of wines chemical analytical methods have been developed that could be used for unveiling wine counterfeiters. In Hungary, it is due to the outstanding chemist Vincze Wartha that wine adulteration was centred to the focus, because he as a chemist knew colorants which were suitable for wine adulteration, and he gave advice to detect these colorants from counterfeit wine. We know from his work that *„The gift of the God, the bulls' blood from Eger, is no longer other than a zing trilogy painted with magenta, boric acid and glycerol. It is no wonder, that the society thus contested gets exasperated and intend to start a smashing war against the army of counterfeiters, who are from dirty profiteering not only unconscionable enough, to be involved in an assassination attempt against the life of fellow man, but they are shameless enough, that the reputation of the Hungarian products are totally spoiled for the foreigners. Real treason of such an act is!”* (Wartha, 1880). Nothing could be added to this, but dejectedly note, that for more than 130 years, little has been changed, counterfeiting today also exist, and the fight against adulteration today is still going on.

How the red wines have been adulterated at the time of Vincze Wartha? For the painting of the wine the different tar paints were used, from which the fuchsine had a special significance, because this material is used for the production of blue, violet, green and yellow colours. It is applied to paint the wine, because at the vegetable inks (mallow, privet, elderberry) much smaller concentrations can be used to achieve the desired effect. 10–12 mg per litre in white wine can change the colour to red, without the change of the taste and flavour, while achieving similar effect with plant dyes much more material should be used, which can change the flavour of the wine. The next version of its acid derivative, the acid fuchsine was used in dyeing, which solved in the water well, and did not precipitate during storage. The derivatives of β -naphthol and

naphthylamine, the aniline brown, the crisotoluidine, the safranin, the grenadine and other aniline dyes, mainly aniline violets, have been used, and the yellow caramel is used for the buffering of the colour.

Vince Wartha has developed methods to detect these tar paints from red wine. According to the first method the wine was oversaturated by magnesium-oxide, and the resulting mixture was extracted by the mixture of amylalcohol and ether, and in the presence of fuchsine the colour became red. At the other method the red wine was precipitated by lead-acetate, and the red colour of the filtrate suggests its presence in the fuchsine. According to his most sensitive method the wine thickened by boiling was treated by ammonium hydroxide medium and then the resulting substance was extracted by ether and the extracted material was filled into a white porcelain dish. The extract also turns red the colour of the white wool. In this way one mg of fuchsine could be detected. The red colour disappeared after the treatment of the wool by hydrochloric acid or ammonia (Wartha, 1883).

The contemporary scientists also noted that the tar colours mixed to the wine caused general physical degradation, besides which they mostly impact negatively the kidneys, leading to an increase in urinary protein content. It is not accidental that with this knowledge we shall consider Vince Wartha the front-line fighter in the counterfeiting war, who 130 years ago stepped up for the production of the high-quality wine which is free from counterfeiting. Among other things, as a result of the work of scientists such as him, the falsification of the wine decreased to a minimum in a couple of decades in Europe.

The current position of the wine adulteration

Are the wines adulterated nowadays, or what tricks are applied by the wine counterfeiters, why are the wines adulterated at all? The age of such falsification that was used in previous centuries has been finished with the use of modern analytical methods. It is also difficult to counterfeit, because the recycling of by-products of wine-making is prohibited by law, so wine from marc or sweeping wine today is no longer possible to be produced. The natural alcohol content of the wine is generated from glucose, which concentration at early vintage or at high production of grape is less than as desirable, therefore, in this case, the use of sugar supplementation is legally possible, but adding larger quantities than legal is considered to be counterfeiting. In the case of high alcoholic content the wine is weakened by adding water to it, which procedure is prohibited in Hungary, but in the United States the water addition, which procedure is called stretching or diluting, takes place within legal frameworks (Buglass, 2011; Hoorfar et al., 2011).

Applying sugar in the must, or applying sugar and water together is a common practice throughout the world. In addition, additives and assistant materials are used during the production of wine, such as cultured yeasts, and nutrients necessary for its reproduction, pastes, pigments, tannin, tartaric acid, citric acid, gum Arabic, glycerol, and such substances which release sulphur dioxide. The removal of the water, the thickening, is not allowed, due to which sweeter and heavier wines can be produced (Hoorfar, 2012).

The supplementation with glycerine is often used to improve the quality of the wine, which is a natural constituent of the wine, but if it exceeds the maximum length allowed, it is considered as counterfeiting.

Otherwise the supplementation with glycerine is not prohibited by American laws. Changes in the natural environment, the transformation of the climate also requires certain legislative changes, for example due to the increased temperature, the organic acid content is considerably smaller than before, so a few years ago only the tartaric acid was allowed to be added to the wine, in the meantime, the supplementation with malic acid and lactic acid are also allowed. The supplementation with tannins and ascorbic acid is allowed, or even the supplement with sugar can also be provided up till the limit established by the law. Who crosses the limit, becomes a wine counterfeiter (Reynolds, 2010).

Cheap foreign wines were often used to counterfeit Hungarian wines, which could be sold more expensively as a domestic product. This method, however, declined because of the checks. To manipulate with the quantity is also difficult, because it can be estimated how many grape can be harvested from one hectare, which is an average of 12 tons. The must is 75–80% of the grapes, that is, 100 kg of grapes will produce 80 litres of grape juice, from which 75 litres of wine can be produced.

Although the different grape varieties might behave differently in this respect, but there are no big differences, the evaluation can be carried out safely.

In Hungary, the National Foodchain Safety Agency oversees the classification of wines, and fights against counterfeiting. For this purpose the laboratory is able to examine the products in order to determine the plant origin at almost atomic level. They can determine some parameters without opening the bottle, but in addition to classical methods, the very sophisticated instrumental analysis is also used for the classification of the wines. These include for example the nuclear magnetic resonance analysis, mass spectrometry, isotope ratio test analysis, gas- and high performance liquid chromatography, atomic absorption spectrophotometry, and inductively coupled plasma emission, and other instrumental analysis. These techniques are suitable for the easier identification and detection of the most common class of tricks, such as

the excessive enrichment with non grape originated sugar, dilution with water or supplementation with artificial sweeteners.

These reliable methods are necessary, because the modern imitations made by the sophisticated methods, are not recognised by the majority of the consumers. One should always beware from the very cheap wines, because there is a high risk of counterfeiting. Until now, the most common tricks for adulteration are the too high sulphur dioxide concentration, incidentally heavy metals may be included in the course, but toxins may be included as well (if apple juice is used for counterfeiting) or ethylene glycol and methanol. Sometimes artificial sweeteners and dyes are added to wine illegally, in order to improve the characteristics of the wines, or to dissemble the corrupt characteristics or bad quality.

Sometimes the State itself participates in the wine falsification. In the 1960s, 13 times as many wines (Leányka) have been sold to the Soviets than the amount of the grape grown in Hungary. In the 1990s a gold-colour counterfeit wine was produced by the soaking of straw, which was the peak of the wine adulteration in Hungary. As a result of the organizational collaboration and action created in 1997 in Hungary, the wine adulteration fall back, but it is estimated that 20% of the wine in the market is still fake (Csapó et al., 2016).

The judgement of the adulteration is made more difficult, if some countries cannot agree on who is entitled to produce certain types of wines. The Tokaji Aszú is adulterated all over the world, causing significant economical and prestige loss for Hungary. It is adulterated by the Italians (Tocai Friulano), the Alsatians (d Alsace Tokay), and we have our arguments in this respect with Slovakia, because in 1920 the Trianon Treaty split the Tokaj wine region as well, since three municipalities of the Hungarian Tokaj Region appended to Slovakia. Altogether seven Slovakian villages require the name of Tokaj, due to which Hungary is protesting, because the peculiar quality of the Hungarian wine is caused by the natural endowments, human factors and traditions, as well as the advanced technology, which is located only in Hungary. In Beregovo in Ukraine a large-scale forgery of the Tokaj wine is under process, which is sold primarily to the Italian market, and the installation of the Tokaj grape has been attempted in the Crimea as well.

Some examples for the detection of wine by instrumental analytical chemistry techniques

In a well equipped food analytical laboratory identifying the rough wine falsifications should not cause any problems. Although the Hungarian wine law currently permits the use of sucrose to improve the quality of must, but only to the extent permitted. The larger amount of sucrose is

easily detected on the classical methods, reducing, and non-reducing sugars, and total sugar, or by HPLC (Kovács and Csapó, 2015ab).

The titratable acidity of the variety of musts changes with broad limits depending on the region, the weather conditions and the date of the harvest, but only L(+) tartaric and L(-) malic acid present in the grape and must. If the D-tartaric acid and D-malic acid can be detected from the wine, say, precolumn derivatization by HPLC, or with chiral column without derivatization, we can be sure that non-grape origin acids were mixed to the wine (Csapó et al., 2013).

The situation is not so easy in the case of the lactic acid, while yeasts production is almost exclusively D(-) lactic acid produced from sugar, but in the course of the biological malic acid fermentation in the lactic acid bacteria from L(-) malic acid L(+) lactic acid is generated, so then the analysis of the enantiomers in this case is not suitable for identification of the wine adulteration.

The sorbitol is also worth mentioning out of the compounds which can be detected by liquid chromatography, which can be found in the juice of the pear, the apple and other fruits, however, grape and wine contains it only in small amounts (up to 80 mg·dm⁻³). In the presence of more significant amounts of sorbitol, the grape must was blended with other fruits juice and the falsification can be clearly demonstrated (Csapó et al., 2008, Moreno and Peinado, 2012).

Liquid chromatography is suitable for the determination of the all mono- and disaccharides, glycerine and ethylene glycol, mono-, di- and tricarboxylic acids, polyphenols, colorants, flavourings and vitamins and the ion-exchange column chromatography is useful for determination of the free and bound amino acids as well as the assumption of biogenic amines formed from wine (Kovács and Csapó, 2015ab). The gluconic acid can be determined by high performance liquid chromatography, which proves the presence of sweet wine originated from grape after fine putrescence. The musts and wines contain 120 mg·dm⁻³ gluconic acid. The glucuronic acid can be detected from grapes passed through fine putrescent or putrescent. Its' concentration in the wines is between 0.40–1.25 mg dm⁻³ quantities (Kovács and Csapó, 2015ab).

The heat stable volatile components and those which can be converted into volatile compounds after precolumn derivatization can be determined by gas chromatography. Capillary gas chromatography coupled with mass spectrometry (GC-MS) is particularly efficient for the analysis of wines, which so far have been able to determine 350 different volatile compounds from different wines. These include alcohols, ketones, aldehydes, volatile and non-volatile fatty acids, esters, fatty acid derivatives, the components of waxes and resins and different aroma compounds produced during the course of the fermentation and

maturation of the wine. The special aroma compounds of the different wine varieties can be determined, with which one can also identify which wines were blended (Cuvee) or what kind of wine was used for falsification with other wines (Cox, 1999; Sun, 2008).

Atomic absorption spectrophotometry and inductively coupled plasma emission photometry is suitable for the determination of the macro and micro elements, but even the less than one $\mu\text{g}/\text{l}$ concentration can be easily detected and determined.

Using the free amino acid content for the identification of wines

According to our own investigations, the 10–40% of the total nitrogen of wines comes from amino acids; at white wines the amino acid nitrogen is 10–25%, at red wines is 20–40% of the total nitrogen. The free amino acids are the nutrient sources of microorganisms taking part in the fermentation, and starting materials of the volatile aroma compounds of the wines. The free amino acid content of the wine is influenced by the wine production technology, the degree of ripeness of the grapes, the grape status, the healthiness, kind and genotype of the grapes, the wine-growing area, the agricultural conditions, including climate conditions, the genotype of bacteria and the yeasts that were used, the technology of wine production and the age of the wine (Csapó et al., 2011, 2013).

The free amino acid content of the wines changes because the yeasts consume them during their metabolism, but during the metabolisms of the yeasts, and the enzymatic breakdown of the proteins they can be formed in significant quantities. The utilization of the free amino acids is affected by the concentration of the nitrogen containing substances, the alcohol and sugar content, the pH, the temperature and the aeration (Csapó and Csapóné, 2004).

The yeasts cannot utilize the proline and can utilize only slightly the lysine and cysteine. Out of the amino acids proline and arginine are present in highest concentrations in the wines. The free amino acid fraction of the Hungarian white wines has 50–54% proline, 10–17% arginine, while this ratio for red wines is 60–80% for proline, and 2–5% in the case of the arginine. In addition to these two amino acids the wines contain significant quantities even γ -amino butyric acid, alanine, glutamic acid, histidine, lysine, aspartic acid and ornithine. The amount and proportion of the amino acids are affected by the type of grapes, the agricultural area, the vintage, the technology used and the combination of these effects (Csapó, 2004).

Figure 1 shows the standard chromatogram of free amino acids after their separation and determination with ion exchange column chromatography, using lithium buffers. We analysed the free amino acid content of some white wines (Villányi Rozé, Tokaji Szamorodni,

Neszmélyi Irsai Olivér, Villányi Olaszrizling) and some red wines (Villányi Kékfrankos, Villányi Cabernet Sauvignon, Villányi Kékoportó, Villányi Zweigelt, Egri Bikavér).

Figure 1. Chromatogram of standard amino acids

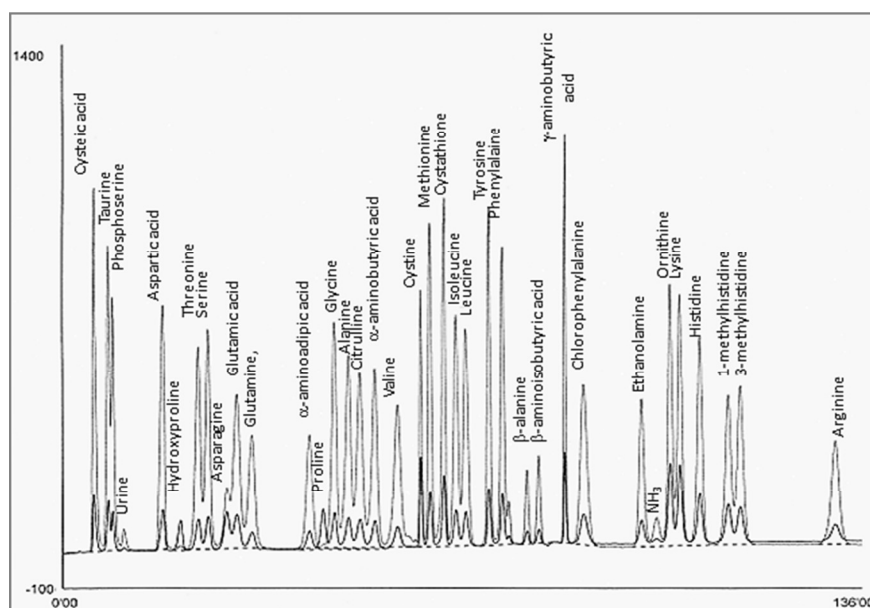


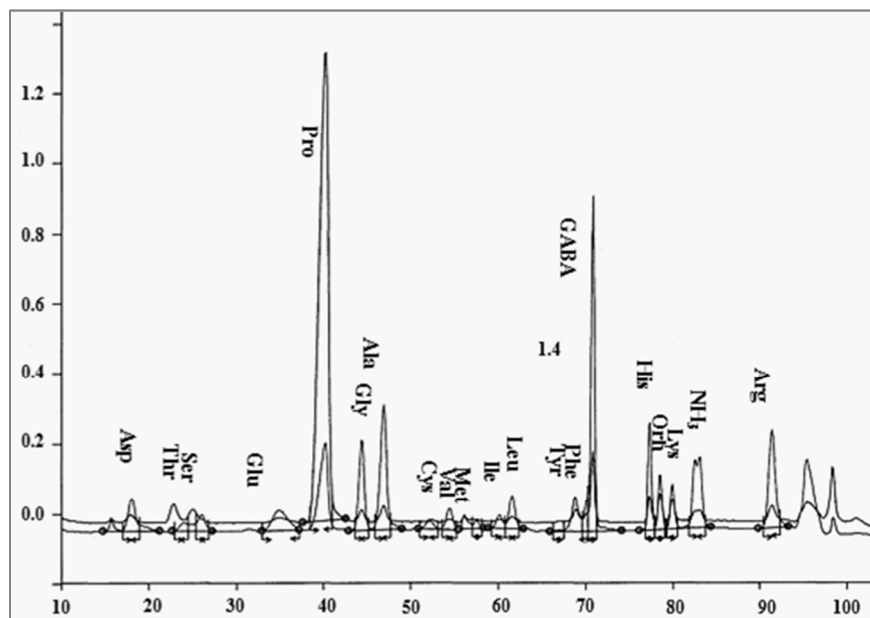
Figure 2 shows the free amino acid content of the Tokaj Szamorodni, Figure 3 the free amino acid content of the Irsai Olivér and Figure 4 shows the free amino acid content of the Olaszrizling. The vertical axis shows the absorbance, the horizontal the retention time of the amino acids (Csapó et al., 2013).

In the case of red wines the proline and arginine content (1105 and 341 $\text{mg}\cdot\text{l}^{-1}$) of the Villányi Kék Oportó was the highest and the Egri Bikavér had the lowest (662 and 11.7 $\text{mg}\cdot\text{l}^{-1}$) concentration from these two amino acids. The concentration of the free glutamic acid varied between 21 and 87 $\text{mg}\cdot\text{l}^{-1}$, the free alanin content varied between 32 and 155 $\text{mg}\cdot\text{l}^{-1}$, and the free γ -amino butyric acid content varied between 12 and 116 $\text{mg}\cdot\text{l}^{-1}$. The total free amino acid content was the highest at the Villányi Kék Oportó (2191 $\text{mg}\cdot\text{l}^{-1}$) and the lowest at the Egri Bikavér (842 $\text{mg}\cdot\text{l}^{-1}$).

In the case of white wines the proline content of the Villányi Rozé was the highest (677 $\text{mg}\cdot\text{l}^{-1}$), in contrast the Neszmélyi Irsai Olivér had the lowest concentration (270 $\text{mg}\cdot\text{l}^{-1}$) from this amino acid. The arginine content of the Neszmélyi Irsai Olivér was the highest (724 $\text{mg}\cdot\text{l}^{-1}$) and the Tokaji Szamorodni had the lowest (67 $\text{mg}\cdot\text{l}^{-1}$) concentration from this

amino acid. The free glutamic acid content varied between 10 and 30 mg·l⁻¹, the free alanine content between 37 and 126 mg·l⁻¹, while that of free γ -butiric acid content between 10.4 and 196 mg·l⁻¹. The total free amino acid content was highest at the Villányi Rozé (1723 g·l⁻¹) and the lowest at the Villányi Olaszrizling (728 mg·l⁻¹).

Figure 2. Free amino acid content of the Tokaji Szamorodni



Note: the abbreviations for the amino acids – Asp=aspartic acid, Thr=treonine, Ser=serine, Glu=glutamic acid, Pro=proline, Gly=glycine, Ala=alanine, Cys=cystine, Val=valine, Met=methionine, Ile=isoleucine, Leu=leucine, Tyr=tyrosine, Phe=phenylalanine, GABA= γ -amino butiric acid, His=histidine, Orn=ornithine, Lys=lysine, NH₃=ammonia, Arg=arginine, Trp=tryptophan (was not present in any sample).

Examining the free amino acid content of approximately 100 different wine samples it can be concluded that the free amino acid content of the red wines is greater than the white wines and that there is a significant difference between the red and white wines both in free amino acid composition and the rate of different amino acids.

A variety of wines in both categories sometimes can have a completely different free amino acid composition, which on the basis of an accepted standard can be used for to identification of wines or for the detection of the counterfeiting. It was concluded, that not only the quantity of free amino acids, but their rates may differ significantly,

which may also be the basis for the identification (Csapó, 2010, 2011; Csapó et al., 2013).

Figure 3. Free amino acid content of the Irsai Olivér

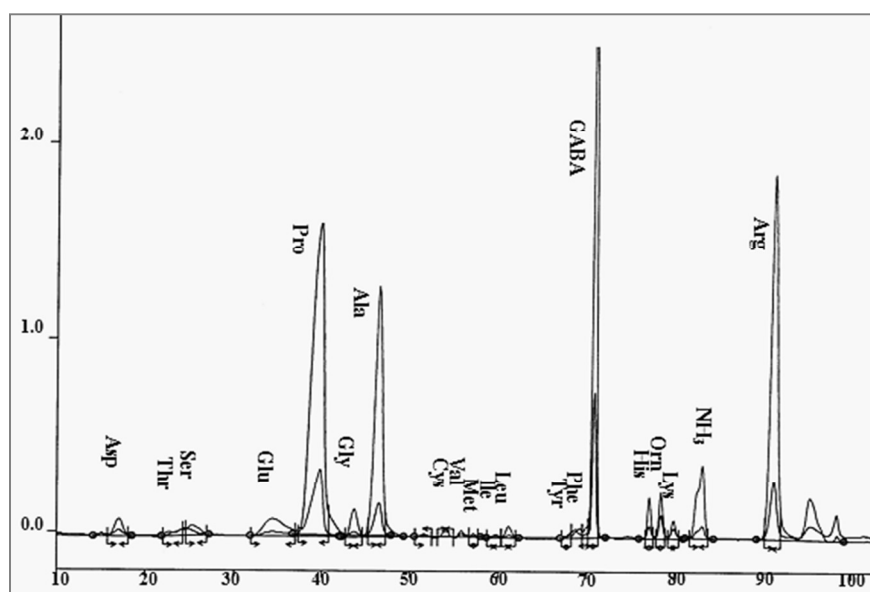
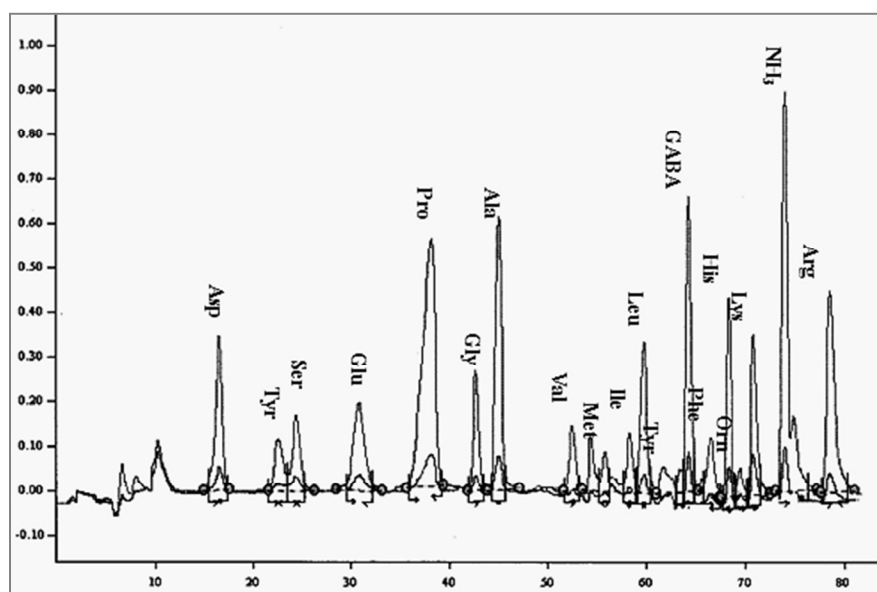


Figure 4. Free amino acid content of the Olaszrizling



Therefore, we can draw the conclusion that with the application of pattern recognition mathematical statistical methods the measurement of free amino acids can be used to identify the different wines and it can also be used to detect the counterfeiting or the dilution of the wine with water.

If we dispose the previously mentioned instrumental analytical techniques, there is almost no such component of the wines that cannot be detected and identified. If we know the amount and proportion of the components, the typical standard for the different wines, there is no such crafted wine forger, whom, with the help of these methods, could not be exposed.

References

2004. évi XVIII. törvény a szőlőtermesztésről és a borgazdálkodásról. (2004) 19.
- Buglass, A. J. (2011): Handbook of alcoholic beverages. Technical, analytical and nutritional aspects. John Wiley & Sons. 1167.
- Cox, J. (1999): From vines to wines. The complete guide to growing grapes and making your own wine. Versa Press. United States. 235.
- Csapó J.–Albert Cs.–Csapóné Kiss Zs. (2008): Élelmiszeranalitika. Válogatott fejezetek. Scientia Kiadó. Kolozsvár. 314.
- Csapó J.–Csapóné Kiss Zs. (2004): Élelmiszerkémia. Mezőgazda Kiadó. Budapest. 492.
- Csapó J. (2004): Biokémia. Scientia Kiadó. Kolozsvár. 468.
- Csapó, J. (2010): Separation and determination of the amino acids from wine by ion exchange column chromatography applying postcolumn derivatization. ERASMUS LLP – The intensive program Ecovitis. Sienna. Italy. July 8–27. 38.
- Csapó J. (2016): Tévhitek az élelmiszer fogyasztásban, avagy miért funkcionális élelmiszer a vörösbor? Magyar-Román Baráti Társaság. 2016. február 18. (68 slide).
- Csapó, J.–Albert, Cs.–Csapó-Kiss, Zs. (2013): Detection and determination of sugars in wine. ECOcycles Erasmus intensive programme on sustainable development through ecological cycles. Palermo. Italy. July 14–August 4. 46.
- Csapó, J.–Csapó, J. jun.–Albert, Cs.–Csapó-Kiss, Zs. (2013): Separation and determination of the amino acids and biogenic amine content of wine by ion exchange column chromatography applying postcolumn derivatization. ECOcycles Erasmus intensive programme on sustainable development through ecological cycles. Palermo. Italy. July 14–August 4. 70.
- Csapó, J. (2011): Amino acids in wine. May this be a possible fingerprint of wines? International Conference of Environmental Technologies, Sustainable Agriculture and Regional Development. Hungary. Gyöngyös. June 06–12. 1–30.
- Csapó J.–Albert Cs.–Csapóné Kiss Zs. (2016): Élelmiszerhamisítás. Debrecen University Press. Debrecen. 217.
- Csapó J.–Albert Cs. (2018): Funkcionális élelmiszerek. Debrecen University Press. Debrecen. 354.

- Ebeler, S. E.–Takeoka, G. R.–Winterhalter, P.* (2011): Progress in authentication of food and wine. ACS Symposium Series: American Chemical Society. Washington DC. 377.
- Hoorfar, J.–Jordan, K.–Butler, F.–Prugger, R.* (2011): Food chain integrity. A holistic approach to food traceability, safety, quality and authenticity. Woodhead Publishing Limited. 367.
- Hoorfar, J.* (2012): Case studies in food safety and authenticity. Lessons from real life situations. Woodhead Publishing Limited. 360.
- Kovács B.–Csapó J.* (2015): Az élelmiszermeghatározás analitikai módszerei. Debreceni Egyetem Mezőgazdaság-, Élelmiszertudományi és Környezetgazdálkodási Kar. Készült a TÁMOP-4.1.1.C-12/1/KONV-2012-0014. projekt keretében. 252.
- Kovács B.–Csapó J.* (2015): Modern methods of food analysis. University of Debrecen Faculty of Agricultural and Food Science and Environmental Management. Készült a TÁMOP-4.1.1.C-12/1/KONV-2012-0014. projekt keretében. 205.
- Lees, M.* (2000): Food authenticity and traceability. Woodhead Publishing Limited. 585.
- Moreno, J.–Peinado, R.* (2012): Enological chemistry. Elsevier. 429.
- Reynolds, A. G.* (2010): Managing wine quality. Volume 1. Viticulture and wine quality. Woodhead Publishing Limited. 606.
- Sun, D. V.* (2008): Modern techniques for food authentication. Elsevier. 714.
- Wartha V.* (1880): A vörös borok hamisításáról. Magyar Kémikusok Lapja. 2011. 66. 364.