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Occurrence of Heavy Metals in Wines for 13 European Countries: A Short Review

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Abstract

The origins of metals found in wine could be both natural or relate to human activities; their concentration impacts the wine's ability to be consumed or conserved, considering that metallic ions are crucial in oxidation-reduction reactions that culminate in wine browning, lack of clarity and potential astringency. Metals in wine may affect human health, and although drinking wine may provide partial quantities of the suggested daily consumption of essential metals, it may also become toxic if the metal concentrations exceed the allowed limits. Strict monitoring is thus recommended throughout the wine-making process. This work provides a comprehensive review of current knowledge on the sources of toxic heavy metals that can be detected in wines. The main focus is set on the limits concerning heavy metals that was observed in wines. Our findings indicated that some countries have exceeded the maximum acceptable copper and lead limits set by OIV. In this light, pure materials such as activated carbon and banana or potato peels could assist in processing the wine, to remove the excess of heavy metals, thus rendering them safe to consume.

Keywords: Heavy metals, Adsorption, wine, Copper, Zinc, Iron, Activated carbon

1. Introduction

Wine is an extremely popular drink around the world, so it has a huge economic impact on the global economy. Researchers have proven that there are some beneficial effects of light to moderate wine consumption [1]. However, the wine comes from vineyards that have been exposed to global pollution and burdened by the widespread use of fertilizers and chemicals to control vine diseases. Many published studies deal with the issue of heavy metals and wine. The present work collects and summarizes published papers and draws aggregated results for a relatively large number of countries; the latter has not been done in literature until low.

The problem with heavy metals lies in the fact that they accumulate more and more in the environment, with adverse consequences for living organisms as they are exposed to them [2], interfering with the functioning of vital cellular components. Some minerals in small amounts are important to human health, but become toxic in higher doses. Typical examples of this group are copper and zinc, while substances such as arsenic and lead, without nutritional value, can become toxic. To prevent intoxication or other negative effects of the organoleptic characteristics of wine, the mineral content is regulated by law. The presence of metals in wines can be attributed to the (i) the soil where the vine grows and (ii) human activity.

The International Organization of Vine and Wine (OIV) has identified the highest limits for the content of trace elements in wine. In this review we provide an illustration of the geographical distribution of the most common heavy metals detected and recorded in the literature, in 13 European countries: Italy, Spain, Ukraine, Poland, Romania, Portugal, Turkey, Serbia, Bulgaria, Croatia, Bosnia-Herzegovina, Kosovo and North Macedonia.

The map (Fig. 1), marking in pink the countries studied. Naturally, the samples of the studies in most cases refer to specific regions of these countries and the findings are a product of generalization throughout the rest of each country.

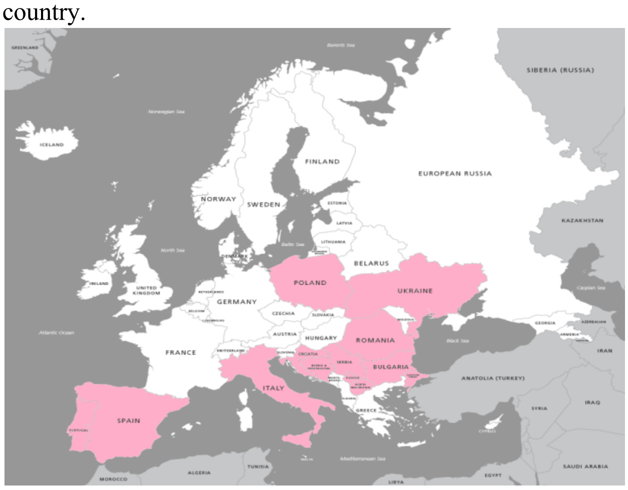

Fig. 1. Map of Europe and countries that are the focus of this review.

2. Heavy Metals Detected in European Wines

For the case of Italian wines, a survey regarding twenty red, white and rose wines, that was conducted in year 2016, was used as a source of information. More than a dozen samples stemmed from the same geographic region in Crotone region [3]. Most of the remaining samples were collected from the "Terre di Cosenza".

In this research, quantitative detection was performed on six heavy metals and specifically on copper, lead, arsenic, boron, zinc and cadmium (Table 1). The results are very interesting and varied. In the case of copper, a lower value

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of approximately 0.014 (mg/L) and a higher value of 0.456 (mg/L) were encountered, averaging at about 0.082 (mg/L). The lowest value for zinc is approximately 0.289 (mg/L) while the highest value reaches almost 0.967 (mg/L), averaging at 0.576 (mg/L). For Lead, the lowest value is 0.003 (mg/L) and the highest value of 0.059 (mg/L), averaging at 0.017 (mg/L). For Boron the lowest value reaches almost 2 (mg/L) and a max of 9.43 (mg/L) averaging typically about 6.4 (mg/L). Similarly, the lowest and highest values for Arsenic were 0.002 (mg/L) and 0.035 (mg/L) respectively, averaging at 0.008 (mg/L). Lastly, the lowest value for cadmium reaches 0.00017 (mg/L), while the maximum ranged around approximately 0.0037 (mg/L), averaging at approximately 0.0085 (mg/L).

Table 1. Determination of iron, copper and lead concentration in Italian wines.

Italian wines	Copper (mg/L)	Zinc (mg/L)	Lead (mg/L)
Lower value	0.014	0.289	0.003
Higher value	0.456	0.967	0.059
Median value	0.082	0.576	0.017

Spanish wines data were obtained through a study regarding both young and aged wines from Córdoba region, Spain, that were assessed for the presence of Iron, copper, zinc, as well as other heavy metals, via atomic absorption spectrophotometry [4]. The findings (Table 2) suggest that in the case of copper, the lowest value reaches 0.10 (mg/L), with the highest value reaching about 1.58 (mg/L), median of the values is 0.547 (mg/L). For zinc the lowest is 0.070 (mg/L), while the highest is approximately 4.400 (mg/L), median of the values is 0.790 (mg/L). In the case of Iron, the lowest value reaches about 1.070 (mg/L) with a max of slightly over 9.830 (mg/L) and median of the values is 3.490 (mg/L).

Table 2. Determination of iron, copper and zinc concentration in Italian wines.

Spanish wines	Copper (mg/L)	Zinc (mg/L)	Iron (mg/L)
Lower value	0.100	0.070	1.070
Higher value	1.580	4.440	9.830
Median value	0.547	0.790	3.490

For Ukrainian wines the data were obtained from a study aiming to evaluate the content levels of trace metals in vines, wine and the environment in a particular region and juxtapose the measurements with the data from other wine-growing locations. The findings indicated (Table 3) that the concentration level of Zinc in Chardonnay wine was 0.340 mg/L, reaching 0.430 mg/L in Muscat white. The concentration of copper reached 0.330 mg/L and 0.400 mg/L in Chardonnay and Muscat white.

Table 3. Concentration levels of copper and zinc concentration in Ukrainian wines.

Ukrainian wines	Zinc Ch/nay (mg/L)	Zinc Muscat (mg/L)	Copper Chardonnay (mg/L)	Copper Muscat
Median value	0.340	0.430	0.330	0.40(mg/L)

A very large survey regarding wines from around the world [5] attempted to assess the impact of the selected parameters (wine type, producer, location) on the concentration levels of 28 metals in 180 samples of wine. The sample consisted of 79 red, 75 white and 26 rose wines. The ICP-MS, ICP-OES and cold vapour atomic absorption spectrometry (CVAAS) methods were used to estimate the concentration levels of metals such as copper, zinc, silver, cadmium, magnesium, titanium, lead, mercury, iron etc. The results (Table 4) indicated that copper was found in concentrations between 0.0049 (mg/L) and 1.071 (mg/L), averaging at about 0.1062 (mg/L). The zinc levels ranged up to 4.046 (mg/L), median of the values is 0.2504 (mg/L). Lastly, the iron content level varied between 0.1549 (mg/L) and 10.25 (mg/L), median of the values is 1.5690 (mg/L).

Table 4. Determination of copper, zinc and iron concentration in wines around the world.

Wines	Copper (mg/L)	Zinc (mg/L)	Iron (mg/L)
Lower value	0.0049	nd	0.1549
Higher value	1.0710	4.0460	10.250
Median value	0.1062	0.2504	1.569

The data for the Romanian wines stemmed from a study [6] on the concentrations of Copper, Lead, Zinc, Cadmium and Nickel in grape juice and wine from three vineyards in Turulung, Romania. The findings (Table 5) suggested that the concentration levels of Zinc in white wine varied between 0.45 mg/L and 0.60 mg/L, while for copper started at about 0.23 mg/L, reaching up to 0.32 mg/L in the same locations.

Table 5. Determination of Cu, Zn in Romanian wines.

Romanian wines	Copper (mg/L)	Zinc (mg/L)
Lower value	0.23	0.45
Higher value	0.32	0.60
Median value	0.28	0.52

The data for Turkish wines (Table 6) were obtained from an analysis [7] of the wine mineral profiles and soil samples procured from the Southeast Anatolia of Turkey. Specifically, it referred to data regarding a well-known, high-quality Turkish wine (Boğazkere) and a set of popular international wines, such as Merlot, Cabernet Sauvignon, Tannat, Cot and Syrah.

The ICP-OES technique was employed to estimate the concentrations of twelve elements including cadmium, copper, iron, magnesium, lead, nickel and zinc accurately. The levels of iron in this site ranged between 9.14 (mg/L) and 29.64 (mg/L), median of the values is 16.28 (mg/L). On the other hand, the concentration of copper varied between 0.43 (mg/L) and 0.99 (mg/L), median of the values is 0.81 (mg/L). Lastly, the levels of zinc ranged from 1.58 (mg/L) and 4.89 (mg/L), median of the values is 3.14 (mg/L).

Table 6. Determination of copper, zinc and iron concentration in Turkish wines.

Turkish wines	Copper (mg/L)	Zinc (mg/L)	Iron (mg/L)
Lower value	0.43	1.58	9.14

Higher value	0.99	4.89	29.64
Median value	0.81	3.14	16.28

A study on Serbian wines (Table 7) was used to procure data [8] about the concentration of Copper, Iron and Zinc in wine samples from Serbia. Spectrometry was employed for the measurements, finding that the concentration of copper ranged between 0.07 (mg/L) and 0.44 (mg/L), median of the values is 0.21 (mg/L). The concentration of Iron ranged between 2.93 (mg/L) and 36.20 (mg/L), median of the values is 7.05 (mg/L). Lastly the Zinc concentration varied between 0.21 (mg/L) and 0.67 (mg/L), median of the values is 0.52 (mg/L).

Table 7. Determination of Copper, Zinc and Iron concentrations in Serbian wines.

Serbian wines	**Copper (mg/L)**	**Zinc (mg/L)**	**Iron (mg/L)**
Lower value	0.07	0.21	2.93
Higher value	0.44	0.67	36.20
Median value	0.21	0.52	7.05

The data for the Bulgarian wines (Table 8) were obtained from a study [9] on the concentrations of Copper, Cadmium, Lead and Zinc in wines that were created in Bulgaria and Italy, as well as in alcoholic fruity drink from Poland. All the elements were detected in the studied samples. The Zinc levels varied between 0.29 (mg/L) and 0.44 (mg/L), median of the values is 0.36 (mg/L). The Copper levels fluctuated between 0.07 (mg/L) and 0.09 (mg/L), median of the values is 0.08 (mg/L). Lastly, the Lead concentration ranged between 0.0 (mg/L) and 1.22 (mg/L), median of the values is 0.36 (mg/L).

Table 8. Determination of Copper, Zin and Lead concentrations in Bulgarian wines.

Bulgarian wines	**Copper (mg/L)**	**Zinc (mg/L)**	**Lead (mg/L)**
Lower value	0.07	0.29	n.d
Higher value	0.09	0.44	1.22
Median value	0.08	0.36	0.36

The data for the Croatian wines (Table 9) were obtained from an analysis [10] on the concentration of heavy metals (copper, iron, zinc, lead) in red wines. The wine used for the study was Plavac mali, a local Croatian product, from various wine-makers in the region. The copper levels in this area ranged between 0.21 (mg/L) and 1.23 (mg/L) median of the values is 0.57 (mg/L). The concentration of Iron varied between 0.81 (mg/L) and 6.20 (mg/L), median of the values is 3.40 (mg/L). Lastly, the zinc levels had a minimum of 0.31 (mg/L) and a maximum of 2.43 (mg/L), median of the values is 0.76 (mg/L).

Table 9. Determination of Copper, Zinc and Iron concentrations in Croatian wines.

Croatian wines	**Copper (mg/L)**	**Zinc (mg/L)**	**Iron (mg/L)**
Lower value	0.21	0.31	0.81
Higher value	1.23	2.43	6.20
Median value	0.57	0.76	3.40

Herzegovinian wines (Table 10) were also studied [11] in regards to the concentration levels of various hazardous elements, such as Lead, Copper, Zinc, and Iron, from two dozen Herzegovinian white and red wines procured from four different vineyards.

The concentration of lead ranged between 0.0 (mg/L) and 0.19 (mg/L), median of the values is at 0.03 (mg/L). Copper fluctuated between 0.0 (mg/L) and 2.87 (mg/L), median of the values is 0.40 (mg/L). The zinc levels extend between 0.36 (mg/L) and 2.10 (mg/L), median of the values is 0.85 (mg/L).

Table 10. Determination of copper, zinc and Lead concentrations in Bonsia-Herzegovinian wines.

Bosnia-Herzegovinian wines	**Copper (mg/L)**	**Zinc (mg/L)**	**Lead (mg/L)**
Lower value	n.d	0.36	n.d
Higher value	2.87	2.10	0.19
Median value	0.40	0.85	0.03

Wines from the region of Kosovo (Table 11) were assessed [12] in order to explain the appearance of heavy metals such as Iron and Copper, since they are detrimental to the product's quality as well as people's health.

A series of 41 samples were obtained and tested during all phases of the wine-making process. Iron was found in concentrations ranging between 0.46 (mg/L) and 2.48 (mg/L), median of the values is 1.23 (mg/L), while Copper was detected in ranges between 0.030 (mg/L) and 5.67 (mg/L), median of the values is 0.63 (mg/L).

Table 11. Determination of copper and iron levels in Kosovo wines.

Kosovo wines	**Copper (mg/L)**	**Iron (mg/L)**
Lower value	0.03	0.46
Higher value	5.76	2.48
Median value	0.63	1.23

Wine samples were also obtained from North Macedonia [12] (Table 12) in an effort to verify quantitatively the occurrence and concentration levels of hazardous heavy metals such as Iron and copper, using 41 samples from various phases of wine-making. The iron concentration varied between 1.346 (mg/L) and 1.576 (mg/L), median of the values is 1.476 (mg/L), while copper levels ranged between 0.127 (mg/L) and 0.158 (mg/L), averaging at 0.150 (mg/L).

Table 12. Determination of copper and iron levels in North Macedonia wines.

North Macedonia wines	**Copper (mg/L)**	**Iron (mg/L)**
Lower value	0.127	1.346
Higher value	0.158	1.576
Median value	0.150	1.476

The data of the afore-mentioned research works were gathered and processed, in an attempt to create a summary report for the overall concentration of heavy metals in wines from all studied regions.

Table 13 provides an overview on the amounts of copper, zinc, lead and iron appearing in wines from mainly the Southern European region, where wine production is popular, due to the favourable climate.

Table 13. Determination of Cu, Zn, Pb and Fe in Herzegovinian wines.

All wines	Copper (mg/L)	Zinc (mg/L)	Lead (mg/L)	Iron (mg/L)
Lower value	n.d	n.d	n.d	0.15
Higher value	5.76	4.89	0.19	36.2
Median	0.35	0.81	0.03	4.93

Fig. 2 depicts the information from (Table 13) regarding the average concentration per metal (mg/L) for the studied regions, showing that iron has the highest average of the studied metals, followed by zinc and copper. Lead concentration is more limited at 0.03 (mg/L).

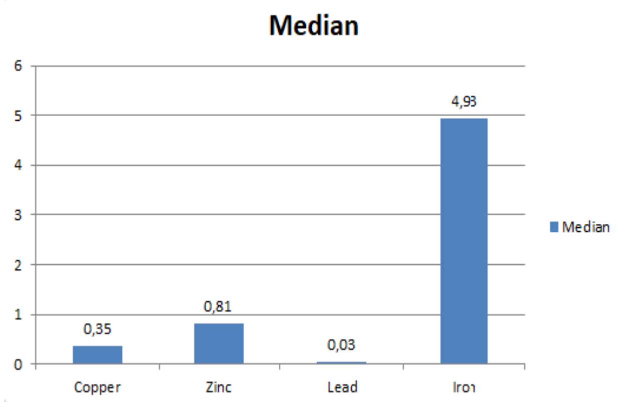

Fig. 2. Median values of copper, zinc, lead and iron concentrations.

In the last table we gathered the median values so that the reader of the research can find the data we used and draw his own conclusions.

Table 14. Median values of all tables.

Median values	Copper (mg/L)	Zinc (mg/L)	Lead (mg/L)	Iron (mg/L)	Lead (mg/L)
Italian wines	0.082	0.576	0.017	n.d	n.d
Spanish wines	0.547	0.790	n.d	3.490	n.d
Ukranian wines	0.330	0.340	n.d	n.d	n.d
Around the world	0.106	0.250	n.d	1.569	n.d
Romanian wines	0.280	0.520	n.d	n.d	n.d
Turkish wines	0810	3.140	n.d	16.280	n.d
Serbian wines	0.210	0.520	n.d	7.050	n.d
Bulgarian wines	0.080	0.360	0.360	n.d	n.d
Croatian wines	0.570	0.760	n.d	3.400	n,d
Bosnia-Herzegovinian wines	0.400	0.850	0.030	n.d	n.d
Kosovo wines	0.630	n.d	n.d	1.230	n.d
North Macedonia	0.150	n.d	n.d	1.476	n.d

3. OIV Maximum Allowed Limits of Heavy Metals in Wine

In 2011 the OIV published instructions regarding the maximum allowed limits of a variety of substances that can be found in wine. According to the regulations, the maximum concentration of Arsenic is 0.2 mg/L, while the corresponding values for Cadmium and Copper are 0.01 mg/L and 1 mg/L respectively. The limit for silver is <0.1 mg/L, while for Lead it reaches 0.15 mg/L, for wines produced after. Lastly, the maximum allowed Zinc concentration is 5 mg/L.

4. Discussion

An important factor that affects the differentiation of the results is the territories in which the studied vineyards are located. It is evident that in these 13 countries, there is a great deal of unevenness in the mechanical composition of the soil. Some of them have sandy soils, others clay, and general average composition. The factors affecting soil structure and consequently their heavy metal content, are majorly (i) wetting and drying, (ii) folding and melting, (iii) organic matter, (iv) the kind of cations that are saturated with the soils, (v) the microorganisms of the soil, (vi) the mechanical recommendation, (vii) the increase and decomposition of roots. The concentrations detected in the studied wines vary considerably. More specifically, the territories of the vineyards from these 13 countries may not have uniformity of mechanical composition. There will certainly be big differences in them and of course it is a factor affecting the fluctuation of the measurements. More recently, materials such as activated carbon [13] emerged, which can remove the excess of many heavy metals and improve wines. Our findings suggested that some countries have higher levels of copper and lead than the maximum acceptable limits of OIV. New studies on pure materials such as activated carbon and banana or potato peels suggest that they could offer a solution in terms of processing the wine to remove the excess of heavy metal in them, ensuring they are safe to consume.

Yet, another important factor to consider that affects the measurements is the climate, and other related regional environmental aspects. For instance, in vineyards located in high elevations, there is lower human activity, thus lower levels of copper and zinc, since those are linked to activities such as the widespread utilization of fertilizers. Coupled with the fact that copper appears in the Earth's bark at concentrations between 25-75 mg/kg, we can easily conclude that in countries with a lot of humidity and soils that do not support vine growth, there will be much higher concentrations of copper and zinc. This is a conclusion that supports prior research that claims that soil from arid environments typically have low copper concentrations [14].

The results can be significantly affected by the impact of human intervention, leading to air, water and soil pollution with heavy metals. According to a study conducted in China, grapes were collected from areas infected with heavy metals, namely an area of disassembly of electronic waste and collected from areas without pesticides to compare [15]. The grape peels were then separated from the pulp to evaluate the effect of accumulation behavior, environmental transport. Copper levels in the grape peel (approximately 5 μg/g) were higher than those found in pulp (approximately 3.75 μg/g). According to a different study, there are important differences in the elementary composition between types of wine. All the studied materials exhibited higher levels in red grapes. [16] This can be attributed due to the fact that their production requires the stay of the grapes in the must, as opposed to white vinification, where in most cases we have the immediate separation of the must from the peels.

A Chinese study concluded that the evaluation showed no significant variance in grape copper concentration between grapes collected from electronic waste dissolution and areas with theoretical or even slight pollution. The concentration levels and potential sources of heavy metals like Lead, Arsenic, Cadmium and Manganese were also analysed. Increased levels of these hazardous elements were detected in the grape peel by the pulp. The aforementioned study rejects the argument related to environmental pollution, but there are other studies that support it. Specifically, a study conducted between 1991 and 1993 using samples of Mavrud, produced in a location with a major industrial pollutant and another with no pollutants, to be used as a control. The heavy metals levels found in wine, grapes and soil were examined for a vineyard situated approximately 3.5 km away from a non-ferrous metal factory (NFMW), where it was noted that the detected levels are significantly higher than the control location close to Pleven region. The amounts found in the surface soil are tenfold for Lead, quadruple for zinc and double for cadmium, while copper levels were practically identical. This research supports the argument regarding the environmental pollution and its impact on the results. It was also found that there are more heavy metals in red wines, and they were detected mainly in the stalks, with lower quantities in the skins, seeds and pulp.

5. Conclusion

One of the most important issues related to heavy metals, is their build-up in the environment and the impact this might have on human health and important cellular functions. The toxicity of heavy metals such as arsenic, lead and mercury were common knowledge for a very long time, with various analytical studies on the subject dating more than 150 years ago. Poisoning caused by heavy metal consumption by humans, is typically addressed via chelating agents. A scientific intervention is thus necessary to manage and regulate the use of heavy metals, aiming to protect the environment and human health. Wine, of course, in not responsible for the overall accumulation of heavy metals in living organisms, but since it is part of the daily life of people, the issue should be addressed accordingly.

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