



Article Total Dealcoholisation of Wines by Very Low Temperature Vacuum Distillation Technology Called GoLo

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Abstract: The use of wine dealcoholisation has multiplied in recent years as a result of various factors, including the increase in alcohol content due to global warming and changes in consumer drinking habits. There are several post-fermentation dealcoholisation methods in the literature which allow for the partial or total elimination of the alcohol content of wine. However, there are no studies on the patented very low temperature vacuum wine distillation technology called GoLo. Therefore, for the first time, this paper evaluates the quality of dealcoholised white, rose and red wines using GoLo technology. For this purpose, alcohol content, pH, total SO₂, free SO₂, total acidity and volatile acidity were measured. There were no significant differences in the variations in pH, total acidity and volatile acidity after the dealcoholisation process using GoLo technology and dealcoholised wines showed a reduction of 22.1% in total SO₂ and a complete absence of ethanol and free SO₂. A model for predicting the total SO₂ content of dealcoholised wines and a model for predicting the amount of sulphites to be added after dealcoholisation were found after the statistical treatment of the data. GoLo dealcoholisation delivers 100% removal of alcohol and free SO₂ in less time, with less loss and energy than other dealcoholisation technologies. The verification and extension of these results will be the focus of future studies.

Keywords: dealcoholisation; GoLo; quality; sulphites; wine

1. Introduction

It is well known that wine, a complex and unique beverage consisting mainly of water and ethanol, is the fermented juice of the grape (*Vitis vinifera*), and that polyphenols are the main compounds associated with the benefits of moderate wine consumption due to their antioxidant and free radical scavenging properties [1,2]. Despite the many health benefits, the excessive consumption of wine can have adverse health effects due to its alcohol content. For example, a negative association has been described between the light-to-moderate consumption of wine and the incidence of certain diseases such as diabetes, ischaemic heart disease or the induction of cancer or allergies, and it is also an addictive substance that can lead to dependence and addiction [3,4]. However, this information should be treated with caution, as there is still a lack of solid "pharmacological" human evidence on the biological effects of wine polyphenols [5], and a reduction in the risk of heart disease (a lower risk of myocardial infarction) seems to be the only disease still recognised in critical reviews of epidemiological data related to wine consumption [6].

Depending on its alcohol content, wines can be classified as alcohol-free [<0.5% (v/v)], low-alcohol [0.5% to 1.2% (v/v)], reduced-alcohol [1.2% to (5.5%)], lower-alcohol [5.5% to 10.5% (v/v)], and alcoholic wines [>10.5% (v/v)]. These concentrations may vary from country to country and are regulated by the International Wine Organisation (OIV) and the regulatory council of each designation of origin (DO) [7-9].



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Wine dealcoholisation processes have been used since the early 1900s [8] and have multiplied in recent years as a result of various factors such as the increase in alcohol content due to global warming and changes in drinking habits, with an increase in consumer demand for beverages without calories, alcohol, or additives [9,10]. Climate change is causing the fruit to ripen more quickly and to a greater extent, thus increasing its sugar content, which in turn leads to a higher alcohol content after fermentation [11,12] and may also lead to a loss of wine typification, as wine typification is based on its alcohol content, as mentioned above. In addition, this rapid increase in grape ripening can lead to early harvests, which can result in the incomplete development of secondary fermentation metabolism and, thus, affect the quality of the wines. The other factors are changes in drinking habits due to the harmful effects of alcohol, excess calories, or additives on health [13]. For example, the non-alcoholic wine market in Germany has grown dynamically in recent years and is expected to grow by 9% per year between 2021 and 2025 [14].

For these reasons, there are several methods of dealcoholisation that allow for the partial or total elimination of the alcohol content of wine, which also eliminates other components of the wine such as volatile compounds, which, in many cases, can be re-introduced into the dealcoholised wine. These methods are used at different stages of the winemaking process, including pre-fermentation, during fermentation and post-fermentation. The pre-fermentation stage uses techniques to reduce fermentable sugars such as viticultural practices (leaf area reduction, pre-harvest irrigation, the application of growth regulators or reduction in photosynthetic activity), early grape harvest, the dilution/blending of grape must, the filtration of grape juice or the addition of enzymes such as glucose oxidase [9,15,16]. The fermentation stage uses techniques to reduce alcohol production, including arrested or limited fermentation, biomass reduction, the use of non-Saccharomyces *cerevisiae* yeasts or the use of modified yeast strains [9,15,16]. The post-fermentation stage uses filtration techniques to remove alcoholic content in wines includes nanofiltration (NF), reverse osmosis (RO), osmotic distillation (OD), also called evaporative perstraction or pervaporation (PV), and non-filtration techniques such as vacuum distillation (VD), spinning cone column (SCC), multi-stage membrane-based systems [9], supercritical fluid extraction with CO₂, extraction with solid CO₂, microbial fuel cell [16], freeze-drying [17] or the patented very low temperature vacuum distillation technology called GoLo. This technology is very similar to SCC and integrates classic multi-batch separation operations into a single, continuous, easy-to-use process, while providing a thin film formation system that operates highly efficiently with no moving parts. In a single pass, GoLo can remove almost 100% of the volatile aromatic compounds, which can be reincorporated into the dealcoholised wine, and dealcoholise the wine to 0.05% (v/v) [18]. Some of these techniques have drawbacks, such as using high temperatures that can significantly degrade the wine, being expensive or using a lot of energy [19].

On the other hand, during the process of the dealcoholisation of wine, the volatile fraction is also removed along with other compounds that can cause allergies. Allergenic substances in wine include sulphites, as the addition of sulphites during the winemaking process is a common practice. Inexpensive and easy to use, sulphiting agents allow for the avoidance of competition between common microorganisms and fermentation yeasts and provide post-fermentation and colour stability [20–22]. However, the use of sulphites cannot be considered a harmless additive for humans, as the ingestion of foods containing elevated concentrations of sulphites may cause food intolerance symptoms and allergic reactions in sensitive individuals [23,24], as aforementioned. The use of sulphites is regulated by the EU 1169/2011 [25], where SO₂ and sulphites at concentrations of more than 10 mg/L, in terms of the total SO₂, must be declared on the label. In addition, the maximum daily intake of sulphites for humans determined by the European Food Safety Agency is 0.7 mg/kg [26]. Currently, the regulatory trend leads to products with lower concentrations of total SO₂, in addition to the commercial rejection of allergen labelling.

There are many analytical determinations that can be carried out on wine to characterise its composition and evaluate its evolution, from sugar content to the presence of heavy metals. However, in terms of wine quality, the most important determinations for practical and legal purposes include alcohol, total acidity, volatile acidity and free and total SO₂ [27,28]. In this sense, there are several studies in the literature on the evolution of the physico-chemical, sensory and quality parameters [13,29–33] of dealcoholised wines using different technologies or on the waste produced by high temperatures for the production of low-alcohol wines [34]. However, there are no studies on the evolution of physicochemical parameters of dealcoholised wines using GoLo dealcoholisation technology. Therefore, the aim of this study was to assess, for the first time, the evolution of alcohol, free and total SO₂, pH, total acidity and volatile acidity using GoLo technology methods in order to achieve the total dealcoholisation of red, rose and white wines on an industrial scale.

2. Materials and Methods

2.1. Sample Selection

A total of 274 samples of different types of wine [183 white (B), 64 red (T) and 25 rose (R)] were obtained from the Spanish commercial market including different DOs. The varieties of white wines were Airen, Sauvignon, Chardonnay, Verdejo, Macabeo, Moscatel, Syrah and *coupage*; the varieties of rose wines were Garnacha, Tempranillo, and *coupage*; and the varieties of red wines were Merlot, Cabernet, Syrah, Tempranillo, Garnacha and *coupage*. Non-dealcoholised wine samples were analysed, dealcoholised using GoLo technology and analysed again over a 12 month period between January 2023 and January 2024. Samples were supplied by a company in central Spain and analysed in the laboratories of the University of Murcia in collaboration with a private laboratory with ISO 17025 accreditation [35] that specialised in wine analysis.

2.2. Chemicals

A buffer solution with pH 7.0, sodium hydroxide 0.1 mol/L and bromothymol blue indicator solution were purchased from Sigma-Aldrich (Stenheim, Germany) to analyse total acidity. Tartaric acid, hydrochloric acid (37%), potassium iodide, starch and sodium tetraborate decahydrate were purchased from J.T. Baker (Deventer, The Netherlands) for the analysis of volatile acidity. Sodium hydroxide 0.1 mol/L, iodine 0.005 mol/L, acetic acid 0.1 mol/L and lactic acid 0.1 mol/L solutions were purchased from Sigma-Aldrich (Stenheim, Germany) for the analysis of free and total SO₂.

Hydrochloric acid (37%), hydrogen peroxide (\geq 30%), methyl red solution and carbonatefree sodium hydroxide [50% (w/w)] were purchased from Sigma-Aldrich (Stenheim, Germany). Ethanol [min 99.9% (v/v)] was purchased from J.T. Baker (Deventer, Netherlands). Water quality type I (>18 MΩ) from Millipore was used.

2.3. Total Dealcoholisation of Wines Using GoLo Technology

GoLo is the most advanced dealcoholisation technology on the market today. This technology integrates classic multiple batch separation operations into a single, continuous, easy-to-use process, while offering a thin film creation system that operates highly efficiently with no moving parts. In a single pass, GoLo can achieve the separation of almost 100% of the volatile aromatic compounds (the essence), dealcoholise the wine up to 0.05% (v/v) and rectify the alcohol up to 85% (v/v) (Figure 1), being able to obtain a final product with the desired alcohol content and the initial aromatic fraction [18].

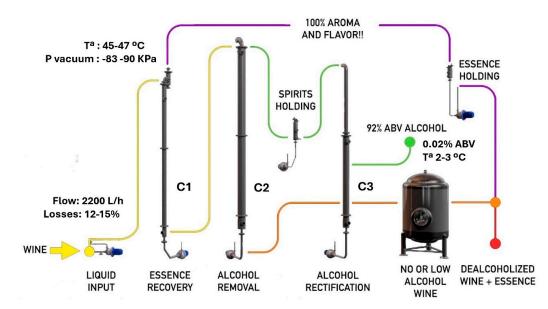


Figure 1. Scheme of the GoLo dealcoholisation process. (ABV: alcohol by volume).

The GoLo dealcoholisation technology is based on vacuum distillation (-83-90 KPa) at low temperatures (45–47 °C) and the outlet temperature of the dealcoholised wine is 2 °C. The process consists of three distillation columns (C1, C2 and C3) which can be described as stainless steel cylinders filled with stainless steel pellets as a filter with a large interfacial area.

The wine to be dealcoholised is introduced at the top of column C1 and by gravity moves down through the column packing to the bottom of the column, where it is collected by a centrifugal pump and sent to the top of column C2. The dearomatised wine coming from the bottom of C1 contains 1 to 3 degrees less alcohol than the input wine. The vapours (100% of wine aroma and flavour, rich in low molecular weight aromatic compounds) are extracted, condensed and stored by the head of column C1 to be reincorporated into the final product at the end of the process.

The product sent to the top of column C2 moves down through the column filler to the bottom where the product would already be dealcoholised and is sent to the storage tanks (this product may be considered a fully dealcoholised wine or a wine with reduced alcohol content). The alcoholic vapours produced in column C2 [40–50% (v/v)] are sent to the top of column C3, which moves to its bottom by passing through the packing. From the head, it is sent to a condenser, and some of the liquid (water) is recirculated to column C2, and some is extracted as concentrated alcohol and sent to another storage tank.

The vapours that move to the top of the columns are produced in the evaporator at the bottom of column C2. This evaporator is heated by boiler steam and boils some of the liquid from the bottom of the column, which flows counter current to the liquid through the packing, acting as an extracting agent for the more volatile components of the product (alcohol and flavourings). These vapours pass to the bottom of column C3, although a certain percentage of them also pass to column C1. Once the dealcoholisation stage has been completed, the aromatic compounds recovered in column C1 (100% of the aroma and flavour of non-dealcoholised wine) are reincorporated into the dealcoholised wine, so that it retains its organoleptic qualities intact.

According to information supplied by the company that provided the samples for this study, which has both SCC and GoLo technology in its facilities, compared to SCC (the dealcoholisation technology most similar to GoLo), GoLo operates at higher flow rates (2200 L/h compared to 1800 L/h for SCC) with lower energy consumption (0.021 kw/L compared to 0.028 kw/L for SCC) and lower losses (15% compared to 30% for SCC).

2.4. Sample Analysis and Determination

The samples of non-dealcoholised wines were analysed after being delivered by the company. The samples were analysed again after the process of dealcoholisation using GoLo technology. Each sample was analysed in triplicate.

According to EC [36] and OIV [27], the method based on electronic densimetry was used for the analysis of alcohol in wines before and after the process of dealcoholisation using GoLo technology.

Free and total SO₂ content in wines before and after the process of dealcoholisation using GoLo technology were analysed via the optimised Monier-Williams method [27,37,38]. It is known that the optimised Monier-Williams method has limitations due to background interference or unsatisfactory accuracy [22]. However, these limitations have not been determined for wines. Finally, pH, total acidity and volatile acidity content were measured according to the Compendium of International Methods of Analysis of Wine and Must OIV [27].

The limit of quantification for each technique was 5 mg/L for free and total SO₂ and 0.1% v/v for alcohol, volatile acidity and total acidity. For calculation purposes, the values of samples with results below the limit of quantification were transformed by dividing the limit of quantification by 2 [39].

2.5. Statistical Analysis

The RStudio IDE Desktop 2023.09.01 tool and R 4.3.1 were used for data analysis and graph generation [40]. This software is free and can be used in both desktop and cloud versions. RStudio allows scripts to be created using code written in R. This can be used to automate optimization for future studies. Different R libraries were used depending on the analysis carried out; for example, the data import into R was performed using the Readxl library, and graph analysis and Student's t-test for paired samples were performed using ggplot2, corrplot, Performance Analytics, dplyr and car. The R script code used in this paper is available on request from the corresponding author. A more detailed analysis of the data contained in the present manuscript can be consulted at the following web address https://rpubs.com/chemavb/1159224 (accessed on 15 March 2024).

3. Results and Discussion

3.1. Analysing the Dealcoholisation Process Using GoLo Technology

Figure 2 shows the composition of all the non-dealcoholised wines analysed where each sub-figure, represented by a grey rectangle, indicates the interquartile range (25% quartile to 75% quartile). Within each rectangle, there is a shaded line representing the median or central value, and above and below the rectangle, there are vertical dashed lines representing the minimum and maximum values for each variable. Each figure also includes rounded points representing values that were identified as outliers because they are far from the maximum and minimum. Of note, the median and interquartile range of the data are presented in a boxplot for a better interpretation of the results obtained after the dealcoholisation process using GoLo technology.

Table 1 shows the composition of all the non-dealcoholised wines analysed broken down by type of wine. As can be seen, the results presented in Table 1 and Figure 1 are similar to those reported in previous studies on commercial white, rose and red wines from Spain [41–46]. On the other hand, the total SO₂ content is sufficiently variable (from a minimum of 52.75 mg/L to a maximum of 115 mg/L) to be able to evaluate, in different ranges, what happens to the SO₂ content after the GoLo process at a medium-high ethanol content between 9.56 and 13.6%.

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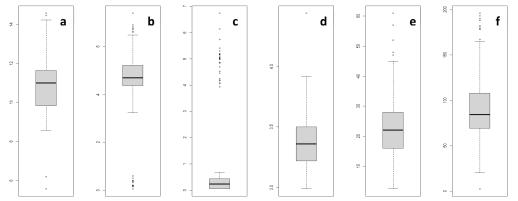


Figure 2. Boxplot of the composition of all the non-dealcoholised wines analysed. (a) Alcohol ((v/v); (b) total acidity (g/L); (c) volatile acidity (g/L); (d) pH; (e) free SO₂ (mg/L) and (f) total SO₂ (mg/L).

Table 1. Characterisation of non-dealcoholised wines studied as median and interquartile range (Q1–Q3).

| Wine | pH | Total Acidity (g/L) | Volatile Acidity (g/L) | Ethanol (% v/v) | Free SO ₂ (mg/L) | Total SO ₂ (mg/L) |
|-------|------------------|---------------------|---------------------------|------------------------|--------------------------------|---------------------------------|
| White | 3.29 (3.20–3.41) | 4.66 (4.27–5.36) | 0.17 (0.05–0.32) | 10.77 (9.56–11.30) | 23.15 (16.5–29) | 93 (77.5–113.5) |
| Rose | 3.22 (3.14–3.32) | 5.10 (4.71–5.55) | 0.18 (0.15–0.25) | 10.92 (10.82–11.61) | 23.0 (12.0–25.0) | 92 (72.0–115.0) |
| Red | 3.61 (3.52–3.69) | 4.65 (4.43-4.95) | 0.46 (0.38–0.52) | 12.23 (11.04–13.06) | 22.0 (16.0–28.0) | 65 (52.75-82.25) |

Figure 3 shows the parameters studied for all the dealcoholised wines using GoLo technology, and Table 2 shows those parameters broken down by the type of wine. The ethanol contents of the 274 non-dealcoholised wines were between 5.57 and 14.59% v/v.

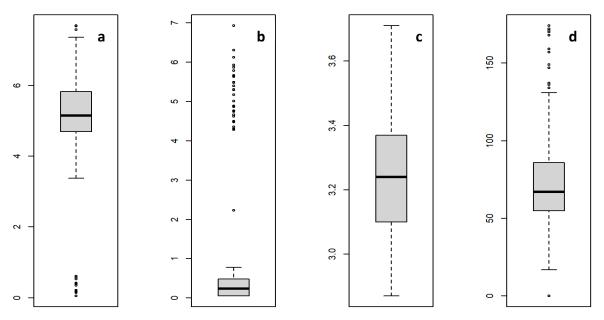


Figure 3. Boxplot of composition of dealcoholised wines using GoLo technology. (a) Total acidity (g/L); (b) volatile acidity (g/L); (c) pH; and (d) total SO₂ (mg/L).

| Wine | рН | Total Acidity (g/L) | Volatile Acidity (g/L) | Alcohol (% v/v) | Free SO ₂ (mg/L) | Total SO ₂ (mg/L) |
|-------|------------------|---------------------|---------------------------|-----------------|--------------------------------|---------------------------------|
| White | 3.18 (3.10–3.30) | 5.05 (4.52–5.89) | 0.17 (0.05–0.35) | <0.1 | <5.0 | 102.0 (84.5–127.5) |
| Rose | 3.10 (3.08–3.19) | 5.76 (4.92–5.96) | 0.19 (0.16–0.29) | <0.1 | <5.0 | 87.0 (70.0–118.0) |
| Red | 3.44 (3.36–3.51) | 5.25 (4.98–5.57) | 0.49 (0.41–0.55) | <0.1 | <5.0 | 88.0 (74.8–104.0) |

Table 2. Characterization of dealcoholised wines studied as median and interquartile range (Q1–Q3).

The results for ethanol are not shown in Figure 3 because the dealcoholisation process using GoLo technology reduced the initial alcohol content of the wines to levels below the method's limit of quantification (0.1% v/v), thus achieving a 100% yield. The total dealcoholisation of wines has also been reported using SCC to achieve a final wine alcohol content of 0.3% v/v with high capital and operating costs due to the number of ancillary devices required [47]; however, technologies such as NF, RO, OD or PV can achieve a final ethanol content in the wine of more than 0.5% v/v [9]. It can, therefore, be said that the GoLo technology achieves the total dealcoholisation of wines in less time, with less losses and with less energy consumption than other dealcoholisation technologies.

The free SO_2 content obtained for the 274 non-dealcoholised wines was between 5 and 61 mg/L. Similar to the alcohol content and as would be expected, the dealcoholisation of wines using GoLo technology also provides the 100% removal of free SO₂ along with its antioxidant and antimicrobial activity. The mechanism by which free SO_2 is removed in the wine dealcoholisation process was previously reported by Belisario-Sánchez, Taboada-Rodríguez, Marín-Iniesta and López-Gómez [31]. Briefly, they reported that after the mass balance of the SCC distillation, ethanol and free SO₂ have a similar relative volatility with respect to water, because the free SO_2 of the non-dealcoholised wine is mainly present in the ethanol fraction. In this sense, if the content of free SO_2 in the wine is less than 15 mg/l, the wine would be unprotected and susceptible to microbial spoilage, which is why dealcoholised wines require the addition of preservatives to achieve the required shelf life on the market [48,49]. These preservatives include SO₂ itself and other traditional preserving agents such as dimethyl decarbonate or potassium sorbate, Valcorin [50] or Nagardo[®](Lanxess Corp., Pittsburgh, PA, USA), the natural preservative for dealcoholised drinks approved by Food and Drugs Agency of USA in 2017 at concentration levels ranging from 2 to 100 ppm [49].

The total SO₂ present in wine is partly gaseous (SO₂) and partly dissolved (HSO₃⁻ and SO₃²⁻), forming what is known as free SO₂, and partly combined with acetic aldehyde, sugars, tannins, colourings, etc., forming combined SO₂. This distinction is important for practical purposes, since the SO₂ with antiseptic action is the free one, while the combined one constitutes the necessary reserve for the free fraction. In other words, the two forms are in equilibrium, which is influenced by pH and temperature (the lower the pH and the higher the temperature, the greater the proportion of free SO₂). The sum of the free and combined SO₂ is equal to the total SO₂ [51].

With this in mind, and knowing that dealcoholised wine contains no free SO₂, only the combined SO₂ should remain as a contributor to the total SO₂ in the dealcoholised wine. In this sense, the total SO₂ content obtained for the 274 non-dealcoholised wines ranged from 2.5 to 196 (mg/L), while for the corresponding dealcoholised wines, it ranged from 55 to 86 (mg/L), which represents a reduction of 22%. A paired samples t-test showed that there were significant differences ($p = 2.35 \times 10^{-8}$) in the reduction in total SO₂ in the dealcoholisation process using the GoLo technology. Significant reductions in total SO₂ were also achieved with other dealcoholisation technologies such as RO or VD [52].

With regard to the other parameters studied, there was a 3.6% increase in the pH value (from 3.24 to 3.36) after the dealcoholisation process using GoLo technology, total acidity decreased by 9.3% (from 5.15 to 4.69) and volatile acidity decreased by 4.3% (from 0.24 to

0.23). However, after statistical analysis using the t-test, it was found that there were no significant differences in the variations in these parameters. Thus, pH, total acidity and volatile acidity were not affected by the dealcoholisation process using GoLo technology. This fact has also been observed using other dealcoholisation technologies such as PV [53], OD [13], RO [54] and SCC [55].

3.2. Variable Correlation Analysis

Since the statistical analysis of parameter reduction did not reveal any differences between the wines taken as a whole or by typology (white, rose and red), a variable correlation analysis was carried out to identify possible relationships between the different quality parameters affecting the GoLo dealcoholisation technology. The same analysis was also used to identify the relationship between both free and total SO₂ before the dealcoholisation process and the total SO₂ after the dealcoholisation process.

Figure 4 shows the scatter plot of the relationship between free and total SO_2 , and by evaluating the regression model, Equation (1) was obtained.

Free
$$SO_2 = 12.20848 + 0.11785 \times \text{total } SO_2$$
 (1)

where free SO_2 and total SO_2 are the contents of free SO_2 and total SO_2 , expressed in mg/L, of the non-dealcoholised wine.

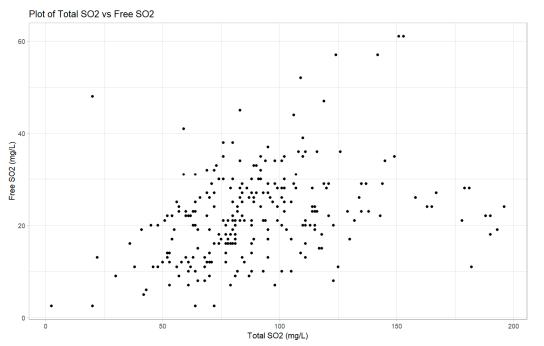


Figure 4. Scatter plot of the relationship between free and total SO₂.

Coefficients and the model were significant (*p*-value < 0.01), indicating a relationship between the response variable (free SO₂) and predictor (total SO₂). However, the value of R^2 was low (0.1437), and the standard residual error was high (9.54), which would allow it to be used not for precise predictions but only for first approximations.

Figure 5 shows the analysis of correlations between the variables studied before the GoLo dealcoholisation process and the total SO₂ content after the dealcoholisation process. The distribution of each variable is shown on the diagonal. At the bottom of the diagonal are the bivariate scatter plots with a fitted line, at the top of the diagonal are the correlation value plus the significance level as stars, and each significance level is associated with a symbol of the *p*-values [0.001 (***), 0.01 (**), 0.05 (*), 0.1 ("."), 1 (".")].

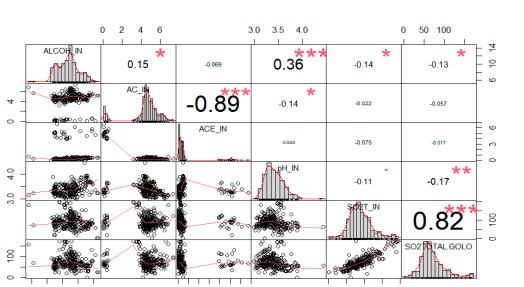


Figure 5. Correlations analysis between the variables studied.

0

Figure 5 shows that the total SO₂ content of dealcoholised wines using GoLo technology is correlated with the total SO₂ (0.82; *p*-value = 0.001), pH (-0.17; *p*-value = 0.01) and the alcohol content (-0.13; *p*-value = 0.05) of the non-dealcoholised wines. Given the existence of significant variables in the correlation analysis, a linear multivariate regression model was adjusted, obtaining the model shown in Equation (2).

0 50

150

$$\text{Total SO}_2\text{GoLo}\left(\frac{\text{mg}}{\text{L}}\right) = 42.95 + 0.45328 \times \text{alcohol}\left(\%\frac{\text{v}}{\text{v}}\right) - 13.13024 \times \text{pH} + 0.77445 \times \text{total SO}_2\left(\frac{\text{mg}}{\text{L}}\right)$$
(2)

2

4 6

where total SO₂ GoLo is the content of total SO₂, expressed in mg/L, that is expected in the dealcoholised wines on the basis of the alcohol content ((v/v), pH and total SO₂ (mg/L) of the non-dealcoholised wines.

The model obtained showed an R² of 0.69, which indicates that the model is capable of explaining 69 percent of the variability observed in the total SO₂ value, which was 2.2×10^{-16} , indicating that the model is significant. In this sense, this model can be used to accurately predict the total SO₂ content after the GoLo process, thus saving the time and cost of laboratory analysis.

Finally, due to the loss of free SO_2 in the dealcoholisation process, and in order to ensure a sufficient level of SO_2 in the wine to guarantee its preservative, antioxidant and microbial functions, Equation (1) together with Equation (2) can be used as an approximation of the amount of SO_2 to be added to the dealcoholised wine to ensure a minimum total SO_2 level to guarantee its quality but without exceeding the maximum permitted level [56].

The use of GoLo technology for wine dealcoholisation allows for the complete dealcoholisation of any type of wine, eliminating all free SO_2 and significantly reducing total SO_2 without altering pH, volatile acidity and total acidity. This provides access to a new type of technology that is economically viable and easy to implement in companies that have a need to dealcoholise wine.

4. Conclusions

6 8 10

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For the first time, this paper studies the quality of wines dealcoholised using the patented dealcoholisation technology called GoLo. There were no significant differences in the variations in pH, total acidity and volatile acidity after the dealcoholisation process. Compared to other dealcoholisation technologies, GoLo achieves the total dealcoholisation and free SO₂ removal of wines in less time, with fewer losses and with less energy consumption. During the process, free SO₂ is also eliminated and sulphites or other preservatives should be added to guarantee the dealcoholised wine's shelf life. The variable correlation analysis revealed a model for predicting the total SO₂ content of dealcoholised wines as a

function of pH, alcohol content and the total SO_2 of non-dealcoholised wines, as well as a model for predicting the amount of sulphites to be added after the dealcoholisation process. These results will be subject to verification and extension in future studies with an increase in the number of parameters under investigation such as colour, phenolic profile, reductor sugars, volatile fraction analysis or sensory evaluation.

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References

- 1. Dias, R.; Pérez-Gregorio, R. Phenolic Compounds in Wine. Beverages 2023, 9, 70. [CrossRef]
- 2. Mitrović, D.; Sredović Ignjatović, I.; Kozarski, M.; Popović-Đorđević, J. Wine is more than just a beverage: Chemical diversity, health benefits, and immunomodulating potential of wine polyphenols. *Food Saf. Health* **2024**, *2*, 196–212. [CrossRef]
- 3. Haseeb, S.; Alexander, B.; Santi, R.L.; Liprandi, A.S.; Baranchuk, A. What's in wine? A clinician's perspective. *Trends Cardiovasc. Med.* **2019**, *29*, 97–106. [CrossRef] [PubMed]
- Díaz, L.A.; Fuentes-López, E.; Idalsoaga, F.; Ayares, G.; Corsi, O.; Arnold, J.; Cannistra, M.; Vio, D.; Márquez-Lomas, A.; Ramirez-Cadiz, C.; et al. Association between public health policies on alcohol and worldwide cancer, liver disease and cardiovascular disease outcomes. J. Hepatol. 2024, 80, 409–418. [CrossRef]
- Visioli, F.; Panaite, S.-A.; Tomé-Carneiro, J. Wine's Phenolic Compounds and Health: A Pythagorean View. *Molecules* 2020, 25, 4105. [CrossRef] [PubMed]
- 6. Wood, A.M.; Kaptoge, S.; Butterworth, A.S.; Willeit, P.; Warnakula, S.; Bolton, T.; Paige, E.; Paul, D.S.; Sweeting, M.; Burgess, S.; et al. Risk thresholds for alcohol consumption: Combined analysis of individual-participant data for 599912 current drinkers in 83 prospective studies. *Lancet* **2018**, *391*, 1513–1523. [CrossRef] [PubMed]
- Saliba, A.J.; Ovington, L.A.; Moran, C.C. Consumer demand for low-alcohol wine in an Australian sample. *Int. J. Wine Res.* 2013, 5, 1–8. [CrossRef]
- 8. Pickering, G.J. Low- and Reduced-alcohol Wine: A Review. J. Wine Res. 2000, 11, 129–144. [CrossRef]
- Sam, F.E.; Ma, T.-Z.; Salifu, R.; Wang, J.; Jiang, Y.-M.; Zhang, B.; Han, S.-Y. Techniques for Dealcoholization of Wines: Their Impact on Wine Phenolic Composition, Volatile Composition, and Sensory Characteristics. *Foods* 2021, 10, 2498. [CrossRef]
- Bucher, T.; Deroover, K.; Stockley, C. Low-Alcohol Wine: A Narrative Review on Consumer Perception and Behaviour. *Beverages* 2018, 4, 82. [CrossRef]
- 11. Afonso, S.M.; Inês, A.; Vilela, A. Bio-Dealcoholization of Wines: Can Yeast Make Lighter Wines? *Fermentation* **2024**, *10*, 36. [CrossRef]
- 12. Catarino, M.; Mendes, A. Dealcoholizing wine by membrane separation processes. *Innov. Food Sci. Emerg. Technol.* **2011**, *12*, 330–337. [CrossRef]
- Liguori, L.; Russo, P.; Albanese, D.; Di Matteo, M. Evolution of quality parameters during red wine dealcoholization by osmotic distillation. *Food Chem.* 2013, 140, 68–75. [CrossRef] [PubMed]
- 14. Schulz, F.N.; Farid, H.; Hanf, J.H. The Lower the Better? Discussion on Non-Alcoholic Wine and Its Marketing. *Dietetics* 2023, 2, 278–288. [CrossRef]
- 15. Schmidtke, L.M.; Blackman, J.W.; Agboola, S.O. Production Technologies for Reduced Alcoholic Wines. *J. Food Sci.* 2012, 77, R25–R41. [CrossRef] [PubMed]
- 16. Mangindaan, D.; Khoiruddin, K.; Wenten, I.G. Beverage dealcoholization processes: Past, present, and future. *Trends Food Sci. Technol.* **2018**, *71*, 36–45. [CrossRef]
- 17. Díaz-Gálvez, I.; Gutiérrez-Gamboa, G.; Plaza, A.; Concha-Meyer, A.A. Effect of Encapsulation Processes by Freeze and Spray Drying on the Antioxidant Properties of Red Wine from cv. Listan Prieto and Syrah. *Foods* **2022**, *11*, 3880. [CrossRef] [PubMed]
- Pienaar, S.W. Process and Apparatus for the Reduction of Alcohol in Fermented Beverages. U.S. Patent US20150132459A1, 2 February 2016.
- 19. Stoleicova, V.S. Comparative analysis of the techniques used to reduce alcohol level in wines. Pomic. Vitic. Şi Vinif. 2015, 56, 37–39.

- Vitali Čepo, D.; Pelajić, M.; Vinković Vrček, I.; Krivohlavek, A.; Žuntar, I.; Karoglan, M. Differences in the levels of pesticides, metals, sulphites and ochratoxin A between organically and conventionally produced wines. *Food Chem.* 2018, 246, 394–403. [CrossRef]
- Lisanti, M.T.; Blaiotta, G.; Nioi, C.; Moio, L. Alternative Methods to SO2 for Microbiological Stabilization of Wine. Compr. Rev. Food Sci. Food Saf. 2019, 18, 455–479. [CrossRef]
- 22. Huang, L.; Lai, L.; Zhang, X.; Lin, S.; Jin, G.; Li, D. Practical assay for determining residual sulfite of the wine in rapid detection or quantitative analysis. *LWT* 2023, *189*, 115503. [CrossRef]
- 23. Fazio, T.; Warner, C.R. A review of sulphites in foods: Analytical methodology and reported findings. *Food Addit. Contam.* **1990**, *7*, 433–454. [CrossRef] [PubMed]
- 24. Nardini, M.; Garaguso, I. Effect of Sulfites on Antioxidant Activity, Total Polyphenols, and Flavonoid Measurements in White Wine. *Foods* **2018**, *7*, 35. [CrossRef] [PubMed]
- 25. EC. Regulation (EU) No 1169/2011 of the European Parliament and of the Council of 25 October 2011 on the Provision of Food Information to Consumers, Amending Regulations (EC) No 1924/2006 and (EC) No 1925/2006 of the European Parliament and of the Council, and Repealing Commission Directive 87/250/EEC, Council Directive 90/496/EEC, Commission Directive 1999/10/EC, Directive 2000/13/EC of the European Parliament and of the Council, Commission Directives 2002/67/EC and 2008/5/EC and Commission Regulation (EC) No 608/2004 Text with EEA Relevance. 2011. Available online: https: //eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32011R1169 (accessed on 10 March 2024).
- 26. EFSA Panel on Food Additives and Nutrient Sources Added to Food (ANS). Scientific Opinion on the re-evaluation of sulfur dioxide (E 220), sodium sulfite (E 221), sodium bisulfite (E 222), sodium metabisulfite (E 223), potassium metabisulfite (E 224), calcium sulfite (E 226), calcium bisulfite (E 227) and potassium bisulfite (E 228) as food additives. *Efsa J.* **2016**, *14*, 4438. [CrossRef]
- 27. OIV. Compendium of International Methods of Analysis of Wine and Must. 2023. Available online: https://www.oiv.int/ standards/compendium-of-international-methods-of-wine-and-must-analysis (accessed on 22 January 2024).
- 28. Navarre, C.; Belly, P. L'oenologie, 8th ed.; Tec & Doc Lavoisier: Paris, France, 2017.
- 29. Ma, T.; Sam, F.E.; Didi, D.A.; Atuna, R.A.; Amagloh, F.K.; Zhang, B. Contribution of edible flowers on the aroma profile of dealcoholized pinot noir rose wine. *LWT* **2022**, *170*, 114034. [CrossRef]
- Belisario-Sánchez, Y.; Taboada-Rodríguez, A.; Marín, F.; Iguaz, A.; López-Gómez, A. Aroma Recovery in Wine Dealcoholization by SCC Distillation. *Food Bioprocess Technol.* 2011, *5*, 2529–2539. [CrossRef]
- Belisario-Sánchez, Y.Y.; Taboada-Rodríguez, A.; Marín-Iniesta, F.; López-Gómez, A. Dealcoholized Wines by Spinning Cone Column Distillation: Phenolic Compounds and Antioxidant Activity Measured by the 1,1-Diphenyl-2-picrylhydrazyl Method. J. Agric. Food Chem. 2009, 57, 6770–6778. [CrossRef]
- 32. Gómez-Plaza, E.; López-Nicolás, J.M.; López-Roca, J.M.; Martínez-Cutillas, A. Dealcoholization of Wine. Behaviour of the Aroma Components during the Process. *LWT—Food Sci. Technol.* **1999**, *32*, 384–386. [CrossRef]
- Pickering, G.J.; Heatherbell, D.A.; Barnes, M.F. GC-MS Analysis of Reduced-alcohol Müller-Thurgau Wine Produced using Glucose Oxidase-treated Juice. LWT—Food Sci. Technol. 2001, 34, 89–94. [CrossRef]
- Nikolaou, A.; Kourkoutas, Y. High-Temperature Semi-Dry and Sweet Low Alcohol Wine-Making Using Immobilized Kefir Culture. *Fermentation* 2021, 7, 45. [CrossRef]
- 35. *ISO/IEC-17025*; General Requirements for the Competence of Testing and Calibration Laboratories. ISO: Geneva, Switzerland, 2017. Available online: https://www.iso.org/obp/ui#iso:std:iso-iec:17025:ed-3:v2:es (accessed on 15 March 2024).
- 36. EC. COMMISSION REGULATION (EC) No 128/2004 of 23 January 2004 Amending Regulation (EEC) No 2676/90 Determining Community Methods for the Analysis of Wines. 2004. Available online: https://eur-lex.europa.eu/LexUriServ/LexUriServ.do? uri=OJ:L:2004:019:0003:0011:EN:PDF (accessed on 24 January 2024).
- UNE-EN 1988-1:2000; Foodstuffs—Determination of Sulfite—Part 1: Optimized Monier-Williams Method. UNE: Biddeford, ME, USA, 2001. Available online: https://www.une.org/encuentra-tu-norma/busca-tu-norma/norma?c=N0023351 (accessed on 24 January 2024).
- AOAC. Official Methods of Analysis. Chapter 47. Monier-Williams AOAC Official Method (Optimized Method) 990.28; NQAC: Dublin, Ireland, 2000; pp. 29–30.
- Keizer, R.J.; Jansen, R.S.; Rosing, H.; Thijssen, B.; Beijnen, J.H.; Schellens, J.H.M.; Huitema, A.D.R. Incorporation of concentration data below the limit of quantification in population pharmacokinetic analyses. *Pharmacol. Res. Perspect.* 2015, *3*, e00131. [CrossRef] [PubMed]
- 40. Aria, M.; Cuccurullo, C. Bibliometrix: An R-tool for comprehensive science mapping analysis. *Informetrics* **2017**, *11*, 959–975. [CrossRef]
- Cadahía, E.; Fernández de Simón, B.; Sanz, M.; Poveda, P.; Colio, J. Chemical and chromatic characteristics of Tempranillo, Cabernet Sauvignon and Merlot wines from DO Navarra aged in Spanish and French oak barrels. *Food Chem.* 2009, 115, 639–649. [CrossRef]
- 42. Sáenz-Navajas, M.-P.; Avizcuri, J.-M.; Ferreira, V.; Fernández-Zurbano, P. Insights on the chemical basis of the astringency of Spanish red wines. *Food Chem.* 2012, 134, 1484–1493. [CrossRef] [PubMed]
- 43. Vilanova, M.; Escudero, A.; Graña, M.; Cacho, J. Volatile composition and sensory properties of North West Spain white wines. *Food Res. Int.* **2013**, *54*, 562–568. [CrossRef]

- 44. Pérez-Magariño, S.; Ortega-Heras, M.; González-San José, M.L. Multivariate classification of rosé wines from different Spanish protected designations of origin. *Anal. Chim. Acta* 2002, 458, 187–190. [CrossRef]
- 45. Pérez-Magariño, S.; Ortega-Heras, M.; González-San José, M.L.; Boger, Z. Comparative study of artificial neural network and multivariate methods to classify Spanish DO rose wines. *Talanta* **2004**, *62*, 983–990. [CrossRef]
- 46. Castrillo, D.; Blanco, P. Peculiarities of the Organic Wine in Galicia (NW Spain): Sensory Evaluation and Future Considerations. *Beverages* **2023**, *9*, 89. [CrossRef]
- Margallo, M.; Aldaco, R.; Barceló, A.; Diban, N.; Ortiz, I.; Irabien, A. Life cycle assessment of technologies for partial dealcoholisation of wines. *Sustain. Prod. Consum.* 2015, 2, 29–39. [CrossRef]
- Taboada-Rodríguez, A.; Belisario-Sánchez, Y.Y.; Cava-Roda, R.; Cano, J.A.; López-Gómez, A.; Marín-Iniesta, F. Optimisation of preservatives for dealcoholised red wine using a survival model for spoilage yeasts. *Int. J. Food Sci. Technol.* 2013, 48, 707–714. [CrossRef]
- 49. Galasong, Y.; Sogin, J.H.; Worobo, R.W. Natural glycolipids inhibits certain yeasts and lactic acid bacteria pertinent to the spoilage of shelf stable beverages. *Food Control* **2023**, *146*, 109544. [CrossRef]
- 50. EFSA. Scientific opinion on the re-evaluation of dimethyl dicarbonate (DMDC, E 242) as a food additive. *EFSA J.* **2015**, *13*, 4319. [CrossRef]
- Giménez-Gómez, P.; Gutiérrez-Capitán, M.; Puig-Pujol, A.; Capdevila, F.; Muñoz, S.; Tobeña, A.; Miró, A.; Jiménez-Jorquera, C. Analysis of free and total sulfur dioxide in wine by using a gas-diffusion analytical system with pH detection. *Food Chem.* 2017, 228, 518–525. [CrossRef] [PubMed]
- 52. Sam, F.E.; Ma, T.; Liang, Y.; Qiang, W.; Atuna, R.A.; Amagloh, F.K.; Morata, A.; Han, S. Comparison between Membrane and Thermal Dealcoholization Methods: Their Impact on the Chemical Parameters, Volatile Composition, and Sensory Characteristics of Wines. *Membranes* **2021**, *11*, 957. [CrossRef] [PubMed]
- Pham, D.-T.; Stockdale, V.J.; Wollan, D.; Jeffery, D.W.; Wilkinson, K.L. Compositional Consequences of Partial Dealcoholization of Red Wine by Reverse Osmosis-Evaporative Perstraction. *Molecules* 2019, 24, 1404. [CrossRef]
- 54. Longo, R.; Blackman, J.W.; Antalick, G.; Torley, P.J.; Rogiers, S.Y.; Schmidtke, L.M. A comparative study of partial dealcoholisation versus early harvest: Effects on wine volatile and sensory profiles. *Food Chem.* **2018**, *261*, 21–29. [CrossRef]
- Puglisi, C.; Ristic, R.; Saint, J.; Wilkinson, K. Evaluation of Spinning Cone Column Distillation as a Strategy for Remediation of Smoke Taint in Juice and Wine. *Molecules* 2022, 27, 8096. [CrossRef]
- 56. EU. Commission Delegated Regulation (EU) 2019/934 of 12 March 2019 Supplementing Regulation (EU) No 1308/2013 of the European Parliament and of the Council as Regards Wine-Growing Areas where the Alcoholic Strength may Be Increased, Authorised Oenological Practices and Restrictions Applicable to the Production and Conservation of Grapevine Products, the Minimum Percentage of Alcohol for by-Products and Their Disposal, and Publication of OIV Files. 2019. Available online: https://eur-lex.europa.eu/eli/reg_del/2019/934/oj (accessed on 24 January 2024).

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