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Production of Syrups from Corinthian Currant Industrial Finishing Side-Stream: Quality Evaluation and Volatilome

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Abstract: The industrial finishing side-stream (FSS) of premium-quality Corinthian currants was used to produce syrups with/without pigment- and tartrate-reduction treatments. The chemical composition, total titratable acidity (TTA; % *w*/*w* as tartaric acid), volatile acidity (VA; % *w*/*w* as acetic acid), total phenolic content (TPC; as gallic acid, GA), antioxidant capacity (AC; as ascorbic acid, AA), volatilome (SPME GC-MS), sensory properties, and microbial stability were compared. All syrups had similar average sugar content (65.4–69.4% *w*/*w*), and no sucrose. Those not treated for tartrate reduction were more acidic (pH ~4.5) than those treated (pH > 5.6), while all syrups had higher pH than similar commercial products (3.0–4.5). On the other hand, the FSS syrups had similar TTA (<1.2 ± 0.3%) despite the applied treatment, and had low VA (0.08–0.27%). The blonde syrups had a lower average TPC (134–143 mg GA/100 g) and AC (0.90–1.0 mg AA/100 g) than the brown syrups (185–213 and 0.3–0.6, respectively), due to the removal of phenolics in the clarification treatments. Totally 144 headspace aroma volatiles were identified, deriving either from the grapes or the raisin-drying process. HMF was not detected. The sensory, microbiological, and VA analyses indicated that FSS can be used to produce high-quality, preservable, and added-value syrups.

Keywords: Corinthian currants; finishing side-stream; valorization; syrup; phenolic content; antioxidant capacity; volatilome

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1. Introduction

Syrup refers to any viscous liquid consisting of a concentrated sugar solution with/without the addition of flavorings. The global syrup market (chocolate, maple, fruit, malt, corn, rice, tapioca, honey, flavored, and other types of syrups) records billion-dollar revenues with increased compound annual growth rates, despite the uncertainties caused by the COVID-19 outbreak [1,2]. The increasing demand for syrups is due to their numerous applications as sweeteners, taste enhancers, and flavoring agents in processed food production (beverages, bakery products, desserts, confectioneries, etc.), as well as due to increasing globalization rates, rising incomes, and adoption of urban lifestyles. However, increased consumer awareness and demands towards healthier processed foods has also increased scientific and industrial interest in the development of low-calorie, low-glycemic index, and additive-free syrups and sweeteners [2]. Other recent research trends related to the production of syrups include optimization of production conditions to minimize the energy requirements [3], use of enzyme immobilization techniques (glucose isomerase, β-galactosidase, etc.) for continuous syrup production, development of combined enzyme methods for single-step carbohydrate hydrolysis and isomerization [4,5], development

of waste biomass pretreatment and hydrolysis methods [6], genetic engineering for the production of efficient low-cost enzymes related to syrup production [7], etc.

Grape syrups are an important category within the global syrup market. Grape syrup is produced either by concentration (boiling) of fresh grape must (grape juice concentrates, such as petimezi in Greece, pekmez in Turkey, etc.) or by concentration of raisin extracts (raisin syrups, such as stafidini in Greece). According to the Greek Food and Drinks Code (art. 66) [8], raisin syrup is the product obtained by concentrating an aqueous raisin extract after removing most of its acid (tartrate) content. Pigment removal and addition of flavorings is allowed as long as it is stated on the product label. The presence of sulfite, which is a common antioxidant and preservative, should not exceed 40 mg/kg in the final product [8]. Raisin syrup is a natural sweetener with a high energy value, and due to its high content of reducing sugars (70–71% *w*/*w*, half of which is fructose), trace elements (K, Na, P, Mg, Cu), and vitamins (C, B3, A), its addition to food meets the requirements of consumers for healthier foods. It can also be used as a natural brown food coloring, while providing a typical fruity aroma [9].

The available scientific literature on the production of raisin syrups is scarce. In Greece, raisin syrup is usually produced from the industrial finishing side-stream (FSS) of raisins (mainly Corinthian currants), which until recently was mainly supplied to the vinegar production sector [10–12]. In this study, the production and quality evaluation, including volatilome, total phenolic content (TPC), and antioxidant capacity (AC), of brown and blonde syrups made from the FSS generated from premium-quality Corinthian currants Vostitsa (Protected Designation of Origin, PDO), grown exclusively in the area of Aeghion in western Greece [11], is presented. These currants have been extensively studied for their nutritional value, and the published works have highlighted their rich composition in antioxidant phenolics, other bioactive components, and micronutrients [12], as well as their potential health benefits, including in Alzheimer's disease [13], atherosclerosis [14], athlete endurance, performance, and blood redox status [15], cancer, diabetes [16], etc.

As recently reported [10,12], a currant processing company produces FSS that accounts for 5–6% of the raw material, additionally to that generated during harvesting in the field. FSS differs from the marketable currants in the size of the raisin berries and the presence of seeds and other plant material (e.g., stems), and has a high nutritional value, rich volatilome, and increased AC compared to the raw material. The exploitation of FSS for added-value products such as syrups is essential for the sustainability of Corinthian currant production, which is gradually being abandoned by farmers due to the expensive and laborious cultivation practices and the absence of governmental subsidies [10,12]. Other products from FSS that have been recently proposed (wines, distilled liqueurs, specialty vinegars, microbial metabolites, etc.) [10,12,17] can also create significant added value for the raisin-processing sector. Also, since raisins are typical components of the Mediterranean diet, this study also aims to contribute to the promotion of and adherence to such dietary patterns, which in combination with sustainable growth of commodities such as the Corinthian currants, will positively affect local biodiversity, agricultural communities, processing companies, and the national, regional, and local economies.

Therefore, in this study, methods for brown and blonde syrup production from FSS extracts of the premium-quality Vostitsa PDO currants, with and without tartrate reduction and depigmentation treatments, are for the first time proposed and compared. No extra sugars or additives were added, except sulfite in the extracts before condensation to avoid spoilage. The condensation was carried out at low temperature (45 °C) under vacuum to avoid nutritional and sensory degradation. The proximate composition, total titratable acidity (TTA), volatile acidity (VA), TPC, AC, volatilome, microbial stability during storage, and sensory properties of the syrups were evaluated and compared.

2. Materials and Methods

2.1. Chemicals

The chemicals used in this study for the treatment of syrups (tartrate reduction, clarification, depigmentation, sulfite addition, and subsequent oxidation) and the analytical methods used (acidity, sugars, protein, TPC, AC, volatiles, microbial counts), were: NaOH (Lach-Ner, Neratovice, Czech Repulic); Na_2CO_3 (Penta, Prague, Czech Republic); $CaCO_3$ (Chemco, Athens, Greece); Ringer tablets, methyl orange, 2-propanol, $NaHSO_3$, dioxane, acetonitrile, and KOH (Merck, Darmstadt, Germany); standard 0.1 M NaOH and HCl solutions, $(NH_4)_2SO_4$, $CuSO_4 \cdot 5H_2O$, phenolphthalein, and fructose (Chem-Lab, Zedelgem, Belgium); glucose, methanol, $K_4[Fe(CN)_6] \cdot 3H_2O$, and $ZnSO_4 \cdot 7H_2O$ (Fisher Scientific, Loughborough, UK); saccharose (Chembiotin, Athens, Greece); 2,2-Diphenyl-1-picrylhydrazyl radical (DPPH) (Duchefa Biochemie, Haarlem, The Netherlands); K_2SO_4, H_2SO_4, and gallic acid (Sigma-Aldrich, St. Louis, MO, USA); H_2O_2 30% and iodine solution (Carlo-Erba, Val de Reuil, France); Folin–Ciocalteau reagent (Scharlab S.L., Barcelona, Spain); ascorbic acid, Plate Count Agar (PCA), Violet Red Bile Glucose Agar (VRBGA) (Fluka BioChemika, Buchs, Switzerland); Potato Dextrose Agar (PDA) (Condalab, Madrid, Spain); starch (Riedel-de Haën, Seelze, Germany); C8-C24 n-alkanes (Niles, IL, USA); $K_2S_2O_5$, bentonite, and activated carbon (Syndesmos S.A., Athens, Greece).

2.2. FSS Extract Production

The FSS was obtained from the Agricultural Cooperatives' Union of Aeghion S.A. (201 Korinthou Str., Aeghion, Greece). It was used for syrup production after extraction by maceration with hot water (70 °C), to which potassium metabisulfite ($K_2S_2O_5$) was added at an amount suitable to yield 1.2 g SO_2/L of extract, to prevent spontaneous fermentation and microbial spoilage, as is a common industrial practice. The extraction was followed by removal of the solid residues (grape skins, seeds, etc.) from the extract by cloth filtration and centrifugation at 5000 rpm for 10 min (Sigma 3K12, Bioblock Scientific, Sigma Larborzentrifugen GmbH, Osterode, Germany). The received FSS extract had a Baumé hydrometer density of about 15 °Be (255.5 ± 2.7 g/L total sugar content), and was used for the production of the various types of syrups, with and without activated carbon treatment to remove pigments (blonde and brown syrups, respectively), and with/without treatment to reduce tartrate (Figure 1). Before condensation, 30% H_2O_2 was added stoichiometrically in order to reduce the excess sulfite in the FSS extract to a residual concentration of less than 40 mg/L [8].

2.3. Syrup Production from FSS

2.3.1. Production of Brown Syrups

The FSS extract was concentrated on a rotary evaporator (Heidolph, WB2001) at low temperature (45 °C) to avoid thermal degradation of sugars (especially the heat-labile fructose) through caramelization reactions, which may degrade the syrup quality. The concentration took place until a syrup of 38–40 °Be density was obtained (~65–70% total sugar content). The produced brown syrups without treatment for tartrate reduction (BrST: Brown Syrup with Tartrate) were placed in sealed containers and stored in a dark place at room temperature until further analysis. For the reduction of tartrate in the FSS extract, 70.4% $CaCO_3$ solution was added under continuous stirring until pH 5.45 was reached (Figure 1). The extract remained at 4 °C for 24 h. The precipitated tartrate was removed by centrifugation. Theoretically, 0.67 g/L $CaCO_3$ is required to reduce tartaric acid by 1.0 g/L [18]. The reduction of sulfite in the FSS extract and the concentration to obtain the brown syrup with reduced tartrate (BrS) were carried out as described above for the BrST syrups.

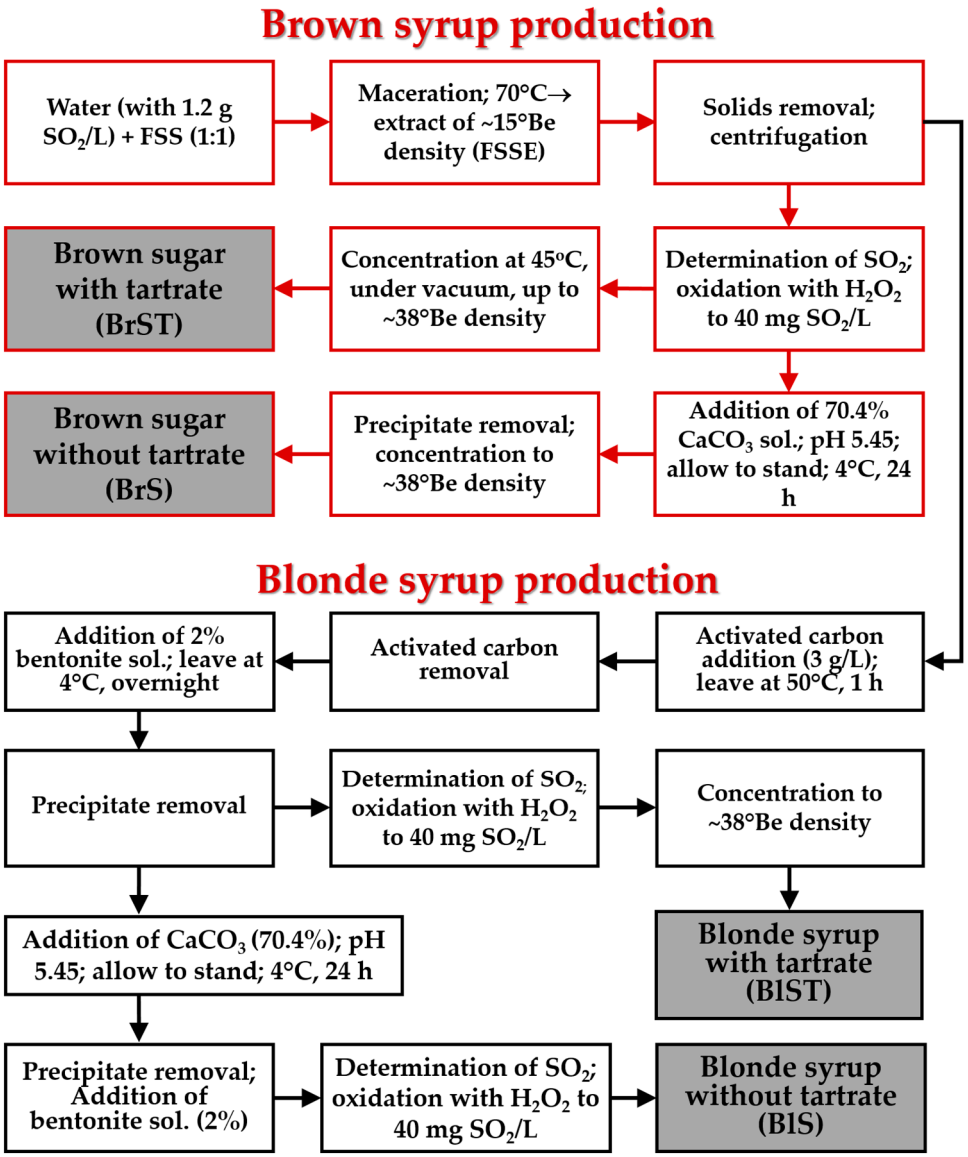

Figure 1. Processes for the production of syrups from the Corinthian currant finishing side-stream (FSS). FSSE: FSS extract. BrST: Brown syrup with tartrate. BrS: Brown syrup with reduced tartrate. BlST: Blonde syrup with tartrate. BlS: Blonde syrup with reduced tartrate.

2.3.2. Production of Blonde Syrups

The same process was followed for the production of blonde syrups, with an additional step for pigment removal (Figure 1). Specifically, 0.3% of activated carbon was added to the FSS extract, which was then heated at 50 °C for 1 h with constant stirring. When the extract was cooled down to room temperature, it was centrifuged and filtered under vacuum to completely remove the carbon and obtain a pale-yellow extract. Then 2% *w*/*v* of bentonite was added and the extract was left at 4 °C for 24 h for stabilization (protein haze and undesirable odor removal) [19]. The reduction of sulfite and the extract concentration were carried out as described above to obtain the blonde syrup with tartrate (BlST). The production of blonde syrup with reduced tartrate (BlS) was carried out as in the case of the brown (BrS) syrup (Figure 1).

2.4. Analytical Methods

2.4.1. Determination of pH, Acidity, and Total Nitrogen

The pH value (Cyberscan 10 pH-meter, Eutech Inst., Singapore) and TTA (expressed as g tartaric acid/100 g syrup) were determined after blending 20 g of syrup with 100 mL of water. For TTA, 10 mL of the solution was titrated with std 0.1 M NaOH solution [20].

The VA (expressed as g acetic acid/100 g syrup) was determined with steam distillation (UDK 129 Kjeldahl distillation unit, Velp Scientifica, Usmate Velate, Italy) of a 50 mL sample, followed by titration with std 0.1 M NaOH solution [20].

Protein as total nitrogen was analyzed using a modified Kjeldahl's method. Specifically, 3 g of syrup was digested on an InKjel mM Kjeldahl digestion unit (behr Labor-Technik GmbH, Düsseldorf, Germany) with the following amounts of catalysts: 0.6 g $CuSO_4 \cdot 5H_2O$, 6 g K_2SO_4, and 15 mL H_2SO_4. NH_3 was released from the digested solution by the addition of 50 mL of 40% NaOH solution, steam distillation in the Kjeldahl distillation unit, trapping in 40 mL of std 0.1 M HCl solution, and back-titration with std 0.1 M NaOH [20].

2.4.2. Determination of Sugars

Sugars (fructose, glucose, sucrose) were determined on a Shimadzu LC-9A HPLC system equipped with a Nucleogel Ion 300 OA column, LC-9A pump, RID-6A refractive index detector, CTO-10A column oven (set at 33 °C), and DGU-2A degassing unit [10,12]. The mobile phase was aqueous 0.017 M H_2SO_4 solution at a flow rate of 0.55 mL/min, and 1% v/v 2-propanol solution was used as an internal standard (IS). The sample dilution was 2% v/v, and the injection volume was 40 μL.

2.4.3. Determination of TPC and AC

For the TPC analysis, 0.6 g syrup were diluted with 10 mL water. Then 0.1 mL of the solution, 5 mL of water, and 1 mL of Folin–Ciocalteau reagent were added to 10 mL flasks and left for 30 min in the dark. Then 1 mL of 7.5% w/v Na_2CO_3 solution was added, and the volume of the flask was fixed to 10 mL with water. The mixture was left again for 30 min in the dark. Likewise, calibration solutions of gallic acid and blank (not including the sample) were prepared. The absorbance was measured at 725 nm on a Jasco V-630 UV-vis spectrophotometer. The TPC was expressed as mg of gallic acid equivalents (GAE)/100 g of syrup [10,12].

AC was assessed using the radical scavenging method, based on the decrease in absorbance at 517 nm of a methanolic solution of DPPH against aqueous methanol solution as blank [10,12]. Specifically, 0.6 g syrup were diluted with 10 mL water. In 8 test tubes, 3 mL of 137.6 μM methanolic DPPH solution and various amounts of syrup solutions (in the range 0.05–1 mL) were added, and the volume was fixed with methanol to 4 mL. The samples were left for 30 min in the dark and the absorbance was measured at 517 nm. Calibration solutions of ascorbic acid were also prepared, and the results were expressed as mg ascorbic acid equivalents (AAE)/g syrup.

2.4.4. HMF Analysis

For the HMF analysis after White, 5 g of syrup was diluted with 25 mL of water and transferred to a 50 mL volumetric flask. Then 0.5 mL of Carrez I solution (15 g $K_4[Fe(CN)_6] \cdot 3H_2O$ in 100 mL water) and 0.5 mL of Carrez II solution (30 g $ZnSO_4$ $7H_2O$/100 mL water) were added. The volume of the flask was then fixed to 50 mL. The solution was paper-filtered and the first 10 mL of the filtrate was rejected. Aliquots of 5 mL were transferred into 2 test tubes, and 5 mL of water was added to the first (sample solution) and 5 mL of sodium bisulfite ($NaHSO_3$) solution 0.2% was added to the second tube (reference solution). The absorbance of the sample solution at 284 nm (A_{284}) was determined versus the reference solution to avoid interference of other components at that wavelength. The absorbance at

336 nm (A_{336}) was measured to subtract the background absorbance. The HMF (mg/kg) was quantified using Formula (1) [21,22]:

$$\text{HMF (mg/kg)} = (A_{284} - A_{336}) \times 149.7 \times 5 \times D/W \quad (1)$$

where A_{284} and A_{336} are the absorbance at 284 nm and 336 nm, respectively, D is the dilution coefficient, and W is the sample weight. The constant 149.7 is calculated using Equation (2):

$$(126 \times 1000 \times 1000)/(16830 \times 10 \times 5) \quad (2)$$

where 126 is the MW of HMF, 16,830 ($L \cdot mol^{-1} \cdot cm^{-1}$) is the molar absorptivity coefficient (ε) of HMF at 284 nm, 1000 is the conversion of g into mg, 10 is the conversion of volume 5 into 50 mL, 1000 is the conversion of g sample to kg, and 5 is the theoretical nominal weight of the sample.

2.4.5. Titrimetric Determination of Sulfite

For the determination of sulfite in the syrups, a titrimetric method was applied. Specifically, 10–11 g (±0.01 g) of sample was diluted with water in a 50 mL volumetric flask, the solution was transferred into a 250 mL conical flask containing 25 mL of 1 M KOH solution, and the mixture was allowed to stand for 15 min. This process aims at releasing, as $KHSO_3$, the sulfite that is bound with other compounds in the sample (e.g., carbonyls), according to reaction (3).

$$\text{R-CH(OH)-SO}_3\text{H} + \text{KOH} \rightarrow \text{R-CHO} + \text{KHSO}_3 + \text{H}_2\text{O} \quad (3)$$

Then, 10 mL of 25% v/v H_2SO_4 solution was added to convert HSO_3^{-1} to SO_4^{-2} (reaction 4) and the mixture was titrated with 0.02 N iodine solution with a starch indicator (reaction 5). The total sulfite was calculated as mg SO_2/kg of syrup.

$$\text{KHSO}_3 + \text{H}_2\text{SO}_4 \rightarrow \text{H}_2\text{SO}_3 + \text{KHSO}_4 \quad (4)$$

$$\text{H}_2\text{SO}_3 + \text{H}_2\text{O} + \text{I}_2 \rightarrow \text{H}_2\text{SO}_4 + 2\,\text{HI} \quad (5)$$

2.4.6. Analysis of Volatile Compounds

The volatile profile of the syrups was determined with GC/MS with headspace solid-phase micro-extraction sampling (SPME; DVB/CAR/PDMS fiber, 2 cm; Sigma Aldrich, Germany), as described in [12]. In brief, a GCMS-QP2010 Ultra (Shimadzu Inc., Kyoto, Japan) instrument was used, with a DB-Wax capillary column (30 m, 0.25 mm i.d., 0.25 μm film thickness; Agilent, Santa Clara, CA, USA), and He as carrier gas (36 cm/s). For the sampling, 2 g of sample was transferred into a 20 mL glass vial containing 4 mL water, 2 g $(NH_4)_2SO_4$, and 10 μL 1,4-dioxane solution (1000 mg/L; as IS). The vial was sealed and equilibrated for 5 min in a water bath at 40 °C. The SPME fiber was then exposed to the vial headspace for 30 min.

The volatiles were desorbed from the SPME fiber by exposing in the GC injection port (with liner; 0.7 mm i.d.; Sigma Aldrich; split ratio 1/10) at 240 °C for 5 min. The column oven temperature program was: 40 °C for 5 min; increase to 180 °C by 5 °C/min; increase to 240 °C by 30 °C/min; hold for 5 min. The MS was operated in the EI mode (70 eV) and 40–300 m/z mass scan range. The source and interface temperatures were 200 °C and 240 °C, respectively. Identification of compounds was achieved by comparing MS data and Retention Indices (RI) based on the homologous series of C8-C24 n-alkanes with those of authentic compounds and those of the NIST14 library (NIST, Gaithersburg, MD, USA) [12]. The software GCMS Solution (ver. 4.30; Shimadzu), AMDIS (ver. 2.72; NIST), and NIST MS Search (ver. 2.2; NIST) were used for the identification. The concentrations of the volatile compounds are expressed as normalized peak areas % (percentage of area corresponding to an Amdis component relative to the sum of areas of all components).

2.4.7. Microbiological Analysis

Amounts of 10 g of each syrup were received aseptically during the 1st and 3rd months of storage. The samples were homogenized with 90 mL sterile ¼ strength Ringer's solution. Each suspension was then subjected to serial decimal dilutions in Ringer's solution. Viable cell counts of total mesophilic bacteria (TMB), yeasts, molds, and enterobacteria were determined on selective media. Specifically, TMB were enumerated on Plate Count Agar (PCA) after incubation at 30 °C for 72 h. Yeasts and molds were enumerated on Potato Dextrose Agar (PDA) after incubation at 30 °C for 72 h. The presence of *Enterobacteriaceae* was determined on Violet Red Bile Glucose Agar (VRBGA) at 37 °C for 24 h. The results of the microbiological analysis were expressed as Colony-Forming Units (CFU) per g of sample, in plates containing from 30 to 300 colonies. All experiments were carried out in triplicate.

2.4.8. Sensory Evaluation

For the sensory evaluations, the syrup samples were stored at room temperature for 1 month after production and were evaluated by 10 untrained individuals who were asked to rate their sensory properties based on a 1–5 preference scale (1—Unacceptable, 2—Bad, 3—Good, 4—Very good, 5—Excellent) (consumer-oriented testing) [23]. The samples were coded randomly and were served to the testers at equal portions at room temperature (~22 °C). The testers were asked to taste each sample both plain and spread on bread (Figure 2). A sample of commercial grape syrup (petimezi) was also evaluated for comparison. The panel was specifically asked to evaluate each product in terms of aroma, taste, aftertaste, metallic aftertaste, and color. Specifically, aroma and taste were evaluated based on the 1–5 preference scale, while for aftertaste, the panelists were asked to comment based on the descriptions short, medium, and long, and the absence or presence of metallic taste. Finally, regarding color, the panelists were given the possibility of free description (description of shade, intensity, clarity, etc.).

(**a**) (**b**) (**c**)

Figure 2. (**a**) Sensory evaluation of the FSS syrups. (**b**) Brown FSS syrup. (**c**) Blonde FSS syrup.

2.4.9. Statistical Analysis

The significance of differences in the means of various data groups was checked with the One-Way Anova or t-Test (two populations), at the 0.05 level of significance, using the Microcal™ Origin® software, version 6.0 (Microcal Software, Inc., Northampton, MA, USA).

3. Results and Discussion

3.1. Composition of the Brown and Blonde Syrups Made from FSS

The results for the composition of the brown and blonde FSS syrups (sugars, acidity, protein, TPC, AC, sulfite, and HMF), are presented in Table 1.

Table 1. Composition of brown and blonde syrups made from the Corinthian currant finishing side-stream.

Parameter	Syrup Type			
	BrST	**BrS**	**BlST**	**BlS**
pH	4.35 ± 0.04 [a]	5.57 ± 0.52 [b]	4.46 ± 0.15 [a]	6.25 ± 0.18 [c]
TTA (% *w*/*w* tartaric acid)	0.88 ± 0.08 [a]	0.91 ± 0.08 [a]	1.18 ± 0.30 [b]	0.75 ± 0.16 [a]
VA (% *w*/*w* acetic acid)	0.16 ± 0.13 [a]	0.11 ± 0.04 [b]	0.27 ± 0.03 [a]	0.09 ± 0.00 [b]
Total sugar content (% *w*/*w*)	65.4 ± 0.5 [a]	69.4 ± 0.5 [b]	66.5 ± 0.6 [a]	66.3 ± 7.5 [a]
Glucose (% *w*/*w*)	32.9 ± 0.7 [a]	31.3 ± 1.5 [a]	35.4 ± 0.6 [a]	34.6 ± 4.1 [a]
Fructose (% *w*/*w*)	32.5 ± 0.2 [a]	34.5 ± 4.1 [a]	33.4 ± 3.3 [a]	30.5 ± 5.1 [a]
Sucrose (% *w*/*w*)	nd	nd	nd	nd
Total SO_2 (mg/kg)	40.7 ± 1.1 [a]	33.0 ± 0.5 [b]	22.1 ± 0.8 [c]	26.1 ± 0.1 [d]
Protein (% *w*/*w*)	1.56 ± 0.05 [a]	1.68 ± 0.16 [a]	1.52 ± 0.18 [a]	1.40 ± 0.18 [b]
TPC (mg GAE/100 g)	213.3 ± 7.7 [a]	184.9 ± 4.5 [b]	134.0 ± 2.9 [c]	143.3 ± 2.4 [d]
AC (mg AAE/g)	0.90 ± 0.09 [a]	1.03 ± 0.03 [b]	0.29 ± 0.02 [c]	0.58 ± 0.04 [d]
HMF (mg/kg)	nd	nd	nd	nd

TTA: Total titratable acidity. VA: Volatile acidity. TPC: Total phenolic content. AC: Antioxidant capacity. HMF: Hydroxymethyl furfural. GAE: Gallic acid equivalent. AAE: Ascorbic acid equivalent. BrST: Brown syrup with tartrate. BrS: Brown syrup with reduced tartrate. BlST: Blonde syrup with tartrate. BlS: Blonde syrup with reduced tartrate. nd: not detected. Superscript letters in a row indicate statistical differences between treatments ($p < 0.05$). All assays were carried out at least in triplicate (n = 3–6).

The total average (av.) sugar content of all syrups after condensation was 65.4–69.4% *w*/*w*, and their Baumé hydrometer density was 38–40 °Be. According to the Greek legislation [8], commercial raisin syrups must have a density of 40–41 °Be, corresponding to 70–71% sugar content; however, at densities higher than 40 °Be, rapid crystallization of the sugars in the samples was observed. In the syrups prepared with the proposed methods, no sugar crystals were formed during the studied storage period and beyond. All syrups had similar contents of glucose (av. 31.3–35.4 % *w*/*w*) and fructose (av. 30.5–34.5% *w*/*w*) ($p < 0.05$), while no sucrose was found in the HPLC analysis (Table 1). Sugars determine both the taste and texture of syrups, and play an important role in their preservation by inhibiting the growth of microorganisms since they are present at high levels. The absence of sucrose was expected as it was not found in the raw material (FSS), as also reported previously [12], and is generally low (av. 0.4%) in the grapes of most vinifera cultivars [24]. The drying, storage, and extraction processes of FSS possibly led to the hydrolysis of any sucrose present in the grapes or currants. Similar commercial raisin syrup products (stafidini) found in the Greek market report reducing sugar levels of 68.0% for brown syrup (CBrS) and 71% for blonde syrup (CBlS). These data were either obtained from the product labelling or the product specifications available online [25,26].

The pH of syrups depends on the composition of the raw material and the method of production. The components of FSS that are responsible for the acidity of grape syrups are mainly tartaric acid, malic acid, and other organic acids that are present in lesser quantities (citric, succinic, etc.) [12,24]. The FSS syrups that were not treated for tartrate reduction were more acidic with pH values around 4.5 (Table 1), while the treated syrups had higher pH (av. 5.6–6.3). Comparing the pH values of the produced FSS syrups with those reported for two commercial brown and blonde syrups available in the local market (3.0–4.0 for CBrS, and 3.70–4.50 for CBlS), it can be observed that the commercial products have lower pH vales (Table 1). Also, the pH values of all blonde syrups made in this study, and the commercial ones, tend to be higher than those of brown syrups. These differences may be due to several factors including the different raw material and production process, the pH determination method, the different levels of tartrate reduction, the presence of sulfite, and the removal of other acidic components by the activated carbon treatment of the blonde syrups.

As in the case of pH, the TTA is mainly due to non-volatile organic acids (tartaric and malic acid, and other organic acids to a lesser extent), which are present in raisins and are transferred to the syrup. All syrups had similar TTA (av. 0.8–0.9% *w*/*w*, as tartaric acid), except BlST syrup, which had slightly higher TTA (av. 1.2%; $p < 0.05$) (Table 1). Despite the fact that half the syrups had undergone treatment to reduce tartrate, all products finally presented similar TTA values. The TTA of commercial brown syrup (CBrS) varies between 3.5 and 6.0% *w*/*w*, which is much higher than the FSS syrups prepared in this study. This difference may be due to different production processes and tartrate precipitation levels. From the above results it can be concluded that the pH and TTA values determined in this study cannot be considered reliable (as a measure of the absolute organic acid concentration), since the syrups had undergone many treatments (tartrate precipitation by $CaCO_3$, sulfite addition and excess reduction by oxidation, etc.) that affect these values. An HPLC analysis of individual organic acids, especially tartrate, would provide more accurate results regarding their final concentration in the syrups; however, for technical reasons, it was not possible. The TTA (as tartrate) was previously found to be 2.19 ± 0.17% *w*/*w* in FSS, 2.60 ± 0.31 g/L in FSS extract (of 11.3 °Be density), and 3.99 ± 0.22 g/L or 4.32 ± 0.11 g/L in dry FSS wine made with free or immobilized yeast cells, respectively [12].

The Greek legislation for commercial raisin syrups requires reduction of tartrate [8], because tartaric and malic acids tend to precipitate at low temperatures, making the product unstable and visually unappealing to the consumer. Indeed, during the various stages of the proposed FSS syrup methodology, tartrate seems to be spontaneously destabilized and precipitate; therefore, the tartrate reduction step may be omitted, which may be advantageous in terms of both cost and process simplicity. On the other hand, it should be noted that tartrate and other organic acids are beneficial as they provide flavor and contribute to the microbial stability of the products [24].

VA is also important for the organoleptic quality, and indicative of the microbiological stability of a product. In the case of grape syrups, VA is developed mainly by the action of yeasts and acetic acid bacteria, which eventually leads to the conversion of sugars to ethanol and acetic acid during the stages of grape harvesting, raisin production, and industrial finishing, and the storage and handling of the generated FSS thereafter [12]. Other volatile acids also contribute to a lesser extent. The average VA (as acetic acid) of the FSS syrups made in this study was 0.09–0.27% *w*/*w* (Table 1), which is low, indicating that the syrups did not undergo significant microbial alterations during production that could reflect on their organoleptic quality. For example, the overnight stay of the FSS extract in the refrigerator for cold stabilization or any other delays and disturbances during the various stages of production could have affected the VA levels by increasing exposure to oxygen and facilitating spoilage by yeasts and acetic acid bacteria. Among the different samples, lower values ($p < 0.05$) were observed in the case of samples that were treated with $CaCO_3$ for tartrate reduction. The VA was previously found to be 0.20 ± 0.04% *w*/*w* in FSS, 0.17 ± 0.01 g/L in FSS extract (of 11.3 °Be), and below 0.60 ± 0.17 g/L in dry FSS wines [12]. No data on the VA of raisin syrups have been reported in the literature.

During grape ripening, inorganic nitrogen is rapidly reduced due to conversion to organic compounds (amino acids, peptides, polypeptides, proteins), and at full ripening only a very small percentage is inorganic [24]. The syrups prepared in this study had similar protein contents (av. 1.5–1.7% *w*/*w*; as Kjeldahl total nitrogen), except the sample BlS, which presented slightly lower values (av. 1.40%; $p < 0.05$), indicating that the production process did not significantly affect this characteristic (Table 1). This small decrease observed in the blonde syrups is possibly due to the application of bentonite, a common clarifying agent that carries negatively charged particles and removes pigments and turbidity by flocculating unstable proteins [27,28].

The addition of sulfite to the FSS extracts before the production of the syrups is essential to avoid microbial spoilage and spontaneous fermentation, and is a common practice in the food industry, including syrups, jams, jellies, and other fruit-based preserves [29]. Sulfite also acts as an antioxidant, protecting from oxidative degradation of the product's

sensory properties, especially color [29]. The average residual sulfite in the FSS syrups after treatment with H_2O_2 was 22–41 mg SO_2/kg (Table 1). The maximum permitted level according to Greek legislation is 40 mg/kg for raisin syrups [8], and according to EU legislation, in jams, jellies and marmalades made with sulfited fruit, it should be less than 100 mg/kg or mg/L [30]. With regard to the sulfite addition and excess oxidation applied in this study, based on the multiple laboratory tests that were carried out in order to regulate its final level in the products within the regulatory limits (<40 ppm), it was concluded that these processes are laborious and have small reproducibility. Taking into account that sulfites are associated with undesirable health effects (such as allergies) [29], the application of other techniques for microbial load reduction should be considered in syrup production (e.g., warm extraction of FSS, pasteurization of the extracts, etc.). Moreover, the final products are stable due to their high sugar content.

HMF was not detected in any of the produced syrups. This makes sense as the syrups were prepared by concentration at low temperature (45 °C). HMF in food is one of the many compounds resulting from heating due to the acid-catalyzed dehydration of sugars such as hexoses, and as an intermediate of the Maillard reaction. HMF can produce other compounds with potential carcinogenicity, such as 5-sulfoxymethyl-2-furfural; however, for HMF this has not been documented in vivo [31]. Another process that could lead to the presence of HMF is the drying of raisins under the sun, and in the case of FSS, this natural drying process does not seem to produce HMF.

Regarding the TPC (GAE equivalent) of the syrups, variation can be observed among the samples ($p < 0.05$). The blonde syrups had lower contents (av. 134–143 mg/100 g) than brown syrups (av. 185–213 mg/100 g) (Table 1). This is possibly due to the removal of pigments from the blonde syrups, including antioxidant phenolics, by the activated carbon treatment [12,32]. Data on the TPC of similar commercial syrups were not found. For grape juice concentrate, TPC values (GAE; dry mass) of 252 ± 33 mg/100 g (from grapes with initial TPC 1619 ± 167 mg/100 g) [33], and 245.6 ± 4.3 mg/100 g [34], have been reported. In traditional Turkish pekmez, a TPC of 335.9 ± 7.2 mg/100 g has been reported [35]. Finally, in grape molasses produced using traditional and industrial methods, the TPC (GAE; dry mass) varied from 1.70 ± 0.13 to 8.31 ± 0.11 mg/g, according to [36].

In [12], the TPC of FSS (same origin and batch as the FSS used in this study) was found to be 476 ± 0.3 mg GAE/100 g, while that of aqueous FSS extract (11.3 °Be) was 107.0 ± 0.02 mg GAE/L. The significant reduction in TPC in the syrups compared to the raw material (FSS) is due to the production process, and specifically to the removal of the solid FSS residues (skins, seeds, and stems) and the clarification and depigmentation steps [12,33], as well as to possible chemical effects of these processes on the FSS phenolics.

For the same possible reasons, the brown FSS syrups presented higher AC (av. 0.90–1.0 mg AAE/g) compared to the blonde syrups (av. 0.3–0.6 mg AAE/g) (Table 1). The same was observed by [33], who determined an 83–92% decrease in AC (as μmol TEAC/100 g, dry mass) from the grape (6910 ± 421) to the concentrate (527 ± 28). According to [36], the AC of grape molasses varied from 68.2 ± 1.7 to 96.6 ± 2.2 mg AAE/ g of dry extract, while according to [34], the AC of grape juice concentrate, which was screened by the DPPH· radical scavenging assay, was 488.6 ± 19.6 μmol Trolox eq./100 g dry mass. In [12], the AC of the FSS was determined to be 2.4 ± 0.04 mg AAE/g and that of aqueous FSS extract (11.3 °Be) was 10.3 ± 0.1 mg AAE/L.

3.2. Volatilome

The volatile profile of the syrups was determined using GC/MS with headspace SPME sampling. In total, 144 compounds were detected, and their concentrations are presented in Table 2 as normalized peak areas %. Specifically, 15 esters (mainly fatty acid ethyl esters, acetate esters, and phenyl esters), 28 alcohols (including phenyl alcohols and a furanyl alcohol), 12 organic acids (straight-chain C2-C10 and 2/3-methylcarboxylic acids), 41 carbonyl compounds (aldehydes, ketones, and furanyl-, phenyl-, and pyrrolyl-aldehydes), 18 terpenes (mainly oxygenated monoterpenes, sesquiterpenes, norisoprenoids), seven lactones, eight hy-

drocarbons (alkanes and straight-chain alkenes, C6-C16), and 15 other compounds (mainly furans and pyrazines) were identified. Odor and taste descriptions for these compounds, as well as their prior identification in grapes, raisins, other parts of the vine plant, or other plant sources, were previously presented in detail in [10,12].

Table 2. Volatiles identified by SPME GC/MS analysis (normalized peak areas %) in the brown (BRST) and blonde (BLST) syrups.

Compound	CAS	RID	RI_{ref}	RI	FSS	BrST	BlST
Esters							
Methyl acetate	79-20-9	A	828	820	0.20	0.19	<0.01
Ethyl acetate	141-78-6	A	888	882	5.29	4.38	9.68
Ethyl 2-methylpropanoate (ethyl isobutyrate)	97-62-1	A	961	958	<0.01	<0.01	<0.01
2-Methylpropyl acetate (isobutyl acetate)	110-19-0	A	1012	1012	<0.01	0.03	0.10
Ethyl butanoate (ethyl butyrate)	105-54-4	A	1035	1034	<0.01	0.02	0.03
Ethyl 3-methylbutanoate (ethyl isovalerate)	108-64-5	B	1068	1066	<0.01	0.01	0.03
3-Methylbutyl acetate (isoamyl acetate)	123-92-2	A	1122	1118	0.05	0.17	1.19
Ethyl hexanoate (ethyl caproate)	123-66-0	A	1233	1229	0.07	0.02	0.02
Ethyl 2-hydroxypropanoate (ethyl lactate)	97-64-3	A	1347	1344	0.02	<0.01	<0.01
Ethyl octanoate (ethyl caprylate)	106-32-1	A	1435	1435	0.70	0.06	<0.01
Ethyl decanoate (ethyl caprate)	110-38-3	A	1638	1640	0.33	<0.01	<0.01
Ethyl 2-phenylacetate (ethyl benzeneacetate)	101-97-3	C	1783	1781	<0.01	0.01	<0.01
2-Phenylethyl acetate	103-45-7	A	1813	1810	0.38	0.44	0.29
2-Phenylethyl butanoate (phenethyl butyrate)	103-52-6	B	1958	1965	0.02	<0.01	<0.01
Octyl octanoate	2306-88-9	B	2009	2014	<0.01	0.03	0.11
Total					<7.12	<5.40	<11.52
Alcohols							
Ethanol	64-17-5	A	932	931	6.54	0.15	0.27
2-Methyl-1-propanol (isobutanol)	78-83-1	A	1092	1098	0.23	<0.01	0.02
1-Butanol	71-36-3	A	1142	1151	0.12	0.07	0.05
1-Penten-3-ol (ethyl vinyl carbinol)	616-25-1	B	1159	1166	0.24	<0.01	0.01
3-Methyl-1-butanol (isoamyl alcohol)	123-51-3	A	1209	1211	1.57	0.65	0.32
1-Pentanol	71-41-0	A	1250	1256	0.59	0.43	0.52
(Z)-2-Penten-1-ol	1576-95-0	B	1318	1324	0.08	<0.01	<0.01
3-Methyl-2-buten-1-ol (prenol)	556-82-1	C	1320	1324	0.05	0.03	0.02
1-Hexanol	111-27-3	A	1355	1357	1.31	0.75	0.28
(E)-3-Hexen-1-ol	928-97-2	B	1367	1370	<0.01	0.02	<0.01
(Z)-3-Hexen-1-ol	928-96-1	B	1382	1387	0.09	<0.01	<0.01
(E)-2-Hexen-1-ol	928-95-0	B	1405	1409	0.09	0.13	<0.01
1-Octen-3-ol	3391-86-4	A	1450	1454	1.99	0.11	0.05
1-Heptanol	111-70-6	B	1453	1460	0.14	0.11	<0.01
2-Ethyl-1-hexanol	104-76-7	A	1491	1493	0.61	4.08	0.34
(E)-2-Hepten-1-ol	33467-76-4	C	1517	1514	0.05	0.01	<0.01
2,3-Butanediol isomer 1	513-85-9	C	1543	1544	0.49	0.30	0.49
1-Octanol	111-87-5	A	1557	1562	1.07	0.45	0.28
2,3-Butanediol isomer 2	24347-58-8	C	1556	1581	0.53	0.26	0.51
(E)-2-Octen-1-ol	18409-17-1	C	1614	1617	0.16	0.01	<0.01
2-Furanmethanol (Furfuryl alcohol)	98-00-0	B	1660	1661	0.11	1.28	0.33
1-Nonanol	143-08-8	B	1660	1665	0.68	0.03	0.10
5-Methylfurfuryl alcohol	3857-25-8	C	1714	1725	<0.01	0.42	0.02
3-(Methylthio)-1-propanol (methionol)	505-10-2	B	1719	1721	<0.01	0.01	0.08
2-Dodecanol	10203-28-8	C	1813	1822	0.14	0.02	0.04
Phenylmethanol (benzyl alcohol)	100-51-6	B	1870	1875	0.53	0.24	0.07
2-Phenylethanol (phenylethyl alcohol)	60-12-8	A	1906	1912	0.63	0.28	0.16
1-Tetradecanol (myristyl alcohol)	112-72-1	C	2165	2181	0.41	0.06	0.15
Total					<18.45	<9.90	<4.11

Table 2. *Cont.*

Compound	CAS	RID	RI_{ref}	RI	FSS	BrST	BlST
Organic acids							
Acetic acid	64-19-7	A	1449	1445	10.75	3.90	4.35
Formic acid	64-18-6	B	1503	1506	0.05	0.05	0.10
Propanoic acid	79-09-4	B	1535	1538	0.08	0.11	0.12
Butanoic acid	107-92-6	B	1625	1628	0.01	0.01	0.01
3-Methylbutanoic acid (isovaleric acid)	503-74-2	B	1666	1670	0.14	0.12	0.03
2-Methylbutanoic acid	116-53-0	C	1662	1671	0.05	<0.01	<0.01
Pentanoic acid (valeric acid)	109-52-4	B	1733	1737	0.12	0.04	0.02
Hexanoic acid (caproic acid)	142-62-1	A	1846	1844	1.14	0.26	0.14
3-Methylhexanoic acid	3780-58-3	C	-	1954	0.16	0.03	0.01
Octanoic acid (caprylic acid)	124-07-2	A	2060	2063	8.48	<0.01	<0.01
Nonanoic acid	112-05-0	C	2171	2174	0.23	0.14	0.04
n-Decanoic acid (capric acid)	334-48-5	B	2276	2251	0.89	0.11	0.11
Total					22.10	<4.77	<4.93
Carbonyl compounds							
Acetaldehyde	75-07-0	A	702	701	<0.01	0.29	0.53
2-Methylpropanal (isobutyraldehyde)	78-84-2	B	819	13	0.14	3.39	0.60
Butanal (butyraldehyde)	123-72-8	B	877	867	0.03	0.02	0.03
2-Butanone (methyl ethyl ketone)	78-93-3	B	907	899	0.04	0.23	0.35
2-Methylbutanal	96-17-3	B	914	908	0.59	8.73	2.63
3-Methylbutanal (isovaleraldehyde)	590-86-3	B	918	911	1.64	12.09	8.66
2,3-Butanedione (Diacetyl)	431-03-8	A	979	970	5.91	0.92	5.43
Hexanal	66-25-1	A	1083	1076	3.32	0.78	2.05
2-Methyl-2-butenal	1115-11-3	B	1095	1089	<0.01	0.05	<0.01
2-Heptanone	110-43-0	B	1182	1178	0.05	0.07	0.02
Heptanal (oenanthic aldehyde)	111-71-7	B	1184	1179	0.17	<0.01	0.09
2-Methyltetrahydrofuran-3-one (coffee furanone)	3188-00-9	B	1268	1262	<0.01	1.47	0.46
3-Hydroxy-2-butanone (acetoin)	513-86-0	A	1284	1281	21.52	2.29	3.95
Octanal	124-13-0	B	1289	1284	0.21	0.15	0.07
1-Hydroxy-2-propanone (hydroxyacetone)	116-09-6	B	1303	1293	<0.01	0.27	0.15
2-Heptenal	18829-55-5	B	1323	1319	0.21	0.01	<0.01
6-Methyl-5-hepten-2-one	110-93-0	C	1338	1335	0.09	0.03	0.01
2-Acetoxy-3-butanone (acetoin acetate)	4906-24-5	C	1378	1381	0.10	0.01	<0.01
1-Hydroxybutan-2-one	5077-67-8	B	1388	1376	0.02	0.05	0.08
2-Nonanone	821-55-6	C	1390	1387	<0.01	<0.01	<0.01
Nonanal	124-19-6	B	1391	1391	0.40	0.36	0.18
5-Ethyl-1-formylcyclopentene (Phoracanthal)	36431-60-4	C	1410	1411	0.40	<0.01	<0.01
3-Octen-2-one	1669-44-9	C	1411	1405	0.31	0.01	<0.01
(E)-2-Octenal	2548-87-0	C	1429	1426	0.16	<0.01	<0.01
2-Furfuraldehyde (furfural)	98-01-1	A	1461	1459	3.36	24.21	38.24
Phenylmethanal (benzaldehyde)	100-52-7	A	1520	1517	0.96	1.09	<0.01
(E)-2-Nonenal	18829-56-6	C	1534	1533	0.07	<0.01	<0.01
1-(2-Furyl)-1-propanone	3194-15-8	B	1563	1576	<0.01	0.11	0.09
(3E,5E)-3,5-Octadien-2-one	30086-02-3	C	1570	1569	0.12	<0.01	<0.01
5-Methyl-2-furfural	620-02-0	B	1570	1572	0.25	10.67	6.03
1-(Furan-2-yl)butan-2-one	4208-63-3	C	1584	1598	<0.01	0.07	0.05
6-Methyl-3,5-heptadiene-2-one	1604-28-0	B	1602	1591	0.27	<0.01	0.01
Ethyl-1H-pyrrole-2-carboxaldehyde	2167-14-8	C	1610	1605	0.14	3.30	2.66
1-Methylpyrrole-2-carboxaldehyde	1192-58-1	C	1626	1620	<0.01	0.20	0.20
Phenylacetaldehyde	122-78-1	C	1640	1636	0.27	0.19	0.16
Acetophenone	98-86-2	B	1647	1646	<0.01	<0.01	0.10
2,4-Nonadienal	6750-03-4	C	1700	1700	0.07	<0.01	<0.01
2-Acetylpyrrole	1072-83-9	B	1973	1971	0.03	0.73	0.19
2-Hydroxyacetylfuran	17678-19-2	C	1995	2003	0.15	0.14	0.27
2,4-Decadienal	2363-88-4	B	1797	1805	0.10	<0.01	<0.01
1H-Pyrrole-2-carboxaldehyde (pyrrole aldehyde)	1003-29-8	B	2030	2023	0.01	0.11	0.11
Total					<41.11	<71.75	<72.87

Table 2. *Cont.*

Compound	CAS	RID	RI_{ref}	RI	FSS	BrST	BlST
Terpenes							
a-Pinene (2,6,6-Trimethylbicyclo [3.1.1]hept-2-ene)	80-56-8	C	1028	1018	<0.01	0.02	0.03
D-Limonene (1-methyl-4-prop-1-en-2-ylcyclohexene)	5989-27-5	A	1200	1185	0.39	0.15	0.10
Eucalyptol (1,3,3-trimethyl-2-oxabicyclo[2.2.2]octane)	470-82-6	B	1213	1200	<0.01	<0.01	<0.01
p-Cymene (1-Isopropyl-4-methylbenzene)	99-87-6	B	1272	1265	<0.01	0.01	0.02
cis-Linalool oxide	5989-33-3	B	1444	1447	<0.01	0.23	0.12
trans-Linalool oxide	34995-77-2	C	1452	1477	<0.01	0.24	<0.01
Linalool (3,7-dimethyl-1,6-octadien-3-ol)	78-70-6	A	1547	1551	0.08	0.06	0.02
Fenchol (1,3,3-trimethylbicyclo[2.2.1]heptan-2-ol)	1632-73-1	B	1582	1588	<0.01	0.03	0.03
L-4-Terpineol (4-methyl-1-propan-2-ylcyclohex-3-en-1-ol)	20126-76-5	B	1593	1606	<0.01	0.06	0.03
β-Cyclocitral (2,6,6-trimethylcyclohexene-1-carbaldehyde)	432-25-7	C	1611	1618	0.08	<0.01	<0.01
(-)-Menthol (5-methyl-2-(1-methylethyl)-cyclohexanol)	2216-51-5	C	1633	1647	<0.01	0.08	0.17
4-Ketoisophorone (2,6,6-trimethyl-2-cyclohexene-1,4-dione)	1125-21-9	C	1676	1690	0.04	0.09	0.05
α-Terpineol [2-(4-methyl-3-cyclohexen-1-yl)-2-propanol]	98-55-5	A	1697	1700	0.07	0.44	0.19
L-Borneol (1,7,7-trimethyl-bicyclo[2.2.1]heptan-2-ol)	507-70-0	B	1702	1706	<0.01	0.03	0.03
β-Damascenone	23726-93-4	C	1823	1818	0.06	0.11	0.14
p-Cymen-8-ol (2-(4-methylphenyl)propan-2-ol)	1197-01-9	C	1852	1851	<0.01	0.14	0.05
trans-Geranylacetone [(E)-6,10-dimethylundeca-5,9-dien-2-one]	3796-70-1	C	1859	1853,9	0.09	0.06	0.10
trans-β-Ionone [(E)-4-(2,6,6-trimethyl-1-cyclohexen-1-yl)-3-buten-2-one]	79-77-6	C	1940	1941	0.08	0.01	0.01
Total					<0.89	<1.74	<1.06
Lactones							
γ-Butyrolactone (dihydrofuran-2(3H)-one)	96-48-0	B	1632	1621	1.92	1.10	1.80
β-Angelica lactone (5-methyl-2(5H)-furanone)	591-11-7	C	1669	1674	0.03	0.01	0.02
γ-Hexalactone (dihydro-5-ethyl-2(3H)-furanone)	695-06-7	C	1694	1698	0.04	0.03	0.01
γ-Crotonolactone (2(5H)-Furanone)	497-23-4	B	1742	1750	<0.01	0.04	0.01
2-Hexen-1,4-lactone (5-ethyl-2(5H)-furanone)	2407-43-4	C	1745	1753	0.06	0.01	<0.01
γ-Octalactone (5-Butyldihydrofuran-2(3H)-one)	104-50-7	C	1910	1915	0.01	0.02	0.01
γ-Nonalactone (dihydro-5-pentyl-2(3H)-furanone)	104-61-0	C	2024	2028	0.06	0.03	0.01
Total					<2.12	1.24	<1.86
Other compounds							
Dimethyl sulfide (DMS)	75-18-3	B	754	739	0.01	0.60	0.78
Furan		B	799	793	<0.01	0.05	0.01
2-Methylfuran	534-22-5	B	869	861	<0.01	0.03	0.01
2-Ethylfuran	3208-16-0	B	950	943	0.04	0.04	0.02
1,4-Dioxane (IS)	123-91-1			1055.5			
2-Pentylfuran	3777-69-3	B	1231	1220	0.22	0.08	0.02
2,6-Dimethylpyrazine	108-50-9	B	1328	1327	<0.01	0.03	<0.01
2-Ethyl-6-methylpyrazine	13925-03-6	B	1386	1385	0.04	0.04	0.06
2,3,5-Trimethylpyrazine	14667-55-1	B	1402	1404	<0.01	0.03	0.03
2,6-Diethylpyrazine	13067-27-1	C	1444	1437	<0.01	0.02	<0.01
2,3-Dimethyl-5-ethylpyrazine	15707-34-3	C	1460	1463	0.08	0.03	0.05
2,3,5,6-Tetramethylpyrazine (Ligustrazine)	1124-11-4	B	1469	1477	0.03	0.04	0.01
2-Acetylfuran (2-furyl methyl ketone)	1192-62-7	B	1499	1500	0.16	3.44	1.75
2-Acetyl-5-methylfuran	1193-79-9	C	1606	1611	<0.01	0.03	<0.01
Guaiacol (2-Methoxyphenol)	90-05-1	B	1861	1856	<0.01	0.03	<0.01
Benzothiazole	95-16-9	C	1958	1953	<0.01	0.02	<0.01
Total					<0.57	3.91	<1.96

Table 2. *Cont.*

Compound	CAS	RID	RI_{ref}	RI	FSS	BrST	BlST
Hydrocarbons (alkanes/alkenes)							
Hexane	110-54-3	A	600	600	5.91	0.05	0.14
Heptane	142-82-5	A	700	700	0.35	<0.01	<0.01
Octane	111-65-9	A	800	800	0.33	0.02	0.01
Nonane	111-84-2	A	900	900	0.07	<0.01	<0.01
Decane	124-18-5	A	1000	999	0.10	<0.01	<0.01
Dodecane	112-40-3	A	1200	1200	0.32	<0.01	<0.01
Tetradecane	629-59-4	A	1400	1400	0.46	0.01	0.02
Hexadecane	544-76-3	A	1600	1600	0.06	0.03	0.01
Total					7.6	<0.11	<0.18

CAS: Chemical Abstracts Service registry number. RI: Retention Index. RI_{ref}: The reference RIs were obtained from the NIST14 library, where they are displayed as the experimental RI median value taken from various sources in the literature. RID: Reliability of identification. RID levels: A, agreement of RI and MS spectra with those of an authentic compound analyzed under identical conditions. B, agreement of RI (ΔRI < 20) and MS (match > 900). C, at least ΔRI < 20 or MS similarity match > 800. FSS: Corinthian currant finishing side-stream. BrST: Brown syrup with tartrate. BlST: Blonde syrup with tartrate.

Among esters, ethyl acetate was found in significant amounts in the syrups' volatilome (above 4%), as well as in the raw material (FSS). Other esters found at levels above 0.1% of total volatiles were isoamyl acetate (fruity, banana) and 2-phenylethyl acetate (floral, rose, honey) [12]. All identified esters were also found in the FSS, but in total, the BlST syrup presented a richer ester profile (Table 2).

The main alcohol found in the syrups (4.1% in BrST) was 2-ethyl-1-hexanol (citrus, fresh, flowery) (Table 2), which has been previously identified in raisins, grapes, and wines [12]. Furfuryl alcohol, 3-methyl-1-butanol, 1-pentanol, and 1-hexanol were found at levels of 0.3–1%. Of the 28 alcohols detected, 21 have been previously found in grapes or raisins [12]. The alcohols (Z)-2-penten-1-ol, 2-dodecanol, 5-methylfurfuryl alcohol, and methionol have only been previously reported in wine [12].

Of the 12 identified organic acids, nine were also previously reported in grapes or raisins [12]. Acetic acid was the major acid identified in the headspace of both the FSS (10.8%) and the FSS syrups (3.9–4.4%) (Table 2). All other acids, except 2-methylbutanoic acid and caprylic acid (below 0.01%), were found at levels 0.01–0.3%.

Carbonyl compounds (in total, 41) were found at higher levels in the syrups than in the FSS (Table 2). The main compounds in the syrups were 2-methylbutanal (3–9%) (rummy, malty, nutty, fermented), 3-methylbutanal (9–12%) (aldehydic, cocoa, fatty, fruity, nutty), acetoin (2–4%) (fatty, creamy, milky), furfural (24–38%) (woody, bready, nutty, caramellic, burnt), 5-methyl-2-furfural (6–11%) (spicy, caramellic, maple, grain), and ethyl-1H-pyrrole-2-carboxaldehyde (3%) (burnt, roasted, smoky) [12]. Isobutyraldehyde, coffee furanone, and benzaldehyde were found at higher levels in BrST, while diacetyl and hexanal were found at higher levels in BlST. Most of the identified compounds are either produced during the dehydration of the raisins as lipid oxidation products or as products of the Maillard reaction [12,37,38].

Terpenes are also important aroma compounds derived mainly from grapes and are key components of varietal aromas. The major terpenes found in the syrups (at levels above 0.1%) were D-limonene, cis- and trans-linalool oxide, α-terpineol, β-damascenone, l-menthol, and D-cymen-8-ol (Table 2). α-Terpineol (pine, woody, lemon, floral) [12] was the major terpene identified at levels 0.2–0.4%. In total, higher levels of terpenes were found in the BrST syrup.

A number of other heterocyclic and aromatic compounds (furan, alkyl furans, alkyl pyrazines, acetyl furans, guaiacol, benzothiazole), were identified in both FSS and in the syrups (Table 2). These compounds are associated with the Maillard and Strecker degradation reactions commonly found in raisins and providing typical aroma descriptions (roasted, nutty, green, floral, fruity, caramellic) [10,12,24,37,38]. The major furan found was 2-acetylfuran (3.5–1.8%). Dimethyl sulfide (asparagus, truffle, molasses) was also identified,

which may be a result of microbial action or may be produced by heat [12]. In total, BrST contained higher amounts of these compounds compared to BlST, and much higher than those identified in the FSS, indicating a correlation with the syrup production process, which involves heating.

Finally, seven lactones (five γ-lactones, β-angelic lactone, and 2-hexene-1,4-lactone), and eight alkanes (C6-C16) were identified in the syrups (Table 2). γ-Butyrolactone (creamy, caramel, milky, fruity, peach) [12] was the major lactone found at levels of 1.1–1.8%. The C6, C8, C14, and C16 alkanes were found at levels above 0.01%, while of the eight identified alkanes, only five have been previously reported in grapes or raisins according to [12].

3.3. Microbial Stability

The microbial load of a food product is the result of the microflora present in the raw material and the microorganisms that are introduced during the stages of processing and storage until consumption. The produced FSS syrups were stored in sealed containers and kept in the dark at room temperature for 3 months. Microbial analysis for TMB, yeasts, molds, and enterobacteria was performed after the first and third months of storage, and the results are presented in Table 3.

Table 3. Microbiological load (cfu/g) of the brown and blonde syrups made from the FSS, and comparison with similar commercial products.

Microbial Group	Month	Syrup Type			
		BrST	**BrS**	**BlST**	**BlS**
TMB	1	$<10^2$	$<10^2$	$12 \times 10^3 \pm 3 \times 10^3$	$<10^2$
	3	$<10^2$	$<10^2$	$7 \times 10^3 \pm 3 \times 10^3$	$<10^2$
Yeasts	1	$<10^2$	$<10^2$	$9 \times 10^3 \pm 2 \times 10^3$	$<10^2$
	3	$<10^2$	$<10^2$	$10^4 \pm 3 \times 10^3$	$<10^2$
Molds	1	$<10^2$	$<10^2$	$<10^2$	$<10^2$
	3	$<10^2$	$<10^2$	$<10^2$	$<10^2$
Enterobacteria	1	$<10^2$	$<10^2$	$<10^2$	$<10^2$
	3	$<10^2$	$<10^2$	$<10^2$	$<10^2$

TMB: Total mesophilic bacteria. BrST: Brown syrup with tartrate. BRS: Brown syrup with reduced tartrate. BlST: Blonde syrup with tartrate. BlS: Blonde syrup with reduced tartrate. EC: *E. coli*. C: Coliforms.

A very low microbial load can be observed for all syrups, in some cases lower than that of similar commercial products. An exception was BlST syrup, which presented elevated levels of TMB and yeasts, possibly due to contamination at some stage of the production process or storage.

The lower microbial stability of a syrup may also be due to its different composition, i.e., the different levels of sugars, organic acids, antioxidants, and antimicrobial components that may have been removed by the applied clarification treatments.

3.4. Sensory Properties

For the sensory evaluation of the syrups, a consumer preference test was performed (Figure 2). The results for descriptions such as aroma, taste, aftertaste, metallic aftertaste, and color are presented in Table 4. A commercial grape syrup (CGS; petimezi) was also evaluated for comparison, and reported data for other commercial raisin syrups (CBrS, CBlS) are also presented in Table 4.

The taste of BrST, BrS, and BlST syrups was characterized as good, and even better for BlS, which received the highest score. On the other hand, the CGS received the lowest score. The taste was described as sweet and sour (BlST) or similar to honey (BrS and BrS), while only for BrST was a characteristic fruit taste pointed out. Regarding the aftertaste perception, it was described as long for all tested syrups except BrS, which was described as having a moderate aftertaste. A metallic aftertaste was not perceived in any of the samples. No data regarding the aftertaste of commercial syrups were found. In terms of aroma, the syrups BrST, BrS, and BlST were rated as good to excellent, while BlS and

CGS received lower ratings. However, for all products the aroma was characterized as not particularly intense.

Table 4. Sensory evaluation of the FSS syrups and typical Greek commercial raisin/grape syrups.

Syrup	Taste	Aftertaste	Metallic Aftertaste	Aroma	Color	Clarity
BrST	3.3 ± 0.8; raisin, typical	Long	No	3.9 ± 0.8; Raisin	Brown, honey-like	Slightly cloudy
BrS	3.8 ± 0.8; honey-like	Medium	No	3.8 ± 1.4; Raisin	Brown-red, honey-like	Clear
BlST	3.2 ± 1.3; sweet-sour	Long	No	3.6 ± 0.4; Raisin	Golden yellow	Clear
BlS	4.3 ± 1.0; honey-like	Long	No	2.9 ± 1.1; Raisin	Orange-yellow, amber	Cloudy
CGS	2.8 ± 0.9	Long	No	3.0 ± 1.0; Raisin	Dark brown	Cloudy

Preference scale 1–5: 1—Unacceptable, 2—Bad, 3—Good, 4—Very good, 5—Excellent. BrST: Brown syrup with tartrate. BrS: Brown syrup with reduced tartrate. BlST: Blonde syrup with tartrate. BlS: Blonde syrup with reduced tartrate. CGS: Commercial grape syrup (petimezi).

Finally, regarding the color and clarity of the syrups, for BrST and BrS it was described as brownish-red, similar to honey, with BrST being described as cloudy and BrS as clear. BlST was characterized as golden yellow and clear, while BlS was characterized as orange-yellow and cloudy. The colors of commercial syrups were described as dark brown for brown raisin syrups and golden brown for blonde raisin syrups. To conclude, the FSS syrups were generally described as products with a strong aftertaste, a good but not particularly strong smell, a good sweet-sour or honey-like taste, and no metallic aftertaste.

4. Conclusions

From the FSS of premium-quality Corinthian currants (Vostitsa PDO), methods for producing brown and blonde syrups are proposed and compared, including must extraction by maceration at 70 °C, tartrate reduction and depigmentation treatments, and condensation at low temperature (45 °C) under vacuum. All syrups had similar sugar contents and no sucrose. Those not treated for tartrate reduction were more acidic in terms of pH, but all had higher pH compared to similar commercial products. On the other hand, all syrups had similar TTA; therefore, the tartrate reduction step could be omitted, which may be advantageous in terms of both cost and process simplicity. The differences in protein, TPC, and AC levels among the FSS syrups and similar commercial products are possibly due to different raw materials and production processes. Sulfite addition and subsequent oxidation processes, applied to the FSS extracts to avoid spoilage, are laborious, and alternative methods to ensure the microbial stability of the syrups should be contemplated in the case of small-scale production. The syrups' volatilome, compared to the raw material (FSS), presented increased levels of compounds that are associated with sugar- and lipid-decomposition reactions. Finally, from the VA, sensory, and microbiological analyses, it can be concluded that the FSS syrups are products of good quality, and during their preparation and storage no significant microbial or chemical alterations occur. They can be considered products of high sensory as well as nutritional quality, taking into account their antioxidant properties and lower glycemic index since half their sugar is fructose. Therefore, syrups from side-streams such as FSS can create an added-value significance for the sustainability of the raisin-processing sector. The production sustainability of local commodities such as the highly nutritious Corinthian currants will also contribute to the promotion and adherence of Mediterranean dietary patterns, positively affecting local biodiversity, agricultural communities, processing companies, and the national, regional, and local economies.

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