

Review **An Overview on Management and Valorisation of Winery Wastes**

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Abstract: As we address important societal needs, the circular economy equips us with the means to jointly combat climate change and biodiversity loss, including the revaluation of waste. The wine-making process is a huge generator of waste, creating problems for manufacturers every year; therefore, an appropriate management and valorisation of winery wastes are crucial, even if it is difficult. This results from the hardship of disposing of grape marc, which is considered a pollutant for the environment. In the past, the simplest option for this waste disposal was the payment of a fee around EUR 3000, which recently increased up to EUR 30,000–40,000. Several environmentally friendly technologies have been proposed for the recovery of cellar waste. Fermentation of grape residue, pruning, or wine-making lees have been reported to yield lactic acid, surfactants, xylitol, ethanol, and other compounds. In addition, grape pulp and seeds are rich in phenolic compounds, which have antioxidant properties, and tartaric acid from vinasse can be extracted and marketed. Additionally, complex phenol mixtures, such as those found in wine residues (seeds, bark, stems, or leaves), are effective as chemotherapeutic agents and can be used in medicine. In this review, the potential of using wine-making by-products, extracts, and their constituent parts as raw materials for adsorbents, biopolymers, natural reinforcing fillers, and sustainable energy production will be a key point of discussion. An overview on how wine producers, based on wine and wastes chemistry, can implement the circular economy as an alternative to the conventional linear economy (make, use, dispose) will be provided.

Keywords: circular economy; grapes; sustainable processing; valorisation; vine; winery waste

1. Introduction

The circular economy presents a compelling argument for how a new economy should use and produce goods. The objective is to achieve "zero waste" by maintaining a constant flow of supplies between producers and consumers. Since this system is supposed to have low environmental impact and high economic activity [\[1\]](#page-16-0), it addresses global issues related to climate change, pollution, waste, and biodiversity [\[2\]](#page-16-1).

Food waste has negative impacts on the environment and the economy, and its reduction is an urgent necessity. Some methods that can be used to accomplish this goal are represented by dehydration, green extraction, and microwave- or ultrasound-assisted extraction, ensuring food safety and recovering bio-compounds from by-products [\[3](#page-16-2)[–5\]](#page-16-3). The production of wine is one of the industries that must support sustainable development. Given its significance to society and the economy, it is imperative that actions be taken in accordance with the circular economy's assertions regarding the proper use of resources and the restoration of the natural order.

Compared with other domains, the wine industry is considered to have a lower environmental influence [\[6](#page-17-0)[,7\]](#page-17-1). Although various studies on sustainability and environmental impact in winemaking technologies have been developed in recent years, one can observe that this was achieved in a disconnected way [\[6\]](#page-17-0). This observation served as an impetus for this overview, which aims to highlight whether the application of the circular economy to winemaking technologies will have positive social, economic, and environmental effects.

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Grapes represents one of the most important fruit crops that are grown on Earth. In Grapes represents one of the most important fruit crops that are grown on Earth. In 2020/2021 grape production was estimated at about 24.52 million metric tons [\[8\]](#page-17-2). Wine-2020/2021 grape production was estimated at about 24.52 million metric tons [8]. Winemaking also produces a high amount of waste (Figure [1\)](#page-1-0), which must be addressed. This making also produces a high amount of waste (Figure 1), which must be addressed. This waste is rich in biodegradable compounds and suspended solids [\[9\]](#page-17-3). Briefly, the resulting waste is rich in biodegradable compounds and suspended solids [9]. Briefly, the resulting residues include plants remains from the de-stemmed grapes, bagasse from pressing, or sediments after clarification. The wastewater resulting from the vinification lees has grape sediments after clarification. The wastewater resulting from the vinification lees has grape pulp, seeds, skins, or dead yeast used in the alcoholic fermentation. pulp, seeds, skins, or dead yeast used in the alcoholic fermentation.

economy to winemaking technologies will have positive social, economic, and environ-

Figure 1. Vinification process diagram. **Figure 1.** Vinification process diagram.

In order to increase the overall efficacity and to minimise the environmental impact In order to increase the overall efficacity and to minimise the environmental impact of the winemaking technologies, various measures have been established, allowing the of the winemaking technologies, various measures have been established, allowing the minimization, management, and efficient recuperation of waste streams in accordance minimization, management, and efficient recuperation of waste streams in accordance with the circular economy perspective $[10]$. This approach allows and integrates each element from the production chain and increases the awareness, which is essential to target a real a real shift towards sustainability, with efficient resource usage and valorisation of by-shift towards sustainability, with efficient resource usage and valorisation of by-products and wastes $[10]$.

In this respect, the wine-making industry is urging recycling options to benefit from In this respect, the wine-making industry is urging recycling options to benefit from the wastes through their use as raw materials for value-added products. the wastes through their use as raw materials for value-added products.

The most important (in terms of abundance and composition) by-product of the The most important (in terms of abundance and composition) by-product of the wine-making process is the grape pomace, which constitutes a major source of bioactive wine-making process is the grape pomace, which constitutes a major source of bioactive compounds, such as dietary fibres, lipids, minerals polyphenols, and proteins [11,12]. It compounds, such as dietary fibres, lipids, minerals polyphenols, and proteins [\[11](#page-17-5)[,12\]](#page-17-6). It can be used as a substitute for obtaining various products, as Figur[e 2](#page-2-0) depicts. can be used as a substitute for obtaining various products, as Figure 2 depicts.

The by-products resulting from winemaking can be applied for various purposes. Thus, grape pomace and seeds can be used to obtain anthocyanin colourants, oils, or catechin polymers [\[13\]](#page-17-7). Moreover, the lees have been used as a supplement in animal feed; however, the yeast content possesses poor nutrient value, and were deemed unsuitable for this purpose [\[13\]](#page-17-7).

The use of grape pomace is a perfect example of a circular economy. It is estimated that more than 79 million tons of grapes are used annually, with about 30% being represented by grape pomace [\[14\]](#page-17-8). It can be used as animal feed and for pharmaceutical purposes, due to its high content of bioactive compounds, mainly phenolics [\[14\]](#page-17-8), which have antioxidant and anti-inflammatory effects [\[15\]](#page-17-9).

Grape marc also contains high quantities of soluble sugars useful for ethanol fermentation, resulting a beverage known as "grape spirit" [\[16\]](#page-17-10). Furthermore, the fermentation of residual sugars can increase its economic value by producing industrial ethanol used in the cosmetic and pharmaceutical industries [\[17\]](#page-17-11). Grape pomace also has the potential to become an important alternative for fossil fuels, by using it for bioethanol production [\[17\]](#page-17-11).

According to the European Council Regulation (EC) 479/2008, grape pomace and lees must be delivered to alcohol distilleries for their transformation into exhausted grape pomace and a liquid waste named vinasse [18]. Small wine-makers usually do not respect this law, generating grape pomace and wine lees, along with grape stalks as wastes.

The main scope of this study was, through a systematic literature review, to clarify how the wine sector has assimilated the assertions of a circular economy. Six topics will be addressed, including chemical use, energy, land use and ecosystems, in different parts of the chain: viticulture, winemaking and distribution, solid waste, as well as water treatment. Moreover, besides its role as dairy fibre or a food ingredient, grape pomace can be considered an important source of natural phytochemicals including polyphenolics, useful as functional compounds for the pharmaceutical and cosmetic industries, due to their antioxidant, anti-inflammatory, antifungal, or scavenging activities [\[5,](#page-16-3)[19\]](#page-17-13). Therefore, the last part of this study focuses on the main vinery waste, grape pomace, highlighting its composition that induces its use for nutritional and health-promoting effects. The goal is to maximize the value of resources while they are being used, extend their useful lives as much as possible, and recover and regenerate goods and materials at the end of their α and α and α and α 479/2008, grape points α 479/2008, grape points α service life.

2. Winery Waste Composition and Source of Bio-Active Compounds

Winemaking by-products contain high amounts of bioactive secondary metabolites this law, generating grape pomace and wine lees, along with grape stalks as wastes. from various phytochemical groups (alkaloids, antibiotics, phenolics, resins, saponins, main various priy concentreal groups (analytical) and volatile review, priencifies, to call the most abundant h_{conv} , manns of a circular term μ_{max} assimilated the assertion μ_{max} and μ_{max} and μ_{max} compounds from various by-products from winery waste.

Table 1. Main characteristics of the by-products.

Therefore, these by-products can be further used for obtaining new products, such as food additives, animal feed, cosmetics, fertilizers, ingredients for foods/dietary supplements, medical remedies, nutraceuticals, antimicrobial components, or biomass for biofuels [\[21,](#page-17-15)[22\]](#page-17-16). Among the bioactive metabolites, polyphenols are one of the most abundant, representing around 70% of the bioactive components [\[23\]](#page-17-17). They can be classified into two major groups, based on their chemical structure, number of aromatic rings, and binding affinity: flavonoids and non-flavonoids [\[23\]](#page-17-17). Flavonoids from grapes are further divided into five subgroups: anthocyanidins, chalcones, flavonols, flavan-3-ols, and flavones [\[14\]](#page-17-8). Non-flavonoids comprise coumarins, lignans, phenolic acids (hydroxybenzoic and hydroxycinnamic acids), and stilbenes [\[23,](#page-17-17)[24\]](#page-17-18). The most abundant winemaking by-product is grape pomace (or grape marc), a solid organic material remaining after the crushing, draining, and pressing steps. It contains a mixture of grape seeds, skins, and stalks. It was estimated that from 6 L of wine, 1 kg of grape marc remains [\[21](#page-17-15)[,22\]](#page-17-16). The chemical composition of grape pomace is, however, influenced by the environment (climate and soil) [\[13](#page-17-7)[,25](#page-17-19)[,26\]](#page-17-20), the viticultural factors (defoliation, fertilization, grape variety, maturity, or harvest time) [\[26\]](#page-17-20), and by the used winemaking method [\[18\]](#page-17-12). One of the most important characteristics of grape pomace is the high polyphenolic content, due to its pharmacological properties. The compounds responsible for these properties are the alcohols, anthocyanins, flavanols, flavanols/catechins, flavonol glycosides, lignans, procyanidins, resveratrol, phenolic acids, and stilbenes [\[21,](#page-17-15)[25\]](#page-17-19). Grape skins are generally recognized as the major component of the grape marc, comprising up to 56% of the dry matter in red pomace, and 28% of the dry matter in white pomace [\[27\]](#page-17-21). The inner layer of the grape skins contains phenolics such as anthocyanins and tannins, but small amounts of gallates compared with other grape marc components [\[16\]](#page-17-10). Anthocyanins are mostly located in skins, the major compound being malvidin-3-O-glucoside, followed by peonidin-3-O-glucoside. The skins also contain neutral polysaccharides (12% hemicelluloses and 20% cellulose), 20% acidic pectin compounds, 15% insoluble proanthocyanidins, 5% compounds soluble in dichloromethane, 5–12% structural proteins, and 2–8% ash [\[27](#page-17-21)[,28\]](#page-17-22).

Another important winemaking by-product is represented by the grape seeds, corresponding to 38–50% of the grape marc on a dry matter basis and approximately 5% of the grape weight [\[29\]](#page-17-23). Every year, almost 3 megatonnes of grape seeds are globally

discharged by the winemaking industry [\[27\]](#page-17-21). Grape seeds are characterized by a high content of carbohydrates, fibre (40% *w*/*w*), lipids (16% *w*/*w*), minerals, polyphenolics $(7\% w/w)$, and proteins $(11\% w/w)$ [\[30\]](#page-17-24). Grape stalks represent the skeleton of the grape raceme, removed before the vinification, representing 14% by weight of the total wine solid waste [\[19\]](#page-17-13). The chemical composition of the stalks includes lignocellulosic materials, such as 20–30% cellulose, 3–20% hemicelluloses, and 17–26% lignin, but also 6–9% ash [\[28\]](#page-17-22). Stalks can also contain tannins (around 80%) associated with the lignin and more than 50% polysaccharides (the main one being glucose and xylose) [\[19,](#page-17-13)[31\]](#page-17-25). In terms of phenolics, grape stalks contain around 6% on a dry weight basis, the main ones being flavan-3-ols, flavonols (e.g., galactoside, glucuronide, glucoside, and rutinoside), hydroxybenzoic acids (gallic and syringic acids), and stilbenes [\[19,](#page-17-13)[32\]](#page-18-0).

Various studies have investigated the antibacterial properties of the extracts from winemaking by-products [\[33](#page-18-1)[–40\]](#page-18-2). The phenolic compounds responsible for the antimicrobial activity from winemaking by-products are alkaloids, coumarins, flavonoids, phenolic acids, quinones, saponins, tannins, or terpenoids [\[24](#page-17-18)[,41\]](#page-18-3). Table [2](#page-5-0) summarizes some studies on the antibacterial activity of winemaking by-products.

Table 2. Winery by-products as bio-active components.

Table 2. *Cont.*

The scarcity of investigation concerning the antimicrobial activity of winemaking by-products and the multitude of methods to obtain them make the effort to highlight conclusions from their analyses more complicated. Winemaking by-products were found to be active against multidrug-resistant strains in various investigations [\[35\]](#page-18-5), but also against bacteria with passive mechanisms of antibiotic exclusion (such as Clostridium or Bacillus) [\[26–](#page-17-20)[33](#page-18-1)[,38](#page-18-8)[,39\]](#page-18-9). Various studies mentioned that a broader inhibitory spectrum was found against Gram-positive bacteria [\[42\]](#page-18-10). It was observed that winemaking byproducts were most often bactericidal [\[43\]](#page-18-11). From Table [2](#page-5-0) it can be deduced that the minimum inhibitory concentrations vary extensively, indicating that not only the resistance or sensitivity of the target bacteria, but that other various factors that affect the type and concentration of the extract phenolics also affect their antimicrobial activity.

The antimicrobial activity of winemaking by-products can be influenced by various factors, such as the extraction solvent or procedure, grape variety, or pomace fraction [\[38](#page-18-8)[,40\]](#page-18-2). For instance, various studies reported that acetone/water/acetic acid extracts had higher

inhibitory activity compared to methanol/water/acetic acid extracts [\[39\]](#page-18-9). Regarding the grape variety, some reports mentioned that winemaking by-products resulting from red grapes had a higher minimum inhibitory concentration than those resulting from white grapes [\[19\]](#page-17-13). Further investigation demonstrated that there is a connection between the phenolic content of the extracts obtained from various pomace fractions and their antibacterial properties [\[38\]](#page-18-8). Comparing some of these results, seeds can be considered as more appropriate sources of antibacterial extracts than other fractions [\[35\]](#page-18-5).

A potential antimicrobial application of polyphenolic extracts resulting from winemaking by-products is as antibiotic adjuvants [\[44\]](#page-18-12). Their potential as multidrug resistance inhibitors may enhance the activity of the antibiotics [\[44\]](#page-18-12). However, the mechanism of plant polyphenols–antibiotic synergism is still unclear Although there seem to be four principal steps: (a) active site modification on and in the bacterial cell [\[45\]](#page-18-13); (b) inhibition of bacterial enzymes that affect antibiotic modification or degradation [\[46\]](#page-18-14); (c) increasing membrane permeability [\[45\]](#page-18-13); and (d) inhibition of the antibiotic outflow pumps [\[45\]](#page-18-13). An in vitro study against clinical multidrug-resistant strains of E. coli and S. aureus highlighted the synergism between grape pomace extracts and some antibiotics [\[47\]](#page-18-15).

The discovery of in vitro bioactivity is the first step on a long and difficult road in terms of drug development. Various factors must be taken into consideration: bioavailability, mode of delivery, toxicity, and the interaction with other compounds [\[44\]](#page-18-12). Furthermore, overcoming the actual lack of knowledge on the synergistic properties of the winemaking by-products and their mechanisms of interaction with the bacteria or antibiotics represents an important milestone [\[48\]](#page-18-16).

3. Grape Pomace as Antioxidant and Anti-Inflammatory Agents, Dietary Fibre, or as Ingredient in Food Products

The grape pomace is usually discarded by the wineries or is used as compost or animal feed without any pre-treatment [\[49\]](#page-18-17), although there is an increasing trend for finding new food sources or ingredients with nutritional and functional characteristics [\[50\]](#page-18-18). In this respect, the grape pomace corresponds with this trend due to its polyphenolic content (70% grape polyphenols) that remains after the vinification process. Phenolic compounds represent the main secondary plant metabolites, having one or more aromatic rings and they usually appear as glycosides, esters, and methyl esters which can be conjugated with mono-, oligo-, or polysaccharides [\[23\]](#page-17-17). The main polyphenolic compounds from grape pomace are the anthocyanins (only in red grape pomace), catechins, flavonol glycosides, or phenolic acids [\[51\]](#page-18-19). Phenolics are the most valuable compounds from grape marc, along with dietary fibre which have health benefits, such as the prevention of chronic diseases or cancer [\[52\]](#page-18-20). Additionally, polyphenols have an increased antioxidant potential and they can be used for food preservation, by inhibiting lipid oxidation and through their antibacterial effects [\[53\]](#page-18-21).

According to the literature, 6 L of wine generates 1 kg of grape pomace [\[27\]](#page-17-21). One tonne of grape pomace contains 249 kg stalks, 225 kg grape seeds, 425 kg grape skins, and water [\[54\]](#page-18-22).

The grape pomace can be regarded as an unconventional pectin source [\[55\]](#page-18-23). The physico-chemical composition and main components of grape pomace are presented in Table [3.](#page-7-0)

Table 3. Main components of grape pomace.

 $\frac{1}{1}$ TPC—total phenolic content; ² TAC—total anthocyanin content; ³ GAE—gallic acid equivalent; ⁴ CE—catechin equivalent; ⁵ AAE—ascorbic acid equivalent.

It is worth mentioning that grape pomace represents an important source of fibre (43–75%), such as cellulose, hemicellulose, lignin, and pectin [\[27\]](#page-17-21). However, few available products from grape pomace extracts are commercialized in various countries, according to the local regulations [\[29\]](#page-17-23); however, considering the low cost for marc processing and integrated usage, more products must be developed.

It is well-known that grape pomace presents high antioxidant properties, but the difference between grape pomace resulted from white or red wines must be established in order for the appropriate valorisation [\[59\]](#page-19-2). The strong antioxidant activity of grape pomace is due to its phenolic compounds and it is related to their chemical structure. The differences between the grape pomaces resulted from white or red whites are influenced by the terroir [\[14\]](#page-17-8) and by the winemaking technology [\[60\]](#page-19-3).

It was demonstrated that red grape pomace is rich in phenolic acids (such as gallic acid and protocatechuic acid), flavanols (such as myricetin-3-O-rhamnoside), stilbenes (mainly resveratrol), and anthocyanins (cyanidin-3-O-glucoside, delphinidil-3-O-glucoside, malvidin-3-O-glucoside, peonidin-3-O-glucoside, and petunidin-3-O-glucoside) [\[61\]](#page-19-4). White grape pomace contains high amounts of phenolic acids (such as homovanillic acid, gentisic acid, p-hydroxyphenylacetic acid, syringic acid, vanillic acid, 3- and 4-O-methylgallic acid, hydrocaffeic acid, hydroferulic acid, and isoferulic acid), flavanols (catechin, epicatechin, and procyanidin B1) [\[61\]](#page-19-4), flavonoid glycosides (such as isoquercitrin, quercitrine, and rutin), and flavonoid aglycons (such as luteolin) [\[62\]](#page-19-5). Overall, it was concluded that red grape pomace possesses a higher polyphenolic content than white ones [\[63\]](#page-19-6).

Various studies have revealed a beneficial impact of grape pomace polyphenols on metabolic syndrome, which represents a key factor in several health-related problems [\[64](#page-19-7)[–66\]](#page-19-8), for example, by reducing cardiovascular disease risk factors [\[67\]](#page-19-9), and decreasing hypertension and hyperglycaemia [\[68\]](#page-19-10).

Grape pomace, due to its antioxidant activity, can be helpful in atherosclerosis development and progression management, preventing LDL cholesterol oxidation and having anti-inflammatory effects. In this respect, red grape pomace effect was studied on ischemic heart disease in rats with induced atherosclerosis [\[69\]](#page-19-11). The results showed that it had antiinflammatory activity, resulting in the decrease in TNF- α (tumour necrosis factor α) and IL-10 (interleukin 10) levels. Moreover, it was observed that, by adding red grape pomace to the diet, the concentration of HDL cholesterol increased, leading to an anti-atherogenic effect and to the decrease in atherosclerotic lesions size and number [\[69\]](#page-19-11). Other in vitro and in vivo investigations observed that that grape pomace, due to its antioxidant activity, is helpful in the management of the pro-oxidant status, by increasing SOD (superoxide dismutase), CAT (catalase), GPx (glutathione peroxidase), and GRx levels (glutathione reductase) [\[70,](#page-19-12)[71\]](#page-19-13), and reducing GSH (glutathione), ROS (reactive oxygen species), and TBARS (thiobarbituric reactive substances) [\[72\]](#page-19-14). The grape pomace antioxidant effect was studied by UV radiation-induced oxidative stress in human keratinocytes, highlighting that the cells pre-treated with grape pomace manifested a significantly lower increase in protein levels and ROS, specific to the apoptosis process [\[73\]](#page-19-15). The antioxidant activity of white grape pomace was also studied on H_2O_2 -induced oxidative damage in human colonic epithelial cells; the grape pomace was found to play an important role in reducing ROS levels [\[59\]](#page-19-2).

Grape pomace has been used for the fortification of meat products, including pork burgers or sausages, pork loin marinade, and chicken meat (Table [4\)](#page-8-0).

| Product | Grape Pomace Addition Effects | | Ref. |
|--------------------|--|--|--------|
| Pork burger | 0.06% grape pomace extract added to product weight | Positive: Increased lipid oxidation inhibition and enhanced colour stability | [74] |
| Pork sausages | Positive: Decreased lipid 0.5 and 1% grape pomace oxidationNegative: Induced incorporated into the recipe colour change | | [75] |
| Pork loin marinade | Pork loin was soaked in 0.5, Positive: Inhibition of lipid oxidation and 1.0, 2.0, 20.0, and 40.0% grape pomace solution microbial growth | | [76] |
| Chicken meat | Positive: Decreased lipid Grape pomace extract oxidationNegative: Induced adding up to $TPC = 60$ mg/kg (in meat) colour and flavour change | | $[77]$ |
| Chicken meat | Grape pomace extract adding up to TPC of 10, 20, 40, and 60 mg/kg (in meat) | Positive: Decreased lipid oxidation | [78] |

Table 4. Grape pomace effect on meat and products.

The investigation of meat product fortification highlighted grape pomace's effects on meat product life, lipid oxidation, and stability. The fortification was achieved by grape pomace extract or powder addition to the recipe [\[74](#page-19-16)[,75,](#page-19-17)[77,](#page-19-19)[78\]](#page-20-0) or by soaking the product in a grape pomace solution [\[76\]](#page-19-18).

For example, 0.06% of grape pomace extract was added to pork burgers [\[74\]](#page-19-16), with a positive effect on the inhibition of lipid oxidation and on the colour stability [\[74\]](#page-19-16). Additionally, 0.5 and 1% grape pomace was included in pork sausages [\[75\]](#page-19-17), leading to decreased colour lightening and to lipid oxidation inhibition after 10 days of refrigerated storage [\[75\]](#page-19-17). The effects of a grape pomace marinade on pork loin quality was studied using concentrations of grape pomace solution of 0.5, 1.0, 2.0, 20.0, and 40.0% [\[76\]](#page-19-18). It was observed that the marinade led to lipid oxidation inhibition after 10 days of storage [\[76\]](#page-19-18). Chicken meat was efficiently fortified by a grape pomace extract added to the minced products up to a TPC (total phenolic content) of 60 mg/kg [\[77](#page-19-19)[,78\]](#page-20-0), observing that the lipid oxidation decreased due to the strong antioxidant activity, but with colour and flavour changes [\[77\]](#page-19-19).

Grape pomace was also used for yogurt fortification [\[55\]](#page-18-23) and production of cheese [\[79\]](#page-20-1). For example, the coagulated milk was fortified with grape pomace solutions (1, 2, and 3%) [\[55\]](#page-18-23). It was observed that the total dietary fibre content increased with grape pomace concentration [\[55\]](#page-18-23). The disadvantage was that the fortified yogurt exhibited a darker colour [\[55\]](#page-18-23). The grape pomace extract (1%) addition to another yogurt formulation increased its antioxidant capacity and TPC [\[51\]](#page-18-19). A slight colour change was observed, but no sensory effects were registered [\[51\]](#page-18-19). Semi-hard and hard cheeses were fortified with 0.8 and 1.6% grape pomace, resulting in increased antioxidant activity and TPC, but no changes in physicochemical characteristics and microbial counts were observed [\[51\]](#page-18-19).

Various compounds from wine may affect its quality and safety, including some metabolites (such as biogenic amines or ochratoxin A) as well as metals [\[80\]](#page-20-2). The best option to remove these harmful substances is fining. The role and behaviour of grape pomace in the process of wine fining and clarification was first investigated in 2013 [\[81\]](#page-20-3) using fibres extracted from Cabernet Sauvignon grape pomace. A decrease in phenolic concentration and in turbidity values in the treated wines was observed [\[81\]](#page-20-3). Furthermore, purified cell wall material was used for wine fining, positively affecting the wine phenolic content [\[82\]](#page-20-4). Cell wall purification and extraction represent a time-consuming process. In this respect, the wine repassage over grape pomace may decrease the ochratoxin A levels in must and wine during the vinification process, but the effects on removing biogenic amines or metals were not studied [\[80\]](#page-20-2). Purified grape pomace was applied for wine fining, inducing a significant reduction in ochratoxin A, but also in histamine, K, and Ca, indicating that it can be considered an alternative for protein-based fining agents [\[80\]](#page-20-2).

4. Winery Waste for Water Treatment

The capitalization of the agri-food industrial wastes constitutes a good opportunity for developing other products in the frame of a circular economy and sustainable waste usage [\[83\]](#page-20-5). Table [5](#page-9-0) presents the most relevant examples of water treatment using winery wastes.

Table 5. Relevant examples of water treatment using winery wastes.

The high organic carbon content from wine processing sludge may lead to excellent adsorbents for heavy metals due to their large surface area and increased binding affinity. For example, an eco-friendly adsorbent was obtained from grape marc and tested for the removal of lead (heavy metal) from contaminated effluents [\[83\]](#page-20-5). It was proven that the adsorption was strongly affected by pH. Maximum lead adsorption capacities were around 40 mg/g for Merlot grape marc and 64 mg/g for Sauvignon Blanc grape marc, at pH = 5.5 and 22 $°C$ [\[83\]](#page-20-5). However, further studies are needed to test the effectiveness of the heavy metal removal. Another study used wine processing waste sludge (WPWS) as an adsorbent for Ni removal [\[84\]](#page-20-6). WPWS proved to be effective due to its properties, such as the high organic matter contents, rough surface texture, and compounds with rich amino and carboxyl functional groups. The adsorption capacity was influenced by the pH, temperature, initial Ni concentration, and waste particle size. According to the Langmuir isotherm equation, the maximum sorption capacity was 66.55 µmol at 50 \degree C [\[84\]](#page-20-6). Further investigation remains necessary to elucidate the interaction between Ni and functional groups from WPWS compounds.

According to the literature, grape bagasse (GB) consisting of seeds and skins, has been used in its natural form for the removal of heavy metals such as Cd(II) and Pb(II) from water, exhibiting adsorption capacities of around 54 and 42 mg/g, respectively [\[85](#page-20-7)[,86\]](#page-20-8). It has also been used to obtain activated carbons, which were applied for the adsorption of dyes (basic blue 9 and acid yellow 36) and Cu(II), reaching adsorption capacities up to 417 mg/g [\[87](#page-20-9)[,88\]](#page-20-10). These results indicate that this type of adsorbent can be efficiently used for water treatment. A heavy metal found in water resources is mercury, a harmful chemical for humans and ecosystems due to its toxicological profile [\[90\]](#page-20-12). Its adsorption is essential, and adsorbents with specific surface functionalities are needed in order to reach a competitive removal performance [\[91\]](#page-20-13). In this respect, alternative adsorbents were prepared for mercury removal via the pyrolysis of grape bagasse [\[89\]](#page-20-11). Mercury adsorption capacities were evaluated through their heterogeneous surfaces containing carboxylic, lactonic, phenolic, and silicon functionalities. The maximum adsorption capacity reached 46 mg/g, with DFT calculations confirming that carboxylic functional groups and Si constituents were the primary active sites for adsorption [\[89\]](#page-20-11).

Wine waste-based sorbents can be green and cost-effective alternatives to commercial ones in terms of pharmaceutical removal from water. Although conventional wastewater treatment systems are efficient in terms of organic matter or nutrient removal, they are still not equipped for the removal of complex micro-pollutants like pharmaceuticals [\[92\]](#page-20-14). Their release into the aquatic environment can lead to chronic toxicity [\[93\]](#page-20-15). Pharmaceutical removal by adsorption is one of the most simple and efficient methods [\[94\]](#page-20-16). The commercial adsorbents ensure high removal rates, but their high cost represents a drawback; thus, rapid discovery of alternative low-cost and biodegradable adsorbents is needed. One of the residual products that remains after wine consumption is the cork stops. An interesting study proposed the use of this waste for low-cost bio-sorbent preparation [\[95\]](#page-20-17). The goal was to compare the adsorption capacities of commercial adsorbents (like activated carbon and synthetic zeolites) with those of bio-sorbents (cork waste) in terms of pharmaceutical (such as fluoxetine) removal from water. The waste-based bio-sorbent exhibited lower maximum adsorption capacity (4.7 mg/g) compared with the commercial adsorbents (233.5 mg/g, 32.1 mg/g, and 21.9 mg/g for active carbon, zeolite 13X, and zeolite 4A, respectively). Although the removal efficiency was lower, the economic feasibility was examined in terms of cost per gram of fluoxetine removed, with commercial adsorbents exhibiting higher costs (6.80 EUR/g, 3.13 EUR/g, and 1.07 EUR/g for zeolite 4A, zeolite $13\times$, and activated carbon, respectively) compared to the low-cost bio-sorbent (0.41 EUR/g) [\[95\]](#page-20-17).

5. Winery Wastes as Precursors for Sustainable Energy

The increasing interest in using biomass as an energy source has resulted in the use of winery wastes for thermal decomposition [\[27](#page-17-21)[,96\]](#page-20-18). The potential energy content of residual biomass resulting from a grapevine hectare was approximated at 19 GJ [\[97\]](#page-20-19). It was demonstrated that pyrolysis was more feasible than combustion from an economic point of view, resulting in minimal amounts of residue. From one tonne of grape marc, 150 kg of biochar and 140 kg of biofuel were produced [\[96\]](#page-20-18).

One disadvantage of the winery wastes is the humidity, which affects energy productivity. In this respect, other processes, such as hydrothermal carbonization, which requires milder operation conditions (180–250 ◦C and 20–40 bar), must be applied [\[98\]](#page-20-20). The resulting products mainly consist in a liquid phase with dissolved organics, a solid phase rich in carbon (hydrochar), as well as a small amount of gases [\[98\]](#page-20-20). Higher energy yield in hydrothermal carbonization was achieved by grape pomace processing [\[99\]](#page-20-21). A recent investigation confirmed that grape marc hydrothermal treatment constitutes an efficacious way to obtain CO_2 neutral solid fuels directly in the wineries [\[100\]](#page-20-22).

The winery waste removal and valorisation of carbons obtained with high efficiency would be an important advance, transforming an environmental problem into an economic would be an important devance, transforming an environmental problem into an economic
advantage [\[100\]](#page-20-22). There are few studies regarding the transformation of winery wastes into carbons, with the majority treating the potential use of biomass as raw material for porous carbons [\[101\]](#page-20-23). One of the more important research projects involved the valorisation of extractions prop. Since of the more important research projects involved the valentiation of winery wastes by one-pot activation [\[101\]](#page-20-23). Stalks, bagasse, and oil-free seeds were used as P intergraphent portugal portugal portugal portugal control of the section of the section of $CO₂$ $\frac{1}{2}$ capture and electric energy storage in supercapacitors $[101]$. not which was feuroval and valorisation of caloons obtained which guidelines in required from the more are constructed to more important research projects in the more important resea

way to obtain C_2 neutral solid fuels directly in the wineries \mathcal{I}_1

results in the use of anaerobic digestion for winery waste removal is an appropriate option, resulting in lower energy costs and improved efficiency [\[102\]](#page-20-24). The valorisation of vineyard residues by anaerobic co-digestion was investigated using activated sludge, resulting in 65% methane and a yield of $0.4 \text{ Nm}^3/\text{kg COD}$ [\[103\]](#page-20-25). The transformation of sludge with low organic matter concentrations led to a 25-30% biogas production [\[104\]](#page-20-26). Grape pomace, pulp, and seeds were processed by anaerobic digestion for methane production, with the is ubstrates being ground to increase their maximum degradability up to 22% [\[105\]](#page-20-27). The biogas and methane generation from organic wine wastes was also investigated, with the one with the highest potential being grape must, resulting in 1.152 and 838 m³/tonne volatile solids of biogas and methane, respectively [\[106\]](#page-21-0). **6. Wine By-Products as Raw Materials for Biopolymers and Natural**

6. Wine By-Products as Raw Materials for Biopolymers and Natural Reinforcing Fillers Reinforcing Fillers

Bio-based polymers may be directly extracted from biomass (Fi[gur](#page-11-0)e 3, Path 1), obtained by the polymerization of building blocks resulting from natural sources [\(F](#page-11-0)igure 3, Path 2), or synthesized by a direct microbial rout[e \(F](#page-11-0)igure 3, [path](#page-21-1) 3) $[107]$.

Figure 3. Bio-based polymers obtaining routes: (1) extraction from biomass; (2) polymerization of bio-based building blocks, and (3) microbial synthesis.

Bio-based polymers extracted from biomass, such as starch or cellulose (Path 1 from Figure [3\)](#page-11-0), are mainly mixed with other polymer types, because their direct usage is difficult due to their intrinsic characteristics. The obtaining of bio-building blocks from renewable sources (Path 2 from Figure [3\)](#page-11-0) is getting serious interest, resulting in sustainable polymers or other key bio-based chemicals. Furthermore, the microbial synthesis of polymers (Path 3 from Figure [3\)](#page-11-0) gained importance due to the obtained polymers' versatile characteristics [\[107\]](#page-21-1).

Among biopolymers, one of the most important is polylactic acid (290 thousand tonnes in 2019) [\[108\]](#page-21-2), which is a biodegradable linear aliphatic polyester synthesized by ring opening polymerization of lactone cyclic di-ester derived from lactic acid. Nowadays, lactic acid is industrially obtained from sugar or starch-rich biomasses, but research was conducted on using lignocellulosic sources to decrease the final polylactic acid price. In this respect, lignocellulosic vine shoots are considered a promising substrate for lactic acid production (Table [6\)](#page-12-0).

Table 6. Microbial synthetic route of lactic acid using various wine wastes as fermentative source.

Vine shoot hydrolysate (18 g/dm³ xylose, 11 g/dm³ glucose, and 4.3 g/dm³ arabinose) was used as a carbon substrate for lactic acid synthesis by *Lactobacillus pentosus*, resulting in a final lactic acid concentration of 21.80 g/dm 3 [\[113\]](#page-21-7). Furthermore, the vine shoot hydrolysate used as the carbon substrate was combined with distilled white wine lees (20 $\rm g/\rm dm^3$) as a source of nutrients, resulting in an encouraging volumetric productivity of 3.10 g/dm^3 h and a yield of 70% [\[112\]](#page-21-6). Several researchers used red and/or white lees as fermentative broth rich in nutrients [\[110,](#page-21-4)[111\]](#page-21-5). For example, 20 $g/dm³$ wine lees resulted from the second decanting (as nutrient) and *Lactobacillus rhamnosus* (as microorganism) produced 105.50 g/dm 3 lactic acid with a volumetric productivity of 2.47 g/dm 3 h. Hemi-cellulosic hydrolysates from vine shoot trimmings were used as a carbon source for a two-stage bioreactor [\[115\]](#page-21-9). In the first stage, 31.50 g/dm 3 lactic acid was produced through glucose conversion by *Lactobacillus rhamnosus*; in the second phase, xylose was fermented into xylitol by *Debaryomyces hansenii* [\[115\]](#page-21-9). Vine shoot hydrolysate was used to obtain, besides lactic acid, phenyllactic acid and biosurfactants by applying simultaneous saccharification and fermentation [\[114\]](#page-21-8).

Polyhydroxyalkanoates (PHAs) consist of biodegradable polyesters derived from 3-, 4-, 5-, and 6-hydroxyl-carboxylic acids. They often result from bacterial fermentation [\[116\]](#page-21-10). Depending on synthesis method, substrate, and process parameters, more than 150 types of hydroxyl-carboxylic acids can be obtained as PHA monomers [\[117\]](#page-21-11), leading to

Table [7](#page-13-0) presents studies in which biocomposites were obtained from biodegradable (polylactic acid, polybutylene succinate, or PHAs) polymers and wine by-products as reinforcing fillers.

| Biodegradable Polymer | Wine Waste | Filler Percent $(\%wt.)$ | Filler Treatment | Reference |
|--|-------------------|------------------------------------|--|------------------|
| Polylactic acid | Grape stalks | $30 - 50$ | Untreated | $[122]$ |
| | Grape pomace | $5 - 20$ | Untreated | $[123]$ |
| Polybutylene succinate | Grape pomace | $40 - 50$ | Reactive extrusion with maleic anhydride-grafted polybutylene succinate | $[124]$ |
| | Wine lees | $10 - 30$ | Reactive extrusion with silane (for the 20% formulation) | $[125]$ |
| $Poly-3-$ hydoxybutyrate-co- 3-hydroxyvalerate | Grape pomace | $5 - 20$ | Both untreated and after polyphenols extraction | $[126]$ |
| | Vine shoots | $5 - 20$ | Both untreated and after polyphenols extraction | $[127]$ |
| | Wine lees | $10 - 30$ | Reactive extrusion with silane (for the 20% formulation) | $[128]$ |

Table 7. Biocomposites obtained from biopolymers and wine by-products as reinforcing fillers.

Vine shoots and grape stalks have low apparent densities (approximately 0.03 g/cm³), affecting the cost of transportation when large-scale operations are required [\[129\]](#page-21-23). Regarding the average particle size, wine lees present lower values (D_{50} of 25 μ m) [\[128\]](#page-21-22). Lignocellulosic fillers, such as vine shoot or grape stalks containing particles with sizes varying between 0.3 mm and 5 mm surrounded by extended split fibres, and grape stalks (containing high amounts of cellulose, up to 30% wt.), have the ability to augment the elastic modulus of polylactic acid up to 65% with an intrinsic elastic modulus of 6–9 GPa [\[122\]](#page-21-16). Other studies have outlined the potential of wine lees to improve the elastic modulus of various biopolymers (for example polybutylene succinate or poly-3-hydoxybutyrate-co-3-hydroxyvalerate) and reported an intrinsic elastic modulus of 4–7 GPa [\[125](#page-21-19)[,128\]](#page-21-22). Vine shoot and grape pomace were used as reinforcing fillers for poly-3-hydroxybutyrate-co-3 hydroxyvalerate, with vine shoots increasing the elastic modulus while grape pomace had no effect on this characteristic [\[127\]](#page-21-21). The identified reason was that grape pomace cellulose and hemicellulose contents were low (10% and 6%, respectively), but it was rich in lignin (up to 42%, acting as a coupling instrument between cellulose and hemicellulose) [\[127\]](#page-21-21). Additionally, the stiffening effect was due to the rigid inorganic particles (for example alumina silicates or potassium tartrates) from the wine lees, which possess small diameter sizes, leading to particles dispersion and homogeneity [\[127\]](#page-21-21).

In order to fully completely the possible plasticizing effect of the wine wastes, lipid extracts must be applied [\[107\]](#page-21-1).

7. Valorisation of Wine Lees

Wine lees are materials that look like sludge, containing dead and living yeast cells, and yeast debris that gradually precipitate at the bottom of wine tanks [\[130\]](#page-22-0).

Wine lees ("heavy" or "light" depending on the decanting stage) were defined as "*the residue that forms at the bottom of recipients containing wine, after fermentation, during storage or after authorized treatments, as well as the residue obtained following the filtration or centrifugation of this product*" [\[131\]](#page-22-1) and represent 2–6% of the total volume of the resulting wine.

The high values of chemical oxygen demand (COD \cong 30,000 mg/L) and organic matter content (OMC \leq 35,000 mg/L) make the lees environmentally friendly [\[132\]](#page-22-2). Despite the various methods that were proposed to recover and valorise ethanol, polyphenols, and tartaric acid from wine lees [\[133\]](#page-22-3), few studies investigated the solid fraction from wine lees, which mainly consists of yeast biomass [\[134\]](#page-22-4).

Three extraction protocols were applied in order to extract glycol compounds from wine lees resulting from the alcoholic fermentation of a white wine [\[130\]](#page-22-0). The purpose of this study was to investigate whether the less could be used to increase wine stability and sensorial characteristics. It was observed that none of the extracts was equally efficient for all the tested characteristics, although some of them influenced various properties. For example, lees extracted by autoclaving influenced the wine tartrate stabilization, wine foaming, and tartrate recovery from the insoluble fraction. Contrarily, lees extracted by ultrasonication or enzymatic techniques aided the protein stabilization in heat-unstable wines [\[130\]](#page-22-0).

The wine lees usage as nutrient supplement for fermentation was investigated. For example, lees resulting from red or white wines from the first or second decanting stage were applied as a nutrient supplement for lactic acid production in the presence of *Lactobacillus* and glucose as the carbon source [\[111\]](#page-21-5). In 2010, a process for tartaric acid recovery was proposed, with the remaining wine lees being applied as fermentation medium for xylitol production [\[135\]](#page-22-5). Nevertheless, all the above-mentioned procedures did not take full advantage of the wine lees potential. In a biorefinery perspective, wine lees could be applied to produce antioxidants, ethanol, and tartaric acid, the remaining fraction rich in yeast cells could be transformed into a generic fermentation feedstock [\[121\]](#page-21-15). Wine lees hydrolysates and crude glycerol were applied as nutrient and carbon sources, respectively, to produce poly(3-hydroxybutyrate). Its production was significantly influenced by the free amino nitrogen content of the wine lees hydrolysates [\[121\]](#page-21-15).

The only actual commercial use of wine lees is for ethanol extraction by distillation, as required by European Regulations [\[18\]](#page-17-12). Nevertheless, this is not sufficient to reduce the chemical oxygen demand of wine lees or for a complete valorisation. In this respect, future investigation is needed to efficiently recover valuable compounds. For example, an integrated procedure for polyphenol extraction together with ethanol and tartaric acid extraction should be designed. The remaining biomass could be applied for obtaining fermentation media supplements, thus targeting a complete exploitation of the lees. One issue for industrial scaling is represented by the variability in wine lees composition. In this respect, the recovery of yeast cell wall polysaccharides may be an interesting option for the biomass valorisation. Indeed, the extraction of mannoproteins (MPs) and β-glucans (β-G) with potential applications was reported [\[136\]](#page-22-6).

8. Conclusions and Future Perspectives

This literature review highlighted a lack of sustainable and integrated usage of winery wastes. This study has reviewed the potential use of winery wastes, such vine shoots, grape pomace, and wine lees, as raw materials for various applications, in order to keep resources in use as long as possible, extracting their maximum value whilst in use, and recovering and regenerating products and materials at the end of their service life.

Due to their antimicrobial and antibacterial activity, and synergism with antibiotics, winery wastes may be used to manage antibiotic resistance while solving another worldwide problem, the environmental sustainability of agricultural activity. The high content of bioactive compounds, such as polyphenols, makes these wastes a promising source for new antibacterial agents.

Agricultural residues such as winery wastes have been pointed out as potential adsorbents for wastewater treatment. Transitional metal ions from wastewater are bound though functional groups such as hydroxyl, amino, or carboxyl, in proteins or phenolic compounds.

Wine by-products have been applied as carbon source for the microbial production of important polymer precursors, such as lactic or succinic acid. Likewise, grape stalks were used a raw material for the microbial synthesis of polymer precursors. However, even if they have a higher conversion yield, they have been used as carbon sources less frequently than vine shoots, due to their high content of antimicrobial tannins bonded with structural lignin, which are difficult to remove. In this respect, further investigation must be done to improve the detoxification treatment for tannin removal. The optimal valorisation path of grape pomace is the recovery of tartaric acid which can be further transformed into succinic acid to produce biopolyesters. Wine lees, predominantly containing salts and dead yeast, have been used as nutrient medium, offering a low-cost option compared with commercial yeast extracts.

Furthermore, wine wastes have been applied as reinforcing filler biopolymers such as polylactic acid, polybutylene succinate, or poly-3-hydoxybutyrate-co-3-hydroxyvalerate. In terms of mechanical characteristics, wine wastes can enhance the biopolymer stiffness and lower their tensile strength. Since each study tested wine wastes without any preliminary treatment, it is rational to imagine that wine filler surface treatment, such as acetylation or silanization, will enhance this characteristic. Further research must be done to test residual wine solids resulting from the biopolymers synthesis, and assessing the possibility of obtaining wine-derived plastics in line with a circular economy.

Even though various studies were achieved using grape pomace for different applications that could be applied by small- or medium-scale industrial producers, an integrated process including chemical, biochemical, and thermal treatments may be essential to maximally valorise this agricultural waste to produce a low-cost raw material. Grape pomace usage as a pectin source represents a promising future perspective. Consequently, its recovery from grape pomace, along with bioactive components and even oil, constitutes an attractive outlook for waste valorisation in terms of economic and environmental sustainability. Furthermore, grape pomace valorisation gained an increased interest from the food industry, with pectin extraction being the compelling argument for further intense investigation. Moreover, the most important components from grape pomace, dietary fibre and polyphenols, were identified as fortification elements for food. It was observed that the addition of grape pomace increased the total polyphenolic content, but, sometimes, the fortification also led to modification of sensory properties. In this respect, individual food commodities must be further tested separately for potential grape pomace fortification.

It was demonstrated that the grape pomace can be considered an alternative for wine fining, reducing ochratoxin A and other harmful compounds, with its use also potentially decreasing allergen-related effects. However, its influence on wine volatile compounds had not been studied, and further research is necessary. Moreover, grape pomace usage must be optimized since, after its addition, a significant loss of wine volume occurs through the lees.

The existing knowledge regarding the total phenolic content and antioxidant activity of grape pomace resulting from red and white wines is still limited, since it is necessary to clearly differentiate between the two types. In this respect, a future direction must be the evaluation of the climate, geographical factors, along with the methods used to obtain the grape pomace, in order to provide a proper valorisation method. Moreover, no sufficient information is available for comparing the effects of grape pomace from red and white wines on humans.

Furthermore, continuing the investigation into wine lees as a raw material for sourcing valuable compounds, could result in an improved exploitation of this by-product, providing a boost to the circular economy perspective within the winemaking industry. This area still needs to be developed to make these by-products viable on a commercial scale; this mainly concerns the methods to be applied for their processing, as they must be food-grade, cost-effective, sustainable, and maintain the expected functionality. Part of the reasons for the lack of efforts in wine lees valorisation can be assigned to their properties, e.g., the high polyphenolic content and the potential presence of adsorbed pesticide residues.

In terms of the production chain details, it is feasible to assess the potential energy content of the wastes resulting from a hectare of grapevines. During combustion, the biomass-based fuel behaviour can be affected by the presence of other fuels; further investigation is necessary for a proper assessment of the consequences from using these materials in co-combustion installations.

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