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A comprehensive approach to winery wastewater treatment: a review of the state-of the-art

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ABSTRACT

Winery industries generate large volumes of high-strength wastewater whose characteristics greatly vary depending on either seasons, production technologies or scale of the wineries. Winery wastewater (WW) is persistent to degrade by means of the conventional activated sludge process because of the high organic loading and polyphenolic content especially during vintage. To face this situation, a number of processes have recently been attempted as alternatives or integrative to biological treatments. However, there is still no agreement on the best practice to treat WW. Despite even more stringent standards, untreated or partially treated effluents continue to be improperly discharged into aquatic or soil matrixes, influencing microbial communities and physicochemical soil properties. This work presents a review on the state-of-the-art of management of wastewater originated from winery industries. Advantages and drawbacks of the treatment technologies at bench-, pilot-, and full-scale applications in the scientific literature have been considered to draw out a sustainable management scheme.

Keywords: Winery wastewater; Wastewater treatment; Aerobic processes; Anaerobic processes; Advanced oxidation processes; Integrated processes

1. Introduction

According to the International Organisation of Vineyard and Wine (www.oiv.int), the global production of wine in 2013 was 265 Mhl, of which 68% came from Europe, 19% from America, 5% from Asia, 4% from Africa, and 5% from Oceania [1]. Fig. 1 shows the worldwide wine production and the proportion produced in Italy [1,2].

Wine production has been traditionally seen as an environment-friendly process. However, it requires a considerable amount of resources such as water, fertilizers, and organic amendments, while producing a large amount of wastewater and organic wastes [3–6].

Wastewater and pollution loads vary greatly in relation to the working period (vintage, racking, bottling), the winemaking technologies adopted (e.g. in the production of red, white, and special wines), and wineries scale [3,7–12]. It is estimated that 1–4 L of wastewater is generated for every liter of wine produced [10]. In addition, there are seasonal peaks in

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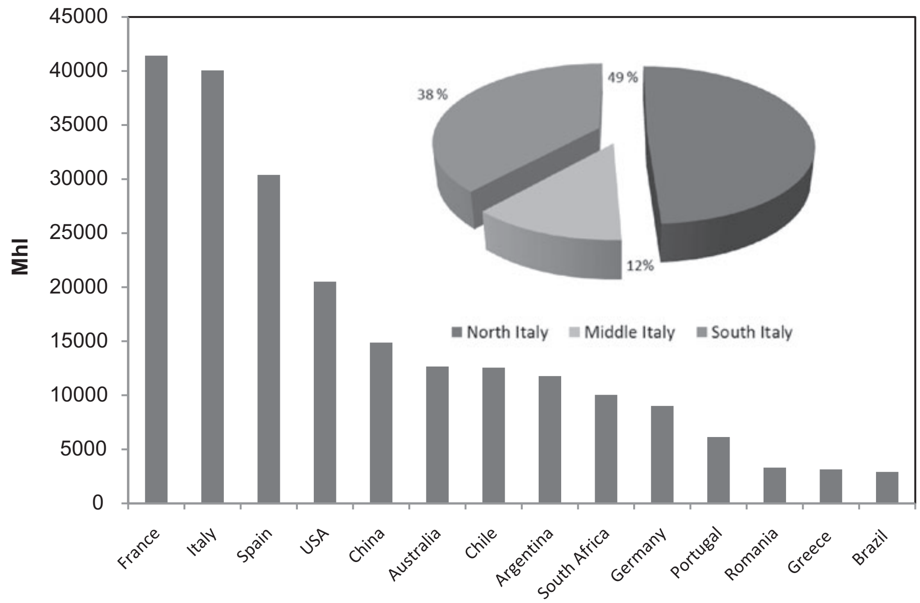

Fig. 1. Worldwide wine production and the proportion produced in Italy [1].

organic load associated with maximum pressing activity and refiltration of the newly fermented wine [13–15]. Over the last few decades, the adverse impact of liquid and solid wastes on the environment has become major concerns for the competitiveness of this industrial sector [5–7], especially for wine-growing countries; therefore, they need to be managed carefully before being discharged into the environment [4–8].

Traditionally, waste management efforts have focused primarily on "end-of-pipe" treatment methods. However, measures aimed at reducing the pollutant loading in winery effluent should be taken into account in order to effectively avoid or minimize the potential adverse environmental impacts associated with pollutants and waste [4]. To fulfill the strengthening legislation requirements, when cannot be conveyed to municipal wastewater treatment plants [10,13–16], winery wastewater (WW) has been managed by the construction of wastewater treatment plants for one single industry or a group of cellars in developed countries [4,17–21]. However, there has been no general agreement on the most suitable method for the management of WW yet.

Hence, this paper aims to gather WW characteristics reported in the literature and to discuss several treatment options, such as aerobic- and anaerobic-based biological processes, constructed wetlands (CW), physicochemical, oxidation processes, and their combinations in order to present the picture of more effective and sustainable management strategies of WWs.

2. WW characteristics

Wine production involves a biotechnological sequence of several unit operations including grape reception, destemming and crashing, wine vinification and maceration, pressing, fermentation, transfer and conservation, finishing, ageing, and bottling, as shown in Fig. 2 [3,8]. Winemaking is seasonal with high activity in the harvest period from early September until the beginning of November in the northern hemisphere, or the middle of February until the beginning of March in the southern hemisphere, a less-important activity on the occasion of transfers and filtrations, and a weak activity during the rest of the year [3]. Table 1 shows the variation of wastewater volumes produced in the process.

WW contains various pollutants such as sugars, ethanol, esters, glycerol, organic acids, polyphenolic compounds such as tannins, and a numerous population of bacteria and yeasts [9,12,22]. Their characteristics vary greatly during harvesting and normal period [10]. WW produced during vintage was reported to have higher BOD total, nutrients, electrical conductivity and toxicity, and more acidic in comparison to non-vintage seasons [5]. The low pH (3–6) of WW during the vintage can be partially ascribed to the presence of organic acids while the high COD is due to ethanol, sugars, and polyphenols [23,24]. Ethanol represents about 90% of influent COD during vintage as measured by Bories et al. [25] for a series of French winery facilities. On the other hand, WW is generally

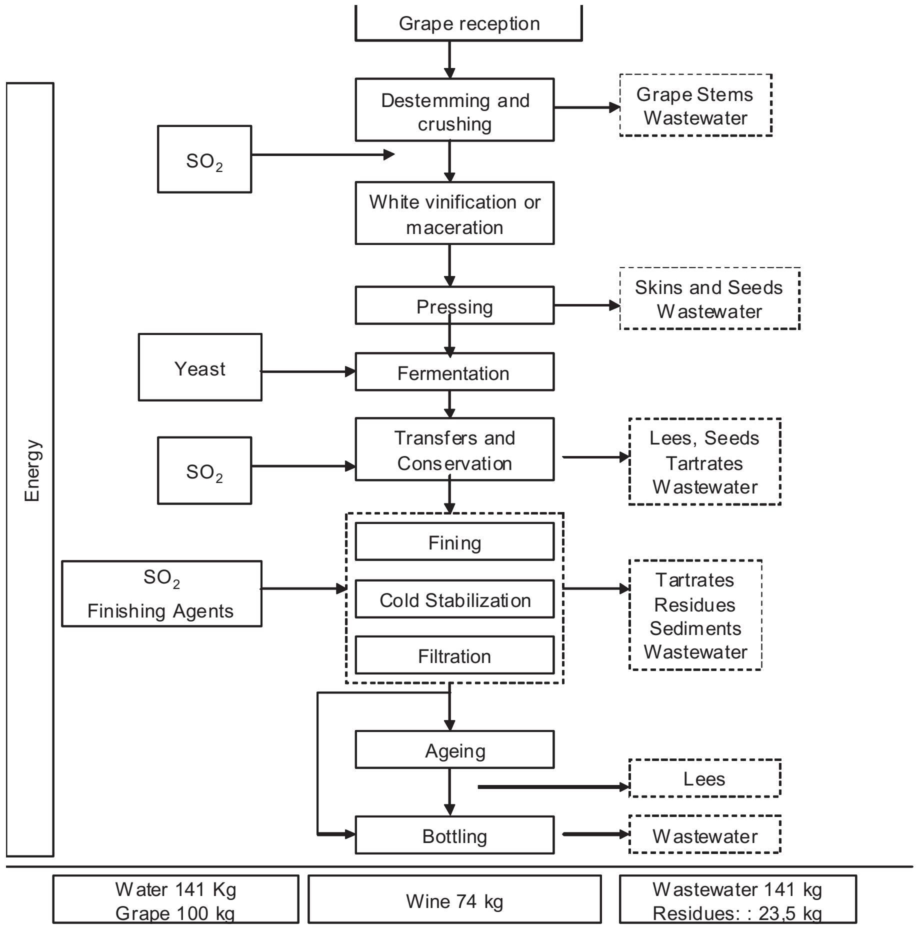

Fig. 2. A schematic mass balance of winemaking process (modified from Brito et al. [3]).

Table 1
WW production in Italy [12]

Wastewater production	Production factor kg/hl of wine	Quantity *t*/year
Vintage	116	6,180,000
Transfers	54	2,877,000
Cleaning operations	31	1,652,000
Bottling	18	959,000
Total wastewater	219	11,668,000

alkaline and saline during non-vintage seasons (pH 11) [15,26]. Agustina et al. [23] reported a detailed fractionating of the organic carbon composition of WW. The BOD level associated with grape crushing, barrel washing, and bottling may be as high as 5,000 mg L^{-1}. Effluents have a pronounced demand in nitrogen and phosphorous, with a BOD/N/P relation often near 100/1/0.3 [3,7]. Bolzonella et al. [10] reported a range of nitrogen and phosphorous concentrations of 50–80 mg $L^{-1}$and 5–25 mg L^{-1}, respectively. The ratio between the readily biodegradable COD and total COD varied from 0.3 (during the non-vintage period) up to 0.9 (during the vintage period) [16,27,28]. Andreottola et al. [19] claimed that soluble and flocculated COD (sfCOD) is the major part of the total COD covering 86% on average during the whole year. Further, the authors observed that there was an amount of non-biodegradable soluble COD (10%) discharged in the effluent.

Jourjon et. al. [29] reported that winery effluents are charged with micro-organisms by a factor that ranged from 105 to 108 UFC/ml. The flora composition was also very dependent on the period of the year and therefore, on the activities of the winery in which acetic and lactic bacteria and the yeasts were dominant at the beginning of the vintage and progressively diminished during the year. Measured typical characteristics of the WW are reported in Table 2.

Table 2
Wastewater characteristics in wine production industries*

Parameters	Units	Values
pH		3.5–12.40
Conducitivity	mS/cm	3.2–3.3
COD	mg/l	4,650–24,500
BOD	mg/l	3,250–13,400
TOC	mg/l	2,674
TKN	mg/l	1,350
TSS	mg/l	485–1,259
TS	mg/l	748–21,410
Polyphenols	$mg_{gallic\ acid}/L$	103–735
Cu	mg/l	0.5–1.63
Ni	mg/l	0.1
Cr	mg/l	0.12
Cd	mg/l	BDL
Zn	mg/l	0.14–1.47

Note: *Values from [52–45,62,64,65] BDL Below detection limit.

3. Treatment technologies for treatment of WW

As Table 2 shows heavy organic loads, WW should not be delivered directly to municipal wastewater treatment plants, or disposal sites [5,10]. The choice of a treatment system is not simple and must take into account a number of elements such as capital investments, retention time, and pollutant removal efficiency [29,30]. Among several technologies attempted for WWs treatment as explained in the following, a "mainstream" technology or process scheme treatment has not been established yet [31]. In other words, most studies in the scientific literature summarized in the following sections, present case solutions.

3.1. Physicochemical treatments

Physicochemical processes have been attempted to improve biodegradability before biological treatment [9] or to reduce residual organic load and color after biological treatment as well as for removal of metal content of WW [24].

Colloidal particles that tend to clog filtering devices are one of the problems in WW. Destabilization of the colloidal dispersion, inducing flocculation of large amount of dispersed matter, might result in an effective pre-treatment that can lower total dispersed solids (usually denoted as "Total Suspended Solids"—TSS), turbidity, and even part of the chemical oxygen demand (COD). A very effective two-step procedure with sepiolite and sepiolite modified with crystal violet, that changes the colloidal properties of winery effluents, inducing TSS and turbidity reduction has been recently reported by Rytwo et al. [32].

Chelating agents (TMT: 2,4,6-trimercaptotriazine) were used for the reduction of Cu (ranged from 0.68 to 1.63 mg L^{-1}) and Zn (varied from 0.14 to 1.47 mg L^{-1}) from raw WW. The efficiency was higher than 96% for Cu and 77% for Zn. Despite a high TSS removal (90%), only a slight COD removal (9%) was measured due to the fact that 92% of COD was in soluble form [24].

3.2. Aerobic treatment processes

Several aerobic process alternatives have been in question for WW as presented in Fig. 3. Main drawbacks and advantages of these treatment methods are summarized in Table 3.

3.2.1. Suspended growth processes

3.2.1.1. Activated sludge processes. The most conventional option continues to be activated sludge process (AS) with the addition of nutrients to improve biological treatment [10,11]. As the WWs show a low content of nutrients, additions of urea and phosphate salts are needed to guarantee the process of cellular synthesis.

A common alternative for WW treatment was reported to be the co-treatment with municipal wastewater in conventional activated sludge process (CAS) [16,17]. Beck et al. [33] carried out the optimization of a municipal activated sludge wastewater treatment plant to face high organic flow due to the input of viticulture effluents (COD of 468–4,240 mg L^{-1}; BOD_5 of 203–2,120 mg L^{-1}). They evaluated the use of a single aerated basin and observed that the optimization of the treatment line could be achieved by the installation of two reactors instead of one and by the introduction of a secondary clarifier between those two reactors. Bolzonella et al. [21] monitored a full-scale activated sludge wastewater treatment plant receiving both municipal and WWs. Influent COD varied from an average of 250 mg L^{-1} up to 500 mg $L^{-1}$with peaks of 1,000 mg L^{-1} during vintage. A COD removal of 75% was observed. The sludge production increased from 4 to 5.5 tons per day during the vintage and winemaking period. The estimated costs were calculated as €0.2–0.3 per m^3 of treated wastewater. Eusebio et al. [34] reported the results of a full-scale wastewater treatment plant with a rotating biological contactor (RBC) in series with an activated sludge process (double line), final filtration, and disinfection where municipal and pre-treated WW were co-treated (COD mass load of 2,200–3,800 kg d^{-1}). The activated sludge process was upgraded with alternate cycles (AC) performed. Accordingly, the coupled treatment was revealed to be outstanding to include a double step in

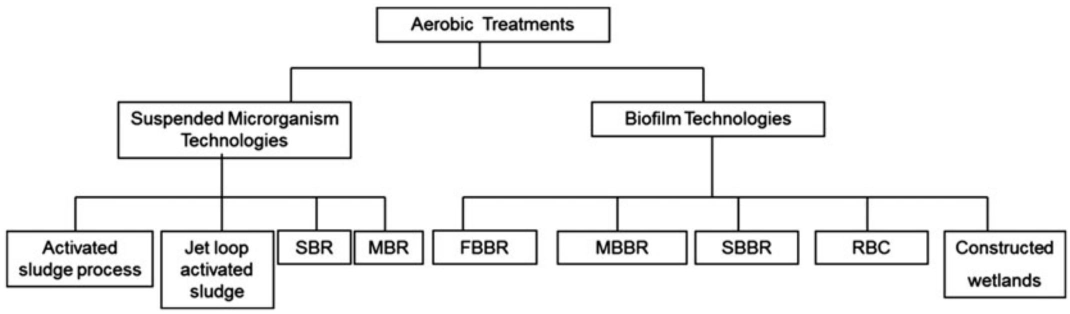

Fig. 3. Aerobic technologies for treatment of WW.
Note: Fixed bed biofilm reactor (FBBR), Membrane bioreactors (MBR), Moving bed biofilm reactors (MBBR), Rotating biological contactor (RBC), Sequencing batch reactor (SBR), Sequencing batch biofilm reactors (SBBR).

biological treatment to cope with the intense variation of influent loadings and to ensure an optimal nutrients removal in the second stage. They could save energy consumption from 13 to 23% by the AC process compared with the total oxidation process.

Although activated sludge treatment plants are suitable to be optimized, they remain strongly influenced by the toxicity of polyphenolic compounds present in these effluents [9]. Furthermore, seasonal operation in wineries may create a problem for aerobic systems leading to decreased sludge settleability, floc disintegration, and increased solids in the effluent [3].

3.2.1.2. Jet loop activated sludge reactors (JLR). The use of jet aeration systems is becoming more common as a means of combining efficient oxygen transfer with high turbulent mixing [35,36]. Jet loop reactors (JLR) usually show advantages of the absence of mechanical devices for aeration (e.g. blower, impellers, turbines, etc.). Petruccioli et al. [13] reported JLR reactors operated continuously with an organic load rate varied from 0.4 to 5.9 $kg_{COD}\ m^{-3}\ d^{-1}$ and 2.1–4.4 d of hydraulic retention time (HRT) achieved a COD removal efficiency (96–98%). A new type of JLR with more than 20 $g_{COD}\ L^{-1}$ d organic loading rate (OLR) was used by Eusebio et al. [37] and a COD removal greater than 80% was proved. Successively, Eusebio et al. [35] investigated the influence of the reactor hydrodynamics, causing high shear stress applied on the nozzle and its influence on the composition of the microbial population.

3.2.1.3. Membrane bioreactors. Recently, the use of membrane bioreactors (MBR) has been considered as a suitable option for WW treatment [10,14,24,27,38,39]. The advantages of the MBR system over CAS include maximum flexibility according to the influent loadings, small footprint, reduced sludge production, and a compact system with better solids removal and disinfection. As shown in Table 4, where their performances in pilot- and full-scale applications are described, COD removal was always higher than 95%. Lobos et al. [39] compared the performance of two immersed membranes operating in continuous mode (CMBR) and in a sequencing manner (SMBR). The results showed high COD removal and suspended solid retention for both systems (COD <125 mg L^{-1}). A better performance with CMBR was found notably in terms of controlling the filtration step while transmembrane pressure (TMP) evolution in SMBR system was greater than in CMBR.

The economic analysis carried out in the studies reported in Table 4 shows that MBR plants are very competitive when considering the treated water for some potential reuse application.

3.2.1.4. Sequencing batch reactor. Sequencing batch reactor (SBR) presents main advantages of the capacity for easily adaptation for continuous variations of pollutant concentrations. However, there has not been wide application of this process for WW treatment. Torrijos and Moletta [40] reported that SBR process was found to be effective during the 1994 vintage in a winery producing 7,300 hl annually located in *Domaine du Mouton* (Bordeaux, France). The SBR was fed once a day following a sequence of aeration/stirring for 20 h, decanting for 3 h, and pumping off of the clarified wastewater for the last hour. The authors indicated a volumetric applied OLR equal to 0.8 $kg_{COD}\ m^{-3}\ d^{-1}$ on average. A 93% removal for total COD, a 95% removal for soluble COD, and a 97.5% removal for BOD_5 were obtained.

3.2.2. Biofilm technologies

3.2.2.1. Fixed bed biofilm reactor. Among biofilm systems fixed bed biofilm reactor (FBBR) is a suitable alternative for WW treatment. Andreottola et al. [19]

Table 3
Main drawbacks and advantages of aerobic technologies used for WW treatment (modified from Andreottola et al. [39])

Technology	Advantages	Drawbacks
Activated Sludge (AS)	• High COD and BOD removal efficiency • Short retention time	• pH control • Floc disintegration, bulking • High operational costs • Inhibition of biomass due to polyphenolic compounds • Nutrients addition can be required
Jet loop reactor	• Absence of mechanical devices for aeration • Reduced energy consumption • No bulking problems	• pH control • Poor sludge settleability can occur • No adequate extent of experience at different scale and especially at full-scale • Nutrients addition can be required
Membrane bio reactor (MBR)	• Great improvements in treated water quality • Effluents free of suspended solids and bacteria (with UF) • Possibility of direct reuse on site • No sedimentation tanks • Small amount of wasted sludge • Quick start-up • Low footprint	• pH control • Fouling • High capital and managing costs, • Inhibition of biomass due to polyphenolic compounds • Nutrients addition can be required
Sequencing biofilm reactors (SBR)	• Operational flexibility • Less land required than AS • Lower cost than AS • Biomass cannot be washed out • Low maintenance	• pH control • storage basin to reduce shock loading • Nutrients addition can be required
Fixed bed biofilm reactor (FBBR)	• Reduction of the required volume with respect to the conventional AS system • Reduction in bulking problems • Absence of return flow and backwashing; • Easier management with respect to AS	• pH control • Plastic media costs • Nutrients addition can be required
Moving bed biofilm reactors (MBBR)	• High empty space of plastic media • Backwashing not required • Simple management • No bulking problems	• pH control • Additional cost of plastic media compared with activated sludge • Nutrients addition can be required

(*Continued*)

Table 3 (*Continued*)

Technology	Advantages	Drawbacks
Sequencing batch biofilm reactors (SBBR)	• High empty space of plastic media • Backwashing not required • Simple management • No bulking problems	• pH control • Additional cost of plastic media compared with activated sludge • Nutrients addition can be required
Rotating biological contactors (RBC)	• Easy to operate • Short start-up • Little maintenance • Effectively oxygenated • Little sloughing of biomass • No bulking problems	• pH control • Low rate of treatment • Nutrients addition can be required
Constructed wetlands (CW)	• Low-energy consumption and costs, • Capacity to treat record high organic loads • High rate of treatment in relatively short adaptation • Time	• High retention time • Large area • pH and TSS influence, • Odor problem • Feasible only in low density of population areas

investigated a full-scale FBBR system located in Trento province (Italy) for the treatment of WW. The treatment plant was consisted of screening, aerated equalization tank, first-stage FBBR (two reactors operating in parallel) followed by a settler, second-stage FBBR followed by a final settler. COD removal was affected significantly by the high fluctuations of the influent concentrations (6,957 ± 4,300 mg L^{-1} as annual average). In general, a high removal efficiency of total COD (91% as average, achieving an effluent COD concentration of 212 mg L^{-1} for the most of operation period) was obtained. However, the authors pointed out the difficulty to achieve lower effluent COD due to the non-biodegradable fraction of total COD (estimated 9.8% on average).

3.2.2.2. Moving bed biofilm reactors. Moving bed biofilm reactors (MBBR) consist of a process tank in which polymeric prism-shaped carriers are immersed and gradually colonized by the attached biomass on their own inner surface. Ødegaard [41] pointed out that MBBR combines positive aspects of both suspended growth and attached growth since no sludge recycle from the clarification compartment to the biological process is needed. However, additional cost of plastic media compared with activated sludge has to be taken into account. Andreottola et al. [42] reported a COD removal of 78–97% in the MBBR.

3.2.2.3. Sequencing batch biofilm reactors. Sequencing batch biofilm reactors (SBBR) result from the combination of MBBR and SBR. They correspond to the management of a fixed biomass grown on plastic media by a sequencing cycle. However, sludge settlement does not occur in the same tank of oxidation. Because the sludge settlement worsens in presence of the plastic elements, one aeration is stopped; the effluent is conveyed in a separate final settler. In this way, the great advantage of SBR given by the reduction of plant volume is lost. Andreottola et al. [43] carried out pilot-scale experiments applying SBBR to treat wastewater generated in a winery factory of Trento province (Italy). The study lasted from September (coinciding with the vintage) to June in order to follow the complete production cycle through the year. Daily influent flow rate varied between 30 and 108 L d^{-1}. COD removal efficiency varied from 85% and 99 at organic loads up to 29 gCOD $m^{-2}d^{-1}$. Urea (0.11 $g_{urea}g^{-1}{}_{COD}$) and orthophosphoric acid (0.018 g_{H3PO4} $g^{-1}{}_{COD}$) were added as N and P sources in order to guarantee the biosynthesis.

3.2.2.4. Rotating biological contactors. RBC reactors offer principal advantage of the large interfacial area which is practically independent of the speed of rotation, unlike in the activated sludge process. Malandra et al. [11] evaluated RBCs for the treatment of the samples taken from various wineries. The results indicated an

Table 4
Findings of MBR applied to WW

Winery wastewater	OLR	MBR description	Main findings	Costs (€/m^3)	References
Screened winery wastewaters.	500–2 kg COD/d	• Kubota (UK), submerged MBR system[b] • Two trains of 200 modules with a surface of 138 m^2 per train • Plate and frame microfiltration membranes (0.4 mm) with a 10 mm space between two adjacent panels	• COD removal around 95%, also for organic loading rate up to 2 kg COD/m^3 of bioreactor per day • Specific energy consumptions 2.0–3.6 kWh/m^3 of treated wastewater or 1 kWh per kg of COD removed		[10]
Synthetic winery wastewater	0.5–2.2 kg COD/m^3	• Zenon ZW-10 submerged MBR system[a] • Nominal surface area of 0.9 m[b] • Hollow fiber ultrafiltration membrane module	• COD removal efficiency was always higher than 97%	0.38	[27]
Synthetic winery wastewater	2 kg COD/m^3	• Zenon ZW-10 submerged MBR system[a] • Nominal surface area of 0.9 m[b]; • Hollow fibre ultrafiltration membrane module	• COD removal efficiency above 97%		[14]
Winery wastewater	Influent COD 100–8,000 mg/L	• Kubota membrane modules[a] • Flat sheet membrane • 20 m[b] membrane surface area	• COD removal efficiency above 97% • 100% removal of biodegradable fraction of the COD • MBR permeate is suitable for urban, agricultural and recreational reuse according to the quality specifications defined by international guidelines and regulations for water reuse and reclamation	0.40	[38]

[a]Pilot plant.
[b]Full scale.

average COD decrease of 43% with a retention time of 1 h. The inflow and outflow COD values varied in the range of 3,380–6,090 mg L^{-1} and 1,715–3,475 mg L^{-1}, respectively.

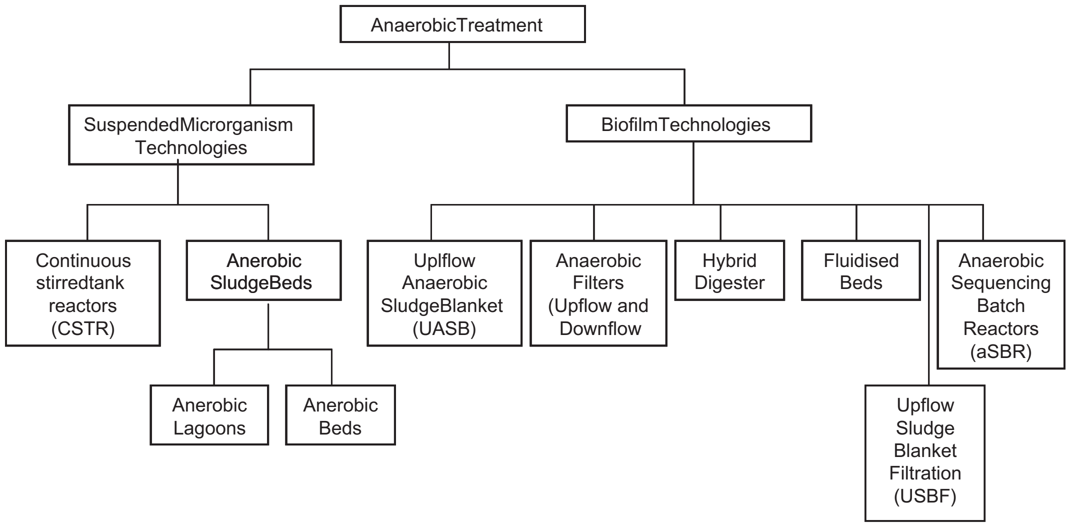

Fig. 4. Anaerobic technologies for WW treatment.

3.2.2.5. Constructed wetlands. In the case of large area availability, constructed wetlands are simplified systems with a low energy consumption [44,45]. From the technical point of view, CWs are realized with low deep basins, with long HRT, filled up with sand, gravel, and earth on a protective coating of waterproof sheath, and planted with macrophytes [46]. Mulidzi [47] showed an average COD removal of 83% in winter and 80% (initial 8,000–12,000 mg L^{-1}; retention time of 14 d) in summer by CWs fed with WW in Goudini, South Africa. Furthermore, several studies indicated that removal of solids from WW and pH adjustment are important in CWs [26,46]. Meunier et al. [48] evaluated the treatment of winery effluents by a combined process of SBR with reed bed filters. An average of 94% of the COD and 89% of SS was eliminated, revealing that CWs may be suitable as secondary treatment options for WW.

3.3. Anaerobic treatment processes

Anaerobic digestion is found particularly suitable for WW having various potential benefits such as low excess of sludge production and can become profitable by cogeneration of useful biogas [22,31,42,49,50]. However, in the case of overloading, volatile fatty acids (VFA) can be accumulated in the reactors producing a drop of pH with a failure of the process. Furthermore, the anaerobic digestion generally needs to be followed by an aerobic treatment to meet the discharge standards. Several configurations of anaerobic processes are reviewed by Moletta [49], as shown in Fig. 4. Main drawbacks and advantages of those process modifications of anaerobic treatment are summarized in Table 5.

The most promising anaerobic process was cited as granular up-flow anaerobic sludge blanket reactor (UASB) for WW [49]. Most of the practical UASB systems are operated under mesophilic conditions; however, thermophilic operation results in higher methanogenic activity [31]. The success of UASB depends on the formation of active and settleable granules, which can further be inoculated with selected natural bacterial strains. One reactor was seeded with granular sludge enriched with *Enterobacter sakazakii* and a 90% COD removal was obtained treating an inflow COD at a HRT of 24 h [22].

Anaerobic filters (AF) are also used in WW treatment but only few applications can be noted [49]. The AF reactor often encounters the problems of clogging and channeling, in particular, when the AF is packed with high specific surface media. Additionally, the anaerobic micro-organisms are not evenly distributed along the height of the filter.

Fluidized bed reactors (FBR) contain an appropriate media such as sand, gravel, or plastics for bacterial attachment and growth. The reactors can be operated either in the upflow or downflow modes [31]. Fernandez et al. [51] treated the vinasse generated in the production of a tropical fruit wine in anaerobic FBRs (OLR: 2–5 $kg_{COD}m^{-3}\,d^{-1}$). The authors showed that there was no accumulation VFA in the process.

Upflow sludge blanket filtration (USBF) reactor configuration combines the main advantages of the upflow anaerobic filter (UAF) and UASB reactors for biomass retention. Molina et al. [52] run a pilot plant,

Table 5
Main drawbacks and advantages of anaerobic technologies used for WW treatment (modified from Andreottola et al. [39])

Process	Advantages	Drawbacks
Continuous stirred tank reactors (CSTR)	• Energy recovery • Enhanced methane production in the case of co-digestion with activated sludge • Low cost	• Settler is required • Sludge recirculation
Anaerobic lagoons	• Easy to operate • Biogas production	• High retention time • Large area • Odor problem
Upflow anaerobic sludge blanket reactor (UASB)	• Elimination of mechanical mixing • Recycling of sludge biomass • Ability to cope up with perturbances caused by high loading rates and temperature fluctuations • Biogas recovery	• Occasionally accumulation of floating scum • Pre-treatment requiring aerobic post treatment for effluent with low COD concentration
Anaerobic filters(AF)	• Long solid retention time • High removal rate per unit of reactor volume and at low HRT • Low sludge production • Biogas production	• Clogging or channeling (especially with high specific surface media • No (or very low)solid concentration required
Upflow sludge blanket filtration (USBF)	• Combination of main advantages of UAF and UASB reactors • Reduction of clogging and biomass flotation problems • Rapid start up • Energy recovery • Low sludge production	• Few applications in the literature
Fluidized bed	• High removal rate per unit of reactor volume • Low sludge production • Biogas production	• Small fluidized particles • High fluid velocities
Anaerobic sequencing batch reactor (ASBR)	• Operational flexibility • No separate clarifier is required • Good effluent quality control • Biogas production and energy recovery • Optimization of cycle length by automation	• Nutrient addition could be required • Online monitoring and modeling needed for optimization

started up with an OLR of 0.5 $kg_{COD}m^{-3}\,d^{-1}$. Once the process was stable and the effluent concentration was lower than 500 mg L^{-1}, the OLR was gradually increased. An OLR of 5 $kg_{COD}m^{-3}\,d^{-1}$ with a COD removal of 98% and a negligible accumulation of intermediates was obtained. An adequate biogas quality was also achieved (70–74% CH_4).

Anaerobic sequencing batch reactor (ASBR), which is a fill and draw process like the aerobic SBR, involves repetition of cycle including four discrete steps: fill, react, settle, and draw [49]. Ruiz et al. [53] worked at a laboratory-scale ASBR at a 8.6 $kg_{COD}\,m^{-3}$ OLR value. They showed a soluble COD removal efficiency greater than 98% at a HRT of 2.2 d.

3.4. Advanced oxidation processes

Advanced oxidation processes (AOPs) involve the generation of highly reactive radical species, predominantly hydroxyl radicals (·OH), which react non selectively with a wide range of organic compounds [9,16,23,54]. The main drawback of AOPs lies in the high cost of reagents such as ozone, hydrogen peroxide, or energy light sources, as shown in Table 6. However, the use of solar radiation can significantly reduce costs [55]. Table 7 summarizes main findings of various AOPs applied to WW.

3.4.1. Fenton and photo-Fenton processes

The Fenton process involves the reaction of ferrous ions (catalyst) with hydrogen peroxide (oxidizing agent) to form the active hydroxyl radicals. The Fenton reaction is markedly accelerated by light, so the photo-Fenton reaction typically gives faster rates and a higher degree of mineralization. Furthermore, the photo-Fenton reaction can be driven with low-energy photons in the visible part of the spectrum. Thus, photo-Fenton processes are potential low-cost AOPs that can be achieved under solar irradiation [16,18,55–62]. According to Table 7, winery effluents can be efficiently degraded by Fenton and photo-Fenton processes.

Lucas et al. [56] run an aerobic biological process followed by a Fenton's reagent. By this combination, a COD removal of 99.5% was achieved when the mass ratio (H_2O_2/COD) was kept equal to 2.5, maintaining constant the molar ratio of H_2O_2/Fe^{2+} at 15. Mosteo et al. [57] applied photo-Fenton treatment in heterogeneous phase under energetic conditions to synthetic WW. They obtained a high purification level, up to 50% of total organic carbon (TOC) removal.

More recently, ferrioxalate-induced solar photo-Fenton treatment of WWs was performed at pilot scale by Monteagudo et al. [58] as a pre-treatment step prior to biological process. Because H_2O_2 has a maximum absorption at 220 nm and can only absorb photons below 320 nm, ferrioxalate can be used to increase the oxidation efficiency of the solar photo-Fenton process as it is a photo-sensitive complex that is able to expand the useful range of the solar spectrum up to 450 nm. High levels of polyphenols compounds have inhibiting characteristics to aerobic biological processes, so it is considered of interest to know the behavior of these compounds when Fenton/photo-Fenton processes are used as pre-treatments. Mosteo et al. [59] observed that the amount of polyphenols increased at the beginning of the oxidation due to the hydroxylation of the aromatic rings, before to down to 40 mg L^{-1}.

3.4.2. Photocatalysis

The photocatalytic oxidation process is based on the formation of hydroxyl radicals that occurs as a catalytic semiconductor is illuminated with near UV radiation ($\lambda < 400$ nm). Although several semiconductors have been tested as photocatalysts, TiO_2 is the widest used one because of its high stability, low-energy band gap, toxicity, and cost. The behavior of the process can be enhanced by adding hydrogen peroxide in order to increase hydroxyl radicals formation, facilitating compliance with the specific treatment requirements. The $H_2O_2/UV/TiO_2$ treatment (H_2O_2 2.5 ml L^{-1} and catalyst 1 g L^{-1}) using, respectively, natural (24 h treatment) and artificial light (80 min. treatment) reached a 52–58% removal ratio of COD in real WW samples [60]. Meanwhile, Agustina et al. [23] reported that the addition of TiO_2 affected negatively the process efficiency of photolysis. As shown in Table 6, the highest photodegradation rate by means of COD and TOC removals was achieved at zero catalyst loading.

3.4.3. Ozone oxidation processes

The treatment of WW by ozonation and ozone-related processes has been demonstrated in laboratory studies and pilot-scale plants [6,28,61,63,64]. The ozone oxidation mechanism follows two different pathways: (1) at low pH, ozone exclusively reacts via direct pathway; (2) at alkaline and neutral pH, the degradation rate is accelerated by the formation of radical species from the decomposition of ozone. As consequence, a low reduction in COD could be observed under the action of ozone at the acidic pH [9,28,61,64]. In order to improve COD removal, Beltran et al. [61] reported

Table 6
Main drawbacks and advantages of AOPs used for WW treatment

Processes	Advantages	Drawbacks
Fenton and photo-Fenton	• No pH adjustments required (vintage season) • Decolorization of the effluent • Biodegradability improvement • Toxicity reduction • High efficiency • Catalyst can even be activated by sunlight	• Inorganic sludge production • Consumption of high energy photons generated by artificial UV light • Chemicals and energy consumption • Technology complexity • No adequate extent of experience at full-scale
Photocatalysis	• Complete mineralization of toxic organics is possible • Removal and recovery of many toxic metals are possible • Catalyst (TiO_2) is relatively cheap • Oxidant is atmospheric oxygen. • Catalyst can even be activated by sunlight	• UV Light is required for surface area for activation • TiO_2 shading effect on the light reaching the organic contaminants • Technology complexity • No adequate extent of experience at full scale
Ozone	• Color removal • Free of sludge production • Biodegradability improvement • High removal of phenol compounds • Toxicity reduction	• Low performances for COD removal • Technology complexity

that adjusting pH in sequential cycles (acidic and alkaline periods) was required. On the other hand, the degradation rate of polyphenol by ozonation was found to be faster at acidic pH than at alkaline pH [6].

3.4.4. Electrochemical treatments

Electrochemical oxidation can be achieved by both the direct and indirect processes and its effectiveness strongly depends on the treatment conditions and on the nature of the electrodes materials. The behavior of the process can be increased through the combination with hydrogen peroxide and ozone. Nowadays, electrochemical technologies have reached such a state that they are not only comparable with other technologies in terms of cost but also are more efficient and more compact. Although the electrochemical oxidation proved uneconomic when applied to raw wastewaters, due to the very high-energy requirement to deplete pollutants to acceptable concentrations, the process was found to be competitive in costs of operation and treatment efficiency when applied as a final polishing step. For the above-mentioned reasons, the electrochemical treatment is widely used to remove the color from winery effluents [54,55]. According to Table 7, the process allowed to achieve high color removal efficiency (>80%) and COD removal higher than 40% in wastewater with the highest values of initial COD [54].

4. Discussion

In order to evaluate the sustainability of the different wastewater treatment options for WW, the location of the winery industry plays a significant role. Because most of winery industries are located in rural areas where there is no sewage collection system, the on-site treatment may considerably reduce the risk to potential impact on the receiving water body and the management costs as well. On-site treatment should start with a pH and flow balance stage prior to submit wastewaters to a sequential chemical treatment one. In the chemical treatment stage, chemical precipitation using biopolymers as coagulant would be a good alternative for organic content–sludge production. Chemical treatment would unavoidably be followed

Table 7
Findings of various AOPs applied to WW

References	Wastewater characteristics	Location	COD influent (mg L^{-1})	AOPS	Type of equipment and experimental details	Highlights of the work
[62]	Partially biological treated effluent (SBR)	Cyprus	1,060	Photo–Fenton	*Pilot description(1)* Bench scale UV-A irradiation provided by a 125 W lamp *Operational conditions* pH 2.5 $[H_2O_2]$: 34–175 mM $[Fe^{2+}]$: 0.5–2 mM	80% of COD removal *t* 4 h
[58]	Chemically pre-treated	Spain	TOC 2,674–50	Solar photo-Fenton	*Plant description (2)* The compound parabolic collector (CPC) pilot plant contains a solar reactor consisting of a continuously stirred tank (50 L), a centrifugal recirculation pump, and a solar collector unit with an area of 2 m^2 *Operational conditions* $[H_2O_2]$: 250 mg L^{-1}; $[Fe^{2+}]$: 10 mg L^{-1}; $[H_2C_2O_4]$: 30 mg L^{-1};	Under optimal conditions, 61% TOC removal from the treated water was achieved in 360 min. The correlation between consumed hydrogen peroxide and removed TOC was found to remain constant. Thus the addition of H_2O_2 can be used to control the degree of mineralization for this type of wastewater
[16]	Effluents already treated by a membrane bioreactor (MBR)	Cyprus	120	Solar photo-Fenton	*Plant description (1)* For the solar photo-Fenton experiments, irradiation was provided by a 1 kW lamp (Xe-OP) of a solar simulator (Newport model 91,193). The irradiation intensity of the simulator was 272.3 Wm^{-2} *Operational conditions* pH 3 Fe^{2+}: 1–5 mg L^{-1} H_2O_2: 25–500 mg L^{-1} *T*: 15–45°C *t*: 20–120 min	Optimum Fe^{2+} and H_2O_2 concentrations were found to be 3 mg L^{-1} and 250 mg L^{-1} respectively 70% COD removal 53% DOC removal 75% color removal *t* 120 min and *T* 25°C The toxic and the phytotoxic compounds still present in the effluent after the biological treatment were reduced or even eliminated through solar photo-Fenton oxidation
[23]	Winery wastewater	Western Australia	not specified	Photocatalysis	*Plant description (1)* Experiments were carried out in batch operation in an annular-type reactor 60 L capacity e with an irradiated volume of 38.5 L. The outer chamber was a stainless steel vessel (i.d. = 30 cm, height = 60 cm), fitted with a UV lamp (Primarc Ltd., PM2326 emitting in the wavelength range 310–435 nm, with a maximum emission at 365 nm) enclosed in a cylindrical quartz tube (i.d. = 5 cm) *Operational conditions* pH 3–11 gas flow rates investigated: 0–10 L min−1 TiO_2 0.5–1–3 g L^{-1}	Optimum gas flow rate was 6 L/min 84% COD removal 75%TOC removal 0 catalyst loading pH 6.5 *t* 60 min *T*: 25°C

(*Continued*)

Table 7 (*Continued*)

References	Wastewater characteristics	Location	COD influent (mg L^{-1})	AOPS	Type of equipment and experimental details	Highlights of the work
[61]	Wine-distillery wastewater diluted with domestic wastewater (1 : 10 v: v)	Spain	not specified	Ozone	*Plant description (1)* Ozone was produced from oxygen with a 500 Fisher laboratory ozone generator. Ozonation runs were carried out in two flow modes: semi-batch and continuously. Semi-batch experiments were completed in a glass bubble column (i.d. 9 cm; length 45 cm) equipped with a diffuser plate (pore diameter, 16 ± 40 mm) at its bottom to supply the oxygen ± air gaseous mixture. A peristaltic pump was used to circulate the wastewater at 20 L h^{-1} flow rate, thus providing good mixing conditions *Operational conditions* Applied O_3 dose 10 and 20 mg L^{-1} pH 4 and 10 *T*: 20°C	pH sequential ozonation with 2 cycles (two acidic ± alkaline periods of 10 and 50 min, respectively) represented optimal conditions COD removed and the ozone efficiency reached 41.4 and 78%, respectively
[63]	Winery wastewater	Spain	800	Ozonation Photocatalysis (UV/TiO_2) Photocatalytic Ozonation ($UV/TiO_2/O_3$)	*Plant description (1)* 1 Liter capacity tubular borosilicate glass photoreactor (450 mm long, 80 mm diameter) was used in all of the experiments. Ozone was produced from oxygen in a laboratory SANDER 301.7 ozone generator with a maximum capacity of 12 g h^{-1} A high-pressure mercury lamp (Heraeus, TQ 718 700 W) immersed in a Pyrex glass well was placed at the middle of the reactor. The lamp bandwidth was located in the range of 238–579 nm with three main wavelengths emitting at 254, 313, and 366 nm *Operational conditions* pH 3–11 TiO_2 0–3 g L^{-1} Ozone concentration 50 mg L^{-1} *t*: 0–120 min	If ozone was applied with a concentration of 50 mg L^{-1}, the final COD conversion reached 37%, whereas the mineralization level is located around 15%. pH 11 slightly enhances the COD removal if compared to neutral or acidic conditions The combinations light/O3 and light/TiO2 do not achieve significantly better results in terms of COD removal (43 and 41%, respectively) The system UV-A/vis/O3/TiO2 leads to the total mineralization of the COD removed. TiO_2 1.5 g L^{-1} pH 7 Ozone concentration 50 mg L^{-1}
[64]	Winery wastewater	Portugal	4,650	Ozone O_3/UV $O_3/UV/H_2O_2$	*Plant description (2)* The ozone-based AOPs were conducted in a bubble-column, semi-batch reactor (internal diameter 0.1 m, height 1 m) made of QVF borosilicate glass equipped with a quartz UV-lamp tube (external diameter 40 mm) mounted in the axial position which houses UV-C lamp used (Philips TUV36 W, 36 W nominal power, bulb dimensions: 1.W156 m long, 28 mm in diameter) emitting predominantly monochromatic radiation at 253.7 nm (15 W UV-C output) *Operational conditions*	At the natural pH of the wastewater (pH 4) the effectiveness of each AOP followed the sequence: $O_3/UV/H_2O_2 > O_3/UV > O_3 >$ UV-C. The rate of COD and total organic carbon(TOC) removal were enhanced by operation at neutral (pH 7) and at alkaline pH (pH 10) With a COD/H_2O_2 (w/w) ratio equal to 4, the COD removal was 35% after 180 min

					COD/H_2O_2 ratios 1, 1.3, 2, 4 pH 4,7,10 *t*: 0–180 min O_3: 100 mg min^{-1}	
[54]	Winery wastewater	Israel	10,147	Electrochemical treatment (EC)	*Plant description (1)* Batch laboratory tests (three sets) were performed in identical 1 L glass containers. Two 11 cm × 8 cm aluminum electrodes, 1.5 cm apart, and connected to a DC-power supply operated at 2.5 A and 10 V, were set in each container *Operational conditions:*	A 10 min batch treatment of WWW produced 16.4–27.9% reduction of biochemical oxygen demand (BOD), 28.2–41.9% of the chemical oxygen demand (COD), and 89.2% the total phosphorus
				EC/O_3	O_3 1 L min^{-1} *t*: 10–40 min	In the case of BOD removal, O3 did not improve the EC performance. In contrast, the EC performance in removing the COD was enhanced by the O_3
				EC/H_2O_2	0.75% H_2O_2	2.5% dilution of WW by 30% hydrogen peroxide, generated a COD rise of 64.7 and 94.2% after 10 and 40 min of EC treatment, respectively. This adverse effect of H_2O_2 was rather unexpected

by a biological treatment stage. Having an agreement on which process or processes to be proper or the best option is not an easy subject since all those above-mentioned processes have their advantages and drawbacks to be evaluated on the basis of the individual process. From the view point of engineering, it is almost the best optimization strategy by means of economics. However, "sustainability" should be evaluated with multi-criteria such as cost of area, investment costs, energy consumption, visual impact, sludge production, smells production, noise, removing nutrients, conventional parameters, and emerging contaminants, possibility of effluent reuse, carbon footprint, management complexity—presence of skilled personnel, maintenance, decentralization, public acceptability, flexibility, durability, promotion of sustainable development, and replicability with regards to local conditions.

One can conclude easily that aerobic processes are favored to operate in the case of fluctuating water quality. Among various biological processes, RBCs with their technological simplicity and short retention time could provide an effective pre-treatment before municipal sewage or to manage the peaks of high COD and acidity during the vintage season, although their efficiency is not comparable to that obtained with anaerobic digesters or activated sludge reactors [11]. Furthermore, to operate this system at high volume could be difficult too. It is explained above that anaerobic processes could operate better as pre-treatment before an aerobic stage.

Being a progressive subject to find a general wastewater management scheme for WWs, biomembrane (MBR) is still being accepted an innovative technology with various favorable reasons, in particular, low sludge production with a high effluent quality and lowered operational costs [10,38]. According to the results of Valderrama et al. [38], MBR can achieve high removal efficiencies in WW treatment and the effluent potentially complies with most of the quality specifications defined by international guidelines and regulations for water reuse and reclamation. Furthermore, the total annual cost of €0.36–0.40 per m^3 treated wastewater for MBR treating WW results to be very close to the co-treatment price of Bolzonella et al. [21] who monitored a full-scale activated sludge wastewater treatment plant receiving both municipal and WWs. They estimated costs as €0.2–0.3 per m^3 of treated wastewater.

Meanwhile, development of low-pressure reverse osmosis (RO) processes has made them an attractive alternative for the treatment of wastewater since they offer high fluxes and solute separations, and can operate over wide temperature and pH ranges [65]. It will also be a good solution for the removal of metals, in most cases especially zinc and copper that did not comply with the discharge limits as reported by Andreottola et al. [24]. Nevertheless, it is well known that membrane separation processes do not really destruct the pollutants, but merely concentrate them into smaller volumes of wastewater. The concentrate contains high levels of refractory organic pollutants and inorganic salts and represents one of the main drawbacks of this technology. When discharged without appropriate treatment, these streams can cause serious ecotoxicological effects. Recently, the use of solar photo-Fenton oxidation (simulated solar light) to treat the concentrate from the RO process proved to be a successful combined process for the integrated treatment of winery effluents [65].

The increasing shortage of water resources in arid zones where grapes are usually grown attracted the interest in reuse of treated WW too [18] while, anyhow, treated effluent quality should be accurately evaluated before irrigation purpose to avoid any negative interaction with the structure of plant and soil.

5. Conclusions

This paper reviewed the options for treatment technologies more appropriate for WW. It has been remarkable that all singular described processes pose advantages and drawbacks to overcome pollution control of WW. High organic load and changing characteristics make these wastewaters choose a mill-specific treatment option. It is for this reason that the environment-friendly and cost-effective combined treatment processes have to be evaluated to attain high removal efficiency in order to meet discharge limits.

Discharge to the end-pipe municipal wastewater treatment plant following an on-site pre-treatment, chemical, or anaerobic treatment that would not need a significant nutrient addition would be an optimized solution to treat WW when the winery mill is relatively close to settlements.

On the other hand, it has been underlined as MBR is the present promising technology to recover treated water for irrigation on site, while future trend technologies such as RO and photo-Fenton combination need to be extended to full-scale applications to optimize WW treatment options as well as costs.

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