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Review

Treatment wetlands and phyto-technologies for remediation of winery effluent: Challenges and opportunities

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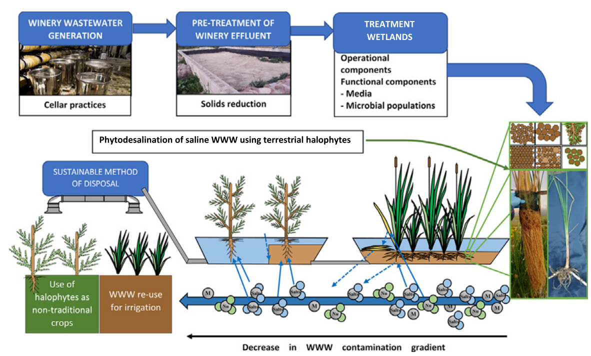
HIGHLIGHTS

- Review of WWW remediation by TWs relative to bio-hydrogeochemical components
- Salinity impacts inorganic remediation by TWs and wastewater reuse.
- Review presents case for novel application of terrestrial halophytes.
- Halophytes are viable alternatives to macrophytes for saline wastewater treatment.
- Review describes solutions to challenges impacting sustainability of wine industry.

GRAPHICAL ABSTRACT



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ABSTRACT

The composition and concentration of contaminants present in winery wastewater fluctuate through space and time, presenting a challenge for traditional remediation methods. Bio-hydrogeochemical engineered systems, such as treatment wetlands, have been demonstrated to effectively reduce contaminant loads prior to disposal or reuse of the effluent. This review identifies and details the status quo and challenges associated with (i) the characteristics of winery wastewater, and the (ii) functional components, (iii) operational parameters, and (iv) performance of treatment wetlands for remediation of winery effluent. Potential solutions to challenges associated with these aspects are presented, based on the latest literature. A particular emphasis has been placed on the phytoremediation of winery wastewater, and the rationale for selection of plant species for niche bioremediatory roles. This is attributed to previously reported low-to-negative removal percentages of persistent contaminants, such as salts and heavy metals that may be present in winery wastewater. A case for the inclusion of selected terrestrial halophytes in treatment wetlands and in areas irrigated using winery effluent is discussed. These are plant species that have an elevated ability to accumulate, cross-tolerate and potentially remove a range of persistent contaminants from winery effluent via various phytotechnologies (e.g., phytodesalination).

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Contents

1. Introduction

The wine industry contributes significantly to the global economy, with a steady increase in the trade value of this product of 25.8 to 31.4 billion € between 2014 and 2018, and a production volume ranging between 2.49 and 2.92 × 10^{10} L over the same period (OIV, 2019). Concurrent with wine-making, the industry generates large amounts of potentially hazardous solid and liquid 'wastes' from various cellar activities (Bolzonella et al., 2019; Bordiga et al., 2019). In the move towards a circular economy, a biorefinery approach has been adopted in many countries for the valorization of grape pomace and other forms of solid residues (Holtman et al., 2018; Khan et al., 2015). However, apart from the beneficial use of winery wastewater (WWW) of adequate quality for crop irrigation (i.e. beneficial irrigation), it is still largely viewed as a form of waste rather than a resource (Holtman et al., 2018). Consumer pressure is driving wineries to adopt best practice measures in cellars, including those that simultaneously reduce the quantity and mitigate the toxicity of WWW (Conradie et al., 2014). Cellars can reduce potable water usage and the amount of solids in the effluent by introducing best available technologies and cleaner production methods (e.g. re-using treated effluent for basic cleaning activities and installing filters to retain solids) (Bolzonella et al., 2019; Welz et al., 2016). Despite these efforts, WWW generally requires some form of primary physicochemical and/or secondary treatment in order to improve the quality to comply with legislated standards before discharge.

The effluent produced by wineries contains a range of organic and inorganic constituents, with considerable inter- and intra-site qualitative and quantitative variability (Bolzonella et al., 2019; Welz et al., 2016; Howell and Myburgh, 2018). Although the volume of WWW differs for each winery, studies conducted between 2003 and 2016 provide a rough estimate of 2.86 L (average) of effluent generated from each L of wine produced (Bolzonella et al., 2019; Mosse et al., 2011; Vlyssides et al., 2005), translating to a global total of approximately 7.5 × 10^{10} L per annum.

The extent and type of treatment that is required depends on the means of discharge (i.e. directly to sensitive environments, subsurface discharge, beneficial irrigation, or discharge to municipal sewage systems) (Bolzonella et al., 2019; Holtman et al., 2018; Johnson and Mehrvar, 2020). Multiple factors need to be considered when opting for any particular WWW treatment system. These include the (i) volume and site-specific character of the effluent that is generated, (ii) availability and value of land, (iii) size of the winery, (iv) availability of skilled operators, and (iv) capital and operational costs (Bolzonella et al., 2019; Howell and Myburgh, 2018).

For smaller wineries that have sufficient land available, but do not have the skills, time, or capital to install and operate more sophisticated systems, treatment wetlands (TWs) are a viable option. Data is available for systems that have been piloted or operated at full scale for WWW remediation in a number of countries, including Greece (Akratos et al., 2020), Portugal (Calheiros et al., 2018), Spain (Flores et al., 2019; Serrano et al., 2011), Italy (Rizzo et al., 2020), France (Kim et al., 2014), the USA (Grismer et al., 2003; Shepherd et al., 2001a; Shepherd et al., 2001b), Mexico (Grismer and Shepherd, 2011), South Africa (Holtman et al., 2018; Mulidzi, 2010), and Canada (Johnson and Mehrvar, 2020; Rozema et al., 2016).

This review identifies the status quo and challenges associated with the (i) characteristics of WWW, (ii) functional components of TWs, (iii) operational parameters of TWs, and (iv) TW performance. Furthermore, opportunities for overcoming these challenges are discussed. A particular emphasis is placed on phytoremediation of WWW, and the rationale for selection of plant species for niche bioremediatory roles.

2. Winery wastewater

Winery wastewater is generated from wine-making processes, including pressing, fermenting, clarification, storage, and bottling (Bolzonella et al., 2019; Vlyssides et al., 2005; Buelow et al., 2015). In terms of quality, many studies report on basic WWW characteristics as an adjunct to the primary aim/s of the research, such as wastewater treatment system performance. Such data may be skewed as criteria are typically limited to once-off batches and/or one site. Results from four comprehensive seasonal WWW characterization studies from multiple wineries in wine-making areas in Spain, France, Portugal, the USA and South Africa are summarized in Table 1. The variability in organic concentration in WWW is reflected in the chemical oxygen demand (COD) measurements, with inter-study averages of around 3500 mg L^{-1} to 7700 mg L^{-1} both in and out of the crush (vintage) periods, but with minimum and maximum values ranging from <100 mg L^{-1} to >70,000 mg L^{-1} being reported (Table 1).

Table 1
Seasonal composition of winery effluent (adapted from (Flores et al., 2019)[a] (Welz et al., 2016)[b] (Buelow et al., 2015)[c] (Malandra et al., 2003)[d]).

Parameter	Crush			Non-crush		
	Minima	Maxima	Averages	Minima	Maxima	Averages
pH	4[a] 4[b] 4[c] 4[d]	7[a] 12[b] 13[c] 5[d]	–	7[a] 2[b] 4[c]	8[a] 12[b] 11[c]	–
EC (mS·cm^{-1})	0.37[c]	9.7[c]	1.3[c]	0.14[c]	3.3[c]	0.97[c]
TSS (mg·L^{-1})	523[a]	2190[a]	1237[a]	<200[a] 267[b]	<1000[a] 21,697[b]	2382[b]
COD (mgO$_2$·L^{-1})	1031[a] 54[b] 320[d]	16,825[a] 13,900[b] 5760[d]	7684[a] 5951[b] 3449[d]	<500[a] 28[b]	<2000[a] 76,900[b]	5489[b]
BOD$_5$ (mgO$_2$·L^{-1})	650[a] 6[c]	10,300[a] 15,400[c]	3542[a] 1790[c]	<250[a] 4[c]	<1000[a] 4100[c]	1390[c]
VFA (mg·L^{-1})	7[b] 1[d]	308[b] 799[d]	123[b] 216[d]	0[b]	11,254[b]	830[b]
Ethanol (mg·L^{-1})	0[b]	1240[b]	338[b]	0[b]	21,000[b]	1693[b]
Sugars (GFMmg·L^{-1})	0[b] 0[d]	1474[b] 4940[d]	195[b] 2006[d]	0[b]	733[b]	33[b]
TPhen. (mg·L^{-1})	5[b] 0[d]	86[b] 27[d]	46[b] 9[d]	3[b]	273[b]	50[b]
Sodium (mg·L^{-1})	8.6[b] 7.8[c]	105[b] 3060[c]	31[b] 137[c]	5.9[b] 5.4[c]	574[b] 714[c]	61[b] 87[c]
Potassium (mg·L^{-1})	5.8[b] 3.2[c]	387[b] 772[c]	187[b] 176[c]	1.7[b] 1.3[c]	393[b] 1270[c]	162[b] 133[c]
TN (mg·L^{-1})	9.7[a] 0.7[b]	109[a] 34[b]	43[a] 10[b]	<20[a] 0.4[b]	<100[a] 176[b]	14[b]
TP (mg·L^{-1})	1.5[a] 0.4[b]	188[a] 31[b]	6.9[a] 11[b]	<10[a] 0.5[b]	<50[a] 280[b]	19[b]
COD:N	–	–	146:1[a] 595:1[b]	–	–	392:1[b]
COD:P	–	–	912:1[a] 541:1[b]	–	–	289:1[b]

Data from studies using respectively unknown [a], 168[b], 360[c], and 9[d] samples from 6[a], 4[b], 18[c], and 9[d] wineries.
EC = electrical conductivity TSS = total suspended solids COD = chemical oxygen demand BOD$_5$ = five-day biological oxygen demand VFA = (short chain) volatile fatty acids GFM = glucose + frutose + maltose TPhen. = total phenolics SAR = sodium adsorption ratio TN = total nitrogen TP = total phosphorous.

The composition and concentrations of organics, which may include alcohols, organic acids, sugars and phenolics, are dependent on the seasonal wine-making processes: for example, ethanol constitutes the highest fraction in the non-crush period, while significant concentrations of grape sugars are generally only produced during periods of crushing (Welz et al., 2016; Malandra et al., 2003). In terms of amenability to secondary remediation technologies, the organics range from being highly biodegradable (e.g. sugars) to more slowly biodegradable or even recalcitrant (e.g. phenolics) (Welz et al., 2016; Mutabaruka et al., 2007). Phenolics also differ in their degree of toxicity to microbes and plants, a factor that requires consideration, particularly for WWW treatment options such as planted TWs (Arienzo et al., 2009; Chen et al., 2009).

In general, if the ratio of COD to nitrogen to phosphorus (COD:N:P) in WWW (Table 1) is higher than 100:5:1, wastewater may be supplemented with nutrients to enhance the biodegradation of organics for efficient secondary wastewater treatment (Welz et al., 2016; O'Flaherty and Gray, 2013). The WWW may also be treated concurrently with domestic wastewater (Ganesh et al., 2010), but the potential introduction of pathogens, such as multi-drug resistant bacteria, may render the effluent unsuitable for land application.

Inorganics in WWW emanate from (i) cleaning and sanitizing agent/s [including sodium hydroxide (NaOH), potassium hydroxide (KOH), sodium metasilicate (Na_2SiO_3), trisodium phosphate (Na$_3$PO4), sodium carbonate (Na_2CO_3), phosphoric acid (H_3PO_4) quaternary ammonium compounds, peracetic acid, hydrogen peroxide (H_2O_2), and sulfur (S)], (ii) filtration and clarification aids (e.g. diatomaceous earth, bentonite clay, perlite), and (iii) grapes (Conradie et al., 2014; Mosse et al., 2011; Lucas et al., 2010; Versari et al., 2014). Grape residues and juice can contribute appreciably to the potassium (K^+) concentration whereas the use of cleaning and sanitizing products are the major contributing factors to the introduction of sodium (Na^+) in WWW as NaOH is still the agent of choice for many wineries due to its effectiveness and low cost (Conradie et al., 2014; Mosse et al., 2011). Salts are not removed during secondary treatment processes, and monovalent cations, particularly Na^+, can lead to soil degradation (via increasing soil sodicity) if the WWW is used for irrigation purposes (Rengasamy and Marchuk, 2011). Best practice therefore advocates the use of K-based, or ideally, organic cleaning and sanitizing products (Rengasamy and Marchuk, 2011).

3. Operation and performance

A literature search (SCOPUS) using the keywords "winery, wastewater, effluent, treatment, wetland, constructed" was used to identify studies pertaining to treatment of synthetic and/or real winery effluent in treatment wetlands. Selected data obtained from all the studies that assessed and monitored the physical set-up, operation, and performance of TWs for the remediation of WWW are provided in Tables 2 and 3. Data may be cross-referenced between these tables using the numbers in the left hand (ID) column. Additional information for these studies is provided in Table S1.

3.1. Lack of data and experimental flaws

It is difficult to holistically evaluate which of the existing TW systems are the most effective options for remediation of WWW by comparing the published data. While it is not possible to standardize factors such as the character of WWW or climatic variables, other fundamental data required to allow meaningful comparisons between systems are often omitted from manuscripts (Table 2). These include the functional surface area (FSA), functional depth, hydraulic loading rate (HLR), organic loading rates (OLR), and measured flow rates (MFR). These can exhibit notable variations, for example, the data available for the HRT range from 1.8 to 24 days. In addition, although the FSA is an important parameter for calculating the spatial footprint of a TW, the standard use of this parameter to describe the HLR in TWs should ideally be augmented by the use of the functional volume (FV), which is more informative when performing inter- and intra-study comparisons of systems containing, for example, different types of media. This is because the saturation level (depth) also varies between systems, in this instance from 0.3 to 1.2 m (Table 2). For example, if a shallow system is compared with a deep system using only the FSA to calculate the HLR and/or OLR, it will give a skewed picture of the contribution of the media to the overall remediation potential of the studied TW (Holtman et al., 2018).

In order to account for the random variability inherent in biological systems, experimental replication is required to ensure the statistical validity of studies comparing different factors (Casler et al., 2015). Replication is more feasible at laboratory-scale, but most studies reported in literature have been conducted using full-scale TWs (Table 3). For TWs treating WWW, a limited number of laboratory studies have been used to compare the presence/absence of plants (Akratos et al., 2020; Grismer and Shepherd, 2011) and/or media (Skornia et al., 2020). However, in all but one instance (Grismer and Shepherd, 2011), experiments were not replicated. This compromises the reproducibility and practical application of studied TWs as the 'snapshot' of the TW's remediation efficacy may be an anomaly compared with a standardized and expected outcome. Going forward, this highlights the need to consider performing well-replicated laboratory studies to improve the performance of TWs treating WWW.

Table 2
Selected operational and performance data of studies pertaining to treatment wetlands used to remediate winery wastewater.

ID	FSA (m^2)** [Depth (m)]	HRT [D] (days)	HLR – FSA** (mm·day^{-1})	HLR – FV** (L·m^{-3}·day^{-1})	COD$_{in}$ (mg L^{-1})	OLR - FSA (g COD·m^2·d^{-1})	OLR - FV (g COD·m^3·d^{-1})	ORR (%)	MFR (m^3·day^{-1})	Ref
1	0.48 [0.35]	2-4	UTC	UTC	788-2985	UTC	UTC	81-92	NG	(Akratos et al., 2020)
2	1.2 [0.6]	NG	UTC	UTC	1258	UTC	UTC	NG	NG	(Calheiros et al., 2018)
3	350* [0.3-1.4]	NG	20 ± 7 (VF&HF) 13-25 (HF$_{1,2,3}$)	14 (VF) 43-82 (HF$_1$) 22-41 (HF$_{2,3}$)	1558 ± 1023	30.4 ± 19.3 (all) 213 (VF) 16.2 (HF$_{1,2,3}$)	152* (VF) 54* (HF$_1$) 27* (HF$_{2,3}$)	71*	5.2	(De la Varga et al., 2013a; De la Varga et al., 2013b)
4	350* [0.3-1.4]	3	77-215 (VF) 13-36 (HSSF$_{1,2,3}$)	55-154* (VF) 43–120* (HF$_1$) 22–60* (HF$_{2,3}$)	NG	30.4 (all) 43–466 (VF) 3.6–55 (HF$_{1,2,3}$)	31–333* (VF) 12–183* (HF$_1$) 6.0–92* (HF$_{2,3}$)	73 (all) 29–70 (VF) 23–79 (HF)	NG	(Serrano et al., 2011)
5	60 [1–1.2]	NG	25.7 (VF&HF)	25.7 VF* 15.4 HF*	1665 VF NG (HF)	162 VF 65 HF	162.0 VF* 39.2 HF*	47–96 VF 7* HF	1.28 ± 1.18	(Flores et al., 2019; Pascual et al., 2021)
6	14.9 [0.9]	9.7	34	28*	993–4720	35–164*	37–176*	97–99	0.5	(Shepherd et al., 2001a; Shepherd et al., 2001b; Grismer et al., 2001)
7	4400 [1.2]	5.5 [10]	31*	26*	7406 ± 2090 (C) 1721 ± 439 (NC)	120–270	100–225*	49 (C) 79 (NC)	137	(Grismer et al., 2003)
8	304 [1.2]	[5]	UTC	UTC	290	553	465*	98	137	(Grismer et al., 2003)
9	NG	2.5–5.0	UTC	UTC	1183	UTC	UTC	49–70	NG	(Grismer and Shepherd, 2011)
10	120* [0.9]	6 ± 1.6 [14]	UTC	UTC	72,965 ± 29,066	UTC	UTC	94–97	NG	(Grismer and Shepherd, 2011)
11	144* [0.9]	18–24 [14]	UTC	UTC	5080 ± 1211 (C)	UTC	UTC	98–99	NG	(Grismer and Shepherd, 2011)
12	7.3* [NG]	1.8	109*	150 (12−313)	1138	110*	152 (23–469)	79 (28–98)	0.41	(Holtman et al., 2018; Welz et al., 2018a)
13	75–164 [1.2]	NG	119–160 (all) 357–480 (HF$_1$)	99–133*	NG	NG	UTC	>98	10–23	(Johnson and Mehrvar, 2020)
14	190–404 [1.2]	NG	39–53 (all) 133–164 (HF$_1$)	33–44*	NG	NG	UTC	>98	10–17 (C)	(Johnson and Mehrvar, 2020)
15	NG	3–5	38 (all) 76 (HF$_1$)	32*	NG	NG	UTC	>98	< 10.0	(Johnson and Mehrvar, 2020)
16	1330* [0.7]	NG	26	18*	4045	329	230*	98	35	(Masi et al., 2002)
17	752* [0.7]	NG	23	16*	1003	236	165*	93	10	(Masi et al., 2002)
18	215 [0.7]	NG	37	26*	722	352	246*	88	8	(Masi et al., 2002)
19	3034* [0.4–0.85]	[2.5]	60–80	71–94* (VF)	1159 ± 432 (VF)	160–230 (D)	188–271*	98 (all) 70 (VF)	61 ± 28 (max. 118)	(Rizzo et al., 2020)
20	1.63 [0.6]	9.6	31* (SA)	51*	171 ± 14 184 ± 11	5	8.8* 9.4*	69–93 86–93	0.050	(Mena et al., 2009)
21	180 [0.9]	1) 14 2) 7	1) 23* 2) 45*	1) 25* 2) 50*	1) 14000 2) NG	1) 315* 2) NG	1)350* 2) NG	1) 77–80 (NC) 1) 83–88 (C) 2) 60	1) 4.1 2) 8.1	(Mulidzi, 2010; Mulidzi, 2007)
22	404 [0.4–0.8]	NG	22.3	56* (VF$_{1,2,4}$) 28* (VF$_3$)	3043 (C) 2117 (NC)	34	85*(VF$_{1,2,4}$) 43*	99	7.3* (C) 11* (NC)	(Rozema et al., 2016)
23	NA [0.75]	14	6.9	9.2*	4997–6189	5.18	6.9*	>99	NG	(Skornia et al., 2020)

FWS = free water surface. FSA = functional surface area D = design FV = functional volume HRT = hydraulic retention time HLR = hydraulic loading rate COD$_{in}$ = influent chemical oxygen demand OLR = organic loading rate ORR = organic removal rate MFR = measured flow rate UTC = unable to calculate (from literature data) NG = not given C = crush season NC = non-crush season.

* Calculated from literature data

** FWS data not included.

Table 3
Selected operational data and details of functional components of treatment wetlands treating winery wastewater (plants excluded).

ID	Study type & duration	Wine/effluent production per annum	Wastewater type/discharge	Pre-treatment	Configuration & mode	Media			HC ($mm \cdot s^{-1}$)	Ref.
						Material	Diameter (mm)	Porosity (%)		
1	Lab study 2 years	NA	Diluted WWW NA	PS → TF	HF	Igneous fine gravel Bulgarian zeolite	6 (D_{50}) 4 (D_{50})	35 25	–	(Akratos et al., 2020)
2	Pilot study 2 weeks	6000 bottles	WWW NG	–	HF	Cork stoppers	3–7	44	–	(Calheiros et al., 2018)
3	Full-scale 2 years	315 m^3	WWW & DWW Municipal sewer	UASB	VF → $HF_{1,2,3}$	Granitic gravel Gravel	8–16 (T,B) 3–6 (M) 6–12	NG 40	- 1.5–1.6	(De la Varga et al., 2013a; De la Varga et al., 2013b)
4	Full-scale 2 years	315 m^3	WWW & DWW Municipal sewer	UASB	VF → $HF_{1,2,3}$	Granitic gravel Gravel	8–16 (T,B) 3–6 (M) 6–12	NG 40	- -	(Serrano et al., 2011)
5	Pilot 2 years	368 m^3/ 1398 m^3	WWW Recirculated	Homogenization tank HUSB	VF → HF →	Granitic gravel Sand Gravel	6–12 (B), 2–4 (M) 1–2 (T) 6–12	NG	–	(Flores et al., 2019; Pascual et al., 2021)
6	Pilot study 2 years	18,200 m^3	WWW NG	Sand pre-filter	HF	Pea gravel	4.7 (D_{50})	36	6.0	(Shepherd et al., 2001a; Shepherd et al., 2001b; Grismer et al., 2001)
7	Full-scale 1 year	Moderately sized winery	WWW Irrigation	Settling pond	HF	Pea gravel	4.0	NG	–	(Grismer et al., 2003)
8	Full-scale 1 year	Small winery	WWW Irrigation	Settling pond	HF	Rock	NG	NG	–	(Grismer et al., 2003)
9	Lab study 30 days	NA	WWW NA	NA	HF & FWS	Volcanic rock	40	50	–	(Grismer and Shepherd, 2011)
10	Full-scale NG	14,000 cases	WWW Irrigation	Septic tank	HF	Pea gravel	<8	NG	–	(Grismer and Shepherd, 2011)
11	Full-scale NG	14,000 cases	WWW Irrigation	Septic tank	HF	Pea gravel	<8	NG	–	(Grismer and Shepherd, 2011)
12	Pilot 3 years	Small winery	WWW Irrigation	Primary settling →balancing tank	HF	Dune sand	0.4 (D_{50}) 0.2–1.0	29	0.04–0.20	(Holtman et al., 2018; Welz et al., 2018a)
13	Full-scale (4) NG	NG	WWW & sewage Sub-surface	Septic tank →AFFR	HF_1 (anoxic) $HF_{2,3}$ (aerobic)	Wood chips Gravel & sand	NG NG	NG NG	–	(Johnson and Mehrvar, 2020)
14	Full-scale (3) NG	NG	WWW & sewage Sub-surface	Septic tank →AFFR	$HF_{1,2,4}$ (aerobic) HF_3 (anoxic)	Gravel & sand Wood chips				(Johnson and Mehrvar, 2020)
15	Full-scale (20) NG	NG	WWW Sub-surface	Septic tank	$HF_{1,2}$	Gravel & sand				(Johnson and Mehrvar, 2020)
16	Full-scale 1 year	NG	WWW Irrigation	Imhoff tank	HF → FWS	Gravel	5–10	NG	–	(Masi et al., 2002)
17	Full-scale 1 year	NG	WWW & DWW Irrigation	Imhoff tank	$VF_{1,2}$ → HF → FWS → pond	Gravel & sand Gravel (HSSF)	8–40 (T-B) 8–12	NG	–	(Masi et al., 2002)
18	Full-scale 1 year	NG	WWW Water body	Imhoff tank	HF	Gravel	5–10	NG	–	(Masi et al., 2002)
19	Full-scale 2 years	Bottling and aging only	WWW (no crush) River	Equalisation tank	VF → HF → FWS	Gravel NG	2–40 (T-B)	NG 35 (HSSF)	- 5.8	(Rizzo et al., 2020)
20	Pilot 5 months	NG	SWW	NA	HF	Gravel	6–9	NG	–	(Mena et al., 2009)
21	Pilot 2 years	NG	WWW Cabbage irrigation	NG	HF	Dolomitic gravel	20–30	35	–	(Mulidzi, 2010; Mulidzi, 2007)
22	Full-scale 6 years	NG	WWW & DWW Subsurface	Dosing tank	$VF_{1,2,3}$ VF_4 (anoxic)	Gravel Wood chips Peat moss (T) 0.3 m	5–10 NG	NG NG	–	(Rozema et al., 2016)
23	Lab column 6 months	Lab study	WWW NA	Primary settling	VF replicates	Gravel Clinopatilolite Tyre chips Oyster shells	6.4 1.2–2.4 10–15 1.2–2.4	NG	NG	(Skornia et al., 2020)

HC = hydraulic conductivity PS = primary settling TF = trickling filter HF = horizontal (subsurface) flow WWW = winery wastewater NA = not applicable NG = not given VF = vertical (subsurface) flow FWS = free water surface UASB = upflow anaerobic sludge blanket reactor AFFR = anaerobic fixed film reactor T = top M = middle B = bottom DWW = domestic wastewater HUSB = hydraulic upflow sludge blanket reactor FWS = free water surface flow SWW = synthetic winery wastewater.

It has been suggested that because of the relatively large spatial footprint of TWs, they are not suited to large wineries that produce copious amounts of effluent, particularly if land values are high. However, the connection between the size of a winery (small, medium, large) and the capacity of a TW system is difficult to establish. Ideally, the volume of effluent should be measured. For a number of reasons, this is practically difficult, which is evidenced by the fact that the effluent volume was only provided in one of the twenty studies included in this review (Table 3). Wineries are alternatively described by the amounts of grapes crushed or wine produced (bottles/cases or volume), but it is difficult to define the size of a winery based on these criteria as some wineries do not bottle, while others do not crush.

3.2. Performance evaluation

Most studies have focused on the universal objectives of removal/reduction of organics and solids from WWW, and pH neutralization (Tables 2 and 3). For reference, the influent COD (COD_{in}) and OLR in terms of FSA and FV, as well as the organic removal rates (ORR) are given in Table 2. These results are not discussed further because the organic removal performances of most of the TWs described in this manuscript have already been adequately reviewed (Johnson and Mehrvar, 2020; Masi et al., 2015). In summary, the authors of these reviews concluded that TWs are able to reduce the organic load sufficiently, provided that the HRT is sufficient to maintain the OLR within design parameters, and that pre-treatment for solids reduction is satisfactory. In addition, TWs have proven to be effective at reducing the total suspended solids (TSS) concentration, increasing the pH of acidic WWW, and decreasing the pH of alkaline WWW (Holtman et al., 2018; Masi et al., 2015; De la Varga et al., 2013a). The pH buffering mechanism/s of WWW in TWs has not yet been elucidated.

Due to potential environmental toxicity, removal of (poly)phenolics from WWW in TWs is an important consideration. However, studies typically limit analysis of organic removal to COD and/or BOD removal, and to our knowledge there are only four studies describing removal of phenolics or tannins from WWW in TWs or simulated TWs. Grismer et al. (2003) found higher removal rates during the non-crush season (average 78%; influent = 55 ± 16 $mg \cdot L^{-1}$) compared with the crush season (average 46%; influent 55 ± 22 $mg \cdot L^{-1}$). The reduced removal rates correlated with a decreased HRT during the crush season. In another system operated with a more consistent HRT, no seasonal trend in phenolic removal rates was noted, and an average removal rate of 77% (range 5.1–44 $mg \cdot L^{-1}$) was obtained. Other studies were conducted using WW with low phenolic concentrations: WWW mixed with domestic WW (9.7 ± 3.0 $mg \cdot L^{-1}$) and synthetic WWW (13 $mg \cdot L^{-1}$) (De la Varga et al., 2013b; Mena et al., 2009). Studies have also been conducted using other forms of agri-industrial wastewaters at moderate influent phenolic concentrations. Although the plant-based phenolic profiles would be expected to differ from those of WWW, these studies can still provide some valuable insights. In gravel-based horizontal flow systems, (Rossman et al. (2012) found that pre-aeration and inclusion of rye grass increased the removal of total phenolics from coffee processing wastewater from 54% to 72%, while Gomes et al. (2018) found that phenolic removal rates decreased when loads in cork boiling effluent exceeded 0.4 $mg\ L^{-1} \cdot day^{-1}$, and that some phenolic molecules were more readily removed than others.

Typically, less importance is placed on the removal of inorganics in biological systems treating WWW (Marchuk and Rengasamy, 2010). The need to remove nutrients and other inorganic elements and molecules from WWW is incumbent on local legislative requirements, place/type of discharge, and WWW character, and is therefore site-specific (Johnson and Mehrvar, 2020; Flores et al., 2019; De la Varga et al., 2013a; De la Varga et al., 2013b). If the effluent is discharged into a sensitive aquatic environment or municipal sewer where nutrient discharge limits are set, then removal mechanisms such as nitrification/denitrification will be important and must be considered. Conversely, the essential plant nutrients, namely N and P, may be seen as such if the effluent is to be re-used for irrigation (Flores et al., 2019). In the case where treated WWW is utilized for irrigation purposes, it will be important to limit the Na^{+} concentration and sodium adsorption ratio (SAR) within permissible limits to prevent soil sodicity and salinization, and associated loss of soil structure (Marchuk and Rengasamy, 2010).

It has been clearly demonstrated that N removal takes place over the long-term operation of full-scale TWs treating WWW. However, as with the character of WWW itself, removal rates vary from site to site and seasonally. Masi et al. (2015) reported an elevated average total nitrogen (TN) removal in three full scale systems with influent concentrations of 15 $mg \cdot L^{-1}$, 27 $mg \cdot L^{-1}$, and 65 $mg \cdot L^{-1}$, and respective removal rates of 81%, 90% and 58%. As with tannins, Grismer et al. (2003) found lower nitrate (NO_3^-) removal rates in the crush compared with non-crush seasons (17% vs 73%, respectively), which could be attributed to reduced HRT during the crush season. Similarly, the authors also reported lower average NH_4^+ removal rates in samples taken during the crush period (average 29% of 37 $mg \cdot L^{-1}$ v/s 62% of 118 $mg \cdot L^{-1}$). Apart from the shorter HRT during the crush season, diminished nitrification may have been exacerbated by a more unfavorable C:N ratio (200:1 v/s 15:1), as nitrification rates decrease with increasing C:N ratios in TWs (Lai et al., 2020). De la Varga et al. (2013a) reported low NH_4^+ removal rates (≤ 29%) in a system treating combined WWW and domestic WW with influent concentrations of 0.6 to 74 $mg \cdot L^{-1}$ in Europe. In a Canadian system designed to treat the combination of WWW and domestic WW by enhancing nitrification-denitrification processes within the TW, elevated NH_4^+ removal rates (>99%) were achieved during the warmer half of the year (6-month season) for three years but decreased to as low as 19% thereafter. The removal rates were consistently >99% for the rest of the year. This result appears to be anomaly, as given the cold winter climate in Canada, it would be expected that nitrification would decrease during this period. The result may be attributable to low sampling frequencies ($n = 2$ per season) that may not have presented a true reflection of overall performance for each season, along with low influent concentrations, particularly in the colder seasons (average 2.2 $mg \cdot L^{-1}$ and 0.9 $mg \cdot L^{-1}$ in the warmer and colder seasons, respectively). Grismer et al. (2003) also measured removal of sulfur (S) species, and found 95% removal of sulfates (SO_4^{2-}) and 78% removal of sulfites (SO_3^{2-}) in a HF gravel-based system with influent concentrations of 35 ± 19 $mg \cdot L^{-1}$ and 0.56 ± 0.20 $mg \cdot L^{-1}$, respectively.

TWs do not appear to effectively remove cations from WWW. For example, Mulidzi (2010) found marginal, but erratic removal of Na^{+} (1–43%, average 12%; influent 101–282 $mg \cdot L^{-1}$; $n = 23$) and K^{+} (1–43%, average 8%; influent 141–615 $mg \cdot L^{-1}$; $n = 23$) in HF gravel-based systems containing plants (*Typha, Scirpus* and *Phragmites* spp.). Removal was attributed to plant uptake, which is supported by the fact that no removal of Na^{+}, K^{+}, or magnesium (Mg^{2+}) was found by Holtman et al. (2018) in an unplanted sand-based WWW treatment system. The selection of certain plant species with the ability to remove elevated concentrations of such contaminants may therefore be utilized in TWs to improve the quality of WWW used for irrigation and is further discussed in Section 4.3.

For interested readers, more detailed data on specific (poly)phenolic and inorganic concentrations and removal efficiencies in TWs, treating WWW, are included in Table S1. The biotic and abiotic mechanisms, present in TWs, for removal of important WWW organics and inorganics are described later in this manuscript.

4. Functional significance and interactions of major biotic and abiotic components

4.1. Background

As alluded to previously, the performance of TWs treating WWW has been reviewed by Masi et al. (2015) in 2015. However, in order to continually improve the efficiency of these systems, there is a need to critically analyse the current fundamental knowledge pertaining to the types of abiotic substrate (media) and plants, as well the microbial community structure and function which may be utilized in these TWs. The selection of plants is particularly important because of the potential phytotoxicity of WWW (Arienzo et al., 2009).

To address this, the relevance and characteristics of (i) the media, microbes, and media-microbe interactions, and (ii) the plants, and plant-media and plant-microbe interactions, are discussed generically, and as they specifically pertain to the treatment of WWW in the following two sub-sections.

4.2. Media and microbial populations

4.2.1. Types of media used in winery wastewater treatment wetlands

Various types of gravel, with diameters ranging from 3 to 40 mm, have historically been the preferred media used in TWs remediating WWW. Sand has also been extensively used, in combination with or without gravel (Table 2). Other substrates have also been used to achieve particular removal functions. For example, Johnson and Mehrvar (2020) used wood chips as a carbon (C) source for denitrification of WWW mixed with sewage, Calheiros et al. (2018) used cork for its general adsorptive capacity, while Skornia et al. (2020) added oyster shells as a buffering agent in a laboratory study.

4.2.2. Biotic and abiotic removal mechanisms

The primary biotic removal mechanisms in TWs are microbial biodegradation, biotransformation, and bioprecipitation (Table 4), and a series of different phytoremediatory mechanisms (Table 4, Section 4.3). These are accompanied by physicochemical (abiotic) mechanisms, most notably adsorption and precipitation (Marchuk and Rengasamy, 2010; Lai et al., 2020). Degradation of organic molecules can also be facilitated by physicochemical catalysis. For example, the oxidation of phenolic acids may be coupled to the reduction of iron and/or manganese present in the substrate particles (Polubesova et al., 2010; Welz et al., 2012).

Table 4
Phyto- and biotechnologies, used as tools for the biological remediation of various contaminants present in winery wastewater (WWW).

Bio- and phytotechnology	Description	Mechanism(s)
Microbial biotechnologies		
Bioprecipitation	Contaminants are precipitated out of solution by various mechanisms including the release of complexing agents by microorganisms, rendering the contaminant biologically inert.	Bioprecipitation of contaminants by reduction-oxidation (redox) reactions, intracellular assimilation, complexation, sorption, release of complexing agents (e.g. ligands), and biosorption to membranes
Microbial biodegradation	Bacteria release extracellular biodegradation enzymes that breakdown contaminants resulting in the release of water-soluble intermediates, CO_2, H_2O, and other metabolites.	Enzymatic biodegradation and mineralization. Extracellular biodegradation enzymes are released by bacteria to break down contaminants. Degradation products (water-soluble intermediates) are released into the environment and/or assimilated into bacterial cells. Bacteria (present within the biofilm) attach to the surface of the contaminant resulting in contaminant degradation and the release of CO_2, H_2O and other metabolic products.
Biomethylation	Transformation of contaminants by microbes (aerobic and anaerobic bacteria, and fungi) into volatile derivatives and can be removed from the TW system via evaporation.	Enzymatic transfer of methyl groups to metals (e.g. As, Hg, Pb, Se, and Sn). For example, microbes may methylate Hg (forming methylmercury, CH_3Hg) which accounts for one of the removal pathways from TWs. It must be noted that the volatile derivative, e.g. CH_3Hg, may be more toxic due to its lipophilic nature and therefore, its bioavailability for plant and animal uptake.
Plant-assisted biotechnologies		
Rhizodegradation	Plant-assisted bioremediation involving the complex interplay between plant roots, plant exudates, rhizosphere chemistry, and microbe species/communities to render contaminants biologically inert via microbial degradation processes.	Plant roots provide surface for microbe colonization. Plants stimulate the growth of microbial species/communities (via release of exudates such as organic acids), enhancing microbial bioremediation pathways (e.g. biodegradation and bioprecipitation).
Phytotechnologies		
Rhizofiltration	Sorption (ad- and absorption), bioconcentration, and precipitation of contaminants from wastewater by plant roots of aquatic and terrestrial plant species.	Phytoextraction and (hyper)accumulation of contaminants (e.g. heavy metals) in roots.
Phytodesalination	Use of halophytic plant species to remove salts, and/or their constituents, from contaminated land or water.	Halophytes hypertolerate (and cross-tolerate) and accumulate elevated concentrations of salts, and their constituents, due to their effective ion homeostasis networks [strict and integrated cross-talk between genetic (e.g. upregulation of similar genes), molecular (e.g. reactive oxygen species, ROS), cellular (e.g. compartmentalization of contaminants), and physiological (e.g. contaminant secretion from specialized glands on the surface of leaves) mechanisms].
Phytostabilization	Establishing plant cover to physically and chemically stabilize soil contaminants thereby mitigating soil and aeolian erosion, and contaminant leaching.	Release of exudates into the rhizosphere to chelate metals which may subsequently be ad/absorbed by plant roots and/or sequestered into the plant's rhizosphere (phytosequestration).
Phytoextraction	Use of plants to sequester (phytosequestration), take up, and accumulate elevated concentrations of contaminants *in planta*.	Once contaminants have been sequestered (via the release of chelating compounds) and taken up by plant roots, the plant may utilize avoidance (root-to-shoot restriction/shoot-to-root re-translocation) and/or tolerance (such as excretion from leaves, contaminant complexation, and translocation of contaminants to storage sites such as vacuoles) strategies to render toxic contaminants biologically inert. The number, and efficiency, of these strategies enable elevated concentrations of contaminants to be accumulated by particular plant species.
Phytohydraulics	Use of plants to intercept and prevent the horizontal migration of contaminants within surface and groundwater (including phytofiltration, and more specifically rhizofiltration).	Rhizosphere reverses the hydraulic gradient forming zone of stagnation which 'captures' contaminants thereby mitigating the transport of contaminants downstream (surface water) and within groundwater.

References: (Truu et al., 2019; Yan et al., 2020; Truu et al., 2015; Flowers and Colmer, 2015; Farzi et al., 2017; Lee and Yang, 2010; Saddhe et al., 2020; Chaudhry et al., 2005; Limmer and Burken, 2016; LeDuc and Terry, 2005; Dushenkov et al., 1995; Manousaki et al., 2008; McGrath and Zhao, 2003; Mendez and Maier, 2008; Pérez-Esteban et al., 2014; Sander and Ericsson, 1998)

Although adsorption and precipitation may be the primary removal mechanisms for some molecules and elements, there is a danger that these may later desorb or re-mineralize due to changes in physicochemical conditions, and/or the saturation of substrate (media) binding sites. In cases where concurrent biotic and abiotic removal takes place, good long-term removal rates may still be achieved, as demonstrated with the phenolics viz - gallic acid and vanillin present in synthetic WWW (Welz et al., 2012). This combined biotic/abiotic approach was also used by Skornia et al. (2020), where media was supplemented with clinopatilolite to adsorb ammonium ions (NH_4^+) (average 13.7 $mg \cdot L^{-1}$) and tyre chips to adsorb NO_3^- (average 4.2 $mg \cdot L^{-1}$) from WWW in gravel columns. The rationale for the experiment was that in colder climates where nitrification rates are low in winter, these forms of N could be adsorbed, and then mineralized to nitrogen gas (N_2) by nitrification-denitrification with the advent of warmer weather. While the approach had merit, the short-term study failed to show significant differences in removal rates between the amended columns and the gravel controls in this instance. In laboratory TW simulations testing systems containing different plants and media, Akratos et al. showed increases in NH_4^+ removal rates from domestic wastewater (Akratos and Tsihrintzis, 2007) and WWW (Akratos et al., 2020). In the latter, rates increased from 30% to 57% to 78% in unreplicated laboratory HF systems containing gravel, gravel and plants, and zeolite and plants, respectively. The authors attributed the increased nitrification to the presence of plants and greater NH_4^+ adsorption of zeolite when compared to igneous gravel.

In contrast to N, where primary removal in TWs is microbially mediated (nitrification-denitrification), and discounting removal by plants (discussed in Section 4.3), removal of P is abiotic (adsorption and/or precipitation) (Lai et al., 2020). Adsorption rates are dependent on the chemistry and morphology of the substrate particles, and can be increased by using highly reactive media such as apatite or steel slag (Akratos et al., 2020; Akratos and Tsihrintzis, 2007; Dell'Osbel et al., 2020; Korkusuz et al., 2005; Zhang et al., 2007). Although many studies have looked at P removal rates in gravel and/or sand filled TWs, in reality, unless significant plant uptake occurs and the plants are subsequently harvested, negative P removal rates may occur at some point due to plant senescence, saturation of binding sites and/or desorption or mineralization due to changes in physicochemistry (e.g. pH, redox, increased sulfate concentrations) (Maine et al., 2007; Dell'Osbel et al., 2020). While Masi et al. (2002) reported excellent total P removal rates over one (1) year in two (2) full scale systems in Europe: average 73% (influent 4.9 $mg \cdot L^{-1}$, $n = 5$) and average 94% (influent 1.9 $mg \cdot L^{-1}$, $n = 10$), Rozema et al. (2016) measured negative P "removal" after 5 years of operation (range: -115% to $>99\%$ $n = 23$) in a full-scale TW remediating WWW combined with domestic WW. Over the first 3 years of operation, >99% removal was achieved, suggesting that the primary reason for the decreased performance was saturation of P binding sites. It is therefore recommended that if P removal is seen as a priority, and in order to recapture this limited resource, specific downstream removal processes are applied. A promising example is provided by Skornia et al. (2020), whom used a proprietary iron-oxide based commercial P adsorbent (sponge) as a polishing step to precipitate and recover P from WWW. Such technologies that support a circular economy could be viable options going forward and should be investigated further.

4.2.3. Biofilm, solids and hydraulic conductivity

The main advantage of using sand instead of gravel in TWs is that the particles provide a larger surface area for biofilm attachment and surface chemistry (Akratos et al., 2020; Akratos and Tsihrintzis, 2007; Torrens et al., 2009). The major comparative disadvantage is the increased risk of clogging because of the smaller matrix pores in the sand milieu (Knowles et al., 2011; Pedescoll et al., 2011). The term 'clogging' in TWs is applied when the hydraulic conductivity (HC) decreases to the extent that the system can no longer function effectively. The risk of clogging can be ameliorated by the (i) popular use of WWW pretreatment (Table 2) to remove suspended solids, (ii) intermittent operation to allow solids and excess biofilm degradation during the resting period, and/or (iii) application of low organic loading rates such as effluent recycling (Torrens et al., 2009; Knowles et al., 2011; Prochaska et al., 2007; Tao et al., 2007).

While clogging with non-biodegradable solids can negatively impact the long-term operation of TWs, it should be noted that some critical decrease in the HC is expected to occur after start up due to the gradual, and necessary build-up of a functional biofilm (Truu et al., 2009; Welz et al., 2018a). Operational HC values of 1.5 $mm \cdot s^{-1}$ (De la Varga et al., 2013a), 5.8 $mm \cdot s^{-1}$ (Rizzo et al., 2020) and 6.0 $mm \cdot s^{-1}$ have been reported in gravel-based systems, and values ranging from 0.04–0.20 $mm \cdot s^{-1}$ (calculated from flow rates measured in-situ) were reported in a sand-based system (Welz et al., 2018a). It has been shown that predicted HC measurements based on grain size distribution may be inaccurate due to most models assuming particle sphericity (Welz et al., 2018a; Grismer et al., 2001). For example, Grismer et al. (2001) found an order of magnitude difference in the HC of poorly graded pea gravel between predicted values and column experiment measurements (6.0 $mm \cdot s^{-1}$).

Temporal changes in the HC of TWs treating WWW are rarely described in literature, making comparisons between systems difficult. Akratos et al. (2020) reported a 15% loss in porosity over 2 years in a HF system containing igneous fine gravel and Bulgarian zeolite, however the HC was not measured. De la Varga et al. (2013a) studied the accumulation of solids in a gravel-based system and found that, although the accumulated TSS and volatile suspended solids (VSS) increased annually to reach maximum values of 8.6 $\pm$ 3.0 kg and 1.01 $\pm$ 0.78 $kg \cdot m^2$, respectively after 2.8 years of operation, the average reduction in HC (1.64 $mm \cdot s^{-1}$ to 1.49 $mm \cdot s^{-1}$) measured by the falling head method was insignificant over three measurement periods (1.6, 2.2, and 2.8 years of operation). In contrast, reductions in HC of up to 50% have been measured in sand-based treatment systems treating WWW (Welz et al., 2018a), highlighting the need to monitor the HC in studies where sand is used as a physical substrate. If HC reductions are factored into the design capacity, and realistic flow rates can still be achieved, the use of sand as a medium may still be a viable option (Welz et al., 2018a). Furthermore, it has been shown that biofilm-related decreases in HC are inversely related to the organic loading rate, and are reversible (Tao et al., 2007; Welz et al., 2018a). Similarly, clogging due to accumulation of organic sludge formed during the crush season is also degraded during the non-crush period, restoring the HC (Masi et al., 2015; Masi et al., 2002).

4.2.4. Microbial community structure and function

The microbial community structure and function in CWs is influenced by the plant species, climatic variables, and physicochemical variables related to the substrate, mode of operation and type of wastewater (Ramond et al., 2012; Ramond et al., 2013a; Truu et al., 2019). In TWs, sedimentary and epiphytic bacteria, as well as planktonic bacteria (in FWS systems) are principally responsible for nitrification, denitrification, SO_4^{2-} oxidation/reduction and hydrocarbon degradation, as well as transformation and mineralization, which follow natural principles for the biogeochemical cycling of C, N and sulfur (S). Studies suggest that it takes around 100 days from start-up for the microbial communities to become established (equilibrated) in TWs (Ramond et al., 2012; Truu et al., 2019; Weber, 2016; Oopkaup et al., 2016). This is an important factor that is not always considered by researchers. In literature reports on TWs treating WWW, full-scale studies have been conducted over ≥2 years. However, the results of many lab- and pilot-scale experiments may not provide an accurate assessment of the remediation potential of the systems because the duration of the experiments was too short to allow effective establishment of the functional microbial communities (Table 3).

It has been shown that under the same climatic conditions and influent characteristics, even small physicochemical differences in the substrate play a significant selective role on the microbial community structure in unplanted TWs (biological sand filters) (Welz et al., 2014a). Even in particles with similar geochemistry, the shape of sand particles can affect biofilm attachment/abundance and consequent C and N removal performance; for instance, it has been demonstrated that natural sand presents a superior biofilm attachment surface to crushed sand (Torrens et al., 2009). Significant differences in the microbial community structure have also been noted in systems containing the same substrate, but with different WWW influent (Ramond et al., 2013a; De Beer et al., 2018; Ramond et al., 2013b). For example, in high rate biological contact reactors treating WWW and operated in series, De Beer et al. (2018) found that the higher pH and lower organic load achieved in the (identical) second reactor allowed the preferential selection of different bacterial and fungal species to the first reactor, including nitrifying and denitrifying bacteria. Similarly, different microbial communities are selected within spatial niches of TWs, with redox status and influent degradation gradients thought to be the primary selective drivers (Ramond et al., 2013a; Ramond et al., 2013b; Welz et al., 2014b). In biological sand filters, and, by inference, TWs receiving high C:N WWW, nutrient limitation may be naturally mitigated by selection of N-fixing bacteria such as *Azotobacter* spp. (Ramond et al., 2013a; Welz et al., 2018b). These results are supported by a recent study by Ospina-Betancourth et al. (2020), whom enriched high C:N effluent with *Azotobacter vinelandii* and achieved appreciable rates of N-fixation. The authors proposed the use of the treated waste as a high N organic fertilizer, which is potentially a novel option for beneficiation of WWW which supports circular economy principles.

4.3. Plants, and plant-microbe plant-media interactions

Phytoremediation (plant-based) and microbial bioremediation (e.g. bacteria- and fungi-based) processes in wetland systems have been demonstrated to effectively remediate a range of inorganic and organic contaminants present in wastewater originating from different sources (e.g. acid mine drainage, sewage, aquaculture, and to a lesser degree, WWW) (Muthusaravanan et al., 2018). Plants and/or microbes, used as tools for phyto- and biotechnologies, have the ability to degrade, take up, transform (e.g. methylate), volatilize, and/or stabilize (i.e. immobilize) contaminants (Table 4) (see Yan et al., 2020 for review). However, various factors, including plant, microbe, and plant-microbe interactions (e.g. plant-assisted bioremediation), physicochemical properties of WWW (e.g. bioavailability, octanol-water partition coefficient, competitive ion adsorption), and environmental factors (viz – pH, redox potential (E_h), salinity, and temperature/solar radiation) influence the efficacy of these remediation processes (Yan et al., 2020; Truu et al., 2015).

4.3.1. Plant factors

Plant species, as well as their genotypes, differ in their ability to remove inorganic and organic contaminants. This is attributed to the number, type, and efficacy of genetic, molecular, cellular, and physiological mechanisms, as well as the interplay between these mechanisms (Manara et al., 2020; Sytar et al., 2020). Phytotoxic concentrations of contaminants present in WWW have been demonstrated to impede plant growth and survival (Nagajyoti et al., 2010). However, as plants are sessile organisms, they have developed various strategies to adapt to abiotic and biotic stresses (Manara et al., 2020). This enables certain plant species with elevated phytoremediation potentials to be utilized in TWs to remediate targeted contaminants. Plants selected for TWs must possess elevated (i) growth rates, (ii) biomass production (especially well-developed root systems), (iii) degree of stress cross-tolerance, and (iv) ability to render targeted contaminants biologically inert via the extraction, volatilization, or sequestration of contaminants (Liang et al., 2017; Jesus et al., 2018). Based on these criteria, commonly selected plant species for TWs include *Phragmites australis* (common Reed), *Typha* spp. (cattails), and *Cyperus* spp. (sedges) (Table 5).

Unlike inorganic contaminants present in WWW, plants can metabolize organic contaminants. The metabolism of organics involves the transformation (e.g. redox reactions which alters the species of contaminant), conjugation (e.g. complexation with other molecules such as low-molecular weight thiols), compartmentalization (e.g. storage of contaminants in vacuoles), and/or volatilization (via evapotranspiration processes) of contaminants (Yan et al., 2020; Tripathi et al., 2020). These metabolic processes may render toxic organics biologically inert. Various studies have investigated the role of plants in TWs, for the removal of organic constituents from WWW, especially COD (*refer to Section 2; see Table S1 for extensive list of studies investigating the remediation of organics, present in WWW using TWs*).

Although numerous inorganic constituents, including salts and their constituents (e.g. Na^+, Cl^-, and heavy metals), have been investigated (Table S1), TWs are generally ineffective in remediating the inorganic fraction of WWW due to its inhibitory effect on biological processes. For example, Na^+ removal efficiency in wetland system ranges from −78 to 43%, of which Na^+ and other salt constituents (e.g. Cl^-) are considered as the most persistent and challenging contaminants to remove from wastewater (Johnson and Mehrvar, 2020; Akratos et al., 2020; Masi et al., 2015; De la Varga et al., 2013a). The removal of other inorganics, namely N and P, is also mediated by wastewater salinity, where the removal efficiency significantly decreases as salinity increases (Wu et al., 2008). This highlights the need to desalinate wastewater in order to effectively reduce the remaining inorganic fraction present in WWW (Li et al., 2018). Moreover, the presence of these inorganics at elevated, phytotoxic concentrations generally requires expensive forms of treatment, viz. - dilution or physicochemical treatment methods (e.g. electrodialysis, reverse osmosis, and/or ion exchange treatments), creating other environmental challenges such as the production of a concentrated brine requiring disposal. Furthermore, these treatment methods have various disadvantages which include the method's low resistance to fluctuating contaminant loading, inhibiting effect of salts on the microbial component present in biological reactors, and inability to effectively treat non-point source pollution (Liang et al., 2017).

The concentrations and composition of inorganics present in WWW, which fluctuates through space (e.g. geographic locations) and time (e.g. according to seasonal cellar activities), creates a selective pressure to utilize cross-tolerant plant species with an elevated ability to remove targeted inorganics. Moreover, WWW must be treated before its disposal as per regulatory guidelines where legislation on permissible inorganic concentrations for disposal differ between countries.

The use of halophytic plants for the remediation of the inorganic fraction of WWW presents an inexpensive and effective alternative to the use of dilution and physicochemical treatment methods. Halophytes are generally characterized as plants which can complete their life cycles in saline environments where salt concentration is greater than 200 mM NaCl (Flowers and Colmer, 2015; Flowers et al., 1986). The use of halophytes has gained increasing attention over the past decades due to their elevated ability to cross-tolerate a range of stresses (Manousaki and Kalogerakis, 2011; Nikalje and Suprasanna, 2018). Elevated Na^+ and Cl^- concentrations in saline soils induce osmotic and ionic stresses in plants – leading to secondary stresses (Fig. 1). Halophytes have evolved various mechanisms to tolerate salinity stress, and are associated with the controlled uptake and assimilation of Na^+, K^+, and Cl^- (Flowers and Colmer, 2008; Dassanayake and Larkin, 2017). These tolerance mechanisms (described below) enable halophytes to maintain homeostasis, preventing the toxic build-up of Na^+ ions, and potential antagonistic interactions between Na^+ and K^+ due to these cations sharing similar physicochemical properties (Wakeel, 2013; Flowers et al., 2019). Although Na^+ is preferentially taken up over K^+ by plants, the elevated capacity of halophytes to accumulate elevated concentrations of these persistent inorganics (consequently

Table 5
Commonly selected plant species for use in TW for the remediation of organic and inorganic contaminants present in winery wastewater (WWW).

Plant species	Reason for selection for WWW remediation	References
Phragmites australis	- Fast growth rate and high biomass productivity (ranges from 0.413 to 9.890 kg dry mass·m^{-2} per annum); - Tolerant to elevated organic and nutrient levels (determined thresholds include COD: 14000 mg·L^{-1}, TKN: 506 mg·L^{-1}, and TP: 95 mg·L^{-1}); - Elevated rate of evapotranspiration (e.g. implications for WWW dewatering); - Flood, salinity-, and metal-tolerant (e.g. Fe^{2+}, Zn^{2+}, and Ni^{2+}) and effectively removes certain metals such as chromium (Cr^{2+}); - Tolerant to range of pH levels (e.g. used for remediation of acid mine drainage and industrial effluent); - Cosmopolitan species with a widespread distribution (i.e. the ability to growth and survive in a range of environmental conditions) - Extensive root system	(Ayyappan et al., 2016; Bonanno and Giudice, 2010; Kumari and Tripathi, 2015; Peruzzi et al., 2009; Vymazal and Kröpfelová, 2011)
Typha spp. (including *T. latifolia*)	- Fast growth rate and high biomass productivity (exceeds 5.00 kg dry mass·m^{-2} per annum); - Tolerant to a range of abiotic stresses (salinity, organic matter, heavy metals, and nutrients); - Effectively remove suspended solids; - Effectively removes Cu^{2+} and Cd^{2+} and is more effective at removing certain contaminants (such as P, Na^{+}, Ca^{2+}, Mg^{2+}, Cu^{2+} and Fe^{2+}), compared with *P. australis;* - Tolerant to a range of pH levels; - Widespread distribution and colonizes anthropogenically-affected habitats.	(Maine et al., 2006; Sukumaran, 2013; Vymazal, 2020; Zingelwa and Wooldridge, 2009; Hadad et al., 2018)
Cyperus spp. (e.g. *C. alternifolius*)	- Fast-growing plant species with a dense root system and is easily propagated; - Utilized in TWs for remediation of organics, pathogens, nutrients, and heavy metals (e.g. Al^{3+}, Cd^{2+}, Cu^{2+}, Pb^{2+}, and Zn^{2+}); - Demonstrated to effectively reduce COD, BOD, TSS, NO_3^-, NH_3, PO_4^{3-}, as well as total coliforms and fecal coliforms.	(Sepúlveda et al., 2020; Cheng et al., 2002; Cui et al., 2009; García-Ávila et al., 2019)

removing these cations from saline WWW) indicates a potential application of halophytes for use in TWs. Moreover, heavy metal hyperaccumulation has been a long reported phenomenon where plants accumulate elevated concentrations of a particular metal *in planta* above a threshold (e.g. 1000 mg/kg in leaves for nickel (Ni) - see Van der Ent et al. (2013) for review). However, the hyperaccumulation threshold for other elements, such as Na^{+}, has not been defined. A study, conducted by Levinsh et al. (2021), investigated the accumulation of Na^{+} in leaves by 102 plant taxa across 77 sampling sites. Based on leaf Na^{+} concentrations, the "Na^{+}-hyperaccumulation threshold" was set at 18–30 g/kg dry mass. It must however be noted that further research on the hyperaccumulation threshold of Na^{+} (as well as elements other than heavy metals) must be conducted to validate these thresholds. This further suggests the potential application of halophytes for use in TWs for the removal of persistent inorganics, namely Na^{+} and K^{+}, from saline WWW.

Halophytes may be divided into different categories based on the mode of salt transport, storage, and potential excretion. These categories include recretohalophytes [divided into *exo*- (excrete excess salts from specialized salt glands) and *endo*-recretohalophytes (store salts in glands)], euhalophytes (compartmentalize salts into leaf/stem vacuoles), and pseudohalophytes (accumulate salts in the vacuoles of the wall or parenchyma of xylem present in roots) (Dassanayake and Larkin, 2017; Deinlein et al., 2014; Litalien and Zeeb, 2020). Halophytes can be further characterized by their dependence on salt (i.e. facultative vs obligate halophytes) and the environments they inhabit (e.g. hydro-halophytes, xero-halophytes). Halophytes may tolerate a range of inorganic contaminants by effectively detoxifying metal ions via heavy metal exclusion (i.e. selective restriction of the uptake and translocation of metals by roots), excretion (i.e. phytoexcretion, where salt glands excrete excess ions thereby contributing to the maintenance of the plant's metal homeostasis network), and/or accumulation (i.e. compartmentalization of metal ions into vacuoles) (Nikalje and Suprasanna, 2018; Christofilopoulos et al., 2016; Wei et al., 2020). These halophytic characteristics have important implications for the removal of inorganics as the use of *exo*-recretohalophytes may re-introduce extracted contaminants into the TW water body, resulting in negative inorganic removal percentages. For example, the presence of salt glands in the photosynthetic plant parts of some *exo*-recretohalophytes are used to detoxify accumulated metal ions where excess salts (and heavy metal constituents) are excreted onto the surface of the leaves (Wei et al., 2020; Sookbirsingh et al., 2010). However, exo-recretohalophytes may also desalinate environments by haloconduction - a process whereby salt excreted onto the leaf surface is mobilized by wind and deposited onto surrounding areas (Yensen and Biel, 2008; Yun et al., 2019). The deposition of salts away from the contaminated site may enable the dispersal of salts over large areas, diluting the impact of salt accumulation within the receiving environment (Yun et al., 2019). Moreover, the dispersal of salts (where some salts are considered macro- and micro-nutrients) at low concentrations over large areas may improve the nutrient content of salt-deficient soils (Yun et al., 2019). Although this provides an additional application of terrestrial exo-recretohalophytes, this phytotechnology is a relatively novel concept requiring further investigation. The implications for treatment of WWW in TWs is that during rainfall events secreted salts may be washed off plant leaves and subsequently re-introduced into the WWW, consequently increasing the concentration of salts in WWW, resulting in negative removal efficiencies (Wei et al., 2020; Yun et al., 2019). Another important implication was demonstrated by Matinzadeh et al. (2019) where euhalophytes accumulated elevated concentrations of Na^{+} compared with K^{+}. This was attributed to the role of Na^{+} in mediating the plant's ion homeostasis network whereas elevated concentrations of K^{+} were accumulated by facultative and pseudo-halophytes (essential macronutrient) thereby differing in their ability to accumulate salts. This study elucidates the importance of identifying viable halophyte candidates with desirable halophytic traits for targeted contaminant removal in TWs. Moreover, the composition and concentration of contaminants present in WWW (refer to Table 1) must be characterized prior to the selection of halophytic plant species.

In many countries, the main means of disposal of treated WWW is via land irrigation, presenting one of the major environmental challenges associated with the wine production industry (Howell and Myburgh, 2018). Land irrigation with WWW containing inorganics

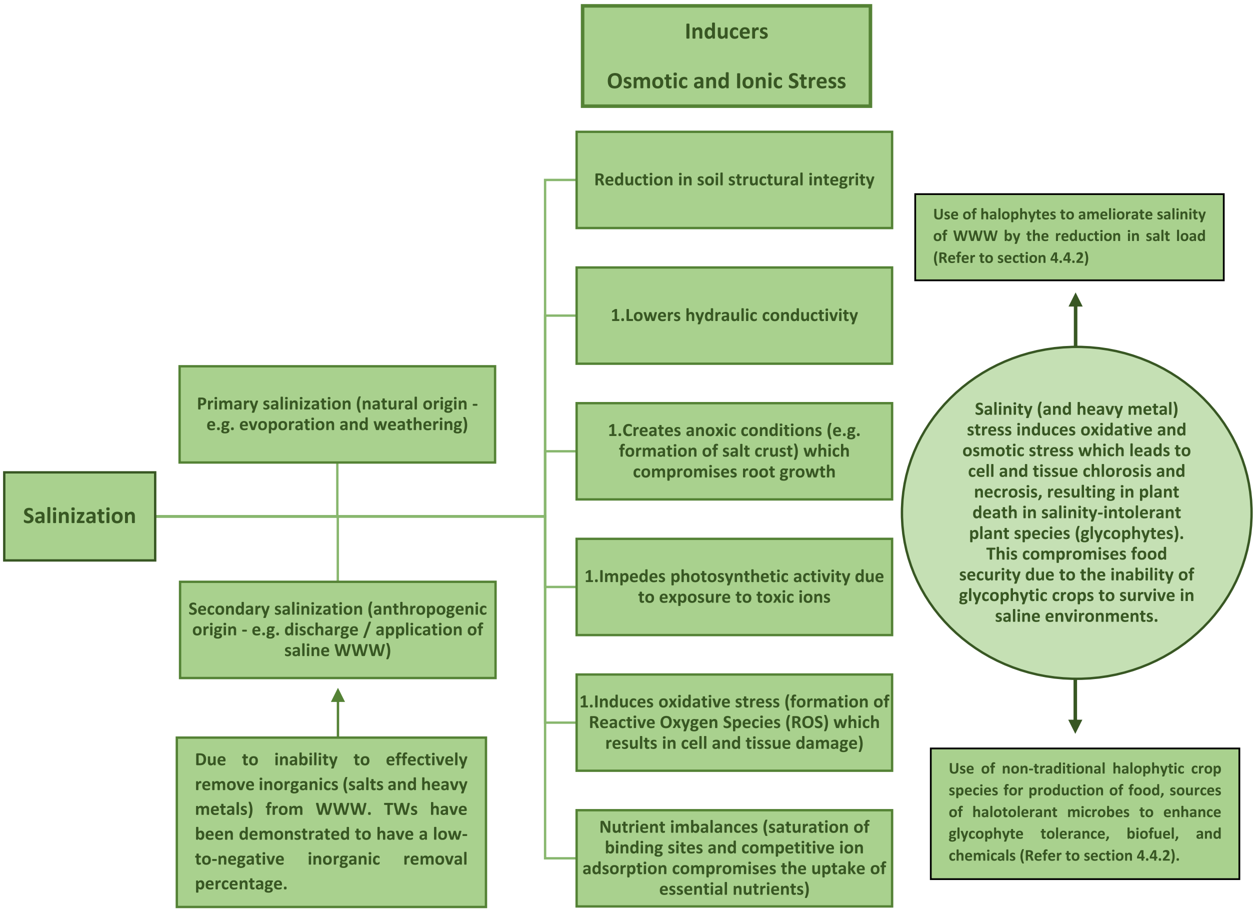

Fig. 1. Pathways of salt (and heavy metal) phytotoxicity. Phytotoxic effects of salinity on plant growth and survival, where salinity stress may induce the combination of ionic and osmotic stress leading to secondary stresses.
References: (Buelow et al., 2015; Litalien and Zeeb, 2020; Acosta et al., 2011; Butcher et al., 2016).

may result in land salinization, increase in soil sodicity, potential eutrophication of receiving water bodies, and assimilation of other inorganics (e.g. heavy metals) in the soil (Howell and Myburgh, 2018; Liang et al., 2021; Mulidzi et al., 2020).

Therefore, to mitigate the negative impacts associated with disposal practices, after secondary treatment, WWW must be effectively managed with a focus on the removal of salts, and their constituents. The management of WWW includes, but is not limited to, (i) treatment of saline WWW by phytodesalination before its disposal (Table 4), and/or (ii) irrigation of cash crops tolerant of saline WWW (i.e. beneficial irrigation). These management options must be considered as they directly impact socioeconomic development (e.g. food security) and the receiving environment. To implement either of these management strategies, halophytes with an elevated degree of cross-tolerance and extraction potential should be utilized in TWs to reduce salt load, as well as their constituents, present in WWW.

4.3.2. Phytoremediation of saline winery wastewater before disposal

Although numerous studies have investigated the phytoremediation of saline and heavy metal contaminated land by halophytes, limited research has been conducted on the phytodesalination and phytoextraction of inorganics present in wastewater originating from various sources (Khanlarian et al., 2020; Mujeeb et al., 2020; Wiszniewska et al., 2019). To the knowledge of the authors, studies investigating the phytoremediation potential of halophytes have been restricted to studies on wastewater originating from aquaculture (Lymbery et al., 2013; Diaz et al., 2020)] where salt concentrations have been reported in the range of 17,000 - 46,000 mg NaCl/L), domestic (e.g. (Fountoulakis et al., 2017)), tannery (e.g. salt concentrations up to 80,000 $mg \cdot L^{-1}$) (Ayyappan et al., 2016; Calheiros et al., 2007; Ashraf et al., 2020), tool factory (Maine et al., 2007), oil production (Stefanakis, 2020), and textile (Saeed and Sun, 2013) saline wastewater. The wine, aquaculture, and other agricultural industries generate relatively similar key contaminants (namely organics, salts, N, and P) compared with the mining, tannery, textile, and petrochemical industries – comprised of relatively complex contaminants and solution chemistry (Liang et al., 2017; Stefanakis, 2020; Zhao et al., 2020). Although, the concentration and composition of these inorganics differ within and between industries, limited studies have investigated the use of halophytes for the treatment of saline wastewater originating from various industries. The majority of these studies have been restricted to the use of hydro-halophytes, such as *P. australis* (Nawaz et al., 2020) and *Typha* spp. (Meitei and Prasad, 2021) which cross-tolerate saline and heavy metal stresses under flooded conditions (which they naturally inhabit) and have therefore received notable attention for their use in TWs.

Halophytes have been reported to effectively (hyper)accumulate a range of inorganics including salts and their constituents (Na^+, K^+, Mg^{2+}, Cl^-, NaCl, and SO_4^{2-}), as well as heavy metals (e.g. cadmium (Cd^{2+}), copper (Cu^{2+}), lead (Pb^{2+}), and zinc (Zn^{2+}) – ecotoxicologically important heavy metals present in WWW from soil and water (Khanlarian et al., 2020; Mujeeb et al., 2020; Wiszniewska et al., 2019). For example, Fountoulakis et al. (Fountoulakis et al., 2017) demonstrated the ability of the terrestrial halophyte, *Atriplex halimus*, to effectively remove elevated salt load from TWs along with high biomass production compared with the *Juncus acutus* and *Sarcocornia perennis*. This highlights the possible role of halophytes, and the need to select certain halophytes with an elevated ability to remediate and cross-tolerate a range of inorganics present in WWW.

4.3.2.1. The novel application of terrestrial halophytes. Flooding in combination with salinity stress are common environmental variables. To survive, plant species must tolerate the combination of these stresses (and secondary stresses such as the reduction in uptake and translocation of nutrients, such as K^+, to aboveground biomass) where the inability to do so results in plant mortality (Sairam et al., 2008). Although tolerance to the combination of these stresses typically involves adventitious root production (maintaining an internal O_2 content), halophytes possess the ability to effectively maintain their ion homeostasis network - regulating shoot ion concentrations independent of anoxic environmental conditions (Colmer and Flowers, 2008). The ability of some halophytes to cross-tolerate these abiotic stresses is elucidated by the elevated productivity of salt marshes (Kelleway et al., 2017). Although no link has been provided, various studies have indicated the ability of terrestrial halophytes to cross-tolerate these stresses. For example, Farzi et al. (2017) demonstrated the use of three terrestrial halophytes, namely *Salicornia europaea, Salsola crassa,* and *Bienertia cycloptera*, in a TW under a salinity dose-dependent experiment. These halophytes completed their life cycles under TW conditions and reduced measured elemental parameters to permissible levels. Aquatic plant species have typically been investigated for their rhizofiltration potential (i.e. the sorption (adsorption/absorption), concentration, and precipitation (onto the root surface) of contaminants from wastewater by plant roots), suggesting the possible role of terrestrial plant species, and halophytes in particular, for rhizofiltration and selection for use in TWs. A study conducted by Lee and Yang (2010) demonstrated that the terrestrial plant, *Helianthus annuus* (sunflower), effectively removed 80% of the uranium (U) present in a hydroponic solution via rhizofiltration. Moreover, terrestrial xerophytes and halophytes (such as *Atriplex halimus* and *Bassia indica*) have been successfully propagated in TWs (Fountoulakis et al., 2017; Shelef et al., 2013). Another study demonstrated that terrestrial halophytes from the genus *Salicornia* possess aerenchyma within their roots, a flooding-tolerance mechanism present in hydrophytes, promoting redox conditions within the rhizosphere. This creates microenvironments conducive to nitrification/denitrification processes within TW systems (Brix, 1994; Haberl et al., 1995; Faulwetter et al., 2009). Furthermore, the growth, survival, and phytoremediation potential of terrestrial halophytes may be enhanced through the control of TW operational and functional components. For example, substrate-less TWs, such as floating (FTW, utilizing larger, emergent hydrophytes planted in a buoyant mat) or vertical up-flow (VUF) TW, modifies the plant's root system architecture (RSA). This may enhance the halophyte's rhizofiltration potential by increasing root growth and root surface for contaminant sorption, concentration, and/or precipitation, biofilm development (see Section 4.3.3), and/or root-contaminant contact time as roots are directly exposed to contaminants present within the WWW (Fig. 2) (Colares et al., 2020; Afzal et al., 2019; Saddhe et al., 2020). As terrestrial halophytes have been demonstrated to grow under TW conditions, the phytoremediation potential of terrestrial halophytes can therefore be enhanced by modifying TW design.

The management of WWW is a major sustainability challenge within the wine industry. Although TWs generate multi-purpose byproducts (e.g., harvesting leaf biomass for biofuel or fertilizer, re-use of treated WWW for irrigation, etc.), WWW is still largely viewed as a waste product (Liu et al., 2012). Moreover, global climate change is expected to negatively impact the quality and quantity of treated WWW available for re-use (Bolzonella et al., 2019; Welz et al., 2016). This predicted impact on water scarcity is also expected to intensify the reliance on re-using treated wastewater – forming a key role in sustainable water management practices (Vo et al., 2014). Thus, WWW sustainability challenges within the wine industry must be addressed in the context of global climate change.

Various climatic drivers (e.g., temperature, evapotranspiration (ET), and wind) place a selective pressure on plants and microbes that can tolerate various hazards associated with the combination of the effects of these drivers, such as environmental salinization (Merloni et al., 2018; Zscheischler et al., 2018). Macrophytes, traditionally used in TWs, possess a low water use efficiency. These inefficient water users have an elevated water loss potential due to elevated ET processes, processes which are expected to be exacerbated by climate change (Headley et al., 2012). Elevated water loss potential directly impacts the quality (i.e., concentration of contaminants within the TW system) and quantity (due to change in hydrologic regime, decreasing the volumetric flow of WWW passing through the system) of treated wastewater available for reuse (Headley et al., 2012; Białowiec et al., 2007). Thus, it is envisaged that climate change is likely to decrease the sustainability and performance of traditional biological treatment systems utilizing salinity-intolerant macrophytes (Vo et al., 2014).

This sustainability challenge may be addressed by the selection of efficient water use plant species and/or decreasing the hydraulic retention time to minimize ET (Freedman et al., 2014). Some terrestrial halophytes possess the ability to cross-tolerate a combination of environmental conditions, such as salt and drought stresses (Nikalje et al., 2019). This is attributed to various adaptations including physiological processes such as the (i) density, size, and location of stomata (small pores regulating gaseous exchange), (ii) waxed epidermal layer in leaves (reducing transpiration water loss from the surface of the leaves), and/or (iii) small-sized leaves/scales (reduction in surface area) which result in reduced transpiration and thus, water loss (Shabala, 2013; Belkheiri and Mulas, 2013; Akcin et al., 2017). This further strengthens the case for utilizing terrestrial halophytes in the treatment of saline WWW, where these adaptations promote water use efficiency by halophytes while simultaneously tolerating a range of abiotic stresses affected by climatic drivers.

Thus, the criteria for selecting terrestrial halophytes in TWs for saline WWW treatment should be expanded to include the selection of species which (i) are easily propagated, (ii) are able to grow and survive under TW conditions, (iii) are able to (hyper)accumulate salts and their constituents (Na^+), (iv) are efficient water users with low ET, (v) have desirable modes of salt transport, accumulation and/or excretion (i.e., haloconduction), and (vi) simultaneously contribute to the removal of other inorganics such as N and/or P (Grismer et al., 2003). Thus, the potential use of terrestrial halophytes should be incorporated as an additional plant selection criterion in future studies for the treatment of saline wastewater by TWs, contributing to sustainable wastewater resource management within the wine industry (Knight et al., 2019).

This review highlights a gap in knowledge and proposes the novel, non-traditional use of terrestrial halophytes (which have been extensively studied for their phytoremediation potential in highly saline and heavy metal-contaminated terrestrial environments) for the remediation of saline WWW. Ultimately, the reduction of WWW salt load, along with other targeted inorganics, may reduce the negative impacts associated with the irrigation of land using treated WWW. This would ameliorate the negative impacts associated with the salinization and increase in soil sodicity – advancing the survivable conditions of numerous glycophytic plant species, including crops, and promoting food security in semi- and arid environments (Hayat et al., 2020). Solutions to these challenges have been described above and illustrated in Fig. 2, a process diagram presenting potential solutions to challenges negatively impacting the TW remediation process and the sustainability of the wine industry.

4.3.2.2. Irrigation of cash crops tolerant of saline winery wastewater. Globally, secondary salinization by irrigation has contributed to the salinization of approximately 20% of total cropland, which is predicted to increase to 50% by 2050 (Nachshon, 2018; Singh, 2021). Two themes to improve crop production under saline conditions have been researched, namely the (i) introduction of salt tolerance traits into glycophytic crops (i.e. genetic engineering), and (ii) domestication of halophytes as non-traditional crops (Mishra and Tanna, 2017; Nikalje

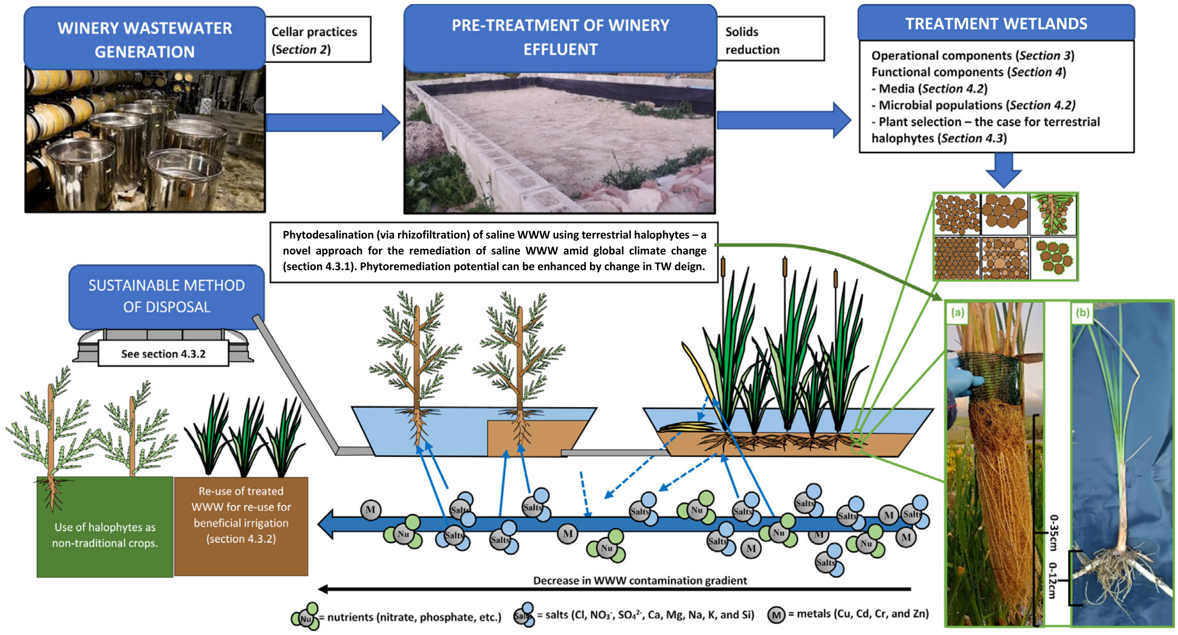

Fig. 2. Schematic depicting the contents of this review.

et al., 2018). The use of halophytes for the non-traditional production of food (e.g. *Chenopodium quinoa* (quinoa)), proposed sources of halotolerant microorganisms (to increase tolerance of glycophytic crops), biofuels, and chemicals in arid areas (i.e. saline water with poor soil quality and high solar radiation) has received notable attention in the last few decades (Debez et al., 2017; Etesami and Beattie, 2018). Halophytes can therefore be grown in non-arable land under saline WWW irrigation without competing for nutrients and resources required by glycophytic crop species, ultimately promoting food security in arid and semi-arid areas where climatic drivers (e.g. evapotranspiration and extreme temperatures) are expected to increase amid global climate change (Fisher et al., 2017).

The practical implications of selecting halophytes for use in phytoremediation or as a cash crop must consider the origin of the plant species. For example, *Eichhornia crassipes* (water hyacinth), a halophyte originating from South America, has been demonstrated to tolerate highly saline wastewater and accumulate various heavy metals and nutrients (Auchterlonie et al., 2021). However, *E. crassipes* is considered one of the world's most prevalent invasive aquatic plant species, negatively impacting socioeconomic development and the ecosystems they inhabit (Enyew et al., 2020). This has led to the legislated prohibition (e.g. South African National Environmental Management: Biodiversity Act – Alien Invasive Species Regulations, Act 10 of 2004) of growing and utilizing alien invasive plant species, such as *E. crassipes*, in phytoremediation trials or for recreational purposes. This highlights the need to select indigenous plant species, where the plant's potential spread outside of the TW would not negatively impact the receiving environment. It must be noted that *P. australis* is considered a cosmopolitan species with an elevated genetic diversity and phenotypic variation (Table 5) (Eller et al., 2017). This suggests that *P. australis* rapidly adapts to various geographic regions (i.e. different environmental conditions) across the world, making the case to use *P. australis* as a model plant species to investigate the remediation potential of TWs across geographical ranges.

4.3.3. Plant-assisted bioremediation (plant-microbe interactions)

Plant-microbe interactions can enhance the remediation of inorganic and organic contaminants in wetland systems. The use of plants, in combination with rhizosphere and/or endophytic bacteria enhances the remediation of organic and inorganic contaminants compared with using phytoremediation in isolation. Plant roots provide (i) a surface for microbial adhesion, (ii) protection of microbial communities from environmental conditions, such as desiccation, (iii) a C source for microbial growth and activity (i.e. release of exudates, such as organic acids, sugars, and amino acids), and (iv) aerobic microenvironments for aerobic (heterotrophic and autotrophic) microbial activities – where plants release O_2 into the rhizosphere via aerenchyma adventitious roots (Gagnon et al., 2007; Hussain et al., 2018; Abdullah et al., 2020). In turn, microbes associated with the rhizosphere promote plant growth and survival by increasing the bioavailability of nutrients, such as P and N, for plant uptake, N fixation, protection against plant pathogens, as well as promoting resistance (i.e. avoidance or tolerance) to phytotoxic elements (Liang et al., 2017; Chaudhry et al., 2005; Stefanakis et al., 2019). The composition of the microbial community is directly influenced by the plant species due to variation in (i) root system architecture, and (ii) rhizosphere pH (influenced by the release of exudates) (Liang et al., 2017; Stefanakis et al., 2019).

Rhizodegradation, or plant-assisted bioremediation, is a primary contributor to the remediation of organic contaminants (Donnelly and Fletcher, 1994). By-products, produced by rhizodegradation processes may be taken up into the plant, translocated to aboveground biomass via the transpiration stream, and subsequently volatilized by evapotranspiration (i.e. phytovolatilization). This process enhances the plant's ability to remove elevated concentrations of contaminants and/or their transformed/degraded species from TWs (Heaton et al., 1998; Limmer and Burken, 2016). Moreover, plant-assisted bioremediation has long been demonstrated to significantly increase the assimilation of contaminants within wetland substrate (Doyle and Otte, 1997) when compared with non-planted wetlands. This reduces the mobility, and thus the toxicity, of contaminants within aquatic and terrestrial (e.g. via treated WWW irrigation) environments, decreasing contaminants entering the food chain through plant uptake and accumulation in aboveground biomass.

Complex biological interactions associated with plant-assisted bioremediation of WWW limit the comparability and reproducibility of studies. This is attributed to the inability to effectively distinguish the role of plants (phytoremediation component), microbes (bioremediation component), microbe-microbe/plant-plant interactions (i.e. intra- and interspecies competition), as well as the plant-assisted bioremediation components utilized in TWs. Moreover, these biological, along with hydrogeochemical, contaminant removal pathways occur simultaneously and fluctuate through space and time (Imfeld et al., 2009). The degree to which plants contribute to the overall remediation potential of TWs differ between studies. For example,Zhang et al. (2014) demonstrated that plants play a significant role in the remediation of certain pharmaceutical contaminants whereas other studies have demonstrated no significant difference between contaminant removal efficiencies between planted and unplanted TWs. This further highlights the complexity of determining the efficacy of TWs treating WWW as the fate of contaminants (e.g. competitive ion adsorption, composition of contaminants), functional components (media, plant species, and environmental conditions) and operational parameters differ between studies (Table S1). In order to ensure that the role of plants is not masked, studies should ideally consider a range of relevant parameters that may affect the speciation, bioavailability, and toxicity of WWW contaminants on plants. These include environmental factors such as pH, electrical conductivity, redox potential, temperature, and O_2 availability of WWW.

5. Conclusion

Treatment wetlands, comprised of biological and hydrogeochemical components, have been used to remediate wastewater originating from various anthropogenic sources. The systems have proven effective for reducing the organic load and neutralising the pH of WWW, but concurrent or downstream removal of inorganics remains a challenge. Removal of inorganic macronutrients (N,P,K) is important if the treated WWW is discharged into aqueous environments, but not if the effluent is to be used for irrigation. While N compounds can be mineralized, P is removed chiefly by media adsorption, so negative removal rates can be found once the binding sites are saturated. In line with circular economy principles, there is a need for studies aimed at capturing P from WWW for re-use. Other inorganics found in high concentrations in WWW (Na^+, K^+) are not removed by microbial action, nor adsorbed by the TW substrate. This creates a selective pressure for the use of halophytes which have the elevated ability to (hyper)accumulate these inorganics (effectively removing these cations from WWW) and cross-tolerate a range of stresses. This review also presents the case for the use of terrestrial halophytes in TWs. Terrestrial halophytes have the ability to (hyper)accumulate and cross-tolerate a range of contaminants, along with the ability to control operational parameters of TWs (i.e. artificial aeration) - creating viable alternatives to the selection of macrophytes currently used world-wide. Salinization is a growing issue compromising food security due to the inability of glycophytes to survive under saline conditions. The non-traditional use of halophytic crops which can be grown in non-arable, saline land under WWW irrigation, is a viable solution ultimately promoting food security in arid and semi-arid environments, where fresh water for irrigation is limited.

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Declaration of competing interest

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Appendix A. Supplementary data

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