Constructed wetlands in the treatment of agro-industrial wastewater: A review

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Abstract

Due to their simplicity and low operation cost, constructed wetlands are becoming more prevalent in wastewater treatment all over the world. Their range of applications is no longer limited to municipal wastewater, but has expanded to the treatment of heavily polluted wastewaters such as agro-industrial effluents. This paper provides a comprehensive literature review of the application of constructed wetlands in treating a variety of agro-industrial wastewaters, and discusses pollutant surface loads and the role of constructed wetland type, prior-treatment stages and plant species in pollutant removal efficiency. Results indicate that constructed wetlands can tolerate high pollutant loads and toxic substances without losing their removal ability, thus these systems are very effective bio-reactors even in hostile environments. Additionally, the review outlines issues that could improve pollutant treatment efficiency and proposes design and operation suggestions such as suitable vegetation, porous media and constructed wetland plain view. Finally, a decision tree for designing constructed wetlands treating agro-industrial wastewaters provides an initial design tool for scientists and engineers.

Keywords: constructed wetlands, agro-industrial wastewater, review, decision tree, design.

Available online at the Journal website: http://www.ache.org.rs/HI/

REVIEW PAPER

UDC 504.5(497.1)"1999":
546.791623.454.8:631.4(497.1)

Hem. Ind. **69** (2) 127–142 (2015)

doi: 10.2298/HEMIND150121018S

Constructed wetlands (CWs) are a low-cost technology that has been used to treat various types of wastewaters for nearly twenty years [1–2]. They are an attractive treatment option because they use solar energy, are simple to construct and operate, have low maintenance cost and are inexpensive and sustainable compared to conventional treatment methods [3]. The water quality improvement observed depends on the wetland design, microbial community, and the different plant species involved [4].

Constructed wetlands are artificial systems that have been designed to operate as natural wetland ecosystems to improve wastewater treatment efficiency. These systems generally fall into two general categories: a) subsurface flow system wetlands, and b) free water surface systems. In recent years, constructed wetland technology has been based on three basic types of CWs: a) free water surface (FWS) wetlands, b) horizontal subsurface flow (HSF) wetlands and c) Vertical flow (VF) wetlands.

Lately, different CW types have been paired into hybrid treatment systems. Hybrid CW systems have also been used extensively in wastewater treatment. These hybrid systems mainly consist of different HSF and VF stages and are used to achieve high organic matter removal efficiency and high nitrification rates from the VF CWs, and high denitrification rates from the HSF CWs.

CWs are mainly used to treat domestic and municipal wastewaters, but more recent applications of CWs include treatment of other types of wastewater, such as industrial, agro-industrial and agricultural wastewaters, various runoff waters and landfill leachate [5].

Agro-industry includes post-harvest activities involved in the transformation, preservation and preparation of agricultural products for intermediary states or final consumption. Agro-industrial wastewaters are usually characterized by their high organic load and their quantity and quality variations over a year [6].

Although previous papers have partially reviewed CWs treating agro-industrial wastewater, they are either dated [7] or agro-industrial wastewater is only a limited part of the paper and not thoroughly discussed [8]. This paper provides a comprehensive literature review of the application of CWs in treating a variety of agro-industrial (*i.e.*, dairy, animal farm, winery, trout farm, sugar production and olive mill) wastewaters, as CW application is no longer limited to municipal wastewaters. To evaluate comprehensively the ability of CWs to treat these wastewaters, certain parameters are examined and discussed thoroughly. These parameters include CW type, prior-treatment stages, and plant

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Paper received: 21 January, 2015
Paper accepted: 16 March, 2015

species. Other significant parameters such as CW surface area and pollutant surface loads for specific toxic pollutants (*e.g.*, phenols) are discussed in detail. Furthermore, the review provides design and operation suggestions including issues such as vegetation, porous media, CW plain view and others, by introducing an innovative decision tree for designing CWs treating agro-industrial wastewaters.

PRIOR-TREATMENT STAGES

CWs were mainly used as a polishing treatment method for agro-industrial wastewater treatment, thus several prior-treatment technologies are employed (Table 1). These technologies aim to remove suspended solids and reduce organic matter loads. Suspended solid removal is essential before the wastewaters enter the CW stage, because high suspended solid concentrations can cause clogging of the porous media. The necessity of pre-treatment stage was also outlined in previous reviews, where it was mentioned that CWs treatment facilities were coupled mainly by settling basins, lagoons, septic tanks in order to mainly remove SS [7]. Therefore, in the majority of experiments/applications referred in the literature, the pre-treatment stages include either simple settling basins [3,9–22] or stabilization lagoons, which can achieve high removal rates of suspended solids and organic matter [4,23–27].

According to the specifications of each wastewater, prior-treatment stages can also include other technologies. Winery wastewater treatment usually includes a sludge digestion stage to remove suspended solids and significantly reduce organic loads [28]. Animal farm wastewaters that contain high organic and nitrogen

Table 1. Prior-treatment stages used in CWs treating agro-industrial wastewater

Prior-treatment stage	Wastewater Type	Reference
	FWS	
Stabilization lagoon	Animal farm	[1,35–39]
	Dairy	[40]
Dilution	Animal farm	[41]
	OMW	[42]
Settling tanks	Dairy	[3,12,16]
	Animal farm	[19]
	Hybrid	
Settling tanks	Dairy	[15]
Stabilization lagoons	Animal farm	[25]
	Vinegar	[26]
Septic tank	Dairy, swine and potato starch	[31]
Sludge digester	Winery	[28]
	HSF	
Biological treatment	Dairy- Cheese whey	[2,43,44]
	Animal	[30]
Stabilization lagoons	Dairy	[4,23,27]
	Animal farm	[24]
Settling tank	Dairy	[9–11,13,17,20,21]
	Animal farm	[18]
	Winery	[14]
	Trout farm	[22]
Solid separation, anaerobic digestion and aerobic oxidation	Animal	[29]
Sludge digestion	Animal	[45,46]
	Winery	[47]
Coagulation	OMW	[33,48]
Dilution	Sugarcane	[49]
	VF	
Dilution	OMW and swine	[50]
	Cheese whey	[51]
Biological filter	OMW	[34]
Electrochemical oxidation	OMW	[32]

loads are mainly treated with various biological treatment methods to reduce both organic and nitrogen loads [29–31]. The main treatment stages for OMW involve advanced treatment methods including coagulation [32], electrochemical oxidation [33], and biological trickling filters [34] to lower toxic phenol concentrations.

Several experiments/applications on agro-industrial wastewater treatment using CWs lack a main treatment stage [41,42,49-52]. In these experiments/applications the wastewaters were diluted with tap water before being introduced into the CW system.

Although the majority of CWs experiments/applications on agro-industrial wastewaters refer to prior-treatment stages and they do not discuss the specific aspects of these stages. Thus, limited information is available concerning the significance and effectiveness of prior-treatment stages on CW performance. Nevertheless, some critical information can be drawn. Specifically, although physicochemical methods (e.g. coagulation) can successfully remove TSS, they use chemical compounds (*e.g.*, calcium hydroxide, lime putty and hydraulic lime). This leads to the increase of pH values to above 8, which could be toxic to CW vegetation [32]. On the other hand, the electrochemical oxidation as a prior-treatment stage to CWs treating OMW results in lower pollutant removal efficiencies than when it is used as a polishing stage after CW treatment [33]. Therefore it seems that a biological treatment method should be applied as a prior-treatment stage to CWs [33,34]. As agro-industrial wastewaters are characterized by high pollutant loads, aerobic suspended growth systems would probably not achieve satisfactory removal efficiencies. In contrast, aerobic attached growth systems (*e.g.*, trickling filters) or anaerobic systems could operate under these high pollutant loads and could achieve high removal rates.

VEGETATION

One of the main issues in CW treatment systems is to identify the precise role of the plant vegetation in pollutant removal and define their toxicity boundaries. Several plant species have been used in CWs treating agro-industrial wastewater treatment (Table 2). The exact contribution of plants in nutrient removal is a controversial issue as almost all related studies give different removal efficiencies. Gottschall *et al*. [36] report that nutrient removal due to plant uptake was significantly lower in their study compared to previous studies [53,54] which reported that plant uptake is responsible for 27–66% of nitrogen removal and 47–65% of phosphorus removal. In addition, Newman *et al*. [11] report that only 3% of nitrogen removal should be attributed to plant uptake. Mantovi *et al*. [13] also attribute most nutrient removal to biofilm biochemical oxidation and plant uptake. Tanner *et al*. [23] reported that planted wetlands showed greater removal efficiencies of N and P from dairy farm wastewaters than unplanted wetlands. They recorded higher TP removal in the summer months due to higher plant biomass growth and temperatures. The unplanted wetland proved to be less efficient at removing both N and P with higher loading rates. Percentage removal of NH_4^+–N increased with retention time in the planted HSF CW, whereas the unplanted wetland showed lower performance. Plant rhizosphere aeration may stimulate aerobic decomposition processes by increasing nitrification and subsequent gaseous losses of N through denitrification [55,56], and by decreasing the relative levels of dissimilatory nitrate reduction to ammonium [57].

Table 2. Plant species used in CWs treating agro-industrial wastewater

Plant species	Influent concentration, mg/L			CW Type	Wastewater type	Reference
	COD	TN	P			
Acorus calamus	1700	360	–	HSF	Animal farm	[46]
Alisma plantago-aquatica	1700	360	–	FWS	Dairy	[16]
				HSF	Animal farm	[46]
Butomus umbellatus	–	–	–	Hybrid	Dairy	[15]
Cacomantis flabelliformis	1034	448	–	FWS	Animal farm	[19]
Carex spp.	682–1700	11–360	2.3–10.4	FWS	Dairy	[16]
				HSF	Dairy	[10]
				HSF	Dairy	[59]
				HSF	Animal farm	[46]
				Hybrid	Vinegar	[26]
Ceratophyllum demersum	721–4045	14.7–65.2	1.9–4.9	FWS	Winery	[14]
Cucurbita maxima	–	–	–	Hybrid	Dairy	[15]
Eichhornia crassipes	1160	200	40	HSF	Animal farm	[29]
Eleocharis obtusa	–	–	–	HSF	Dairy	[9]

Table 2. Continued

Plant species	Influent concentration, mg/L			CW Type	Wastewater type	Reference
	COD	TN	P			
Elodea canadensis	721–4045	14.7–65.2	1.9–4.9	FWS	Winery	[14]
Filipendula ulmaria	1700	360	–	HSF	Animal farm	[46]
Glyceria spp.	682–1500	97	10.4	FWS	Dairy	[16]
				HSF	Dairy	[10]
				HSF	Dairy	[59]
				HSF	Dairy	[60]
Holcus lanatus	–	–	–	HSF	Dairy	[60]
Iris pseudacorus	1700	360	–	FWS	Dairy	[16]
				HSF	Animal farm	[46]
Juncus spp.	–	20–120	20–50	FWS	Animal farm	[1]
				FWS	Animal farm	[61]
				HSF	Animal farm	[62]
Lemna spp.	254	29	17	FWS	Dairy	[3]
				HSF	Dairy	[27]
Litaneutria minor	1900	164	53	FWS	Dairy	[12]
				HSF	Dairy	[9]
Lythrum salicaria	1700	360	–	HSF	Dairy	[9]
				HSF	Animal farm	[46]
Mentha aquatica	1700	360	–	HSF	Animal farm	[46]
Nymphea rustica	721–4045	14.7–65.2	1.9–4.9	FWS	Winery	[14]
Nuphar lutea	–	20–120	20–50	HSF	Animal farm	[62]
Phalaris arundinaceae	6.79–1500	6.16	0.34	FWS	Dairy	[16]
				HSF	Dairy	[59]
				HSF	Trout farm	[26]
Phormium tenax	–	–	–	HSF	Dairy	[60]
Phragmites spp.	6.79–14000	6.16–506	0.34–95	FWS	Dairy	[16]
				FWS	Dairy	[12]
				FWS	Dairy	[40]
				Hybrid	Dairy	[15]
				Hybrid	Dairy	[63]
				HSF	Dairy	[64]
				HSF	Dairy	[17]
				HSF	Dairy	[4]
				HSF	Dairy	[10]
				HSF	Dairy	[13]
				HSF	Dairy	[9]
				HSF	Dairy	[11]
				HSF	Cheese whey	[44]
				HSF	Animal farm	[24]
				HSF	Animal farm	[45]
				HSF	Animal farm	[46]
				HSF	Animal farm	[30]
				VF	Animal farm	[48]
				VF	Animal farm	[65]
				HSF	OMW	[48]
				VF	OMW	[34]
				FWS	OMW	[42]
				FWS, HSF, VF	Winery	[14]

Table 2. Continued

Plant species	Influent concentration, mg/L			CW Type	Wastewater type	Reference
	COD	TN	P			
Phragmites spp.	6.79–14000	6.16–506	0.34–95	Hybrid	Vinegar	[26]
				HSF	Trout farm	[26]
				VF	OMW and swine	[50]
				Hybrid	Winery	[28]
				Hybrid	Dairy, swine and potato starch	[31]
				Hybrid	Winery	[47]
Pontederia spp.	254–1200	20–120	22–50	HSF	Dairy	[27]
				HSF	Animal farm	[62]
				HSF	Sugarcane	[49]
Schoenoplectus americanus	445–796	66–171	56–71	FWS	Animal farm	[39]
				FWS	Animal farm	[61]
				FWS	Animal farm	[37]
				FWS	Animal farm	[66]
				FWS	Animal farm	[38]
				Hybrid	Animal farm	[25]
Scirpus spp.	285–2240	20–907	20–53	FWS	Dairy	[12]
				FWS	Dairy	[40]
				HSF	Dairy	[4]
				HSF	Dairy	[23]
				FWS	Animal farm	[1]
				FWS	Animal farm	[61]
				HSF	Animal farm	[62]
				HSF	Animal farm	[18]
				HSF	Animal farm	[24]
				HSF	Animal farm	[46]
Solanum americanum	–	–	–	FWS	Animal farm	[1]
				FWS	Animal farm	[61]
Sparganium erectum	1700	360	–	FWS	Dairy	[16]
				Hybrid	Dairy	[15]
				Hybrid	Animal farm	[25]
				HSF	Animal farm	[46]
Senecio sylvaticus	–	–	–	Hybrid	Dairy	[15]
Sotalia fluviatilis	1160	200	40	Hybrid	Dairy	[67]
				HSF	Dairy	[21]
Suillus pungens	2700	102	26	HSF	Dairy	[9]
				HSF	Dairy	[11]
Stuckenia pectinata	682	97	10.4	HSF	Dairy	[10]
Typha spp.	150–4045	14.7–360	10.4–71	FWS	Dairy	[16]
				FWS	Dairy	[3]
				FWS	Dairy	[12]
				FWS	Dairy	[40]
				Hybrid	Dairy	[15]
				HSF	Dairy	[2]
				HSF	Dairy	[10]
				HSF	Dairy	[59]
				HSF	Dairy	[9]
				HSF	Dairy	[11]
				HSF	Dairy	[20]

Table 2. Continued

Plant species	Influent concentration, mg/L			CW Type	Wastewater type	Reference
	COD	TN	P			
Typha spp.	150–4045	14.7–360	10.4–71	FWS	Animal farm	[36]
				FWS	Animal farm	[1]
				FWS	Animal farm	[39]
				FWS	Animal farm	[61]
				FWS	Animal farm	[37]
				FWS	Animal farm	[66]
				FWS	Animal farm	[38]
				Hybrid	Animal farm	[25]
				HSF	Animal farm	[62]
				HSF	Animal farm	[24]
				HSF	Animal farm	[46]
				HSF	Animal farm	[30]
				VF	OMW	[33]
				FWS, HSF, VF	Winery	[14]
				VF	Cheese whey	[52]
Urtica dioica				Hybrid	Dairy	[15]

The selection of appropriate vegetation in CWs treating agro-industrial wastewater is an important issue as toxic effects caused by high organic and nutrient loads can occur. *Typha* spp. and *Phragmites* spp. are most commonly used for various agro-industrial wastewaters (*i.e.*, dairy, animal farm, winery, vinegar, trout farm, potato starch and OMW). Apart from *Typha* and *Phragmites* thirty-one other species have been used in CWs treating agro-industrial wastewater. Of these 31 species, *Carex* spp. and *Scirpus* spp. are the most frequently used in CWs treating dairy and animal farm wastewaters. Although a significant number of plant species have been used in CWs treating agro-industrial wastewaters no significant observations have been made concerning the effect of the different species on pollutant removal efficiencies. On the contrary, the comparative studies with different species showed that all were able to grow in CWs treating dairy wastewater without showing toxicity [15] and did not show any significant differences on organic matter removal [58]. Cronk [7] also suggested that CWs vegetation should be native and tolerant to extreme pollutant loads, while they should also have rapid growth and significant nutrient uptake.

Phragmites

Phragmites spp. is recognized as the most popular plant used in CW applications [68]. Its extensive use is due to: a) its high biomass productivity (up to 9,890 g dry mass/m^2 per year), b) its ability to grow in fresh and saline waters and c) its natural widespread distribution [68]. *Phragmites* spp. is also extensively used in CW applications for agro-industrial wastewater treatment. *Phragmites* spp. appears to be extremely tolerant to high concentrations of organic matter and nutrients and does not show toxic effects to COD, TKN and TP concentrations of up to 14000, 506 and 95 mg/L, respectively (Table 2).

Typha

Along with *Phragmites* spp., *Typha* spp. is the most commonly used plant in CWs, and they share common characteristics of high biomass productivity, tolerance to brackish waters and a widespread distribution [68]. *Typha* spp. is used in all CW types to treat a variety of agro-industrial wastewaters (i.e. dairy, cheese whey, animal farm, OMW and winery). *Typha* spp. is tolerant to high organic matter and nutrient concentrations, as it has been used in experiments/applications in which COD, TKN and TP concentrations were up to 4000, 360 and 71 mg/L, respectively (Table 2). These concentrations are rather high and prove that *Typha* spp. can be used successfully in CWs treating high strength wastewaters. On the contrary, Ghosh and Gopal [2] examined plant tolerance to dairy wastewater and found that young *Typha* plants yellowed when wastewater with high EC values was applied to the CWs. Ghosh and Gopal [2] also mentioned that plant density and height were maximum near the CW's inlet and attributed this to the higher nutrient concentrations present in these areas which promote plant growth. Concerning nutrient uptake from *Typha* spp., Gottschall *et al.* [36] found that N was absorbed in *Typha* spp. biomass mainly as NH_4^+ and not as NO_3^-, while P uptake was significant lower.

FWS CWS TREATING AGRO-INDUSTRIAL WASTEWATERS

FWS CWs have been extensively used to treat a variety of agro-industrial wastewaters (*i.e.*, dairy, animal farm and OMW) (Table 3). These applications range from pilot-scale experiments to full-scale facilities as their areas vary from 120.6 to 4000 m^2 (Table 3). FWS CWs have been operated with various pollutant surface loads ranging from 1.9 to 259.4 g/m^2 per day for organic matter, from 0.4 to 77 g/m^2 per day for TKN, from 0.05 to 12.7 g/m^2 per day for phosphorus, and from 2.55 to 949 g/m^2 per day for TSS. Removal efficiencies in these CWs show great variations, ranging from 3 to 98% for organic matter, from 26 to 96% for TKN, from 8 to 92% for TP, and from 26 to 99% for TSS, depending on the HRT applied and the metrological conditions. In most cases the high removal efficiencies were achieved when pollutant surface loads were low (Table 3).

A common characteristic of all FWS CWs treating agro-industrial wastewater is that influent COD concentration does not exceed 2000 mg/L, as at higher concentrations FWS CWs cannot efficiently remove pollutants due to the anoxic or anaerobic conditions created in the water column, which reduces the amount of oxygen available for microbial organic matter oxidization. According to Schaafsma *et al.* [12], organic matter and nutrient removal efficiencies are also significant, but in some cases, nitrate and nitrite concentrations are increased in the effluent. To enhance denitrification, Schaafsma *et al.* [12] suggested increasing plant density and partially recirculating wastewater. Concerning nitrogen removal, Gottschall *et al.* [36] showed that TKN removal was lower than NH_4^+ removal. The wetland was NH_4^+-dominated and showed a greater uptake of NH_4^+ than NO_3^-. The only attempt to treat OMW using FWS [42] showed significant removal efficiency for organic matter (86%). However, the organic surface load applied was the lowest for OMW reported in the literature (5–15 g/m^2 per day).

HRTs applied on FWS CWs range from 4 to 120 days and significantly affect wetland performance. When low HRTs were used (4 to 15 days) removal efficiencies were low and ranged from 40 to 60% [37,38,40]. On the other hand, high removal efficiencies were achieved for HRTs exceeding 60 days [3,36,61].

HSF CWS TREATING AGRO-INDUSTRIAL WASTEWATERS

HSF CW systems appear to be favoured for agro-industrial wastewater treatment compared to FWS CWs, as they have been used to treat a greater variety of wastewaters (*i.e.*, dairy, animal farm, OMW, winery, sugarcane and trout farm). They have been used at several scales from laboratory experiments to full-scale applications with surfaces areas ranging from 0.25 to 7600 m^2 (Table 4). HSF CWs operated under similar pollutant surface loads with FWS CWs varying from 0.17 to 376 g/m^2 per day for organic matter, 0.007 to 2.7 g/m^2 per day for TKN, 0.004 to 4.7 g/m^2 per day for phosphorus, and from 0.2 to 62.4 g/m^2 per day for TSS. Removal efficiencies in HSF CWs are similar to those recorded in FWS CWs as they range from 28 to 99% for organic matter, 10 to 99% for TKN, 2 to 99% for TP, and from 76 to 99% for TSS, depending on the applied HRT and the meteorological conditions (Table 4).

Although pollutant surface load and removal efficiencies of HSF CWs do not differ greatly from those in FWS CWs, HRTs applied in HSF CWs were lower and ranged from 2 to 60 days. Even with a 2-day HRT, HSF CWs can achieve high organic matter removal efficiencies (up to 90%) [44,49] and this establishes them as far more efficient bio-reactors than FWS CWs.

Table 3. FWS CWs treating agro-industrial wastewater treatment by CWs

Ref.	Wastewater type	CW surface area, m^2	Surface load, g/m^2 per day				Removed surface load, g/m^2 per day			
			C	TKN	TP	TSS	C	TKN	TP	TSS
[1]	Animal farm	120.6	2.74	3.5	–	–	–	–	–	–
[3]	Dairy	100	6.5	0.8	0.12	4.8	6.37	0.64	0.11	4.61
[12]	Dairy	500	–	–	–	–	–	–	–	–
[16]	Dairy	4265	6.84	–	0.05	2.55	6.22	–	0.04	2.52
[36]	Animal farm	327	–	0.95–1.62	0.33–0.34	–	–	0.38–0.65	0.05	–
[37]	Animal farm	220	7.14	1.23	1.1	24.2	4.2	0.7	0.32	15.0
[38]	Animal farm	440	1.9–6.1	0.7–4.0	0.8–1.6	4.4–8.2	0.9–2.7	0.3–1.8	0.16–0.32	1.8–3.3
[39]	Animal farm	440	8.13–87.1	0.4–3.5	–	3425–35109	3.3–34.5	0.2–1.6	–	1370–14044
[40]	Dairy	630	20.36	23.2	–	67.36	0.61	6.0	–	24.2
[42]	OMW		5–15	–	–	–	4.3–12.9	–	–	–
[61]	Animal farm	241.2	10.7–12	3.7–4.4	1–1.6	11.5–18.8	6.4–7.2	2.9–3.5	0.3–0.5	10.6–17.3
[66]	Animal farm	440	–	2.3	1.2	–	–	0.9	0.09	–

Table 4. HSF CWs treating agro-industrial wastewater treatment by CWs

Ref.	Wastewater type	CW surface srea, m^2	Surface load, g/m^2 per day				Removed surface load, g/m^2 per day			
			C	TKN	TP	TSS	C	TKN	TP	TSS
[9]	Dairy	138.6	52.6	1.9	0.514	14.8	14.7	0.5	0.14	6.7
[10]	Dairy	10–15	26.8–45.3	–	–	–	24.1–40.8	–	–	–
[11]	Dairy	138.6 (each cell)	51.3	1.96	0.5	24.6	38.9	0.55	0.23	22.1
[13]	Dairy	72	110.1	5.84	1.15	62.3	101.3	2.83	0.70	56.6
[14]	Winery	127–850	23.6–35.2	0.6–2.7	0.08–0.2	–	22.4–33.4	0.4–1.9	–	–
[17]	Dairy	50–1900	5.7–8.3	0.56–1.97	0.17–1.71	–	4.5–6.6	0.2–0.8	0.12–1.2	–
[18]	Animal farm	18	9.7–47.3	0.7–4.54	–	–	4.85–23.6	0.42–2.7	–	–
[2]	Dairy	1.63	8.6–34.5	1.3–4.9	OP: 0.3–1.1	0.2–0.7	5.2–20.7	0.78–2.94	OP: 0.09–0.33	0.16–0.56
[20]	Dairy	100	17	–	–	–	16.7	–	–	–
[21]	Dairy	892	68.5	–	0.6	16	–	–	–	–
[22]	Trout farm	23.9	9.8	8.9	0.49	–	19.8	1.1	0.27	–
[23]	Dairy	19	0.9–4.1	0.6–2.7	0.2–0.8	1.9–8.5	0.76–3.5	0.4–1.8	0.12–0.48	1.5–6.5
[24]	Animal farm	6000	0.17–0.26	0.15–0.68	–	0.21–1.85	0.12–0.18	0.09–0.48	–	0.07–0.93
[27]	Dairy	398	29.4	3.4	2	8.4	11.2	1.45	0.5	4.9
[29]	Animal farm	31.1	39–137	6.9–26.2	1.5–4.7	30.3–62.4	31.2–110	1.03–3.9	0.7–2.1	29.4–60.5
[30]	Animal farm	0.9	4.2	–	0.2016	6.1	–	–	–	–
[4]	Dairy	600	–	–	–	–	–	–	–	–
[43]	Dairy	20	–	–	–	–	–	–	–	–
[44]	Cheese whey	1.1	19–110	–	–	–	2–109	–	–	–
[45]	Animal farm	0.25	3.2	0.104	0.265	–	2.24	0.08	0.21	–
[46]	Animal farm	4500	0.14	0.03	0.005	–	0.13	0.03	0.005	
[47]	Winery	350	2–49	–	–	–	1.4–34.3	–	–	–
[48]	OMW	0.85	77.03 Phenols: 16.85	1.08	0.42	9.32	53.1 Phenols: 13.2	0.13	0.23	4.6
[49]	Sugarcane	0.9	47.2–94.8	–	–	–	37.8–75.8	–	–	–
[59]	Dairy	7600	1.2	–	–	0.21	1.14	–	–	0.2
[60]	Dairy	260	–	0.7– 2.9	0.004–0.013	–	–	0.4–1.7	0.0004–0.0013	–
[62]	Animal farm	4500	–	0.007	0.004	–	–	0.005	0.001	–
[64]	Dairy	160	–	–	–	–	–	–	–	–

Nevertheless, HRT and pollutant surface loads seem to be important parameters in HSF CWs as, according to Lee *et al.* [29], removal efficiencies are higher when surface loads are low, while Meers *et al.* [45] suggested that by increasing the HRT and the plant root depth zone in HSF CWs, removal efficiencies would increase. HSF CWs appear to be more efficient at treating OMW [32], as removal efficiencies reach 69% for COD, 12% for nitrogen, 55% for phosphorus, 50% for TSS and 79% for phenols. It should be mentioned that these removal efficiencies were achieved with pollutant surface loads higher than those applied to FWS systems, but lower than those applied to VF systems.

HSF CW operation is greatly influenced by HRT, as extremely low HRTs can lead to insufficient treatment, while high HRTs do not improve CW performance and lead only to higher area demand. Ghosh and Gopal [2] state that while organic matter removal increased nearly 3-fold when HRT increased from 1 to 2 days, when HRT was further increased to 3 days organic matter removal increased only by 18%. Sultana et al. [44] also state that HRTs ranging from 2 to 8 days have no significant effect on organic matter removal, while an HRT of 1 day was found to be inadequate for organic matter removal. Almost the same HRT range (2 to 7 days) was also proposed by Tanner *et al.* [23] for successful organic matter removal from dairy wastewater. On the other hand, N and P removal seems to be more affected by HRT, as when HRT increases from 1 to 4 days, N and P removal rates increase up to 4 times [2,23]. Munoz *et al*. [21] suggest that artificial aeration (1.13 m^3/min) leads to clogging elimination. Although clogging is a major problem in CW operation, continuous aeration greatly increases operation cost.

VF AND HYBRID CWS TREATING AGRO-INDUSTRIAL WASTEWATERS

In contrast to FWS and HSF, VF CWs (Table 5) can treat extremely higher pollutant surface loads ranging from 10 to 6589 gr/m^2 day for organic matter, 0.6 to 575 g/m^2 per day for TKN, 0.08 to 20 g/m^2 per day for phosphorus, and 35 to 1836 g/m^2 per day for TSS. Although VF CWs received pollutant surface loads up to 10 times greater than FWs and HSF CWs, they appear to be extremely efficient as removal rates range from 24 to 95% for organic matter, 10 to 99% for TKN, 47 to 95% for TP, and from 21 to 99% for TSS. For some specific agro-industrial wastewaters such as OMW [32––34,51], VF CWs show high removal efficiencies for all pollutants (72–86% for COD, 75% for nitrogen, 88–95% for phosphorus, 79% for phenols), while the pollutant surface loads applied were among the highest reported for all agro-industrial wastewaters. It appears from the literature that VF CWs are more efficient at organic matter and phenol removal, but attention should be paid to the existence of TSS, which can cause porous media to clog and thus damage the CW system. Yalcuk *et al*. [51] attributed VF CW treatment ability to the efficient diffusion of oxygen in CW pores. The higher oxygen concentration in VF CWs results in increased organic matter oxidation and ammonia nitrification. Nevertheless, TN removal in VF CWs is limited due to insufficient denitrification. A viable solution to this problem is to recirculate treated wastewater, which leads to increased denitrification rates [28,31].

While the majority of the published experiments/applications used common gravel as porous media, Yalcuk *et al*. [51] examined zeolite efficiency in OMW treatment. They reported that the use of zeolite increased organic matter and ammonia removal, which is consistent with the results of other research groups [68,69].

Although VF CWs usually operate under aerobic conditions, the extremely high pollutant loads in agro-industrial wastewater could lead to anoxic/anaerobic conditions. For this reason Poach *et al*. [37] tried to increase DO concentrations in VF CWs by increasing the number of drainage periods. From these experimental results it was concluded that increased drainage periods do not increase pollutant removal efficiencies [37]. To overcome the problem of low DO concentrations, Babatunde and Zhao [65] used VF CWs with tidal flow strategies which can promote oxygen supply. When

Table 5. VF and hybrid CWs treating agro-industrial wastewater treatment by CWs

Ref.	Wastewater type	CW surface area, m^2	Surface load, g/m^2 per day				Removed surface load, g/m^2 per day			
			C	TKN	TP	TSS	C	TKN	TP	TSS
					VF					
[14]	Winery	127–850	23.6–35.2	0.6–2.7	0.08–0.2	–	22.4–33.4	0.4–1.9	–	–
[32]	OMW		15	–	–	–	12.9	–	–	–
[33]	OMW		114.71	–	OP: 2.74	–	82.5	–	OP: 2.6	–
[34]	OMW		6589 Phenols: 997	175	20.0	–	4810 Phenols: 748	131	17.6	–
[41]	Animal farm	0.5	51.7	22.5	1.84	14	31.0	15.8	1.1	13.3
[48]	Animal farm	n.a.	36–474	46–79	8–174	73–1836	21.6–284	20.7–35.6	6.4–139	–
[52]	Cheese whey	0.03	10	–	–	–	3	–	–	–
[65]	Animal farm	575	–	575	–	35		491	–	32.8
					Hybrid					
[15]	Dairy	1990	1.28	0.74	0.10	1.96	1.2	0.68	0.09	1.92
[19]	Animal farm	0.5	51.7	22.5	1.84	14	31.0	15.8	1.1	13.3
[25]	Animal farm	120	–	1.4–1.5	–	–	–	0.98–1.05	–	–
[26]	Vinegar	730	23.4	0.3	0.06	–	67	83	62	–
[28]	Winery	350	30.4	–	–	–	22.3	–	–	–
[31]	Dairy, swine and potato starch	168–2151	24–92	–	–	–	19.2–73.6	–	–	–
[63]	Dairy	80	50–1500	50–1500	1.5–40	20–400	45–1350	32.5–975	0.8–21	18.8–376
[67]	Dairy	1.87	173.5	–	2	22.3	156	–	1.6	20.1

treating wastewaters containing high organic loads, where the oxygen supply from the plant root zone is insufficient, the tidal flow VF CWs increases DO concentration in the biofilm and thus enhances organic matter degradation. Thus organic surface load can increase up to 112g BOD/m^2 per day, and still maintain aerobic conditions [37]. VF CW tolerance to such high organic loads has also been attributed to tidal flow operation and not vegetation as *Phragmites* can only release 0.02 g O_2/m^2 per day [37].

The most efficient CW system for agro-industrial wastewater treatment appears to be a hybrid system (Table 5) of both VF and HSF stages. This system has achieved the highest removal rates among all CWs types, ranging from 83 to 96% for organic matter, 55 to 92% for TKN, 52 to 96% for TP, and 83 to 99% for TSS, while receiving high pollutant surface loads (1.28 to 1500 g/m^2 per day for organic matter, 0.3 to 1500 g/m^2 per day for TKN, 0.06 to 40 g/m^2 per day for phosphorus and 1.96 to 400 g/m^2 per day for TSS).

DISCUSSION – DESIGN AND OPERATIONAL SUGGESTIONS

As agro-industrial wastewaters have common characteristics (*i.e.*, high organic loads, low pH values, toxic effects) the CWs treating them present the same problems. Thus, general suggestions can be made on the design and operation of CWs treating agro-industrial wastewaters. The following suggestions concern issues such as temperature, prior-treatment stage, chosen vegetation, porous media and CW plain view.

Temperature usually affects CW performance as the main pollutant mechanism, biological degradation, is temperature-dependent. Lee *et al*. [67] examined organic matter and TSS temporal variations, and found that temperature did not significantly affect their removal. This was also observed by Akratos and Tsihrintzis [70], who state that organic matter degradation is not affected by temperature, because aerobic and anaerobic bacteria responsible for organic matter degradation can function even at low temperatures (5 °C). Contrary to these observations, Newman *et al*. [11] and Mustafa *et al*. [59] reported that nitrogen and phosphorus removal are strongly temperature-dependent. Akratos and Tsihrintzis [70], who also reported a strong correlation between nitrogen removal and temperature, claim that this phenomenon could be attributed to plant uptake and that nitrifying bacteria increase their performance in temperatures above 15 °C.

Concerning vegetation in CW systems treating agro-industrial wastewater, although numerous plant species have been tested, no specific species has been proved to be superior over the others for pollutant removal. The main concern is to select a species resistant to the toxic wastewaters involved and which is indigenous to the geographical area. Tests indicate that the most tolerant plant species is *Phragmites* spp. (the common reed), as it appears resistant to the toxic effects of dairy, animal farm, winery and olive mill wastewaters, and demonstrates high pollution removal efficiencies. Furthermore, it can be found free-growing in most areas. To overcome vegetation species limitations and achieve higher removal rates, Harrington and McInnes [71] propose the use of a variety of vascular plants as long as pollutant concentrations have no toxic effects on vegetation.

Another issue concerning vegetation is the density of the plants sown. Research results show that increased plant density increases pollutant removal. Furthermore, initial dense vegetation could also minimize the adjustment period and possible toxic effects. Therefore, in CWs treating agro-industrial wastewater, the initial planting density should increase from 4 [70] to 6 or 8 plants/m^2, depending on the toxicity of the wastewater concerned. As the plants themselves are responsible for only a small percentage of nutrient removal (around 3%) [11,36], there is no need for their periodic removal. On the contrary, this action would cause a decrease in pollutant removal efficiency, as the oxygen levels in the CW would decrease. The CW should only be replanted when the initial vegetation turns yellow and dies. Removal of dead plants avoids increasing nutrient concentrations in the CW caused by the deposition of decaying plant biomass.

Despite the numerous experiments/applications of CWs treating agro-industrial wastewater, in the majority of cases effluent pollutant concentrations remain above EU recommended limits (Table 6), thus prohibiting its direct use for irrigation or disposal. Effluent concentrations below EU limits were reported only in cases where influent concentrations were very low [2,3,10,12,13,17,59,61]. Therefore, when designing CWs for agro-industrial wastewater treatment various parameters should be considered. Figure 1 presents a decision tree for the design of CWs treating agro-industrial wastewaters and provides a rough tool for scientists and engineers. The first designing step should be the appropriate selection of the prior-treatment stage, as the majority of the experiments/applications presented included either a prior-treatment stage or used diluted wastewaters to eliminate toxic effects on CW vegetation. Depending on the presence of colour and toxic substances and the low or high organic load in agro-industrial wastewaters, prior-treatment stages could include just settling tanks for COD and TSS removal when initial organic loads are low. While biological pre-treatment stages should be used when organic loads are high and toxic substances are present in the wastewaters. We propose the use of biological treatment systems that can tolerate high pollutant loads

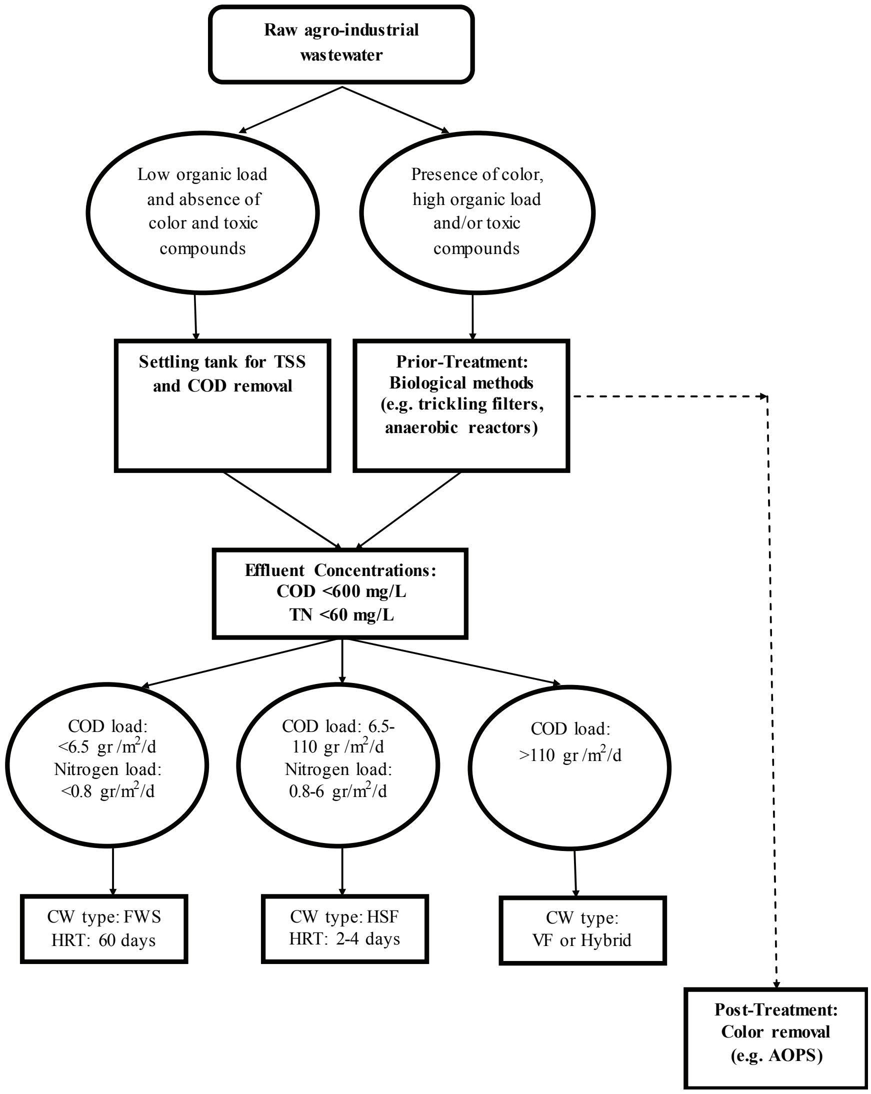

Figure 1. Decision tree for designing CWs treating agro-industrial wastewater.

(*e.g.*, trickling filters, anaerobic reactors and common suspended growth reactors). From the published research it is deduced that CWs were used mainly as polishing treatment stages for agro-industrial wastewater treatment. Bearing in mind that the main advantage of CWs is their low operational cost, they should be coupled with other low-cost treatment technologies (*e.g.*, biological trickling filters, coagulation-flocculation). Biological trickling filters have been successfully used in combination with CWs for OMW treatment [34,72]. The presence of colour in agro-industrial wastewater complicates design as a post-treatment stage should also be included for colour removal. This post-treatment stage usually involves physicochemical methods (*e.g.*, electro-oxidation, ozonation, advanced oxidation processes – AOPS) [33].

Selection of the suitable CW system is also a crucial issue for agro-industrial wastewater treatment. This issue should be considered together with pollutant surface load, as excessive loads negatively affect pollutant removal rates [2,19]. Therefore, CW selection should be based on the maximum pollutant loads that each CW type can tolerate, while effluent concentrations should be below the legislated limits for discharge into a municipal sewerage system. Although all CWs present a wide range of pollutant surface load removal rates (Tables 2–5), design suggestions (Fig. 1) are based only on pollutant loads that lead to effluent concentrations below permitted limits for discharge into a sewer system (Table 6). Specifically, for low pollutant loads (up to 6.5 gr COD/m^2 per day and 0.8 g N/m^2 per day) FWS operating with an HRT of 60 days can achieve effluent concentrations below the permitted limits for discharge into a municipal sewer system (Fig. 1). Cronk [7] has proposed some guidelines for CWs treating wastewater either from animal farms or dairy factories. These guidelines concerned mainly BOD removal and could be characterized as rather conservative, as for a FWS CW

Table 6. Pollutant influent and effluent concentrations compared with EU standards (EU Directive 1991/271/EEC)

Ref.	Influent concentration, mg/L					Effluent concentration, mg/L				
	COD	TKN	$N–NH_4^+$	$N–NO_3^-$	TP	COD	TKN	$N-NH_4^+$	$N–NO_3^-$	TP
EU standards						120		10		2
[2]	150	20	40	4.5	4	15	3.2	0.2	0.14	2.2
[3]	1747	237	188	3.7	37	34	19	14	0.6	17
[10]	682	97	74	0.6	10.4	89	63	25	32	4.7
[11]	2700	102	7.7	0.3	26	611	74	52	0.1	14
[12]	1900	164	72	5.5	53	53	3.3	32	9.9	2.7
[13]	1219	65	22	0.5	13	98	33	25	0.5	5
[18	2240	135	118	0	–	658	34	14	7.5	–
[19]	1034	–	448	–	–	310	–	134	–	–
[23]	–	38	–	–	11	–	23	–	–	7.5
[24]	–	907	366	–	–	–	248	221	–	–
[27]	254	29	18	–	17	158	17	11	–	13
[29]	1160	200	185	3.7	40	190	156	144	1.7	21
[32]	129100	90	–	–	–	95100	560	–	–	–
[34]	14000	506	123	–	95	3500	99	36	–	12
[35]	3220	1333	1072	–	–	2200	333	313	–	–
[37]	445	66	–	–	71	280	30	–	–	65
[39]	796	171	139	0.6	–	471	87	66	0.6	–
[40]	285	296	196	<2	–	277	247	128	<2	–
[41]	2500	–	90	2	40	625	–	60	–	19
[43]	2000	–	0.3	–	5.3	210	–	0.1	2.3	6.4
[44]	1200–3500	–	–	–	–	10–120	–	–	–	–
[45]	3167	104	4.4	101	265	950	26	0.6	9.3	53
[46]	1700	360	–	–	58	115	7.8	–	–	0.8
[51]	2880	–	0.9	–	68	750	–	0.5	–	3.5
[59]	1500	–	40	3.4	–	75	–	0.4	1	–
[62]	–	20–120	–	–	20–50	–	5–15	–	–	10–30
[66]	–	116	86	–	56	–	70	53	–	48

the maximum BOD surface was proposed to be 73 kg/ha per day, with minimum HRT of 12 days.

Subsurface flow CWs (HSF and VF) are more efficient bioreactors and are preferred for the treatment of heavily polluted wastewaters as they can receive high loads of both organic matter and N. Specifically, HSFs appear to be rather effective at agro-industrial wastewater treatment as they can tolerate pollutant loads up to 110 g COD/m^2 per day and 6.5 g N/m^2 per day (Fig. 1). Additionally, the HRTs tested in HSF CWs are significantly lower compared to those used in FWSs and range from 2 to 4 days.

Although VF and hybrid CWs have not been used extensively for agro-industrial wastewater treatment they have been recorded to receive high organic loads (over 110 g COD/m^2 per day), and achieve high removal rates (up to 90%, Table 5). Although final effluents are above permitted limits for discharge into municipal wastewater systems, VF and hybrid CWs should be preferred when organic loads exceed 110 g COD/m^2 per day (Fig. 1; Table 6).

To overcome the limitations of each CW system, several points should be highlighted:

- The main advantage of FWSs is the higher DO concentration observed compared to that of HSF and VF systems. On the other hand, FWS CWs are not preferred for agro-industrial wastewater treatment as they require higher HRTs than HSF and VF CWs. This problem was overcome by Stefanakis and Tsihrintzis [73], who combined FWS and HSF by raising the water level in an HSF system. This new CW type achieved similar removal rates to the HSF system despite receiving 15-20% increased pollutant loads.
- During the operation of many CWs treating agro-industrial wastewaters, plant density was recorded to be higher near the CW inlet. This is attributed to higher nutrient concentrations at and around the inlet. This phenomenon confirms that horizontal flow CWs (FWS and HSF) function as plug-flow reactors and

therefore the biochemical processes occurring are more intense near the inlet. This phenomenon could be exploited to increase CW efficiency in two ways. The first is to change the design of the CW plain view from the common rectangular shape to trapezoidal. This would increase the area near the inlet where nutrient availability is higher, plant density is higher and biochemical processes are more intense. This trapezoidal design was tested by Kotti *et al.* [74], who used FWS CWs for municipal wastewater treatment and found that the trapezoidal CW was cca. 8% more efficient at pollutant removal than the rectangular CW. The second method is to use multiple inlet points along the units. In this way nutrient availability increases along the length of CW, therefore increasing both plant density and pollutant removal. This scenario was tested by Stefanakis *et al.* [75] who used three different inlet points and two different schemes (33:33:33 and 60:25:15). Their results showed that a gradual wastewater inflow from multiple inlet points (60:25:15) significantly increased pollutant removal efficiencies (up to 5%).

Another CW design issue is the origin of the soil (FWS) or porous media (HSF and VF) used. The type of soil/porous media is important as it could positively affect N and P removal. Most studies have not examined thoroughly the effect of CW substrate, however one attempt was made to use zeolite [68]. Zeolite, which is a natural absorbent, has also been used as a substrate in post-treatment filters that increase organic matter and ammonia removal [68,69]. Other substrate materials tested include bauxite, flying ash, river gravel and quarry gravel [68–70,76,77]. From the literature it can be concluded that substrate origin and chemical composition is critical, as minerals with reactive Fe and Al or calcareous materials that promote Ca phosphate precipitation are rather efficient at phosphorus removal, and materials with high cation exchange ability promote ammonium removal.

The major problem in CW application is porous media clogging, especially when treating wastewaters containing high TSS concentrations, such as agro-industrial wastewaters. Future research should focus on testing materials that would increase CW porosity and thus limit the clogging effect. These materials could include the plastic materials already used in biological trickling filters [72], where biofilm density is higher than in CWs.

CONCLUSIONS

CWs appear to be rather effective in treating agro-industrial wastewaters after a prior treatment stage. Although CWs were originally designed to receive low pollutant loads they have been proved to tolerate extremely high pollutant loads and still achieve high pollutant removal rates. Although the presence of vegetation positively influences pollutant removal, different species do not significantly affect CW performance. Based on the above literature review, some initial design guidelines for CWs treating agro-industrial wastewaters can be suggested including:

a) Prior treatment stages are imperative before the CW stage. When initial organic loads are low and no toxic substances are present in the wastewater, a simple settling tank suffices for COD and TSS removal. However, when organic loads are high and toxic substances are present, a biological treatment system should be used before the CW stage.

b) To achieve effluent concentrations that meet permitted limits for discharge into a municipal sewerage system, the appropriate CW system and HRT should be selected according to the organic and N loads of the wastewaters concerned.

c) A post-treatment stage is imperative in cases where colour is present in the wastewaters.

d) To further improve CW performance on agro-industrial wastewater treatment specific design and operation parameters should be examined, including CW plain, vegetation density, step feeding, special porous media, etc.

Acknowledgement

The first author gratefully acknowledges a graduate studies scholarship from the Hellenic State Scholarships Foundation.

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IZVOD

UPOTREBA PLANSKIH MOČVARA U TRETMANU AGROINSTUSTRIJSKIH OTPADNIH VODA: PREGLED

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(Pregledni rad)

Širom sveta se planske močvare upotrebljavaju u tretmanu otpadnih voda zbog svoje jednostavnosti i niskih troškova izrade. Njihova upotreba se više ne ograničava samo na komunalne otpadne vode, već se širi i na tretman jako zagadjenih otpadnih voda kao što su agro industrijske otpadne vode. Ovaj rad pruža sveoubhvatan pregled literature koja se bavi aplikacijom planskih močvara za potrebe tretiranja različitih agro industrijskih otpadnih voda, kao i pitanja nagomilanog otpada na površini vode, ulogu tipova konstruisanih tresetišta, faze pre samog tretmana, biljne vrste koje su efikasne u otklanjanju zagađivača. Rezultati ukazuju da konstruisana tresetišta mogu da tolerišu visok nivo zagadjivača i toksičnih supstanci bez gubljenja svojih osobina, i na taj način su ovi sistemi veoma efikasni kao bio reaktori u zagadjenom okruženju. Ovaj rad predlaže pitanja koja mogu da poboljšaju efikasnost tretmana i predlaže dizajn i predloge u vidu odgovarajuće vegetacije, poroznih materijala i pregleda konstruisanih tresetišta. Na kraju se predlaže šematski prikaz kao pomoć u dizajniranju tresetišta neophodnih za tretiranje agro industrijskih otpadnih voda koji treba da posluži kao alat za naučnike i inžinjere.

Ključne reči: Konstruisana tresetišta • Agroindustrijske otpadne vode • Pregled • Šematski prikaz • Dizajn