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Potential of wastewater treatment systems based on constructed wetlands for agricultural reuse under the EU framework

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Abstract

Wastewater reuse was recognized as one of the solutions for the problems regarding increasing water scarcity and pollution of water resources. Constructed wetlands (CWs) are a sustainable and cost-effective technology for wastewater treatment. If able to produce effluent of a needed quality, they can be a valuable addition for wastewater reuse schemes. This review studied 39 treatment systems based on CWs, and it assessed their characteristics and performance on pollutants removal. Moreover, their potential to reach the future European Union standards for agricultural wastewater reuse was evaluated. The results showed that the combination of CWs with additional techniques (e.g. UV treatment, anaerobic reactors) can further increase their performance and provide better removal efficiencies in comparison with conventional HSSF and VSSF CWs. Particularly, hybrid systems showed a better removal of organic matter and bacterial indicators than single-stage CWs. Most of the systems considered could reach some of the limits for agricultural reuse in the terms of biochemical oxygen demand and total suspended solids, although improved single-stage CWs and hybrid systems were able to meet stricter requirements. However, that was often not the case with *Escherichia coli* and therefore it is recommended to combine them with disinfection technologies in order to reach the levels required for agricultural reuse.

Key words: wastewater treatment, constructed wetland, agricultural wastewater reuse

1. Introduction

Currently, water scarcity is becoming a worldwide risk (Mekonnen and Hoekstra, 2016). The severe pressure on water resources is mainly attributed to global population growth, expansion of irrigation

agriculture, economic development and climate change (Gosling and Arnell, 2016; Hamadeh et al., 2014; Mekonnen and Hoekstra, 2016; Tao et al., 2017). In fact, existing natural freshwater resources seem to be inadequate to satisfy various ever-increasing demands (Almuktar et al., 2018) and therefore imbalance between water demand and water supply (Ghaitidak and Yadav, 2013). Moreover, discharging wastewater without previous treatment may not only lead to a certain “waste” of water resources, but can also harm different ecosystems (Lavrnica et al., 2018). On the other hand, treated wastewater is regarded as an alternative resource for water supply, and also has the potential to be used for different purposes (Hamadeh et al., 2014; Tao et al., 2017). Under such circumstances, research on wastewater reclamation strategies is being carried out in order to address increasing demand for water resources and to prevent further deterioration of water quality (Almuktar et al., 2018; NAS, 2016).

Apart from conventional wastewater treatment technologies, nature-based solutions, especially CWs, have been used worldwide for treating wastewater and improving its quality (Hamadeh et al., 2014). CWs are engineered ecological system that can treat wastewater through different natural processes which are under the influence of the combined action of aquatic plants, soils and microorganisms (Hamadeh et al., 2014; Tao et al., 2017).

CWs are viewed as a cost-effective and sustainable option for wastewater treatment (Arden and Ma, 2018; ElZein et al., 2016; Lee et al., 2009). Their main advantages are good removal efficiency, simple and low cost construction, running and maintenance, nutrients recycle and esthetic values (Brix et al., 2011; Chen, 2011; Hamadeh et al., 2014; Liu et al., 2015; Rousseau et al., 2008). However, CWs also have certain shortcomings. For example, there is a certain risk of bed clogging, especially under high loading rates of organic and suspended solids (SS). It can later cause hydraulic malfunction and decrease overall treatment performance, even further shortening the lifespan of systems (Aiello et al., 2016; Jozwiakowski et al., 2018; Kim et al., 2016; Ruiz et al., 2010). Besides, nitrogen removal efficiency of CWs is sometimes limited due to insufficient conditions for denitrification and nitrification (Jozwiakowski et al., 2018; Liu et al., 2015; Wu et al., 2014). Also, pathogen removals could be low due to the lack of disinfection treatment or other chemical agents (Andreo-Martinez et al., 2017), and unsatisfactory performance of single-stage CW (Toscano et al., 2015; Zurita and White, 2014). In order to overcome these problems, it can be a good solution to integrate CWs with right disinfection measures (Arden and Ma, 2018), artificial aeration technologies, anaerobic baffled reactor (ABR), even combining different types of CWs, etc.

CWs effluents can be reused in irrigation, gardening, flushing toilet, groundwater replenishment and other public and industrial utilizations (Angelakis and Snyder, 2015; Barbagallo et al., 2014; Dou et al., 2017; Rousseau et al., 2008). Being one of the biggest consumers of freshwater resources in the world, agriculture is under a constant threat of climate change and water scarcity, and in order to ensure sufficient crop production, additional water resources (e.g. treated wastewater) need to be used. Nevertheless, although wastewater reuse can be beneficial for agricultural irrigation, negative effects of this practice (e.g. on soil or plants) should be considered. Thus, it is important to regulate this area and prevent negative consequences on the environment. Several countries (e.g. Italy, Spain, U.S.) have already implemented regulations and limits for wastewater reuse in agriculture (Andreo-Martinez et al., 2017; Jokerst et al., 2011; Licciardello et al., 2018). Some of these countries provide detailed classification of irrigation water quality according to crop categories, irrigation methods and areas (Ayaz et al., 2015; Russo et al., 2019). Also, the European commission proposed a regulation on minimum requirements for water reuse in agriculture (European Commission, 2018). However, wastewater treated by CWs cannot always satisfy these standards related to safe reuse (Arden and Ma, 2018; Garcia et al., 2013; Jokerst et

al., 2011; Lavrnic et al., 2017).

Therefore, the main objective of this paper is to provide an overview on the design, characteristics, as well as performance of CWs treating domestic wastewater in order to assess their potential for wastewater reuse within the new EU framework (European Commission, 2018).

2. Materials and methods

This review mainly focuses on domestic wastewater treatment systems based on CWs. It is a result of the search of Web of Science database using keywords such as “constructed wetland”, “domestic water/wastewater treatment” and “water/wastewater reuse (in agriculture)”. The time period of publication was set from 2008 to 2019. According to the main objective of this review, 39 research publications which focus on domestic wastewater treatment, pollutants removal effect and reuse in agriculture were selected to be discussed. On the other hand, papers which did not report pollutant concentration or removal were excluded from the selection.

The 39 articles analyzed cover 19 countries. Regular domestic wastewater (a mixture of kitchen, shower, toilet etc.) treatment is the main topic of 77% of the articles considered, while greywater (domestic wastewater from non-toilet sources) and blackwater (domestic wastewater from toilets) amount to 18% and 5%, respectively. Hybrid CWs or CWs combined with other systems are 64% of the total, while single-stage CWs are 36%. They were mostly of pilot-scale (51%), but full-scale (31%) and lab-scale studies (18%) were also well represented. Regarding experimental duration, 49% of the research studies lasted for more than one year, 33% were between six months and one year long, 10% were shorter than six months, while the remaining 8% did not provide a specific time. Plant species applied or tested in these studies were either a mixture of various species or different species planted separately.

In Table 1 are listed 39 selected experimental case studies. The efficacy of these various wastewater treatment systems was analyzed and conformity of their effluents to reuse limits evaluated. This research did not introduce studies on free surface flow CWs, since not many cases suitable for this review were reported in the time frame considered. It was also noted that some studies in hybrid system section (Table 1) tested more than one system, either with similar (No. 16, 17, 18, 20, 35 and 37) or different configurations (No. 27a, 27b, 36a and 36b).

In the case when the paper did not provide influent, effluent concentration or removal efficiency, the following equation was used to calculate the missing variable:

$$\frac{C_i - C_e}{C_i} = E \quad (1)$$

Where C_i = influent concentration of a pollutant (mg L^{-1}), C_e = effluent concentration of a pollutant (mg L^{-1}), and E = removal efficiency of a pollutant in a system.

Table 1. 39 Selected case studies of wastewater treatment systems recorded in the literature from 2008 to 2019.

CW types	System No.	Experimental scale	Country	Influent	Experimental period	Vegetation	Reference
Single-stage CW							
HSSF	1	Pilot-scale	Italy	RDW	>1 year	<i>Cyperus alternifolius</i> L. (unit 1), <i>Typha latifolia</i> L. (unit 2), unplanted (unit 3)	Tuttolomondo et al. (2016)
	2	Pilot-scale	Italy	RDW	6-12 months	<i>Vetiveria zizanioides</i> , <i>Miscanthus x giganteus</i> , <i>Arundo donax</i> , <i>Phragmites australis</i>	Toscano et al. (2015)
	3	Pilot-scale	Spain	RDW	>1 year	<i>Phragmites australis</i>	Morato et al. (2014)
	4	Pilot-scale	Turkey	RDW	>1 year	<i>Cyperus</i>	Ayaz (2008)
	5	Full-scale	Poland	RDW	>1 year	<i>Salix viminalis</i> L.	Jozwiakowski et al. (2018)
	6	Pilot-scale	Spain	RDW	6-12 months	<i>Phragmites australis</i>	Andreo-Martinez et al. (2017)
VSSF	7	Pilot-scale	Italy	RDW	>1 year	<i>Typha latifolia</i> , <i>Phragmites australis</i>	Morari and Giardini (2009)
	8	Pilot-scale	Egypt	RDW	>1 year	<i>Canna</i> , <i>Phragmites australis</i> and <i>Cyperus papyrus</i>	Abou-Elela and Hellal (2012)
	9	Pilot-scale	Greece	RDW	6-12 months	<i>Atriplex halimus</i> , <i>Juncus acutus</i> and <i>Sarcocornia perennis</i> , <i>Phragmites australis</i>	Fountoulakis et al. (2017)
	10	Lab-scale	UK	RDW	>1 year	<i>Phragmites australis</i>	Almuktar et al. (2017)
Improved system GROW	11	Pilot-scale	India	GW	>1 year	8 varieties of Indian native plant species	Ramprasad et al. (2017)
Improved system	12	Lab-scale	Israel	GW	>1 year	<i>Juncus alpigenus</i> and <i>Cyperus haspen</i>	Sklarz et al. (2009)
RVFCW	13	Lab-scale	Israel	GW	<6 months	<i>Hydrocotyle leucocephala</i> and <i>Cyperus papyrus</i>	Travis et al. (2010)
Improved system upflow subsurface CW using green sorption media	14	Pilot-scale	USA	GW	<6 months	<i>Juncus effuses</i> (cell 1), <i>Panicum hemitomon</i> (cell 2), <i>Zizaniopsis miliacea</i> (cell 3)	Xuan et al. (2009)
Hybrid systems							

VSSF-HSSF	15	Full-scale	South Korea	RDW	>1 year	<i>Phragmites australis</i> and <i>Phragmites japonica</i> (VF), <i>Miscanthus sacchariflorus</i> , <i>Carex dispalata</i> , <i>Juncus effuses</i> , and <i>Iris pseudacorus</i> (HF)	Kim et al. (2016)
Two VSSF-HSSFs in parallel ^a	16	Pilot-scale	Spain	RDW	6-12 months	<i>Phragmites australis</i> and <i>Scirpus</i> sp.	Melian et al. (2010)
Four separate HSSF-VSSFs ^b	17	Pilot-scale	Iran	RDW	6-12 months	<i>Phragmites australis</i> , <i>Typha latifolia</i> , <i>Arundo donax</i> , unplanted	Haghshenas-Adarmanabadi et al. (2016)
HSSF-HSSFs, VSSF-VSSFs, VSSF-HSSFs ^c	18	Lab-scale	Colombia	RDW	Not provided	<i>Papyrus</i>	Garcia et al. (2013)
FWS-SSF	19	Pilot-scale	USA	GW	6-12 months	<i>Typha latifolia</i> (FWS), <i>Scirpus acutus</i> (SSF)	Jokerst et al. (2011)
HSSF-VSSFs ^d	20	Lab-scale	Spain	RDW	>1 year	Common reed and <i>Papyrus</i> (HSSF), not provided (VSSF)	Herrera-Melian et al. (2018)
IVFCW-HSSF	21	Pilot-scale	China	RDW	6-12 months	<i>Canna indica</i> L. (down-flow VSSF), <i>Juncus effusus</i> L. (up-flow VSSF), <i>Scirpus validus</i> Vahl (HSSF)	He et al. (2018)
Saturated VSSF-free-drain VSSF-HSSF	22	Pilot-scale	Czech Republic	RDW	>1 year	<i>Phragmites australis</i> (saturated VSSF), <i>Phragmites australis</i> (free-drain VSSF), <i>Phalaris arundinacea</i> (HSSF)	Vymazal and Kropfelova (2015)
VSSF-HSSF-FWS	23	Full-scale	Spain	RDW	Not provided	<i>Phragmites australis</i> (VSSF), <i>Phragmites australis</i> (HSSF), <i>Typha</i> spp., <i>Scirpus</i> spp., <i>Iris pseudacorus</i> , <i>Carex flacca</i> , <i>Cyperus rutundus</i> and <i>Juncus</i> spp. (FWS)	Avila et al. (2015)
Settling cum equalization tank-UFDF sand filter-	24	Full-scale	India	GW	6-12 months	<i>Canna indica</i>	Patil and Munavalli (2016)

HSSF-charcoal filter-water hyacinth system							
BCO pretreatment-greenhouse-structured HSSF	25	Full-scale	China	RDW	>1 year	ornamental plants including <i>Hemerocallis lilioasphodelus</i> L., <i>Iris tectorum</i> , <i>Oxalis violacea</i> , <i>Sedum erythrostictum</i> Mig and <i>Hosta ensata</i>	Gao and Hu (2012)
Ice-block unit ^e -VSSF	26	Pilot-scale	Mongolia	GW	6-12 months	not provided	Uddin et al. (2016)
HSSF-lagooning	27a	Full-scale	Italy	RDW	6-12 months	<i>Phragmites australis</i> , <i>Typha latifolia</i>	Russo et al. (2019)
HSSF-UV treatment	27b	Full-scale	Italy	RDW	6-12 months	<i>Phragmites australis</i> , <i>Typha latifolia</i>	Russo et al. (2019)
RVFCW-UV disinfection	28	Lab-scale	Israel	RDW	>1 year	<i>Cyperus haspen</i> , <i>Juncus alpigenus</i> and <i>Hydrocotyle vulgaris</i> L.	Sklarz et al. (2013)
SSF-UV/TiO ₂ /O ₃	29	Pilot-scale	Brazil	BW	>1 year	<i>Hymenachne grumosa</i>	Horn et al. (2014)
Anaerobic pretreatment-HSSF-VSSF	30	Pilot-scale	Turkey	RDW	<6 months	<i>Phragmites australis</i>	Ayaz et al. (2015)
HUSB reactor-VSSF-HSSF-FWS	31	Lab-scale	Spain	RDW	>1 year	<i>Phragmites australis</i>	Avila et al. (2016)
OP-FWS-Cascade-FWS-SSF	32	Full-scale	China Taiwan	RDW	6-12 months	<i>Typha latifolia</i> and <i>Phragmites australis</i> (FWS), <i>Phragmites australis</i> (SSF)	Yeh et al. (2010)
FP-FWS-SSF	33	Full-scale	Spain	RDW	6-12 months	<i>Typha latifolia</i> (SF), <i>Salix atrocinerea</i> (SSF)	Reinoso et al. (2008)
Sedimentation tank-HSSF-VSSF	34	Pilot-scale	Egypt	BW	Not provided	<i>Phragmites</i>	Abdel-Shafy et al. (2017)
Combinations of HSSF, VSSF or stabilization pond ^f	35	Pilot-scale	Mexico	RDW	>1 year	<i>Zantedeschia aethiopica</i> , after 8 months (replaced with) <i>Canna indica</i> (HSSF), <i>Strelitzia reginae</i> (VSSF)	Zurita and Carreon-Alvarez (2015)

HSSF-biological pond-storage reservoir-sand and disk filters	36a	Full-scale	Italy	RDW	<6 months	<i>Phragmites australis</i>	Licciardello et al. (2018)
HSSF-sand and disk filters-UV treatment	36b	Full-scale	Italy	RDW	<6 months	<i>Phragmites australis</i>	Licciardello et al. (2018)
ABR-VSSF/HSSF-FWS ^g	37	Full-scale	Pakistan	RDW	6-12 months	<i>Typha latifolia</i> , <i>Phragmites australis</i> and vetiver grass (VSSF, HSSF), <i>Pistia stratiotes</i> (FWS)	Ali et al. (2018)
Two settling tanks in series-VSSF-a zeolite tank	38	Full-scale	Greece	RDW	>1 year	Unplanted (cell 1), <i>Phragmites australis</i> (cell 2)	Gikas and Tsihrintzis (2012)
A septic tank-an Imhoff tank-two parallel VSSFs-HSSF	39	Full-scale	Spain	RDW	>1 year	<i>Typha latifolia</i> (HSSF)	Vera et al. (2013)

RDW = regular domestic wastewater, GW = greywater, BW = blackwater, GROW = green roof-top water recycling system, RVFCW = recirculating vertical flow constructed wetland, FWS = free water surface constructed wetland, SSF = subsurface flow constructed wetland, IVFCW = integrated vertical flow constructed wetland, UFDF = up-flow down-flow filter, BCO = bio-contact oxidation, UV = ultraviolet, HUSB = hydrolytic up-flow sludge blanket, OP = oxidation pond, FP = facultative pond, ABR = anaerobic baffled reactor

^a System 1: VSSF-HSSF both with lapilli, system 2: VSSF-HSSF both with gravel

^b System 1: *Phragmites* HSSF-*Phragmites* VSSF, system 2: *Typha* HSSF-*Typha* VSSF, system 3: *Arundo* HSSF-*Arundo* VSSF, system 4: unplanted HSSF-unplanted VSSF

^c System 1: HSSF-HSSF planted, system 2: HSSF-HSSF unplanted, system 3: VSSF-VSSF planted, system 4: VSSF-VSSF unplanted, system 5: VSSF-HSSF planted, system 6: VSSF-HSSF unplanted

^d System 1: Mulch-based HSSF-gravel-based VSSFs, system 2: Mulch-based HSSF-mulch-based VSSFs

^e The system functioned as the storage pond of frozen wastewater in winter and transferred to septic tanks for treating melted wastewater in summer

^f System 1: HSSF-stabilization pond, system 2: HSSF-VSSF, system 3: VSSF-HSSF

^g System 1: ABR-Saturated VSSF-FWS, system 2: ABR-HSSF-FWS

3. Single-stage CW

On the basis of wetland flow, CWs are classified into free water surface (FWS) and subsurface flow (SSF). SSF CWs are the most widely used systems (Fonder and Headley, 2013) and can be subdivided into two specific types, horizontal (HSSF) and vertical (VSSF) one. Generally, SSF systems show a better performance than FWS ones, especially when hydraulic loading rate (HLR) is high (Liu et al., 2009). Certain modifications of the original CW types yielded also some novel technologies such as a green roof-top water recycling system (GROW) CW (Avery et al., 2007) and a recirculating vertical flow constructed wetland (RVFCW) (Gross et al., 2007). Improved systems are thought to be an option to save land resource (Ramprasad et al., 2017), optimize organic matter and biogenic compounds removal (Sklarz et al., 2009; Xuan et al., 2009), decrease the likelihood of human contact with wastewater (Sklarz et al., 2009) and environmental risks of its reuse (Travis et al., 2010), meanwhile achieving a relatively good overall treatment efficiency (Ramprasad et al., 2017; Sklarz et al., 2009; Travis et al., 2010). Furthermore, CWs can be filled with different substrates and vegetated by aquatic plants (Chen, 2011; Melian et al., 2010; Toscano et al., 2015), which were suggested to be able to improve pollutants removal (Arden and Ma, 2018; Liu et al., 2009).

3.1. HSSF

Until now, many studies on HSSF CWs have been implemented and reported. More HSSF systems were operated in Europe and United states than VSSF CWs (Nivala et al., 2019), and showed reliable capacity for total suspended solids (TSS) and biochemical oxygen demand (BOD) removal (Lavrnic et al., 2017). However, it is worth noting that these systems may not ensure a stable and good removal efficiency of phosphorus, nitrogen and organics (Andreo-Martinez et al., 2017), as a result of a lack of metal ions (e.g. Ca, Mg, Fe and Al) in conventional substrates (Vohla et al., 2011) and a lack of dissolved oxygen (DO) in water (Vymazal, 2007).

In the west of Sicily, Italy, Tuttolomondo et al. (2016) carried out a two-year experiment on a pilot-scale HSSF CW system (containing three independent units) where one unit was planted with *Cyperus alternifolius*, one with *Typha latifolia* and the third was unplanted. The system was filled with silica quartz river gravel (a particle size of 20 to 30 mm) and operated at a HLR of 12 cm d⁻¹. The mean concentration removal rates of parameters chemical oxygen demand (COD) and BOD₅ are shown in Table 2 (Table 2, No. 1). Evapotranspiration (ET), as a primary factor of the system water balance, had an influence on available treated water volume. The findings showed that the observed removal efficiency of BOD₅ and COD was negatively correlated with ET. Therefore, ET should be taken into consideration especially for arid areas when wastewater reuse in irrigation is the objective. Similarly, in Sicily, Toscano et al. (2015) tested pilot HSSF CWs filled with volcanic gravel to a depth of 0.6 m for tertiary treatment of domestic wastewater from March to November 2012. The system contained two lines, each of which consisted of five parallel HSSF beds. Four were planted with macrophytes (*Vetiveria zizanoides*, *Miscanthus x giganteus*, *Arundo donax*, *Phragmites australis*, respectively) and the fifth one was unplanted. The authors indicated that the vegetated CWs were more effective in contaminants removal. The average concentration removal efficiencies for planted CWs were 92.8% for TSS, 68.1%

for COD and 61.3% for total nitrogen (TN), and they were higher than the ones of unplanted systems that were 89.4%, 55.4% and 43.1%, respectively. The best performance of pollutants removal was attained by the system planted with *Phragmites australis*, with efficiencies of 99.9% for *Escherichia coli* (*E. coli*), 88% for TSS, 63% for COD and 61% for TN (Table 2, No. 2), thus regarded as the most suitable plant species for wastewater treatment in this case. Similarly to the previous study, it was reported that vegetation has affected water balance of systems leading to different ET values measured in planted and unplanted wetlands.

In Spain, Morato et al. (2014) evaluated the effect of design factors (water depth and gravel granulometry) on treatment efficiency of HSSF systems. As can be observed in Table 2, the mean removal efficiencies of COD and BOD₅ were 63.8% and 65%, respectively (Table 2, No. 3). It was found that the system with the water depth of 0.27 m and the size of granular medium of 3.5 mm, was more effective for microbial removal in comparison with other systems (0.5 m water depth, 10 mm size of granular medium). The removal effectiveness in the system with a fine medium may be explained by a larger proportion of water volume contacting with root systems of the vegetation, beneficial to microbial reduction. Microbial removal was primarily attributed to mechanisms of filtration and sedimentation occurring near the inlet of HSSF CWs. Moreover, seasonal variations affected removal of some bacterial groups - a higher removal of *E. coli*, total coliforms (TC) and *Clostridium* spores was achieved in summer, while for heterotrophic plate count it was during winter. Ayaz (2008) also demonstrated the effect of seasonal changes on removal efficiency, since removals of BOD₅ and COD were greater during summer. Furthermore, the author indicated that HSSF CWs were more effective for SS (80%), BOD₅ (65%) and COD (50%) removal in comparison with VSSF and FWS CWs. The only exception was total organic carbon (TOC), which obtained low removal efficiency (sometimes even negative one) in all wetlands due to the additional generation of organic carbon by planted vegetation. The mean removal efficiency of fecal coliforms (FC) and TC both amounted to more than 94% (Table 2, No. 4).

Jozwiakowski et al. (2018) carried out a 14-year investigation on a HSSF wetland in Poland operated under a HLR of 0.6 cm d⁻¹. The 1.2 m deep system was filled with sand and *Salix viminalis* L. was planted in the humus layer distributed over the sand layer. It was observed that for TN and total phosphorus (TP) it was not possible to achieve a continuous and satisfactory removal in the long-term (Table 2, No. 5). It could be explained by the fact that HSSF system did not provide sufficient conditions for nitrification, which in turn negatively affected TN removal. Regarding TP, the sorption capacity of substrate declined over time, causing a lower abatement efficiency of TP during later period of the experiment.

3.2. VSSF

VSSF CWs differ from horizontal ones mainly by flow direction. They have a greater oxygen transfer rate that is beneficial for nitrification and organic matter removal (Sklarz et al., 2009), and that is leading to a lower surface area required in comparison to HSSF CWs (Herrera-Melian et al., 2018; Lavrnic et al., 2017). Several research studies have recently focused on them.

In Italy, two type of pilot-scale VSSF CWs planted with *Typha latifolia* and *Phragmites australis*, respectively, were evaluated for two years by Morari and Giardini (2009). The systems were both filled with gravel (30-50 mm, 4-8 mm and 8-12 mm size of granular medium) and topped with sand (effective size of 0.16 mm). The mean removal efficiency of parameters tested in two years is shown in Table 2 (No.7). It was also found that the systems performed much better in the second experimental year,

especially regarding COD (>93%), BOD (>92%), N (>90%) and K (>86%) removal. The authors attributed such a result to macrophytes that were completely established by the second year. The vegetation showed a positive effect during a treatment process by uptake of nutrients, providing a habitat for microbial populations, etc. However, the treatment efficiency did not differ for tested macrophyte species.

In Egypt, Abou-Elela and Hellal (2012) conducted a pilot-scale VSSF CW experiment for two years. There were three types of macrophytes (*Canna*, *Phragmites australis* and *Cyperus papyrus*) in different sections of this wetland unit. The top 60 cm of the bed were filled with 10 mm gravel and the bottom 25 cm with 20 mm gravel. The average removal rates of TSS, BOD and COD were 92%, 90%, 88% (Table 2, No. 8), respectively. However, different mean concentrations of pollutants accumulated in roots of *Canna*, *Phragmites australis* and *Cyperus papyrus* also proved different vegetation species could influence the removal rates of some contaminant. It was found that *Cyperus papyrus* increased removal of heavy metals, TN and TP, while *Canna* was better for pathogen reduction.

Fountoulakis et al. (2017) reported on the use of a VSSF system planted with halophytes, namely *Atriplex halimus*, *Juncus acutus* and *Sarcocornia perennis*, for treatment of primary treated domestic wastewater in Heraklion, Greece, and compared it with another VSSF CW planted with *Phragmites australis*. Both beds were filled with a 15 cm depth drainage layer of 20-40 mm gravel, a 10 cm depth transition layer with 8-20 mm gravel and a 55 cm depth main layer of 1-3 mm coarse sand, while HLR was 95 mm d⁻¹. The mean removal efficiency of all systems were 78.5% for COD, 26.5% for TN and 30% for TP (Table 2, No. 9). The authors indicated that *Atriplex halimus* was better for salt accumulation (especially Na ones), biomass production and pathogen removal. However, there was no significant difference on the removal efficiency of phosphorus and organic matter among CWs planted with halophytes and common reed, except for slightly lower TN removal rate achieved by the system planted with halophytes.

In the UK, Almuktar et al. (2017) adopted a completely randomized design when testing ten different VSSF CWs located at an aerated greenhouse. They were filled with pea gravel (a depth of 60 cm) differing in four parameters (aggregate diameter, loading rate, contact time, resting time). They were operated for more than 4 years (June 2011 to September 2015). The findings indicated that effluent concentrations of TP (4.2 ± 0.48 mg L⁻¹), NH₃-N (4.2 ± 2.64 mg L⁻¹), potassium (7.0 ± 3.03 mg L⁻¹) and TC (69647 ± 64852.6 cfu 100 mL⁻¹) were significantly higher than the irrigation limits, despite a good treatment efficiency for other pollutants. However, nutrients from the effluents were recycled for irrigation and lead to greater chilies weights and dimensions, consequently more marketable profits. Furthermore, the best quality fruits came from the chilies irrigated by the systems of small aggregate diameters, long resting and contact time, under high inflow loading rates.

3.3. Improved single-stage constructed wetlands

In recent years, there were studies focusing on novel constructed wetlands for wastewater treatment. After some improvements (e.g. location of wetlands, recirculation and aeration), these systems could achieve a greater treatment capacity for some contaminants (e.g. TN, TP) than common single-stage CWs (Martinez et al., 2018; Xuan et al., 2009), while also reducing surface area needed in comparison with hybrid CWs (Ramprasad et al., 2017).

For instance, based on HSSF CWs, GROW was first established in the UK. The system was made up of interconnected weirs and troughs functioning as the beds for wastewater treatment. Besides, it was placed

on the roof in order to save ground space. Thus it was proposed as a viable alternative to treat greywater, addressing the limitation of land resource (Ramprasad et al., 2017). Similarly, a research in the Netherlands proposed the design of a shallow constructed wetroof (Zapater-Pereyra et al., 2016). It was also reported that TN (>87%), TP (>86%), TSS (>80%), COD (>79%) and BOD₅ (>95%) were reduced effectively in the tested system at a low organic loading rate, predominantly due to the treatment of roots, organic soil and sand in the system. In India tropical conditions, from November 2013 to April 2015 Ramprasad et al. (2017) monitored a pilot-scale GROW system planted with eight kinds of local common plants and filled with a 15 cm depth layer of mixed substrates, containing sand, brick bats and gravel (1:1:1). The results showed that overall removal rate of all tested parameters was very high, in particular COD (92.5%), BOD (90.8%), TSS (91.6%), TN (91.7%), TP (87.9%) and FC (91.4%) (Table 2, No. 11). The author pointed out that the high removal of solids was attributed to the baffled CW configuration, which increased the flow path and improved filtration process. It was also concluded that 3.125 cm day⁻¹ was the most suitable HLR for organics removal (BOD and COD) in comparison with other tested HLRs. A higher HLR than 3.125 cm day⁻¹ caused shorter hydraulic retention time (HRT), consequently less removal. Moreover, the system was affected by the factor of seasonal changes, attaining the highest treatment efficiencies of pollutants (i.e. organics, nutrients and FC) in summer, which was consistent with Ayaz (2008).

In Israel, the research related to RVFCW reported that the recirculation was beneficial to organics abatement (Sklarz et al., 2009). In this study, Sklarz et al. (2009) tested two RVFCW systems with or without soil-plant component (a layer of peat vegetated with *Juncus alpigenus* and *Cyperus haspan* for a depth of 8 cm) and revealed that even in the recirculating system without soil-plant component, organics from wastewater can be removed with a high efficiency, namely 95% for BOD₅ and 84% COD on average, besides a 90% TSS removal (Table 2, No. 12). Similarly, in another 40-day study (Travis et al., 2010) on a lab-scale RVFCW planted with *Hydrocotyle leucocephala* and *Cyperus papyrus* and fed with domestic greywater, it was observed that TSS decreased by around 95% and BOD₅ by about 99% (Table 2, No. 13).

In addition, artificial aeration and innovative media were introduced into some CW systems to address a lack of oxygen and the unfavorable adsorption rates of conventional media. Andreo-Martinez et al. (2017) improved a HSSF wetland in Spain by filling it with blast furnace slags (BFS) and feeding it with artificially aerated municipal sewage. Those changes produced higher quality effluents. The data demonstrated that the application of BFS and aeration optimized TP removal. Moreover, the average removal efficiency of turbidity ($99.5 \pm 0.3\%$), TSS ($97.5 \pm 1.3\%$) and TN ($91.5 \pm 5.3\%$) had also been improved due to the aeration (Table 2, No. 6). Sklarz et al. (2009) also pointed out the favorable effect of passive aeration on organics removal in their research. Additionally, Xuan et al. (2009) introduced green sorption media (recycled and natural materials) into upflow subsurface CWs and tested them for three months. It was found that TP and TN were effectively reduced by 94.9% and 75.4% (Table 2, No. 14) in the CWs and reduced by 95.8% and 81.3% in the integrated system with the combination of a septic tank and CWs, respectively. It was verified the tested CWs had a good capacity for TN and TP abatement, especially for TP, which was hardly affected by the septic tank.

Table 2 shows the mean concentration removal efficiency of investigated systems coming from previously discussed 14 single-stage CW studies. Actually, the removal efficiency of pollutants can be assessed by two methods - using concentration or mass load for calculation. Values attained from the two methods were similar when ET was low, while obviously different under very high ET in summer, as Tuttolomondo et al. (2016) stated in their study. High ET losses caused unsatisfactory residual

concentration of pollutants, as they concentrated the elements in effluents, offsetting the effect of treatment (Morari and Giardini, 2009). Therefore, mass load removal efficiency is considered to be more accurate (Tuttolomondo et al., 2016). However, most researchers provide concentration removals in their studies due to easier measurement of related parameters.

Among all the systems considered (Table 2), improved systems GROW and RVFCW displayed superior overall removal efficiency, beyond 90% for the most of contaminants measured (Table 2, Nos. 11-13). According to Table 2, the majority of VSSF and HSSF CWs reached a good effectiveness of removing solids. Generally, VSSF systems (Table 2, Nos. 7-10) showed better effect on organic matter removal in comparison with HSSF ones (Table 2, Nos. 1-5). One exception was the improved HSSF system (Table 2, No. 6), that showed a greater capacity of nutrients removal comparing with other HSSF and VSSF CWs. Regarding bacterial indicators, the HSSF systems No. 2, 4 and the VSSF system No. 8 (Table 2) removed them most effectively. Their abatement higher than 94% could be related to different factors: i) longer HRT (Ayaz, 2008), ii) high temperature and oxygen concentration in the VSSF CW, providing aerobic environment unfavorable for coliforms survival (Abou-Elela and Hellal, 2012). It was revealed that organics and solids removal were also high in the system No. 8 (Table 2). Settleable organics could be eliminated through filtration and deposition, while the high removal of organic compounds can be explained by the fact that this system provided both oxygen and a more favorable habitat for microorganisms due to the presence of various plants. Moreover, the diversity of roots increased HRT, beneficial for pollutants removal (Abou-Elela and Hellal, 2012).

The role of plants was reported in several studies. Plant types (i.e. macrophytes and halophytes) affected pollutant removal rates (Fountoulakis et al., 2017). Some research stated the positive effect of plants (Morari and Giardini, 2009; Toscano et al., 2015). In contrast, plants can also result in the pollutant concentrations increase by the generation of organic carbon (Ayaz, 2008) and the higher ET values (Toscano et al., 2015). Furthermore, Morari and Giardini (2009) highlighted the macrophytes did not show their full capacity by the time they were mature.

Seasonal variations affect the removal capacity of systems. It is found that higher removal efficiencies of organics, microbes (e.g. *E. coli*, FC and TC) and nutrients can be achieved in summer (Ayaz, 2008; Morato et al., 2014; Ramprasad et al., 2017).

Interestingly, the water depth also plays a role in removal performance. Shallow CWs (e.g. GROW, constructed wetroof), providing high aerobic conditions for pollutants and a larger fraction of water volume in contact with plants roots, were favorable for enhancing removal efficiencies in comparison with conventional CWs (Morato et al., 2014; Zapater-Pereyra et al., 2016).

The continuous recirculation can also improve organics abatement (Sklarz et al., 2009), while the aeration can help reducing the values of nutrients, turbidity, TSS and organics (Andreo-Martinez et al., 2017; Sklarz et al., 2009). The system filled with a smaller size medium performed better in microbial removals than the one filled with a bigger size medium (Morato et al., 2014).

Table 2. The mean concentration removal efficiency of main pollutants in single-stage systems analyzed.

System No.	HLR (cm d ⁻¹)	Removal efficiency (%)							
		Organic matter		Nutrients		Solids	Bacterial indicators		
		COD	BOD ₅	TN	TP	TSS	<i>E. coli</i>	FC	TC
1	12	60.5	53.8	-	-	-	-	-	-
2	36	63	-	61	-	88	99.9	-	-
3	3.6	63.8	65	-	-	-	-	-	-
4	-	50	65	-	-	-	-	>94	>94
5	0.6	-	-	51.3	72.7	-	-	-	-
6	2.62	92.7	97.8	91.5	96.9	97.5	-	-	-
7	1.8-4	79	76.5	-	62.8	59.3	-	-	-
8	-	88	90	-	-	92	94-99.9	94-99.9	94-99.9
9	9.5	78.5	-	26.5	30	-	-	-	-
10 ^a	-	-	-	-	-	-	-	-	-
11	1.94-3.75	92.5	90.8	91.7	87.9	91.6	-	91.4	-
12	-	84	95	-	-	90	-	-	-
13	-	-	99	-	-	95	-	-	-
14	-	-	-	75.4	94.9	-	-	-	-

^aThe research did not provide either removal efficiencies of main pollutants or influent concentrations, so the efficiency values could not be calculated

4. Hybrid systems

In the expectation of strengthening the treatment capacity of pollutants and attaining effluents of higher quality, some researchers combined different CW types or CWs with other techniques into hybrid systems, making use of their respective advantages (Avila et al., 2015; Haghshenas-Adarmanabadi et al., 2016; Ramprasad and Philip, 2018; Zurita and White, 2014).

4.1. Hybrid constructed wetlands

The most widely used hybrid CW system is the combination of two subsurface flow CWs - horizontal and vertical one (Ramprasad et al., 2017). For example, in South Korea, Kim et al. (2016) carried out a full-scale experiment on VSSF-HSSF CWs from 2002 to 2013. The VSSF system was planted with *Phragmites australis* and *Phragmites japonica*, while the HSSF CW was planted with *Miscanthus sacchariflorus*, *Carex dispalata*, *Juncus effuses* and *Iris pseudacorus*. Both of them were filled with coarse sand. The study revealed that the hybrid system had a stable TN removal in the whole 12-year operation period, with the average removal rate of 71.8% (Table 3, No. 15). TN removal efficiency was associated with the factors like season (greater removal rates in summer), operating stage (greater removal rates during the middle stage of operation, between 2006 and 2009) and nitrogen load. The highest removal rate simulated was at the inflow nitrogen load of under $2.8 \text{ g m}^{-2} \text{ day}^{-1}$ in summer. Besides, the VSSF had a better removal efficiency of TN than the HSSF. It can be explained by the fact that HSSF cannot satisfy the needs of denitrification/nitrification for a complete anaerobic/anoxic condition, while VSSF can provide a aerobic condition that favors nitrification process.

Melian et al. (2010) tested two pilot VSSF-HSSF systems filled with different substrates (gravel and lapilli) for eight months in Spain. Both systems were effective in municipal wastewater treatment, achieving the removal efficiency of more than 86%, 78%, 84%, 95%, 96% and 98.7% for BOD₅, COD, NH₄⁺-N, SS, turbidity and fecal indicators, respectively. The best COD and FC removal was attained by gravel-based system and lapilli-based system, respectively, at a high HLR of 7.9 cm d^{-1} , while the best BOD₅ removal was from lapilli-based system, at low HLRs in the range of $3.7\text{-}4.1 \text{ cm d}^{-1}$. Pollutants (COD, FC and NH₄⁺-N) removal efficiencies were not significantly different between two hybrids with gravel and lapilli when HLRs varied. However, for BOD₅, the lapilli-based system achieved significantly higher removal than gravel-based one. This better performance on BOD₅ removal could be attributed to a higher porosity and smaller diameter of lapilli particles applied, thus leading to greater removal during the treatment processes (e.g. filtration, sedimentation and BOD₅ degradation). Hence, the marginally greater system is lapilli-based hybrid CWs at high HLR, with the removal of 82% for COD, 89 % for BOD₅ and 99.9% for FC (Table 3, No. 16).

In Iran, Haghshenas-Adarmanabadi et al. (2016) evaluated four pilot HSSF-VSSF CWs from September 2013 to August 2014, three of them planted with different vegetation types and the last one unplanted. The average HLR was 5.3 cm d^{-1} . It was found that the hybrid systems can abate the main contaminants with high removal rates. The best removal efficiencies belonged to *Phragmites* hybrid CW, 84% for BOD₅, 79% for COD, 78% for TSS, 99% for FC, 43-97% for TP. (Table 3, No. 17). However, no significant difference was found among various hybrid systems for those pollutants removal, except for

TP removal. Planted CWs performed significantly better than unplanted CW for TP reduction. It can be attributed to plants uptake and sequestration in microbial biomass. Furthermore, adding VSSF CWs after the HSSF CWs was highly effective for optimizing main pollutants removal except for nitrates, which can be produced by ammonium nitrification in the VSSF stages.

In Colombia, Garcia et al. (2013) reported the high performance of a series of two-stage CWs in pathogen removal. These systems differed by feeding modes, order or combination of CWs (i.e. VSSF, HSSF) and vegetation conditions. The detailed description can be found in Table 1 (No. 18). It was shown that the type of planted VSSF-HSSF combination can reduce 99.984% *E. coli*, 99.987% TC and 90.741% Helminth eggs. The system had the best nitrogen removal (>90%) of all, also providing high removals of organics and solids (>90% for both BOD₅ and COD, >85% for TSS), regarded as the system with the best overall performance (Table 3, No. 18).

In Spain, Herrera-Melian et al. (2018) tested two hybrid systems, i) mulch-based HSSF followed by gravel-based VSSFs, ii) mulch-based HSSF followed by mulch-based VSSFs, under different feeding modes (continuous and intermittent). The highest removals of various pollutants (82% for COD, 97% for BOD₅, 99% for TSS, 99.8% for FC) were provided by the second hybrid system, the combination of HSSF and mulch-based VSSFs, under the continuous feeding mode (Table 3, No. 20). The authors argued that the intermittent feeding mode could have caused a shorter HRT compared to the continuous one, thus the lower removal efficiencies were attained. Furthermore, the greater performance of VSSFs with mulch than the ones with gravel can be attributed to its characteristics (e.g. compressibility and small particle size). A small particle size provides several benefits, longer HRT, greater water distribution on the reactor surface and retention of pollutants (e.g. TSS, turbidity).

In USA, Jokerst et al. (2011) operated a pilot-scale FWS-SSF hybrid CW in a semi-arid, temperate climate for one year. The FWS and SSF beds were planted with *Typha latifolia* and *Scirpus acutus*, respectively. The FWS CW was filled with amended soil up to a depth of 0.9 m, a mixture of 50% sandy-loam soil and 50% sphagnum peat, while the SSF CW was filled with about 15 mm-diameter granite stone. The author reported the seasonal performance of the system. It was shown that during non-winter periods the mass removal efficiency of contaminants BOD₅, TN and TP was as high as 92%, 85% and 78% on average, respectively. However, the treatment effect seemed to decrease in winter season. The mean yearly concentration removal efficiency is given in Table 3 (Table 3, No. 19). In addition, TSS removal was probably negatively affected by a considerable number of growing algae during warm months of spring in the FWS bed.

In recent years, multistage hybrid CWs have been applied. In China, He et al. (2018) tested for nine months a system consisting of a down-flow VSSF CW, an up-flow VSSF CW and a HSSF CW. The highest intensity of nitrification and denitrification of the media was in the down-flow VSSF bed and the HSSF bed, respectively, which may be associated with the abundance of nitrifying and denitrifying bacteria in these wetlands. During the experimental period the removal rates of COD, TP, TN and NH₄⁺-N reached 59%, 82.8%, 57.7% and 79.2% on average, respectively (Table 3, No. 21). COD removal was negatively affected by the application of media with relatively big diameters. Since the media could not have provided sufficient surface area for biofilm growth, microbial activities were limited and unsatisfactory COD removal efficiency was obtained. Despite this, the media decreased the possibility of clogging in the system.

In Czech Republic, a three-stage hybrid CW comprised of a saturated VSSF CW, a free-drain VSSF CW and a HSSF CW, was investigated by Vymazal and Kropfelova (2015) for nineteen months. They reported good efficiencies of the hybrid system - 92.5% for BOD₅, 96% for TSS, 88.8% for NH₄-N, 83.8% for

COD and 79.9% for TN (Table 3, No. 22). A similar removal robustness of a lab-scale three-stage hybrid system (two VSSF CWs operating alternatively, a HSSF CW and a FWS CW in series) located in Spain has been reported by Avila et al. (2013). The removal efficiencies of that system were 91% for BOD₅, 97% for TSS, 94% for NH₄-N, 78% for COD and 46% for TN. A full-scale research (Avila et al., 2015) also demonstrated the superb overall pollutants abatement capacity of hybrid systems (Table 3, No. 23), even for some emerging pollutants (more than 80% removal, e.g. analgesic and anti-inflammatory drugs, personal care products). These high removals of different pollutants in hybrid system were aided by high temperatures and synergies and combination of removal mechanisms (e.g. nitrification, denitrification, biodegradation and sorption) under different physicochemical conditions of CW configurations (Avila et al., 2015; Vymazal and Kropfelova, 2015).

4.2. CWs combined with additional technologies

Some investigations have concentrated on the addition of different technologies to existing CWs, such as filtration, ultraviolet (UV) treatment, etc. These physical, chemical or biological technologies were applied to improve wastewater treatment, especially for some pollutants like turbidity or microbial indicators (Patil and Munavalli, 2016; Russo et al., 2019; Toscano et al., 2013).

In Sakharale, India, a tropical zone, Patil and Munavalli (2016) reported a three-stage treatment system, containing preliminary treatment, HSSF CW treatment and a post treatment. Pretreatment was done by a settling cum equalization tank and an up-flow down-flow filter (UFDF), and could significantly decrease turbidity while also partially removing COD. The post treatment included a vertical flow charcoal filter and water hyacinth system, improving further the quality of final effluents to fulfill the outflow requirements mainly attributed to the role of adsorption and plant uptake. The overall removal rates of the system achieved were 70% for COD, 70% for total kjeldahl nitrogen and 85% for pathogen (Table 3, No. 24).

In Heilongjiang, China, Gao and Hu (2012) combined bio-contact oxidation (BCO) technique with a greenhouse-structured HSSF CW. The utilization of double solar panels in the greenhouse provided a stable temperature for treating wastewater, and improved effectively pollutants removal rates especially during winter season. The overall removal efficiency of the parameters evaluated in the combined system was 85.01% (COD), 70.98% (NH₃-N), 36.48% (TP) (Table 3, No. 25). The BCO treatment was responsible for 74.6% and 85.4% of overall removal of COD and NH₃-N, respectively while the wetland contributed to 59% of overall TP removal.

In Ulaanbaatar, Mongolia, an 'ice-block unit' consisting of a storage tank, septic tanks, a VSSF CW and a collecting tank showed a great potential for application in cold climates. It stored greywater throughout freezing period in an ice-block, melting and treating it during warm months. The removal rates of main pollutants (e.g. COD, NH₄⁺, TSS, *E. coli*) ranged from 87% to 100% (Uddin et al., 2016) (Table 3, No. 26).

In Italy, Russo et al. (2019) tested for one year a full-scale HSSF CW combined with an UV unit to treat domestic wastewater. The findings indicated that the whole system eliminated thoroughly microbial indicators such as *E. coli*, somatic coliphages and *C. perfringens* spores (Table 3, No. 27b). Sklarz et al. (2013) proved the effectiveness of UV light disinfection treatment through agricultural reuse experiments, since there was no *E. coli* detected in the soil irrigated with the treated wastewater. Moreover, wastewater treated after the integrated operation of the sedimentation tank, RVFCW, the filter and the UV

disinfection unit caused a large decrease on concentration of BOD₅, COD, TSS and *E. coli* in comparison with the raw wastewater (Table 3, No. 28). Horn et al. (2014) concluded that the combination of photocatalytic ozonation (UV/TiO₂/O₃) techniques and SSF CWs was capable of improving disinfection efficacy of the system and removed effectively microbial contaminants, reducing the microbial load under the detection limit. Besides, it eliminated 88.7% of BOD₅, 62.1% of COD and 63.4% of TP (Table 3, No. 29). However, the ramp of UV/TiO₂/O₃ reactor was observed to saturate after running for 4 h as a result of physisorption and chemisorption.

In Turkey, Ayaz et al. (2015) reported a three-stage hybrid pilot system built in a small community, consisting of anaerobic pretreatment, a HSSF CW and a VSSF CW. The pretreatment, an ABR and an upflow anaerobic sludge bed (UASB) reactor running in parallel, removed partially organic matter and SS. The combined system removed 90% of nitrogen and 95% of organic matter on average and the mean removal rates of pollutants BOD₅, COD and TSS are shown in Table 3 (Table 3, No. 30). The authors stated that the combination of HSSF-VSSF CWs optimized the removal of organics and SS, aided denitrification process of HSSF, effectively reduced phosphorus and stimulated nitrification in the VSSF, and in general showed greater performance than a single CW. Another way to increase TN removal is the recirculation of effluents in the system, which can also contribute to organics reduction (Sklarz et al., 2009).

Likewise, the study provided by Avila et al. (2016) proved that an experimental integrated system containing an anaerobic reactor was a good alternative for wastewater treatment in small communities, particularly in warm zones. The system comprised of two alternating VSSF CWs, a HSSF CW and a FWS CW operating in series, following an anaerobic reactor (a HUSB reactor). The results showed the system was able to effectively reduce BOD₅ (93% removal), TSS (96% removal), COD (82% removal) and NH₄-N (75% removal), whereas it did not perform well in PO₄-P (11%) and SO₄²⁻ (10%) removal (Table 3, No. 31). Another application of a HUSB reactor reported in Spain showed that it did not perform as well as a conventional settler before HSSF CWs (Pedescoll et al., 2011).

Yeh et al. (2010) carried out an experiment on the system made up of an oxidation pond, two FWS CWs with a cascade between them and a SSF CW operating in series. The findings (Table 3, No. 32) showed the system removed 81% of BOD and 48% of COD on average, mainly owing to microbial degradation, and reduced 65% of TN, primarily attributed to nitrification and denitrification occurring in the treatment processes.

Reinoso et al. (2008) operated a facultative pond (FP) followed by a FWS and a SSF system for 10 months in Spain and the results revealed that FP was the most effective in bacterial removal (e.g. TC, *E. coli*), while the SSF CW had the most robust capacity for removing protozoan pathogens and coliphages among the three techniques tested. It was observed that 99.33% of *E. coli* and 97.12% of TC were removed on average (Table 3, No. 33).

In Egypt, Abdel-Shafy et al. (2017) tested a pilot system including a sedimentation tank, a HSSF CW and a VSSF CW running in series for treating blackwater. The hybrid system was proven to be capable of treating a high hydraulic and organic load, with the removal efficiency of 98.5%, 98%, 97.4%, for COD, BOD and TSS, respectively (Table 3, No. 34). Furthermore, it was found that a high surface area and a low velocity of the integrated system (HSSF and VSSF CWs) principally contributed to the improvement of wastewater quality. However, a disadvantage of the system was exactly its high surface requirement.

In Greece, a three-stage hybrid system consisting of two serial settling tanks, a VSSF CW and a zeolite tank, was operated for 40 months. The overall removal efficiencies were satisfactory for BOD (95.8%),

COD (94.9%), TSS (96%) and TC (99.97%) (Table 3, No. 38). It was observed that organics removal was mostly attributed to the septic tanks followed by the CW. Finally, the zeolite tank further enhanced the treatment performance, since relatively large pores of zeolite were favorable for the adsorption of organics. Besides, the superb removal of TC was achieved mainly by sedimentation and filtration in the septic tanks and the CW, respectively. It was also reported that plants played a significant role on removal of organics and nutrients (except for TSS and TC). Moreover, their growth and movement may be beneficial for preventing clogging (Gikas and Tsihrintzis, 2012). Similarly, delaying or preventing system clogging can also be achieved by the application of media with bigger sizes (He et al., 2018), the setup of pretreatment (e.g. a HUSB reactor, a UFDF sand filter), adjustment of treatment operation and systems structure (e.g. intermittent discharge, upflow structure of CWs) (Pedescoll et al., 2011; Vera et al., 2013), as well as the application of earthworms or a low organic loading rate (Lavrnjic et al., 2019; Zapater-Pereyra et al., 2016).

Vera et al. (2013) reported a system composed of a septic tank and an Imhoff tank in series as pretreatment, and two parallel VSSFs followed by a HSSF as secondary treatment. It was found that the overall removal rates were 98% for TSS, 93% for BOD₅, 89% for COD, 61% for TN and 47% for TP (Table 3, No.39). Despite variability in removal efficiency among the stages affected by different factors (e.g. seasonal change and influent quality), overall performance was relatively stable during the experimental period of 2 years.

4.3. Comparison of various hybrid systems

As reported in section 4.1, Haghshenas-Adarmanabadi et al. (2016) tested four pilot HSSF-VSSF hybrid CWs for one year, three units planted with different emergent vegetation (*Phragmites australis*, *Typha latifolia* and *Arundo donax*) and one left unplanted, with a mean HLR of 5.3 cm d⁻¹. The authors pointed out that the hybrid systems offered better conditions for reaching reuse standards than single CWs. Besides, there were no significant differences among four systems in removal capability of BOD₅, COD, TSS and coliforms. However, with regards to nutrient removal, the planted systems (especially the unit planted with *Phragmites australis*) performed better in comparison with the unplanted one.

As reported in section 4.1, Garcia et al. (2013) assessed the performance of six planted or unplanted two-stage hybrid systems consisting of HSSF or/and VSSF CWs. The results highlighted that the combination VSSF-HSSF was the most efficient for *E. coli* removal (4 log units), TC (3 log units) and nitrogen (>90%), whether planted or not. In addition, it statistically displayed that vegetation probably contributed a lot to the reduction of nutrients and *E. coli*.

In Mexico, Zurita and Carreon-Alvarez (2015) reported on the application of three integrated systems running for two years, namely HSSF+stabilization pond (SP), HSSF+VSSF and VSSF+HSSF systems. During the first year, it was observed the HSSF+VSSF and VSSF+HSSF system showed the best efficacy of TC removal (2.2 log units) and *E. coli* removal (3.8 log units), respectively. During the second year, both HSSF+VSSF and VSSF+HSSF, performed well in TC and *E. coli* removal that was in the range 2.34-2.44, 3.44-3.74 log units, significantly better than HSSF+SP system. As a result, HSSF+VSSF system was the most effective for *E. coli* (99.94%) and TC (99.5%) removal during the two years study (Table 3, No. 35).

Multistage hybrid systems were also utilized in some research. For instance, Licciardello et al. (2018) evaluated two systems with similar costs, whose primary difference was whether UV disinfection was

utilized or not. One system was comprised of a HSSF CW, a biological pond, a storage reservoir followed by sand and disk filters, while the other one was made up of a HSSF CW, sand and disk filters and UV treatment, both of them operating in series. It was found that the system including UV treatment was more effective. The removal efficiencies achieved were 76.48%, 80.43% and 90.87% for COD, BOD₅ and TSS, respectively. Moreover, *E. coli* was reduced by 5.49 log units, mostly owing to the UV disinfection (Table 3, No. 36b).

According to Ali et al. (2018), the two full-scale systems tested in Pakistan, system I made up of an ABR, a saturated VSSF CW and a FWS CW and system II consisting of ABR, a HSSF CW and a FWS CW were both influenced by seasonal factors. They achieved greater removal efficiency in summer, similar with several findings concerning single or hybrid CWs (Ayaz, 2008; Kim et al., 2016; Ramprasad et al., 2017). Generally, the system I provided higher overall removals of COD (73.6%), BOD₅ (76.2%) and NH₄-N (52.8%). The exception was TSS removal (82%), that was slightly lower than in the system II (91%) (Table 3, No. 37). The differences of removal efficiency between the two systems were mainly caused by the use of HSSF or VSSF as the secondary treatment. It was also found that the first stage treatment (ABR) of both systems predominantly contributed to solids removal and organics degradation. Table 3 shows the overall pollutants removal efficiency of hybrid systems from 25 studies previously discussed. These integrated systems showed nearly complete removal of bacterial indicators and quite a good effectiveness in TSS abatement (>85% for most systems), except for the system HSSF-lagooning (Table 3, No. 27a). In comparison with single-stage CWs (Table 2), they had a better overall performance in organics reduction. These superb removal rates can be explained by the combination of techniques in hybrid systems that contributed to a greater overall removal efficiency. However, regarding nutrients, the hybrid systems displayed a wide range of removal variation, from 29.5% (Table 3, No. 28) to 94% (Table 3, No. 23) for TN and from 7% (Table 3, No. 28) to 82.8% (Table 3, No. 21) for TP. Low TP removal observed in a few studies could be attributed to a limited sorption of some substrates applied (e.g. crushed rock), and to the fact that no additional measures for enhancing TP removal were used (Vymazal and Kropfelova, 2015). Interestingly, there is one system HSSF-lagooning (Table 3, No. 27a) that showed low removal efficiencies for the most pollutants, although a majority of hybrid systems performed well. It can be attributed to the fact that the algae growth and decomposition in the lagooning unit contributed to the increase of TSS, BOD₅ and COD concentrations. Besides, the lagooning unit was not able to further reduce nutrients (TN and TP), due to the anaerobic decomposition of algae.

The same as section 3, the research discussed in this section also pointed out the role of plants, substrates, seasonal variation in hybrid systems. The presence of plants can improve the removal efficiencies of organics and nutrients (Garcia et al., 2013; Gikas and Tsihrintzis, 2012), while the reduction of TSS could be negatively affected by a large number of algae (Jokerst et al., 2011). Regarding *E. coli* removal, the influence of plants was not consistent. The role of plants is still an open question and while Garcia et al. (2013) reported there was significant difference between planted and unplanted systems tested, Headley et al. (2013) stated the opposite results.

In terms of substrates, the application of lapilli (Melian et al., 2010), mulch (Herrera-Melian et al., 2018) and zeolite (Gikas and Tsihrintzis, 2012), was regarded as favorable for pollutants removal, due to their characteristics of a high porosity, small media sizes and good compressibility. He et al. (2018) indicated that the media with bigger sizes cannot provide enough surface area for microbial activities, thus resulting in the lower COD removal.

In addition, the seasonal variation also had an impact on treatment performance of the hybrid systems. Jokerst et al. (2011) highlighted the better removal efficiencies of BOD₅, TN and TP during the warmer

part of the year. Particularly, the greatest performance can be generally achieved in summer (Ali et al., 2018; Kim et al., 2016).

Moreover, in comparison to single-stage systems, the use of UV treatment can enhance the disinfection efficacy of hybrid systems, as a result of excellent microbial removals.

Table 3. The mean overall concentration removal efficiency of main pollutants in hybrid systems analyzed.

System No.	HLR (cm d ⁻¹)	Removal efficiency (%)							
		Organic matter		Nutrients		Solids	Bacterial indicators		
		COD	BOD ₅	TN	TP	TSS	<i>E. coli</i>	FC	TC
15	4.5-22.7	-	-	71.8	-	-	-	-	-
16	3.7-7.9	82	89	-	-	-	-	99.9	-
17	5.3	79	84	-	43-97	78	-	99	-
18	10	>90	>90	>90	-	>85	99.984	-	99.987
19	1.3-3.4	-	80.5	74.8	66.3	-	-	-	-
20	-	82	97	-	-	99	-	99.8	-
21	15-24	59	-	57.7	82.8	-	-	-	-
22	3.8-61.1	83.8	92.5	79.9	30	96	-	-	-
23	4.4	89	99	94	47	98	99.999	-	-
24	1.5-9.3	70	-	-	-	-	-	-	-
25	-	85	-	-	36.5	-	-	-	-
26	-	98-100	-	-	-	97	98	-	-
27a	-	28.26	19.23	50.93	11.86	22	99.937	-	99.899
27b	-	-	-	-	-	-	>99.99	-	99.996
28	-	85.5	96.8	29.5	7	90	99.999	-	-
29	-	62.1	88.7	-	63.4	-	-	-	-
30	11.1-21.9	94.3	91.9	-	-	97	-	-	-
31	27	82	93	-	-	96	-	-	-

32	-	48	81	65	-	-	-	-	-
33	-	-	-	-	-	-	99.33	-	97.12
34	-	98.5	98	-	-	97.4	-	-	-
35	6.8-14.5	-	-	-	-	-	99.94	-	99.5
36a	-	69.02	68.01	-	-	90.15	99.99	-	-
36b	-	76.48	80.43	-	-	90.87	99.999	-	-
37	-	73.6	76.2	-	-	82	-	-	-
38	-	94.9	95.8	-	67.3	96	-	-	99.97
39	-	89	93	61	47	98	-	-	-

When the research tested more than one hybrid system (No. 16, 17, 18, 20, 35 and 37), the one with the best overall removal efficiencies was reported in this table.

The study (No. 26) provided only the maximum removal rates.

5. Wastewater reuse in agriculture

Wastewater reuse can not only alleviate water scarcity, but it can also relieve pressure on conventional wastewater treatment plants (Ghaitidak and Yadav, 2013). Treated wastewater offers a more sustainable and stable use in comparison with natural resources, especially when seasons and climate change are considered (Zhang and Shen, 2019). Moreover, treated wastewater is rich in inorganic elements and organic compounds that can increase crop yields, while at the same time reducing use of fertilizers (Castro et al., 2011). Similar findings were also made by Almuktar et al. (2017) that recycled nutrients from wastewater in agriculture, resulted in greater chilies weights and dimensions, as discussed in section 3.2.

Nevertheless, wastewater could negatively impact irrigated soil and plants, likely to be a potential threat on the environment and human health. Travis et al. (2010) presented hydrophobicity development of soil as a result of the application of raw wastewater in irrigation. Moreover, it can cause heavy metals accumulation as reported by Gola et al. (2016) and shallow groundwater pollution (Zhang and Shen, 2019). However, the use of suitable wastewater treatment before irrigation can effectively prevent modification of soil properties and diminish environmental risks (Sklarz et al., 2013). Further information on the effect that wastewater reuse can have on soil and crops can be found in Al-Isawi et al. (2016); Almuktar et al. (2017) and Sklarz et al. (2013).

5.1. EU standards on wastewater reuse for agricultural purposes

Currently, more and more attention is given to the issues of wastewater treatment and reuse. Treated wastewater is reused for crop irrigation in many areas of the world (Licata et al., 2017). Specific standards about use of reclaimed water in agriculture already exist, including those in developed countries (e.g. United States, Italy, Spain), developing countries (e.g. Egypt, Pakistan, Turkey, Iran, Mexico, Thailand, Colombia) and even some international institutions such as the World Health Organization. The European Commission is currently in the process of adopting its own guidelines (European Commission, 2018), that are characterized by detailed classification limits depending on crop types and irrigation methods (Table 4).

Table 5 shows main pollutants concentration in effluents from 39 studies analyzed in this review. According to the future European criteria, parameters BOD₅, TSS and *E. coli* are classified into different levels that correspond to different agricultural purposes. It can be seen that a majority of studies presented here achieved Class B, C and D (25 mg L⁻¹) for BOD₅ concentration in effluents, suitable for crops irrigation (e.g. industrial, energy, and seeded crops, processed food crops and non-food crops) under any method (Table 4). Besides, the better BOD₅ removals were exhibited by the improved single-stage CWs - GROW and RVFCW (Table 5, No. 11, 13) and several multistage hybrid systems (Table 5, No. 22, 23, 28, 32, 36b), consequently meeting the strictest standard of irrigation reuse (Class A). As expected, most treatment systems showed a good performance on TSS removal leading to low effluent concentrations, except for two hybrid systems (Table 5, No. 27a, 37) exceeding the range of the guideline. The high TSS concentration in research No. 27a can be attributed to algae growth in the lagooning unit. The similar finding was also observed by Jokerst et al. (2011) as reported in section 4.1. Regarding *E. coli*, four

studies (Table 5, No. 6, 27b, 33, 36b) meet Class A (10 cfu 100 mL⁻¹) of the irrigation limits. Those results can be mainly explained by the positive effects of artificial aeration (Andreo-Martinez et al., 2017), UV treatment (Licciardello et al., 2018; Russo et al., 2019) and the combination of different treatment systems (Reinoso et al., 2008). Moreover, it is found that not many studies can achieve excellent *E. coli* removal without the application of disinfection measures or other chemical agents (Andreo-Martinez et al., 2017). Thus, several researchers indicated that, in order to improve effluents quality and to reach the criteria recommended for agricultural reuse, at least two stages of wastewater treatment in hybrid systems are necessary for pathogen removals (Toscano et al., 2015; Zurita and White, 2014). In terms of COD and TN, effluent concentrations among various systems vary considerably (Table 5). As concluded in section 3.3, artificial aeration and recirculation treatment can improve the removal of nutrients and organics, respectively. Furthermore, as reported in section 4.3, plants can also be beneficial for both pollutants removal.

5.2. Irrigation applications of treated wastewater

Among the 39 experimental research selected, there are several papers focusing on reclaimed wastewater reuse in irrigation, besides treatment methodologies. Some researchers (Almuktar et al., 2017; Almuktar and Scholz, 2015) have carried out lab-scale experiments on an overall process of wastewater treatment and reuse in irrigation for a few years. The findings indicated that chilies were able to grow successfully if irrigated with effluents from a VSSF CW, that for the majority of parameters complied with the irrigation criteria. Sklarz et al. (2013) explored the influence on soil of using treated wastewater from RVFCW for irrigation, and concluded that after 3 years physical and chemical properties of soil were similar to a soil undergoing usual agricultural treatment (irrigated with fresh water and enriched with fertilizers). Travis et al. (2010) also stated that the RVFCW effluents did not have any obvious adverse impact on plants growth and soil, thus it could be considered as an effective irrigation source.

Table 4. The European guidelines on pollutants threshold values of reclaimed water for agricultural irrigation (European Commission, 2018).

Pollutants	Reclaimed water quality class			
	Class A	Class B	Class C	Class D
<i>E. coli</i> (cfu 100 mL ⁻¹)	10	100	1000	10000
BOD ₅ (mg L ⁻¹)	10	25	25	25
TSS (mg L ⁻¹)	10	35	35	35

Class A: All food crops, including root crops consumed raw and food crops where the edible portion is in direct contact with reclaimed water. Irrigation method: All irrigation methods allowed.

Class B: Food crops consumed raw where the edible portion is produced above ground and is not in direct contact with reclaimed water, processed food crops, non-food crops including crops to feed milk- or meat-producing animals. Irrigation method: All irrigation methods allowed.

Class C: Crop category applicable is the same as Class B. Irrigation method: Drip irrigation only.

Class D: Industrial, energy, and seeded crops. Irrigation method: All irrigation methods allowed.

Table 5. The mean concentration of tested main pollutants in effluents of analyzed systems and the class level for agricultural purpose of treated wastewater according to parameters BOD₅, TSS and *E. coli* referring to the guidelines of European Commission (2018).

System No.	Effluent concentration								
Parameters required by European Commission (2018)						Other parameters			
Single-stage CW	<i>E. coli</i> (cfu 100 mL ⁻¹ unless stated otherwise)	Class	BOD ₅ (mg L ⁻¹)	Class	TSS (mg L ⁻¹)	Class	COD (mg L ⁻¹)	TN (mg L ⁻¹)	TP (mg L ⁻¹)
1	-	-	12.2	B, C, D	-	-	21.1	-	-
2	-	-	-	-	7.4	A	28.6	10.8	-
3	-	-	49	None	-	-	61.5	-	-
4	-	-	3.9	A	-	-	16.5	-	-
5	-	-	21.7	B, C, D	29.7	B, C, D	57.8	32.9	6.1
6	~0	A	16.5	B, C, D	<20	B, C, D	100.3	16.1	1
7	-	-	-	-	-	-	-	-	-
8	1.1×10 ³ MPN 100 mL ⁻¹	-	13.2	B, C, D	8.5	A	30.6	-	0.4-2
9	2.3 log MPN 100 mL ⁻¹	-	-	-	-	-	48	62	7
10	-	-	19.1	B, C, D	7.1	A	51	-	-
11	-	-	<10	A	-	-	<20	-	0.8-1.4
12	-	-	-	-	-	-	-	-	-
13	-	-	1.2	A	8.5	A	38	0.5	7.9
14	-	-	-	-	-	-	-	11.2	0.35
Hybrid system									
15	-	-	-	-	-	-	-	10.8	-
16	-	-	16.2	B, C, D	-	-	79.46	-	-
17	-	-	14.4-96	B, C, D	21.34-127.6	B, C, D	42-336	-	-

18	1×10 ³	C	<11.2	A, B	-	-	<33.9	-	-
19	-	-	16.8	B, C, D	8.2	A	-	3.4	1.4
20	-	-	17	B, C, D	-	-	99	-	-
21	-	-	-	-	-	-	62.3	8	0.1
22	-	-	7.7	A	2.6	A	39	6.5	2.8
23	<40	B	4	A	3	A	43	2.2	3.1
24	-	-	46	None	-	-	58	-	-
25	-	-	-	-	-	-	22.4	-	2.1
26	9.2×10 ⁴	None	-	-	2.5-11.2	A, B, C, D	0-19.2	-	-
27a	1.8 log	B	21	B, C, D	39	None	33	10.6	5.2
27b	<1 log	A	-	-	-	-	-	-	-
28	-	-	5	A	8.9	A	25	25.3	6.5
29	-	-	25.3	None	-	-	100.8	-	3.1
30	-	-	11	B, C, D	5.6	A	28.4	26.2	-
31	-	-	21	B, C, D	8	A	73	-	-
32	-	-	3.2	A	-	-	8.8	4	-
33	3.23	A	-	-	-	-	-	-	-
34	-	-	18	B, C, D	7.6	A	18	-	8.9
35	1.11×10 ³ MPN 100 mL ⁻¹	-	-	-	4.1-4.6	A	35-56.9	58.6-111.6	5.5-12.2
36a	34	B	10.3	B, C, D	8.3	A	24.1	-	4.6
36b	1.6	A	6.3	A	7.7	A	18.3	-	4.8
37	-	-	30	None	84	None	47	-	-
38	-	-	20.2	B, C, D	14.9	B, C, D	48.9	-	2.9
39	-	-	48.8	None	8.46	A	138.3	45.2	8

In the studies (No. 16, 17, 18, 20, 35 and 37) when the research tested more than one hybrid system, the one with the least pollutants concentrations was reported in this table.

6. Conclusions

Constructed wetlands are recognized as an effective and inexpensive technology for wastewater treatment. This review analyzed recent experimental studies on single-stage and hybrid CWs, that tested different scales, operating times, influent strengths, plant species, etc. According to the 39 studies considered, it can be concluded that improved single-stage CWs mainly had a better performance on pollutants removal (i.e. solids, nutrients and organics) than conventional systems. The multiple-stage treatments (e.g. hybrid CWs) and in particular the application of additional techniques (e.g. UV treatment, anaerobic reactors) combined with CWs, were able to further increase and optimize overall removal effectiveness.

In addition, seasonal variation can affect pollutants removal and generally the highest removal efficiency was achieved in summer. Plants could be beneficial for removal of organics, nitrogen and phosphorus, especially after they are fully established. However, it was noted that they could also cause the negative effect on treatment performance - additional generation of organic carbon, higher ET values and increased TSS concentration due to algae growth. Regarding the substrate type, it could be concluded that the ones with a higher porosity, small media sizes and good compressibility were favorable for removal efficiencies. Also, ET, depending on plant species and climate, was shown to be able to offset the effect of wastewater treatment.

The potential of considered treatment systems for irrigation was different to a large extent. Effluent quality of systems analyzed varied in a wide range, and it could not always meet the standards for agricultural reuse imposed by the new European regulations. The improved single-stage CWs and multistage hybrid systems generally had more possibilities to produce effluents with lower BOD₅ concentration (class A) than single-stage CWs. Also, hybrid systems showed a better overall performance on TSS reduction. However, *E. coli* concentration could not always be reduced to a level needed for agricultural reuse without the application of specific disinfection measures. Therefore, additional techniques and treatment steps should be introduced before irrigation application in order to decrease the environmental and public health risks.

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