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Review

A Review on Constructed Treatment Wetlands for Removal of Pollutants in the Agricultural Runoff

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Abstract: Constructed wetland (CW) is a popular sustainable best management practice for treating different wastewaters. While there are many articles on the removal of pollutants from different wastewaters, a comprehensive and critical review on the removal of pollutants other than nutrients that occur in agricultural field runoff and wastewater from animal facilities, including pesticides, insecticides, veterinary medicine, and antimicrobial-resistant genes are currently unavailable. Consequently, this paper summarized recent findings on the occurrence of such pollutants in the agricultural runoff water, their removal by different wetlands (surface flow, subsurface horizontal flow, subsurface vertical flow, and hybrid), and removal mechanisms, and analyzed the factors that affect the removal. The information is then used to highlight the current research gaps and needs for resilient and sustainable treatment systems. Factors, including contaminant property, aeration, type, and design of CWs, hydraulic parameters, substrate medium, and vegetation, impact the removal performance of the CWs. Hydraulic loading of 10–30 cm/d and hydraulic retention of 6–8 days were found to be optimal for the removal of agricultural pollutants from wetlands. The pollutants in agricultural wastewater, excluding nutrients and sediment, and their treatment utilizing different nature-based solutions, such as wetlands, are understudied, implying the need for more of such studies. This study reinforced the notion that wetlands are effective for treating agricultural wastewater (removal >90%) but several research questions remain unanswered. More long-term research in the actual field utilizing environmentally relevant concentrations to seek actual impacts of weather, plants, substrates, hydrology, and other design parameters, such as aeration and layout of wetland cells on the removal of pollutants, are needed.

Keywords: constructed wetlands; agricultural runoff; chemicals of emerging concern; veterinary antibiotics; antibiotic resistant genes

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1. Introduction

Agricultural runoff contains excess quantities of diverse pollutants, such as sediments, nutrients, pathogens, veterinary medicines, pesticides, and metals. Modern agriculture heavily relies on agro-chemicals, such as pesticides, herbicides, and hormones, that would grant a greater yield in a shorter period [1]. As the demands for food have increased, so has the intensity of agricultural activities and animal feed operations [2]. As a result, agricultural practices over the past years have included more pesticides and inorganic fertilizers [3,4]. Carvalho and colleagues (1997) reported that North American farmers relied on herbicides 43.3% of the time, while European farmers used it slightly less at 26.3% in 1993 [5]. In 2005, there were more than 800 newly registered pesticides in the European Union [6]. Additionally, approximately two million tons of pesticides were used globally in 2019, with China and the USA being the two major users [7].

These chemicals are perfect for increasing yield but are ecologically detrimental when they leave agricultural ecosystems in runoff water following storms [8]. Studies have shown that only 1% of pesticides applied to crops are effective, the other 99% enter the atmosphere, soils, and bodies of water through non-targeted contamination [9]. In livestock production, animal waste can act as reservoirs for antibiotic resistant genes (ARGs) and antibiotic resistant bacteria (ARBs) [10–12]. Another study found the prevalence of veterinary pharmaceuticals to be higher in soil than in water, indicating likeliness of movement to water resources through agricultural runoff [13].

In the long run, these chemicals have the ability to negatively impact food security and agricultural sustainability [14]. Termed chemicals of emerging concern (CECs), these compounds have long been a threat for water quality. According to the Environmental Protection Agency (EPA), CECs include but are not limited to nanoparticles, pharmaceuticals, personal care products (PCPs), estrogenic compounds, flame-retardants, detergents, and other industrial chemicals. All of these contaminants, many of which have agricultural origin, significantly influence human health and aquatic life [15].

Treatment of diffuse source pollution, such as agricultural runoff, requires a low-cost, passive, and nature-based approach known as an ecological engineering approach. Constructed wetland (CW) is a natural ecological alternative to the conventional methods for treating various types of wastewater, including agricultural runoff [16]. The EPA (1993) defines CWs as engineered systems that are designed and constructed to utilize natural processes [17]. Specially designed CWs could be used to treat wastewater in a system that mimics their natural components. The use of wetland plants to treat wastewater is a technique that was firstly studied in the 1950s by German scientist Dr. Ka the Seidel; since then, the idea has expanded greatly and is a very sustainable way of naturally treating many sources of wastewater [18]. CWs are more beneficial than conventional wastewater treatment methods because they require lower energy and less operational effort, but they are also land intensive [16]. CWs are versatile in their functioning, serving as a tool for water quality improvement, hydrological buffers, reservoirs, and nature development/recreational areas [19]. Through imitation of natural wetland systems, such as marshes with wetland plants, soils, and soil microorganisms, CWs are capable of removing diverse contaminants from different wastewater sources [20].

However, there is still very little known about the biotic and abiotic influences and interactions that allows this treatment of water and soil to take place [17]. While much of the previous reviews focused on how CWs are used to efficiently remove nutrients, such as nitrogen and phosphorus, and sediments from wastewaters [21,22], this paper focuses on the occurrence of pollutants in the agricultural runoff and how this cost-effective green approach [23] can be used to remove pollutants from agricultural runoff for mitigation of the negative environmental impacts of agricultural intensification. Focus pollutants include veterinary medicines, antimicrobial resistant genes, insecticides, herbicides, and pesticides.

2. Approach and Definitions

In this article, we reviewed global literature that focused on CWs used for the treatment of agricultural runoff or wastewater and the characteristics of their design. Scholarly databases were searched using keywords, such as constructed wetlands, agricultural runoff, ARGs, ARBs, pesticides, veterinary antibiotics, chemicals of emerging concern, and their combination to source relevant articles, reports, books, and conference proceedings published in recent years. Both lab-scale and field-scale experiments that studied effective removal rates of contaminants in these systems were considered. The search resulted in over 60 publications that were examined and subsequently summarized in this article directly or indirectly.

CWs are generally classed based on the life form of the dominating large aquatic plant or macrophyte in the system [24] or water-flow regime [25]. Figure 1 shows the classification of CW and their characteristics, which includes flow and flow direction [18,25,26]. Search results were screened based on their relevancy to include CWs that were subsurface horizontal flow (SSHF), subsurface vertical flow (SSVF), surface flow (SF) and hybrid and were used to remove contaminants that were not nutrients (nitrogen (N), phosphorous (P), total nitrogen (TN), total phosphorous (TP), and sediment).

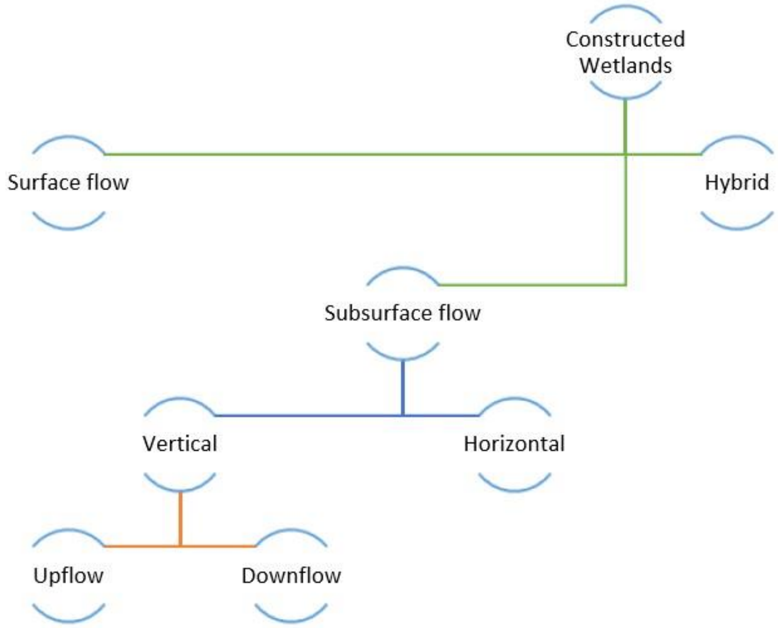

Figure 1. Classification of constructed wetlands.

Hydrological factors dictate the functioning of wetlands as they are directly linked to the ecosystem's biotic and abiotic processes. These processes are what influences both the biological (nutrient availability, microbial community, plant community) and physicochemical (soil pH, water pH, oxidation-reduction potential (ORP)) parameters in CWs [27]. Success of CWs is heavily dependent on the hydraulic residence time (HRT) [28,29] and the hydraulic loading rate (HLR) [30,31]. Various factors, such as wetland design, scale, size, water depth, HRT, HLR, substrate, experiment duration, source of pollutant, pollutant influent concentration, removal percentage, and major mechanisms responsible for removal of pollutants were tabulated, represented in graphs, or analyzed further.

3. Occurrence of Pollutants in Agricultural Runoff

Diverse pollutants have been measured in agricultural runoff. Pesticides, herbicides, and veterinary pharmaceuticals are present in agricultural runoff and are major threats to water quality health [32]. Concentrations of CECs have been found in quantities in excess of 0.01 mg/L, especially during storm events [33]. The antibiotics found mostly in agricultural runoff from the reviewed articles are mainly tetracyclines, sulfamonomethoxine, enrofloxacin, and trimethoprim, which are either used for disease prevention or as growth promoters in the industry [34–41]. A Chesapeake Bay study found high concentrations of antibiotics (azithromycin (AZI), clarithromycin (CLA), difloxacin (DIF), enrofloxacin (ENR), norfloxacin (NOR), roxithromycin (ROX), and sulfamethoxazole (SMX)), and hormones (mainly estrogen derivatives) due to wastewater effluents and agricultural runoff [33]. Antibiotics in both swine and dairy cattle farm effluents were found at high concentrations in China, which implies frequent application of these antibiotics during the production process [42]. Since China is one of the largest producers of animals in the world, significant consumption and release to the environment are expected.

Animal husbandry is a major source of environmental ARGs and ARBs [12]. ARG dissemination from flowing water normally happens from ground or surface water sources receiving effluents from domestic, municipal, and agricultural sources, such as livestock farms [43,44]. Through horizontal gene transfer, bacteria are able transfer resistance from one organism to the other. Oliver and colleagues studied dairy manure systems and found the presence of bacteria, such as *Enterobacteriaceae* (specifically nontyphoidal *Salmonella*), antibiotic-resistant *Campylobacter*, methicillin- and vancomycin-resistant *Staphylococcus*, and vancomycin-resistant *Enterococcus*, which the Centers for Disease Control and Prevention (CDC) deemed clinically dangerous to be prevalent. Additionally, they also found that some of these bacteria were able to resist up to five antibiotics [45]. A Chinese study (2018) found 18 types of ARGs from swine feedlots in the surrounding environment, namely streams and agricultural soils [46]. Genes dominant in swine manure were found to be those that were resistant to tetracycline (TC), aminoglycoside (AGR), chloramphenicol (CPR), multidrug (MDR), sulfonamide (SNR) and beta-lactam (BLR) [46–59]. SNR genes were also found abundantly in dairy manure storm runoffs [60]. Background bacterial DNA concentrations were indicated by 16S rRNA data as high as 4.10×10^{13} copies/mL [61].

The occurrence of pesticides was also found to be prevalent in agricultural runoff effluent [62–65]. A Mexican study found priority pollutants, such as endosulfan, an insecticide that is authorized for use in the country, to be in excess of 8.656×10^{-3} mg/L in runoff water during storms [66]. Other major contaminants in agricultural runoff include veterinary pharmaceuticals and personal care products (PPCPs), such as naproxen, estrone (and other estrogenic derivatives), which are used mainly for pain suppression or growth enhancement for animals [65–69].

A summary of occurrence of these pollutants in the agricultural wastewater (Figure 2) indicate presence of greater than 1000 mg/L biochemical oxygen demand (BOD), chemical oxygen demand (COD) and total suspended solids (TSS); sub part-per-trillion to 30 part-per million of antibiotics, hormones, and veterinary pharmaceuticals, and up to 4.1×10^{12} cells per mL of bacteria [42,65,70–72]. Herbicides were found to be more dominant in the agricultural runoff as it had been found as high as 530 mg/L. The prevalence of other contaminants, such as metals and fungicides, were much lower than the other CECs considered [42,66,68,70].

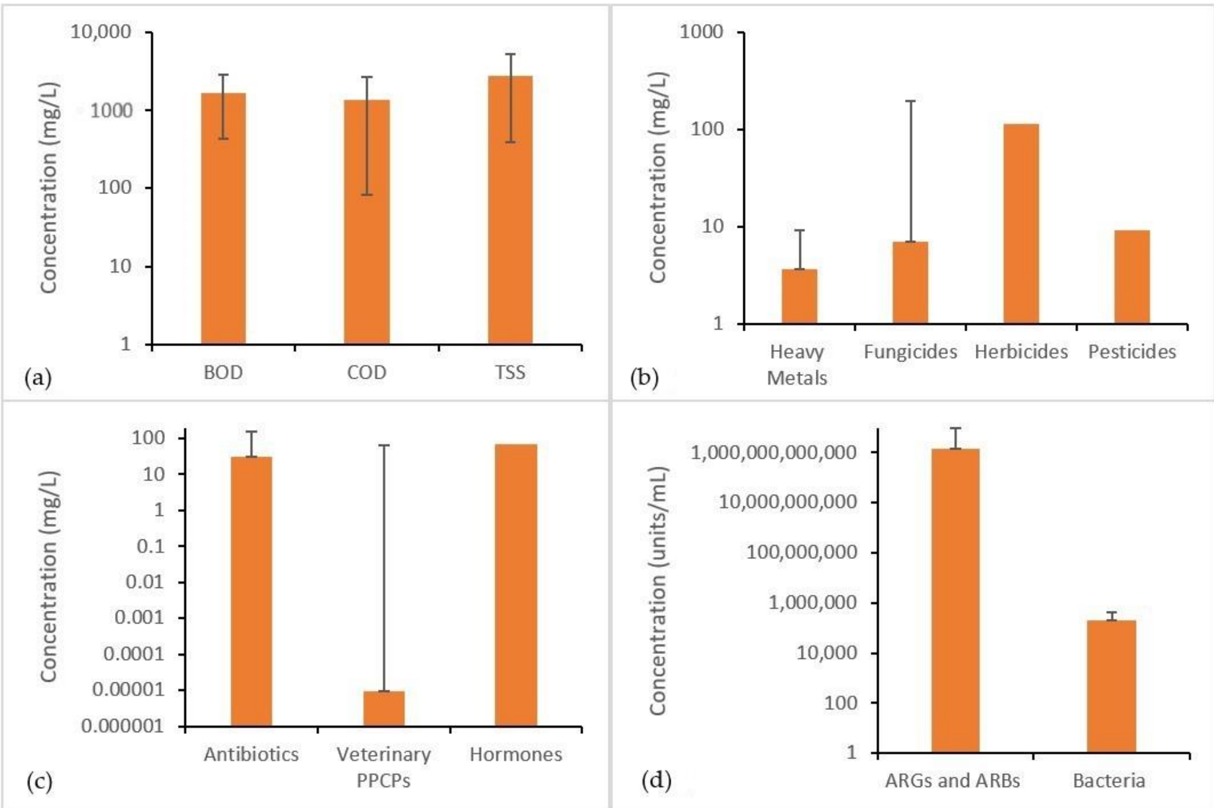

Figure 2. Occurrences of pollutants in the agricultural runoff: (**a**) Total suspended solids, biochemical oxygen demand, and chemical oxygen demand, (**b**) metals, fungicides, herbicides and pesticides, (**c**) antibiotics, hormones and veterinary pharmaceuticals, (**d**) antibiotic resistant bacteria and genes, and bacteria. Data are mean ± standard deviation. Note logarithmic y-axis.

4. Removal of Pollutants by Wetlands and Processes for Their Removal

Many studies have been conducted on the applications of constructed wetlands to remove pollutants from agricultural runoff and wastewater. Based on the study scales, this section has been divided into lab-scale and field-scale for further discussion.

4.1. Lab-Scale

Scientists in Portugal conducted laboratory-scaled microcosm studies to evaluate the removal performance of constructed wetlands for veterinary antibiotics for many years [39,42,73]. In their CW microcosms, multiple layers were set up (from bottom to top) as gravel, lava rock, root bed substrate (which was a mixture of soil and sand to help the vegetation's establishment) and *Phragmites australis* were planted. They used wastewater from swine farms/saline aquaculture facilities as their influent water with antibiotic concentrations spiked-up to 100 μg/L. The results showed that the removal efficiency for vet antibiotics-enrofloxacin (ENR), tetracycline (TET) [39], oxytetracycline (OXY) [73] and ceftiofur (CEF) [42] were over 90% after 9 to 20 weeks treatment period. The major mechanisms for the removal processes were adsorption to the substrate and plant's root (physical process), microbial metabolization and degradation (biological and chemical processes) and plant uptake (biological process) [39,42,73]. Studies conducted using the wide range of pollutants and various influent water types (fresh water and saline water) proved that CW microcosm design was adaptable to various wastewater treatments with satisfying removal efficiencies. Their study in 2020 using the same system even achieved toxic metal removal while maintaining the nutrient levels for agriculture reuse [70]. Another study in 2018 using the same system observed removal of organic micropollutants, such as atrazine, clarithromycin, fluoxetine, and norfluoxetine, from the freshwater aquaculture effluents [74]. Evidence from other studies suggested that such removal was accomplished through microbial degradation [75,76]. Another study conducted in Canada also found that subsurface horizontal flow constructed wetlands could remove 42%, 49% and 49%, respectively, of poultry pharmaceuticals monensin, salinomycin and narasin through sorption onto the soil surface and microbial degradation [77]. This indicates that with successful CW design, we can treat wastewater containing various contaminants in an efficient and economical manner. Such small-scale laboratory studies may not be sufficient for direct field application of constructed wetlands, but they serve as a good role at the proof-of-concept stage for future larger scaled studies.

Besides antibiotics, pesticides, such as chlorpyrifos, have been studied intensively and shown to be highly removable through constructed wetlands [78–82]. Most of the studies showed that biodegradation and adsorption were the primary removal mechanisms of such chemicals from the CW system. In addition, studies have also looked into the removal performance for antibiotic resistance genes [83–85]. According to the study by Song et al. (2018), the accumulation of antibiotics in different layers within the constructed wetland resulted in an abundance of ARGs with a positive correlation relationship [84]. Later studies proved that some CW systems could reduce ARG concentrations as they remove the antibiotic contaminations. Chen et al. 2019 study showed that while antibiotics' major removal mechanism was microbial degradation, ARGs main removal mechanisms were substrate sorption and biological reactions [83]. Another study investigated the comparisons of substrate medium by Du et al. (2020) and observed better removal rates (>95%) for both antibiotics and ARGs when zeolite medium and plant (*Arundo donax*) were used [85].

4.2. Field-Scale

For field-scale studies, it can be further divided into two groups: pilot-scale CW studies and full-scale in situ CW studies. The former one typically had a smaller dimension (within an average volume of 1 m^3 for each CW), while the latter took a greater surface area (typically over 100 m^2) and served as a functional water treatment system for agricultural wastewater or farm runoff. The pilot-scale studies can be seen as a scaled-up version of laboratory-scale studies, as they are in larger volumes and typically operated in greenhouses or open fields receiving more real-world weather conditions than lab studies, but they can still be modified timely during operation to achieve better performance since their scale is still manageable. Therefore, during the pilot-scale study time (ranging from 1 to 16 months), water samples were collected periodically to evaluate the CW performance over time [38,62,63,86–89]. While the full-scale studies were based on fully established CWs that have been operating for several years, therefore, the system typically already reached a steady state for removal performance requiring little manipulation during study time. Compared to the smaller scaled studies that typically collect water samples at the influent and effluent points with multiple and periodic sampling events, the larger scaled studies tend to have more sampling points throughout the system within only one or few sampling events to monitor the removal performance over the entire treatment system [40,41,66,90,91]. Another major difference between pilot-scale and full-scale CW studies was that pilot-scale studies often spiked up the target contamination concentrations even if they already existed in the influent water, but the full-scale field studies treated the existing concentrations and measured field concentrations. Therefore, full-scale studies might show relatively lower removal efficiencies since it is more challenging to achieve high removal performance at lower influent concentrations.

4.2.1. Pilot-Scale

A research group from China studied applying CWs to remove veterinary antibiotics and antibiotic resistant genes from swine wastewater for many years. In their studies, various CW types and their combinations as well as substrate medium and target contamination compounds were investigated. Their 2013 study results found that SSVF-CWs could efficiently remove target antibiotics and ARGs (68–95% and 50–90%, respectively) with the major mechanism being the physical sorption towards the wetland medium [38]. In another study, the results showed that removal performance ranged from high to low in the order of SSVF-Low water level > SSVF-High water level > SSHF > SF (based on average removal rates) indicating that the various design, flow path and water level led to different antibiotic removal rates through impacting the parameters, such as temperature, oxygen transfer, oxidation-reduction potential, sorption sites, etc. [89]. Another key finding from this study was that the seasonality might pose different impacts on different veterinary antibiotics (significant effect on sulfamethazine (SMZ) while no significant effect on TC) and different CW types (significant effect on SF while no significant effect on SSVF) [89]. Another long-term study indicated that high removal rates, ranging from 69.0% to 99.9%, were achieved for the target contaminants in all three treatments with different initial concentrations [86]. In another short-term study, flow direction showed no significant influence since they obtained comparable removal rates, but accumulation of antibiotics and ARGs in the surface soil was observed in down-flow treatments indicating a concern to the local environment due to likeliness of antibiotics enrichment and ARGs abundance [87].

Besides antibiotics, studies have also been conducted on removal of herbicides via application of CW systems, as Gikas et al. demonstrated up to 74% removal of terbuthylazine [92] and 60% removal of S-metolachlor [93] in horizontal subsurface flow CWs. Other researchers also investigated the removal performances of hybrid, SSHF and SF CW systems using various substrate and vegetations [62,63,88]. In a 2019 study, different combinations of CW units (SSHF-SSVF (up-flow); SSHF-SSVF (down-flow); SSFV (down-flow)-SSVF (up-flow)) were run for 84 days to treat antibiotics, ARGs and nutrients from goose wastewater. The researchers reported that the comparable antibiotic removal performance of different combinations of hybrid CWs was probably due to the highly spiked-up influent concentrations (2500 μg/L for tilmicosin (TMS) and 30 μg/L for doxycycline (DOC)), which likely concealed the differences on effluent concentrations among different treatments [63]. This may indicate the importance of conducting full-scale field studies receiving much lower antibiotic concentrations to simulate the real-world scenario, instead of pursuing the high removal efficiency results by dosing up the influent water to an unrealistic level. Besides livestock and poultry, CW has also been applied to treat wastewater from aquaculture. For example, Huang et al. conducted a study using SSHF with different vegetations (single or mixture of *Iris pseudacorus* and *Phragmites australis*) to remove ENR, SMZ and AGRs from wastewater of a local fish farm achieving up to 80% removal performance [88].

4.2.2. Full-Scale

For the full-scale field studies, multiple treatment units either incorporating both traditional water treatment processes (such as filtration, sedimentation, anaerobic digestion, etc.) and constructed wetland treatment processes, or a hybrid system with a series of different CW cells (such as SF, SSHF, SSVF, etc.) were used [40,91]. For assessing treatment performance, an entire system's performance over a long time was monitored [40,91] or the performances at various stages within the system were compared by collecting samples at multiple sampling points [72,90,94]. The application of CW in the field could be an entire system, or sometimes just one unit in addition to the traditional treatment units. For example, the Chen et al. (2012) study compared a traditional swine wastewater treatment system (A) with another system (B) containing additional aquatic vegetation ponds (serving as SF CWs) as a final polishing unit [40]. The results showed that biological activities had a significant impact on the degradation of target contaminants but less impact on the dissipation of contaminants at low concentrations [40]. One common challenge for field studies compared to the pilot-scale or lab-scale studies is there is no perfect "control treatment" to refer to. Therefore, background/influent concentration data for such field studies are extremely important, as they can serve for the comparison of pre and post CW treatment. As an example, Locke et al. (2011) simulated a runoff study which sampled before and after the flushing events for comparisons of the removal rate [91]. Their results indicated that CW could help to protect the downstream water quality through degradation and sorption of the pollutants and retention caused by adsorption and/or uptake by vegetation even after the flushing event throughout the entire 21-day study period [91]. For the integrated/hybrid CW system using multiple treatment units with different designs, the concentrations of target contaminants typically showed a decreasing trend along water flow through various stages. For example, in the study by Chen et al. (2015), which utilized the field CW system containing six units in series and receiving rural wastewater, the antibiotic concentrations decreased continuously along the treatment train as each unit's effluent concentrations were lower than that of the previous unit [90]. This indicated that with careful design and reasonable arrangements, multiple CW treatment units could run in series to achieve better overall removal performance. More complicated systems, such as in the Abdel-Mohsein et al. study (2011), which applied various CW types in series and operated in parallel at each stage with three different treatments in a rotational mode, proved to further enhance the retention time and achieve remarkable removal efficiencies of antibiotic-resistant bacteria with zero residues in the effluent water [72]. Besides these studies looking into the performance at different stages of CW system, some full-scale field studies investigated the removal ability of a single established CW system for various contaminants. For example, Choi et al. (2016) monitored a mature CW system receiving livestock wastewater without any spikes and their results showed various removal rates for the eight antibiotics present in the wastewater [41]. Therefore, it is important to consider whether CW is suitable for the target pollutants based on its chemistry and properties. Conversely, unsatisfying operation performance may occur if the target pollutants are out of the scope from the designed CW's treatment ability.

5. Factors Impacting CW Performance

Based on the literature research, 34 relevant studies have been reviewed for the removal of CECs by CWs. The information has been summarized in Table 1 and various parameters and their impacts on CW performance are discussed in the following section in random order.

Table 1. Removal of pollutants in agricultural runoff wastewater by constructed wetlands.

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Abdel-Mohsein et al., 2020 [72]	Hybrid	Field	Total surface area 111 m^2, depth 0.7 m Depth of VSF is 0.65 and HSF 0.15 m	2009	Sand and gravel	Coarse sand, small gravel and large gravel for both and concrete is additional for Vertical CW	NA	1.8 cm/d	6–7 d	90 days	ampicillin-resistant bacteria	NA	100%	Combination of physical (sedimentation, mechanical filtration, adsorption to organic matter), chemical (oxidation, exposure to biocides), and biological removal means (predation, competition for nutriments, lytic activity)	Full-scale hybridized CWs, with five stages (first four stages as SSVF and last one as SSHF), each stage has three units in parallel with three cells in rotation for each unit, received raw dairy wastewater, in Japan
											gentamicin-resistant bacteria		100%		
											kanamycin-resistant bacteria		100%		
											streptomycin-resistant bacteria		100%		
											vancomycin-resistant bacteria		99.9% (3.2 log 10 removal)		
Agudelo et al., 2010 [78]	SSHF	Lab	Length 1 m, width 0.6 m, height 0.6 m	NA	Gravel, igneous rock	Phragmites australis, igneous rock	0.2 m	1.1 cm/d	NA	180 days	chlorpyrifos	209.7 μg/L	96.2%	Mineralization process, adsorption into plants and gravel, and biological decomposition.	The wetlands were performed at pilot scale and treated with synthetic wastewater. Four wetlands were designed for this experiment at the university research campus of the University of Antioquia, Columbia.
												305.5 μg/L			
												425.6 μg/L			
Borges et al., 2009 [95]	SSHF	Lab	Length 24 m, width 1 m, height 0.35 m	NA	Fine gravel	Typha latifolia, fine gravel	NA	3.2 cm/d, 2.5 cm/d, 1.9 cm/d	3.8 d	77 days	ametryn	NA	39%	Biodegradation, plant uptake and desorption process	Four constructed wetland cells were used with one being a controlled cell. The wetlands were given ametryn-contaminated water. The country of origin for this experiment was Brazil.
Boto et al., 2016 [73]	SSVF	Lab	Length 0.4 m, width 0.3 m, height 0.3 m	2015	Gravel, lava rock, roots bed substrate (mixture of sand and rhizosediments (1:2))	*Phragmites australis*, roots bed substrate, lava rock and gravel	0.16 m	NA	7 d	63 days	enrofloxacin	100 μg/L	>99%	Substrate adsorption, microbial biodegradation, and plant uptake	Batch mode CWs with three treatments and triplicate for each treatment, received saline aquaculture wastewater, in Portugal
											oxytetracycline		>99%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Carvalho et al., 2013 [39]	SSVF	Lab	Length 0.4 m, width 0.3 m, height 0.3 m	2012	Gravel, lava rock, roots bed substrate	*Phragmites australis*, roots bed substrate, lava rock and gravel	0.16 m	NA	7 d	91 days	enrofloxacin	100 µg/L	98%	Adsorption and/or microbial degradation in the microcosms' substrate	Batch mode CWs with three treatments and triplicate for each treatment, received swine farm wastewater, in Portugal
											tetracycline		94%		
Chen et al., 2012 [40]	SF	Field	Surface area 2000 m^3	NA	NA	*Inter alia*, the family *Lemnaceae*; *Eichhornia crassipes*; and *Alternanthera philoxeroides*	NA	NA	20 d	350 days	tetracycline	41.6 µg/L	27.0–97.1%	Sorption, biological degradation, photolysis, and phytoremediation	Aquatic vegetation ponds (SF) applied as the last unit of a treatment system, received swine farm wastewater, in China
											oxytetracycline	23.8 µg/L	94.1–100.0%		
											chlortetracycline	13.7 µg/L	82.8–90.2%		
											doxycycline	685.6 µg/L	57.1–74.3%		
											sulfadiazine	98.8 µg/L	NA		
Chen et al., 2015 [90]	Hybrid	Field	Total surface area 981 m^2	2012	Chaff and soil	*Pontederia cordata* and *M. verticillatum L.*	NA	7 cm/d	1.5 d	NA	leucomycin	0.12 µg/L	100%	Adsorption onto medium, photodegradation, biodegradation (especially anaerobic degradation)	CW system with five units in series (consisting of SF, SSVF, SSHF), received both domestic sewage (70%) and livestock (swine) sewage (30%), in China
											ofloxacin	0.193 µg/L	100%		
											lincomycin	0.061 µg/L	78.0%		
											sulfamethazine	0.054 µg/L	95.1%		
											ARG *sul1*	2.64×10^6 copies/mL	97.2%		
											ARG *sul2*	1.14×10^6 copies/mL	95.4%		
											ARG *tetM*	1.47×10^6 copies/mL	99.8%		
											ARG *tetO*	1.02×10^6 copies/mL	98.9%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Chen et al., 2019 [83]	Hybrid	Lab	Length 0.8 m, width 0.6 m, height 0.8 m	NA	zeolite	Iris tectorum maxim, zeolite	0.77 m	40 cm/d	NA	240 days	sulfamonomethoxine	4.05–6.24 μg/L	87.8% to 99.1% ARG	Microbial degradation, substrate adsorption, and plant uptake	Eight mesocosm scale were used for the hybrid system. Four were HSSF and four were VSSF. The wetlands were constructed in the campus of Guangzhou Institute of Geochemistry. Guangzhou, Guangdong, China. Raw domestic sewage was used to treat the wetlands. The hybrid systems treated antibiotic spiked wastewater for two weeks before the first sampling.
											sulfamethazine				
											sulfameter				
											trimethoprim				
											norfloxacin				
											ofloxacin		87.4% to 97.3% antibiotics		
											enrofloxacin				
											erythromycin-H_2O				
											roxithromycin				
											oxytetracycline				
											lincomycin				
											sul 1 and sul 2				
											tetG and tetO				
											ermB				
											qnrS and qnrD				
											cmlA and floR				
Choi et al., 2016 [41]	Hybrid	Field	Total surface area 4492 m^2, total storage volume 4006 m^3	2007	NA	*Phragmites australis* (PA) and *Miscanthus sacchariflorus* (MS)	0.89 m	44 cm/d	2 d	240 days	sulfamethoxazole	10.03–11,583.33 μg/L	49.43%	Biodegradation (uptake into wetland soil and plants by microorganisms) and direct adsorption into soil and plants	CW system with six units in series (consisting of SF, SSVF, SSHF), received secondary piggery wastewater and stormwater runoff, in Korea
											sulfathiazole	1263.33–57,833.33 μg/L	81.86%		
											sulfamethazine	1055–30,033.33 μg/L	85.00%		
											trimethoprim	1.76–673.33 μg/L	2.32%		
											tetracycline	8.41–69.5 μg/L	NA		
											oxytetracycline	12.33–48.83 μg/L	NA		
											chlortetracycline	4300–16,100 μg/L	29.47%		
											enrofloxacin	34.26–262.16 μg/L	27.26%		
Du et al., 2020 [85]	SSVF	Lab	Length 0.4 m, width 0.2 m, height 0.4 m	2016	Clinoptilolite zeolite, quartz sand	*Arundo donax*	N/A	9.3 cm/d	7 d	365 days	sulfamethoxazole	6×10^{-4} μg/L	56.1–68.8%	Substrate adsorption	Four treatments, each with two separated cells (a down flow cell followed by an up flow cell), received swine wastewater, in China
											sulfamethazine	10 ng/L 0.01 μg/L			
											sulfadiazine	22.1 ng/L 0.022 μg/L			
											tetracycline	0.539 μg/L	85.9–96.4%		
											oxytetracycline	0.233 μg/L			
											antibiotic resistant genes	N/A	71.7–95.3%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Feng et al., 2021 [58]	SSVF	Lab	Height 0.65 m, diameter 0.2 m	N/A	Gravel and biochar with or without air	Gravel, biochar, and *Iris pseudacorus* with or without air	N/A	N/A	3d	180 days	tetA	5.2×10^3–7.03×10^4 copies /mL	26.2–99.3%	Biodegradation	CWs with four treatments and triplicate for each treatment, received synthetic wastewater and swine wastewater, in China
											tetM	2.7×10^5–7.15×10^6 copies /mL			
											tetO	9.87×10^4–4.82×10^6 copies /mL			
											tetW	1.51×10^5–9.42×10^5 copies /mL			
George et al., 2003 [96]	SSHF	Field	Length 4.9 m, width 1.2 m or 2.4 m height 0.1–0.3 m	1992	Quartz gravel	S. Validus, quartz gravel	0.3 m	4 cm/d	2.2 d	70 days	simazine	750 µg/L and 1400 µg/L	59%	Plant absorption and microbial degradation	Twelve cells were used with half containing vegetation and half with no vegetation. The water used in this experiment was a synthetic mix of water and pesticides. This experiment took place in Baxter, Tennessee, USA.
													59%		
								4 cm/d	2 d				78%		
													54%		
								2 cm/d	4.9 d				96%		
													78%		
								2 cm/d	3.8 d				53%		
													63%		
								1 cm/d	12.2 d				51%		
													79%		
								1 cm/d	7.3 d				57%		
													96%		
								2 cm/d	7.3 d				70%		
													96%		
								4 cm/d	3.5 d		metolachlor	3866 to 300 µg/L	62%		
													34%		
							0.46 m	4 cm/d	3.1 d				90%		
													68%		
								2 cm/d	7.7 d				96%		
													93%		
								2 cm/d	6.1 d				76%		
													60%		
								1 cm/d	19.3 d				46%		
													89%		
								1 cm/d	11.6 d				75%		
													95%		
								2 cm/d	19.3 d				87%		
													97%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Gikas et al., 2017 [92]	SSHF	lab	Length 3 m, width 0.75 m, height 1 m	2003	Medium gravel	*Phragmites australis*, gravel *T. latifolia*, gravel	0.55 m	2.4 cm/d, 1.8 cm/d	6 d, 8 d	420 days	terbuthylazine	NA	73.7% 58.4%	Uptake through plants	Two constructed wetlands were used each containing a different wetland plant. The wetlands were treated with water enriched with terbuthylazine. The experiment took place at the laboratory of Ecol. Eng. and technology, Department of Environmental Engineering, Democritus University of Thrace.
Gikas et al., 2018 [93]	SSHF	lab	Length 3 m, width 0.75 m, depth 1 m	2003	Medium gravel (carbonate rock)	*Phragmites australis*, gravel *T. latifolia*, gravel	1 m	2.4 cm/d, 1.8 cm/d	6 d, 8 d	420 days	S-metolachlor	NA	68.9%, 47.8%, 40.8%	Plant uptake and sorption on substrate. Biodegradation	Three constructed wetlands were used with one being left as the control. Water enriched with S-metolachlor was used to treat the wetlands. This experiment was conducted at Department of Environmental Engineering, Democritus University of Thrace.

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Goritio et al., 2018 [74]	SSVF	Lab	Length 0.4 m, width 0.3 m, height 0.3 m	2017	Gravel, lava rock, root bed substrate	Phragmites australis, root bed substrate, lava rock, gravel	0.22 m	N/A	7 d	30 days	Alachlor atrazine chlorfenvinphos isoproturon PFOS simazine azithromycin clarithromycin erythromycin diclofenac methiocarb acetamiprid clothianidin thiacloprid thiamethoxam EHMC atorvastatin carbamazepine cephalexin ceftiofur citalopram clindamycin clofibric acid diphenhydramine enrofloxacin fluoxetine ketoprofen metoprolol norfluoxetine ofloxacin propranolol tramadol trimethoprim venlafaxine warfarin	0.1 µg/L	>87% <87% for EHMC	Absorption into soil, plant uptake, microbial degradation	. Six microcosms assembled with three spiked and three not spiked each treating 2 L of aquaculture effluent. Aquaculture effluent added at the beginning of each week, microcosms were weekly drained and refilled with spiked or non-spiked aquaculture effluents. This experiment took place in Portugal.

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Hsieh et al., 2015 [94]	SF	Field	Length 3.4 m, width 1.4 m, height 1.5 m	NA	Soil, gravel	Cyperus, phragmite, vetiveria, gravel, soil	SF 1: 0.8 m	NA	2.2 d	540 days	chloramphenicol	0.59 ± 0.464 μg/L	98.2%	Removal process must be additionally studied though it is shown wetland was successful in removing antibiotics and other chemicals.	Three free water surface constructed wetlands were used along with a lotus pond and filter bed to achieve purification. The wetlands are field scale and usually treat wastewater on a regular basis which flow through each cell. This experiment was conducted in Taiwan.
											oxolinic acid	NA	100%		
											chlortetracycline	NA	NA		
											oxytetracycline	0.218 ± 0.170 μg/L	97%		
											tetracycline	NA	NA		
											enrofloxacin	NA	NA		
											ciprofloxacin	0.018 ± 0.009 μg/L	100%		
							SF 2: 0.8 m				sulfamerazine	NA	NA		
											sulfamonomethoxine	0.093 ± 0.020 μg/L	87%		
											sulfadimethoxine	0.0843 ± 0.0507 μg/L	90.6%		
											sulfamethazine	NA	NA		
											malachite green	NA	NA		
											leucomalachite green	NA	NA		
											nonylphenol di-ethoxylate	0.173 ± 0.275 μg/L	85.2%		
							SF 3: 0.8 m				nonylphenol mono-ethoxylates	0.291 ± 0.457 μg/L	76.5%		
											nonylphenol	1.65 ± 1.81 μg/L	89.7%		
											octylphenol	1.12 ± 3.02 μg/L	85.1%		
											bisphenol A	0.932 ± 0.684 μg/L	88.8%		
											17b-estradiol	0.189 ± 0.274 μg/L	95.2%		
											estriol	0.156 ± 0.140 μg/L	76.6%		
											17a-ethynylestradiol	0.025 ± 0.039 μg/L	31.8%		
Huang et al., 2015 [86]	SSVF	Field	Height 0.8 m, diameter 0.4 m	2012	Oyster shell, bricks, and red soil	Phragmites australis, oyster shell and red soil	0.7 m	4 cm/d	5 d	480 days	oxytetracycline	14, 64, 164 μg/L	92.7–99.9%	Substrate absorption, plant uptake, microbial degradation, hydrolysis and photodecomposition	Three outdoor VSSF (up flow) CWs served as three treatments, received swine wastewater, in China
											tetracycline	5.56 μg/L	69.0–99.7%		
											chlortetracycline	4.32 μg/L	88.4–98.3%		
											target antibiotic resistant genes	NA	45.4–99.9%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Huang et al., 2017 [87]	SSVF	Field	height 0.6 m, diameter 0.25 m	2015	Brick particle or oyster shell, red soil, and humus soil (2:1)	*Phragmites australis*, brick particle/oyster shell, red soil, and humus soil	0.55 m	5.1 cm/d	1.6–5.8 d	90 days	oxytetracycline	250 µg/L	>33%	Substrate adsorption, plant uptake, microbial degradation, hydrolysis and photodegradation	Four CWs served as four treatments (different substrates and flow directions), received swine wastewater, in China
											difloxacin	250 µg/L	>33%		
											tetracycline resistance genes	NA	>33.2 to 99.6%		
		Lab	Height 0.4 m, diameter 0.03 m		Brick and oyster shells	Brick and oyster shells	N/A	2 cm/d	N/A	20 days	oxytetracycline and difloxacin	250 µg/L	N/A		
Huang et al., 2019a [63]	Hybrid	Field	SSHF: length 0.7 m, width 0.43 m, height 0.9 m	2018	Gravel, ceramsite, zeolite, red soil	*Phragmites australis*, red soil, ceramsite, zeolite, gravel	0.9 m	300 cm/d	6 d	84 days	tilmicosin	2500 µg/L	100%	Adsorption and degradation of antibiotics	Six CW cells with three treatments, each treatment with two cells in series (SSHF-SSVF(U); SSHF-SSVF(D); SSFV(D)-SSVF(U)), received goose wastewater, in China
			SSVF: diameter 0.62 m, height 0.9 m								doxycycline	30 µg/L	98–99%		
											intt1, ermB, ermC	NA	>90%		
											tet genes except tetX	NA	>50%		
Huang et al., 2019b [88]	SSHF	Field	Length 1.5 m, width 0.4 m, depth 0.8 m	2014	Gravel and zeolite	*Iris pseudacorus* and/or *Phragmites australis* (50 plants/m^2), zeolite, gravel	0.6 m	8.4 cm/d	3 d	120 days	enrofloxacin	0.026–0.067 µg/L	75.6–81.1%	Adsorption for ENR; degradation, transformation and anaerobic fermentation for SMZ	Four SSHF cells served as four treatments (different plant species and planting patterns), received aquaculture farm wastewater, in China
											sulfamethoxazole	0.064–0.211 µg/L	54.3–68.7%		
											antibiotic resistant genes	NA	36.5–58.2%		
Hussain, 2011 [77]	SSHF	Lab	Length 6.1 m, width 1.5 m, height 0.75 m	2011	Sandy soil	Phalaris arundinaceae, Typha Latifolia, Sandy Soil	4.17 m cubed	NA	4 d	30 days	monensin, salinomycin, narasin	10,50,100, and 500 µg/L	Monensin 42%, salinomycin 49%, narasin 49%	Sorption into soil, biodegradation, microbial dissipation	SSHF constructed wetland performance was compared to the performance of free water surface performance wetlands. A mixture of contaminated wastewater was used to treat the wetlands. Three CW units were assembled for the experiment. Water was supplied through an inflow manifold. This experiment took place in Canada.
Liu et al., 2013 [38]	SSVF	Field	Height 0.7 m, surface area 1 m^2	NA	Volcanic rocks/zeolite	Red soil; volcanic rocks (CW1)/zeolite (CW2); gravel	0.7 m	3 cm/d	1.25 d	120 days	ciprofloxacin HCl	All at 40 µg/L	82% and 85%	Sorption to wetland medium	Two SSVF CWs served as two treatments (different substrate medium), received swine farm wastewater, in China
											oxytetracycline HCl		91% and 95%		
											sulfamethazine		68% and 73%		
					Volcanic rock						tet gene and 16S rRNA	N/A	50%		
					Zeolite						tet gene and 16S rRNA	N/A	90%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Liu et al., 2014 [89]	Hybrid	Field	SF and SSVF: height 0.8 m, surface area 6 m^2; SSHF: height 0.8 m, surface area 12 m^2	NA	Oyster shell, red soil	*Phragmites australis* (16 stems/m^2), red soil and oyster shell (optional)	SF: 0.3 m; SSVF: 0.1–0.4 m; SSHF: 0.4 m	SF and SSVF: 2 cm/d; SSHF: 4 cm/d	SF: 15.5 d; SSVF: 7.3–14.2 d; SSHF: 16.4 d	428 days	sulfamethazine	25–35 µg/L	40% for SF; 59% for SSHF; 87% for SSVF-L; 70% for SSVF-H	Mainly dependent on the physicochemical process (adsorption)	Four pilot-scale CWs served as four treatments (SF, SSHF, SSVF-L, SSVF-H), received swine farm wastewater, in China
											tetracycline	3.5–6 µg/L	92% for SF; 92% for SSHF; 99% for SSVF-L; 98% for SSVF-H		
Locke et al., 2011 [91]	Hybrid	Field	Length 180 m, width 30 m (average), depth 0.45 m (average)	2003	Sediment	Alligatorweed [*Alternanthera philoxeroides* (Mart.) Griseb.], and two grass plants, junglerice [*Echinochloa colona* (L.) Link] and barnyardgrass [*Echinochloa crus-galli* (L.) Beauv.]	0.3–1 m	2667 cm/d	NA	21 days	atrazine	Both at 10,650 µg/L	89%	Adsorption/uptake by plants	A full-scale CW consisted of one sediment trap and two treatment cells in series, received simulated agricultural runoff, in the United States
											fluometuron		81%		
Lyu et al., 2017 [97]	NA	Lab	Height 0.2 m and diameter 0.2 m	2014–2015	Gravel, geotextile, sand	Juncus effusus, typha latifolia, berula erecta, phragmites and iris pseudacorus, gravel, sand geotextile, gravel	About 0.18 m	1.7, 3.4, 6.9, 13.8 cm/d	2, 1, 0.5, 0.25 days	57 days	tebuconazole	10 to 100 µg/L	99.8%	Plant uptake (biodegradation and metabolization inside plant) substrate sorption	In total 36 mesocosm scale constructed wetlands were used. Water used was artificially spiked with tebuconazole.
Moore et al., 2001 [79]	NA	Lab	10 m width, 66 m length, 0.24 m depth	NA	Sandy loam	Juncus effusus, leersia, ludwigia, sandy loam	0.24 m	NA	NA	84days	chlorpyrifos	147 µg/L	>83%	Sorption by plants and sediments.	Eight CW cells were used, 4 experimental, 1 controlled and 3 as simulated rainfall. Wetlands ere set up at the University of Mississippi Field Station in Mississippi, USA. Chlorpyrifos of various concentration were used to treat the wetlands.
												73 µg/L			
		Field	36 m wide, 134 m length	1991	Silty loam	Typha capensis, Juncus kraussii, Cyperus dives, silty loam	0.3–1 m					147 µg/L			
												733 µg/L			
												1.3 µg/L			
Papaevangelou et al., 2017 [98]	SSHF	Lab	3 m length, 0.75 m width, 1 m depth	2003	Fine gravel	Phragmites australis, gravel	1 m	2.4 cm/d, 1.8 cm/d	6 d, 8 d	1 year	boscalid	NA	75.1%	Adsorption of organic matter in substrate material and plant uptake	Two constructed wetlands were used in this experiment each containing different substrate media. Water enriched with boscalid was used for the experiment. This experiment was performed at Department of Environmental Engineering, Democritus University of Thrace.
					Cobbles	Phragmites australis cobble							72.5%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Santos et al., 2019 [42]	SSVF	Lab	Length 0.4 m, width 0.3 m, height 0.3 m	2014	Gravel, lava rock, root bed substrate	*Phragmites australis* (Cav.) Trin. ex Steud, root bed substrate, lava rock and gravel	0.3 m	NA	7 d	140 days	enrofloxacin	both at 100 µg/L	>90%	Substrate adsorption, microbial biodegradation, and plant uptake	Batch mode CWs with four treatments and triplicate for each treatment, received swine farm wastewater, in Portugal
											ceftiofur		>90%		
											antibiotic resistant bacteria	N/A	>90%		
Sherrard et al., 2003 [80]	NA	Lab	Length 1.85 m, width 0.63 m, height 0.63 m	NA	Sand/organic matter	C. Dubia, P. Promelas, organic mixture/sand	About 0.3 m	NA	3 d	Performed on consecutive weeks	chlorpyrifos	0.90 µg/L, 19.9 µg/L, 19.4 µg/L	98%	Plant uptake	Four experiments were carried out to assess the impact of pesticide removal in constructed wetlands. Well water spiked with pesticide were used for this experiment. This experiment took place in the USA.
											chlorothalonil	148 µg/L, 326 µg/L, 296 µg/L	100%		
Souza et al., 2017 [81]	SSHF	Lab	0.35 m height, 0.5 m length, 2 m width	NA	Pea gravel, biofilm (sewage)	Cyon spp., M. aquatica, P. punctatum, biofilm mixture, pea gravel.	0.35 m	NA	1 d, 2 d, 4 d, 6 d, 8 d	30 days	chlorpyrifos	1000 µg/L	98.6%	Adsorption due to the biofilm and plants. Biodegradation	Four wetlands were used with different vegetation. One was a controlled. Water used in this experiment was spiked with chlorpyrifos. The experiment took place at the Department of Agricultural Engineering, Federal University of Vicosa.
Tang et al., 2018 [82]	SSVF	Lab	Diameter 0.24–0.27 m, height 0.3 m	NA	Gravel	C. Alternifolius, C. Indica, I. pseudacorus, J. effusus, T. Orientalis, gravel media	0.15 m	NA	7 d	42 days	chlorpyrifos	50 µg/L and 500 µg/L	94–98%	Sorption and biodegradation. Plants enhance removal through biodegradation	Constructed wetlands that were planted performed better in removing pesticides than the control groups which had no plants. Synthetic wastewater was used in the experiment which was conducted in China.
Wu et al., 2016 [99]	SSHF	Lab	Length 1.2 m, width 0.4 m, height 0.8 m	NA	Ceramsite	C. Indica, Ceramsite	0.8 m	20 cm/d	NA	365 days	triazophos	100 µg/L	96%	High urease activity of the substrate	Four constructed wetlands were used with one a control. Relationships between pollutant and microbial communities were discussed. Different concentrations of triazophos and raw water were pumped into the wetland systems.
												1000 µg/L	97%		
												5000 µg/L	75%		

Table 1. *Cont.*

References	Wetland Type	Scale	Size	Year of Construction	Substrate	Vegetation/ Layering	Water Depth	HLR	HRT	Run Time	Pollutant Studied	Influent Concentration	Removal %	Major Removal Processes	Factors/Conditions Studied/Wastewater Type/Country
Xian et al., 2010 [62]	SF	Field	Length 0.5 m, width 0.4 m, height 0.4 m	2009	HDPE foam plates	Ryegrass (*Dryan*, *Tachimasari* and *Waseyutaka*)	0.3 m	NA	NA	35 days	sulfadiazine	100 μg/L	98.7–99.2%	Sorption, abiotic transformation and biotic transformation	Three SF CWs served as three treatments (different plant species), received raw swine wastewater, in China
											sulfamethazine	100 μg/L	88.8–91.8%		
											sulfamethoxazole	10 μg/L	99.0–99.5%		
Zhu et al., (2020) [56]	Hybrid	field	Surface area 600 m^2	2004	N/A	N/A	N/A	N/A	7 d	N/A	antibiotic resistant genes	N/A	62%	Microbial degradation and physical adsorption	CW applied as the last unit of a treatment system, received swine farm wastewater, in China

Note: SF—surface flow; SSHF—subsurface horizontal flow; SSVF—subsurface vertical flow; HLR—hydraulic loading rate; HRT—hydraulic retention time; NA—not applicable.

5.1. Target Contaminant Property

Based on the various physicochemical properties, such as pKa, molecular weight, solubility, and functional groups, different contaminants showed different levels of removal by CW systems. From the study by Choi et al. (2016), the major removal mechanism was the adsorption to soil, which was favored for compounds with lower molecular weights and higher pKa values [41]. Gorito et al. (2018) also suggested that high removal of azithromycin through sorption onto the soil and plant uptake were likely due to its high octanol–water coefficient (K_{ow}) and pK_a values [74]. In addition, contaminants with low solubility and high soil adsorption coefficient (K_{oc} > 1000) would have better sorption and retention in soils. For example, Gikas et al. (2018) found poor adsorption of selected pesticides due to moderate solubility and low K_{oc} [92]. This was also supported by a pesticide study conducted by Agudelo et al. (2010) as target contaminant chlorpyrifos with low solubility and high adsorption coefficient showed great sorption into the soil substrate or the humic colloids suspended in the water [78]. Vystavna et al. (2017) indicated that compounds, such as propranolol, tend to accumulate in sediments due to its hydrophobicity, therefore, the utilization of porous filter materials with high sorption ability could improve the removal percentages for such compounds [100]. Functional group and structure could also impact pollutant removal mechanism, as the Lyu et al. (2018) study showed that hydrolysis was negligible for tebuconazole removal due to its chemical properties [97]. Overall, to achieve optimal removal performance by CW systems, one should consider the physical and chemical properties of the target contaminant during the design of the CWs as those properties are likely to impact their removal mechanisms.

5.2. Aeration

Depending on the type of CW system and the specific spot within a CW unit, aerobic or anoxic conditions may exist favoring certain pollutant removal processes. The removal of antibiotics, such as monensin, salinomycin and naracin, through microbial degradation was most active at the water/air interface or within the root zone under aerobic conditions [77]. Other works have also shown aerobic conditions to support the removal of veterinary pharmaceuticals from wastewaters [101]. On the contrary, the biodegradation of chloroacetanilide herbicides might be favored under anoxic conditions, as Elsayed et al. (2015) found that bacterial communities were most abundant and active at anoxic rhizosphere zone and anaerobic degradation accounted for the most dissipation of chloroacetanilides [102]. Besides the natural established aerobic/anaerobic conditions, some studies also introduced artificial aeration to promote the removal rates. For example, Chen et al. (2019) compared four different hybrid CW systems with/without the addition of aeration from an air blower and their results showed enhanced ARGs removal rates in both VSSF and HSSF with additional aeration [83]. Similar findings were also observed in a Feng et al. (2021) study, as they also noticed improved target ARGs removal with aerated treatments [58]. This indicated that for future applications, aeration units should be considered in the CW system design to improve the ARGs removal efficiencies. Alternatively, better designs to enhance aeration naturally in the CWs will likely enhance removal of ARGs.

5.3. Types and Design of CWs

Out of the 32 studies listed in Table 1 with identifiable CW type/design, 3 of them (around 9%) were surface flow (SF), 10 of them (around 31%) were subsurface horizontal flow (SSHF), 11 of them (around 35%) were subsurface vertical flow (SSVF), and 8 of them (around 25%) were hybrid systems containing more than one type (SF/SSHF/SSVF). In general, SSHF and SSVF are more widely applied in single CW type studies comparing to SF. This is due to how SSHF CW provides an anoxic system which promotes denitrification and other anoxic microbial processes; whereas SSVF CW provides an aerobic system which supports nitrification and other aerobic microbial processes [72,102]. In addition, SSVF CW can also remove organic compounds and suspended solids effectively [72]. In order to achieve better removal efficiency for various pollutants, a lot of studies applied hybrid system containing SSVF and SSHF [72,90,102]. For example, Huang et al. (2019a) showed that all three two-stage CW systems removed over 98% of the antibiotics without significant differences among treatments [63]. SSFV (down-flow) and SSVF (up-flow) had a better performance for ARGs and nutrients (especially for N) removal due to its establishment of anaerobic ammonium oxidation condition and limitation of bacterial growth [63].

Besides CW types, studies have also investigated the impacts of different flow directions (up-flow vs. down-flow) [85,87] as well as the water level (high-level vs. low-level) [89]. Their results showed that the configuration of down-flow SSVF followed by up-flow SSVF provided best pollutant removal performance, however, they also expressed concern about accumulation of ARGs in the surface soil for down-flow SSVFs [85,87]. Meanwhile, Liu et al. (2014) reported relatively higher removal efficiencies for SSVF with low water level [89] and this was supported by Lyu et al. (2018) as they found significantly higher tebuconazole removal in unsaturated CWs than saturated CWs [97]. For larger field scale studies consisting of multiple CW units, various configurations are utilized. The most common one was connecting CW units in series [40,41,90,91] but there were more complicated setups in some studies. For example, George et al. (2003) first had eight cells connected in parallel as the first stage and the remaining six cells connected in parallel as the second stage with series connection between stages [96]. Another study conducted in Japan had five stages connected in series and three treatments connected in parallel for each stage; and within each treatment there were three cells operating in rotational mode [72,102]. Such sophisticated design could not only provide better removal performance but also allow the avoidance of cross-contamination between different cells and provide the chances to perform operation and maintenance on a specific cell without disturbing the entire system.

5.4. Hydraulic Parameters (HLR and HRT)

Compared to wastewater treatment plants, CWs typically need lower HLR and longer HRT to achieve the similar level of removal performances. The hydraulic parameters are very important to consider during CW system design since lower HLR/longer HRT may provide better treatment but require much larger land area, while higher HLR/shorter HRT may occupy a smaller footprint but face low treatment efficiency and frequent clogging events and need more operation and maintenance inputs. Based on the study data listed in Table 1, Figures 3 and 4 were plotted to show the relationships between HLR/HRT and target contamination removal percentage. It is apparent from Figure 3 that removal efficiency had a positive correlation with HRT, meaning a greater removal rate with the longer retention time. After 7 days, an average removal efficiency of 90% was achieved, which is in agreement with the findings from previous research that a hydraulic retention time of 6–7 days was adequate for the removal of pollutants [93,103]. Meanwhile, Figure 4 showed that with the increase in HLR, removal efficiency would first increase, reach to a steady level (>90%), and later start to decrease. That is to say, the ideal HLR should be 10–30 cm/d for best pollutant removal performance, as greater or lesser HLR would both result in reduced removal efficiency. This was supported by findings from Lyu et al. (2017) as they reported decreasing removal rates over increased HLR [97]. Therefore, choosing the appropriate HRT/HLR for CW system has great impacts on the system performance. Furthermore, in some studies, hydraulic retention times were adjusted based on seasons, with them being longer in warmer seasons (8 days) and shorter in colder seasons (6 days) to address the water requirement variations due to evapotranspiration [92,93,98].

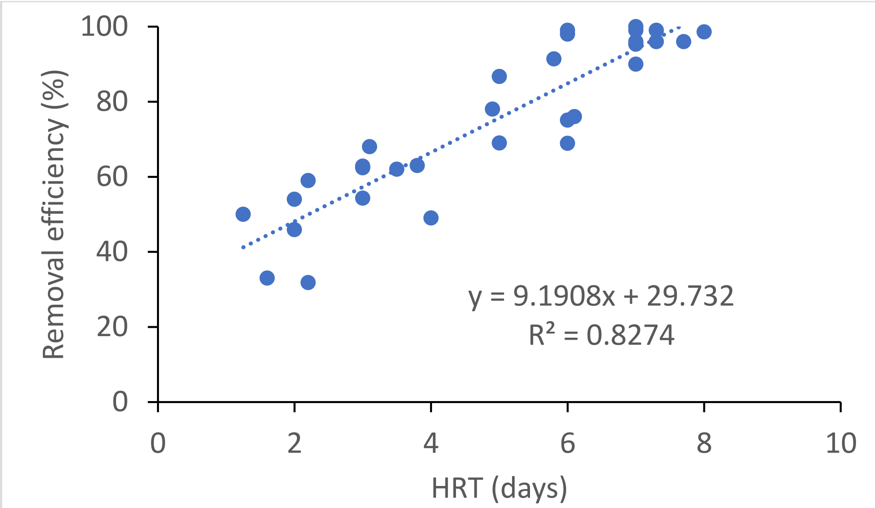

Figure 3. Relationship between HRT and removal efficiency in CWs.

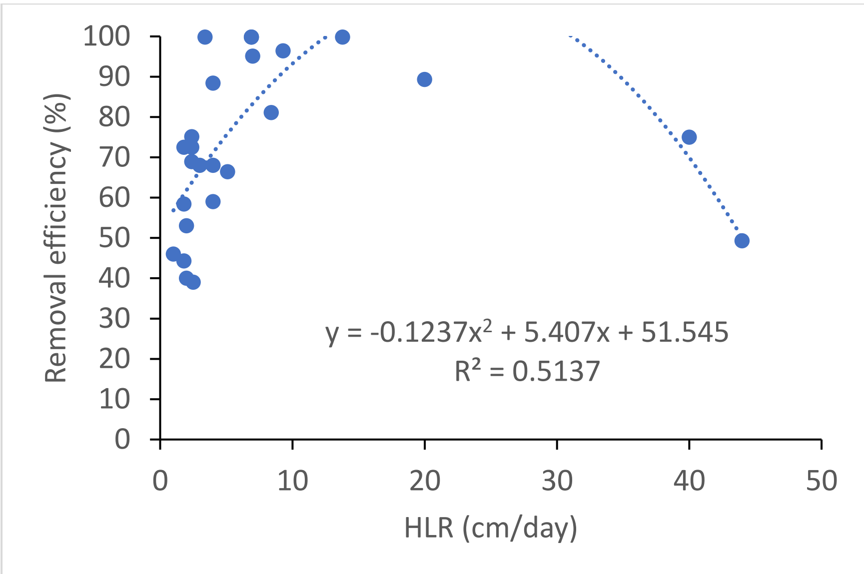

Figure 4. Relationship between HLR and removal efficiency in CWs.

5.5. Substrate Medium

Since adsorption is one of the major mechanisms for pollutant removal in CW systems, the substrate medium's physical and chemical properties would have a huge impact on the removal performances. The Papaevangelou et al. study (2017) compared two substrates (fine gravel and cobbles) from the same riverbed with various sizes and found better removal performance of fine gravel for target pollutant-boscalid (fungicide) in the preliminary tests but no significant difference in performance of the substrates over long-term field study [98]. A lot of previous research have compared the removal efficiency of specific contaminants with various substrate medium. For example, Liu et al. (2013) showed that compared to volcanic rock, zeolite had a lower point of zero charge (PZC) indicating a higher affinity to cationic form of antibiotics at neutral pH levels and had smaller pore size indicating greater sorption sites; therefore, zeolite showed better removal efficiencies for selected antibiotics and ARGs [38]. Similar observations were made by Du et al. (2020) as zeolite medium had a better removal performance for both antibiotics and ARZs compared to quartz sand medium [85]. In the Huang et al. study (2017), brick-based substrate achieved better antibiotic removal performance compared to oyster shell-based substrate due to two major reasons: (1) greater porosity and average pore size that provided more surface areas for sorption processes; (2) higher iron oxides contents in brick that provided better adsorption capacity [87]. Besides zeolite and brick material that were widely applied in CW substrates, medium with high organic matter content was also investigated since it could potentially increase pollutant removal through interactions with organic functional groups (such as phenolic and carboxyl groups), hydrogen bonding, and ion exchange [104]. For example, Feng et al. (2021) compared biochar and gravel based CWs and found that while treatment with only biochar-based substrate had no significant improvement in target contaminants removal, treatment with both biochar-based substrate and aeration showed much higher removal rates [58]. They also measured abundance of ARGs in the substrate indicating the accumulation of antibiotics in the substrate and proliferation of ARGs during the long-term operation [58]. That is to say, appropriate operation methods need to be taken to address the potential risks of ARGs development in such substrates. Therefore, to achieve better elimination of ARGs in practical approaches, the suitable selection of CW substrate medium is an important decision.

5.6. Vegetation

Vegetation is another key component in CW systems as plants cannot only directly uptake pollutants, but also modify the surrounding environment, for example, by transporting oxygen into a rhizosphere to enhance the diversity and biomass of microorganisms, microbial degradation, and sequestration [104]. Several studies have compared treatment with plants versus without plants and most of them showed higher removal performance for herbicides [92,93,96] and pesticides [82,97] with plants as treatment, while one study presented no significant difference with plant treatment for veterinary antibiotics [39]. Research has also been performed to compare the removal performances of various plant species. For example, Lyu et al. (2018) compared five plant species (*Typha latifolia*, *Phragmites australis*, *Iris pseudacorus*, *Juncus effusus* and *Berula erecta*) for pesticide removal and found *Berula* to contribute to significantly higher removal efficiency compared to the rest four plant species [97]. Moreover, Tang et al. study (2019) indicated that *Canna indica*, *Cyperus alternifolius* and *Iris pseudacorus* had better removal performance for pesticides than *Juncus effusus* and *Typha orientalis* [82]. However, Souza et al.'s study (2017) showed no significant differences in pesticide removal among *Polygonum punctatum*, *Cynodon spp.* and *Mentha aquatica* [81].

Another study also confirmed that vegetation type impacts antibiotic removal efficiencies in surface flow CWs. The authors compared three varieties of ryegrass (*Dryan*, *Tachimasari* and *Waseyutaka*) to treat three antibiotics (sulfadiazine, sulfamethazine, and sulfamethoxazole) and found that *Dryan* outcompeted the other two types of plants due to its highest removal rates for both nutrients and antibiotics [62]. Gikas et al. (2018) compared treatments planted with *Phragmites australis* and *Typha latifolia*, with an unplanted control and the results showed that *Phragmites australis* had the highest removal capacity for both herbicide (S-metolachlor) [96] and pesticide (terbuthylazine) [92].

Besides the plant species, studies have also shown that various planting patterns may impact the removal performance. Huang et al. (2019) compared CW treatments with single plant species and mixed plant species and found that CWs with single plant type performed better in reducing antibiotic and ARG concentrations [88]. These findings imply that different plant species and planting patterns should be applied to achieve best performance depending on the target contaminant. Furthermore, studies indicated that after a certain time of exposure to the pollutant, the plant would uptake the pollutants with more concentrations in the root part than in the shoot part [62]. Harvesting the vegetations planted in the CW reduced the concentration of antibiotics in the soil, implying plant harvest as an effective procedure to maintain sustainable efficient removal performance [86].

6. Research Bottlenecks and Prospects

As stated in previous sections and presented in Table 1, numerous studies and reviews have been performed either on the topics of constructed wetlands pollutant removal performance or chemicals of emerging concern (such as pesticides, herbicides, veterinary antibiotics, etc.), but fewer studies have been focused on the overlapping research area of these two topics, which is using constructed wetlands to remove CECs. Among those studies, even fewer are related to agricultural runoff, since most of them studied treating domestic sewage or effluent from wastewater treatment plants. Even those on agricultural runoff, the studies are dominated by nutrients and sediment. Therefore, most future research needs to be performed on the application of CWs to remove CECs from agricultural runoffs. In addition, compared to livestock and poultry wastewater treatment applications, even fewer data were collected and reported from aquacultural wastewater and farm runoff either due to irrigation or precipitation. That is to say, more studies need to be conducted in these specific areas to safeguard our water resources, environmental, and human health.

From the scale's perspective, the majority of current studies are mainly in lab scale or pilot field scale, with only a few papers reporting the data from full-scale field studies. Small-scale studies in a controlled environment in the laboratory or greenhouse setting are valuable to serve as the first step attempt to address the research questions, but eventually large-scale studies fitting the real-world scenario are still needed for future applications. The designed CW system needs to be tested under real field conditions with fluctuating temperatures, flow rate, redox state, etc. to prove its durability. Nowadays, climate change has resulted in more extreme weather conditions happening more frequently; therefore, future research should also take consideration of the impacts of extreme weather, such as flooding and drought, on designed CW systems. To assist the optimization of design parameters, predication models coupled with remote sensing data could be built for simulating various conditions and potential extreme weather events. With the screening feedback from such models, suggestions could be provided for future application development.

In addition, after a period of operation, the CWs could accumulate the CECs within the system and lead to the development of ARGs into the local environment by self-developing and transferring to other microorganisms [101]. Especially for the down-flow SSVF CWs, the enrichment of pollutants and ARGs in the surface soil could become problematic in the long term [86,87]. Therefore, it is necessary to investigate methods to periodically remove and safely treat the accumulated contaminations from the CW system in order to maintain a sustainable high removal performance in the long term. Currently, very few papers have reported such operation and maintenance practices for CW applications.

For the theoretical investigation part, it is broadly accepted that the CW removes pollutants through a variety of processes, including adsorption to the substrate and soil, plant uptake, and biological degradation. The physiochemical sorption process has been well studied based on parameters, such as solubility (S), sorption coefficient (K_d), octanol–water coefficient (K_{ow}), oxidation-reduction potential (ORP), pH and pKa, with a lot of studies reporting certain correlations between the above parameters and removal efficiencies. However, most of the current studies failed to provide detailed explanations on biological processes and their role in the pollutant removal [40,41,86,89]. Therefore, further research is also needed for understanding the mechanisms of microbial biodegradation and plant uptake of CECs within the CW systems. For example, more research can explore various microorganisms' functions under aerobic/anaerobic conditions and compare contaminant uptake at different plants parts (root/stem/leave/shoot/etc.). The identification of optimal conditions for biodegradation and extraction of plant tissues with highest accumulation could be beneficial for future CW system applications by providing suggestions of ideal set-up conditions as well as operation protocols, such as harvesting the plant parts with greatest pollutant accumulations to maintain a high removal rate throughout the entire treatment period.

Current studies have also reported contradictory results of ARG occurrence and removal within the CW systems as some of the studies showed significant removal of ARGs with CWs since they arrest and inhibit the growth of bacteria, while others reported increases of ARGs due to the exposure and adaptations to accumulated contaminants in the substrate/soil. Therefore, future research is also needed to determine the internal complicated processes and mechanisms underlying various conditions of ARG sequestration and removal within the CW system. Based on these results, application suggestions of CW could be provided to avoid ARG accumulation during operation. In addition, further studies could be performed on the evaluation of potential impacts of ARG accumulation within the CW system, such as whether accumulated ARGs are going to change the structure of microorganisms within the system and the system performance; or whether the accumulation may lead to increase in effluent ARG concentrations. If severe impacts are noticed from such accumulation, future research on appropriate approaches to prevent the ARG accumulations will be needed.

With successful CW design, we can treat wastewater containing various contaminants in an efficient and economical manner. However, there are several ways we can improve the performance removal of the pollutants by CWs. For example, finding ways to promote aeration in the CWs can enhance aerobic biodegradation. Additionally, selection of substrate medium is key to achieving better elimination of ARGs. Studies also showed hybrid setup to perform differently based on the order of SF or SSF. Moreover, plant species affect the performance of the CWs. Consequently, screening of plants and plant selection is important for improving the removal efficiency. Another potential method is to improve the design of CWs, for example, CWs in series to boost performance.

Because nature-based systems need time to establish and function, real field studies over longer period without spiking concentrations are needed. As short-term studies with spiked concentrations may not represent the true removal efficiency, real field studies conducted for a long time are required. In addition, sampling strategies, for example, before vs. after in long-term study rather than treatment vs. control, may be needed to represent the efficiency of removal.

7. Conclusions

The paper reviewed recent findings on the applications of wetlands for the treatment of agricultural wastewater that contained pesticides and herbicides, veterinary medicines, antimicrobial resistant bacteria, or ARGs. By the volume of the search results, it can be concluded that these topics are understudied but are gaining major attention lately, likely due to concerns with ARGs. Wetlands are nature-based treatment systems, which are capable of treating many pollutants in the agricultural wastewater simultaneously by utilizing several physico-chemical and biological mechanisms. For example, adsorption to the substrate and plant's root (physical process), microbial metabolization and degradation (biological and chemical processes), and plant uptake (biological process) were found to be responsible for removal of veterinary medicines. While a major removal mechanism for antibiotics was microbial degradation, substrate sorption was a major mechanism for ARGs. The major parameters, such as target contaminants' property, aeration condition, types and designs of CWs, hydraulic parameters, substrate medium and vegetation that impact the CW system's removal performances, were also discussed to provide suggestions for successful future designs. Since CWs are adaptable to various wastewater treatments with satisfying removal efficiencies, CWs can be a key tool to fight against current and emerging environmental problems, especially when resilient and climate smart solutions are needed more than ever.

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