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Review

Vertical and Hybrid Constructed Wetlands as a Sustainable Technique to Improve Domestic Wastewater Quality

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Abstract: Developing safer and environmentally friendly methods for wastewater management is a crucial issue worldwide. Pollutants stemming from pure elemental, organic or inorganic compounds, or microbial sources, are an increasing problem in domestic wastewater. Constructed wetlands (CWs) have been used as an effective and low-cost method of treating different types of polluted water. This review paper focuses on the effectiveness of pollutant-removal from domestic wastewater using vertical flow constructed wetlands (VFCWs) and hybrid constructed wetlands (HCWs). Meta-analysis and ANOVA tests were conducted to analyse the potentiality of VFCW and HCW as a remedy for domestic wastewater and the effect of using different substrates and plant species. Meta-analysis shows a high significance (p = 0.001) between the interactions (method, plant, and substrate) on the pollutant's removal efficiency. In both analysis methods, there were no significant differences between VFCW and HCW for the same pollutant (p > 0.05); the average removal percentages when using VFCW and HCW (according to ANOVA analysis) were 80% vs. 90% for BOD, 78% vs. 77% for COD, 75% vs. 83% for ammonium-N, 48% vs. 56% for TN, and 60% for TP, respectively. Moreover, this review article presents a comprehensive overview of the removal mechanisms for organics, inorganics, and metals from domestic wastewater using VFCW, and the effects of environmental parameters including substrate type, plant species, and dissolved oxygen which have direct and indirect impacts on physical, chemical, and biological removal mechanisms. In conclusion, VFCWs and HCWs seem to be an excellent approach, offering economical and environmentally friendly techniques for domestic wastewater treatment, but VFCW is considered simpler and more applicable for setting up on-site near houses, as there is no significant difference (p > 0.05) between applying VFCW or HCW on removal percentages for most pollutants, according to ANOVA testing. More work is needed to study the effect of non-planted VFCWs and HCWs on removal efficiency.

Keywords: constructed wetlands; wastewater; domestic wastewater; heavy metal; organic pollutants; inorganic pollutants; vertical flow constructed wetlands; hybrid constructed wetlands

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1. Introduction

The shortage of freshwater availability is a worldwide issue caused by urbanisation and population explosion; climate change which results in the drying of freshwater sources; and industrial and agricultural activities, besides the use of fresh water for domestic purposes [1]. About 1.2 billion per capita worldwide do not have access to potable water [2], and nearly two-thirds of the world's population suffers from water scarcity at least one month per year [2]. Some regions such as the Middle East, Central and Southern Asia, North Africa, and East and South Africa are considered vulnerable to climatic change and water scarcity [3–5].

Wastewater treatment is a vital need to mitigate environmental pollution and to meet the growing demand of the global population for freshwater resources [6]. Recycled water can be reused in industrial and agricultural activities, saving freshwater for human consumption [7]. Wastewater is defined as grey, turbid water with an unfavourable odour and a profound change in its physical, chemical, and biological properties after being used by humans or exposed to pollution sources [8]. Globally, the annual production of wastewater is approximately 359.4 billion m^3 [9], with about 75.4% equal to 271 billion m^3 for domestic wastewater. The seepage of wastewater into the groundwater and surface water seriously threatens the environment. The contamination of soil with wastewater increases its salinity, alkalinity, heavy metal concentration, and electrical conductivity [7,10,11]. Several studies found that soil polluted with wastewater can adversely affect plant growth and productivity [10,12,13]. Wastewater introduces organic and inorganic pollutants into the environment and poses a serious threat to terrestrial ecosystems and the food chain [14,15]. Wastewater is divided into domestic, industrial, and stormwater runoff (Figure 1).

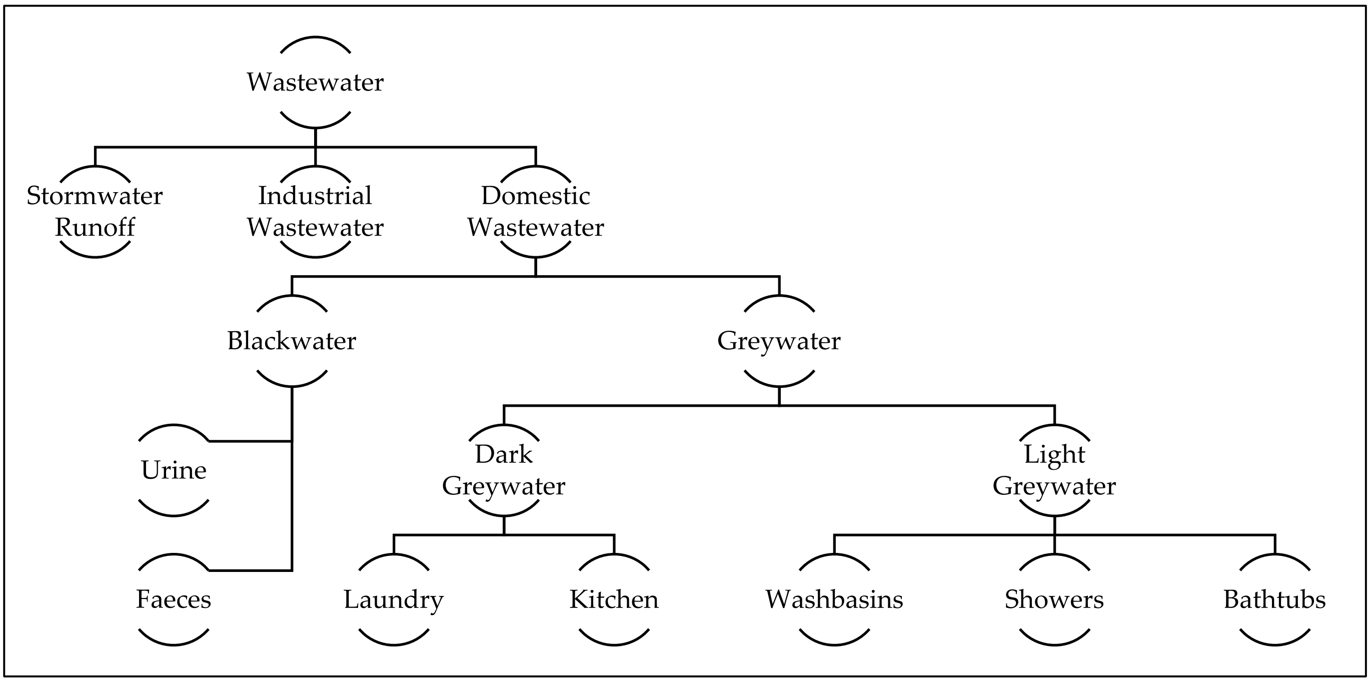

Figure 1. Schematic drawing depicts the classifications of wastewater and illustrates the sources of domestic wastewater.

The purpose of wastewater treatment is to minimise the organic, inorganic, pathogenic, and heavy metal pollutants as much as possible to match the limits set by WHO or a specific country [16]. Numerous remediation technologies have been employed to remove different types of pollutants from contaminated water to minimise its hazardous impacts [17]. The conventional ways of treating wastewater involve physical, chemical, and biological techniques, such as activated sludge, a trickling filter, rotating biological contactor, and the aerobic rotating biological contactor (RBC) [6,11]. Some conventional remediation methods are considered ineffective and improper because of their high construction and maintenance costs, difficulty of operation, and high energy consumption. In addition, these methods are mostly not environmentally friendly as they produce undegradable sludge as a byproduct [18–22].

In recent decades, growing interest in sustainable natural methods for wastewater treatment has emerged; among these methods is constructed wetlands (CWs), developed early in the 1950s in Germany [23,24]. CWs are characterized by low operational and maintenance costs and energy-use efficiency. It is considered an effective treatment technology for several types of polluted water, including domestic wastewater, agriculture wastewater, industrial wastewater, landfill leachates, and stormwater [1]. Pollutant-removal in CWs occurs through chemical, physical, and biological mechanisms among the CW components, water, substrates, plants, and microorganisms [25].

The main objective of this review article is to summarise the removal-efficiency of various pollutants (organic matter, inorganic, and heavy metals) from domestic wastewater using VFCWs and HCWs by utilising meta-analysis and ANOVA tests. Moreover, this article also discusses the removal mechanisms for the previously mentioned pollutants from wastewater and the effect of environmental parameters on removal mechanisms using VFCWs.

2. Pollutants in Domestic Wastewater

Domestic wastewater contains numerous pollutants mainly consisting of biological and chemical contaminants. Biological pollutants are pathogenic microorganisms derived from human waste (faeces and urine) via the toilet [26]. On the other hand, chemical pollutants consist of organic matter such as hydrocarbons, proteins, carbohydrates, detergents, fats, and inorganic pollutants including phosphorous compounds, nitrogenous compounds, and heavy metals [26,27]. Domestic wastewater is subdivided into greywater and blackwater [28]. Greywater stems from bathtubs, sinks, washing machines, and kitchens, and contains no human or animal excreta. In general, it contains chemicals such as oils, hydrocarbons, aromatics, surfactants, fabric softeners, chloride, and sulphates contained in household detergents [14,26,29]. By comparison, blackwater originates from toilet effluent and is considered more polluted than greywater due to its high content of microorganisms [7,28]. The average production of domestic wastewater and its characteristics vary from one region to another. The living standards of the family members determine the amount and the ingredients of the consumed products [28,30–32]. Greywater produces high organic pollutants, suspended solids, turbidity, and pH higher than 6 [29,33]. As much as 60–80% of the domestic wastewater from households is classified as greywater; it contains up to 30% organic matter, and the nutrient contents range from 10 to 20% [7,28,30]. Based on its concentration of contaminants, greywater is categorized as light greywater or dark greywater (Figure 1) [28,30,32]. Additionally, greywater contains high concentrations of inorganic pollutants such as heavy metals (i.e., Zn, Cu, and Pb), N, and P [7,34]. Dark greywater effluents from the kitchen and laundry have more pollutants than light greywater as they may contain microorganisms from food residue, including bacteria such as Salmonella, viruses, fungi, and parasites, which cause diseases like Diarrheal [7,28,30].

Water comprises the majority of domestic wastewater with a percentage of 99.9%, while the rest consists of solid compounds which are divided into organic matter (70%) and inorganic (30%). Organic matter consists primarily of proteins (65%), carbohydrates (25%), and fats (10%) [8].

According to UN-Habitat and WHO [35], 56% of global domestic wastewater is treated in safe ways. The proportions of the safe-treated domestic wastewater range from 25% to 80%, varying according to the economic classification of the region. The percentage of safe-treated domestic wastewater in North America and Europe, Western Asia and North Africa, and Sub-Saharan Africa are 80, 63, and 28%, respectively [35]. Since almost 70% of global freshwater is used in agriculture, the on-site recycling and reusing of treated wastewater for irrigation of edible or non-edible crops, washing vehicles, cleaning toilets, and other domestic tasks will save clean water for drinking and cooking purposes, thus minimising the increased demand for freshwater [28,29,33]. Recycling and reusing greywater can save up to 30% of drinkable water [7,30,33].

2.1. Heavy Metals Pollutants Occurrence in Domestic Wastewater

Heavy metals are metallic non-degradable chemical elements with a relatively high atomic weight and high density, greater than $5 gm/cm^3$. They are toxic and hazardous even at low concentrations [26,36]. The source of heavy metals could be natural, such as the weathering of parental rocks, volcanic, and hydrothermal vents [37,38]. In addition, the anthropogenic sources that emerge from different industries include mining, leather, textiles, plastic, pesticides, and pharmaceuticals, the processing of raw metals, fossil fuels burning, and the discharge of non-treated agricultural and domestic wastewater [37,39,40].

Although some heavy metals are essential for metabolic and biological functions within the human body (i.e., copper, iron, and zinc), they become harmful and toxic when they exceed the standard limits [41,42]. On the other hand, others like arsenic, cadmium, mercury, and lead are non-essential and hazardous elements for the environment as well as for human health, even at low concentrations [43,44]. Heavy metals can cause severe environmental problems [41,44]. Their accumulation in plants and human tissue may cause serious health problems such as renal dysfunction, birth defects, lung damage, cardiovascular disorders, neuronal disorders, and the risk of cancer and diabetes when it exceeds the permissible limits [41,42,45,46]. The most-detected heavy metals in domestic wastewater are Zn, Pb, Hg, Ni, and Cu, which mostly come from detergents used in the kitchen, laundry, and bathrooms [10,31,34,47]. Other household sources of heavy metals are cosmetic products, plumbing pipes, and stainless steel products [48]. The percentage of heavy metals in urban domestic wastewater could reach up to 60% for some metals like Cd [31].

Previous studies showed that some vegetables and forage crops may accumulate heavy metals upon irrigation with domestic wastewater; certain heavy metals such as As, Cd, Cr, Zn, Ni, and Pb were found in high concentrations [13,49,50]. On the other hand, Kim et al. [10] have discovered no harmful impact on human health from consuming vegetables irrigated with effluent from domestic wastewater treatment plants containing small quantities of heavy metals (Pb, Zn, Cd, and Cu).

2.2. Organic Pollutants, N and P Occurrence in Domestic Wastewater

Organic pollutants in domestic wastewater refer to all the decomposition pollutants, including hydrocarbon, washing and cleaning compounds, food waste, and preservatives. The biochemical oxygen demand (BOD) and the chemical oxygen demand (COD) tests are used to determine the organic matter content in wastewater. In addition, the physical method is used to determine the total suspended solids (TSS) [14]. Some of these organics dissolve in the water but others stay as solid particles which are called TSS [14]. The occurrence of organic matter in domestic wastewater can potentially cause diseases, where these organic matter components can be considered as nutrient sources for pathogenic microorganisms, such as bacteria, viruses, protozoa, and parasitic worms, which are capable of causing infection or disease in humans and animals [51]. Moreover, the accumulation of organic matter in the food chain has indirect effects on human health [6,52].

Nitrogen compounds mostly exist in the forms of nitrate, nitrite, or ammonia in domestic wastewater [53]. Ammonia has a poisonous impact on aquatic organisms and human health when it exceeds EPA limits [52]. High levels of nitrate in the human body may cause blue baby syndrome, birth defects, thyroid diseases, and colon cancer [6,52].

Polyphosphates and orthophosphates are the main forms of P in wastewater [6]. Water bodies overly enriched with nutrients can cause the rapid growth of plants and algae i.e., eutrophication, that leads to the death of aquatic animals due to a lack of dissolved oxygen [52].

3. Constructed Wetland Types

There are three main types of CWs based on water flow: surface flow or free water surface flow (FWS), which resembles the natural wetlands; horizontal flow constructed wetland (HFCW); and vertical flow constructed wetland (VFCW). The HCWs system consists of a combination of two or more of the aforementioned types [54] (Figure 2). The differences in flow types of CWs are illustrated in Figure 3.

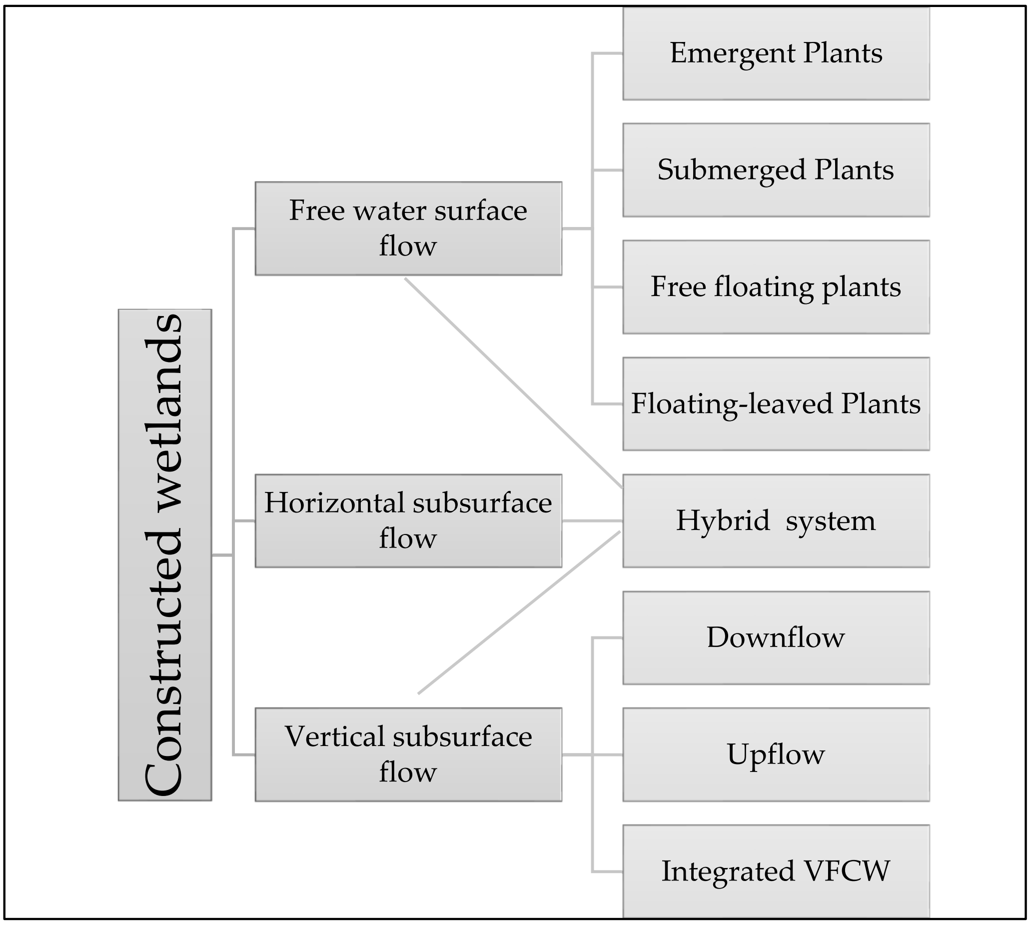

Figure 2. Types of constructed wetlands for wastewater treatment and their interconnections.

3.1. Vertical Flow Constructed Wetlands (VFCWs)

Since their emergence in 1965 [24], VFCWs have become of great interest in research due to their role in enhancing the amount of oxygen that penetrates the filtration media [55]. This available oxygen promotes microbial activity and boosts other removal mechanisms such as organic decomposition and nitrification [56]. The particle size of the filter medium decreases gradually from the bottom to the top, where the small particle size on the top facilitates oxygen-permeability through the media [57].

Generally, three main types of VFCWs have been classified based on water flow direction through the vertical axis by the substrate: down-flow VFCW, up-flow VFCW [58], and the integrated VFCW (IVFCW), where both previous systems are integrated to enhance the pollutants-removal mechanisms [59,60]. Several studies demonstrated the capacity of VFCWs to remove BOD, COD, nutrients (P and N), and TSS from domestic wastewater [56,61–67]. Additionally, VFCWs were tested for treating municipal wastewater to be reused for irrigation purposes [68,69].

Generally, using gravel, soil, and sand as a substrate in the VFCWs is common in these experiments [56,62,64,66,70,71]. In this context, many studies were conducted to find the optimum removal-efficiencies of organic and inorganic pollutants, by trying different types of substrates [56,72,73], plant species [62,64,66], and hydraulic loading rates (HLR) [62]. Meanwhile, another study has investigated the impact of intermittent aeration [71] and the combination of intermittent aeration and biochar on enhanced organic and nitrogen pollutants-removal from domestic wastewater [70].

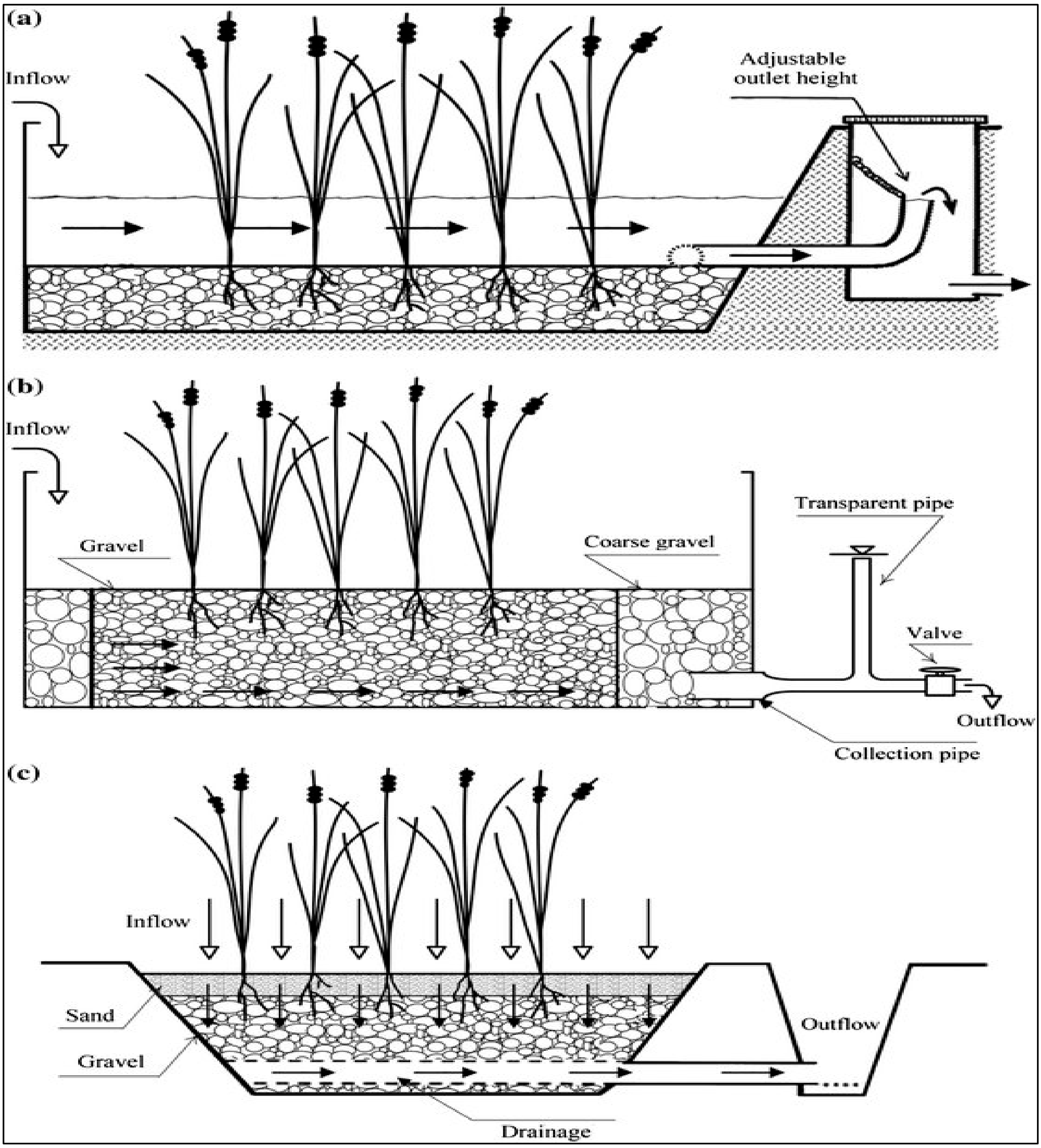

Figure 3. Schematic figures present the types of CWs based on water flow: (**a**) free water surface flow CW; (**b**) horizontal flow CW; and (**c**) vertical flow CW [74]. With permission from authors.

Several studies indicate that VFCW is better than HFCW for overall removal-efficiency [18,75–77]. In contrast, others concluded that HFCWs are more effective for the removal of organic pollutants and total nitrogen [54] while VFCWs are better at ammonium removal [78,79]. The advantages of VFCWs are they need less energy and space area than HFCWs and produce better-quality treated water [21,55,56,76,77,80,81]. However, the cost of operation and maintenance for VFCWs is higher than HFCWs' [55,82].

3.2. Hybrid Constructed Wetland (HCW)

Using the HCWs system combines the advantages of other types of CWs, the VFCW, HFCW, or FWS. Such a combination aims to increase the efficiency of pollutant-removal, especially for total nitrogen [15,82,83] and to reduce the clogging effect of the media [58]. However, these systems are usually used on a large scale and are not suitable for on-site treatment systems [82]. Most of the HCW systems used for domestic wastewater treatment consist of two units, VF followed by HF; the multi-stage systems of HCW are mostly used for industrial and agricultural wastewater, which contain more contaminants than domestic wastewater and demand sophisticated treatment [84].

4. Removal of Organic and Inorganic Pollutants from Domestic Wastewater by VFCW and HCW-Meta-Analysis

4.1. Data Collection and Analysis

Among twenty-one published articles investigating the removal-efficiency for organic and inorganic pollutants from domestic wastewater of VFCW and HCW techniques (Supplementary Tables S1 and S2), nearly half of them can provide the information needed for meta-analysis. A random-effect model was used to calculate the mean effect size; therefore, the mean, sample number, and standard error or standard deviation were collected from eight articles and fit on JASP 0.16.3.0 (Created by Eric-Jan Wagenmakers, University of Amsterdam, Amsterdam, Netherlands) by using the Restricted ML method, $p < 0.05$.

The average removal-efficiency from the twenty-one studies was analysed by variance test (ANOVA), in order to reveal the effects of the method, plant and substrate (and their interaction) on the pollutant's removal-efficiency. Data from more than two replicates were conducted in the analysis as study units. Microsoft EXCEL 365 was used for bar chart graphics.

4.2. Results and Discussion

4.2.1. The Effect of the Interaction between Method, Plant, and Substrate on the Pollutant's Removal-Efficiency

The meta-analysis of the pollutant's removal-efficiency from domestic wastewater using both VFCW and HCW methods, and different substrates and plants, shows a high significance ($p < 0.05$) in the interactions (among method, plant, and substrate) on pollutant removal efficiency (Figure 4).

Within the same method and using the same plant, the effect of applying different substrates is evident. Wu et al. [71] show higher recovery for COD, NH_4, and TN than the results obtained from the work of Korkusuz et al. [73]. In addition, a slight increase in the removal-efficiency of the organic pollutants is observed by adding one more layer to the filtration media [73]. Moreover, there are no significant differences between the two substrate variables used in both TSS and TN. However, a high significance appears for ammonium and total phosphorus removal, mostly due to the increase in the filtration-media surface area, which plays a critical role in the adsorption mechanism. On the other hand, the increase in the removal percentage is obvious when compared to the work of Wu et al. [71] for all pollutants, despite the fact that they used the same plant and only one filtration media (gravel); this increase mostly refers to the intermittent aeration technique which they used.

Both CW established by Korkusuz et al. [73] and Ye and Li [85] are planted with *Phragmites australis*; however, the vertical flow with three substrate layers (gravel, slag, and sand) was applied by Korkusuz et al. [73], while Ye and Li [85] have used gravel and soil as a substrate in their HCW system. Even though the NH_4 removal percentage shows no significant difference between the two systems, the removal percentage for TN increases approximately to double in the hybrid system; these results mostly refer to the advantage of the hybrid system in dealing with nitrite with the multi-system, while in the VFCW the removal of total nitrogen is low because of the low viability of the denitrification process [86,87]. Moreover, the COD and TSS parameters show highly significant differences, with advantages for the hybrid system, while the TP is mostly the same.

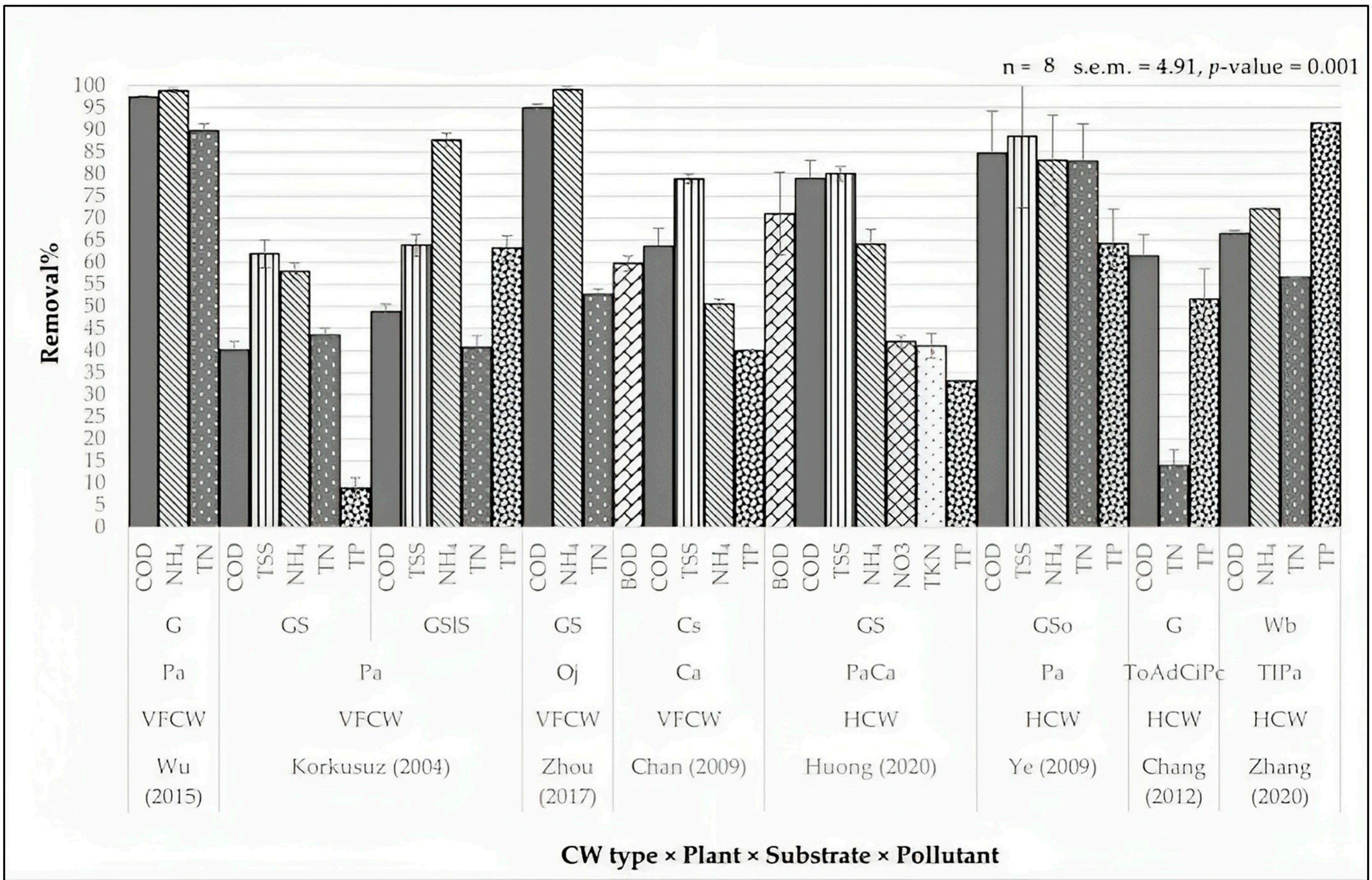

Figure 4. The effect of the used plants, substrates, and their interaction when applying VFCW and HCW to treat domestic wastewater on the average removal percentage for different types of pollutants. [Where, V; Vertical Constructed wetland, H: Hybrid Constructed wetland, Pa; *Phragmites australis*, Oj; *Oenanthe javanica*, Ca; *Cyperus alternifolius*, To; *Typha orientalis*, Ad; *Arundo donax* var. *versicolor*, Ci; *Canna indica*, Pc; *Pontederia cordata*, Tl; *Typha latifolia*, G; Gravel, S; Sand, Sl; Slag, Cs; Coal Slag, So; Soil, Wb; Waste bricks, COD; Chemical Oxygen Demand, TN; Total Nitrogen, TSS; Total suspended solids TP; Total Phosphorus, BOD; Biochemical Oxygen Demand, TKN; Total Kjeldahl Nitrogen, n; the number of studies, s.e.m.; Standard Error of the Analysis, Wu (2015); [71], Korkusuz (2004); [73], Zhou (2017); [70], Chan (2009); [88], Huong (2020); [15], Ye (2009); [85], Chang (2012); [65], Zhang (2020); [89]].

By applying the VFCW technique and using gravel and sand as filtration media, Zhou et al. [70] reveal an apparent increase in removal efficiency for COD, ammonium-N, and TN over the results of Korkusuz et al. [73], mostly referring to the used plant species which differentiates between the two studies. On the other hand, another study applied HCW with the same plant and substrate [15], and the hybrid system showed more efficiency in removing all pollutants than the VFCW system; these results may be explained by the multi-layers with different particle sizes which have been used in the hybrid system and which enhance the removal mechanisms; in addition, they use an earthworm which enhances the biological mechanisms for organic pollutants [15]. In contrast, the vertical method with the intermittent aeration technique in the work of Wu et al. [71] shows more efficiency than the towery hybrid system applied by Ye and Li [85] even though both studies have the same plant (*Phragmites australis*) and gravel as a substrate.

Using industrial byproducts as substrates in CWs rather than conventional substrates is a new technique. As shown in Figure 4, two studies have used different types of byproducts [88,89], where both systems were planted with different species of plants. For all pollutants, the hybrid system shows more efficiency in removal than the vertical one except for COD, where there is no significant difference between the two studies.

In addition to the effect of the used substrate and plant, other environmental parameters have an impact on the efficiency of the CW system. We talk in detail about these parameters in Section 6.

4.2.2. The Efficacy of the Method Used on Pollutant Removal

Regardless of the type of used media, plant species, and any other environmental parameters, a comparison between applying vertical and hybrid systems for domestic wastewater treatment on the removal percentage average for different pollutants is illustrated in Figure 5. The analysis shows no difference ($p > 0.05$) between applying VFCW or HCW on removal percentages for BOD, COD, NH_4, TN, TP, and Cu. TSS shows a slightly significant difference with an advantage for the hybrid system. On the other hand, when applying the same technique, there are highly significant differences between the remediation of NH_4 and TN removal, which mostly refer to the increase of dissolved oxygen VFCW which influences the nitrification process (converting ammonium to nitrate). In contrast, the denitrification mechanism has a low chance of taking place, given the availability of oxygen.

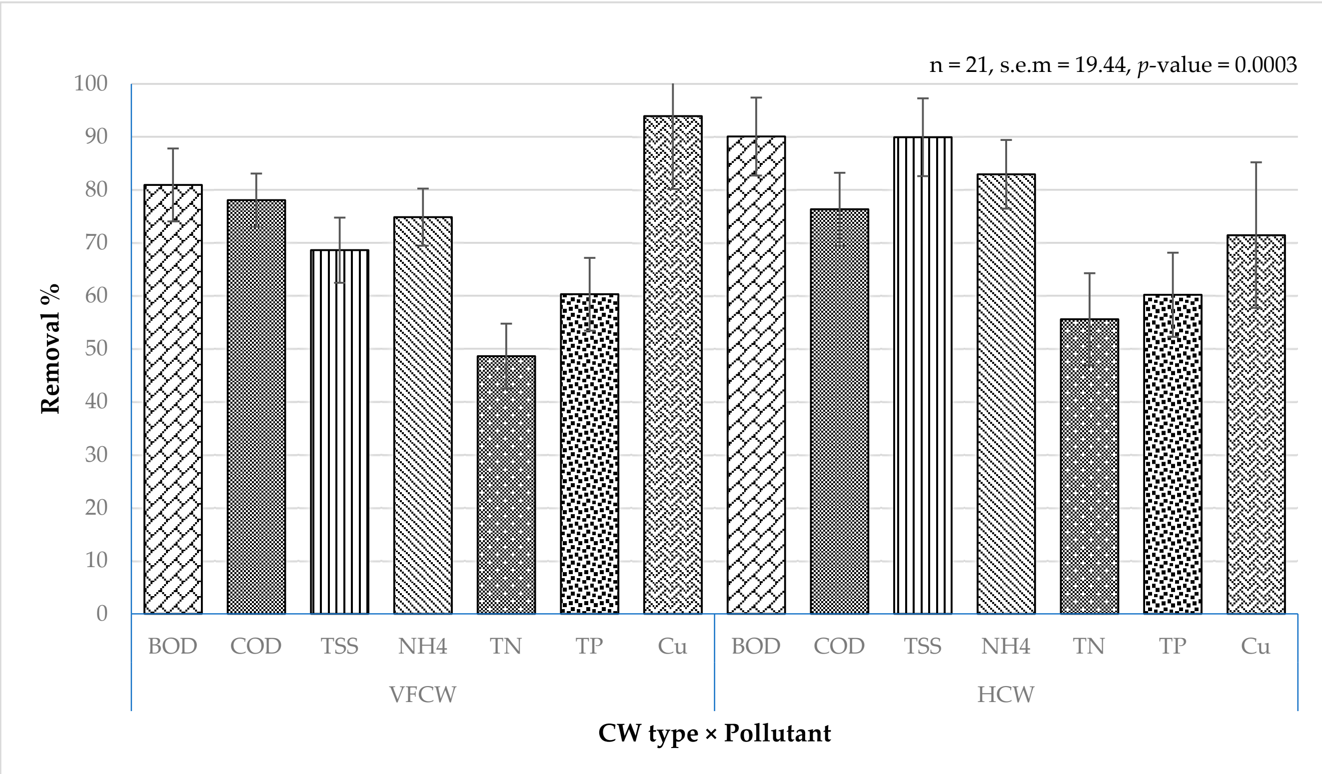

Figure 5. The efficacy of CW type for domestic wastewater treatment on the average removal percentage for different types of pollutants. [Where, BOD; Biochemical Oxygen Demand, COD; Chemical Oxygen Demand, TSS; Total suspended solids, TN; Total Nitrogen, TP; Total Phosphorus, n; number of studies, s.e.m.; Standard Error of the Analysis].

The removal average percentage for organic pollutants (COD and BOD) from domestic wastewater by any of the two systems is usually more than 80%. By contrast, in VFCW the removal average for ammonium nitrogen and TN is 75 and 47%, respectively. On the other hand, the percentage is slightly higher for TN removal in the HCW system.

4.2.3. The Efficient Removal of Pollutants by Different Plant Species Used in Two Different CWs

Phytoremediation is an environmentally friendly and cost-effective technique through VFCW; it involves using plants to remove both organic and inorganic pollutants from contaminated soil or water by absorbing them through their roots [77,90]. It is important to note that the specific plant species used in CW systems vary, depending on the location, natural environment, and specific treatment goals [77,90]. However, the most used macrophyte in VFCWs is the common reed (*Phragmites australis*), which adapts to various environmental conditions, and which is planted alone in some systems [71,73,91], or with other plants [66,68]. Other commonly used plants include yellow iris (*Iris pseudacorus*) [64,86], umbrella sedge (*Cyperus alternifolius*) [62,88], rush (*Juncus* sp.), and bulrush (*Scirpus* sp.) [90].

The effect of using various plant species on the pollutant-removal percentage from domestic wastewater is illustrated in Figure 6. It's obvious that the unplanted system possesses the minimum removal percentage. A system that contains over 10 plant species including *Carex riparia*, *Phragmites australis*, *Typha latifolia* L., *Iris pseudacorus* L., etc., shows the optimum efficiency. This efficiency corresponds to the enhancement of each kind of plant in the removal mechanism (plant uptake) and supports the whole system [86]. In contrast, a system with four plant species reveals the worst performance among planted systems due to the very high HLR (250 mm/d) [65].

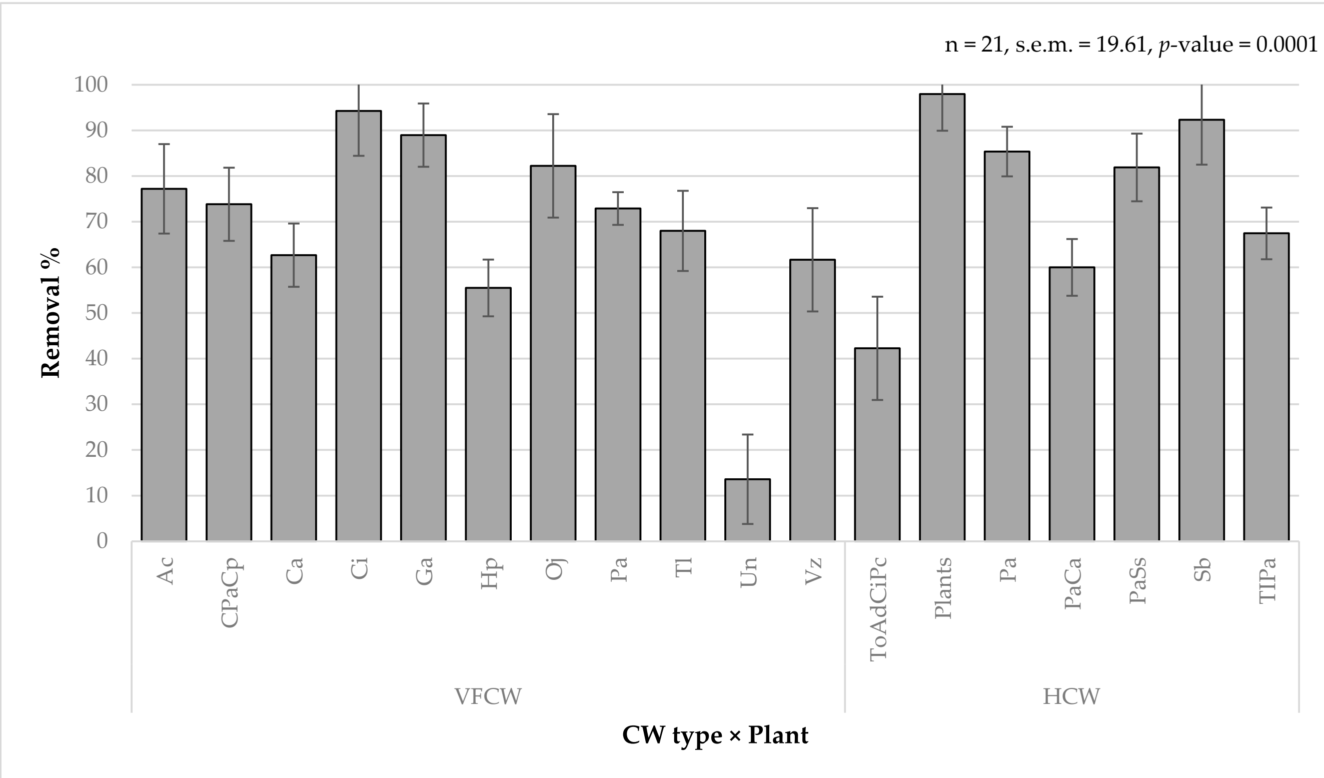

Figure 6. The efficacy of CW type in the removal percentage of pollutants from domestic wastewater based on the used plant species. [Where, Ac; *Acorus calamus*, C; *Canna*, Pa; *Phragmites australis*, Cp; *Cyprus papyrus*, Ca; *Cyperus alternifolius*, Ci; *Canna indica*, Ga; *Glyceria aquatica* L., Hp; *Heliconia psittacorum*, Oj; *Oenanthe javanica*, Tl; *Typha latifolia*, Un; Unplanted, Vz; *Vetiveria zizanioides*, To; *Typha orientalis*, Ad; *Arundo donax* var. *versicolor*, Pc; *Pontederia cordata*, Ss; *Scirpus* sp., Sb; *Salix babylonica*, n; number of studies, s.e.m.; Standard Error of the Analysis].

The efficiency of pollutant-removal by plants is mostly dependent on the capacity of the plant to uptake and tolerate a maximum amount of different pollutants within its tissues. In addition, it depends on the strength of the plant's root and its distribution within the media [68]. Moreover, other environmental parameters in each system including the dissolved oxygen, temperature, pH, HLR, and HRT are determinant factors [65]. *Phragmites australis*, which is generally the most used-plant in CW, shows higher performance in hybrid than vertical, which is mostly related to the multi-systems in hybrid.

4.2.4. The Efficient Removal of Pollutants by Different Substrates Used in Two Different CWs

The effect of substrate type on the pollutant's removal-efficiency from domestic wastewater is illustrated in Figure 7. In general, the conventional filtration media, i.e., gravel, sand, soil, or a mixture of them, are mostly used in CW systems due to their availability and low cost. The removal percentage average by CW based on the conventional substrate is higher than 65%, while other natural substances such as zeolite, wood mulch, and crushed rock raise the percentage up to ~87% [87,91,92]. However, we have to take into account other environmental conditions and the availability of substrate.

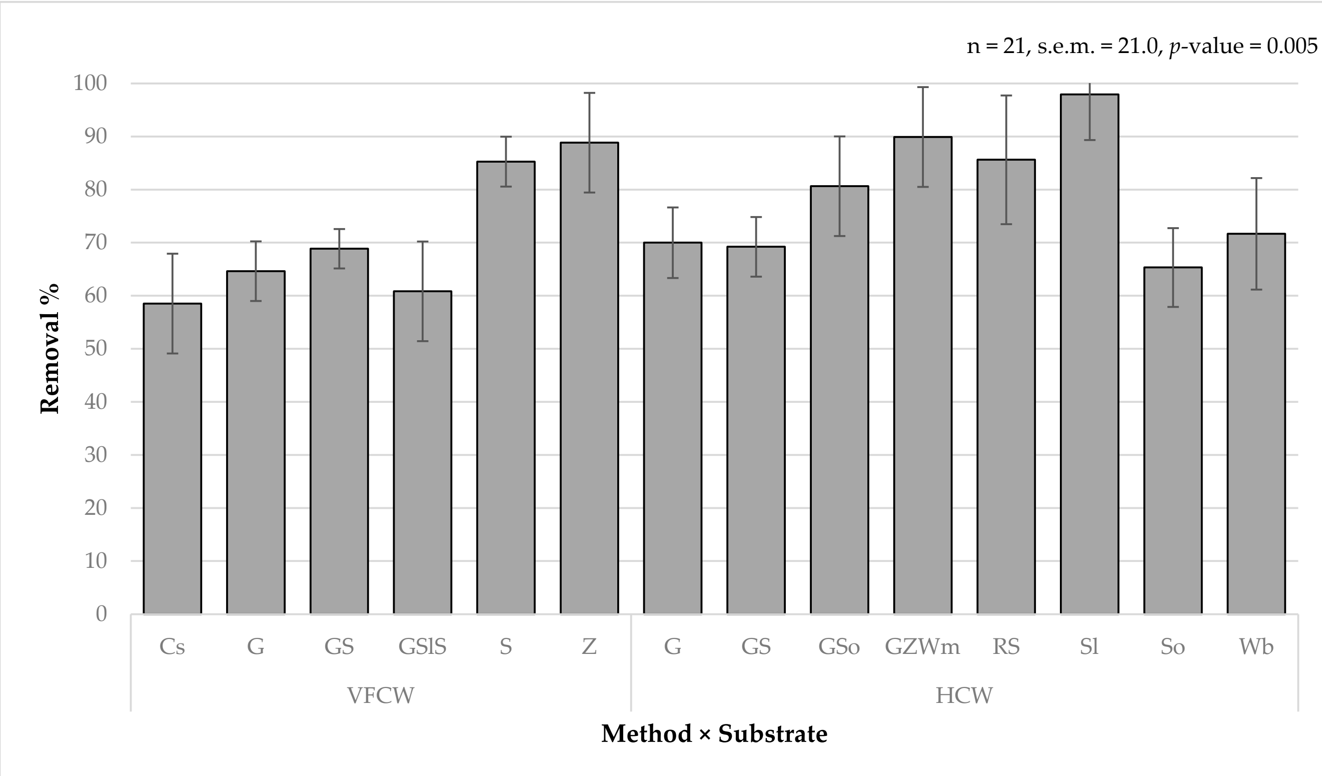

Figure 7. The efficacy of CW type in the removal percentage of pollutants from domestic wastewater based on the used substrates. [Where, Cs; Coal Slag, G; Gravel, S; Sand, Sl; Slag, Z; Zeolite, So: Soil, Wm; Wood mulch, R; crushed Rock, Wb; Waste bricks, n; the number of studies, s.e.m.; Standard Error of the Analysis].

On the other hand, using industrial byproducts shows variability in the efficiency, with high efficiency for slag [86]. However, other byproducts like waste bricks [89] and coal slag [88] show no significant differences when compared to conventional substrates.

High removal percentages for organic matter (BOD, COD, and TSS) from domestic wastewater have been achieved by applying VFCW systems [66]. Moreover, the system has achieved a high performance with the biological contamination represented in two strains of bacteria (*coliform* bacteria and faecal *coliform* bacteria).

5. Mechanisms Involved in Pollutant Removal from Domestic Wastewater Using VFCWs Technology

There are three pathways to removing pollutants in CWs: physical, chemical, and biological. Physical mechanisms involve sedimentation, filtration, and adsorption. The chemical mechanisms include precipitation, adsorption, decomposition (UV irradiation, oxidation-reduction), volatilisation for NH_3, and ion exchange. The removal via biological processes is achieved through plant uptake, degradation of organic pollutants by microorganisms, bacteria metabolisms (nitrification/denitrification), and plant absorption [93].

The process of removing heavy metals using CWs involves filtration, sedimentation, soil adsorption, oxidation/reduction, ion exchange, precipitation, complexation, and plant uptake [20,94]. However, adsorption by the substrate [58,95] and precipitation as insoluble salt [96,97] are considered the main mechanisms for removing heavy metals in CWs [58]. Adsorption is a physical mechanism that occurs by binding metal ions to special sites on the surfaces of the media or sediment particles by electrostatic interaction, i.e., van der Waals forces, removing them from the wastewater [93,98]. The adsorption capacity of the media depends on their composition and surface characteristics [95]. Some factors affect the efficiency of adsorption, such as temperature and pH [95,98]. On the other hand, heavy metals in wastewater can be eliminated by precipitating them (mainly sulphides and oxyhydroxides) [98,99]. This occurs as the pH of the wastewater increases due to microbial activity and chemical processes within the CW [98]. The precipitated metals then settle out and become trapped in the CW's media [100]. Therefore, it's important to choose suitable media to remove the target heavy metals [20,101]. Moreover, the selection of appropriate filtration media is essential for providing high hydraulic permeability. The optimum adsorption capacity of the different pollutants and suitable conditions are important for optimum vegetation growth [1,102]. The cation exchange is a chemisorption process through chemical interaction between the positive ion of metal and the negatively charged site on the substrate [22]. Although the removal of heavy metals by plant uptake is considered a small fraction of the overall heavy metals removal in VFCW, the presence of emergent plants in the CW system is essential for reducing the dredging soil depth for final heavy metal removal [95,100]. However, the capacity of the plants to uptake heavy metals depends on the plant species and the type of metal [83,103]. For example, it has been found that water hyacinth (*Eichhornia crassipes*) has the ability to uptake Hg with an efficiency of up to 95% [104], and bioaccumulation for Cu, Zn, Pb, and Cd with different percentages [105], while duckweed could remove the Cd [106]. It has been found that the sedimentation process of the substrate is affected by the presence of vegetation [96]. Microorganisms play an important role in the removal of heavy metals in VCWs. They can transform toxic metals into less harmful forms through processes such as microbial reduction, volatilisation, and complexation [98,100].

The mechanisms that work on removing organic pollutants from wastewater using VFCWs are manifold and include sedimentation, microbial degradation, volatilisation, oxidation, sorption, photochemical and bioaugmentation of the sediment, and absorption by plants [107]. It has been found that the removal of most TSS (up to 75%) happens in the first layers of CWs, through sedimentation, filtration, aggregation, adsorption of the filter media, and vegetation roots. The removal of organic compounds in CWs is accomplished through degradation by microorganisms whether in aerobic or anaerobic conditions indicated by a decline in the values of COD and BOD parameters [93,108]. COD and BOD removal in VFCW is achieved through a combination of physical, chemical, and biological processes. Physical processes such as sedimentation and filtration help in the removal of suspended solids and particulate organic matter [109,110]. The chemical mechanisms are mainly represented in adsorption, chelation, and precipitation. Adsorption involves the attachment of organic molecules to the surfaces of substrate or plant roots; this process helps to trap and remove the organic pollutants from wastewater [108]. Chelation is another chemical process that contributes to the removal of organic pollutants and metals. It involves the formation of complexes between organic molecules and metal ions; these complexes are relatively stable and can be easily removed from wastewater [5]. Organic matter is subjected to biodegradation by microorganisms present in wetlands. These microorganisms play a vital role in the breakdown and conversion of organic matter, resulting in its removal from the water [110,111]. The efficiency of removing organic pollutants in constructed wetlands can be affected by several parameters, including hydraulic conductivity, the size of the surface area of the substrate, vegetation type, and operating conditions [93,99,112]. The presence of vegetation, particularly macrophytes, is essential for enhancing the removal of organic pollutants by providing a substrate for microbial growth and facilitating oxygen transfer through their root systems [113]. On the other hand, it has been reviewed that the plant uptake mechanism has a negligible effect on the organic pollutants removal process [93]. The amount of decomposing organic matter depends on several factors including temperature and dissolved oxygen [99].

Concerning removing nutrients via VFCWs, adsorption and precipitation are considered the major mechanisms. Therefore, the substrate plays the main role in the nitrogen and phosphorous removal process [114]. Ammonia, nitrate and nitrite are the most common forms of inorganic nitrogen dealt with in wetland systems. There are various mechanisms to remove nitrogen from water using VFCW, such as volatilisation, nitrification, denitrification, ammonification, fixation, plant uptake, microbial decomposition, and ammonia adsorption [57]. Due to the converting process of ammonia-nitrogen into nitrite-nitrogen and then nitrate through the VFCW systems, the removal percentage of ammonia is usually high [53]. The nitrification process is influenced by major factors such as the presence of denitrifiers, moisture, dissolved oxygen, pH, and temperature [53,99]. However, the removal of total nitrogen is low because of the low success of the denitrification process, which is usually lower than in HFCWs [57,86,87]. Therefore, the denitrification mechanism is common in HFCWs. Therefore, the HCW system is considered the best for removing the maximum percentage of total nitrogen [53,112]. Meanwhile, VFCW provides an effective solution for the removal of phosphorus from wastewater [54]. In general, the efficiency of phosphorus-removal in these systems is mostly dependent on several factors such as the nature of the substrate, environmental conditions, and plant species [53,57,92]. The major mechanisms for phosphorus-removal are natural sedimentation, precipitation, adsorption, and uptake by plants [108,112,115]. The precipitation process involves the conversion of dissolved phosphorus into a solid form that can settle to the bottom of the wetland, being later incorporated into sediment. While adsorption plays a crucial role in phosphorus-removal, in the context of a VFCW system, phosphorus in wastewater can attach to the surfaces of substrate particles or plant roots, thereby reducing phosphorus concentrations in the treated water [53,115].

6. Effects of Environmental Parameters on the Removal of Pollutants Using VFCWs

Several operational and environmental factors affect the effectiveness of removing mechanisms for different pollutants when using VFCWs. These factors can be chemical, physical, or biological parameters. They include but are not limited to pH, temperature, hydraulic loading rates (HLR), hydraulic retention time (HRT), dissolved oxygen, type of filtration media, presence of vegetation, and type of vegetation [102,116].

The treatment process of heavy metals using CWs is affected by several factors; among these are the chemical pH [5,39,117], heavy metals' initial concentration [118], and concentration of dissolved oxygen [39]. On the other hand, numerous physical parameters affect decontamination processes, such as temperature [119], the type and design of CWs [97], HLR, HRT [118,120], recirculation of effluent [83], bed depth [118,121], and layers of the substrate [15]. In addition, biological factors include microbial activity [39,80,96], the presence of vegetation [94], and plant species [121–123]. The most effective parameter is the pH [5] and the most recommended range for pH is 5–7 for most of the metals removed [124]. The aforementioned situation is due to the charge of most metals being positive (cations). Therefore, at lower pH values, the media will contain a higher concentration of H+ which will compete with metals for the adsorption sites of the substrate and/or soil particles (clay) which are entirely negatively charged. However, at high pH values (5–7), the metals will be adsorbed more effectively since the concentration of free H+ will be less than at lower pH (4–6). [22]

The amount of dissolved oxygen affects some chemical mechanisms of oxidation/reduction reactions as well as microbial activities [39]. An increased temperature will enhance the growth of microorganisms. Correspondingly, the increased dissolved oxygen will be urgently in demand for better microbial growth and biofilm performance. Under these conditions, the microorganisms will be very effective in decomposing organic matter and nitrification of inorganic nitrogen—indeed, in a positive effect on increasing the adsorption process [22]. This is assessed by an easier flow of dissolved oxygen into the media through a vertical flow [53]. The HRT has a direct effect on removing-efficiency for BOD, COD, TN, and TP [5]. There is no perfect HRT for all metals; mainly, it depends on the type of substrate and the target metal [122]. As the retention-time increases, the removal-efficiency will increase too until a certain point where the substrate becomes saturated. The initial concentration of the heavy metal is considered another important parameter; as the heavy metal's initial concentration increases, the removal percentage decreases, mostly because the number of metallic ions increases but the number of capturing sites within the substrate and plant does not change [39,118].

The presence of the plant in the VFCW plays a major role in pollutant-removal. In this context, rhizomes and roots help prevent clogging, stabilize the surface of the substrate, and positively influence hydraulic conductivity and microorganism community [54]. In addition to the effect of vegetation, some more parameters have effects on the efficiency of the plant's performance in pollutant-removal, including the type of media, contact time, water load, pollutant concentration, and other environmental parameters [5].

Substrates Used in VFCW

A suitable media can lead to maximising the heavy metals removal capacity. Commonly, the chosen substrate must have high porosity to minimise the clogging problem and enhance the removal process [81]. At the same time, it is preferable if it is cheap and available [102]. The substrate can be divided into three main types: natural products, industrial byproducts, and man-made materials [125].

Natural materials such as gravel, soil, and sand are commonly used as CW substrates because of their low cost and high availability [97,123,126,127]. More recently, industrial byproducts and recycled materials have been used as CW media. These materials include rubber tyre chips, steel slag, fly ash, and brick [83,89,128,129]. To remove heavy metals from polluted water, industrial and agricultural byproducts have been used, because of their abundance, low cost, and being environmentally friendly as an effective solution for safe disposal [22,58,83]. Chemical (e.g., acidic modified, alkaline modified, oxidation agents, and organic compounds) [58,130] or physical (e.g., pyrolysis and grinding) modifications [131] for the waste materials before applying as an adsorbent can minimize their hazardous impact on the environment and enhance their adsorption capacity by increasing the surface area [22,58]. The media with chemical modification is considered the most effective type [22].

In VFCWs, using industrial and agricultural waste as a substrate to maximise the capture of heavy metals from diverse types of polluted water is evident [132]. In this regard, some plant waste used as filtration media in CWs, such as cattails leaves, oil palm shells and cocopeat, has the capacity to adsorb specific kinds of heavy metals with different percentages [132]. Correspondingly, Saeed et al. [83] have indicated the removal capacity for four heavy metals from landfill leachate by using an HCW system consisting of VF-HF, where the VF is based on a coconut waste called cocopeat, followed by HF packed with sand. In addition, the system was operated under 50% recirculation. The results reveal high removal efficiency for Zn, Cr, Ni, and Pb at 80, 100, 56, and 97%, respectively. Some CWs consist of more than one substrate layer with varied materials or different granular sizes, where the size increases from top to bottom and increases towards the bottom [80,83,101].

Clogging Problem Facing CWs

Clogging is considered a natural process occurring within filtration media in CWs [133] and is the major challenge facing the long-lasting effectiveness of CWs [102]. It is an operational and maintenance problem facing the effectiveness and validity of CWs by causing a shrinking in the porosity of the filtration beds. The latter leads to a system with poor hydraulic conductivity [134]. The clogging process is caused and affected by several parameters, including the type and particle size of the substrate, the accumulation of suspended solids, the microbial community, and the growth and distribution of the plant's roots within the filtration media [134]. While some authors having concluded that suspended solids are the major reason for clogging [134], others have found that microbial activities are the main factor causing clogging.

Wu et al. [1] have reported that surface flow CWs have a relatively long lifetime compared to subsurface CWs due to the clogging in the latter. As time goes on, the clogging problem in CWs gradually increases, leading to a decrease in the hydraulic conductivity of the media, and a decrease in the adsorption efficiency of the substrate. Some applied solutions could delay the clogging and reduce its consequences in VFCWs, such as biological ones, i.e., earthworms [133]. Moreover, other suggestions include physical treatments [133,134].

7. Conclusions

VFCWs and HCWs are considered sustainable, effective techniques for a safe, economical, and environmentally friendly method of treating domestic wastewater. In this review article, a meta-analysis of the efficiency of using VFCWs and HCWs for domestic wastewater treatment reveals highly significant ($p < 0.05$) interactions among methods, plants, and substrate on the pollutant's removal efficiency. ANOVA tests show the effect of the kind of substrate, and the presence of the plant and its species, on the pollutant's removal-efficiency. Unconventional substrates show more efficiency in the removal process than conventional ones. On the other hand, planted systems are more efficient in the removal efficiency for all pollutants than unplanted ones. However, ANOVA analysis shows no significant difference ($p > 0.05$) between applying VFCWs or HCWs on removal percentages for most pollutants.

There are only a few studies that discuss the potentiality of both VFCWs and HCW systems in removing heavy metals from domestic wastewater. The authors recommend the conduct of more studies for a full-scale system and a non-planted CW system.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15193348/s1, Table S1. Removal-efficiency of organic and nutrients from domestic wastewater by using VFCWs. Table S2. HCW and their removal-efficiency for different pollutants from domestic wastewater. References [135–137] are cited in the Supplementary Materials.

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