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Application of Circular Economy in Oil and Gas Produced Water Treatment

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Abstract: The circular economy (CE) is a promising model in industrial waste management, offering viable long-term resource sustainability. The rising costs of the oil and gas industry make circularity a reliable approach for saving materials, money, and energy. In recent years, attention has risen to the need to apply CE within oil and gas produced water (PW) treatment. The most common treatment practice for PW is based on mechanical treatment, with optional disposal of treated water into deep wells. However, this procedure consumes a lot of energy, increases operational costs, and causes environmental risks. This research aims to propose sustainable treatment technology promoting circularity by introducing a novel nature-based solution to treat PW. The main research objective is to develop a circular model for PW treatment by investigating the treatment of PW using constructed wetlands (CWs) to sustainably reduce the amount of waste in oil and gas fields. Additionally, investigate the use of industrial wastes as filtration materials for CW systems. In this study, eight different laboratory-scale CWs models were designed and tested. The CWS operated in two different types of flow directions: vertical (VF) and horizontal flow (HF). The main filter media for the CW system included aggregates, activated carbons, plastic, and shredded tires. The study investigated the removal rates of Total suspended solids (TSS), Total dissolved solids (TDS), Oil and Grease (OG), and Total Petroleum Hydrocarbon (TPH) from the PW. Testing the CWs, it was found that the results of the PW treatment were promising, with the potential for more future shredded tires and plastic applications. All systems were effective at removing contaminants from produced water, with the highest recorded removal efficiencies of 94.8% TSS, 33.7% TDS, 90.2% OG, and 98.4% TPH. The research results were efficient and promoted the circular use of CW in PW treatment in addition to the possibility of reusing the treated effluent in agriculture and irrigation.

Keywords: produced water treatment; circular economy; sustainability; nature-based solutions; constructed wetland; industrial waste management

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1. Introduction

The linear economic model of take-make-dispose is a historical economic approach that has been used in various industrial sectors since the industrial revolution, promoting economic benefits over all other measures [1]. The literature revealed that this model caused major environmental problems, including the depletion of natural resources, the accumulation of waste, and climate change [2]. However, to avoid such consequences, initiatives to develop alternative economic models emerged, introducing the circular economy (CE) [3]. CE is a sustainable management approach proposed in the 1960s with the potential to reduce waste generation and mediate global warming [2]. CE is based on keeping the used materials in the economy as long as possible by ensuring that the resulting wastes of one process are not discarded directly but are restored or regenerated to become resources for other applications [4–6]. Hence, in contradiction to the linear economy, in a circular economy, waste does not exist and is considered a new raw material, as shown in Figure 1 [7].

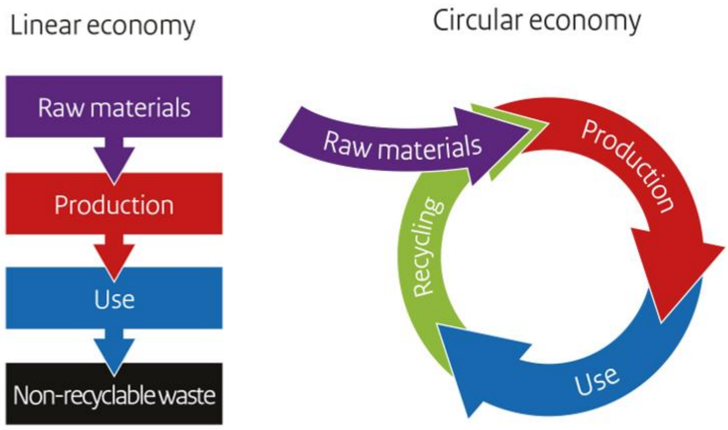


Figure 1. Linear versus circular economy flow charts [7].

Water plays a significant role in achieving CE, where it is a major resource in industrial activities [8,9]. Additionally, the resulting wastewater that has been contaminated during industrial use can, fortunately, be recycled and safely used after a variety of treatment processes, including biological, physical, chemical, or a combination [10,11]. The appropriate treatment method is determined based on the source of wastewater, contaminants constituents, and regulating standards [12,13]. Currently, there is a challenge to promote sustainable treatment solutions that save energy at a lower cost while efficiently treating industrial wastewater [14]. In the oil and gas industry, produced water (PW) is the water associated with oil during the extraction process [15]. It is one of the largest streams of wastewater generated, and its estimated global value reached 250 million barrels per day in 2020 [16], resulting in an annual estimate for 1999 of 77 billion barrels of produced water [17]. By volume, it is therefore the largest waste stream associated with oilfield activities. Thus, CE application and PW proper management are imperative in the oil and gas industry to avoid the risk of environmental degradation and damaging ecosystems.

1.1. Produced Water Composition and Treatment

PW originates from two main sources: the first is during the extraction of oil, which usually provides a mixture of oil and water that comes from the seawater surrounding the oil well [18]. The second involves water being injected into the oilfield to bring the deep oil to the surface, and it ultimately becomes part of the PW, or wastewater [19]. Subsequently, produced water can be classified as a substance derived from natural gas, oilfields, or coal bed methane [20]. The environmental impact of dumping produced water is harmful and a huge waste of valuable resources. PW is not a single product but one that ranges from a simple to a complex composition that is variable and is deemed to be a mixture of dissolved and particulate organic and inorganic chemicals [21]. Chemical and physical properties of produced water vary considerably depending on several factors, such as the location of the field; age and depth of the geological formation; hydrocarbon-bearing formation geochemistry; extraction method, the type of the produced hydrocarbon; and the chemical composition of the reservoir [16].

The toxicity of PW discharged from gas platforms is 10 times more dangerous than the toxicity of oil wells' discharge [22,23]. However, the amounts of oil that are produced are much larger than gas production. Generally, the major constituents that are present in produced water include salt (measured as salinity), total dissolved solids (TDS), oil and grease (OG), total petroleum hydrocarbons (TPH), benzene, toluene, ethylbenzene, and xylenes (BTEX), phenols, organic acids, natural organic and inorganic compounds that cause hardness and scaling (e.g., calcium, magnesium, sulfates, and barium); and chemical additives such as biocides and corrosion inhibitors employed during drilling, fracturing and operation of the well [16,24–26].

Treatment of PW can be done through various methods, including physical (membrane filtration, adsorption, etc.), chemical (precipitation, oxidation), and biological (activated sludge, biological aerated filters, and others) methods. Since PW contains several different contaminants with varying concentrations, numerous treatment technologies have been devised for water treatment [17,22]. Subsequently, it is challenging to choose the type of treatment system best able to remove most of the contaminants from produced water. Generally, the cheapest method is the most preferable, and the cost of the produced water treatment mainly depends on influent quality, the price of electricity, a plant's capacity, and the effluent's desired quality [27,28].

1.2. Promoting Constructed Wetlands for Produced Water Treatment

Of the wide variety of PW treatment methods, filtration is a relatively simple technique used in water and wastewater treatment processes, and it is based on employing porous filter media to allow only the water and not the impurities to pass through them. Various porous materials can serve as filter media, for instance, sand, crushed stone, and activated carbon [29–32]. However, the most widely used material is sand due to its availability, low cost, and efficiency. Hence, Constructed Wetlands (CWs) present an ideal, sustainable, nature-based solution for PW treatment and reuse. CW uses gravitational flow and filtration for wastewater treatment, simulating the naturally created wetlands [33–36]. CWs are generally classified based on the presence or absence of water on the surface as free surface flow or subsurface flow and based on flow direction as vertical (VF CW), horizontal (HF CW), or hybrid (VF + HF CWs) [35,37–39]. Selecting the appropriate CW flow should be according to the wastewater type (industrial or domestic) and the local economic and environmental conditions, as shown in Figure 2 [40,41]. Vymazal [42,43] reported that the application of the constructed wetlands CWS technology in various industrial wastewater treatments has been tested since 1975. Stefanakis [35,44,45] confirmed that CWs present a promising CE with proven high efficiency in the treatment of PW in addition to the reduction of the energy consumed through the treatment process by 99% compared to the conventional deep-well disposal site approach.

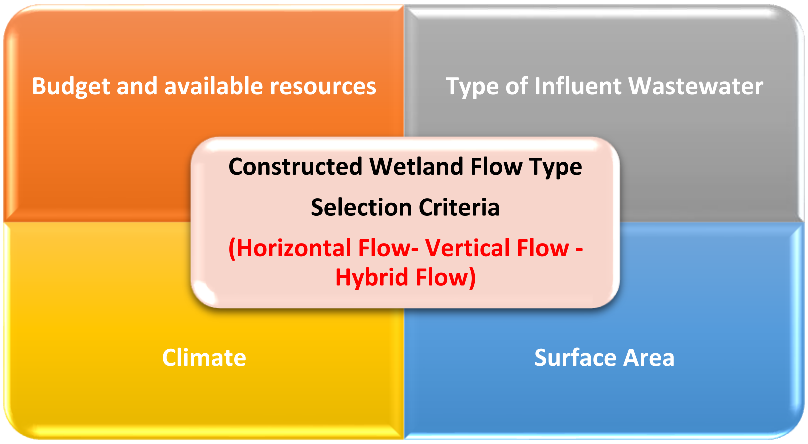


Figure 2. Types of constructed wetlands based on wastewater flow and selection criteria (developed by Vymazal [40]).

In this research, a novel CE proposal for PW is presented by designing and testing eight lab-scale CWs using new filter materials that can be found in the oil and gas fields as wastes like plastic and shredded tires to ensure circularity of both solid and water wastes, besides using activated carbon and aggregates due to their known water treatment capabilities as the control for the rest of the materials. Filtration materials play an important role in the subsurface flow of a constructed wetland [46,47]. The choice of filtration materials is crucial for hydraulic conductivity and the removal of suspended solids and phosphorus [48]. Vymazal [47] summarized the different nonconventional filtration materials used in several pieces of research that investigated the use of different filter media materials in CW, such as rice husk, waste bricks, shredded tires, alum slag, oyster shell, and plastic pipes as well as crushed rock and gravel. Hence, implementing the proposed CE approach in the treatment of PW using CWs with a novel filter medium can help facilitate additional options for petroleum industry water management, including its reuse for agriculture and industry.

1.3. Study Objectives

The main aim of this study is to develop a circular model for PW treatment by investigating the treatment of PW using CWs with the application of waste filtration materials. To achieve this aim, the specific objectives of this research work are listed as follows:

1. Preliminary testing and determining the optimum configuration for the lab scale CWs;
2. Design and installation of a pre-treatment septic tank to decrease the percentage of oil and grease in the effluent;
3. Design and fabrication of eight individual VF and HF CWs units to test four different filter media materials;
4. Chemical analysis of the produced water effluent collected from the Kuwait oil fields;
5. Operation and monitoring of the CWs using main filter media from aggregates, activated carbon, plastic, and shredded tires. The lab-scale wetlands were operated unplanted first, then *Bamboo* plants were added to the systems, and finally, *Bamboo* was replaced with *Cyperus* plants to test the efficiency of the different materials with and without plants;
6. Examine the effectiveness of CWs for the treatment of petroleum-contaminated wastewater concerning Total Petroleum Hydrocarbons (TPH), Oil and Grease (OG), Total Suspended Solids (TSS), and Total Dissolved Solids (TDS).

2. Research Methodology

This section explains in depth the methods followed in this study. The initial stage of the research was to determine the optimum depth of VF CWs; the same configurations were applied to HF in the next stage; three different proposed depths were tested in treating synthetic wastewater, and the optimum depth was selected based on the preliminary study results. Following this was a full chemical analysis of real produced water collected from oil fields based on Kuwait Environmental Public Authority (KEPA) guidelines for wastewater treatment and reuse in irrigation. To evaluate the efficiency of the CW systems, the levels of contaminants before and after the treatment process were compared against the KEPA guidelines for wastewater reuse for irrigation.

Furthermore, a full chemical analysis of the collected PW from the oil field was conducted at the beginning of the study to identify the most critical contaminants that should be regularly tested during every treatment process. Of all the contaminants found in the produced wastewater, the following four types proved to be significantly higher than the KEPA permitted limits: TPH, OG, TDS, and TSS. The other types of contaminants were evidently within the KEPA permitted limits. For this reason, only the above-mentioned four contaminants were examined regularly before and after each treatment process.

2.1. Design and Materials

2.1.1. Lab-Scale Constructed Wetlands Prototype Design

The goal of the preliminary study is to: firstly, determine the best depth for the filter media materials; and secondly, characterize the influent produced water to determine the chemical analysis needed to test the wastewater before and after treatment. The design profile views of the lab-scale prototypes are shown in Appendix A of Figure A1. It consisted of three vertical columns with dimensions of 10 cm length × 10 cm width and varying heights: 30 cm, 50 cm, and 70 cm, respectively. This scenario was tested for 3 weeks using a continuous flow mixture of real waste and synthetic wastewater, with chemical concentrations presented in the results section. It consisted of three VF CWs using the same filter media (*Bamboo* plant, sand, and gravel) to study variations in heights of the gravel layer using 30 cm, 50 cm, and 70 cm. This helped to evaluate the impact of height on the efficiency of treatment.

According to the chemical assessments of the resulting water from the three depths used for the VF CW, the most efficient depth for treatment was 70 cm. Therefore, four VF CW with dimensions of 10 cm length × 10 cm width × 135 cm height, plus four HF CW with dimensions of 135 cm length × 10 cm width × 10 cm height were constructed following the design are shown in Appendix A of Figure A2a,b.

2.1.2. Construction of the Grease Trap Tank

The focus of the major study was to evaluate the VF and HF CWs' performance, using different filter media made from waste materials, and efficiency in removing pollutants from wastewater produced from the oil extraction process. The wastewater produced from oil extraction contains a large amount of oil, grease, and heavy suspended particles. Consequently, a grease trap, presented in Figure A2c, was designed and installed as a preliminary step before the main treatment procedure commenced. This stage is very important to help in separating oil, grease, and heavy suspended particles before introducing the wastewater to the VF and HF CW systems.

The grease trap makes it possible for the oil and grease to float on the surface of the wastewater, while the heavy suspended particles sink to the bottom of the trap. The walls' openings allow only the wastewater to pass through after entrapping the oil, grease, and suspended particles inside the tank. Figure 3 shows the eight VF and HF CW systems, including the four different filter materials selected for the major study.

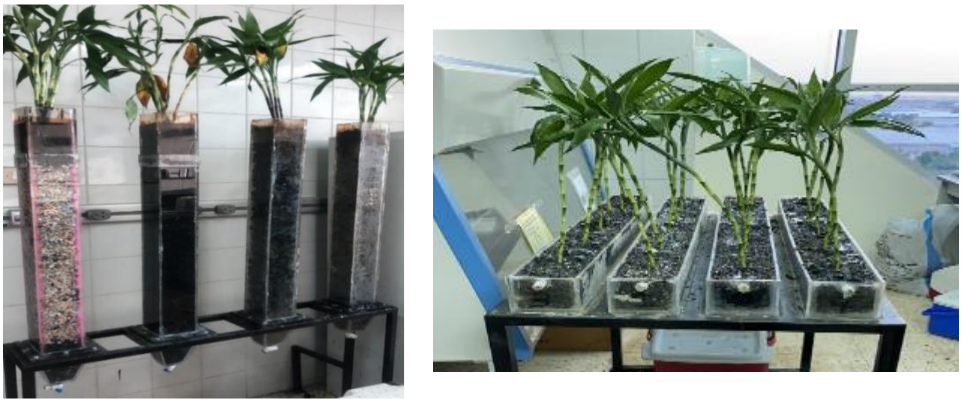


Figure 3. CW systems with four different filter materials.

2.1.3. Constructed Wetlands Filter Media Materials and Plants

Four types of filtration media materials were used (shown in Figure 4), and each material was applied to one HF and one VF CW. These materials were rubber made from shredded tires (gradation in Table 1), corrugated pieces of plastic flexible polyethene tubes used in our experiment were 15–19 mm in length and 17 mm in diameter, coarse aggregates media were stratified by coarse gravel 40–60 mm in diameter, and activated carbon with a uniform length of 15 mm and a diameter of 4 mm. On the top and bottom of each cell, a layer of fine sand of 8–15 mm was placed to facilitate planting and stabilizing the filter media. The eight HF and VF CW systems were operated for three weeks with filtration media only, and then *Bamboo* and *Cyperus* plants were cultivated in the lab-scale wetlands for three weeks each.

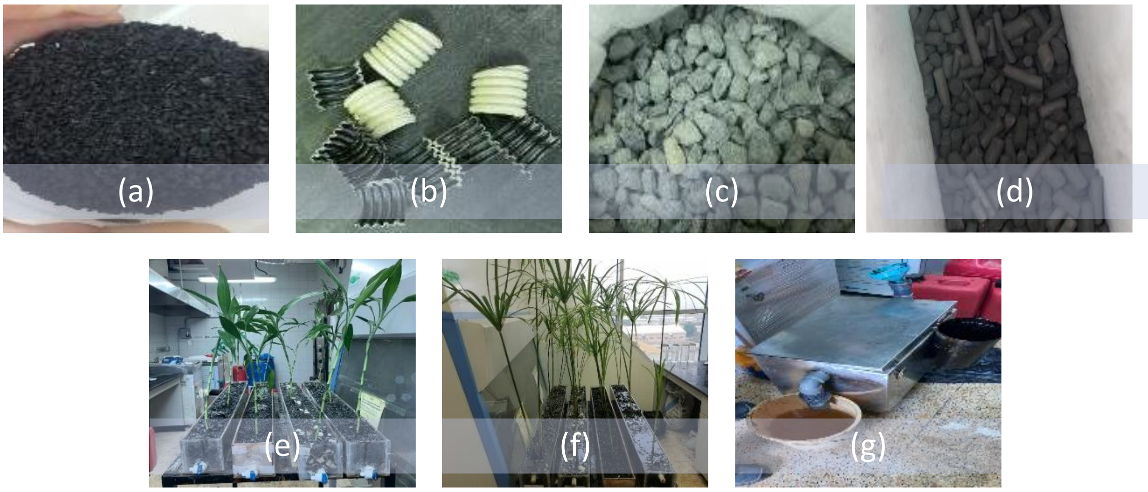


Figure 4. Samples of filtration media and plant. (**a**) Shredded tires (**b**) Plastic (**c**) Aggregates (**d**) Activated Carbon (**e**) *Bamboo* Plant (**f**) *Cyperus* Plant (**g**) grease trap.

Table 1. Shredded rubber gradation.

Sieve Size	Percent Retained	Percent Passing
2.36 mm (No. 8)	0.0	100.0
2.0 mm (No. 10)	0.0	100.0
1.18 mm (No. 16)	0.0	100.0
0.600 mm (No. 30)	30.6	69.4
0.425 mm (No. 40)	65.0	35.0
0.300 mm (No. 50)	82.2	17.8
0.150 mm (No. 100)	95.6	4.4

2.2. Collecting, Sampling, and Characterizing Produced Water

The PW was collected weekly from KOC and chemically analyzed to assess the parameters of contamination then compared to the acceptable limits determined by the KEPA guidelines for reuse in agricultural purposes. Subsequently, the PW was injected into the CWs once every week. Over a 7-day period, the efficiency of the CW systems in reducing each parameter was continuously investigated. Each parameter was measured three times, and the average values were recorded.

The Mass Removal Rate (*MRR*) is the main parameter that determines the removal efficiency of contaminants concentration and difference in influent and effluent after subsequent stages of a constructed wetland using the following formula:

$$MMR = [(\ C_{in} Q_{in}) - (C_{out}\ Q_{out})]/A \quad [\mathrm{g\,m^{-2} d^{-1}}], \tag{1}$$

where A is the area of the constructed wetland bed [m^2], Q_{in} and Q_{out} are the average influent and effluent flow rates, respectively [m^3 d^{-1}], and C_{in} and C_{out} are the average influent and effluent contaminant concentrations, respectively [mg L^{-1}] [49].

The removal performance of any CW is a function of the contaminant decay rate, where a kinetic experiment is normally conducted to account for the rate that varies from one contaminant to another. This, in turn, determines the detention time that the design of the specific CW should provide to achieve full contamination decay at the designed rate [50]. The first-order decay rate is normally assumed in reference to other parameters. The CW design parameters include the retention time, flow rates, surface bed area, contaminant concentrations, and the decomposition constant coefficients (k) for wastewater treated in HF and VF beds, which are normally obtained by applying the first-order equation [51]:

$$(C_{out}/C_{in} = e^{-k\,T}), \tag{2}$$

where k is the contaminant decay rate in d^{-1}, and T is the hydraulic retention time in days.

3. Results

This section describes the results obtained from the preliminary and major studies.

3.1. Preliminary Study and Determination of CW Configuration

To evaluate the impact of the height of the filter media on the efficiency of treatment, three VF CWs with the same media consisting of *Bamboo* plants, sand, and gravel were tested for three weeks. The filter media height was the only variable for testing the VF CW's treatment of synthetic wastewater with the composition given in Table 2. Based on the evaluation shown in Table 3, the maximum contaminant removal percentages resulting from the 70 cm filter media are as follows: Ammonium Nitrogen (54%); Chemical Oxygen Demand improvement (74%); and Total Nitrogen (58%) by CW.

Table 2. Synthetic wastewater composition.

Chemical Components	**Amount/5 L of Wastewater**
Sodium Acetate (CH_3COONa)	4.75 g
Monopotassium Phosphate ($KH_2\,PO_4$)	0.125 g
Dipotassium Phosphate (K_2HPO_4)	0.125 g
Potassium Chloride (KCl)	3.7 g
Sodium Chloride (NaCl)	2.9 g
Ammonium Chloride (NH_4Cl)	1.0905 g
Magnesium Sulfate ($MgSO_4$)	0.5 g
Calcium Chloride Dihydrate ($CaCl_2{\cdot}2H_2O$)	0.5 g

Table 3. The efficiency of pollutants' removal in the preliminary study.

Average Pollutant Removal Efficiency			
Water Quality Parameters	**30 cm Filter**	**50 cm Filter**	**70 cm Filter**
Ammonium Nitrogen (NH_3-N)	29.7%	41.3%	54.1%
Nitrite Nitrogen (NO_2-N)	3.6%	4.6%	11.3%
Total Nitrogen (TN)	26.8%	42.0%	57.8%
Total Phosphate (TP)	1.1%	18.2%	28.0%
Chemical Oxygen Demand (COD)	38.9%	63.0%	73.7%

3.2. Produced Water First Sample Characterization

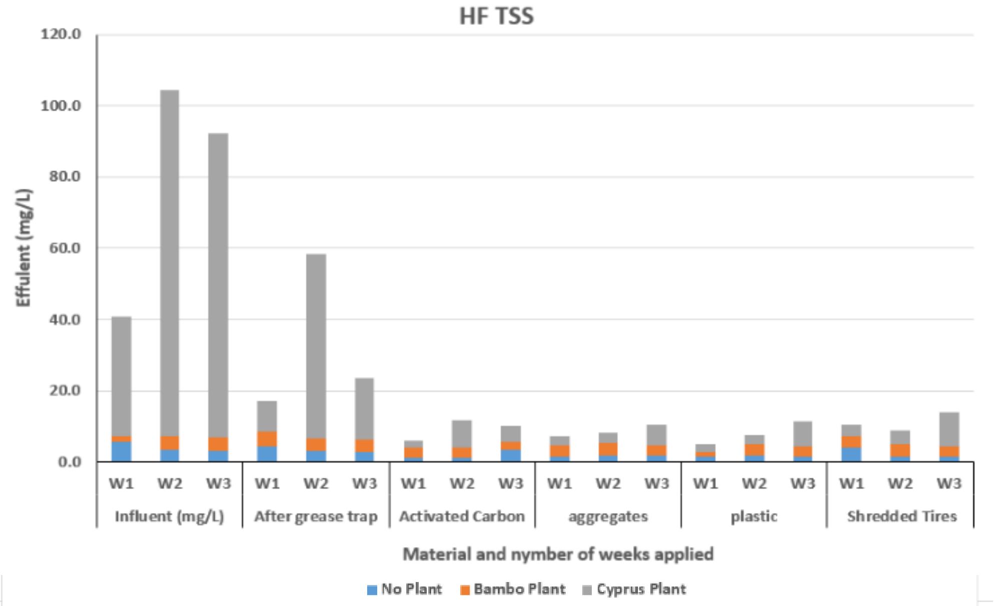
As mentioned earlier in the methodology section and confirmed by the attached results, four levels of contaminants, highlighted in blue, have already exceeded the KEPA limits. Therefore, as shown in Table 4 only the four parameters highlighted in blue (OG, TPH, TSS, and TDS) were regularly tested for the raw produced water, the discharge of the grease trap, and the effluent of all the CWs.

Table 4. Summary of the first produced water sample analysis.

No.	Test Method and Parameters	Standards	KEPA Limit	Result	Unit
1	pH	APHA 4500 HB	6.5–8.5	6.9	-
2	BOD (5 days, 200 °C)	APHA 5210 B	20	13.2	mg/L
3	COD	APHA 5220 C	100	45	mg/L
4	Dissolved Oxygen (DO)	APHA 4500-O G	>2	3.2	mg/L
5	Residual Chlorine	APHA 4500 CL B	0.5–1.0	0.12	mg/L
6	Floatables	APHA 2530	Nil	Nil	mg/L
7	Oil and Grease	APHA 5520 B	5	6.2	mg/L
8	Total Suspended Solids (TSS)	APHA 2540 D	15	450	mg/L
9	Total Dissolved Solids (TDS)	APHA 2540 C	1500	59,500	mg/L
10	Phosphates as PO_4	APHA 4500-P D	30	1.59	mg/L
11	Ammonia	APHA 4500 NH3 D	15	9.5	mg/L
12	Total Kjeldahl Nitrogen	APHA 4500 NORG B	30	19.6	mg/L
13	Total Nitrogen	APHA 4500 NORG B	65	46.5	mg/L
14	Total Recoverable Phenol	APHA 5530 C	1	0.15	mg/L
15	Fluoride	APHA 4500-F D	25	<0.05	mg/L
16	Sulfide	APHA 4500-S2F	0.1	<0.05	mg/L
17	Aluminum as Al	USEPA 6010B	5	<0.01	mg/L
18	Arsenic as As	USEPA 6010B	0.1	<0.01	mg/L
19	Barium as Ba	USEPA 6010B	2	1.1	mg/L
20	Boron as B	USEPA 6010B	2	<0.01	mg/L
21	Cadmium as Cd	USEPA 6010B	0.01	<0.01	mg/L
22	Chromium as Cr	USEPA 6010B	0.15	<0.01	mg/L
23	Nickel as Ni	USEPA 6010B	0.2	<0.01	mg/L
24	Mercury as Hg	USEPA 6010B	0.001	0.001	mg/L
25	Cobalt as Co	USEPA 6010B	0.2	<0.01	mg/L
26	Iron as Fe	USEPA 6010B	5	0.3	mg/L
27	Antimony as Sb	USEPA 6010B	1	<0.01	mg/L
28	Copper as Cu	USEPA 6010B	0.2	0.05	mg/L
29	Manganese as Mn	USEPA 6010B	0.2	0.1	mg/L
30	Zinc as Zn	USEPA 6010B	2	0.2	mg/L
31	Lead as Pb	USEPA 6010B	0.5	<0.01	mg/L
32	Total Petroleum Hydrocarbon	ASTM 1664 A	5	9.4	mg/L
33	*Faecal Coliform*	APHA 92212017,23rd	100	90	CFU/100 mL
34	*E.Coli*	USEPA 1603:2014	50	30	CFU/100 mL
35	*Faecal Streptococci*	ISO 7899-2:2000	50	<1	CFU/100 mL
36	Egg Parasite	APHAMICROSCOPIC	Nil	Nil	-

3.3. Major Study Results

The following Figure 5 highlights the results of the PW contamination analysis before and after treatment. The results represent parameter reductions in mg/L (TSS, TDS, OG, and TPH) from the influent (raw PW before and after grease trap) and observed in effluents of the HF and CF CWs in three operation scenarios (unplanted filter media, *Bamboo* plant, and *Cyperus* plant).



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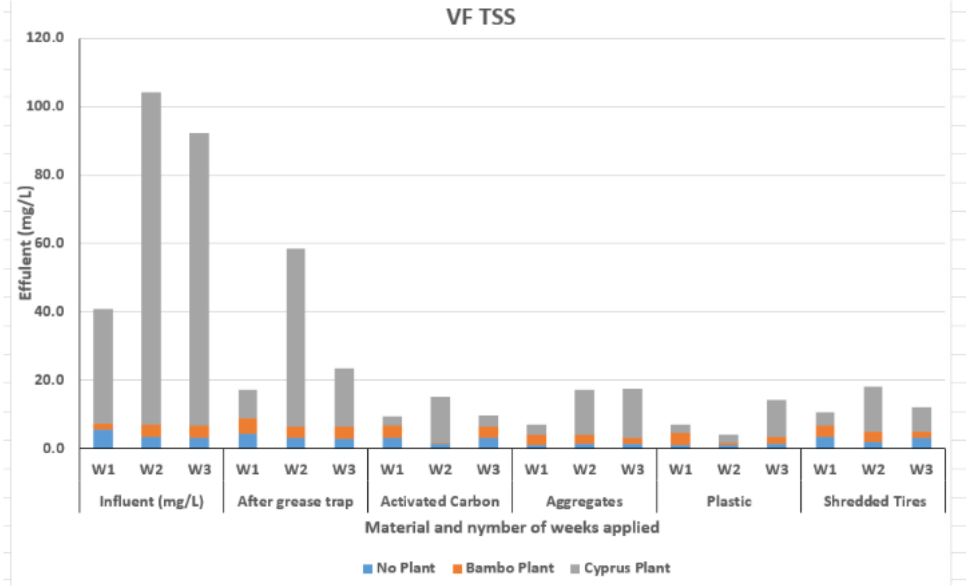

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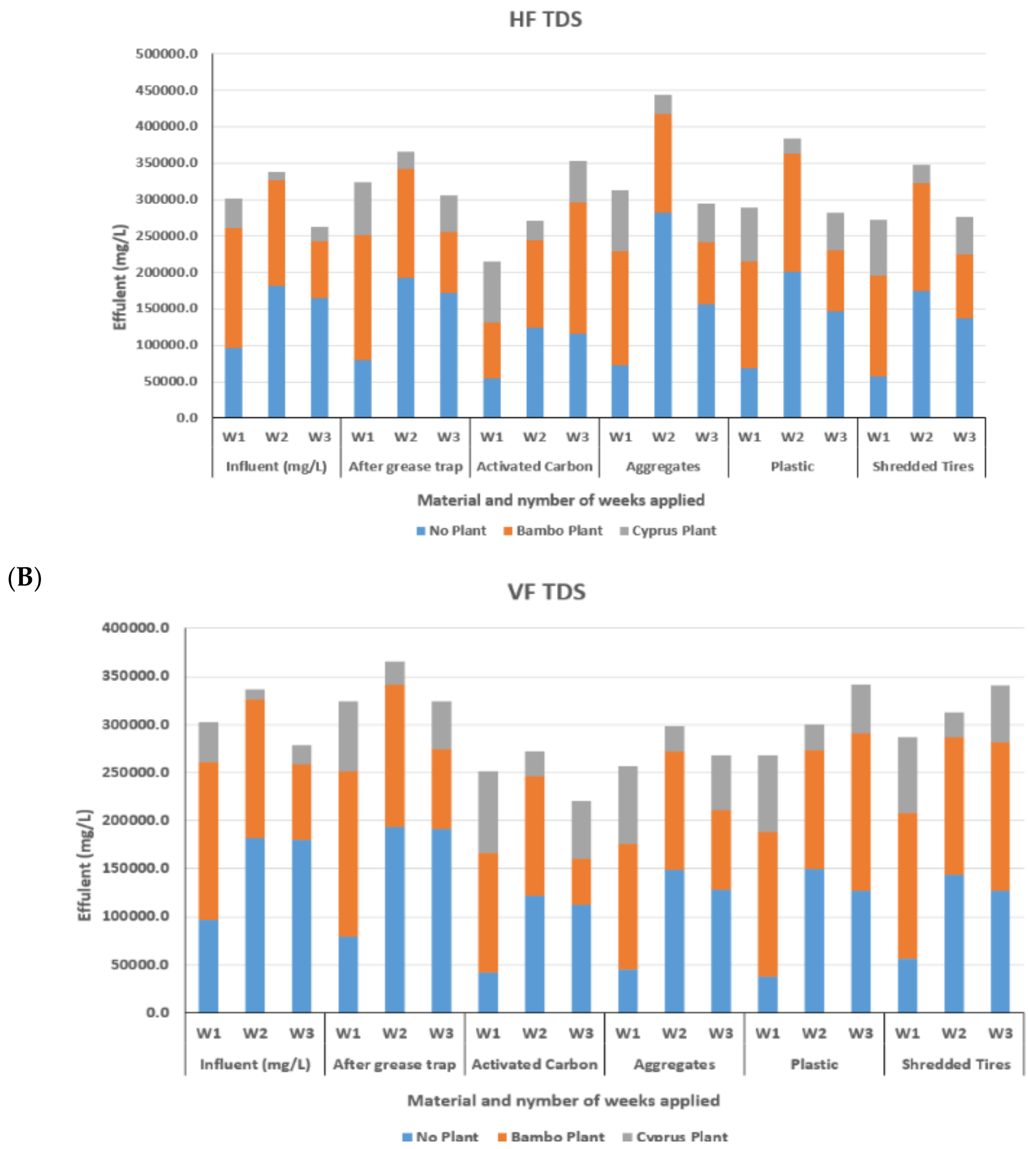

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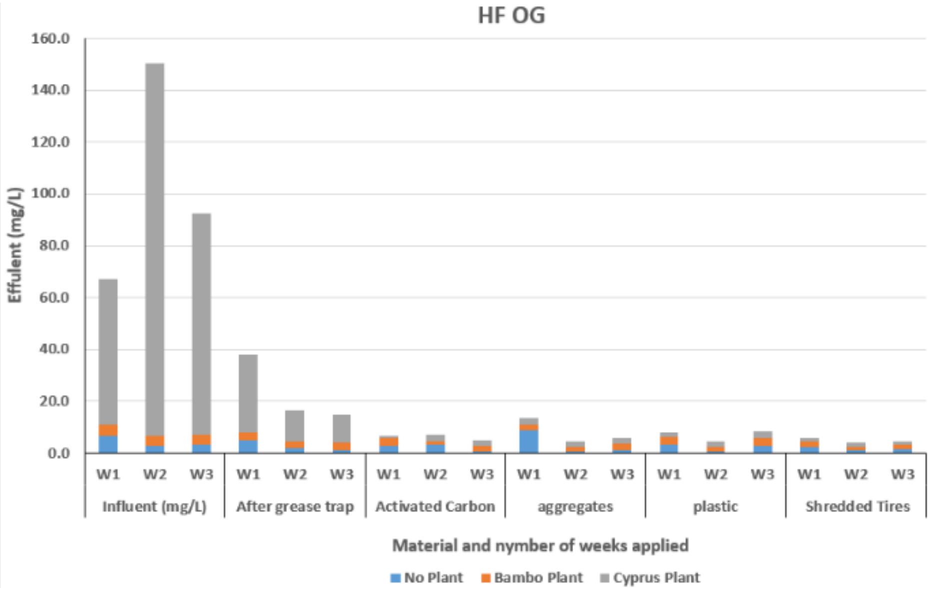

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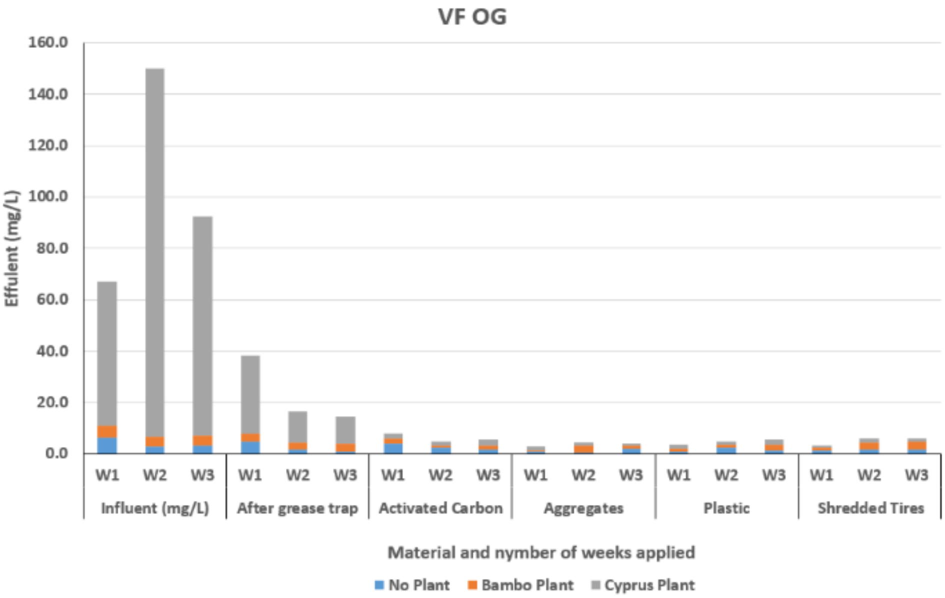

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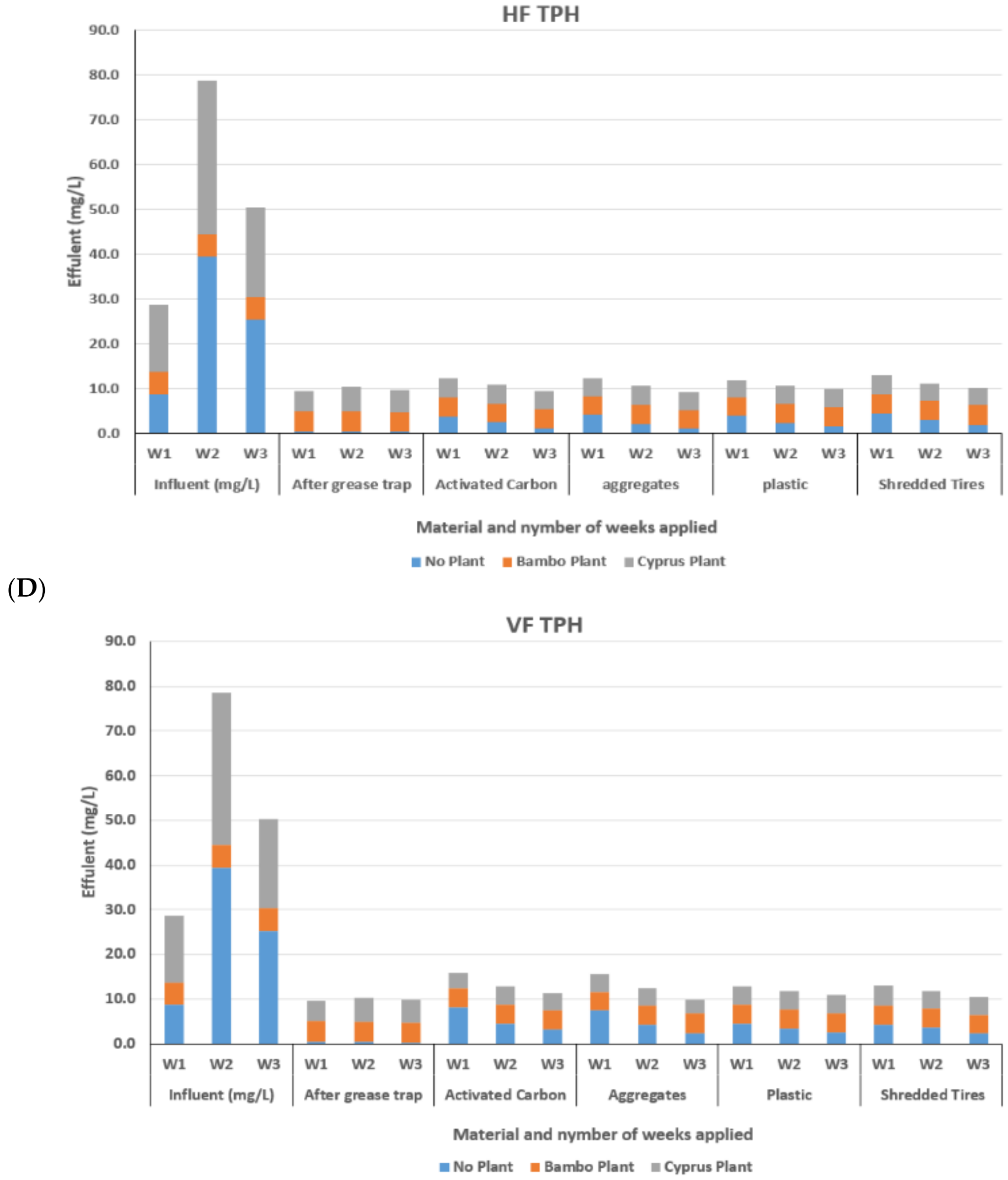


Figure 5. Reduction of contaminants: (**A**) TDS, (**B**) TSS, (**C**) OG, and (**D**) TPH from PW by HF CWs and VF CWs.

4. Discussion and Analysis

In the results obtained in HF CWs, the highest removal of TSS from PW was achieved using aggregates and plastic filtration with the *Cyperus* plant, where TSS average concentration dropped from 72.1 to 33.7 and 3.9 mg/L, respectively (removal percentages of 94.8% and 94.7%). Additionally, using the same operational scenario, activated carbon and shredded tires also showed great TSS reduction, from 72.1 to 4.5 and 5.5 mg/L, respectively (removal percentages of 93.7% and 92.4%). In the case of TDS, the highest concentration reduction was achieved by activated carbon filtration media in the CWs scenario operating unplanted, where TDS dropped from 147,533.3 to 97,861.7 mg/L (a removal percentage of 33.7%), followed by shredded tires and plastic, where TDS values dropped from 147,533.3 to 122,591.7 and 138,288.3 mg/L, respectively (a removal percentage of 16.9% and 6.3%).

The concentrations of PW distinctive pollutants expressed as OG and TPH and the HF CWs combined with grease traps showed great performance in the parameters' removal percentages, with values over 90% in the filter material and *Cyperus* scenario. The grease trap reduced a noticeable amount of OG by 26.2–81.6%. This was further reduced by various filter media, with the highest values recorded by shredded tires in all the operating scenarios, with reduction percentages of 59–98.4%, followed by activated carbon at 47.9–97.9%, plastic at 47.7–97.77%, and aggregates at 15.4–97.6%. Interestingly, the performance of the HF CWs in TPH removal follows the same behavior as OG, where most of the reductions occur in the grease trap phase. However, the filter media with the highest TPH removal recorded was aggregates, with reduction percentages of 15.2–90.2%, followed by activated carbon at 12.5–89.9%, plastic at 14.6–89.4%, and shredded tires at 15.2–87.2%. For all filtration materials, the upper limit of removal percentages was achieved during the scenario operating unplanted.

In VF CWs, the highest removal of TSS from PW was achieved using plastic and activated carbon filtration with the *Cyperus* plant, where TSS average concentration dropped from 72.1 to 5.2 and 6.5 mg/L, respectively (removal percentages of 92.8% and 91%). During the same operational scenario, shredded tires and aggregate showed TSS reductions from 72.1 to 8 and 10.2 mg/L, respectively (removal percentages of 88.9% and 85.8%). In the analysis of TDS, the highest concentration reduction was achieved by activated carbon filtration media in the CWs scenario operating unplanted, where TDS dropped from 152,929.7 to 92,166.7 mg/L (a removal percentage of 39.7%) followed by plastic, aggregates, and shredded tires with values of 104,802.7, 107,201.0, and 108,951.7 mg/L, respectively (removal percentages of 31.5%, 29.9%, and 28.8%, respectively).

Regarding PW distinctive pollutants reduction, the VF CWs combined with the grease trap showed similar performance in the OG and TPH decay rates with HF CWs results, where most of the reduction was by the grease trap. The grease trap reduced OG percentages by 35–98.7%. This was further reduced by various filter media, with the highest values recorded by shredded tires with reduction percentages of 44.2–98.7%, aggregates at 63.1–98.77%, plastic at 60.2–98.6%, and activated carbon at 35.0–98.0%. Lastly, the highest TPH removal recorded by VF CWs was by shredded tires and plastic during the unplanted filter medium scenario with reduction percentages of 86.2% and 85.8%, respectively, followed by aggregates and activated carbons with reduction percentages of 84.4% and 83.4%, respectively, using filtration materials and a *Cyperus* plant.

This may indicate that the removal of petroleum water compounds depends not only on the conditions occurring in the wetland, such as the filtration materials, plant of the system, and configuration, but also on the pretreatment and the presence of a septic tank.

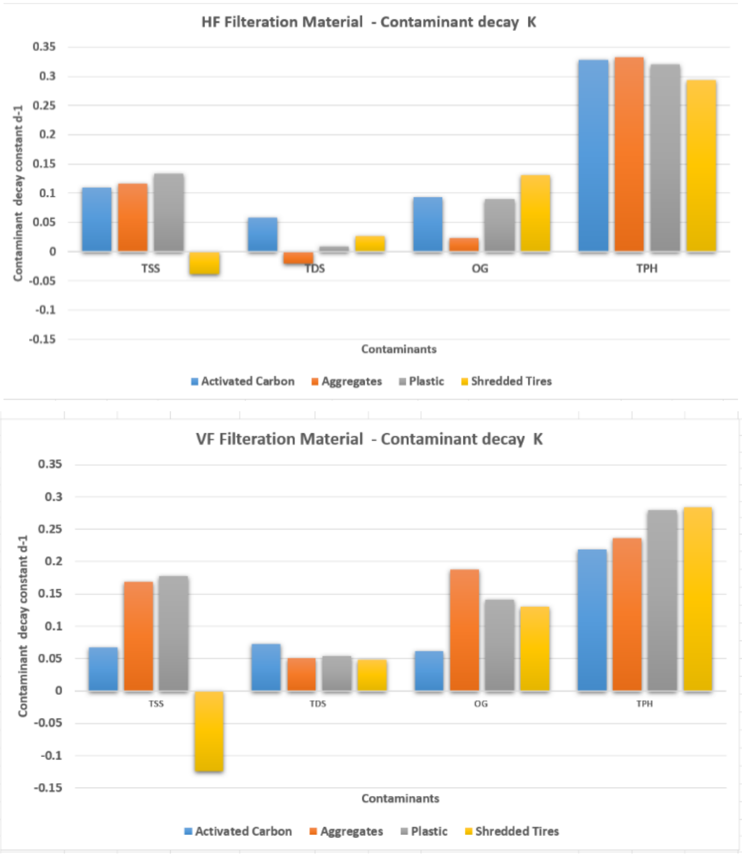
The data presented in Table 5 below offers an attempt to perform a data regression to extract a trend of the treatment efficiency after using the *MRR* equation for the three treatment scenarios for various materials.

Although some of the results showed a high regression coefficient R^2 for the four materials with most of the values nearly equal to 1, this correlation may not precisely reflect the actual test due to the limited number of sampling points for each material. Nevertheless, the data presented for the four filter materials proved good potential for the addressed contaminants. However, from the R^2 results, the waste materials showed potential as soil amendments and filters for CWs.

The TSS, TDS, OG, and TPH removal k-rates in horizontal and vertical flow with different bed filter mediums were calculated and given separately for each operating scenario. Based on the graph shown in Figure 6, reasonable TSS, TDS, OG, and TPH removal efficiency by plastic and shredded tires in comparison to the control materials aggregates and activated carbon. The contaminants' decay rate results were also found to agree with the *MRR* and regression coefficient R^2 results. The low efficiency of the shredded tires compared to the other materials in TSS and TDS could be due to the tire materials' properties, which might not be fully utilized for such applications [52]. More research on the waste materials in the same field could be taken to further understand their behaviour.

Table 5. Correlation coefficient (R^2) of CW models using different filter materials for PW parameters.

R^2		HCW				VCW			
		TSS	TDS	OG	TPH	TSS	TDS	OG	TPH
Activated Carbon Effluent	**No Plant**	0.995	0.795	0.315	0.875	0.922	0.650	0.818	0.819
	Bamboo Plant	0.595	0.999	0.956	0.579	0.512	0.696	0.999	0.509
	Cyperus Plant	0.901	0.792	0.934	0.655	0.901	0.792	0.943	0.635
Aggregates Effluent	**No Plant**	0.720	0.147	0.588	0.886	0.809	0.397	0.953	0.866
	Bamboo Plant	0.580	0.945	1.000	0.617	0.632	0.986	0.379	0.298
	Cyperus Plant	0.944	0.820	0.915	0.650	0.915	0.820	0.940	0.701
Plastic Effluent	**No Plant**	0.663	0.019	0.977	0.871	0.147	0.358	0.897	0.875
	Bamboo Plant	0.953	0.378	0.844	0.298	0.295	0.922	0.999	0.579
	Cyperus Plant	0.647	0.745	0.958	0.958	0.957	0.745	0.939	0.659
Shredded Tires Effluent	**No Plant**	0.216	0.216	0.891	0.887	0.546	0.994	0.514	0.877
	Bamboo Plant	0.216	0.216	0.891	0.886	0.994	0.546	0.514	0.878
	Cyperus Plant	0.935	0.680	0.918	0.686	0.918	0.680	0.935	0.686



(A)

Figure 6. *Cont.*

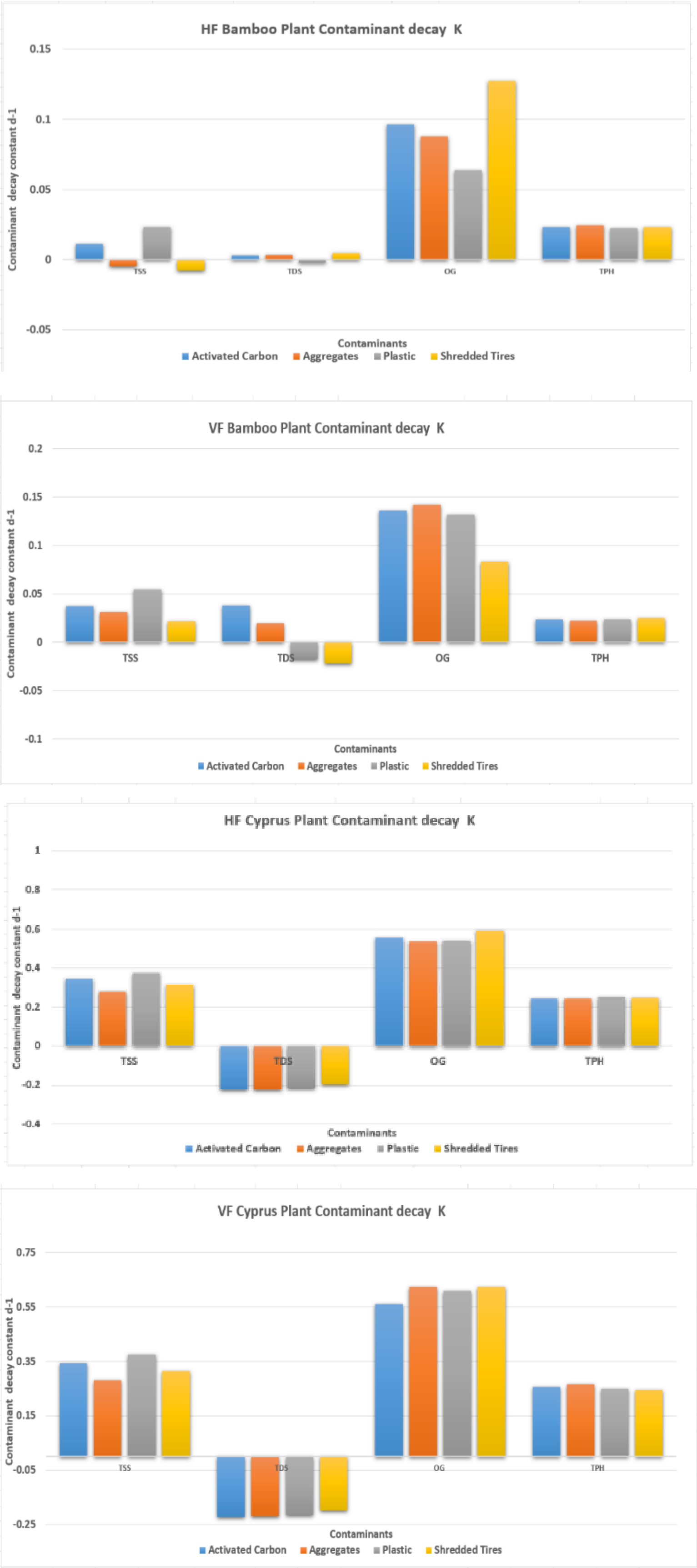


Figure 6. Decay rates (k) for various contaminants by HF and VF CWs through different scenarios (**A**) Filtration materials are not planted (**B**) *Bamboo* planted (**C**) *Cyperus* planted.

5. Conclusions and Recommendations

5.1. Conclusions

The preliminary results showed that the best possible depth of the VF CW systems, 70 cm depth, was found to be the most efficient in wastewater treatment. Chemical analysis of the produced water effluent collected from the oil field showed that Total Petroleum Hydrocarbons (TPH), Oil and Grease (OG), Total Suspended Solids (TSS), and Total Dissolved Solids (TDS) were the contaminants exceeding the KEPA allowable limits for wastewater. Adding a grease trap oil separator system combined with the VCW materials removes a high percentage of oil, grease, and hydrocarbon chemical materials from the produced water. The combined system proved effective at removing contaminants, with the highest removal percentages of 94.8% for TSS, 33.7% for TDS, 90.2% for OG, and 98.4% for TPH over the 7 day retention time. The analysis involved particularly mass removal rates (MRR) and first-order removal rate coefficients of TSS, TDS, OG, and TPH for HF and VF CWs using different filters (aggregates, activated carbon, plastic, and shredded tires). On the basis of the charts presented in Figures 5 and 6, both HF and VF constructed wetlands showed similar efficiency in PW treatment. Additionally, it was found that the waste materials used (plastic and shredded tires) proved to have a good potential in comparison to conventional materials (aggregates and activated carbon) for the removal efficiency of contaminants. Based on the previous conclusions, CW succeeded in presenting circular treatment technology for both solid waste and wastewater.

5.2. Recommendations

The extravagant use of finite water resources and materials is not sustainable in the long term. A current growing concern now is discussing the applications of nature-based solutions in wastewater treatment. Additionally, there is great interest in promoting CE in the industrial sector and water management to ensure a sustainable future for the coming generations. Hence, more efforts and further research should cover the applications of CE in different industrial activities and processes.

Investing in CW implementations will increase water resources, prevent further global pollution, and boost the social and economic benefits. The performance of CW in the treatment of PW proved, in practice, its high treatment capacity. Furthermore, to support CE in the petroleum industry, we encouraged the use of waste recycling applications in wetland technology. The research has shown potential for new CWs filter materials (plastic and shredded tires), although their treatment efficiency has not been fully investigated due to testing one source of wastewater (PW). The use of other alternative influents such as municipal, agricultural, and other industrial wastewater is among the research areas that can be investigated in the future. The presented CWs were tested on a laboratory scale, and it is recommended to consider the analysis of PW treatment by CWs in experiments at field scale to check the CWs' operational efficiency with continuous PW extraction and filling processes.

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Appendix A

The experimental layout of the laboratory constructed wetlands are presented in this section.

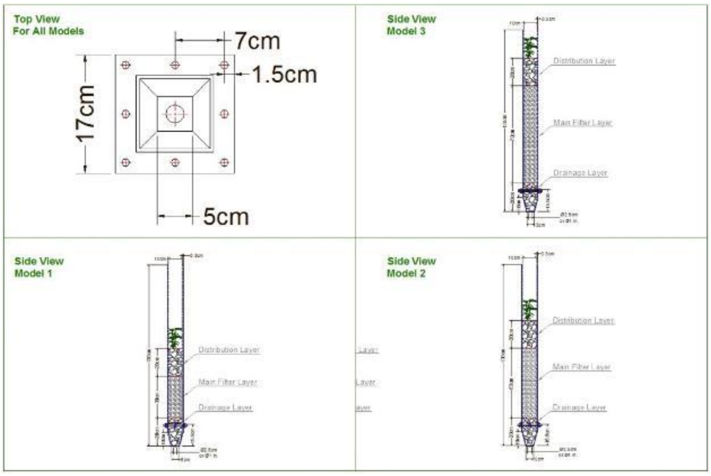

Figure A1. Preliminary study Vertical Constructed Wetlands Model.

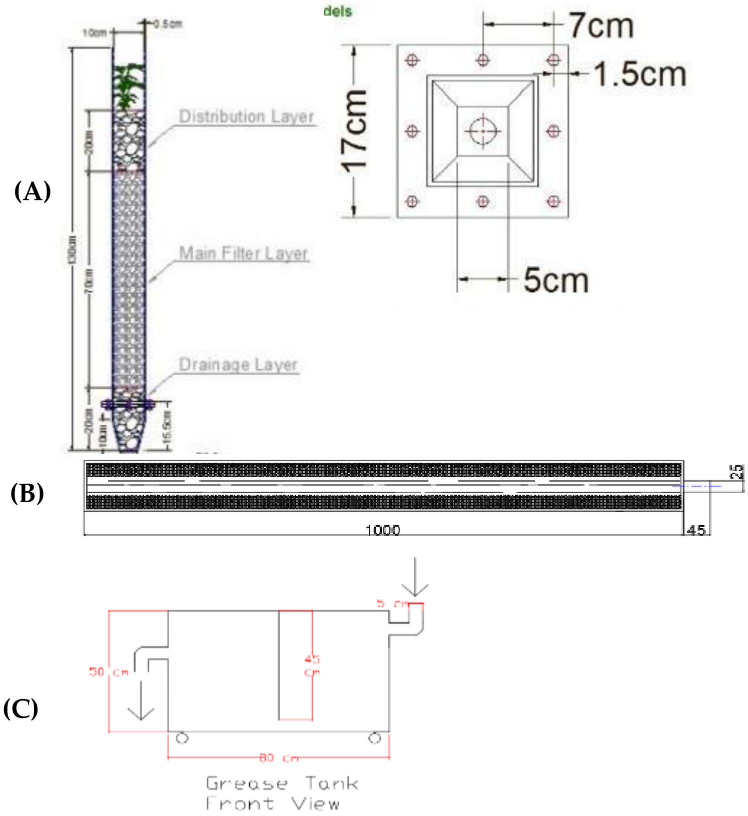

Figure A2. Design layout of constructed wetlands. (**A**) Vertical wetland (**B**) Horizontal Wetlands (**C**) Grease Trap.

References

1. Ciasullo, M.V.; Castellani, P.; Rossato, C.; Troisi, O. Sustainable business model innovation. *Sinergie Ital. J. Manag.* **2019**, *37*, 213–237. [CrossRef]
2. Velenturf, A.P.; Purnell, P. Principles for a sustainable circular economy. *Sustain. Prod. Consum.* **2021**, *27*, 1437–1457. [CrossRef]
3. Reike, D.; Vermeulen, W.J.V.; Witjes, S. The circular economy: New or Refurbished as CE 3.0?—Exploring Controversies in the Conceptualization of the Circular Economy through a Focus on History and Resource Value Retention Options. *Resour. Conserv. Recycl.* **2018**, *135*, 246–264. [CrossRef]

4. Kalmykova, Y.; Sadagopan, M.; Rosado, L. Circular economy—From review of theories and practices to development of implementation tools. *Resour. Conserv. Recycl.* **2018**, *135*, 190–201. [CrossRef]
5. Ellen MacArthur Foundation (EMF). News. 2015. Available online: https://www.ellenmacarthurfoundation.org/news/circular-economy-would-increase-european-competitiveness-and-deliver-better-societal-outcomes-new-study-reveals (accessed on 20 December 2022).
6. EMF. *Towards a Circular Economy: Business Rationale for an Accelerated Transition*; Ellen MacArthur Foundation: Cowes, UK, 2015; p. 20. Available online: https://emf.thirdlight.com/link/ip2fh05h21it-6nvypm/@/preview/1?o (accessed on 20 December 2022).
7. *Circular Economy for Sustainable Development in Serbia*; United Nations Development Programme: New York, NY, USA, 2019; Available online: https://www.undp.org/serbia/news/circular-economy-sustainable-development-serbia (accessed on 14 October 2022).
8. Water in Circular Economy and Resilience (WICER). Available online: https://www.worldbank.org/en/topic/water/publication/wicer (accessed on 17 October 2022).
9. Mbavarira, T.; Grimm, C. A systemic view on circular economy in the water industry: Learnings from a belgian and dutch case. *Sustainability* **2021**, *13*, 3313. [CrossRef]
10. Englande, A.J., Jr.; Krenkel, P.; Shamas, J. Wastewater Treatment &Water Reclamation. *Ref. Modul. Earth Syst. Environ. Sci.* **2015**. [CrossRef]
11. Lipponen, A.; Nikiforova, N. *Wastewater: The Untapped Resource*; UNESCO: Paris, France, 2017.
12. EPA U.S. Office of Research and Development, Technology Transfer and Support Division; Paris, L. *US EPA Guidelines for Water Reuse*; US EPA: Washington, DC, USA, 2004.
13. Yaqoob, I.; Parveen, A.A.; Umar, T.; Nasir, K.; Nasir, M. Role of Nanomaterials in the Treatment of waste water. *Water* **2020**, *12*, 495. [CrossRef]
14. Ghimire, U.; Sarpong, G.; Gude, V.G. Transitioning wastewater treatment plants toward circular economy and energy sustainability. *ACS Omega* **2021**, *6*, 11794–11803. [CrossRef]
15. Beyer, J.; Goksøyr, A.; Hjermann, D.Ø.; Klungsøyr, J. Environmental effects of offshore produced water discharges: A review focused on the Norwegian continental shelf. *Mar. Environ. Res.* **2020**, *162*, 105155. [CrossRef]
16. Salem, F.; Thiemann, T. Produced Water from Oil and Gas Exploration—Problems, Solutions and Opportunities. *J. Water Resour. Prot.* **2022**, *14*, 142–185. [CrossRef]
17. Zolghadr, E.; Firouzjaei, M.D.; Amouzandeh, G.; LeClair, P.; Elliott, M. The Role of Membrane-Based Technologies in Environmental Treatment and Reuse of Produced Water. *Front. Environ. Sci.* **2021**, *9*, 629767. [CrossRef]
18. Clark, C.E.; Veil, J.A. *Produced Water Volumes and Management Practices in the United States*; (No. ANL/EVS/R-09-1); Argonne National Lab.(ANL): Argonne, IL, USA, 2009.
19. Miranda, M.A.; Ghosh, A.; Mahmodi, G.; Xie, S.; Shaw, M.; Kim, S.; Krzmarzick, M.J.; Lampert, D.J.; Aichele, C.P. Treatment and Recovery of High-Value Elements from Produced Water. *Water* **2022**, *14*, 880. [CrossRef]
20. Veil, J.A.; Puder, M.G.; Elcock, D.; Redweik, R.J.J. *A White Paper Describing Produced Water from Production of Crude Oil, Natural Gas, and Coal Bed Methane*; Prepared by: Argonne National Laboratory. Prepared for: U.S. Department of Energy National Energy Technology Laboratory Under Contract W-31-109-Eng-38; Argonne National Laboratory: Lemont, IL, USA; U.S. Department of Energy National Energy Technology Laboratory: Pittsburgh, PA, USA, 2004; p. 87. Available online: http://www.osti.gov/servlets/purl/821666/%0A (accessed on 20 November 2022).
21. Olajire, A.A. Recent advances on the treatment technology of oil and gas produced water for sustainable energy industry-mechanistic aspects and process chemistry perspectives. *Chem. Eng. J. Adv.* **2020**, *4*, 100049. [CrossRef]
22. Danforth, C.; Chiu, W.A.; Rusyn, I.; Schultz, K.; Bolden, A.; Kwiatkowski, C.; Craft, E. An integrative method for identification and prioritization of constituents of concern in produced water from onshore oil and gas extraction. *Environ. Int.* **2020**, *134*, 105280. [CrossRef] [PubMed]
23. Shining New Light on the Toxicity of Chemicals in Produced Water. Available online: https://blogs.edf.org/energyexchange/2019/11/13/shining-new-light-on-the-toxicity-of-chemicals-in-produced-water/ (accessed on 18 October 2022).
24. Fakhru'L-Razi, A.; Pendashteh, A.; Abdullah, L.C.; Biak, D.R.A.; Madaeni, S.S.; Abidin, Z.Z. Review of technologies for oil and gas produced water treatment. *J. Hazard. Mater.* **2009**, *170*, 530–551. [CrossRef]
25. Alley, B.; Beebe, A.; Rodgers, J.; Castle, J.W. Chemical and physical characterization of produced waters from conventional and unconventional fossil fuel resources. *Chemosphere* **2011**, *85*, 74–82. [CrossRef]
26. Neff, J.; Lee, K.; DeBlois, E.M. Produced Water: Overview of Composition, Fates, and Effects. In *Produced Water*; Springer: New York, NY, USA, 2011; pp. 3–54.
27. Rajasulochana, P.; Preethy, V. Comparison on efficiency of various techniques in treatment of waste and sewage water—A comprehensive review. *Resour. Technol.* **2016**, *2*, 175–184. [CrossRef]
28. Igunnu, E.T.; Chen, G.Z. Produced water treatment technologies. *Int. J. Low-Carbon Technol.* **2014**, *9*, 157–177. [CrossRef]
29. de Medeiros, A.D.M.; Junior, C.J.G.D.S.; de Amorim, J.D.P.; Durval, I.J.B.; Costa, A.F.D.S.; Sarubbo, L.A. Oily Wastewater Treatment: Methods, Challenges, and Trends. *Processes* **2022**, *10*, 743. [CrossRef]
30. Wei, X.; Zhang, S.; Han, Y.; Wolfe, F.A. Treatment of petrochemical wastewater and produced water from oil and gas. *Water Environ. Res.* **2019**, *91*, 1025–1033. [CrossRef] [PubMed]

31. US Energy Information Administration. *Kuwait: Country Analysis Brief*; U.S. Energy Information Administration: Washington, DC, USA, 2016; pp. 1–20. Available online: http://www.eia.gov/beta/international/analysis_includes/countries_long/Nigeria/nigeria.pdf (accessed on 20 October 2022).
32. Aljuboury, D.A.D.A.; Palaniandy, P.; Abdul Aziz, H.; Feroz, H.B. Treatment of petroleum wastewater by conventional and new technologies—A review. *Glob. NEST Int. J.* **2017**, *19*, 439–452. [CrossRef]
33. Heneghan, E.; Collier, M.; Kelly-Quinn, M. An evaluation of the potential applications of nature-based solutions for water quality protection: Ireland as a case study. *Boil. Environ.* **2021**, *121B*, 147–162. [CrossRef]
34. Vymazal, J. Wastewater Treatment and Control Through Wetlands. In *Encyclopedia of Sustainability Science and Technology*; Meyers, R.A., Ed.; Springer: New York, NY, USA. [CrossRef]
35. Stefanakis, A.I.; Prigent, S.; Breuer, R. Integrated Produced Water Management in a Desert Oilfield Using Wetland Technology and Innovative Reuse Practices. In *Constructed Wetlands for Industrial Wastewater Treatment*; John Wiley & Sons, Ltd.: Hoboken, NJ, USA, 2018; pp. 23–42. [CrossRef]
36. Vymazal, J.; Greenway, M.; Tonderski, K.; Brix, H.; Mander, Ü. Constructed Wetlands for Wastewater Treatment. In *Wetlands and Natural Resource Management*; Springer: Berlin/Heidelberg, Germany, 2006; pp. 69–96.
37. Programme, J.O. Educational material. *Dent. Radiogr. Photogr.* **1966**, *39*, 39. [CrossRef]
38. Vymazal, J. Constructed wetlands for wastewater treatment. *Encycl. Ecol.* **2018**, 14–21. [CrossRef]
39. Brix, H. Wastewater Treatment in Constructed Wetlands: System Design, Removal Processes, and Treatment Performance. In *Constructed Wetlands for Water Quality Improvement*; CRC Press: Boca Raton, FL, USA, 1993; pp. 9–22. [CrossRef]
40. Vymazal, J. Constructed wetlands for wastewater treatment. *Water* **2010**, *2*, 530–549. [CrossRef]
41. Rahman, E.; Bin Halmi, M.I.E.; Samad, M.Y.B.A.; Uddin, K.; Mahmud, K.; Shukor, M.Y.A.; Abdullah, S.R.S.; Shamsuzzaman, S.M. Design, Operation and Optimization of Constructed Wetland for Removal of Pollutant. *Int. J. Environ. Res. Public Health* **2020**, *17*, 8339. [CrossRef]
42. Vymazal, J. Constructed Wetlands for Wastewater Treatment: Five Decades of Experience. *Environ. Sci. Technol.* **2011**, *45*, 61–69. [CrossRef]
43. Vymazal, J. The Historical Development of Constructed Wetlands for Wastewater Treatment. *Land* **2022**, *11*, 174. [CrossRef]
44. Stefanakis, A.I. The Role of Constructed Wetlands as Green Infrastructure for Sustainable Urban Water Management. *Sustainability* **2019**, *11*, 6981. [CrossRef]
45. Stefanakis, A.I. Constructed wetlands for sustainable wastewater treatment in hot and arid climates: Opportunities, challenges and case studies in the Middle East. *Water* **2020**, *12*, 1665. [CrossRef]
46. Vymazal, J.; Kröpfelová, L. *Wastewater Treatment in Constructed Wetlands with Horizontal Sub-Surface Flo*; Google Books; Available online: https://books.google.com.kw/books?hl=en&lr=&id=IfqerCqRvg8C&oi=fnd&pg=PR11&ots=rxwr4pLE2i&sig=FluxObknFLjBHggIztDDAAy2xc8&redir_esc=y#v=onepage&q&f=false (accessed on 18 October 2022).
47. Vymazal, J.; Zhao, Y.; Mander, Ü. Recent research challenges in constructed wetlands for wastewater treatment: A review. *Ecol. Eng.* **2021**, *169*, 106318. [CrossRef]
48. Villaseñor, J.; Capilla, P.; Rodrigo, M.; Cañizares, P.; Fernández, F. Operation of a horizontal subsurface flow constructed wetland—Microbial fuel cell treating wastewater under different organic loading rates. *Water Res.* **2013**, *47*, 6731–6738. [CrossRef] [PubMed]
49. Gajewska, M.; Skrzypiec, K.; Jóźwiakowski, K.; Mucha, Z.; Wójcik, W.; Karczmarczyk, A.; Bugajski, P. Kinetics of pollutants removal in vertical and horizontal flow constructed wetlands in temperate climate. *Sci. Total. Environ.* **2020**, *718*, 137371. [CrossRef] [PubMed]
50. Pastor, R.; Benqlilou, C.; Paz, D.; Cardenas, G.; Espuña, A.; Puigjaner, L. Design optimisation of constructed wetlands for wastewater treatment. *Resour. Conserv. Recycl.* **2003**, *37*, 193–204. [CrossRef]
51. Hassan, I.; Chowdhury, S.R.; Prihartato, P.K.; Razzak, S.A. Wastewater treatment using constructed wetland: Current trends and future potential. *Processes* **2021**, *9*, 1917. [CrossRef]
52. Kaushik, M.K.; Kumar, A. Beneficial Use of Shredded Tires of Different Sizes with Gravel as Drainage Layer of Landfill Leachate Collection Syastem. In Proceedings of the Indian Geotechnical Conference 2012, Delhi, Indian, 13–15 December 2012; Available online: https://www.researchgate.net/publication/305723985 (accessed on 20 December 2022).