Performance of vertical subsurface flow constructed wetlands for commercial laundry greywater treatment: Effects of design and operational parameters

Javiera Gutiérrez[1 2], Ana María Leiva[1 2], Gloria Gómez[1 2] and Gladys Vidal[1 2*]

[1] Engineering and Biotechnology Environmental Group, Environmental Science Faculty and EULA–Chile Center, Universidad de Concepción. Concepción, Chile

[2] Water Research Center for Agriculture and Mining (CRHIAM), ANID FONDAP Center, Concepción, Chile

* Corresponding author: Gladys Vidal

E-mail address: glvidal@udec.cl

Telephone Number: +56412204067

Submitted to: *Journal of Water Process Engineering*

Abstract

Constructed wetlands (CWs) are being increasingly considered for the treatment of greywater (GW) from commercial laundry sources. However, limited information is available on how operational and design parameters influence CW performance. This study aimed to evaluate the performance of vertical subsurface flow constructed wetlands (VSSFCWs) for the treatment of commercial laundry GW with a focus on the effects of design and operational parameters. The operation of two lab-scale VSSFCWs was divided into three stages, during which ORLs progressively increased and HRTs decreased. One VSSFCW was filled with zeolite (Zeo), while the other used gravel (Gra). In Stage III, both systems achieved removal efficiencies of 73–86% for chemical oxygen demand (COD), 72–84% for ammonium-nitrogen (NH_4^+-N), and 68–89% for phosphate-phosphorus (PO_4^{3-}-P), indicating a significant improvement ($p < 0.05$) as OLR increased from 12.3 to 49.9 g·COD/m²·d and HRT decreased from 10 to 3.5 days. In all stages, the Zeo system outperformed Gra by 19–33% in terms of organic matter and nutrient removal ($p < 0.05$). Additionally, the Freundlich isotherm model revealed a reduction in the sorption capacity of the used zeolite to 0.004 mg/g, which was 1 to 5 times lower than the control zeolite. These findings emphasize the importance of optimizing operational parameters and selecting appropriate support media for the effective and sustainable reuse of commercial laundry GW.

Keywords: greywater, constructed wetland, adsorption capacity, organic loading rate, hydraulic retention time.

Highlights

1. Organic loading rate of 50 g·COD/m^2·d improved performance of constructed wetland

2. Zeo system achieved removal efficiencies between 19 – 33% higher than Gra system

3. Zeolite loses its adsorption capacity to 0.04 mg/g during the operational time

4. The adsorption capacity of zeolite was not influenced by the CW's zone

1. Introduction

The reuse of greywater (GW) for multiple purposes, such as garden irrigation, recreational green areas, and domestic uses that include car washing, floor cleaning, and toilet flushing, has emerged as a viable solution to water scarcity [1]. Studies have shown that GW constitutes 50 – 80% of the total wastewater generated in a household [2,3,4,5] and it can generally be categorized as light (bathrooms, hand sinks, and showers) and dark GW (laundry, kitchen, and dishwashers), each differing in their contaminant profiles [6]. While many studies have focused on characterizing and treating domestic GW, little attention has been paid to GW generated by commercial laundry, which is typically more concentrated in organic, nutrients, and ionic compounds [7].

Before reuse, commercial laundry GW must undergo adequate treatment to mitigate its potential environmental and health impacts [8]. Laundry GW contains a diversity of chemical compounds from detergents, oils, solvents, bleaches, and non-biodegradable fibers from clothing, that is traduced in high chemical oxygen demand (COD), turbidity, total nitrogen (TN) and total phosphorus (TP) concentrations with values ranged between 58 – 4155 mg/L, 34 – 510 NTU, 3 – 31 mg/L, 0.2 – 51.6 mg/L, respectively [3,6,9]. Constructed wetlands (CWs), as nature-based solutions (NBS), have been proven to be sustainable and innovative technologies for treating various effluents, including GW [6,10]. Numerous studies have shown that CWs achieve removal efficiencies of 70 – 97% for COD, 47 – 97% for turbidity, 44 – 59% for TN, and 24 – 74% for TP in different type of GW [3,6,7,11,12]. In the case of laundry GW, Punyapwar and Mutnuri [6] studied the performance of vertical subsurface flow constructed wetlands (VSSFCW) preceded by an anaerobic filter tank treating this type of GW. This study achieved removal efficiencies of 95%, 83%, and 95% for COD, ammonium-nitrogen (NH_4^+-N), and phosphate-phosphorus (PO_4^{3-}-P). Although high performances were achieved in this study, further research is needed to evaluate the removal of other contaminants (turbidity, ions, etc.).

To ensure the safe reuse of GW for domestic and agricultural purposes, optimizing CW performance by identifying appropriate design and operational parameters is essential [2]. Moreover, the effects of seasonal variation and environmental pressures on the removal efficiencies of CWs are another important factor to consider in the optimization of this technology [13]. Hydraulic retention times (HRTs) and organic loading rates (OLRs) are critical factors that influence contaminant removal efficiencies in CWs. The influence of these parameters on removal efficiencies has been extensively studied in CWs treating wastewater [14,15,16], but research focusing on CWs treating GW, especially from commercial laundry, remains underexplored. For instance, Ramprasad and Philip [17] explored the effects of HRT and OLR on the performance of horizontal (HSSFCW) and VSSFCW for GW treatment. Their findings revealed an increase in organic matter and nutrient removal rates from 80% to 90% with a higher HRT and lower OLR. Similarly, Ramprasad et al. [11] observed a decline in organic matter removal efficiency from 96% to 80% as the OLR increased and HRT decreased, though nitrogen removal remained unaffected by these operational changes. However, both studies used synthetic or mixed GW, leaving the specific behavior of commercial laundry GW in CWs unaddressed.

Another key parameter for enhancing CW performance is the choice of support media. Beyond providing structural support for macrophyte root systems and facilitating biofilm formation for biodegradation, the support media contribute to pollutant removal through adsorption processes [18]. This mechanism has been extensively studied for phosphorus removal in CWs [19-21]. Commonly studied support media include zeolite, activated carbon, and anthracite [22]. However, the role of support media in CWs treating commercial laundry GW has been insufficiently explored. Moreover, commercial laundry GW showed a higher concentration of PO_4^{3-}-P (> 20 mg/L), and thus, it is necessary to study different support media that remove this contaminant through the adsorption process in CWs. Dalahmeh et al. [23], for instance, investigated the adsorption capacities of bark, charcoal, and sand in biofilters treating mixed GW. Their results showed that charcoal outperformed bark and sand in removing COD, nitrogen, and phosphorus, likely due to its high porosity, carbon content, and density, making it a promising support media. Zeolites are crystalline microporous aluminosilicates widely used as adsorbents and ion exchangers in CWs. Due to its high porosity, zeolites have a large surface area where adsorption process occurs [24]. Zeolite has charged negative and from the substitution of cations, has excellent adsorption capacity for removing cations from water (e.g. ammonium (NH_4^+)), but lower affinities for anions (e.g. phosphate (PO_4^{3-})) [25]. However, the presence of NH_4^+ displaces cations such as Al, Fe, Mg, and Ca, and favors the precipitation of PO_4^{3-} with these anions following by the adsorption onto the zeolite surface [24]. Moreover, the adsorption capacity of zeolite is important to be studied because the saturation of support media increases during the operational time of CWs. For this reason, isotherm models are commonly applied and different studies showed that zeolite from CWs showed PO_4^{-3} adsorption capacity ranged between 0.007 – 7.128 mg/g [26-31].

Given the limited information about the treatment of commercial laundry GW in terms of the influence of operational and design parameters on enhancing CW performance and the necessity to optimize these systems to ensure a safe reuse, this study is focused on i) evaluating the impacts of different OLRs and HRTs on the performance of lab-sale VFSSFCWS treating commercial laundry GW; ii) determining the effects of two different support media (gravel and zeolite) on the removal efficiencies of contaminants of CWs and iii) evaluating the adsorption capacity of the zeolite used a support media in CWs in terms of PO_4^{3-}-P concentration, due to it higher concentration as contaminant in GW from commercial laundries. This is one of the first studies that focuses on enhancing CW's performance for treating commercial laundry GW and lays the groundwork for implementing these systems on-site at full scale in commercial laundry.

2. Materials and Methods

2.1. Design and operational characteristics of CWs

Two VSSFCWs were implemented at lab scale, with dimensions of 0.71 m x 0.15 m x 0.15 m (height, width, and length), providing a surface area of 0.47 m^2. One of the VSSFCWs was filled with gravel (Gra) and the other with zeolite (Zeo). The macrophyte species *Schoenoplectus californicus* was planted in both systems. For each system (Gra and Zeo), a replicate was constructed with identical design and operational parameters. The diameters and porosity of the zeolite and gravel comprising the support media were 1.81 mm and 19-25 mm, and 25% and 0.6%, respectively [32,33].

The influent used to feed the VSSFCW systems was collected from a commercial laundry located in downtown Concepción, Biobio Region, Chile. Table 1 shows the operational and design conditions of the CWs. Both systems underwent a 60-day setup stage before being operated in three distinct stages with varying OLRs, hydraulic loading rates (HLRs), and HRTs. During Stage I, the OLR, HLR, and HRT were 12.3 gCOD/m^2·d, 1.06 m^2/d and 10 d, respectively. In Stage II, these parameters were adjusted to 24.6 gCOD/m^2·d, 2.12 m^2/d, and 5 d, respectively. Finally, Stage III operated with an OLR of 49.9 gCOD/m^2·d, an HLR of 3.03 m^2/d, and an HRT of 3.5 d. The design and operational characteristics of VSSFCWs were based on the CW systems described in González et al. [33,34].

Table 1

2.2. Monitoring strategies

The CW systems were fed with GW from a commercial laundry using peristaltic pumps, and the effluents were collected through an outlet valve located 0.0015 m above the bottom zone, as shown in Figure 1. During the operating time (232 d), in situ parameters such as pH, temperature (T), reduction-oxidation potential (redox), total dissolved solids (TDS), turbidity, and electrical conductivity (EC) were measured daily. Physicochemical parameters in both influent and effluents were determined weekly, while biological oxygen demand (BOD_5), total nitrogen (TN), and total phosphorus were measured biweekly.

Figure 1

2.3. Analytic methods

In situ parameters including pH, T, redox, TDS, turbidity, EC and dissolved oxygen (DO), and physicochemical parameters such as COD (colorimetric method, 5220-D), BOD_5 (Modified Azid-Winkler method, 5210-B), ammonium-nitrogen (NH_4^+-N, colorimetric method at 640 nm, 4500-NH_3), nitrate-nitrogen (NO_3^--N, colorimetric method at 220 nm, 4500-NO_3), nitrite-nitrogen (NO_2^--N, colorimetric method at 543 nm, 4500-NO_2), TN (Spectroquant-Nova 60, kits Merck, Darmstadt, Germany), phosphorus-phosphate (PO_4^{3-}-P, colorimetric method at 890 nm, 4500-P-E) and TP (Spectroquant-Nova 60, kits Merck, Darmstadt, Germany) were measured using the protocols and equipment described by the Standard Methods [35]. For anions (chloride (Cl^-), nitrate (NO_3^-), nitrite (NO_2^-), phosphate (PO_4^{3-}), sulfate (SO_4^{2-}) and bromide (Br^-), and cations (ammonium (NH_4^+), lithium (Li^+), sodium (Na^+), potassium (K^+)) determinations were carried out using ionic chromatography (930 compact IC flex Methrhm, Herisau, Switzerland), for which the water samples were previously filtered through a 0.02-µm membrane filter. Removal efficiencies were determined with these concentrations in the influent and effluents from Gra and Zeo.

2.4.Adsorption/desorption analysis

As PO_4^{-3}-P is one of the principal pollutants in laundry GW due to the presence of detergent, the adsorption analysis of this compound was conducted. At the end of the operating time (232 d), 100 g of zeolite was extracted from different zones (top, medium, and bottom) of the Zeo to evaluate this material's adsorption/desorption capacity. The zeolite samples were agitated in 100 mL of distilled water at 100 rpm and 20°C for 20 h. Water samples were collected every hour, and the PO_4^{3-}-P concentration was measured until reaching equilibrium. Moreover, 50 g of zeolite that was not used in GW treatment was employed as a control. This analysis was based on the methods described by Alver et al. [36] and Araya et al. [37]. The concentration adsorbed/desorbed by the zeolite was calculated using Eq.1.

$$qe = {}^{(Co - Ce)V}/_{m}) \qquad \text{(Eq.1)}$$

Where, qe is adsorption capacity in mg/g, Co and Ce are the initial and final concentrations for a certain period (mg/L), V is the volume of solution in mL, and m is the zeolite mass in g used in the analysis.

The equilibrium of adsorption/desorption was evaluated using the Freundlich isotherm models (Eq.2).

$$qe = KfCe^{1/n} \quad \text{(Eq.2)}$$

Where Kf is the Freundlich constant related to the adsorption capacity of the adsorbent. Generally, the adsorption capacity of an adsorbent for a given adsorbate increases as Kf increases; $1/n$ is the Freundlich exponent related to surface heterogeneity.

2.5. Statistical analysis

The effects of OLRs, HRTs, and support media on VSSFCW performance were studied using RStudio version 4.4.2 and a significance level of $p = 0.05$. First, the data were tested for normality using the Shapiro-Wilk test and for homogeneity of variance using the Fligner-Killeen test. To determine the effects of different stages and support media, PERMANOVA analysis was performed. Finally, principal component analysis (PCA) was conducted using the principal functions from the "*ggfortify*" R package (https://cran.r-project.org/web/packages/ggfortify/index.html).

3. Results and Discussion

3.1. Influent Characterization

Table 2 shows the physicochemical characterization of the influent. In this GW from commercial laundry, the pH, COD, BOD_5, turbidity, TN, NH_4^+-N, TP and PO_4^{-3}-P concentrations fluctuated between 6.0 – 8.6, 176 – 907 mg/L, 93 – 210 mg/L, 14 – 215 NTU, 4.3 – 18 mg/L, 1.1 – 15 mg/L, 1.2 – 13 mg/L and 0.8 – 9.5 mg/L, with average values of 6.8, 473 mg/L, 150 mg/L, 110 mg/L, 13 mg/L, 8.3 mg/L, 4 mg/L and 2.5 mg/L, respectively. Regarding the characterization of commercial laundry GW in literature, the concentrations of different parameters were within the ranges reported by Li et al. [3], Punyapawar and Mutnuri [6], Shaik et al. [9], and Braga et al. [38] with pH, COD, BOD_5, turbidity, TN, NH_4^+-N, TP and PO_4^{-3}-P values ranged between 5.0 – 10, 58 – 4155 mg/L, 44 – 3330 mg/L, 34 – 510 NTU, 2.8 – 31 mg/L, 4.3 – 17 mg/L, 0.2 – 52 mg/L, and 0.5 – 24 mg/L. In the present study, the influent corresponds to GW from a commercial laundry shop and consequently, it had moderate concentrations of organic matter, turbidity, nitrogen and phosphorus. Although GW from laundry typically contains high concentrations of phosphorus due to its presence in many detergents (e.g., surfactants) [17]. The detergent used in this laundry shop was phosphate-free and biodegradable and can explain this trend. Moreover, the COD/BOD_5 ratio values varied between 1.9 – 4.3, with an average of 3.3, indicating that the influent is difficult to biodegrade. This could be attributed to the high content of recalcitrant compounds such as surfactants and personal care products, representing a challenge for biological treatment as CWs [11,39].

Table 2

Table 3 shows the ion characterization of the influent used in the CWs. Lower concentrations of NO_3^-, NO_2^-, PO_4^{3-}, and SO_4^{2-} ranged between 0.1 – 22 mg/L, were observed compared to those found by Ammari et al. [8], who reported concentrations that were approximately 10 to 470 times higher. This difference may be explained by the fact that their study used mixed GW, which includes water from baths, showers, bathroom sinks, laundry, and kitchen [2]. However, Na^+ and Cl^- concentrations were the highest in the present study, with values of 65 and 169 mg/L, respectively (Table 3). These levels could be related to the bleaching agents and disinfectants contained in detergents [40]. Regarding international guidelines, such as USEPA [41], this influent cannot be directly reused for urban or agricultural purposes due to the high turbidity and BOD concentrations (< 2 NTU and < 30 mg/L, respectively). For this reason, the application of treatment technologies such as CWs with optimized operational and design parameters is needed for removing pollutants.

Table 3

3.2. Effects of operational and design parameters on CW performance

Figure 2 shows the concentrations and removal efficiencies for organic matter, expressed as COD and BOD_5, and nutrients, expressed as TN and TP, in Zeo and Gra, respectively, during the different operational stages. Regarding the effects of operational parameters, in both VSSFCWs, the organic matter and nutrient removal efficiencies increased by 1.1 and 1.4 times, respectively, from Stage I to Stage III ($p < 0.05$). The increase in OLR from 12.3 to 49.9 $gCOD/m^2 \cdot d$ (from 15.6 to 43 $gCOD/m^3 \cdot d$), and at the same time the decrease in HRT from 10 to 3.5 d, generated an increase in organic matter and nutrient removal efficiencies to values between 79 – 88% and 40 – 60%, respectively. As mentioned above, the influent used in this study presented lower biodegradability with a COD/BOD_5 ratio of 3.3. As a consequence of increasing OLR, the availability of organic matter for heterotrophic bacteria of CWs also increased, stimulating the biodegradation of organic matter. Moreover, the changes of OLRs in both VSSFCWs were applied gradually, allowing the biomass present in both Gra and Zeo to adapt to fluctuations in organic matter. In the same line, optimal OLR values have been reported to be in the range of 10 – 60 $gCOD/m^2 \cdot d$ [27,42,43]. In this study, the OLR values of 12.3 to 49.9 $gCOD/m^2 \cdot d$ fall within that optimal range. In the case of TN, VSSFCWs promote nitrification, as their aerobic conditions facilitate the conversion of NH_4^+ to NO_3^- [44]. In both systems (Zeo and Gra), NH_4^+ was transformed into NO_3^-, with average effluent concentrations of 0.45 mg/L and 0.40 mg/L, respectively (see Table 3). Although nitrifying bacteria compete with heterotrophic bacteria for oxygen [44,45], TN removal was not affected by the increase in OLR.

The main mechanism for TP removal in CWs is adsorption [46], which will be discussed later. In this study, TP removal was enhanced by the increase in OLRs, and consequently, by the higher phosphorus load. In line with this, Carrillo et al. [47] investigated phosphorus removal in a pilot-scale CW for wastewater treatment and, through PCA, reported a positive correlation between TP removal and phosphorus loading rates. Other mechanisms contributing to phosphorus removal in CWs include biodegradation and plant uptake. However, these processes could not be analyzed in the present study. Several studies have described biological phosphorus removal by polyphosphate-accumulating organisms (PAOs) and phosphorus-solubilizing bacteria (PSBs) [48-51]. PAOs can assimilate phosphate and store it intracellularly, while PSBs convert inorganic phosphorus forms that are not readily available to plants into soluble and absorbable forms [50,51]. In this study, an increase in organic loads may stimulate the growth and activity of these bacteria, promoting phosphorus bioassimilation by both bacteria and plants, and thus enhancing TP. Nevertheless, future studies are needed to explore the microbial communities involved and their metabolic activity.

These findings contrast with those reported by Ramprasad and Philip [17] and Ramprasad et al. [11], who reported that organic matter and nutrient removal efficiencies decreased as the OLR increased from 15.8 to 35 $gCOD/m^3 \cdot d$. This discrepancy may be attributed to their use of sucrose as an external carbon source to enhance CW performance. Sucrose is readily degraded by microorganisms in water treatment technologies, potentially saturating their removal capacities.

Figure 2

As for the effects of support media on organic matter and nutrient removal efficiencies in each operational stage, Zeo performed better than Gra ($p < 0.05$), achieving 19 – 33% higher average BOD_5, COD, TN, and TP removal values. Vera et al. [28] observed a similar trend when comparing the performance of HSSFCWs filled with zeolite and gravel for treating wastewater. Moreover, various studies have demonstrated that CWs with zeolite as the support media achieve higher pollutant removal efficiencies, particularly for PO_4^{-3} and NH_4^+ [26,27,29,52]. Zeolite is a kind of silicate that has a large specific surface area, a strong adsorption and ion exchange capacity, and a composition of Ca, Al, and Fe oxides [29]. In CWs, zeolite plays a crucial role because it promotes filtration and adsorption processes, and the growth of the biofilm responsible for the biodegradation process [27]. The adsorption process can be physical (physisorption) or chemical (chemisorption) depending on the strength of the interaction between the adsorbate and adsorbent surface. Chemisorption is highly specific, occurs in a monolayer, and is irreversible, while physisorption is nonspecific, multilayered, and reversible. The latter is the mechanism commonly occurring in zeolite used in CWs [24]. In this study, the difference between Zeo and Gra can be related to the particle size and porosity of each support media. Zeolite has a diameter and porosity 10 – 14 times and 4 times higher than gravel, respectively. Thus, the Zeo system has a high surface area where the physisorption process can be carried out, showing higher performance of removing COD, BOD_5, TN, and TP. Regarding the average removal efficiencies of Zeo during Stage III, these values were within the ranges reported as optimal for CWs in the literature with COD, BOD_5, TN and TP removals of 85%, 86%, 29%, and 72%, respectively [1,3,6]. These results highlight the relevance of optimizing design and operational parameters for improving pollutant removal in CWs.

Figure 3 shows the EC concentrations and reductions in VSSFCW systems during the different stages. In contrast to the removal of organic matter and nutrients, EC reductions decreased from stage I to stage II by 7% and 17% in Gra and Zeo, respectively. However, reductions increased during stage III, reaching 0.19 and 0.45 in Gra and Zeo, respectively ($p < 0.05$). One possible explanation for this behavior is that the increase of OLRs, and consequently EC levels, from stage I and stage II may have negatively affected the macrophytes and biomass in both systems. Sepúlveda et al. [10] studied the effect of different EC levels on the performance of HSSFCWs planted with *Cyperus papyrus* and reported that elevated EC levels adversely affect both biomass and macrophyte growth in CWs. Nevertheless, the further increase in EC levels from stage II to stage III did not impact system performance. On the contrary, the reduction increased to 0.19 and 0.45, showing that VSSFCWs were adapted to EC fluctuations. Significant differences between the analyzed support media (zeolite and gravel) during all stages were also reported, with average removals of 21% and 10% in Zeo and Gra, respectively ($p < 0.05$). In other studies, Ammari et al. [8], Morandi et al. [4], Nema et al. [53], and Stefanatou et al. [54] observed negative EC removal efficiencies, indicating that the concentration increases throughout the treatment. This parameter is related to dissolved ions in water and is correlated with the evapotranspiration process in CWs during warm seasons (spring and summer) [10,54]. As for positive removal values in CWs, Arunbabu et al. [55] reported a performance close to 14%, similar to the results for Gra.

Figure 3

Regarding anion and cation removal efficiencies, it is important to mention that the values of the overall treatment were considered in both VSSFCWs (Table 3). In this case, some ion concentrations were below detection limits during different treatment stages. For NO_2^-, NO_3^-, NH_4^+, Na^+, and K^+, the removal efficiencies were 11 – 33% higher in Zeo than in Gra ($p < 0.05$). These results align with the trends observed for organic matter, nutrient, and EC removal values, further highlighting the positive impact of zeolite on CW performance. The highest removal values were achieved for NH_4^+ and PO_4^{3-}, with 63% and 71% in VSSFCW-Zeo, respectively. These results are within the range reported in the literature of 50 – 100% [8,29,53]. The dissolved PO_4^{3-} in CWs that microorganisms or plants do not assimilate tends to precipitate and be adsorbed by elements present in zeolite such as Fe, Ca, or Al [19,27]. In contrast, gravel as a support media has a lower adsorption capacity of nutrients, reporting values close to 0.02 mgP/g [22,28].

Figure 4a shows the principal component analysis (PCA) of VSSFCWs during the different stages. This analysis confirms the significant impact of operational stages on system performance, with principal components 1 and 2 (PC1 and PC2) explaining 63% and 25% of the total variance, respectively. Samples were spatially grouped according to operational stage (Stage I, II, and III). This tendency reflects the effects of increasing OLRs and decreasing HRTs on VSSFCW performance. The most different samples were those from Stage II. Generally, this stage presented the greatest differences with Stage I regarding removal efficiency ($p < 0.05$). For BOD_5 removals, the performance increased 30% from Stage I to Stage II in both VSSFCWs while from Stage II to Stage III, the increment was only close to 20%. In contrast, for EC removals, the performances decreased 20% from Stage I to Stage II and then increased 20% from Stage II to Stage III. The results demonstrated that the increase of OLRs from Stage I and Stage II had a crucial role in the overall performance of VSSFCWs systems. In the same line, PCA also showed strong associations between Stage II samples and CODs, CODt, and TDS concentrations. These concentrations were higher in Stage II than in Stages I and III, with average values of 667.9 mg/L, 583.5 mg/L, and 582.0 mg/L in influent samples, respectively (see Figure 2). Figure 4b showed the same spatial separation when GW samples were grouped as type of support media (Zeo and Gra). This result confirms that Zeo system improved the removal efficiency of VSSFCWs, obtaining better performances than Gra system. Zeolite as a support media played a critical role given the adsorption surface area for removing phosphorus, nitrogen, ions, and organic matter. However, this capacity is limited by the operational time that generates the saturation of the pores and micropores of the support media [29]. This phenomenon will be discussed in the next section.

Based on the results of this study, the VSSFCW system operated at OLR of 49.9 $g{\cdot}COD/m^2{\cdot}d$, with an HRT of 3.5 d and using zeolite as a support media is the system, achieved the highest removal efficiencies for COD, BOD_5, TN and TP with values of 85%, 85%, 29% and 72%, respectively. In comparison, Punyapwar and Mutnuri [6] implemented two VSSFCWs preceded by an anaerobic filter for treating GW from commercial laundry and reported organic matter and nutrients removal efficiencies of VSSFCWs ranging between 33 -36% and 16 – 25%, respectively. These results suggest that the selected VSSFCW configuration (Zeo system with OLR and HRT of 49.9 $g{\cdot}COD/m^2{\cdot}d$ and 3.5 d) could be a viable treatment option for commercial laundry GW. Notably, this study is among the first to optimize CW for this specific application. The results highlight the relevance of evaluating operational and design parameters in system performance. However, it is important to note that the study was conducted at a lab scale, where environmental and operational variables were controlled. Therefore, future research should focus on scaling up the VSSFCW prototype and assessing its performance under real conditions for treating commercial laundry GW.

Figure 4

3.3. Sorption Analysis in Zeo

Given the superior contaminant removal efficiencies achieved by Zeo, a sorption analysis was conducted to evaluate the PO_4^{3-}-P adsorption capacity of the zeolite. Moreover, the detergents used in commercial laundry are commonly composed of surfactants, builders, and enzymes that contain higher PO_4^{3-}-P concentrations [9]. For this reason, the adsorption capacity of this specific parameter was investigated in this study. This capacity was measured across different zones within Zeo (top: Zeo-Top; middle: Zeo-Middle; and bottom: Zeo-Bottom).

Figure 5 and Table 4 show the Freundlich isotherm models and sorption constants of the zeolite in Zeo, respectively. According to this model, a higher *Kf* value indicates greater adsorption capacity, while 1/n values above 1 suggest a heterogeneous material [29]. The 1/n values obtained for the zeolites confirm their heterogeneity, with all values exceeding 1.0. When compared to the results of Zeo-Control, which was not used for GW treatment, the zeolite's adsorption capacity decreased during the treatment. Zeo-Control presented *Kf* values of 0.006 mg/g, while Zeo-CW exhibited lower values of 0.004 mg/g. Furthermore, the zeolite from different CW zones (Zeo-Top, Zeo-Middle, and Zeo-Bottom) did not present differences, with *Kf* values varying between 0.001 – 0.002 mg/g. In previous studies, the sorption capacities of zeolite from CWs resulted in *Kf* ranges between 0.007 – 7.128 mg/g [26-31]. The lower values observed here may be attributed to the fact that most of these studies employed modified zeolite through thermal and chemical treatments to enhance adsorption capacity [26,30]. Moreover, zeolite has charged negative and has lower affinity for anions (PO_4^{3-}). However, the presence of NH_4^+ displaces cations such Al, Fe, Mg, and Ca, and favors the precipitation of PO_4^{3-} with these anions following by the adsorption onto the zeolite surface [24,25]. For future studies, it would be interesting to analyze the zeolite's adsorption capacity with a mixed solution of NH_4^+-N and PO_4^{-3}-P.

Additionally, the sorption analyses were performed at the end of the operational period (232 d), when the Zeo-CW was already saturated. These results confirm what has been reported in the literature: Zeolite loses its ability to retain and release PO_4^{-3}-P during the operational time of CWs. This process may be accelerated with the increase of OLRs and, therefore, the PO_4^{-3}-P loads, which raised from 2.6 to 7.6 g·PO_4^{3-}-P/m^2·d. With the results obtained in adsorption analysis on zeolite, the physisorption mechanism dominated the PO_4^{3-}-P removal in this support media. Due to its small particle size (1.81 mm) and 25% of porosity, zeolite possesses a large surface area for interaction between PO_4^{-3}-P and the active sites. Once all the active sites of the zeolite are saturated, PO_4^{-3}-P is released into the medium, resulting in a desorption process. This demonstrates that the interactions between substrate and adsorbent are weak and reversible [24].

Figure 5

Despite the results of this study showed that VSSFCWs with an OLR and HRT of 49.9 g·COD/m^2·d and 3.5 d, respectively, and filled with zeolite as support media had the better performance and could be an alternative for treating commercial laundry GW, more studies are needed for assuring a safe reuse with no risks for human health and the environment. In this line, it would be interesting to focus on the use of modified zeolite in CWs treating GW for enhancing the adsorption of PO_4^{3-}-P and other anions and therefore, improving the performance of these treatment systems. Moreover, the occurrence of pathogenic microorganisms such as bacteria, protozoa, and viruses should be incorporated in the CW performance analysis for determining the use of a disinfection process on the GW treatment.

Although several aspects still require further investigation, this study provides a valuable foundation for improving the treatment quality of commercial laundry GW. Despite being a relatively underexplored topic, laundry GW represents a promising resource for reuse in various applications, including urban practices (e.g., irrigation of green spaces or reuse within the laundry process) and agricultural activities. This study offers initial guidelines emphasizing the importance of evaluating key operational parameters such as OLR, HRT, and the type of support media, to enhance treatment performance and promote safe reuse.

Table 4

4. Conclusions

This study is among the first to optimize CWs for the treatment of GW from commercial laundry by evaluating key operational parameters (OLR and HRT) alongside design factors such as the type of support media. The results demonstrated that higher OLR (49.9 g $COD/m^2·d$) combined with a shorter HRT (3.5 d) significantly improved the removal efficiencies of COD, BOD_5, TN, and TP in both VSSFCW systems (Zeo and Gra), achieving removal rates of 73–85%, 70–86%, 21–29%, and 34–72%, respectively ($p < 0.05$). The type of support media also played a critical role in system performance. The Zeo system outperformed the Gra system, with organic matter and nutrient removal efficiencies 19–33% higher. However, the phosphate sorption capacity of zeolite decreased over time, reaching a saturation of 0.04 mg/g, likely due to the increasing PO_4^{3-}-P loads. This highlights the need for future research on media regeneration or replacement strategies. Overall, the VSSFCW system operated at 49.9 g $COD/m^2·d$ and 3.5 days HRT using zeolite as the support medium achieved the highest removal efficiencies for COD (85%), BOD_5 (85%), TN (29%), and TP (72%). Compared to existing studies, this system demonstrated superior performance and presents a promising prototype for the decentralized treatment of commercial laundry GW.

Although conducted at laboratory scale under controlled conditions, these findings establish a strong foundation for future full-scale implementation. Scaling up this system will require further research into enhancing zeolite adsorption capacity and exploring the microbial communities involved to better understand the biological and physicochemical mechanisms behind contaminant removal. Importantly, this study contributes to advancing sustainable wastewater management by demonstrating that optimized CW systems can effectively improve water quality and support safe water reuse. Adjusting operational and design parameters to specific GW sources is essential for achieving reliable treatment outcomes, protecting public health, and reducing the environmental footprint of wastewater discharge.

CRediT authorship contribution statement

Gutiérrez, J.: Investigation, Methodology, Writing- original draft; **Leiva. A.M.:** Validation, Conceptualization, Writing – review & editing, Data curation; **Gómez, G.:** Validation, Conceptualization; **Vidal, G.:** Validation, Conceptualization, Writing – review & editing, Supervision, Resources.

Declaration of Competing Interest

The authors declare that there are no competing financial interests on the publication of this article.

Data availability

Data are contained within the article.

Acknowledgements

This study was funded by an ANID/FONDAP/1523A0001 grant.

References

[1] N. Nurmahomed, T. Sobhun, A.K. Ragen, C.M. Sheridan, Performance of a constructed wetland treating synthetic greywater, Bioresour Technol Rep (2022) 17, 100930. https://doi.org/10.1016/j.biteb.2021.100930

[2] S. Arden, X. Ma, Constructed wetlands for greywater recycle and reuse: A review, Sci Total Environ (2018) 630, 587–599. https://doi.org/10.1016/j.scitotenv.2018.02.218

[3] Y. Li, S. Zhu, Y. Zhang, M. Lv, J.R. Kinhoun, T. Qian, B. Fan, Constructed wetland treatment of source separated washing wastewater in rural areas of southern China, Sep Purif Technol (2021) 272, 118725. https://doi.org/10.1016/j.seppur.2021.118725

[4] C. Morandi, G. Schreiner, P. Moosmann, H. Steinmetz, Elevated Vertical-Flow Constructed Wetlands for Light Greywater Treatment, Water (2021) 13, 2510. https://doi.org/10.3390/w13182510

[5] U. Uthirakrishnan, V. Manthapuri, A. Harafan, P.V. Chellam, T. Karuppiah, The regime of constructed wetlands in greywater treatment, Water Sci Technol (2022) 85, 3169–3183. https://doi.org/10.2166/wst.2022.159

[6] S. Punyapwar, S. Mutnuri, Laundry greywater treatment by anaerobic filters and vertical flow constructed wetlands, Water Environ J (2022) 36, 504–512. https://doi.org/10.1111/wej.12782

[7] E. Comino, V. Riggio, M. Rosso, Grey water treated by an hybrid constructed wetland pilot plant under several stress conditions, Ecol Eng (2013) 53, 120–125. https://doi.org/10.1016/j.ecoleng.2012.11.014

[8] T.G. Ammari, Y. Al-Zu'bi, A. Al-Balawneh, R. Tahhan, M. Al-Dabbas, R.A. Ta'any, R. Abu-Harb, An evaluation of the re-circulated vertical flow bioreactor to recycle rural greywater for irrigation under arid Mediterranean bioclimate, Ecol Eng (2014) 70, 16–24. https://doi.org/10.1016/j.ecoleng.2014.03.009

[9] I. N. Shaikh, I. N., M. M. Ahammed, Quantity and quality characteristics of greywater: a review, J Environ Manag (2020), 261, 110266. https://doi.org/10.1016/j.jenvman.2020.110266

[10] R. Sepúlveda, A.M. Leiva, G. Vidal, Performance of Cyperus papyrus in constructed wetland mesocosms under different levels of salinity, Ecol Eng (2020) 151, 105820. https://doi.org/10.1016/j.ecoleng.2020.105820

[11] C. Ramprasad, C.S. Smith, F.A. Memon, L. Philip, Removal of chemical and microbial contaminants from greywater using a novel constructed wetland: GROW, Ecol Eng (2017) 106, 55–65. https://doi.org/10.1016/j.ecoleng.2017.05.022
[12] M. Sotiropoulou, A. Stefanatou, S. Schiza, I. Petousi, A.S. Stasinakis, M.S. Fountoulakis, Removal of microfiber in vertical flow constructed wetlands treating greywater, Sci Total Environ (2023) 858, 159723. https://doi.org/10.1016/j.scitotenv.2022.159723
[13] C. Dykes, J. Pearson, G. Bending, S. Abolfathi, Impact of seasonal climate variability on constructed wetland treatment efficiency. J Water Process Eng (2025) 72, 107350. https://doi.org/10.1016/j.jwpe.2025.107350
[14] V. Burgos, F. Araya, C. Reyes-Contreras, I. Vera, G. Vidal, Performance of ornamental plants in mesocosm subsurface constructed wetlands under different organic sewage loading, Ecol Eng (2017) 99, 246–255. https://doi.org/10.1016/j.ecoleng.2016.11.058
[15] H. Ilyas, I. Masih, E.D. van Hullebusch, Prediction of the removal efficiency of emerging organic contaminants based on design and operational parameters of constructed wetlands, J Environ Chem Eng (2021), 9(1), 104592. ttps://doi.org/10.1016/j.jece.2020.104592
[16] D. Minakshi, P.K. Sharma, A. Rani, P. Malaviya, V. Srivastava, M. Kumar, Performance evaluation of vertical constructed wetland units with hydraulic retention time as a variable operating factor, Groundw Sustain Dev (2022), 19, 100834. https://doi.org/10.1016/j.gsd.2022.100834
[17] C. Ramprasad, L. Philip, Surfactants and personal care products removal in pilot scale horizontal and vertical flow constructed wetlands while treating greywater, Chem Eng J (2016), 284, 458–468. https://doi.org/10.1016/j.cej.2015.08.092
[18] L.C. Sandoval Herazo, A. Alvardo-Lassman, J.L. Marín-Muñiz, J.P. Rodríguez-Miranda, G. Fernández-Lambert, A critical review of mineral substrates used as filter media in subsurface constructed wetlands: costs as a selection criterion, Environ Technol Rev (2023) 12, 251–271. https://doi.org/10.1080/21622515.2023.2198146
[19] V. Carrillo, A.M. Leiva, G. Vidal, Constructed Wetlands: An Emerging Green Technology for the Phosphorus Treatment in Industrial Wastewaters, in: Constructed Wetlands: An Emerging Green Technology for the Treatment of Industrial

Wastewaters. Springer, Singapore, (2019) pp. 21–44. https://doi.org/10.1007/978-981-15-1390-9_2

[20] R. Fahim, L. Cheng, S. Mishra, Structural and functional perspectives of carbon filter media in constructed wetlands for pollutants abatement from wastewater, Chemosphere (2023) 345, 140514. https://doi.org/10.1016/j.chemosphere.2023.140514

[21] C. Vispo, F.K. Geronimo, M. Jeon, L.H. Kim, Performance Evaluation of Various Filter Media for Multi-Functional Purposes to Urban Constructed Wetlands, Sustainability (2024) 16. https://doi.org/10.3390/su16010287

[22] V. Carrillo, B. Fuentes, G. Gómez, G. Vidal, Characterization and recovery of phosphorus from wastewater by combined technologies, Rev Environ Sci Biotechnol (2020) 19, 389–418. https://doi.org/10.1007/s11157-020-09533-1

[23] S.S. Dalahmeh, M. Pell, L.D. Hylander, C. Lalander, B. Vinnerås, H. Jönsson, Effects of changing hydraulic and organic loading rates on pollutant reduction in bark, charcoal and sand filters treating greywater, J Environ Manage (2014) 132, 338–345. https://doi.org/10.1016/j.jenvman.2013.11.005

[24] E. Andrés, F. Araya, I. Vera, G. Pozo, G. Vidal, Phosphate removal using zeolite in treatment wetlands under different oxidation-reduction potentials. Ecol Eng (2018) 117, 18-27. https://doi.org/10.1016/j.ecoleng.2018.03.008

[25] D. López, D. Fuenzalida, I. Vera, K. Rojas G. Vidal, Relationship between the removal of organic matter and the production of methane in subsurface flow constructed wetlands designed for wastewater treatment. Ecol Eng (2015) 83, 296-304. https://doi.org/10.1016/j.ecoleng.2015.06.037

[26] H. He, Z. Duan, Z. Wang, B. Yue, The removal efficiency of constructed wetlands filled with the zeolite-slag hybrid substrate for the rural landfill leachate treatment, Environ Sci Pollut Res (2017) 24, 17547–17555. https://doi.org/10.1007/s11356-017-9402-x

[27] A.I. Stefanakis, V.A. Tsihrintzis, Effects of loading, resting period, temperature, porous media, vegetation and aeration on performance of pilot-scale vertical flow constructed wetlands, Chem Eng (2012) J 181–182, 416–430. https://doi.org/10.1016/j.cej.2011.11.108

[28] I. Vera, F. Araya, E. Andrés, K. Sáez, G. Vidal, Enhanced phosphorus removal from sewage in mesocosm-scale constructed wetland using zeolite as medium and artificial

aeration, Environ Technol (2014) 35, 1639–1649. https://doi.org/10.1080/09593330.2013.877984

[29] H. Wang, J. Xu, L. Sheng, Purification mechanism of sewage from constructed wetlands with zeolite substrates: A review, J Clean Prod (2020) 258, 120760. https://doi.org/10.1016/j.jclepro.2020.120760

[30] Y. Zhang, J. Dong, F. Guo, Z. Shao, J. Wu, Zeolite Synthesized from Coal Fly Ash Produced by a Gasification Process for Ni2+ Removal from Water, Minerals (2018) 8, 116. https://doi.org/10.3390/min8030116

[31] J. Zhao, Y. Zhao, Z. Xu, L. Doherty, R. Liu, Highway runoff treatment by hybrid adsorptive media-baffled subsurface flow constructed wetland, Ecol Eng (2016) 91, 231–239. https://doi.org/10.1016/j.ecoleng.2016.02.020

[32] A.M. Leiva, R. Núñez, G. Gómez, D. López, G. Vidal, Performance of ornamental plants in monoculture and polyculture horizontal subsurface flow constructed wetlands for treating wastewater. Ecol Eng (2018), 120, 116-125. https://doi.org/10.1016/j.ecoleng.2018.05.023

[33] T. González, J. Puigagut, G. Vidal, Organic matter removal and nitrogen transformation by a constructed wetland-microbial fuel cell system with simultaneous bioelectricity generation. Sci Total Environ (2021) 753, 142075. https://doi.org/10.1016/j.scitotenv.2020.142075

[34] T. González, J.P. Miranda, G. Gómez, J. Puigagut, G. Vidal, Saturated constructed wetland-microbial fuel cell system and effect on dissolved oxygen gradient, electricity generation and ammonium removal. Environ Technol (2024), 45(4), 624-638. https://doi.org/10.1080/09593330.2022.2119170

[35] APHA, Standard Methods for the Examination of Water and Wastewater, American Public Health Association, (2005) Washington, DC, USA. https://doi.org/10.1088/1751-8113/44/8/085201

[36] E. Alver, A.Ü. Metin, Anionic dye removal from aqueous solutions using modified zeolite: Adsorption kinetics and isotherm studies, Chem Eng J (2012) 200–202, 59–67. https://doi.org/10.1016/j.cej.2012.06.038

[37] F. Araya, I. Vera, K. Sáez, G. Vidal, Effects of aeration and natural zeolite on ammonium removal during the treatment of sewage by mesocosm-scale constructed

wetlands, Environ Technol (2016) 37, 1811–1820. https://doi.org/10.1080/09593330.2015.1133715

[38] J.K. Braga, M.B.A. Varesche (2014). Commercial laundry water characterisation. Americ J Anal Chem (2014), 5(1), 8-16. https://doi.org/10.4236/ajac.2014.51002

[39] Metcalf and Eddy, Wastewater Engineering: Treatment and Reuse, (2003) McGraw-Hil. ed. New York, USA.

[40] A. Van de Walle, M. Kim, M.K. Alam, X. Wang, D. Wu, S.R. Dash, K. Rabaey, J. Kim, Greywater reuse as a key enabler for improving urban wastewater management, Environ Sci Ecotechnol (2023) 16, 100277. https://doi.org/10.1016/j.ese.2023.100277

[41] USEPA, Guidelines for Water Reuse, EPA/600/R-12/004, USEPA Office of Wastewater Management, Washington DC, 2012.

[42] W.H. Bang, Y. Jung, J.W. Park, S. Lee, S.K. Maeng, Effects of hydraulic loading rate and organic load on the performance of a pilot-scale hybrid VF-HF constructed wetland in treating secondary effluent, Chemosphere (2019) 218, 232–240. https://doi.org/10.1016/j.chemosphere.2018.11.110

[43] H. Ilyas, I. Masih, The performance of the intensified constructed wetlands for organic matter and nitrogen removal: A review, J Environ Manage (2017) 198, 372–383. https://doi.org/10.1016/j.jenvman.2017.04.098

[44] T. Liu, D. Li, Y. Tian, J. Zhou, Y. Qiu, D. Li, G. Liu, Y. Feng, Enhancing nitrogen removal in constructed wetlands: The role of influent substrate concentrations in integrated vertical-flow systems. Environ Sci Ecotechnol (2024) 21, 100411. https://doi.org/10.1016/j.ese.2024.100411

[45] A. M. Leiva, C. Reyes-Contreras, C., G. Vidal, G. Influence of Agapanthus africanus on nitrification in a vertical subsurface flow constructed wetland. Int J Phytorem (2018), 20(6), 530-537. https://doi.org/10.1080/15226514.2017.1393390

[46] V. Verma, A. Soti, N.M. Kulshreshtha, A. Rampuria, U. Brighu, A.B. Gupta, Strategies for enhancing phosphorous removal in vertical flow constructed wetlands. J Environ Manag (2022) 317, 115406. https://doi.org/10.1016/j.jenvman.2022.115406

[47] V. Carrillo, C. Collins, J. Brisson, G. Vidal, Evaluation of long-term phosphorus uptake by Schoenoplectus californicus and Phragmites australis plants in pilot-scale

constructed wetlands. Int J Phytorem (2022), 24(6), 610-621. https://doi.org/10.1080/15226514.2021.1960478

[48] S. Kim, K. Gil, Evaluation of Phosphate Solubilization Microbe (PSM) Activity During Initial Operation of Constructed Wetland. Wetlands (2024) 44(4), 40. https://doi.org/10.1007/s13157-024-01795-7

[49] M. Ji, J. Zhang, S. Wang, S.Liang, Z. Hu, Enhanced phosphorus removal of constructed wetland through plant growth-promoting rhizobacteria (PGPR) addition. Environ Sci Pollut Res (2021) 28(37), 52124-52132. https://doi.org/10.1007/s11356-021-14364-w

[50] S. Chu, F. Zhao, Y. Liu, J. Xiao, X. Ma, Z. Yan, Phosphorus activation from phosphorus-saturated substrates by constructed wetlands plants: Performance and mechanism. J Water Process Eng (2025) 71, 107219. https://doi.org/10.1016/j.jwpe.2025.107219

[51] L. Du, Q. Chen, P. Liu, X. Zhang, H. Wang, Q. Zhou, D. Xu, Z. Wu, Phosphorus removal performance and biological dephosphorization process in treating reclaimed water by Integrated Vertical-flow Constructed Wetlands (IVCWs). Bioresour Technol (2017) 243, 204-211. https://doi.org/10.1016/j.biortech.2017.06.092

[52] W. Xu, B. Yang, W. Hui, S. Wang, K. Jiao, C. Zhang, F. Li, W. Haixia, Improving the removal efficiency of nitrogen and organics in vertical-flow constructed wetlands: the correlation of substrate, aeration and microbial activity, Environ Sci Pollut Res (2022) 30, 21683–21693. https://doi.org/10.1007/s11356-022-23746-7

[53] A. Nema, R. Prasad, D. Sharma, K.D. Yadav, R.A. Christian, H. Ibrahim, Performance evaluation of different macrophytes in small-scale vertical flow constructed wetlands for greywater treatment using principal component analysis, Can J Chem Eng (2023) 101, 1321–1334. https://doi.org/10.1002/cjce.24527

[54] A. Stefanatou, L. Vouzi, I. Petousi, A. Koukoura, G. Gatidou, A.S. Stasinakis, M.S. Fountoulakis, Treatment of real laundry wastewater using vertical flow constructed wetland planted with the ornamental climbing plant Trachelospermum jasminoides: assessing the removal of conventional pollutants and benzotriazoles, Environ Sci Pollut Res (2024) 31, 43281–43291. https://doi.org/10.1007/s11356-024-34035-w

[55] V. Arunbabu, S. Sruthy, I. Antony, E.V. Ramasamy, Sustainable greywater management with Axonopus compressus (broadleaf carpet grass) planted in sub surface flow constructed wetlands, J Water Process Eng (2015) 7, 153–160. https://doi.org/10.1016/j.jwpe.2015.06.004

Figure Caption

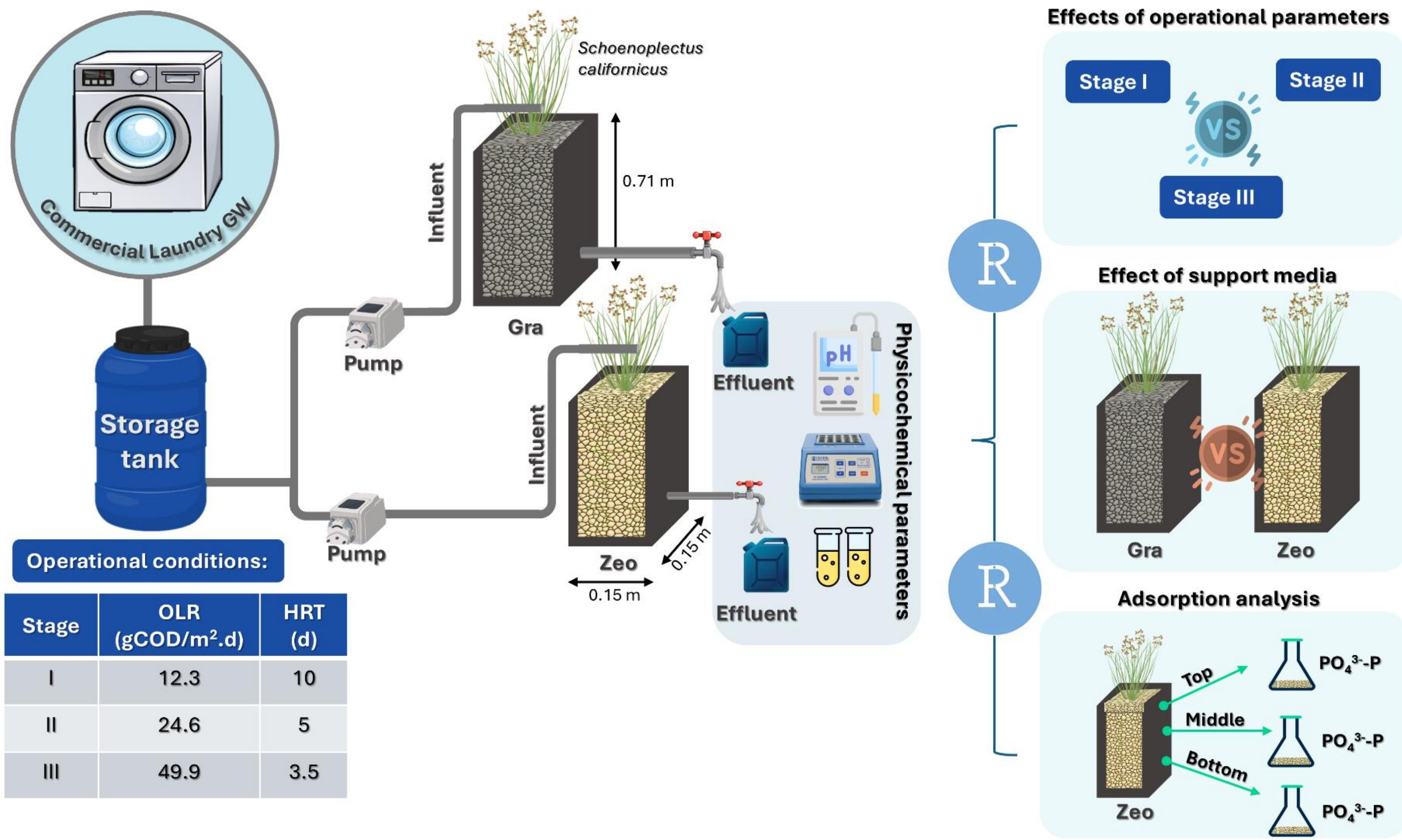
Figure 1. Schematic diagram of VSSFCW treating greywater.

Figure 2. Concentration (bar) and removal efficiency (scatter) of organic matter as **a)** COD; **b)** BOD_5, **c)** TN and **d)** TP. Where ■ Influent, ■ Zeo effluent and ■ Gra effluent.

Figure 3. Concentration (box pot) and reduction (bar, ΔC: difference between initial and final concentration of CE; Ci; initial concentration) of electrical conductivity. Where ■ influent, ■ Zeo effluent and ■ Gra effluent .

Figure 4. Principal component analyses (PCA): **a)** samples in represent Stage I, Stage II, and Stage III, and **b)** samples in represent Zeo and Gra systems.

Figure 5. Freundlich Isotherm models for the desorption of phosphorus as phosphate (PO_4^{-3}-P) on zeolite from VSSFCW-Zeo (Zeo-CW: zeolite samples from all zones of VSSFCW-Zeo; Zeo-Top: zeolite samples from the top zone of Zeo; Zeo-Middle: zeolite samples from the middle zone of Zeo; Zeo-Bottom: zeolite samples from the bottom zone of Zeo: Zeo-control: zeolite samples used as a control that was not used in greywater treatment.



Stage	OLR (gCOD/m².d)	HRT (d)
I	12.3	10
II	24.6	5
III	49.9	3.5

Figure 1

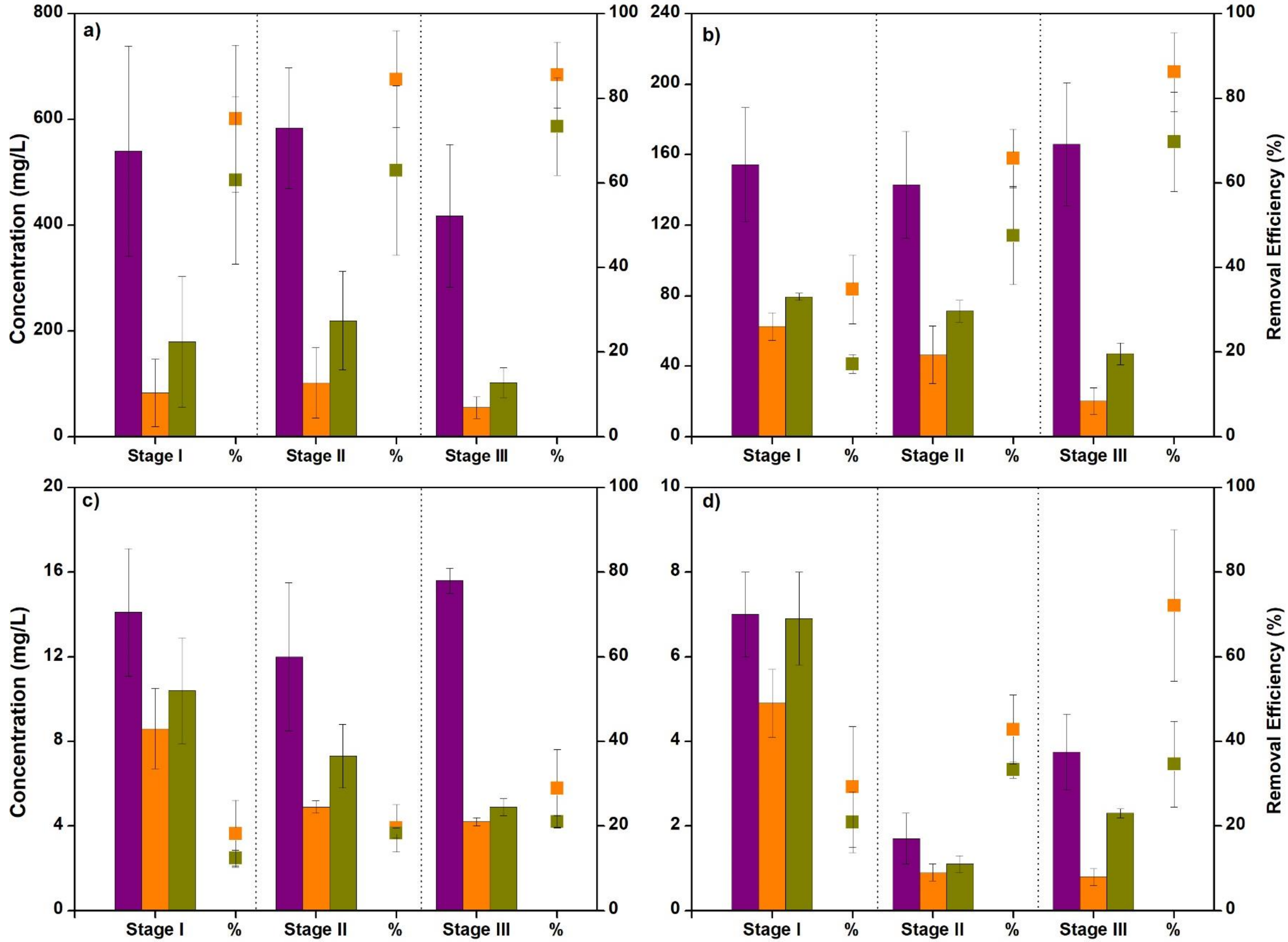

a)
b)
c)
d)
Concentration (mg/L)
Removal Efficiency (%)
Stage I
Stage II
Stage III
%

Figure 2

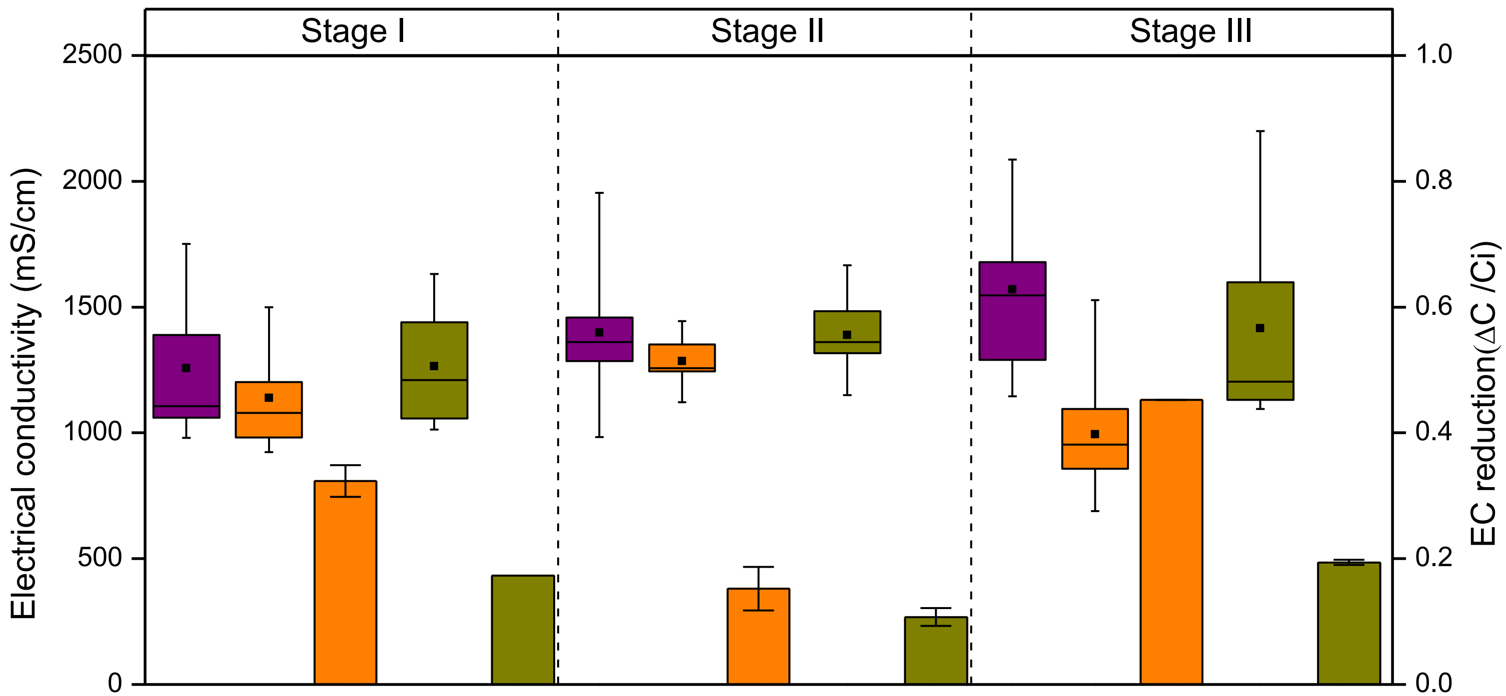

Stage I
Stage II
Stage III
Electrical conductivity (mS/cm)
EC reduction(ΔC /Ci)
2500
2000
1500
1000
500
0
1.0
0.8
0.6
0.4
0.2
0.0

Figure 3

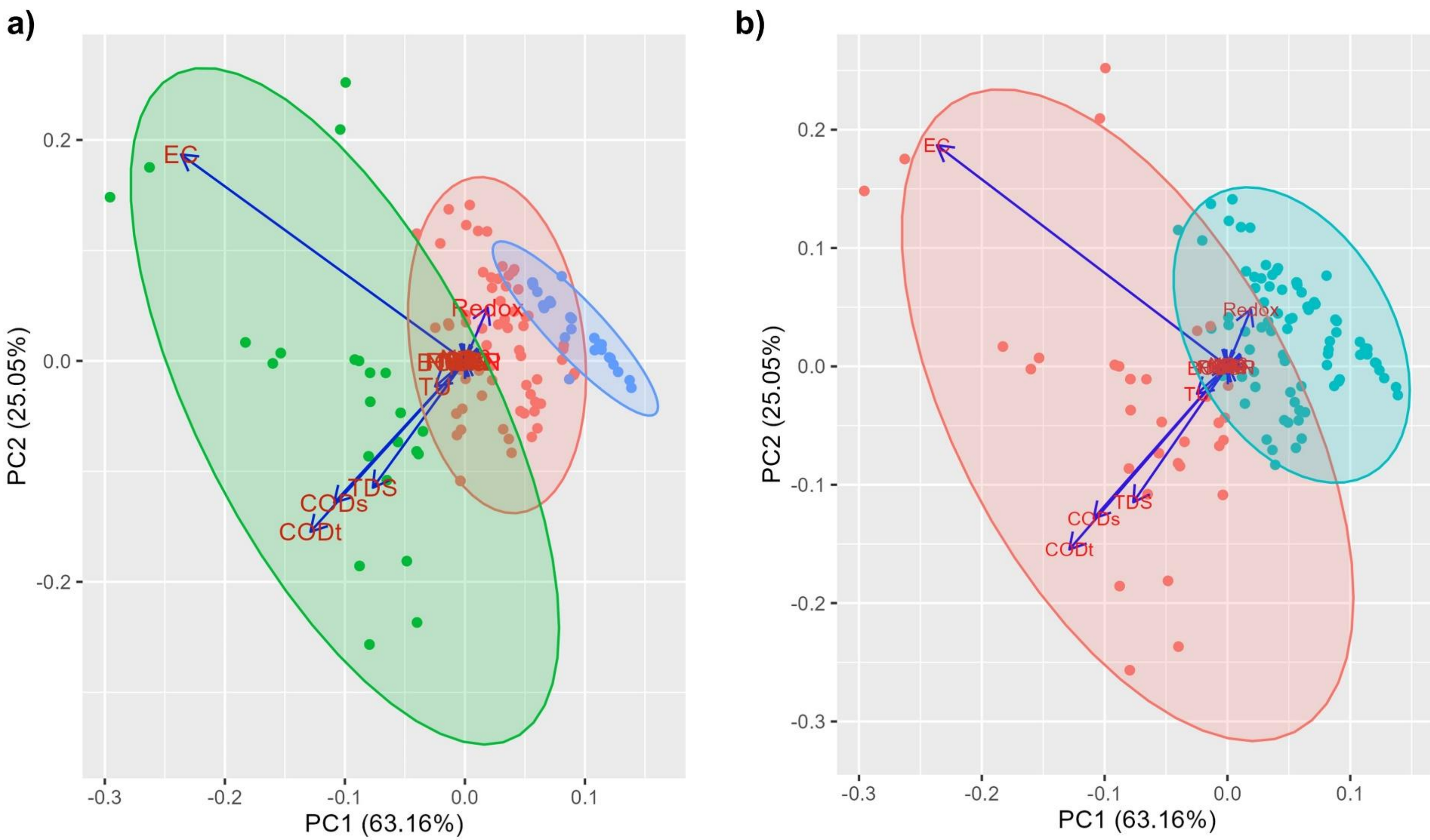


Figure 4

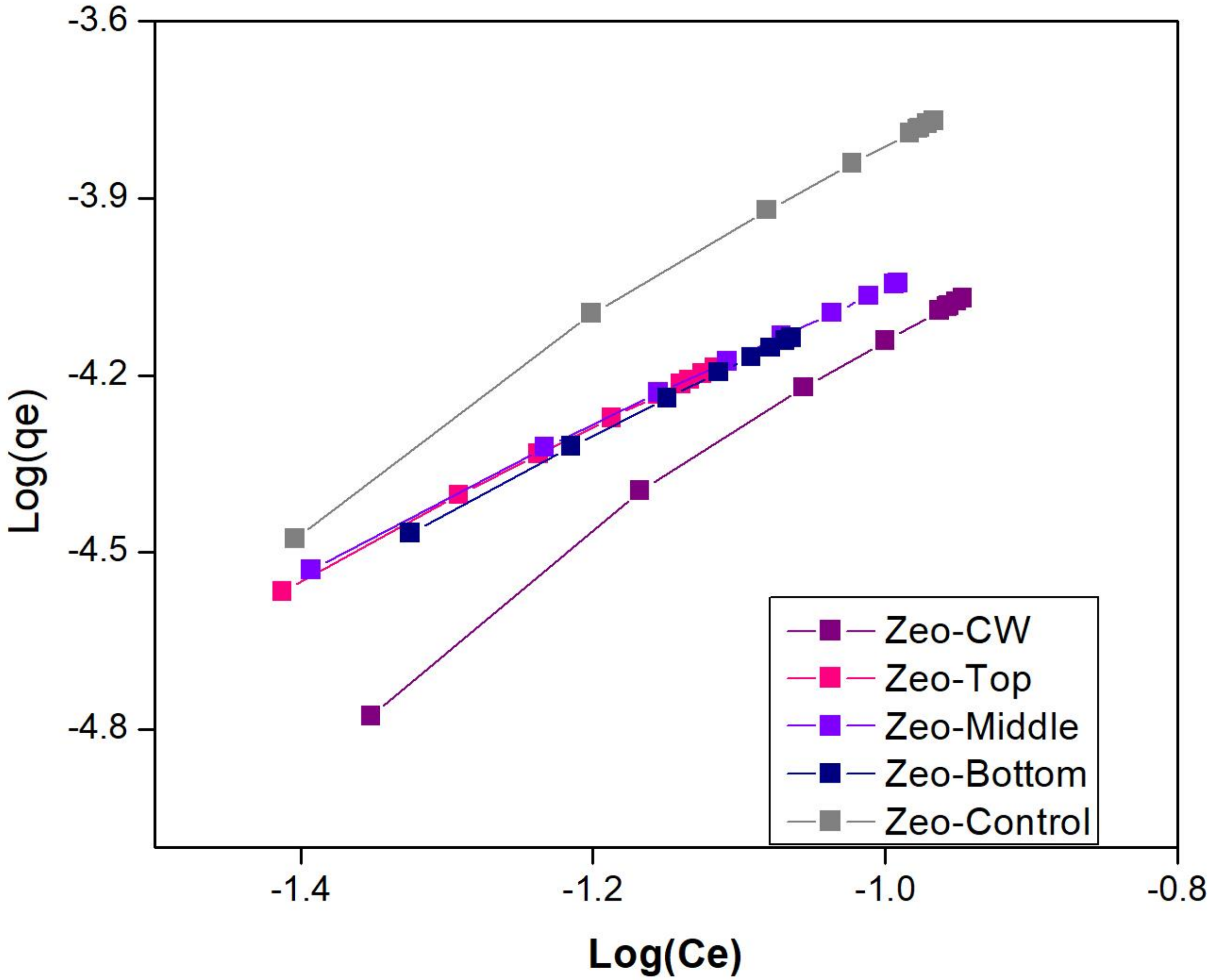

-3.6
-3.9
-4.2
-4.5
-4.8
Log(qe)
-1.4
-1.2
-1.0
-0.8
Log(Ce)
Zeo-CW
Zeo-Top
Zeo-Middle
Zeo-Bottom
Zeo-Control

Figure 5

Table Captions

Table 1. Operational strategies of Zeo and Gra

Table 2. Physicochemical characterization of the influent

Table 3. Ion characterization of the influent and effluents of different constructed wetlands

Table 4. Freundlich desorption constants of zeolite from Zeo.

Parameter	**Unit**	**Acclimation**	**Stage I**	**Stage II**	**Stage III**
Operating time	d	60	69	50	35
Water flow	L/d	0.5	0.5	1.0	1.4
HRT	d	10	10	5	3.5
OLR	$gCOD/m^2 \cdot d$	12.3	12.3	24.6	49.9
HLR	m/d	1.1	1.1	2.1	3.0

Note: HRT: Hydraulic retention time; OLR: Organic loading rate; HLR: Hydraulic loading rate.

Table 1

Parameter	Unit	Range	Average ± SD
pH	-	6.04 – 8.57	6.79 ± 0.55
T	°C	9.30 – 19.80	15.13 ± 3.30
Redox	mV	-128.60 – 151.60	27.60 ± 97.45
DO	mg/L	0.50 – 4.86	1.77 ± 1.10
EC	μS/cm	979.50 – 2826.00	1521.12 ± 485.91
TDS	mg/L	122.00 – 728.70	473.49 ± 144.70
Turbidity	NTU	13.97 – 215.00	110.07 ± 48.22
BOD_5	mg/L	93.75 – 210.00	150.41 ± 47.90
COD	mg/L	175.98 – 907.05	492.95 ± 155.70
TN	mg/L	4.30 – 18.00	13.50 ± 3.85
NH_4^+-N	mg/L	1.07 – 14.73	8.28 ± 4.27
TP	mg/L	1.20 – 12.60	3.88 ± 4.03
PO_4^{3-}-P	mg/L	0.76 – 9.50	2.51 ± 1.98

Note: T: Temperature; Redox: Oxidation-reduction potential; DO: Dissolved oxygen; EC: Electrical conductivity; TDS: Total dissolved solids; BOD_5: Biological oxygen demanded; COD: Chemical oxygen demand; TN: Total nitrogen; TP: Total phosphorus; NH_4^+-N: Nitrogen of ammoniacal nitrogen; PO_4^{-3}-P: Phosphorus of phosphate.

Table 2

Ion concentration in influent and both VSSFCWs						
Ion	**Unit**	**Influent**	**Zeo**	**Removal efficiencies (%)**	**Gra**	**Removal efficiencies (%)**
Anions						
NO_2^-	mg/L	0.19±0.09	0.17±0.11	10*	0.15±0.08	21*
NO_3^-	mg/L	0.33±0.23	0.45±0.25	-	0.40±0.22	-
SO_4^{2-}	mg/L	22.04±6.59	19.18±17.04	13	19.47±17.65	12
PO_4^{3-}	mg/L	0.70±0.52	0.26±0.18	63*	0.37±0.27	47*
Cl^-	mg/L	168.80±66.94	141.22±129.94	16	143.59±141.78	15
Cations						
NH_4^+	mg/L	5.52±2.94	1.61±1.50	71*	3.37±3.08	39*
Na^+	mg/L	64.75±19.18	42.34±15.48	35*	54.20±13.04	16*
K^+	mg/L	6.11±4.58	3.91±1.41	36*	5.93±3.55	3*

Notes: NO_2^-: nitrite; NO_3^-: nitrate; SO_4^{2-}: sulphate; PO_4^{3-}: phosphate; Cl^-: chloride; NH_4^+: ammonium; Na^+: sodium; K^+: potassium; " – ": negative value; * significant difference between VSSFCW-Zeo and VSSFCW-Gra ($p < 0.05$).

Table 3

Zeolite Samples	Equation	1/n	*Kf*	Correlation coefficient (R^2)
Zeo-CW	$y = 1.7102x - 2.349$	1.71	0.004	0.993
Zeo-Top	$y = 1.2733x - 2.763$	1.27	0.002	0.999
Zeo-Middle	$y = 1.2009x - 2.849$	1.20	0.001	0.999
Zeo-Bottom	$y = 1.2622x - 2.792$	1.26	0.002	0.999
Zeo-Control	$y=1.5895x - 2.222$	1.59	0.006	0.994

Notes: Zeo-CW: zeolite samples from all zones of Zeo; Zeo-Top: zeolite samples from the top zone of Zeo; Zeo-Middle: zeolite samples from the middle zone of Zeo; Zeo-Bottom: zeolite samples from the bottom zone of Zeo: Zeo-control: zeolite samples used as a control that was not used in greywater treatment

Table 4