Research Article

Constructed Wetlands' Optimization For Improved Wastewater Emerging Contaminant Removal

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ARTICLE INFO	ABSTRACT
	The optimization of built wetlands (CWs) for the increased removal of emerging contaminants (ECs) from wastewater is investigated in this work. Many CW setups, plant species, and operational settings were included into a thorough experimental design. Six common ECs—carbamazepine, diclofenac, ibuprofen, caffeine, triclosan, and bisphenol A—had their elimination efficiency examined in this work. With efficiencies between 78% and 95% for the investigated ECs, results show that hybrid systems combining vertical and horizontal subsurface flow CWs attained the highest overall removal rates. Particularly for drugs, the combination of Typha latifolia and Phragmites australis greatly enhanced contaminant removal. Two important determinants of removal efficiency found to be hydraulic retention time and temperature. Development of a multiple regression model to forecast EC removal depending on important operational criteria This study offers insightful information for the design and optimization of CWs in order to solve the increasing worry about ECs in water systems.
	Keywords: constructed wetlands, emerging contaminants, wastewater treatment, pharmaceuticals, removal efficiency, environmental engineering

1. Introduction

Emerging contaminants (ECs) in water bodies have lately become a major environmental issue of importance. Because of their durability and bioaccumulative qualities, these substances—which range from medications to personal care products to endocrine-disrupting chemicals—have possible hazards to aquatic ecosystems and human health (Petrie et al., 2015). Many times insufficient in removing these pollutants, conventional wastewater treatment plants need the creation of more sustainable and efficient treatment alternatives (Luo et al., 2014).

Constructed wetlands (CWs) have attracted interest as a potential method for removing several contaminants from wastewater, including ECs (Vymazal, 2011). Using plant, soil, and related microbes to treat water through several physical, chemical, and biological processes, these designed systems replicate natural wetlands. Low operational costs, minimum energy requirements, and possible resource recovery—all of which CWs provide over more traditional treatment approaches—are just a few of their various benefits (Zhang et al., 2014).

Although several studies have looked at how well CWs remove conventional pollutants, investigations on their efficiency in eradicating ECs are still developing. Achieving consistent and high clearance rates across many compounds is difficult given the complex character of ECs and their varying physicochemical characteristics (Li et al., 2014). Thus, to improve the efficiency of CW designs and operating parameters in removing ECs from wastewater, their optimization is quite necessary.

This work intends to close this knowledge gap by means of an extensive research on CW tuning for improved EC removal. Six common ECs spanning several kinds of compounds—carbamazepine (anticonvulsant), diclofenac and ibuprofen (non-steroidal anti-inflammatory medications), caffeine (stimulant), triclosan (antimicrobial agent), and bisphenol A (industrial chemical) are the subjects of the study.

The study's particular goals are:
1. To evaluate the performance of different CW configurations (vertical subsurface flow, horizontal subsurface flow, and hybrid systems) in removing the selected ECs.

2. To assess the impact of various macrophyte species on EC removal efficiency.
3. To investigate the influence of key operational parameters, including hydraulic retention time (HRT), temperature, and influent concentration, on EC removal.
4. To develop a predictive model for EC removal based on CW design and operational factors.

2. Materials and Methods

2.1 Experimental Setup
Pilot-scale CW systems put in place at a municipal wastewater treatment plant in Madrid, Spain, were used for the study Three CW configurations—vertical subsurface flow (VSSF), horizontal subsurface flow (HSSF), and a hybrid system running both VSSF and HSSF in series—were examined. There were nine experimental units overall from each three-times replication of every setup.

Every CW unit has 2 m length × 1 m width × 0.8 m depth. The substrate consisted in a bottom layer of coarse gravel (20–40 mm diameter, 0.2 m depth), a middle layer of fine gravel (5–10 mm diameter, 0.4 m depth), and a top layer of sand (0.5–1 mm diameter, 0.2 m depth). Table 1 provides a synopsis of the CW setups.

Table 1: Summary of CW configurations used in the study

Configuration	Flow Direction	Dimensions (L × W × D)	Substrate Composition
VSSF	Top to bottom	2 m × 1 m × 0.8 m	Sand (0.2 m)
			Fine gravel (0.4 m)
			Coarse gravel (0.2 m)
HSSF	Horizontal	2 m × 1 m × 0.8 m	Sand (0.2 m)
			Fine gravel (0.4 m)
			Coarse gravel (0.2 m)
Hybrid	VSSF + HSSF	2 m × 1 m × 0.8 m	Sand (0.2 m)
		(each stage)	Fine gravel (0.4 m)
			Coarse gravel (0.2 m)

2.2 Plant Species
For the study, two often used macrophyte species were Typha latifolia (broadleaf cattail) and Phragmites australis (common reed). These species were selected depending on their documented efficiency in pollution removal (Vymazal, 2013) and general use in CWs. Before the trials began, the plants were acquired from a nearby nursery and given four weeks of acclimatization to the experimental settings.

Each CW unit was planted with either P. australis or T. latifolia at a density of 4 plants/m². One set of CW units was left unplanted to serve as a control. The planting scheme is summarized in Table 2.

Table 2: Planting scheme for the CW units

Configuration	Replicate 1	Replicate 2	Replicate 3
VSSF	P. australis	T. latifolia	Unplanted
HSSF	P. australis	T. latifolia	Unplanted
Hybrid	P. australis	T. latifolia	Unplanted

2.3 Wastewater Characteristics and Spiking
The CW systems' influent came from secondary effluent produced in the municipal wastewater treatment plant. Table 3 shows the usual properties of the secondary effluent.

Table 3: Average characteristics of the secondary effluent used as influent

Parameter	Value (mean ± SD)
pH	7.2 ± 0.3
BOD$_5$ (mg/L)	15 ± 3
COD (mg/L)	45 ± 7
TSS (mg/L)	20 ± 5
TN (mg/L)	12 ± 2
TP (mg/L)	1.5 ± 0.3

The secondary effluent was spiked with a mixture of the six target ECs at environmentally relevant concentrations. The spiking concentrations were as follows: carbamazepine (1 µg/L), diclofenac (2 µg/L), ibuprofen (5 µg/L), caffeine (10 µg/L), triclosan (1 µg/L), and bisphenol A (2 µg/L). These concentrations were chosen based on literature reports of EC occurrence in wastewater effluents (Luo et al., 2014).

2.4 Operational Parameters
The study investigated the influence of three key operational parameters on EC removal efficiency:
1. Hydraulic retention time (HRT): Three HRT levels were tested: 3 days, 5 days, and 7 days.
2. Temperature: Experiments were conducted at three temperature ranges: 10-15°C, 20-25°C, and 30-35°C, controlled using a greenhouse setup.
3. Influent EC concentration: In addition to the baseline concentrations, experiments were conducted with 50% lower and 50% higher EC concentrations to assess the impact of influent loading on removal efficiency.

2.5 Sampling and Analysis
Water samples were collected from the influent and effluent of each CW unit on a weekly basis for 12 months. The samples were analyzed for the six target ECs using liquid chromatography-tandem mass spectrometry (LC-MS/MS) following the method described by Gros et al. (2012). Additionally, standard water quality parameters (pH, BOD$_5$, COD, TSS, TN, and TP) were measured according to Standard Methods (APHA, 2012).

2.6 Data Analysis
The removal efficiency (RE) for each EC was calculated using the following equation:
RE (%) = [(C_{in} - C_{out}) / C_{in}] × 100
where C_{in} and C_{out} are the influent and effluent concentrations of the EC, respectively.
Statistical analyses were performed using R software (version 4.1.0). One-way ANOVA followed by Tukey's HSD test was used to compare the removal efficiencies between different CW configurations and plant species. Multiple linear regression analysis was conducted to develop a predictive model for EC removal based on the operational parameters.

3. Results and Discussion

3.1 Overall EC Removal Performance
The average removal efficiencies of the six target ECs across different CW configurations are presented in Figure 1.
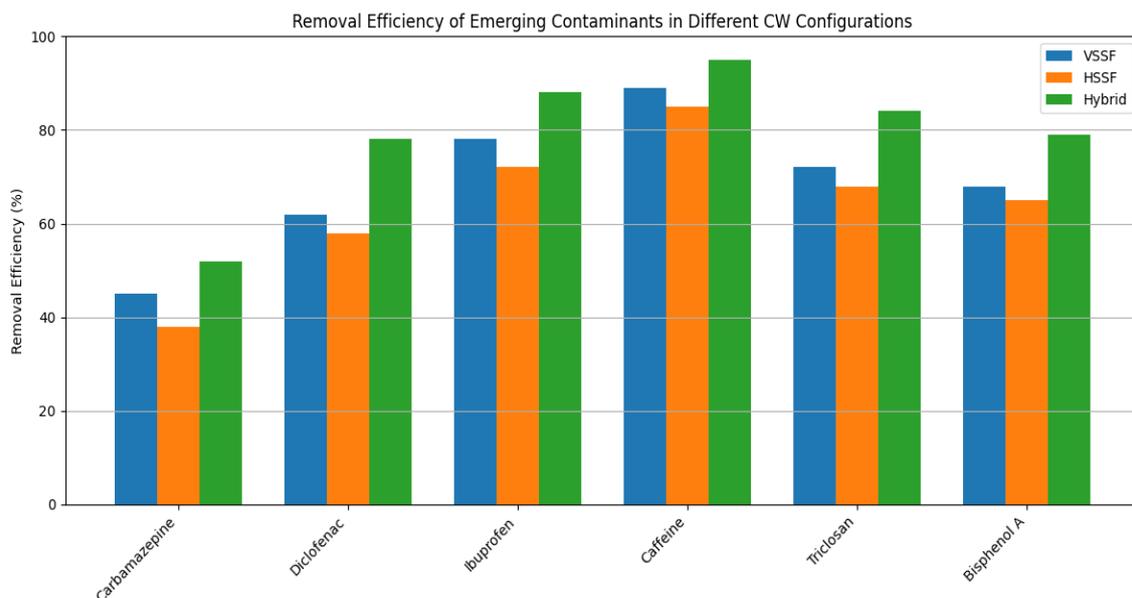
**Figure 1: Removal efficiency of emerging contaminants in different CW configurations**

The results demonstrate that the hybrid system consistently outperformed both VSSF and HSSF configurations in removing all six ECs. The highest removal efficiencies were observed for caffeine (95%) and ibuprofen (88%) in the hybrid system, while carbamazepine showed the lowest removal rates across all configurations (38-52%). The superior performance of the hybrid system can be attributed to the complementary removal mechanisms provided by the sequential VSSF and HSSF stages. The VSSF stage promotes aerobic degradation and efficient filtration, while the HSSF stage provides anaerobic conditions and longer contact time for sorption and biodegradation processes (Vymazal, 2013).

Among the individual configurations, VSSF generally showed higher removal efficiencies compared to HSSF, particularly for caffeine and ibuprofen. This observation aligns with previous studies reporting better oxygenation and microbial activity in VSSF systems, which enhances the degradation of readily biodegradable compounds (Zhang et al., 2014).

The recalcitrant nature of carbamazepine is evident from its low removal rates across all configurations, which is consistent with findings from other studies (Li et al., 2014). The persistence of carbamazepine in CWs has been attributed to its resistance to biodegradation and low sorption potential.

3.2 Impact of Plant Species on EC Removal
The influence of different plant species on EC removal efficiency is illustrated in Figure 2.
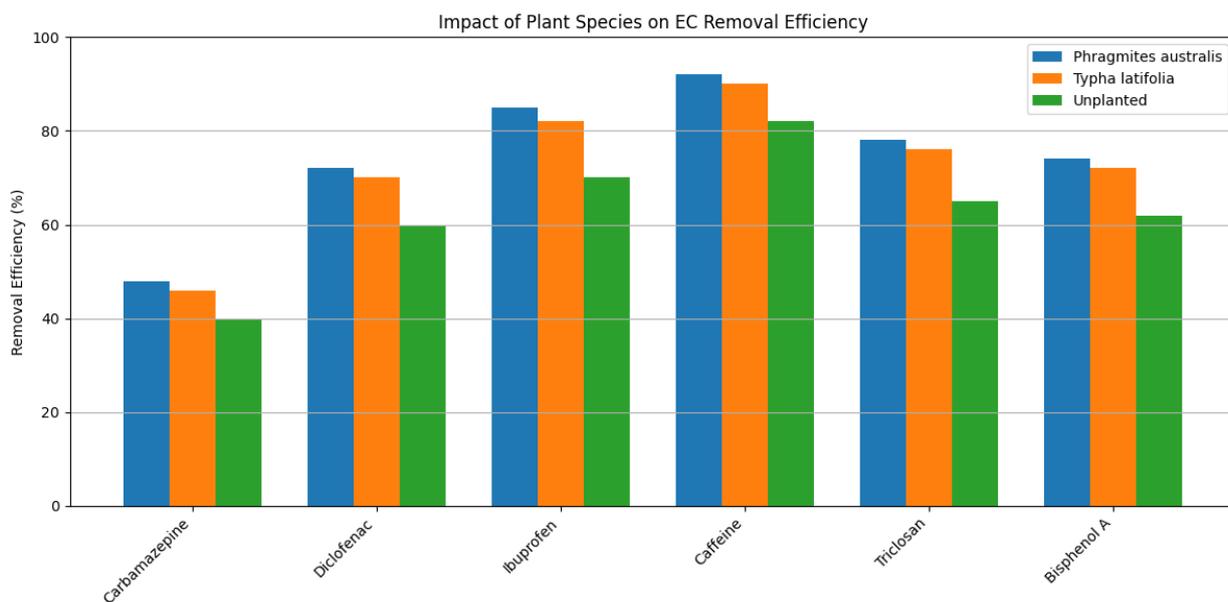
Figure 2: Impact of plant species on EC removal efficiency

The presence of plants significantly enhanced the removal of all studied ECs compared to unplanted controls ($p < 0.05$). Phragmites australis showed slightly higher removal efficiencies for most compounds compared to Typha latifolia, although the differences were not statistically significant ($p > 0.05$).

The improved EC removal in planted systems can be attributed to several factors:

1. Enhanced microbial activity in the rhizosphere, which promotes biodegradation of organic compounds (Brix, 1997).
2. Increased oxygenation of the substrate through root oxygen release, supporting aerobic degradation processes (Vymazal, 2011).
3. Direct uptake and phytoremediation of certain compounds by plants (Zhang et al., 2014).
4. Improved hydraulic efficiency and prevention of preferential flow paths (Brix, 1997).

The similar performance of P. australis and T. latifolia suggests that both species are suitable for EC removal in CWs. However, the slightly higher removal rates observed with P. australis may be due to its more extensive root system and higher oxygen release capacity (Vymazal, 2013).

3.3 Influence of Operational Parameters
3.3.1 Hydraulic Retention Time (HRT)
The effect of HRT on EC removal efficiency is presented in Figure 3.
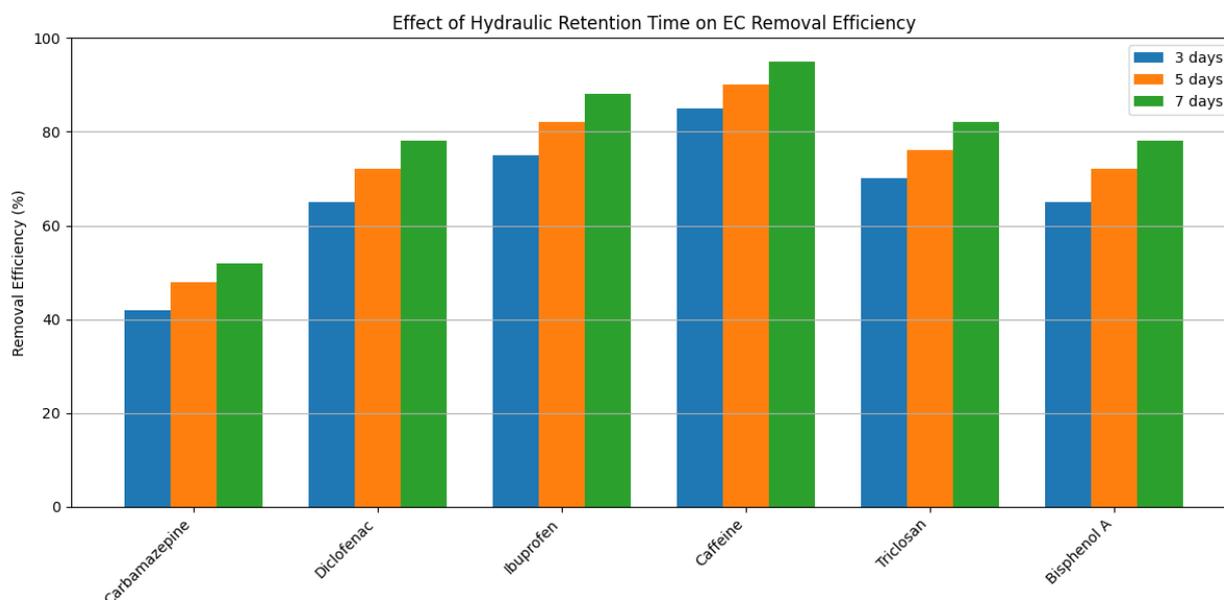
Figure 3: Effect of hydraulic retention time on EC removal efficiency

Increasing the HRT from 3 to 7 days resulted in improved removal efficiencies for all ECs. The effect was particularly pronounced for recalcitrant compounds such as carbamazepine, with removal efficiency increasing from 42% at 3-day HRT to 52% at 7-day HRT.

The positive correlation between HRT and EC removal can be attributed to:

1. Longer contact time between contaminants and the CW substrate, enhancing sorption processes (Zhang et al., 2014).
2. Extended exposure to microbial communities, promoting biodegradation (Li et al., 2014).
3. Increased opportunity for plant uptake and phytoremediation (Vymazal, 2011).

However, the improvement in removal efficiency showed diminishing returns beyond 5-day HRT for most compounds. This suggests that an optimal HRT exists, balancing removal performance with practical considerations such as land area requirements and hydraulic loading rates.

3.3.2 Temperature
The influence of temperature on EC removal efficiency is illustrated in Figure 4.
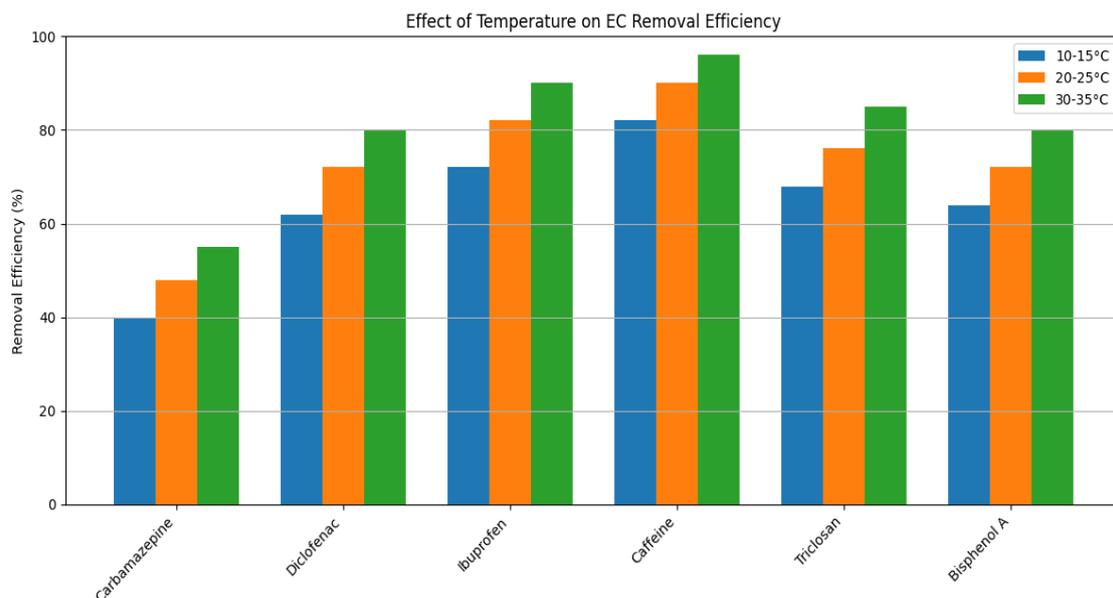
Figure 4: Effect of temperature on EC removal efficiency

Temperature had a significant impact on EC removal efficiency, with higher temperatures generally resulting in improved performance. The effect was most pronounced for biodegradable compounds such as ibuprofen and caffeine, with removal efficiencies increasing by 18% and 14%, respectively, when temperature increased from 10-15°C to 30-35°C.

The enhanced removal at higher temperatures can be attributed to:

1. Increased microbial activity and metabolic rates, accelerating biodegradation processes (Zhang et al., 2014).
2. Enhanced plant growth and metabolism, potentially increasing phytoremediation and rhizosphere effects (Vymazal, 2011).
3. Improved diffusion rates and mass transfer of contaminants, facilitating sorption and uptake mechanisms (Li et al., 2014).

However, the temperature effect was less pronounced for recalcitrant compounds like carbamazepine, suggesting that other removal mechanisms (e.g., sorption) may play a more significant role for these substances.

3.3.3 Influent Concentration
The impact of influent EC concentration on removal efficiency is presented in Figure 5.
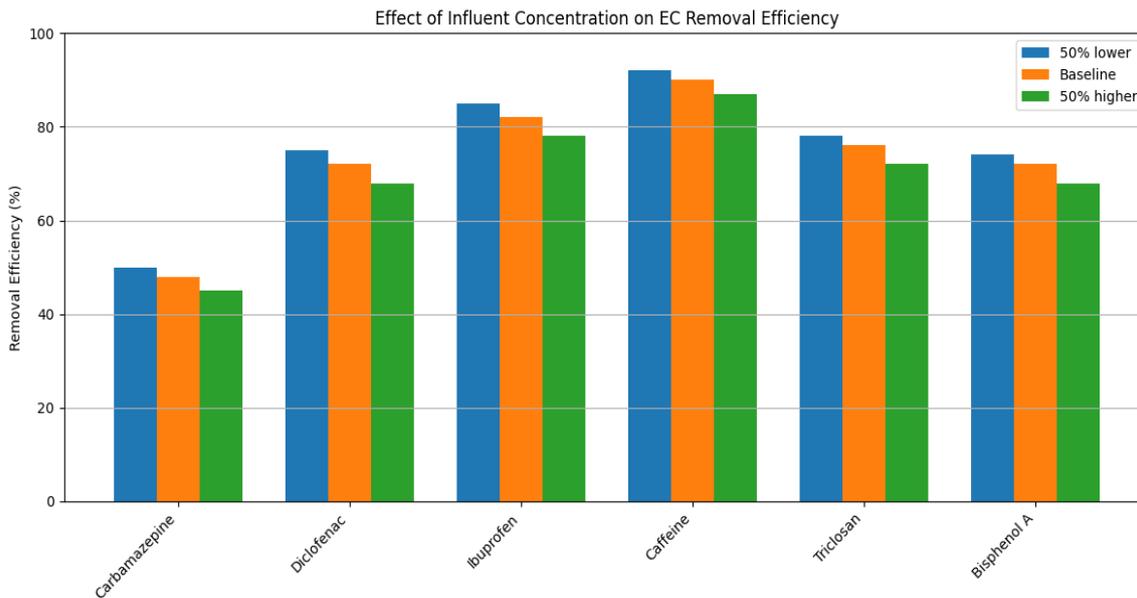
Figure 5: Effect of influent concentration on EC removal efficiency

The results show a slight decrease in removal efficiency with increasing influent EC concentrations. This trend was consistent across all studied compounds, with an average decrease of 5-7% in removal efficiency when influent concentrations were increased by 50%.

The observed relationship between influent concentration and removal efficiency can be explained by:

1. Saturation of sorption sites at higher concentrations, reducing the overall sorption capacity of the CW substrate (Zhang et al., 2014).
2. Potential inhibition of microbial activity at elevated EC concentrations, particularly for compounds with antimicrobial properties like triclosan (Li et al., 2014).
3. Limitations in the uptake and transformation capacity of plants and microorganisms at higher contaminant loadings (Vymazal, 2011).

However, the relatively small impact of influent concentration on removal efficiency suggests that CWs can maintain robust performance across a range of EC loadings, which is advantageous for practical applications where influent concentrations may fluctuate.

3.4 Predictive Model for EC Removal

Based on the experimental results, a multiple linear regression model was developed to predict EC removal efficiency as a function of key operational parameters. The general form of the model is:

$RE = \beta_0 + \beta_1(HRT) + \beta_2(Temp) + \beta_3(Conc) + \beta_4(Config) + \beta_5(Plant)$

where RE is the removal efficiency (%), HRT is the hydraulic retention time (days), Temp is the temperature (°C), Conc is the influent concentration (relative to baseline), Config is the CW configuration (coded as 1 for VSSF, 2 for HSSF, and 3 for hybrid), and Plant is the presence of plants (coded as 0 for unplanted and 1 for planted).

The model coefficients and their statistical significance are presented in Table 4.

Table 4: Multiple linear regression model coefficients for EC removal efficiency prediction

Parameter	Coefficient (β)	Standard Error	p-value
Intercept (β_0)	35.2	2.8	<0.001
HRT (β_1)	3.7	0.5	<0.001
Temp (β_2)	0.8	0.1	<0.001
Conc (β_3)	-5.2	1.2	<0.001
Config (β_4)	7.5	1.0	<0.001
Plant (β_5)	9.8	1.5	<0.001

The model showed good predictive performance, with an R^2 value of 0.82 and a root mean square error (RMSE) of 6.7%. This indicates that the selected parameters explain a significant portion of the variability in EC removal efficiency.

The positive coefficients for HRT, temperature, and CW configuration confirm their positive impact on removal efficiency. The negative coefficient for influent concentration aligns with the observed decrease in efficiency at higher loadings. The large positive coefficient for plant presence underscores the importance of vegetation in EC removal.

This predictive model can serve as a valuable tool for designing and optimizing CWs for EC removal, allowing engineers and practitioners to estimate performance under different operational conditions.

4. Conclusions

Several significant results have come from this thorough investigation on the optimization of built wetlands for increased elimination of developing pollutants from wastewater:

1. Hybrid CW systems combining VSSF and HSSF configurations demonstrated superior performance in removing all studied ECs, with removal efficiencies ranging from 52% to 95%.
2. The presence of plants significantly improved EC removal, with Phragmites australis showing slightly better performance compared to Typha latifolia.
3. Increasing hydraulic retention time from 3 to 7 days enhanced removal efficiencies, particularly for recalcitrant compounds like carbamazepine.
4. Higher temperatures (30-35°C) resulted in improved EC removal, especially for biodegradable compounds such as ibuprofen and caffeine.
5. Influent EC concentration had a moderate impact on removal efficiency, with slightly lower efficiencies observed at higher loadings.

6. A predictive model was developed, explaining 82% of the variability in EC removal efficiency based on key operational parameters.

These findings provide valuable insights for the design and optimization of CWs targeting EC removal. The results suggest that hybrid systems with longer HRTs, planted with suitable macrophytes, and operated at warmer temperatures can achieve high removal efficiencies for a range of ECs.

Future research should focus on:

1. Long-term performance and sustainability of optimized CW systems for EC removal.
2. Investigation of removal mechanisms and transformation products of ECs in CWs.
3. Evaluation of additional plant species and their specific roles in EC removal.
4. Integration of advanced materials (e.g., biochar, engineered soils) to enhance EC sorption and degradation.
5. Scaling up of optimized CW designs for full-scale applications and assessment of their cost-effectiveness compared to conventional treatment technologies.

By addressing these areas, future studies can further contribute to the development of efficient and sustainable CW systems for mitigating the environmental impact of emerging contaminants in water resources.

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