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Review article

Optimizing constructed wetland design and operation for dual benefits: A critical review to enhance micropollutant removal while mitigating greenhouse gas emissions

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ABSTRACT

Constructed wetlands (CWs) are increasingly considered for secondary wastewater treatment, removing both conventional contaminants and emerging pollutants, notably pharmaceutical and personal care products (PPCPs). However, the CW design and operational conditions to biodegrade PPCPs as micropollutants may promote greenhouse gas (GHG) emissions, raising sustainability concerns. This meta-analysis investigates the relationship between PPCP removal (caffeine, ibuprofen, naproxen, diclofenac, ketoprofen, carbamazepine, sulfonamide compounds) and GHG emissions (methane, carbon dioxide, nitrous oxide) in CWs. We uniquely integrate two sets of studies, as prior research has not linked PPCP biodegradation with GHG emissions. Data from 26 papers identify factors driving PPCP removal and 26 publications inform GHG emission factors. Spearman's correlation coefficient and multiple linear regression assess parameter effects and interlinkages. Results highlight biological processes, particularly secondary metabolism or co-metabolism, as pivotal for PPCP removal and GHG emissions, with inlet PPCP concentration, carbon load, and temperature being significant influencers ($p < 0.05$). Challenges persist in optimizing operations to improve PPCP removal and abate GHG emissions simultaneously. Still, CW depth, influent chemical oxygen demand (COD), hydraulic retention time, and subsurface flow wetland configuration emerge as strategic parameters. This study underscores the need for integrated approaches to enhance PPCP removal and decrease GHG emissions in CWs, thereby advancing sustainable water management practices.

1. Introduction

Sustainable urban development faces significant challenges in managing water resources, particularly in treating municipal wastewater. This is in part because of our current engineering practices. Conventional wastewater treatment systems, developed under a linear material and energy management approach, often have large land footprints, energy-intensive operations, and produce harmful byproducts such as greenhouse gas (GHG) emissions (Dijst et al., 2018). In contrast, the concept of circular cities aims to reduce waste and pollution while maximizing resource efficiency. Within this framework, nature-based solutions like constructed wetlands (CWs) have emerged as promising technologies for sustainable water management.

CWs, particularly as secondary treatments, could offer several advantages, such as effective and simultaneous removal of conventional pollutants and emerging contaminants, potential carbon sequestration, and enhancement of urban ecosystems, including promoting water reclamation locally (Mander et al., 2014; Ilyas et al., 2020). However, assessing the sustainability of CWs is not straightforward due to their potential contribution to GHG emissions, mainly carbon dioxide (CO_2), methane (CH_4), and nitrogen oxide (N_2O), generated during wastewater treatment operations (Mander et al., 2014; Ilyas et al., 2020). In urban wastewater, emerging pollutants from various sources, such as pharmaceuticals and personal care products (PPCPs), can limit water reclamation. PPCPs tend to persist in treated effluents (Kim and Zoh, 2016), which can disrupt aquatic ecosystems and potentially cause health

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issues in humans through chronic exposure (Ma et al., 2018). In domestic and municipal wastewaters, PPCPs are found in mixtures and encountered as micropollutants (<1 ppm). Some highly persistent PPCPs which are extensively studied are caffeine (CAF), ibuprofen (IBU), naproxen (NPX), diclofenac (DCF), ketoprofen (KTP), carbamazepine (CBZ) and sulfonamide compounds (sulfamethazine and sulphapyridine, SUL) (Yang et al., 2014) and are the focus of our work.

CWs have shown promise in effectively degrading recalcitrant compounds, especially through co-metabolic processes (Garcia-Becerra and Ortiz, 2018; Men et al., 2017). However, the conditions required for PPCP removal, such as longer retention times, and mixed aerobic/anaerobic or feast/famine conditions, may inadvertently promote higher GHG emissions or produce more potent GHGs (CH_4 and N_2O vs. CO_2). Fig. 1 summarizes the potential removal mechanisms that have been individually reported for the studied PPCPs, along with the release of GHGs. While the mineralization of PPCPs alone is unlikely to contribute significantly to GHG emissions compared to the degradation of conventional organic matter, measured as chemical oxygen demand (COD), biological oxygen demand (BOD), or total organic carbon (TOC), the overall impact of treatment conditions optimized for PPCP removal on GHG production requires investigation. Currently, the relationship, or potential tradeoff, between PPCP removal and GHG emissions in CWs remains poorly understood. However, understanding this relationship in CWs is crucial for developing truly sustainable water treatment solutions that address emerging contaminants without exacerbating climate change concerns.

Investigating the link between PPCP removal and GHG emissions in the design and operation of CWs can help us improve their designs. Determining how CW parameters, such as depth (D), surface area, hydraulic loading rate (HLR), hydraulic retention time (HRT), configuration, and plant species, influence the CW performance is a larger active area of research (Frazer-Williams, 2010; Wu et al., 2015; Ilyas and van Hullebusch, 2019; Rahman et al., 2020). More specifically, multiple critical review papers have identified the main factors affecting the removal of micropollutants or the emission of GHGs in CWs as two independent research areas. In the set of studies focused on PPCPs, it has been observed that the removal of micropollutants depends on their chemical and physicochemical properties, as well as CW design and operational parameters, which in turn promote biodegradation (primary and secondary metabolisms, as well as co-metabolisms), adsorption, and photodegradation, among other removal mechanisms (Zhang et al., 2014a,b; He et al., 2018; Ilyas and van Hullebusch, 2019). As for studies on emissions of GHGs in CWs, GHG production is reported to be highly promoted by aerobic conditions in the presence of high loading rates (CO_2) and in anoxic/anaerobic conditions (CH_4 and N_2O) (Mander et al., 2014; Jahangir et al., 2016; Schalk et al., 2019). That is, both PPCP removal and GHG production have been observed to be biological-driven processes. However, to our knowledge, no previous study has focused on linking PPCPs removal and GHG emissions in the context of secondary treatment of domestic wastewater, which are biologically-driven outcomes taking place simultaneously.

In this work, we studied the possible relationship between removing PPCPs as micropollutants and abating GHG emissions in CWs as secondary treatments of domestic wastewater. The same design and operating parameters were followed in both types of studies (PPCPs and GHGs). These parameters were selected due to their influence on biologically driven processes. Both data sets were assessed individually using the Spearman's correlation analysis and multiple linear regression (MLR) model. This was done to identify the most influential parameters for each treatment outcome (PPCP removal and GHG emissions). We then compared the identified factors affecting the two sets of studies to observe their possible interactions, such as co-metabolism degradation mechanisms.

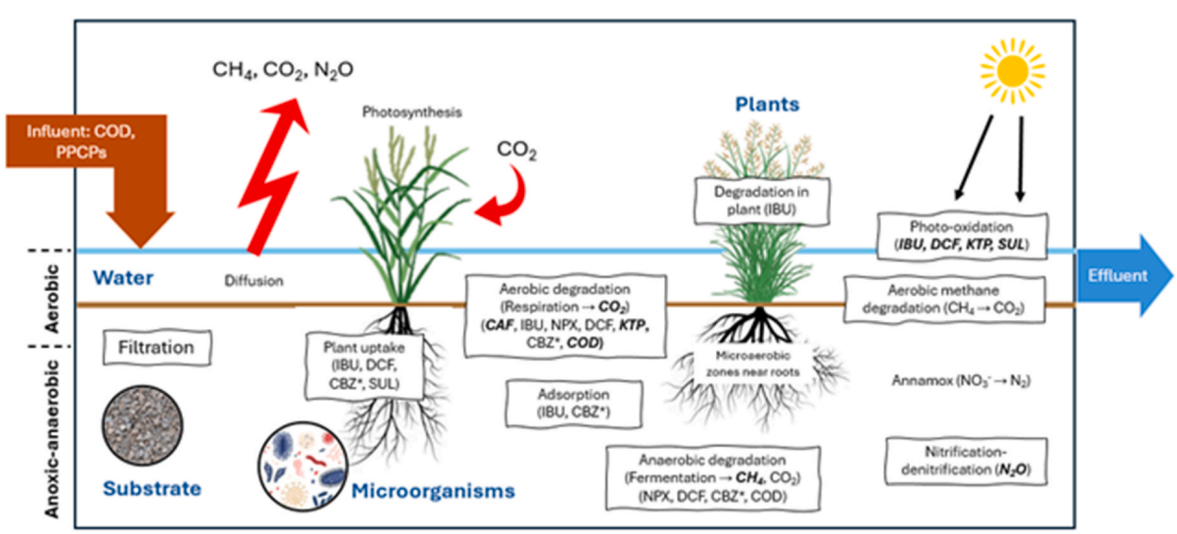
This critical review aims to provide insights that can guide the optimization of CW systems, enabling effective micropollutant removal while minimizing environmental impact through GHG emissions. Through this work, we aim to contribute to the development of next-generation CWs that can play a pivotal role in circular water management, addressing both water quality and climate change mitigation objectives simultaneously.

2. Materials and methods

2.1. Data set formation

Data used for this study was obtained from peer-reviewed journal research papers using the search engines Elsevier Science Direct, Springer Link, Google Scholar, and PubMed. The search was limited to articles written in English and published from 2010 to 2023. The following keywords were used: "treatment wetlands", "constructed wetlands", "domestic wastewater treatment", "municipal wastewater treatment", "micropollutants", and "greenhouse gases", as well as selected PPCPs (CAF, IBU, NPX, DCF, KTP, CBZ and SUL) and GHGs (CO_2, N_2O, and CH_4.)

A screening process was then employed to select studies of CWs used only for secondary treatment of domestic/municipal wastewater or



Note: Bold-italic letters represent the PPCPs' major elimination and GHG production routes.

Fig. 1. PPCPs removal and GHG production mechanisms in CWs.

synthetic wastewater with domestic features. For PPCPs studies, pilot-scale and large-scale systems exposed to the elements were selected to consider their performance under environmental factors. Due to the difficulty of measuring GHGs in outdoor CWs, studies for GHGs included greenhouse conditions and laboratory experiments. Thus, 26 studies were selected for PPCPs, and the same amount was found for GHGs. The average design, operational, and physicochemical parameters of the two sets of studies are indicated in Tables 1 and 2. Twenty-two studies examine CW sizes ranging from 0.35 to 20 m^2, while only four focus on larger scales between 500 and 18,000 m^2. These four studies pertain to PPCPs, as GHG-related studies found in this work were only conducted at a laboratory scale. The selected studies were carried out in various countries worldwide, including China, Colombia, the Czech Republic, Germany, Italy, Mexico, Singapore, Spain and the UK.

Data mining included both design and operational parameters. The design variables collected were: depth (D, m), length:depth ratio (L:D, unitless), length:width ratio (L:W, unitless), flow configuration as subsurface horizontal (HSFW), subsurface vertical (VSFW) and superficial (SFW), and vegetation as planted or unplanted systems. The operational parameters collected were: inlet and outlet PPCPs concentration (Cin and Cout, mg/L), PPCPs removal efficiency (RE, %), GHG emission rate (mg/m^2h), inlet total COD concentration (CODin, mg/L), COD removal efficiency (RE, %), mean water temperature (T, °C), hydraulic loading rate (HLR, m/d), hydraulic retention time (HRT, days), inlet pH, dissolved oxygen (DO, mg/L), and redox potential (Redox, mV).

Where the GHGs emission rate was found in figures and not expressed numerically in their respective research papers, a grid was superimposed to the figure to estimate the values. The values were then homogenized to the same units (mg/m^2h) for comparison. For those studies that did not report mean water temperature, it was calculated from the reported air temperature using the linear correlation proposed by Morrill et al. (2005) as Twater (°C)=Tair (°C)*0.93 + 0.56. Where needed, HLR and HRT were calculated as HLR = flow/A and HRT=V/flow, where V is the volume of the CW (m^3), and A is the surface area of the CW (m^2). COD was approximated using the COD/BOD ratio (COD/BOD = 2) and COD/TOC ratio (COD/TOC = 3) (Aziz and Tebbutt, 1980) when the authors presented either BOD or TOC instead of COD. These parameters were selected due to their known influence on biological wastewater treatments.

Table 1
Average values (and standard deviation) of the selected design, operational, and physicochemical parameters of the selected PPCPs.

Variable	CAF	IBU	NPX	DCF	KTP	CBZ	SUL
Cin (μg/L)	39.9 ± 23.5	57.2 ± 58.3	33.3 ± 42.8	27.8 ± 40.7	42.6 ± 47.8	22.6 ± 46	0.5 ± 0.9
CODin (mg/L)	293 ± 183	262 ± 148	258 ± 159	259 ± 153	225 ± 103	272 ± 176	175 ± 62
T (°C)	16 ± 5.8	18.7 ± 7.0	18.4 ± 7.1	18.5 ± 6.8	18.3 ± 7.6	18.4 ± 5.8	20.2 ± 7.0
D (m)	0.97 ± 0.18	0.58 ± 0.23	0.59 ± 0.24	0.58 ± 0.24	0.56 ± 0.25	0.67 ± 0.17	0.82 ± 0.05
L:D ratio	13.8 ± 21.4	6.4 ± 12.8	7.7 ± 14	6.7 ± 13.2	5.2 ± 5.5	4.2 ± 5.5	1.2 ± 2.9
L:W ratio	2.8 ± 1.5	2.8 ± 5.1	3.3 ± 5.9	2.9 ± 5.3	3.5 ± 6.8	3.5 ± 6.8	1.4 ± 3.5
HLR (mm/d)	287 ± 860	201 ± 635	239 ± 701	208 ± 642	80.9 ± 60	273 ± 808	245 ± 136
HRT (d)	3.8 ± 2.4	3.47 ± 2.3	3.1 ± 2.3	3.4 ± 2.4	3.4 ± 2.5	3.0 ± 1.0	1.0 ± 0.0
TNin (mg/L)	80 ± 10.2	87.4 ± 3.9	87.4 ± 3.9	87.4 ± 3.9	82.3 ± 0	82.7 ± 6	–
TOCin (mg/L)	23 ± 11	30.7 ± 39.9	18.1 ± 12.6	35.9 ± 42.5	37.5 ± 2.6	18.1 ± 12.6	–
pHin (mg/L)	7.4 ± 0.2	7.5 ± 0.2	7.3 ± 0.2	7.3 ± 0.2	7.5 ± 0	7.32 ± 0.21	–
Redoxin (mV)	−98 ± 129	−151 ± 117	−151 ± 117	−151 ± 117	0.1 ± 0	−151 ± 117	–
DOin (mg/L)	0.8 ± 0.8	3.2 ± 3.2	1.6 ± 1.9	1.4 ± 1.9	1.8 ± 2.9	1.4 ± 1.9	–
N	48	85	68	81	47	52	60

Note: Cin is inlet concentration; CODin is inlet total chemical oxygen demand; T is water temperature; D is depth; L:D is length depth ratio; L:W is length width ratio; HLR is hydraulic loading rate; HRT is hydraulic retention time; TNin is inlet total nitrogen; TOCin is inlet total organic; pHin is inlet pH; Redoxin is inlet redox potential; DOin is inlet dissolved oxygen; N is the total number of data points per pollutant.

Table 2
Summary of selected design, operational, and physicochemical parameters of the selected GHGs.

Variable	CH_4	CO_2	N_2O
Emission rate (mg/m^2h)	13.4 ± 42.2	170.2 ± 495.3	0.32 ± 0.78
CODin (mg/L)	154 ± 127	165 ± 117	128 ± 105
T (°C)	17.3 ± 6.9	15.9 ± 6.8	19.6 ± 7.1
D (m)	0.47 ± 0.16	0.5 ± 0.15	0.5 ± 0.12
L:D ratio	5.9 ± 6.9	5.1 ± 5.7	1.8 ± 4.1
L:W ratio	1.4 ± 2.5	0.9 ± 0.6	1.6 ± 2.5
HLR (mm/d)	100.9 ± 123	213 ± 111	79 ± 123
HRT (d)	9.5 ± 14.4	2.4 ± 1.4	11 ± 15
TNin (mg/L)	31 ± 21	40 ± 22	29 ± 17
TOCin (mg/L)	43 ± 33	37 ± 11	49 ± 39
pHin (mg/L)	7.3 ± 0.2	7.3 ± 0.2	7.2 ± 0.2
Redoxin (mV)	23.5 ± 5.2	–	–
DOin (mg/L)	5.2 ± 3.6	5.2 ± 3.6	6.1 ± 3.3
N	259	130	147

Note: CODin is inlet total chemical oxygen demand; T is water temperature; D is depth; L:D is length depth ratio; L:W is length width ratio; HLR is hydraulic loading rate; HRT is hydraulic retention time; TNin is inlet total nitrogen; TOCin is inlet total organic; pHin is inlet pH; Redoxin is inlet redox potential; DOin is inlet dissolved oxygen; N is the total number of data points per pollutant.

The parameters initially searched were selected due to their influence on the biologically driven processes of PPCP removal and GHG emissions. However, since not all parameters are reported in the surveyed studies, the rest of the meta-analysis no longer included L:D ratio, L:W ratio, TNin, TOCin, pHin, Redoxin and DOin. Overall, the number of data points reviewed for the removal of the pollutants is: CAF (48), IBU (85), NPX (68), DCF (81), KTP (47), CBZ (52) and SUL (60). The data points for the emission of GHGs are CH_4 (266), CO_2 (132), and N_2O (145).

2.2. Data analysis

For each set of studies, the Spearman's correlation analysis was calculated to quantify the relationship between the change in PPCPs concentration (ΔC = Cin-Cout, mg/L) and GHG emission rate (mg/m^2h) with respect to Cin, CODin, T, D, HLR, HRT. Vegetation and configuration parameters were not included since they are considered categorical variables. This way, Spearman's correlations capture non-linear trends, quantifying the links between changes in PPCPs concentration and GHG emission rate with various input variables.

The MLR analysis has been applied to study the effect of operational and design parameters on CW performance, with a focus on nutrients and organics removal (Li et al., 2018; Pavlineri et al., 2017). In this study, ΔC and GHG emission rate are the dependent variables for the PPCP analysis and GHG analysis, respectively, as shown in equation 1

$$\Delta C = \beta_1 C_{in} + \beta_2 COD_{in} + \beta_3 T + \beta_4 D + \ldots + \beta_n x_n \quad (1)$$

Where ΔC was the dependent variable, whereas the independent variables were Cin, CODin, T, D, HLR, HRT, vegetation and CW configuration with their respective coefficient value (*β*). For the GHG analysis, the GHG emission rate replaces ΔC in Eq. (1), and there is no Cin. Due to independent variables being presented in different units/dimensions, they were standardized for input into the MLR. The enter method was used considering that all variables inserted in the model were significant at $p < 0.05$. However, only significant coefficients ($p < 0.05$) in the MLR output are reported. The R^2 and adjusted R^2 values were calculated to assess the model fitness with the training data. MLR identifies significant linear relationships between the dependent variables (ΔC and GHG emission rate) and the mentioned independent variables.

Spearman's correlation and the MLR analyses were conducted using the IBM SPSS Statistics version 26 software (IBM Corp, 2019).

3. Results and discussion

3.1. PPCPs removal

In CWs, multiple physical, chemical, and biological processes simultaneously take place leading to four main degradation mechanisms for PPCPs: photolytic degradation, adsorption, phytodegradation, and microbial degradation (Vo et al., 2018). Each mechanism operates differently, with different variables influencing PPCP removal, which is a significant challenge for the technological development of CWs. Vo et al. (2018) reviewed works that have quantified via black box models the contribution of these degradation mechanisms to individual PPCPs. In this work, we use data-driven methods, the Spearman's correlation and MLR analyses, to identify and evaluate the correlation between variables to the removal of the selected PPCPs.

During our search for articles on PPCP removal in CWs, we identified that most studies do not report or monitor key design and operational parameters such as L:D ratio, L:W ratio, TNin, TOCin, pHin, Redoxin or DOin. These variables are key to identify degradation mechanisms. For most articles, we are unable to quantify the contribution of these mechanisms directly. Indirectly, we have linked certain parameters to degradation mechanisms through assumptions. We consider that photolytic degradation is likely to occur in SFW configuration, since it implies exposure to sunlight, and within this configuration lower D values can also be linked to sunlight exposure. We link adsorption more strongly to HSFW and VSFW configurations, where higher D values can provide more adsorption surfaces. We also relate phytodegradation to the presence of vegetation in CWs and higher T values (Li et al., 2020). Regarding microbial degradation, we can associate it to a range of pathways (e.g., aerobic, anaerobic, which can be led by a plethora of microbes such as bacterial, fungi, algae, etc.). Consequently, different variables can be related to different biodegradation pathways. Higher CODin and HLR values can imply an availability of alternative carbon sources during PPCP degradation, suggesting a possible co-metabolic process. Higher values in T, HLR and HRT can also be involved in enhancing biological activity or interactions between microbes and PPCPs. Aerobic biodegradations could be supported by the presence of vegetation, VSFW configuration and low D values. Anaerobic conditions can be found in HSFW and SFW configurations and high D values.

Fig. 2 shows the spread of the removal efficiencies of the selected PPCPs in CWs. On average, the removal of CAF was the highest (81.3 ± 21.4%), indicating this is the most degradable PPCP. While IBU, NPX, DCF, KTP and SUL presented average removal efficiencies of 54.4 ± 29.8%, 56.9 ± 26.9%, 39.1 ± 23.7%, 49.9 ± 29.7%, and 51.4 ± 35.4%, respectively. On the other hand, CBZ had the lowest removal with 24.78% ± 21.96%, indicating it as the most persistent pollutant from the list. These removal rates are consistent in other wastewater treatment processes, where CAF is highly biodegradable, while CBZ is a recalcitrant micropollutant (Hijosa-Valsero et al., 2010; Garcia-Becerra and Ortiz, 2018). Additionally, the average removal for the different configurations was assessed (Fig. S1). While the highest removal was observed in the VSFW at 55%, followed by the HSFW at 50% and the SFW at 48%, the average values are not significantly different. *Typha angustifolia* exhibited the highest average removal rate at 78%, whereas the combination of *Heliconea Zingiberales* and *Cyperus haspan* had an average removal rate of 16% (Fig. S2). Gravel was the most commonly used substrate due to its widespread availability globally. However, in certain studies, zeolite and vesuvianite have demonstrated higher performance (Dan et al., 2013; Chen et al., 2016; Zhang et al., 2018). Batch feeding mode has been found to enhance the removal of certain PPCPs significantly; however, higher removal efficiencies have been observed in continuous mode too (Zhang et al., 2012). Due to the insufficient data distribution on substrate types and operation modes, these factors are not considered in the further analysis of this study.

3.2. Design and operational parameters affecting PPCPs removal

Overall, Spearman's correlation and MLR analyses results suggest that the removal of the micropollutants in this study are primarily associated with biological processes, such as secondary metabolism or co-metabolisms (Tables 3 and 4). Except for SUL, in the Spearman's correlation analysis, the micropollutant Cin has the strongest positive correlation to the removal of all PPCPs, while the CODin has a negative correlation to most of the micropollutants' removal or has a small correlation value for those with a positive correlation. This could suggest that the degradation of these recalcitrant PPCPs is based on secondary metabolisms, which become activated in response to the presence of these pollutants. This is also observed in the MLR analysis, where Cin is the independent variable with the highest coefficient values for all the PPCPs. In the case of SUL, in the Spearman's correlation analysis, the CODin has the highest correlation to the removal of this micropollutant, and in the MLR analysis, the only variables with associated coefficients are the Cin and HLR. These results could suggest that the removal of SUL is through a co-metabolic degradation, where an additional carbon source is required. Further, in the MLR analysis, T appears with a

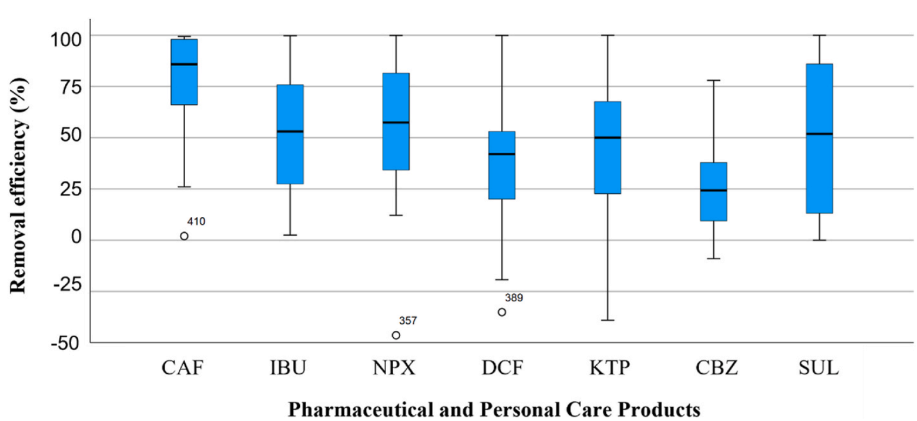


Fig. 2. Removal efficiencies ([Cin-Cout]/Cin) of PPCPs in CWs. The number of data points reviewed for the removal of the pollutants was as follows: CAF (48), IBU (85), NPX (68), DCF (81), KTP (47), CBZ (52) and SUL (60).

Table 3
Spearman correlation statistics among the studied factors and concentration reduction of the studied PPCPs and COD.

Variable	C_{in}	COD_{in}	T	D	HLR	HRT
ΔCAF	**0.880**[b]	**0.433**[b]	−0.025	−0.042	**−0.405**[b]	0.082
N	*42*	*42*	*42*	*42*	*42*	*42*
ΔIBU	**0.911**[b]	0.025	**0.424**[b]	**−0.292**[b]	−0.161	−0.031
N	*79*	*78*	*79*	*79*	*79*	*77*
ΔNPX	**0.934**[b]	**−0.327**[b]	**0.597**[b]	**−0.735**[b]	−0.118	**−0.433**[b]
N	*64*	*63*	*64*	*64*	*64*	*62*
ΔDCF	**0.915**[b]	**−0.363**[b]	**0.493**[b]	**−0.687**[b]	−0.075	**−0.400**[b]
N	*77*	*73*	*77*	*74*	*77*	*72*
Δ KTP	**0.955**[b]	**−0.503**[b]	**0.588**[b]	**−0.799**[b]	**0.668**[b]	**−0.541**[b]
N	*45*	*46*	*47*	*47*	*47*	*46*
ΔCBZ	**0.817**[b]	−0.166	**0.346**[a]	−0.243	−0.172	−0.037
N	*45*	*48*	*48*	*48*	*48*	*47*
ΔSUL	**0.650**[b]	**0.777**[a]	**−0.336**[b]	−0.076	0.148	–
N	*60*	*56*	*60*	*57*	*60*	*1*
ΔCOD	–	**0.945**[b]	**−0.256**[b]	**0.172**[b]	**−0.212**[b]	0.106
N	-	*406*	*417*	*411*	*417*	*347*

[a] Correlation is significant at the 0.05 level (2-tailed). In bold.
[b] Correlation is significant at the 0.01 level (2-tailed). In bold.

Table 4
MLR equations for the selected PPCPs and COD.

PPCPs	R^2	R^2 Adj.	P value	Equation
CAF	0.89	0.87	<0.001	$\Delta C = 0.91C_{in} + 0.23T + 0.13D + 0.12HRT + 0.15VSFW - 0.13SFW$
IBU	0.90	0.89	<0.001	$\Delta C = 1.00C_{in} + 0.15D - 0.19SFW$
NPX	0.93	0.93	<0.001	$\Delta C = 0.91C_{in} + 0.20T - 0.17SFW$
DCF	0.98	0.97	<0.001	$\Delta C = 0.91C_{in} + 0.07T + 0.1SFW$
KTP	0.96	0.95	<0.001	$\Delta C = 0.80C_{in} + 0.17T + 0.17SFW$
CBZ	0.86	0.73	<0.001	$\Delta C = 0.79C_{in} + 0.27T$
SUL	0.48	0.46	<0.001	$\Delta C = 0.68C_{in} + 0.24HLR$
COD	0.92	0.92	<0.001	$\Delta C = 1.02C_{in} + 0.09T + 0.05D + 0.08VSFW - 1.00\ SFW$

positive coefficient in the equations of most of the PPCPs, which can be indicative of biological activity. Apart from the effect of the PPCPs' initial concentration, the possible phenomena associated with the removal of each micropollutant are described below.

3.2.1. Caffeine (CAF)

Previous research has shown that CWs exhibit a high capacity for the degradation of CAF, with removal rates exceeding 80% (Sgroi et al., 2018). This high removal efficiency has been attributed to the presence of aerobic conditions, which facilitate various oxidation pathways during bacterial biodegradation of CAF (Nguyen et al., 2019). Notably, VFSWs have demonstrated superior performance in CAF removal compared to other configurations (Sgroi et al., 2018; de Oliveira et al., 2019).

Furthermore, it has been observed that higher temperatures contribute to enhanced CAF removal (Truu et al., 2009; Hijosa-Valsero et al., 2011). For instance, Hijosa-Valsero et al. (2011) found significantly higher degradation of CAF during summer, with removal efficiency ranging from 80 to 100%, compared to winter, where removal efficiency ranged from 20 to 80%. In addition, a study by Matamoros et al. (2016) focusing on VSFW reported considerably higher removal efficiency in summer (75%) at 19 °C compared to winter (2%) at 10 °C. Moreover, the HRT was found to play a crucial role, as a longer HRT allows more time for pollutant adaptation/assimilation by microbes and physicochemical processes like adsorption. Consistent with these observations, the MLR model indicates that key parameters in CAF removal are T, D, VSFW and SFW configurations, and HRT, in that order.

Although our analyses do not indicate a significant effect of plants on CAF removal in either Spearman's correlation or the MLR model, some studies have suggested the possibility of CAF adsorption onto plant root surfaces as a removal mechanism (Hijosa-Valsero et al., 2016). It is important to consider these various factors collectively to gain a comprehensive understanding of CAF removal in CW systems.

3.2.2. Ibuprofen (IBU)

Aerobic biological removal of IBU in CW has been extensively identified as an effective operational condition (Zwiener and Frimmel, 2003; Zhai et al., 2013; Li et al., 2016; Zhang et al., 2017; AL Falahi et al., 2021). Our study's dataset corroborates the positive influence of aerobic conditions in IBU removal, where VSFW present the highest removal efficiency with 74 ± 28%, followed by HSFW with 52 ± 28%, and SFW with 28 ± 13%. Further, our Spearman's correlation analysis shows a positive correlation between IBU removal and T ($r = 0.424$, $p < 0.01$) and negative correlations with D ($r = -0.292$; $p < 0.01$), where a higher D can lead to less aerobic conditions. As such, these correlations corroborate the significance of aerobic environments during the microbial degradation of IBU. However, it is worth highlighting that the effect of D on IBU removal remains inconclusive in our study, with a negative correlation in Spearman's correlation but a positive coefficient in the MLR model.

Photodegradation has also been considered an important removal process for IBU in CWs (Zhang et al., 2014a,b). However, our results do not support this observation, as both Spearman's correlation analysis and the IBU MLR model show negative correlation and coefficient values, respectively, with respect to the SFW configuration, which is the configuration that can facilitate sunlight exposure, and thus photodegradation processes. Along this line, other works have observed the physical IBU removal process, such as adsorption, to be negligible (Zhang et al., 2017). Hence, further investigation is required to fully comprehend these phenomena.

Plant uptake is considered the major IBU removal process in CWs, as macrophytes can enhance IBU removal efficiency through several mechanisms. Firstly, plants release oxygen from their roots, promoting high microbial activity and thereby accelerating IBU degradation under oxic conditions (Zwiener and Frimmel, 2003). Secondly, plants release root exudates that enhance microbial activity in the rhizosphere, leading to increased IBU degradation (Brix, 1997; Zhai et al., 2013). Thirdly, plants can take up and metabolize IBU themselves (Zwiener and Frimmel, 2003). However, our analyses do not observe a significant effect of plants on IBU removal in either Spearman's correlation or the MLR model.

In summary, our results indicate that aerobic conditions play a crucial role in IBU removal in CW, while photodegradation's relevance is yet to be determined. Moreover, plant-related processes appear promising but require further investigation to establish their significance accurately.

3.2.3. Naproxen (NPX)

NPX removal in CWs has been reported to be primarily driven by biological processes (Zhang et al., 2017; Petrie et al., 2018). It has been observed that NPX is transformed by oxygenases by a range of aerobes in soils and wastewater (Domaradzka et al., 2015; Vulava et al., 2016; Garcia-Becerra and Ortiz, 2018; Zapata-Morales et al., 2020). At the same time, it has also been observed that NPX biodegradations can take place under anaerobic conditions (Alvarino et al., 2018). Thus, NPX can be simultaneously transformed through aerobic and anaerobic processes (Tiwari et al., 2017). In our results, we only observe a significant correlation between aerobic conditions and NPX removal. The Spearman's correlation analysis shows positive relationships between NPX removal and T ($r = 0.597$, $p < 0.01$), and negative correlations with D ($r = -0.735$; $p < 0.01$), HRT ($r = -0.433$, $p < 0.01$), and CODin ($r = -0.327$; $p < 0.01$). Furthermore, our study's dataset with aerobic conditions positively influencing NPX removal, with VSFW exhibiting the highest mean removal efficiency (71 ± 14%), followed by HSFW (56 ± 31%), and SFW (42 ± 16%). To make more accurate observations, DO would need to be followed in future studies, and these values could then be included in data-driven models such as the MLR tool used in this work.

Consistent with the significance of biological processes in NPX removal, T has been identified as a key variable affecting NPX removal (Zhang et al., 2018). Studies have reported removal efficiencies of NPX ranging from 55.6% to 82.3% in summer, 51.6%–74.9% in autumn, and 22.1%–50.5% in winter (Zhang et al., 2018). Our findings are in line with these results, revealing a positive relationship between T and NPX removal in both Spearman's correlation and MLR analysis.

3.2.4. Diclofenac (DCF)

Removal mechanisms of DCF in CWs encompass photodegradation, biodegradation (both aerobic and anaerobic), and plant uptake (D. Zhang et al., 2014). Photodegradation, mainly driven by sunlight, is considered the major removal pathway for DCF (Andreozzi et al., 2003; Méndez-Arriaga et al., 2008; Matamoros et al., 2016). However, in planted systems, the progressive growth of biomass (leaves and roots) can lead to shading, hampering photodegradation and subsequently reducing removal efficiencies (Matamoros et al., 2012). Additionally, DCF biodegradation appears to occur through multiple pathways, with aquatic plants playing a role in removal through plant uptake (21%) and involvement of arbuscular mycorrhizal fungi (AMF) (Matamoros et al., 2012). These complex relationships are evident in the Spearman's correlation analysis, where DCF removal shows positive correlations with T ($r = 0.493$, $p < 0.01$) and negative correlations with D ($r = -0.687$; $p < 0.01$), HRT ($r = -0.400$; $p < 0.01$), and CODin ($r = -0.363$; $p < 0.01$).

The findings from Spearman's correlation and MLR analyses support the observations that photodegradation promotes DCF removal. The MLR model identifies SFW as the most efficient configuration, which in turn can lead to photodegradation processes. Furthermore, both Spearman's correlation and MLR analyses highlight T as a key variable favoring DCF removal. Zhang et al. (2018) reported varying DCF removal efficiencies during summer (42.2–68.3%), autumn (42.1–52.9%), and winter (41.7–47.7%), likely influenced by the increased T and sunlight exposure in warmer seasons, promoting DCF photodegradation.

3.2.5. Ketoprofen (KTP)

KTP has been reported to be removed by photodegradation (Salgado et al., 2013). The KTP MLR model shows the SFW configuration and T as key variables promoting KTP removal. Our dataset agrees with Salgado et al. (2013), where SFW has the highest mean removal efficiency (63 ± 18%), followed by the VSFW (49 ± 2%) and HSFW (45 ± 36%). Further, T has been shown to significantly influence KTP removal (Matamoros and Bayona, 2006; Zhang et al., 2018). Different authors have discussed that temperature influence KTP removal due to higher solar radiation at warm temperatures that can in turn promote effective photolysis.

The influence of temperature in KTP removal can also be related to higher biological activity in warmer conditions. It has been observed that biological-driven degradation in KTP removal, primarily include reduction of the keto group, deoxygenation and oxidation (Rastogi et al., 2021). Moreover, fungal microorganisms such as white-rot fungi, which are strict aerobes, have been identified as highly effective in KTP removal (Domaradzka et al., 2015; Rastogi et al., 2021).

The positive influence of aerobic biological processes in KTP removal can be observed in the Spearman's correlation analysis, where there are positive correlations with HLR ($r = 0.668$, $p < 0.01$), T ($r = 0.588$, $p < 0.01$), and negative correlations with D ($r = -0.799$; $p < 0.01$), HRT ($r = -0.540$, $p < 0.01$), and CODin ($r = -0.503$; $p < 0.01$).

3.2.6. Carbamazepine (CBZ)

In general, CBZ has been reported as a highly recalcitrant pollutant in wastewater treatments, including CWs (Chen et al., 2018). This was also observed in our dataset, as CBZ presented the lowest removal efficiency (24 ± 21%) with respect to the other PPCPs. Overall, it is uncertain which removal mechanism leads to the degradation process. Some authors have discussed that CBZ is removed mainly by physicochemical processes such as adsorption and sorption, while other possible removal mechanisms include plant uptake, aerobic biodegradation and reductive enzymatic transformations (Chen et al., 2018; Nasir et al., 2018). Previous works have shown that temperature affects the removal of the pollutant, including (Hijosa-Valsero et al., 2011), who reported higher CBZ removal in summer (0–58 ± 21%) than in winter (0–9±100 %). This can be because the mentioned biological mechanisms are temperature dependent. Spearman's correlation and CBZ MLR analysis agree with these findings, indicating T as one of the key variables that enhance CBZ removal.

Although nitrogen was not included in this work due to a lack of data, it has been reported that CBZ might be removed by co-metabolism during nitrification (Men et al., 2017; Wang et al., 2023). Oxygen levels have also been reported to influence the removal of CBZ. Tejeda et al. (2015) reported higher efficiencies (60 ± 4.5%) in systems with low DO concentrations, Redox negative values and pH conditions near to 8. In contrast, aerobic conditions demonstrated less efficiency (36 ± 4.4%). In general, it is important to highlight that significant gaps still need to be addressed to understand CBZ removal in wastewater treatments (Nasir et al., 2018; Chen et al., 2018).

3.2.7. Sulfonamide compounds (SUL)

Dan et al. (2013) found that increasing the HLR from 0.125 to 0.25 m/d resulted in higher average removal efficiencies for sulfamethazine (from 0 to 13%), while sulfapyridine remained constant at 85–86%. Overall, a higher HLR can lead to a higher influx of organic matter into the wetlands, subsequently boosting microbial activity and biomass, and ultimately promoting pollutant degradation. The SUL MLR model supports these findings, with HLR identified as one of the main predictive parameters for pollutant removal. However, it is worth noting that if the HLR keeps on increasing, the HRT may then reduce, leading to a reduction in pollution removal efficiencies. Furthermore, the Spearman's correlation analysis revealed a positive correlation between SUL removal and T ($r = 0.346$; $p < 0.01$), suggesting a temperature-driven removal process.

3.2.8. Chemical oxygen demand (COD)

COD is an indicator of conventional contaminants, which tend to be removed mainly biologically. This is observed in the COD MLR model, where the increase in CODin and T promotes COD removal, which has been seen by Murat Hocaoglu et al. (2010). Depth and VSFW also positively correlate to COD removal, while SFW is negatively correlated, indicating again the presence of microbial driven processes. For the most part, the Spearman's correlation analysis indicates that COD removal depends on the same variables as those in the COD MLR model. However, T and HLR have a negative correlation with COD removal. In the case of higher T values, these might lead to microbial overgrowth or other possible operational upsets that could lead to an increase in suspended solids in the effluents, and hence lower COD removal rates. As already mentioned in section 3.2.7, high HLR values could also lead to insufficient contact time to effectively remove COD.

3.2.9. Commonalities among PPCPs

3.2.9.1. Operational parameters. MLR analysis indicates that T greatly influences PPCP removal rates (Table 4). Higher T values will improve the elimination of most PPCPs, except for SUL and IBU, where no statistically significant impact was observed. Therefore, it is desirable to monitor this parameter to better manage or improve the removal PPCPs on their own or as mixtures. As temperatures lower, other operating conditions could be modified, such as HLR, HRT, to compensate for the loss of biological activity.

CODin is also an important parameter to monitor. While the MLR analysis does not show significant influence of CODin on PPCP removals, the Spearman's correlation analysis suggests that CODin negatively correlates with the removal of PPCPs, except for CAF and SUL. COD measures the primary carbon and energy source for microbial growth; when sufficient COD is available, microorganisms are less inclined to co-metabolize other compounds, such as PPCPs. This may indicate the need to set sequential treatment stages, where COD can be reduced in one stage to promote PPCP removal in a follow-up stage. However, further analysis is needed to evaluate this strategy.

Both HLR and HRT do not have significant effect on PPCP removal, except for SUL and CAF, respectively. This is important to consider, since increasing these values may increase operational costs, which may be unnecessary if CAF removal is a treatment objective, since CAF removal can be promoted by other parameters. However, SUL removal appears to be influenced by fewer variables, and thus, increasing HLR would be a suitable approach if SUL removal is a treatment objective.

3.2.9.2. Design parameters. The MLR model results indicate that the SFW configuration is negatively correlated with the removal of half of the PPCPs. Therefore, it is advisable to use subsurface flow configurations for treating PPCP mixtures, if feasible. SFW is recommended only when DCF and KTP are the specific pollutants targeted for removal.

D was observed to affect PPCP removal. However, D can be related to a range of removal mechanisms, mainly aerobic degradation, adsorption, and photolysis. Thus, further studies are needed to better understand the phenomena being promoted through this parameter.

Finally, in the works we analyzed, we did not find a significant influence of planted or unplanted designs in the removal of PPCPs.

3.3. Greenhouse gases (GHGs)

Wastewater represents an important source of C and N for microbial development. GHGs are produced during wastewater treatment mainly due to microbial metabolisms. Consequently, parameters affecting the microbial activity would be the ones governing the GHGs emission rates. CH_4 and CO_2 are linked to the carbon cycle, while N_2O generation is associated to both nitrogen and carbon cycles.

Seasonal variability may affect plant productivity and represent temperature changes that may influence GHGs emission rates. Both parameters promote higher activity during summer when temperatures are higher and most of the vegetation has bloomed. The presence of plants influences GHG emission rates in three ways: (1) root exudates result favorable for microbial communities' establishment, (2) plants allow gas transportation from and into the water, increasing oxygen levels in the rhizosphere and releasing the gases produced in the CW substrate/water, (3) plants play a role in carbon sequestration due to their photosynthetic metabolism. According to Picek et al. (2007), plants can contribute significantly to the net carbon balance, representing half of that coming from the influent wastewater. This carbon includes decomposing material from roots (15%), rhizomes (7%), and root exudates (78%). These parameters influence the development of microenvironments in CWs by promoting the formation of different zones in the same system, ranging from completely anaerobic to aerobic. Specifically, oxygen availability, reflected in DO concentration, is one of the main parameters which in turn will be affected by temperature, HLR, and other factors.

Our data analysis found that the most recurrent GHG investigated was CH_4, with 259 data records, followed by N_2O (147) and CO_2 (130). The average emission rates for the three GHGs were 170.2, 13.4 and 0.32 mg/m^2h for CO_2, CH_4 and N_2O, respectively. Although CO_2 represented the GHG with the highest value (2.2×10^3 mg/m^2h, Fig. 3), it is known that the warming effect of CH_4 and N_2O on climate change is higher, with CO_2 equivalent values of 28 and 300, respectively (IPCC, 2013). Hence the importance of monitoring these other two gaseous products, as reflected in the number of studies and data from the last 10 years. We found that the VSFW configuration, *Cyperus papyrus* and *Typha sp.* exhibited the highest average GHG emissions compared to other configurations and plant species (Figs. S3 and S4).

3.4. Design and operational parameters affecting GHG emission rates

Tables 5 and 6 show the Spearman's correlation and the MLR analysis results for the three GHGs evaluated in this study. Since these GHGs are mainly produced through different microbial metabolic pathways, parameters such as CODin, T, and D demonstrated strong positive correlations in most of the cases. Therefore, when microbial activity is enhanced by higher T and inlet carbon concentration (CODin), it is natural to observe an increase in global and specific emission rates. All three GHGs can be generated in a wide range of environments, from aerobic to strictly anaerobic. Consequently, D becomes an important design parameter, as modifying it could represent the development, switch or adaptation, of a microbial community towards specific metabolic routes.

The HLR is a crucial parameter for the design and operation of wastewater treatment systems. It can control and regulate the efficiency of the system in treating specific pollutants and determines their distribution in the CW, which in turn affects the rate of microbial uptake. Its impact on GHG emission rates varies. Spearman's correlation shows a positive effect for CH_4, while demonstrating an opposite effect for CO_2 and N_2O. This means that although reducing HLR can improve treatment and pollutant removal, it can result in higher GHG emissions.

3.4.1. Methane (CH_4)

The emission rate of CH_4 in CW involves both production and consumption processes (Segers, 1998). The production of CH_4 is primarily carried out by methanogenic archaea in anaerobic conditions. Conversely, CH_4 biodegradation mainly occurs in environments where oxygen is not limited, but it can also take place in molecular oxygen-deprived conditions (anoxic) coupled with the nitrification process (Schalk et al., 2019). Another strategy for CH_4 removal in CW is the use of substrates that can absorb this pollutant. Ji et al. (2020a) investigated the influence of using biochar to improve water treatment and reduce net CH_4 emissions. Interestingly, their results show that, while using the same substrate, intermittent air addition decreased the CH_4 emission rate more efficiently; since the additional oxygen supply at the bottom of the system inhibited the methanogenic activity and promoted methane oxidation metabolism.

According to Spearman's correlation and MLR analysis (Tables 5 and 6), CODin is one of the main parameters affecting positively ($r = 0.285$, $p < 0.01$; 0.207) the CH_4 emission rate in CWs. Most of the organic carbon comes from the wastewater. However, it has been demonstrated that there are more sources of CH_4 to be considered. Firstly, dissolved CH_4 can be found in anaerobic effluents from pretreatment processes such as UASB reactors or septic tanks (Diaz-Valbuena et al., 2011; Schalk et al., 2019). These effluents can apport additional CH_4 to the emissions in a wetland when treating incoming water from anaerobic systems.

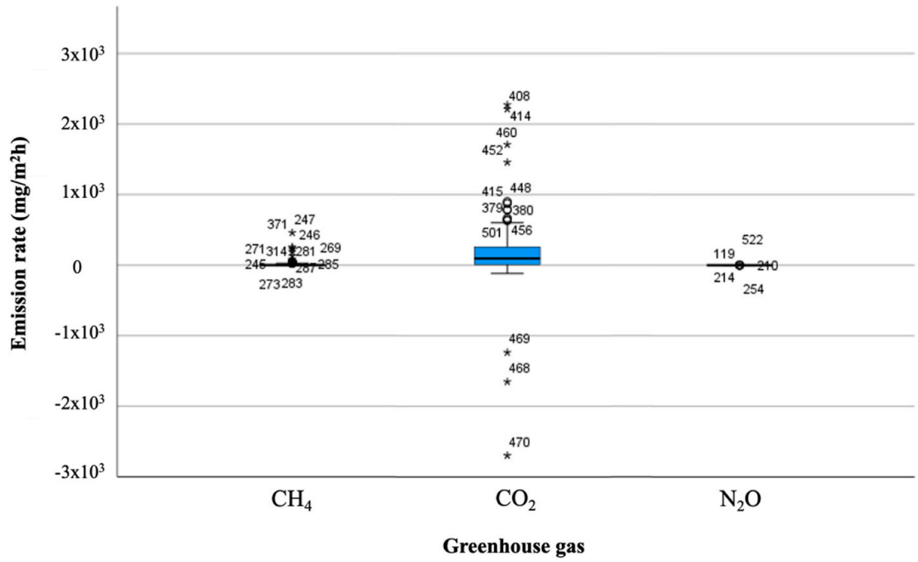


Fig. 3. GHG emission rates in CWs. The number of data points reviewed for the emission rates of GHG was as follows: CH_4 (259), CO_2 (130) and N_2O (147).

Table 5
Spearman's correlation statistics among the studied factors and the emissions of the selected GHGs.

Variable	COD_{in}	T	D	HLR	HRT
CH_4	**0.285**[b]	0.071	**0.170**[b]	**0.286**[b]	−0.077
N	*189*	*240*	*259*	*149*	*175*
CO_2	**0.503**[a]	0.123	0.121	**−0.292**[a]	0.115
N	*82*	*117*	*130*	*57*	*74*
N_2O	0.012	**0.318**[b]	0.027	**−0.293**[b]	0.0
N	*131*	*130*	*147*	*109*	*135*

[a] Correlation is significant at the 0.05 level (2-tailed). In bold.
[b] Correlation is significant at the 0.01 level (2-tailed). In bold.

Table 6
MLR equations for the GHGs.

Pollutant	R^2	R^2 Adj.	P value	Equation
CH_4	0.40	0.36	<0.001	C = 0.207CODin +0.364HLR - 0.698HRT +0.907SFW
CO_2	0.52	0.48	<0.001	C = 0.620CODin +0.322T - 0.180HRT
N_2O	0.37	0.33	<0.001	C = 0.201CODin +0.329T + 0.762D −1.101HLR −0.518HSFW

Also, planted CWs favored CH_4 emissions because the carbon released from dead biomass and root exudates promoted organic matter degradation (Wu et al., 2017). However, Niu et al. (2015) compared planted CWs with an unplanted control, finding values four times higher in the planted systems. They also found that organic compounds released from plants have a greater influence over methanogenic communities' development in contrast to their effect on methanotrophic organisms. In general, from our review, unplanted and planted systems presented average CH_4 emission rates of 4.3 (n = 46) and 15.4 (n = 213) mg/m^{2}h, respectively.

HLR is an important design parameter in CWs and has a strong influence on CH_4 emission rates according to Spearman's correlation (r = 0.286, p < 0.01) and MLR analysis (0.364). HLR positively affects CH_4 emission, whereas larger HRT will decrease the emission rates, as suggested by the negative correlation in the MLR analysis. This occurs because HRT may influence both production and consumption metabolisms of methane. So, although longer HRTs imply more time for carbon uptake/transformation into CH_4, the methanotrophic rate usually surpasses the methanogenic ones (Wu et al., 2017; Zheng et al., 2018; Chen et al., 2020).

It is evident why D promotes methane production (r = 0.170, p < 0.01), as anaerobiosis occurs far from the surface where oxygen limitation is predominant. Deeper CW means less oxygen availability from the exchange with the ambient (air), but also from the roots of the plants (Huang et al., 2013).

Since CH_4 is mainly produced in strictly anaerobic conditions, it is expected to be primarily found in HSFW and SFW, where oxygen levels are usually low throughout the entire system (Ji et al., 2020b). In HSFW, a water table D above 30 cm will generate conditions that promote anaerobic microbial activity (Aguirre et al., 2005). However, Zhou et al. (2017) observed that VSFW could exhibit low DO values in the effluent since the availability of carbon can lead to high biodegradation rates and, thus, the consumption (reduction) of DO. Furthermore, a study by Ding et al. (2012) demonstrated that higher COD/N ratios using synthetic wastewater resulted in lower DO concentrations in the effluent, likely due to oxygen consumption for the excess of organic matter availability.

3.4.2. *Carbon dioxide (CO_2)*

CO_2 is another byproduct of anaerobic metabolism, but it is also the main product of the aerobic mineralization of organic matter. Its emission rate is also a quantification of antagonistic processes since the net flux will include the CO_2 produced from the decomposition of the organic matter (from water and biomass in the system), and the sequestration by plants and autotrophic microorganisms, that can even result in a net negative emission rate value (De Klein and Van der Werf, 2014; Kasak et al., 2015).

Spearman's correlation analysis demonstrated a significant positive effect of CODin (r = 0.503, p < 0.05) over CO_2 emission rates. Similarly to CH_4, higher amounts of organic compounds in the inlet water (CODin) favor the emission rate of CO_2. Yan et al. (2012) corroborated this affirmation in their study, where they found that CO_2 emissions were positively correlated to the C loading, regardless of the N concentration, while testing a VSFW using synthetic municipal wastewater. On the other hand, HLR (r = −0.292, p < 0.05) presented a significant negative correlation over the net CO_2 flux in CWs. Mburu et al. (2013) found a negative correlation between the efficiency of COD removal and HLR in an HSFW CW. Their results showed that higher HLR reduced the system's efficiency to eliminate organic matter. Since CO_2 comes directly from organic matter conversion, it makes sense that its emission is reduced when less is degraded. Results from MLR analysis showed a contradiction in the HRT effect over CO_2 net flux compared to Spearman's correlation analysis. The MLR analysis showed a significant negative impact, while Spearman's correlation found a non-significant positive correlation. The contrasting results are examples of the counter processes happening in a CW and related to CO_2 production and capture.

T is another factor influencing CO_2 emissions, as demonstrated by the MLR analysis. As previously mentioned, temperature (often linked to seasonal variations) will affect not only microbial activity but also vegetation performance, which in turn regulates the amount of root exudates, plant biomass, and gas transportation from and into the system (Wu et al., 2016). Although vegetation was not included in our analysis, the average results from planted and unplanted systems' CO_2 emission rates were 182.8 and 92.1 mg/m^{2}h.

From the data collected, the HSFW configuration showed the most variability in CO_2 emission rates, ranging from -2.7 to 2.2×10^3 mg/m^{2}h. Therefore, it exhibited significantly lower average values for CO_2 emission rates (83.5 mg/m^{2}h) compared to VSFW (526 mg/m^{2}h) and SFW (402 mg/m^{2}h). Despite these differences, the MLR analysis did not find a significant effect of the CW configuration on the overall CO_2 emission rate.

3.4.3. Nitrous oxide (N_2O)

The formation of N_2O is directly related to N removal in CWs. It can occur either as a byproduct of incomplete nitrification or as an intermediate during denitrification microbial processes. As these processes can take place simultaneously in different microenvironments in the CWs, depending on the C and N load and oxygen availability, N_2O release can be achieved in any configuration (Hu et al., 2023). This was clearly corroborated by our dataset as unlike CO_2 and CH_4 emissions, N_2O average emission rates were in the same magnitude order for the 3 configurations. N_2O emission rates mean values were 0.35, 0.17, and 0.34 mg/m^{2}h in HSFW, VSFW, and SFW.

However, N assimilation and removal can also be achieved by plants or physical mechanisms such as sedimentation, adsorption, and volatilization (Vymazal, 2007). Therefore, the influence of a wider range of factors affecting the N_2O emission rate was found after performing the Spearman's correlation and more clearly in the MLR analysis. For the first, the T ($r = 0.318$, $p < 0.01$) was positively related to N_2O fluxes, while HLR ($r = -0.293$, $p < 0.01$) presented a negative correlation. Wu et al. (2013) performed a study that corroborated a seasonal effect with the highest N removal and N_2O production rates during summer. The latter is addressed to changes in temperature, which directly affect microbial activity and vegetation. On the other hand, the correlation between emission rate and D was found to be positive, meaning that CWs with higher water table D are prone to emit more N_2O. Deeper CWs could lead to the development of a more varied community of N-removal microorganisms, such as anammox, allowing the system to assimilate N without generating N_2O emissions (Dong and Sun, 2007).

Similarly to the other two GHGs, N_2O is affected by CODin, as N biotransformation depends on the presence of organic sources. There are several studies relating to the C and N ratios to improve water treatment and evaluate N_2O emissions. A C/N ratio of 5 has been described as optimal when the purpose is pollutant removal and N_2O reduction (Niu et al., 2023). Xu et al. (2021) studied different substrates and their effect on N_2O production. Results showed that the CW packed with walnut shell emitted less N_2O than gravel, Manganese ore, and activated albumin as substrates. The explanation was that walnut shell has a higher organic content, providing an additional carbon source and therefore, favoring complete denitrification of nitrates to N_2, limiting N_2O generation. Similarly, the HSFW water flow regime, with a negative effect according to MLR analysis, could allow the development of larger anaerobic zones within the CW, promoting the reduction of nitrates into N_2 and decreasing the concentration of intermediate compounds, such as N_2O.

3.5. Coupling PPCPs removal & GHG abatement

When assessing CWs as a suitable technology for circular cities, the treatment objectives are to enhance the removal of PPCPs while mitigating the emission of GHGs. Our meta-analysis reveals several key parameters that influence both PPCP removal and GHG emissions, providing insights into potential strategies for optimizing CW design and operation (Table 7).

T emerged as the variable with the most influence in the studied CWs, showing a positive correlation with both PPCP removal and GHG emission. Higher T values enhance the removal of most PPCPs but will increase emissions of N_2O and CO_2. This dual effect is primarily due to augmented microbial activity and vegetation metabolism at higher temperatures. While temperature cannot be directly controlled in most CW systems, understanding its impact is crucial for predicting and managing treatment performance across seasons. In colder climates or seasons, other operational parameters such as HLR or HRT could potentially be adjusted to compensate for reduced biological activity. In our work, we did not focus on the carbon sequestration capabilities of CWs, which could be a countermeasure to GHG emissions.

The SFW configuration also showed significant but often contrasting effects on PPCPs and GHGs. It was found beneficial for the elimination of DCF and KTP but decreased the removal of CAF, NPX, DCF and COD while promoting CH_4 production. This suggests that SFW may not be the optimal configuration to simultaneously remove PPCPs and reduce GHG emissions. Subsurface flow configurations (HSFW and VSFW) generally showed more favorable results for PPCP removal with less promotion of GHG emissions, indicating they may be preferable for achieving both objectives.

Other important parameters are D, HLR, and HRT with decreasing relevance. Our analysis showed that increasing D showed potential for improving the removal of some PPCPs (CAF, IBU, COD) but also promoted N_2O emissions. Higher HLR values promoted CH_4 emissions without significantly affecting most PPCP removals (except SUL). Since HLR does not significantly affect the removal of micropollutants (except SUL), it is recommended to operate CWs at lower HLR to avoid GHG emissions. HRT primarily affected CAF removal and CH_4/CO_2 emission rates, with longer HRTs potentially beneficial for reducing the CH_4 emission rate while maintaining PPCP removal efficiency. Therefore, HRT can be useful mainly when CH_4 emissions need to be reduced, since CAF removal is influenced by other 5 variables.

The VSFW configuration was also observed to be strategic. This configuration promotes the removal of CAF and COD; however, it also enhances the production of CO_2. Hence, the choice of using this configuration must be in accordance with the target PPCPs and ability to offset CO_2 emissions.

The CODin parameter also emerged as a critical parameter to monitor. While it did not significantly influence PPCP removals in the MLR analysis, it showed negative correlations with several PPCPs in the Spearman analysis and significantly fostered GHG emissions. This suggests that a two-stage treatment approach, where COD is reduced in an initial stage before focusing on PPCP removal, could be beneficial for overall treatment goals and GHG mitigation.

These findings highlight the complex interplay between PPCP removal and GHG emissions in CWs. Optimizing CW design and operation requires careful consideration of these parameters to balance treatment efficacy with environmental impact. For example, while subsurface flow configurations may offer a good compromise for most PPCPs and GHGs, specific treatment goals (e.g., targeting certain PPCPs) may necessitate different approaches. Similarly, strategies to manage temperature effects, such as adjusting HLR or HRT seasonally, could help maintain treatment efficiency year-round while minimizing GHG emissions.

Future research should focus on developing integrated models that can predict both PPCP removal and GHG emissions based on these critical parameters. This would allow for more precise optimization of

Table 7
Coefficients associated to operational and design parameters affecting PPCP removal, COD removal and GHG emission.

	T	SFW	D	HLR	HRT	VSFW	HSFW	Cin	COD_{in}
CAF	0.23	*−0.13*	0.13		0.12	0.15		0.91	
IBU		*−0.19*	0.15					1.00	
NPX	0.20	*−0.17*						0.91	
DCF	0.07	0.10						0.91	
KTP	0.17	0.17						0.80	
CBZ	0.27							0.79	
SUL				0.24				0.68	
COD	0.09	*−1.00*	0.05			0.08			1.02
CH_4		*0.91*		*0.36*	−0.70				*0.21*
CO_2	*0.32*				−0.18				*0.62*
N_2O	*0.33*		*0.76*	−1.10			−0.52		*0.20*

Note: Values in italics indicate a negative impact (i.e., decrease the PPCPs removal and promote GHG emissions).

CW systems for specific climates, influent characteristics, and treatment goals. Additionally, exploring novel design elements or operational strategies that can enhance PPCP removal while actively sequestering carbon or promoting methane oxidation could further improve the sustainability of CW systems.

By leveraging our understanding of these interconnected processes, we can work towards developing next-generation CW systems that not only effectively remove emerging contaminants but also contribute positively to climate change mitigation efforts. This holistic approach to CW design and operation is essential for realizing the full potential of these systems for sustainable urban water management practices.

4. Conclusions

This meta-analysis provides insights into the complex relationships between PPCP removal and GHG emissions in CWs. Our findings highlight several key conclusions and directions for future research.

1. Biological processes, particularly secondary metabolism and co-metabolism, play a crucial role in both PPCP removal and GHG emissions. This underscores the importance of understanding microbial community dynamics in CW design and operation.
2. T emerged as a critical factor influencing both PPCP removal and GHG emissions. Since T cannot be practically modified in CWs, it can be considered a major disturbance from an operational control perspective. Future CW designs could incorporate strategies to optimize performance through adaptive control systems.
3. Subsurface flow configurations (HSFW and VSFW) generally showed more favorable results for balancing PPCP removal and GHG mitigation compared to SFW systems. This suggests a preference for subsurface designs.
4. Influent characteristics, particularly organic matter content (CODin), significantly impact both treatment efficacy and GHG emissions. Developing pre-treatment strategies or multi-stage systems to manage influent composition could enhance overall CW performance.
5. Hydraulic parameters (HLR, HRT) offer potential for operational optimization, allowing for adjustments to balance PPCP removal and GHG emissions based on seasonal or other environmental factors.
6. The study reveals the need for integrated monitoring approaches in CWs, simultaneously tracking PPCP removal, GHG emissions, and carbon sequestration to fully assess system sustainability.

A limitation of our work lies in coupling the observations of PPCPs and GHGs studies, which were not originally conducted together. Thus, we cannot fully elucidate the direct link between the PPCP removals and the GHG emissions in the case when the same factor indicates to have a significant effect in these two outcomes. To overcome this limitation, future research should strive to perform simultaneous measurements of PPCP removal, GHG emissions and possibly carbon sequestration capabilities in CWs. We recommend studying these three phenomena together while monitoring additional parameters such as nitrogen, redox potential, pH, and dissolved oxygen to gain a deeper understanding of the underlying processes. Other future research directions could include:

1. Development of comprehensive models integrating PPCP fate, GHG emissions, and carbon sequestration in CWs to enable more precise system optimization.
2. Investigation of novel materials or design elements that can enhance PPCP removal while actively mitigating GHG emissions or promoting carbon sequestration.
3. Long-term studies assessing the stability and resilience of optimized CW systems under varying environmental conditions and influent compositions.
4. Exploration of microbial community engineering strategies to promote desired metabolic pathways for both PPCP degradation and GHG mitigation.
5. Assessment of the potential for coupling CWs with other green technologies (e.g., algal systems, biochar amendments) to further enhance sustainability outcomes.

This study underscores the potential for CWs to play a significant role in sustainable urban water management, addressing both emerging contaminant concerns and climate change mitigation. By adopting an integrated approach to CW design and operation, informed by the parameters and relationships identified in this meta-analysis, we can work towards developing next-generation nature-based solutions for water treatment that are both effective and environmentally sustainable. The path forward requires collaborative efforts between researchers, engineers, and policymakers to translate these insights into practical, implementable strategies for the water sector.

CRediT authorship contribution statement

M.A. Salinas-Toledano: Writing – review & editing, Writing – original draft, Validation, Software, Methodology, Formal analysis, Data curation, Conceptualization. **T.L. Gómez-Borraz:** Writing – review & editing, Writing – original draft, Validation, Supervision. **M.A. Belmont:** Writing – review & editing, Validation. **F.Y. Garcia-Becerra:** Writing – review & editing, Supervision, Resources, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:F. Y. Garcia-Becerra reports financial support was provided by the Mexican National Council for Science and Technology (CONACYT). If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2024.120144.

Data availability

Data will be made available on request.

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