Constructed wetlands efficiency in removing pollutants and organic matter.

María Hijosa- Valsero
Departamento de Química y Física Aplicadas
Facultad de Ciencias Biológicas y Ambientales, Universidad de León
24071, León, Spain
mhijv@unileon.es
Maria Cruz Valsero Blanco
Departamento de Estadística e Investigación Operativa
Facultad de Ciencias, Universidad de Valladolid
47005, Valladolid Spain
mcruz@eio.uva.es
Ricardo Sidrach-Cardona
Instituto de Medio Ambiente,
Universidad de León, C/La Serna 58
24007, León, Spain
rsidm@unileon.es
Javier Martín-Villacorta
Departamento de Química y Física Aplicadas
Facultad de Ciencias Biológicas y Ambientales, Universidad de León
24071, León, Spain
jmarv@unileon.es
Josep M. Bayona
IDAEA-CSIC,
C/Jordi Girona 18-26
E-08034, Barcelona, Spain
jbtqam@cid.csic.es
Eloy Bécares IDAEA-CSIC,
Departamento de Biodiversidad y Gestión Ambiental,
Facultad de Ciencias Biológicas y Ambientales, Universidad de León,
24071, León, Spain
ebecm@unileon.es

ABSTRACT

Constructed wetlands (CWS) have been used and studied for several decades for the treatment of urban sewage and industrial wastewaters. Few works are centered on the topic of which type of CW is the most appropriate for the removal of organic pollutant under the same climate and wastewater conditions.

Eight CWs were set up in the open air inside the facilities of the León conventional activated sludge wastewater treatment plant (WWTP), in the northwest of Spain.

By processing our data we pointed out the importance of temperature, dissolved oxygen concentration, redox potential values, plant presence and chemical structure in the elimination of most pollutants.

1. Introduction

Constructed wetlands (CWs) have been used and studied for several decades for the treatment of urban sewage from small communities and for the treatment of several kinds of industrial and agricultural wastewaters. They consume little energy and their maintenance costs are relatively low; in addition, they show a high capacity to remove organic micropollutants, like pharmaceuticals and personal care products

(PPCPs) (Matamoros et al., 2005, 2008; Park et al., 2009). But the mechanisms involved in this removal are largely unknown. The coexistence of several micro environments in CWs allows for a variety of microbiological communities, which might be able to offer different metabolic pathways leading to PPCP degradation. This coexistence is related to the variation of physico-chemical parameters on different gradients inside CWs (Imfeld et al., 2009). Some of these variations may be generated by the organisms inhabiting the CW (Stottmeister et al., 2003).

Our aim was to investigate the existence of relationships between physico-chemical parameters in CWs (temperature, pH, conductivity, dissolved oxygen concentration and redox potential) and pollutant removal, by using statistical tools. Statistics can help us in the interpretation of elimination processes inside these treatment systems. The studied pollutants were PPCPs, as well as conventional organic pollutants like chemical oxygen demand (COD), filtered COD (FCOD), biological oxygen demand (BOD_5), total suspended solids (TSS), volatile suspended solids (VSS) and nutrients. In order to achieve this objective, exhaustive data from a previous two-year experiment were statistically treated.

Several authors have tried to model the removal of pollutants in CWs (Langergraber et al., 2009; Llorens et al., 2010). To the best of our knowledge, the employment for the first time of statistical tools such as forward stepwise regression models, clustering tree diagrams, regression trees and direct gradient analysis (specifically, RDA) in the study of pollutant removal inside different types of CWs can be found in(Hijosa-Valsero et al., 2011). Those results together with the presented here would shed some light on elimination mechanisms inside CWs and would simplify design decisions to optimize pollutant removal from urban wastewaters.

2. Material and methods

A database obtained during a previous experiment (Hijosa-Valsero et al., 2010a,b) and comprising physico-chemical parameters, organic loading rates and pollutant removal efficiencies in eight different mesocosm-scale CWs was used (Figure 1). This database contained a total of 168 entries and included removal efficiency values (%) of PPCPs [analgesics (ketoprofen, naproxen, ibuprofen, diclofenac, salicylic acid), anti-epileptics (carbamazepine), stimulants (caffeine) and fragrances (methyl dihydrojasmonate, galaxolide, tonalide)], COD, FCOD, BOD_5, TSS, VSS, ammonia nitrogen (NH_4-N), total Kjeldahl nitrogen (TKN) and orthophosphate. For every entry, physico-chemical parameters (temperature, pH, conductivity, dissolved oxygen concentration and redox potential) were recorded at two depths: 5 cm below the surface of the CWs and 5 cm above the bottom. In addition, the characteristics of the influent wastewater were represented by the input areal loading rates (g m^{-2} day^{-1}) of COD, FCOD, BOD_5, TSS, VSS, NH_4-N, TKN and orthophosphate. The presence or absence of plants (*Typha angustifolia* and *Phragmites australis*) was also considered (Table 1).

León WWTP consists of a primary treatment (screening, sand removal, fat removal and primary clarifier) and a secondary treatment (plug-flow activated sludge with nitrification/denitrification and secondary clarifier). The plant was designed to treat the wastewater of 330,000 equivalent inhabitants with an inflow of 123,000 m^3 d^{-1} and a HRT of about 6 hours. Urban wastewater coming from the primary clarifier of León WWTP was conducted to a homogenisation tank of 0.5 m^3. All the CWs except CW6 were fed with this homogenised wastewater at a continuous flow rate of 50 L d^{-1} (HLR 50 mm d^{-1}). CW6 received a triple flow rate (150 L d^{-1}). CWs received a higher pollutant concentration during cold months (the organic input mass load was approximately 3 g m^{-2} d^{-1} BOD in summer and 10 g m^{-2} d^{-1} BOD in winter). This fact could not be avoided, since our influent wastewater came directly from the primary clarifier and the season-variability of its pollutant load was related to the operational regime of the WWTP.

The systems started up in May 2007. After a stabilization period, three sampling campaigns were carried out, one in summer 2007 (July-September 2007), one in winter 2008 (January-March 2008) and the other in summer (July-September 2008). Influent and effluent grab samples were collected once a week (n=6 during the first summer, n=8 in winter and n=7 during the second summer) at the eight CWs. Wastewater samples were always collected on the same day and at the same time. Influent and effluent samples were collected in one-litre amber glass bottles, which were transported refrigerated (4ºC) to the laboratory, where they were analysed within 24 hours. In addition, physico-chemical parameters (temperature, pH, conductivity, dissolved oxygen and redox potential) were measured *in situ* at two different depths (5 cm below water surface and 5 cm above the bottom of the tank) in each CW and the homogenisation tank. Conventional wastewater quality parameters (COD, FCOD, BOD_5, TSS, VSS, TKN, NH_4-N and orthophosphate) were controlled weekly to characterise the wastewater.

3. Results and discussion

A general linear regression model was applied to our data, to estimate the behaviour of a dependent variable based on several independent variables and/or factors, provided that a linear relation exists between that dependent variable and the rest of variables or factors.

In the present work, the forward stepwise method was used to build the general linear regression models. A different model was calculated for the removal efficiency (%) of every variable (PPCPs, COD, FCOD, BOD_5, TSS, VSS, NH_4-N, TKN and orthophosphate). The independent variables included in the model were the surface and bottom values of temperature (ºC), pH, conductivity (µS cm^{-1}), dissolved oxygen (mg l^{-1}) and redox potential (mV), as well as the input areal loading rates (g m^{-2} day^{-1}) of COD, FCOD, BOD_5, TSS, VSS, NH_4-N, TKN and orthophosphate. The presence or absence of plants was included as a factor.

The parameters of the method were set up at p = 0.05, F = 1, maximum steps = 100, sweep delta = 10^{-7}, inverse delta = 10^{-12}. Table 2 shows the calculated model for every pollutant and their respective R-square values. R-square values above 0.50 (indicating the existence of acceptable linear relations) have been shown. Hence, in our case, only ketoprofen, caffeine, galaxolide, tonalide, NH_4-N and TKN models are to be considered valid. This does not necessarily mean that the removal of the other pollutants is not related to physico-chemical parameters, loading rates and plant presence; it only means that the relationship between them is not linear. To confirm this statement, other statistical tests have been performed (Hijosa-Valsero et al., 2011) i.e. regression trees offer more information about these non-linear relationships (figure 2). In general, it was observed that variables like temperature, conductivity, dissolved oxygen and redox potential affected removal efficiencies. The presence of plants and the characteristics of the influent wastewater (especially orthophosphate loading rate) also modified these efficiencies. The influence of the exterior weather is also recollected in the model (table 3)

Macrophytes enhanced the removal of organic matter and some PPCPs. Temperature conditioned the removal of salicylic acid, caffeine, fragrances and nutrients. The pH value was involved in the elimination of tonalide and nitrogen. Dissolved oxygen concentration and redox potential affected the removal of some PPCPs, COD, BOD_5 and nutrients. The applicability of the obtained results is limited to CWs operating under similar conditions to those registered in the database used.

Table 1. A simplified scheme of the database

Treatment system	Ketoprofen removal efficiency	...	Caffeine removal efficiency	...	BOD_5 removal efficiency	...	Surface temperature	Bottom temperature	...	COD input	BOD_5 input	...	Plants
CW1-day 1	81		92		99		18.3	15.9		7.1	4.3		Yes
CW1-day 2	83		98		99		18.3	15.9		7.1	4.3		Yes
...													...
CW8-day 21	-		99		91		19.5	17.2		7.0	4.1		No

Table 2. Multiple regression equation models and R-square values for the removal of every pollutant in CWs

removal efficiency,	Equation	R-squared
Ketoprofen	$y=2.52*DO_{SUP}+0.12*RP_{SUP}+19.96*NH_{load}-228.83*oP_{load}+11.79*p+35.75$	0.59
Caffeine	$y=2.3*T_{INF}-1.11*COD_{load}+7.008*FCOD_{load}+49.45$	0.65
Galaxolide	$y=1.79*T_{SUP}-0.037*C_{SUP}+2.92*DO_{SUP}+0.053*RP_{SUP}-147.99*oP_{load}+13.443*p+44.48$	0.60
Tonalide	$y=1.79*T_{SUP}-0.042*C_{SUP}+0.06*RP_{SUP}+5.27*DO_{INF}-124.24*oP_{load}+8.22*p+45.98$	0.57
NH_4 -N	$y=0.037*C_{SUP}+5.63*DO_{SUP}+0.048*RP_{SUP}-32.51*pH_{INF}-0.77*COD_{load}+12.4*p+256.38$	0.55
TKN	$y=0.019*C_{SUP}+3.56*DO_{SUP}+0.057*RP_{SUP}-27.93*pH_{INF}-49.89*oP_{load}+8.08*p+243.68$	0.50

Table 3. Unusual Residuals for each adjusted variable

NTK	**NH_4-N**	**Tonalide**	**Galaxolide**	**Caffeine**	**Ketoprofen**
CW 8, Summer 2007	CW4, Winter 2008	CW 2, Winter 2008	CW 3, Winter 2008	CW 2, Winter 2008	CW 1, Winter 2008
CW 6, Winter 2008	CW 4, Winter 2008	CW 3, Winter 2008	CW 3, Winter 2008	CW 4, Winter 2008	CW 4, Winter 2008
CW5, Summer 2008	CW5, Winter 2008	CW 3, Winter 2008	CW 4, Winter 2008	CW 5, Winter 2008	CW 6, Winter 2008
	CW 5, Winter 2008	CW 3, Winter 2008	CW 5, Winter 2008	CW 6, Winter 2008	CW 7, Winter 2008
	CW 6, Winter 2008	CW 4, Winter 2008	CW 7, Winter 2008	CW 8, Winter 2008	CW 7, Winter 2008
	CW5, Summer 2008	CW 5, Winter 2008	CW4, Summer 2008	CW 8, Winter 2008	
		CW 7, Winter 2008	CW4, Summer 2008	CW 8, Winter 2008	
		CW2, Summer 2008			
		CW 4, Summer 2008			

Figure 1. Schematic diagram of the studied constructed wetlands. Notes. FM: floating macrophytes, FW: free-water layer, SF: surface flow, SSF: subsurface flow, 3Q: triple flow. (Hijosa-Valsero et al., 2011)

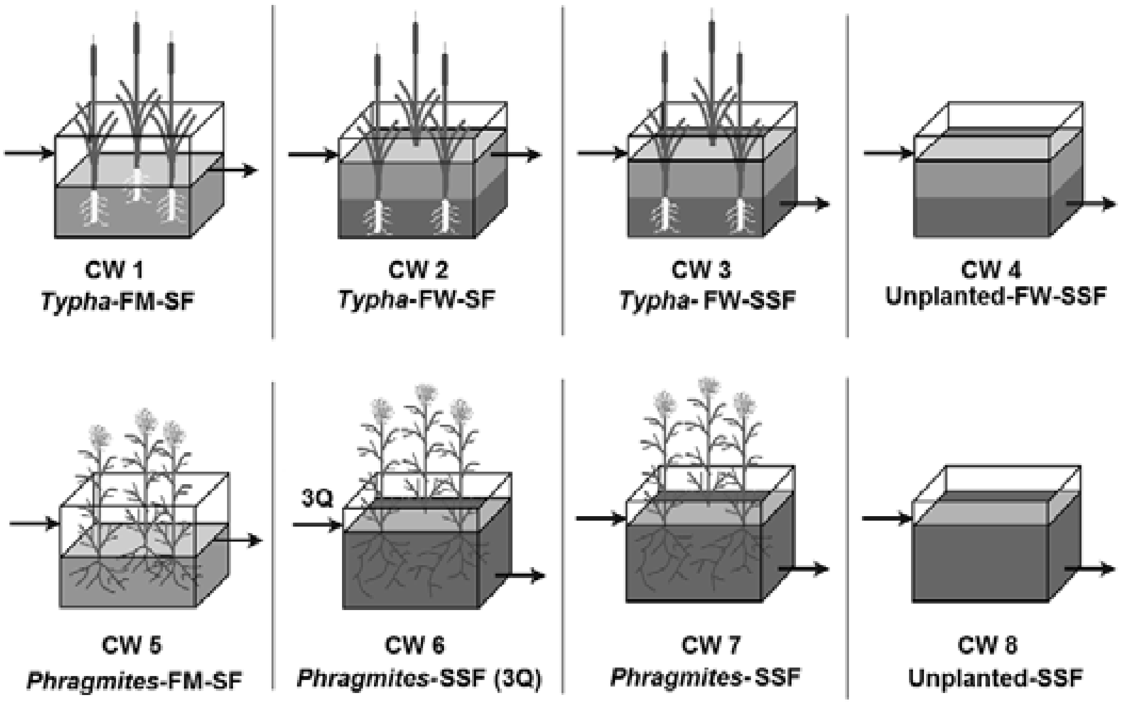
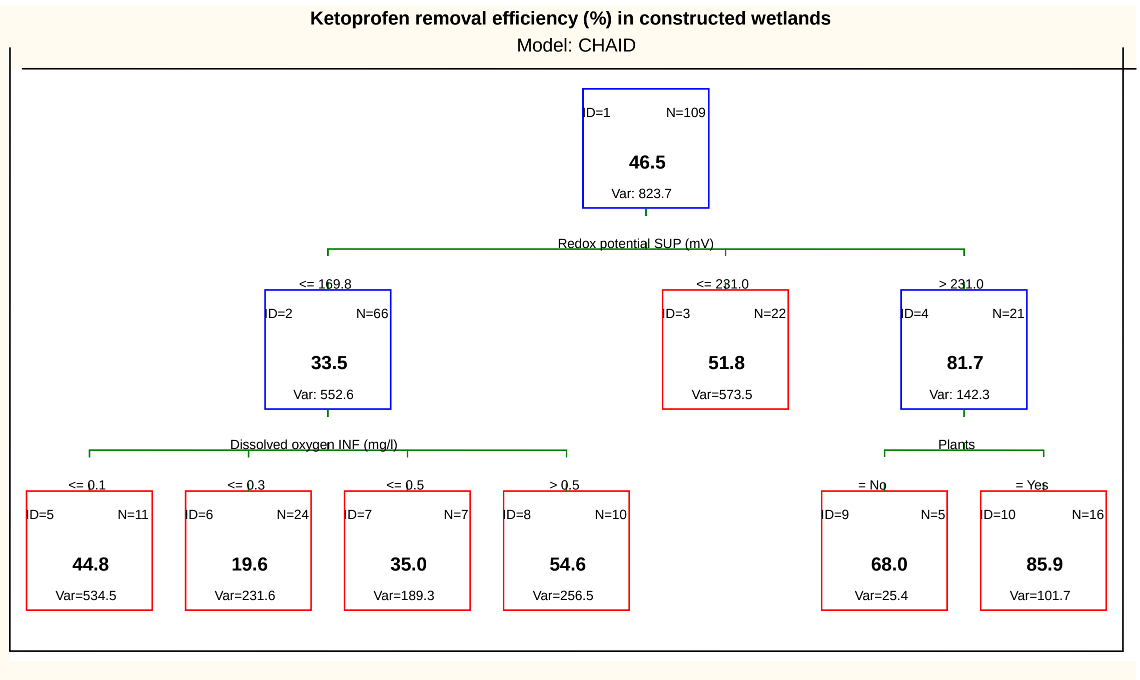


Figure 2. CHAID regression tree models for ketoprofen (Hijosa-Valsero et al., 2011)



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RÉSUMÉ

Les marais artificiels (CWS) ont été étudiés et utilisées depuis plusieurs décennies pour le traitement des eaux usées urbaines et industrielles. Peu de travaux sont centrés sur le thème de quel type de CWS est le plus approprié pour l'élimination des polluants organiques sous le même climat et les conditions de traitement des eaux usées.

Huit WCS ont été mis en place en plein air dans l'installation de traitement classique des eaux usées (WWTP) de León (cité dans le nord-ouest de l'Espagne).

Par le traitement de nos données nous avons souligné l'importance de la température, concentration en oxygène dissous, les valeurs du potentiel redox, la présence des plantes et la structure chimique pour l'élimination de la plupart des polluants.