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Emerging organic contaminant removal in a full-scale hybrid constructed wetland system for wastewater treatment and reuse

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ABSTRACT

A full-scale hybrid constructed wetland (CW) system based on three stages of different wetlands configurations showed to be a very robust ecotechnology for domestic wastewater treatment and reuse in small communities. It consisted of a 317-m^2 vertical subsurface flow (VF), a 229-m^2 horizontal subsurface flow (HF), and a 240-m^2 free water surface (FWS) CWs operating in series. VF and HF wetlands were planted with *Phragmites australis* and the FWS contained a mixture of plant species. An excellent overall treatment performance was exhibited on the elimination of conventional water quality parameters (98–99% average removal efficiency for TSS, BOD_5 and NH_4–N; $n = 8$), and its final effluent proved to comply with existing Spanish regulations for various reuse applications. The removal of studied emerging contaminants, which included various pharmaceuticals, personal care products and endocrine disruptors, was also very high (above 80% for all compounds), being compound dependent ($n = 8$). The high rates were achieved due to high temperatures as well as the differing existing physico-chemical conditions occurring at different CW configurations, which would allow for the combination and synergy of various abiotic/biotic removal mechanisms to occur (e.g. biodegradation, sorption, volatilization, hydrolysis, photodegradation). While aerobic metabolic pathways and solids retention are enhanced in the VF bed, other removal mechanisms such as anaerobic biodegradation and sorption would predominate in the HF bed. At last, photodegradation through direct sunlight exposure, and less importantly, sorption onto organic matter, seem to take an active part in organic contaminant removal in the FWS wetland.

1. Introduction

The occurrence of emerging organic contaminants (EOCs), such as pharmaceutical and personal care products (PPCPs), pesticides or antiseptics in poorly treated wastewater and eventually in other watercourses constitutes nowadays an increasing concern worldwide due to their possible toxicological effects to the environment and living organisms (Cunningham et al., 2006; Daughton, 2005; Kümmerer, 2009).

On the other hand, constructed wetlands (CWs) are wastewater treatment systems that emphasize the processes happening in natural wetlands so as to improve their treatment capacity (Kadlec and Wallace, 2009). They constitute a cost-effective alternative to conventional wastewater treatment plants (WWTPs), especially in the context of small communities with less than 2000 people equivalent (Puigagut et al., 2007). Various types of constructed wetlands have been combined in order to achieve higher treatment efficiency, especially for nitrogen removal. These hybrid systems are normally comprised of vertical subsurface flow (VF) and horizontal subsurface flow (HF) beds arranged in different possible manners, including recirculation from one stage to another. While in HF wetlands nitrification is not achieved due to their low oxygen content, in VF aerobic conditions prevail, which provide good conditions for nitrification, but little to negligible denitrification occurs in these systems. Thenceforward, the strengths and weaknesses of each type of system can balance each other out when combined, and in consequence it is possible to obtain an effluent low in total nitrogen concentrations, as well as other pollutants (Cooper, 1999; Masi and Martinuzzi, 2007; Vymazal, 2007).

Since EOCs are often poorly removed in conventional WWTPs (Heberer, 2002), advanced water reclamation technologies have been studied (e.g. advanced oxidation processes (AOPs) such as

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photo-Fenton, ozonitzation) (Klavarioti et al., 2009; Rosal et al., 2010). However, these AOPs often require a high level of energy and have high O&M costs, and thus are very unlikely to be implemented in the context of wastewater treatment of small communities. To this regard, several studies have shown a great capacity for EOC removal of constructed wetland systems at full-scale for domestic wastewater treatment of small communities in warm climates. These studies were conducted at systems consisting of a single wetland configuration at a time, namely VF, HF (Matamoros et al., 2009) or free water surface (FWS) (Llorens et al., 2009; Matamoros et al., 2008b). However, studies which evaluate the contribution to EOC removal of different wetland types within a hybrid system through potential synergies in treatment processes are very scarce, and include other types of treatment units (e.g. conventional WWTPs, waste stabilization ponds) as a treatment step prior to CWs (Hijosa-Valsero et al., 2010a; Matamoros and Salvadó, 2012). Evaluating the physicochemical properties and behavior of EOCs in different constructed wetland units belonging to hybrid systems remain, thus, a future challenge to be developed. This will help in refining CW design and operation modes, which in turn may increase CW acceptance and implementation as a cost-effective and operational alternative to conventional wastewater treatment technologies in decentralized areas (Imfeld et al., 2009).

In the context of a collaborative project between the Universitat Politècnica de Catalunya-BarcelonaTech (Barcelona) and the Foundation Center for New Water Technologies-CENTA (Seville), which aimed at the treatment of domestic wastewater up to quality standards appropriate for reuse through the sole use of CWs, an experimental meso-scale hybrid constructed wetland system was constructed. The system combined different CW configurations (VF, HF and FWS) and showed an excellent performance, both in terms of water quality parameters but also on EOC removal (Ávila et al., 2013a, 2014). Parallely, and in the context of the same collaboration, a comprehensive approach implemented at pilot full-scale with identical wetland configuration (VF, HF and FWS) in a Mediterranean climate area of south Spain (Seville) proved to be a highly efficient ecotechnology for an integrated sanitation of small communities in warm climates, holding very low O&M requirements (Ávila et al., 2013c). The treatment technology, which received combined sewer effluent, exhibited a great performance on solids, organic matter and total nitrogen removal, and showed to be very resilient to water flow fluctuations when evaluated during stormy periods and first-flush events. The final effluent of this proved to be of sufficient quality for its further reuse in various applications (i.e. silviculture and irrigation of forests and other green areas non accessible to the public, etc.), according to the quality criteria settling in the Royal Decree 1620/2007 establishing the legal regime for the reuse of treated water in Spain (maximum admitted values equal to 1000 CFU/100 mL of *Escherichia coli* and 35 mg L^{-1} of TSS).

However, the disposal into the aquatic environment of EOCs due to incomplete wastewater treatment has been of great concern for more than a decade (Cunningham et al., 2006). Additionally, in recent times there is a clear need to include irrigation as an additional exposure route for chemicals in terrestrial ecosystems. As an example, recent research is being done to explore whether these contaminants can be incorporated to crops irrigated with reclaimed water (Calderón-Preciado et al., 2013; Matamoros et al., 2012b). Although concentrations are low, questions have been raised about the potential impacts of these substances in the environment and animal and public health after long-term exposure (Matamoros et al., 2012b).

In this scenario, the aim of this study was to evaluate the treatment performance of a full-scale hybrid CW system located in a Mediterranean climate from southern Spain on the elimination of various EOCs from a combined sewer effluent. The selected compounds consisted of various commonly used pharmaceuticals and personal care products (PPCPs), as well as a high-production chemical widely used in epoxy resins lining food and beverage containers. These were: three non-steroidal anti-inflammatory drugs (ibuprofen – IB, diclofenac – DCF, acetaminophen – ACE), three personal care products (tonalide – AHTN, oxybenzone – OXY, triclosan – TCS) and two endocrine disrupting compounds (bisphenol A – BPA and ethinylestradiol – EE2).

2. Materials and methods

2.1. Plant description

The hybrid treatment system was part of a larger research, innovation and development center (R&D&i) Center for wastewater treatment and reuse (41,000-m^2) of the Foundation Center for New Water Technologies (CENTA) that received the wastewater from 2500 PE from the municipality of Carrión de los Céspedes (Seville) together with the urban runoff collected in a combined sewer system. Average annual rainfall in the area is around 650 mm and the average temperature is 17.4 °C. The center contains a great variety of both extensive and intensive technologies for wastewater treatment and reuse from small rural communities in the Mediterranean area (Fadh et al., 2007; Martín et al., 2009a), which are submitted to analysis and validation, and are used for knowledge dissemination and outreach (http://www.centa.es). Pretreatment chambers are common to all technologies and its effluent is diverted toward each of them. Pretreatment consists of screening (3 cm and 3 mm), and sand and grease removal. After pretreatment, the effluent is conveyed toward a pumping chamber, from which the water is distributed through submersible pumps to the different treatment technologies present in the center. The constructed wetland system started operation in 2005, though the treatment line as it is now began to operate in July 2009. In particular, a hybrid system consisting of a combination of various CW configurations was set in order to balance out the strengths and weaknesses of each type of system. The treatment line consisted of an Imhoff tank followed by a vertical subsurface flow CW (VF), a horizontal subsurface flow constructed wetland (HF) and a free-water surface CW (FWS) connected in series (Fig. 1).

The VF wetland had a surface area of 317 m^2 and was designed for an organic loading rate (OLR) of about 9 g BOD_5 m^{-2} d^{-1}. Note that this was rather a conservative design because of the lack of experience at full-scale in the area. It was fed intermittently at about 20 pulses d^{-1} to an average inflow of 14 m^3 d^{-1}. The bed consisted of a top layer of 0.05 m of sand (1–2 mm), followed by a 0.6 m layer of siliceous gravel (4–12 mm) and an underlying 0.15 m siliceous gravel (25–40 mm). Feeding of the VF was done through five lengthwise pipes (diameter = 125 mm) perforated with 1 cm diameter holes every 1.8 m distance. Five draining pipes were installed lengthwise at the bottom of the wetland within the 15 cm-thick gravel layer. Every draining pipe had three 1 m-tall chimneys so as to provide oxygen transfer into the wetland bed.

The HF unit had a surface area of 229 m^2 and consisted of a siliceous gravel bed of 0.4 m depth (4–12 mm), with an inlet and outlet zone of stones (40–80 mm) to facilitate the flow. Feeding of the bed was done through a 63 mm diameter polyethylene pipe perforated with 1 cm holes every 1 m distance. The outlet of the wetland was done by means of two 125 mm-diameter draining pipes located at the bottom of the stone layer and connected to a flexible pipe, which held the water level 5 cm below the top of the gravel. Theoretical hydraulic retention time (HRT) within this bed was of 2.3 d, unlike the VF bed one which is supposed to be of just a few hours due to the operation regime of this type of wetlands