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Removal of organic carbon, nitrogen, emerging contaminants and fluorescing organic matter in different constructed wetland configurations

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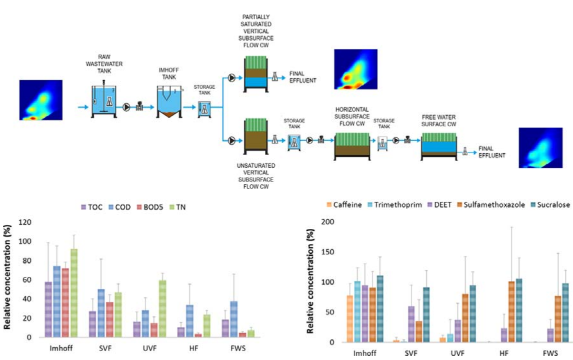
GRAPHICAL ABSTRACT



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ABSTRACT

The elimination of organic carbon, nitrogen, five emerging organic contaminants (EOCs) and fluorescence signature was evaluated in two treatment lines comprising different constructed wetland (CW) configurations: (i) partially saturated vertical subsurface flow (SVF) wetland (treatment line 1) and (ii) unsaturated vertical subsurface flow (UVF), horizontal subsurface flow (HF) and free water surface (FWS) wetlands in series (treatment line 2). Results showed important differences between the different CW configurations. The highest removal of BOD_5 (81%), COD (67%), TOC (72%) and fluorescing organic matter were observed in the UVF wetland, whereas the HF and FWS wetlands were the most efficient units for total nitrogen removal (60 and 69%, respectively). The SVF wetland showed a greater performance in the reduction of total nitrogen than the UVF bed (52 vs. 35%). In addition, the SVF wetland exhibited a higher removal of the EOCs caffeine (95 vs. 90%), trimethoprim (99 vs. 87%) and sulfamethoxazole (64 vs. 4%), as opposed to DEET (34 vs. 63%), whose removal was superior in the UVF unit. Sucralose was negligibly removed in all the CWs. PARAFAC analysis of fluorescence measurements revealed that the proteinaceous tryptophan-like fluorescent component was the most highly removed one in all the investigated CWs (> 28%) and, particularly, in the UVF wetland (66%), whereas humic and fulvic-like components resulted recalcitrant to decomposition. Increases of fluorescence intensities were often observed for fulvic-like substances in CWs operating with saturation of the bed, and these were particularly relevant in the SVF unit. Finally, important correlations ($r > 0.7$) between the tryptophan-like fluorescent

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component and the wastewater quality parameters COD and BOD_5 suggest fluorescence spectroscopy as an useful monitoring tool for water treatment efficiency in CW systems.

1. Introduction

Constructed wetlands (CWs) are engineered systems that emphasizes the physical-chemical and microbiological processes occurring in natural wetlands in a controlled manner to treat wastewater [1]. The use of this technology has developed rapidly over the last decades for the treatment of wastewater in decentralized areas both in industrialized and low-income countries due to their various advantages over conventional wastewater treatment systems. These include low to no energy consumption, ease of maintenance and operation, and good integration into the landscape and promotion of biodiversity, among others [2,3].

CWs have exhibited a great capacity to degrade contaminants from many different origins and also fecal microbial indicators, oftentimes complying with the requirements for potential reuse applications [4]. The removal of emerging organic contaminants (EOCs), including pharmaceuticals and personal care products (PPCPs), and other priority substances has also been lately explored, displaying a remarkable degradation capacity, mainly owed to the complex microbial interactions occurring within the bed media and the rhizosphere promoted by a large range of redox conditions [5–8]. However, more studies are still needed for a complete and thorough understanding of the behavior of these micropollutants in CW systems, which should also shed some light into the transformation pathways of these contaminants in treatment systems and the environment.

Despite CWs have been traditionally considered as 'black boxes', the research and technical development in the technology over the last decades has shown that the system's design and operation parameters influence the dominating environmental conditions inside the wetland, which, in turn affects degradation processes [9,10]. While dissolved oxygen concentration is one of the main limiting factors for biodegradation processes in traditional horizontal subsurface flow CW (HF) operating mostly under anoxic/anaerobic conditions due to the permanent saturation of the wetland bed, unsaturated vertical flow CW (VF) units were developed to increase the oxygen transfer capacity by specific design and operational conditions, such as intermittent feeding and resting periods [1,11]. Various design and operational alternatives have been proposed to improve the performance of CWs, comprising from the use of hybrid systems (VF and HF wetlands operated in series) to other strategies or intensifications that involve the use of induced energy through active aeration, increased pumping or recirculation [10]. Recently, the use of a saturated zone at the bottom of a classic VF wetland aims at creating anaerobic/anoxic conditions in the lower part of the bed, and aerobic conditions in the top part, so as to increase the microbial diversity in the wetland and promote various contaminant removal pathways. This strategy is especially targeted to enhance the removal of total nitrogen through the promotion of the nitrification-denitrification (NDN) processes [12]. Although presumably no studies have yet evaluated the removal of PPCPs in saturated VF wetlands, their transformation might also be benefited by the strategy and their behavior should be addressed.

On the other hand, the monitoring of wastewater treatment plants, including CWs, and its compliance with regulatory standards is generally assessed using physical, chemical and microbiological tests. Among these techniques, reliance is often placed on biochemical oxygen demand (BOD), chemical oxygen demand (COD) and total organic carbon (TOC) [13]. However, these parameters depend on time-consuming methods, offering only snapshots of moment in time, which makes them unsuitable for online monitoring [14]. An alternative promising approach is the use of fluorescence spectroscopy, which could be used for wastewater quality assessment as a tool for discharge detection in natural systems and for a continuous process control during wastewater treatments [15–17]. Fluorescence spectroscopy is a rapid, cost-effective, reagentless technique that requires little or no sample preparation prior to analysis. The acquisition of 3-dimensional excitation–emission matrices (EEMs) provides a 'map' of contributions of different component classes comprising dissolved organic matter (DOM). This has resulted in the development of fluorescence indexes that have been shown to be useful indicators of water treatment efficacy [15–18], or surrogate parameters useful for monitoring the fate of EOCs during conventional and advanced wastewater treatments [19–23]. However, to the best of our knowledge, there are no studies dwelling on the use of fluorescence measurements as indicator of treatment performance and EOC surrogate in CW systems.

In the current study, two CW treatment lines operating in parallel were evaluated for a period of 4 months. One of the lines consisted of a

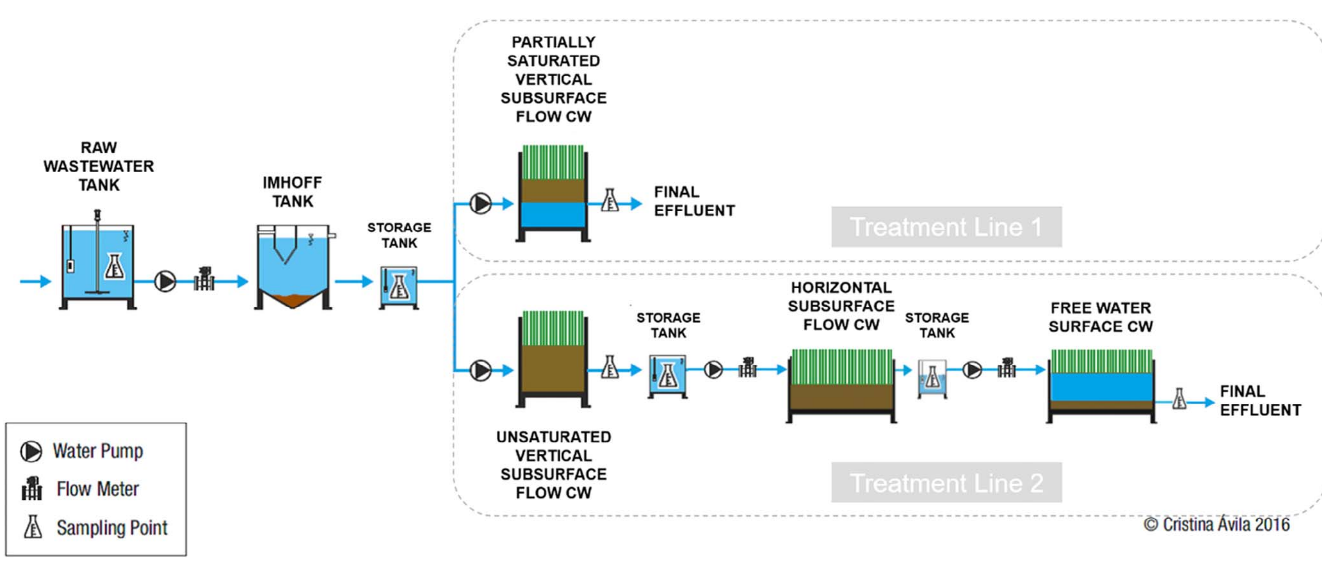

Fig. 1. Diagram of the experimental wastewater treatment plant.