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Effects of constructed wetland design on ibuprofen removal – A mesocosm scale study

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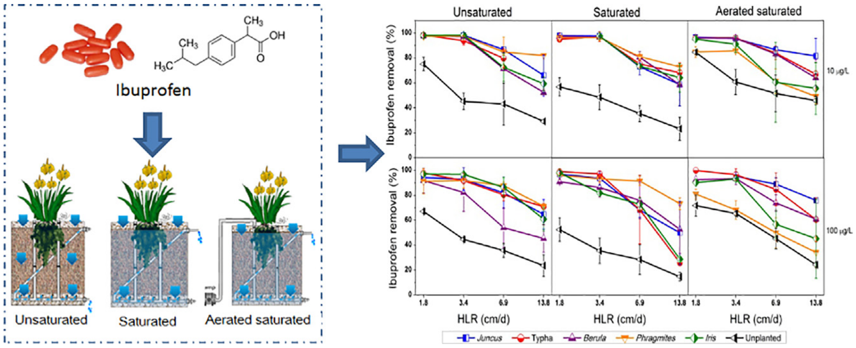
HIGHLIGHTS

- Ibuprofen was efficiently removed in three different CW designs.
- Presence of plants accelerated ibuprofen removal.
- Forced aeration enhanced ibuprofen removal in unplanted CWs.
- Ibuprofen removal followed an area-based first-order kinetics model.
- Biodegradation was the main removal pathway.

GRAPHICAL ABSTRACT



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ABSTRACT

This study aimed to investigate the effects of constructed wetland design (unsaturated, saturated and aerated saturated) and plant species (*Juncus*, *Typha*, *Berula*, *Phragmites* and *Iris*) on the mass removal and removal kinetics of the pharmaceutical ibuprofen. Planted systems had higher ibuprofen removal rates (29%–99%) than in the unplanted ones (15%–85%) in all designs. The use of forced aeration improved ibuprofen removal only in the unplanted mesocosms. In general, ibuprofen removal followed an area-based first-order removal kinetics model with removal rate coefficients (k_A) varying between 3 and 35 cm/d. The ibuprofen removal was mainly attributed to microbial degradation by the fixed bed biofilm, but plant uptake and degradation within plant tissues also occurred. The ibuprofen removal was positively correlated with the oxygen concentration in the water and the removal of nutrients, indicating that degradation may be due to co-metabolisation processes.

1. Introduction

Trace concentrations of pharmaceuticals and personal care products (PPCPs) have been detected in many types of fresh and reclaimed waters, such as seawater, drinking water, surface water, and wastewater effluents (Matamoros et al., 2007). The widespread occurrence of these compounds in natural waters may have toxic effects on aquatic organisms and potentially also long-term threats on human health (Dussault et al., 2008; Schnell et al., 2009; Han et al., 2010).

PPCPs enter into the environment mainly from the effluents from wastewater treatment plants (WWTPs) (Joss et al., 2005). Conventional

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WWTPs are only designed to remove typical pollutants, such as organic matter, nitrogen and phosphorus compounds, and PPCPs are reported to be removed with low efficiencies (Carballa et al., 2004; Sim et al., 2010; Repice et al., 2013). Although many advanced treatment technologies such as ozonation, membrane biofilm reactors, advanced chemical oxidation and UV radiation have recently been assessed for PPCPs removal (Clara et al., 2005; Esplugas et al., 2007; Bolong et al., 2009; Li et al., 2014), the high cost for operation and maintenance of these advanced technologies is the critical bottleneck for their wide application.

Constructed wetlands (CWs) have proven to be effective in removing several PPCPs from wastewaters (Matamoros et al., 2007; Breitholtz et al., 2012; Carvalho et al., 2013). To date, most published work, however, focuses on the removal efficiencies of PPCPs (Matamoros et al., 2007; Hijosa-Valsero et al., 2010; Zhang et al., 2012a; Verlicchi and Zambello, 2014) rather than removal mechanisms and kinetics. The few studies that have included removal kinetics are typically focused in a single type of CWs. The area-based first-order kinetic removal rate constants of several PPCPs have been estimated in pilot-scale horizontal subsurface flow CWs (Matamoros and Bayona, 2006) and pilot vertical flow CWs (Matamoros et al., 2007). Furthermore, Zhang et al. (2012a) estimated the area-based first-order kinetic removal rate constants of some PPCPs in horizontal subsurface flow CWs. Also Zhang et al. (2017) estimated the removal kinetics of two pharmaceuticals (ibuprofen and iohexol) in water saturated CW mesocosms. However, the factors influencing the removal kinetics remain unclear, and the effects of wetland design on the removal kinetics of PPCPs are missing.

Horizontal subsurface flow and vertical flow CWs are mainly used for the secondary and tertiary treatment of domestic and municipal wastewater in Europe (Brix, 1994; Vymazal, 2010). The treatment performance of these CWs can however be intensified by using forced aeration (Ouellet-Plamondon et al., 2006; Nivala et al., 2013). This study utilized three CW designs, (i) an unsaturated system simulating vertical flow CWs, (ii) a water saturated system simulating horizontal subsurface flow CWs, and (iii) a water saturated CW with forced aeration simulating CWs with forced aeration. To our knowledge, the direct comparison of the three CW designs in a single experiment has never been conducted before.

Ibuprofen, a non-steroidal anti-inflammatory drug, was chosen as a model target PPCP (physico-chemical properties shown in Table S1). Ibuprofen is of high consumption worldwide, it has potential ecological impacts due to its frequent detection in freshwater and might also have potential long-term adverse effects on aquatic life (Han et al., 2010; Parolini et al., 2011). Moreover, ibuprofen has been proposed as a marker of wastewater contamination (de Sousa et al., 2014). Ibuprofen is one of the most studied organic micro-pollutants, which allows better comparison with results from other studies.

The objectives of this study were to: (1) compare the ibuprofen removal efficiency and removal kinetics among the three CW designs: water unsaturated (simulating vertical flow), water saturated (simulating horizontal flow) and aerated water saturated (simulating systems with forced aeration); (2) identify the main ibuprofen removal processes in the different CW designs; and (3) investigate possible correlations between ibuprofen removal and various environmental and operational variables.

2. Materials and methods

2.1. CW mesocosm set-up and operation

A total of 54 mesocosms were used and operated under conditions simulating three different CW designs: water unsaturated, water saturated and aerated water saturated (Fig. 1). For each CW design, 18 mesocosms were used, and were divided into six mesocosm types (three replicates for each type) according to the presence or absence of plants and plant species. Five species of wetland plants, *Juncus effusus* (*Juncus*), *Typha latifolia* (*Typha*), *Berula erecta* (*Berula*), *Phragmites australis* (*Phragmites*) and *Iris pseudacorus* (*Iris*) were used, and one set of mesocosms were left unplanted as a control. The experimental systems had been continuously operated for approximately 1.5 years prior to the study (Zhang et al., 2017), and with slight operation modifications were further used in the present study.

Each mesocosm consisted of a black plastic container (20 cm diameter by 20 cm height) with a surface area of 0.03 m^2. The substrate in the mesocosms was layered; 4 cm of coarse gravel (ø 8–12 mm) in the bottom, a geotextile, 10 cm washed quartz sand (ø 0.5–1 mm, porosity 37%) and a 4 cm layer of coarse gravel (ø 8–12 mm) on top to avoid light exposure of the sand. The pore volume of each mesocosm was 1.25 L.

The influent water was loaded onto the surface of the substrate layers and trickled through the substrate to a collection system in the bottom gravel layer. Effluent was evacuated from a bottom outlet in the unsaturated group or from an upper outlet placed just below the surface of the substrate in the saturated and aerated saturated groups (Fig. 1). The influent was fed through a Ø 16 mm PE pipe fitted with pressure compensated drippers (0.5 L/h) connected to a timer-controlled pump. In the aerated saturated group, atmospheric air (2.2 L/min) was continuously injected into the systems from the bottom of the mesocosms by an air pump (SLL-40, SECOH Shanghai Mec Ltd.).

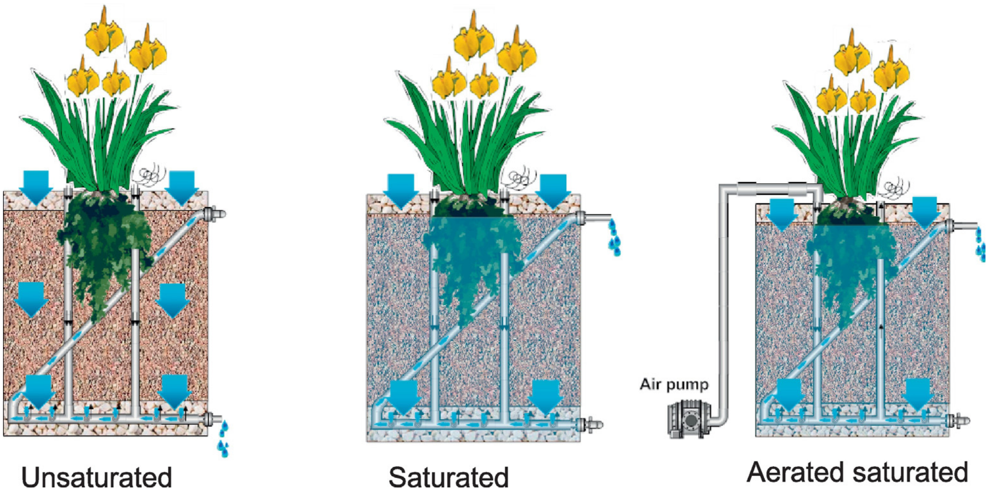

Fig. 1. Simplified diagram of the unsaturated, saturated and aerated saturated mesocosms (modified from Lv et al., 2016).