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Removal of pharmaceuticals by a pilot aerated sub-surface flow constructed wetland treating municipal and hospital wastewater

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ABSTRACT

Constructed wetlands are often used as decentralized wastewater treatment and as treatment systems for small communities. The effluent is usually discharged in small water courses in which the aquatic ecosystem can be put at risk by pharmaceutical residues present in the effluent. This study describes the performance of a pilot-scale aerated subsurface-flow constructed wetland treating municipal and hospital wastewater. Especially the effect of active aeration on the removal of selected pharmaceuticals is assessed. The removal of metformin and valsartan is significantly increased when continuous aeration is applied (99 ± 1% vs. 68 ± 32% for metformin and 99 ± 1% vs. 17 ± 19% for valsartan), although the micro-organisms can adapt to degrading metformin also in anoxic conditions. At the hospital site high concentrations of pharmaceuticals are measured in the influent. Atenolol and bisoprolol are efficiently removed (>75% and >50%, respectively), although the effluent concentrations of these compounds remain high (up to 0.5 μg/L). Only limited removal of carbamazepine, diclofenac, gabapentin and sulfamethoxazole is achieved. Intermittent aeration (50%) provides equally efficient removal of the selected pharmaceuticals as continuous aeration.

1. Introduction

Several studies have shown that effluent from urban wastewater treatment is an important source of organic micropollutants such as pharmaceutical residues in surface waters (Michael et al., 2013). In Flanders, some rivers were investigated for the occurrence of selected pharmaceuticals by VMM (the Flemish Environment Agency) in the context of a European project FATE EUMORE. A third (34%) of these samples contained a higher concentration of pharmaceuticals than the warning level (mostly 100 ng/L) (European Commission, 2015; Loos et al., 2009). Recently, also (Vergeynst et al., 2015, 2014) detected quite a number of pharmaceuticals in Flemish surface waters and wastewater effluents.

Pharmaceuticals are partly metabolized but not completely degraded by the human body and therefore, the unaltered parent compounds and their metabolites are excreted (Ternes et al., 2004; Trautwein and Kümmerer, 2011). At the wastewater treatment plant these compounds can undergo biological degradation in which their molecular structure can be further transformed but they are rarely completely mineralized (e.g. Constructed wetlands (CWs)), which are mainly used as decentralized wastewater treatment and as treatment systems for small communities, can contribute efficiently to decreasing pharmaceutical load to the environment. The ecosystem of the small water courses where CWs discharge (Rousseau et al., 2004) can easily be compromised if dilution in the receiving water is low. Therefore, it is important to study the performance and improve the CW technology towards more efficient pharmaceutical removal.

Extensive investigations on the effects of different types of wetland configurations, plants and treatment strategies on the behavior of different pharmaceuticals have already been

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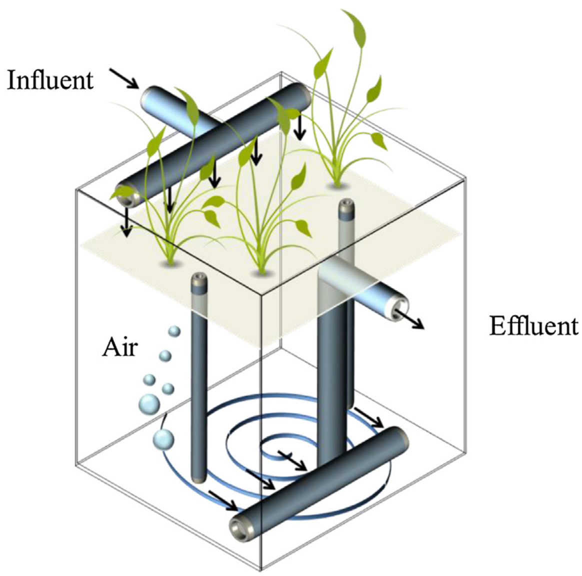

Fig. 1. A schematic representation of the pilot CW.

performed, for example, in Spain (Ávila et al., 2015, 2014a, 2013, 2010; Matamoros et al., 2012; Matamoros and Bayona, 2006; Reyes-Contreras et al., 2012; Hijosa-Valsero et al., 2016, 2011a, 2010) and Germany (Ávila et al., 2014b; Carranza-Diaz et al., 2014). However, further research on different types of CWs is needed to gain insight into the factors affecting the degradation of pharmaceuticals.

The aim of this study was to assess the performance of an aerated pilot-scale sub-surface flow (SSF) CW treating municipal and hospital wastewater. In the first part of the experiment, the pilot CW was fed with settled municipal wastewater of a small community. The effects of active aeration on its pharmaceutical removal efficiency as well as the robustness of its performance during increased hydraulic loading were studied. In the second part of the experiment, the pilot CW was relocated to a hospital site and its treatment performance was assessed during continuous and intermittent aeration. Hospital wastewater has typically a higher concentration of pharmaceutical residues than municipal wastewater. Separate treatment of this water would be advisable for several reasons: firstly, to reduce the load of pharmaceuticals on the wastewater treatment plant (WWTP), secondly, to avoid dilution of the wastewater and thereby, decreased removal efficiency during treatment and finally, to prevent discharge of pharmaceutical residues to the environment due to combined sewer overflows and sewer leakages.

2. Materials and methods

2.1. Experimental setup and location

A transportable pilot-scale subsurface flow constructed wetland (Fig. 1) was built using a plastic container with an approximate total volume of 1 m^3. The cubic tank was filled with an 80 cm layer of coarse Rhine gravel (8–16 mm, approximate porosity 40%; Kranendonk N.V.) and evenly planted with mature *Phragmites australis* plants obtained from laboratory-scale CWs treating WWTP effluent. The plants were allowed to re-root and biofilm to develop for 8 weeks before the sampling was started. During this adaptation period the wetland was fed with wastewater (hydraulic retention time (HRT) 1 d) and aerated continuously.

Influent was evenly distributed on the CW surface over the horizontal length of the tank on one side. On the opposite side, effluent was collected at the bottom of the tank in a perforated inversed T-shaped pipe connected to an elbow which discharged the effluent at a level approximately 5 cm below the gravel surface. As a consequence, the water flow direction was mainly vertical, although the applied aeration was expected to cause mixing. Two perforated PVC tubes were positioned in the middle parallel to each other to allow the measurement of on-site parameters (temperature, pH, dissolved oxygen) inside the wetland. Aeration of the wetland was provided with a fish pond air pump (Secoh, JDK-20; design capacity 23 L/min) connected to a perforated aeration tube that was placed on the bottom of the tank. At setting the flow rate and defining the HRT, a nominal organic loading rate of 80 g O_2/d/m^2 (BOD_5) was used. The value was chosen based on earlier experiments on actively aerated CWs (D. Van Oirschot, personal communication, 12.12.2014 and 5.8.2016). An online database was consulted for the BOD_5 data that has been measured earlier regularly at the site (VMM, 2016).

In March 2015, the pilot CW was placed at the wastewater treatment plant of Aalbeke (Kortrijk, Belgium) which treats combined sewer municipal wastewater from a small community (design capacity 450 IE). This treatment plant is the property of Aquafin N.V. (www.aquafin.be), the company responsible for municipal wastewater treatment in Flanders. Wastewater for the pilot CW was taken batch-wise (10 times per day) from the existing primary settler by using an immersion pump (POW67915, PowerPlus). The operational parameters of the pilot CW can be found in Table 1. After the measurement campaigns, the pilot CW was kept operational (regular feeding and continuous aeration) until the transfer to the hospital site.

In October 2015, the setup was transported to the Campus Kennedylaan of the hospital AZ Groeninge (Kortrijk, Belgium). Raw wastewater was taken directly from the hospital sewer (approximately 300 L/d) by means of a macerator pump (DRK 10-1K, Duijvelaar Pompen) and delivered to a 1 m^3 settling tank. The settling time was 7 h and the accumulated sludge was removed every 2–3 days by manually opening a bottom valve. The settled sewage was pumped to the pilot CW batch-wise (5 times per day) by using a peristaltic pump (530S, Watson-Marlow). The intake of the pump was attached to a piece of Styrofoam to assure that only the supernatant was pumped into the experimental wetland. The operational parameters of the pilot CW can be found in Table 1.

In both locations, the aeration was controlled by a timer. In Aalbeke, three measurement campaigns were conducted: In the first (AER-HRT1) and second campaign (AER-HRT0.5), the aeration was continuously on. The second campaign differed from the first one by a lower HRT (0.5 d and 1 d, respectively). The third campaign (N-AER), with no aeration, was started after an adaptation period of one week and an HRT of 1 d was utilized. It lasted for two weeks: N-AER1 indicating the first week of the campaign and N-AER2 the second. In AZ Groeninge, there were two measurement campaigns: In the first one (AER100), the aeration was continuously on, and in the second one (AER50), aeration time was reduced to 50%. The sampling for AER50 was begun after 2 weeks of adaptation.

2.2. Sampling

In Aalbeke, influent and effluent samples were taken simultaneously, but in AZ Groeninge, the effluent sample was taken 2 days (duration of the nominal HRT) after the influent sample, to be able to take the changing composition of the influent into account. Approximately 80 mL of influent and effluent samples were collected during each feeding/discharge event to obtain a mixed sample from that day by using a peristaltic pump (Sci-Q323, Watson Marlow). The sampling was automated to occur from an overflow beaker at programmed time points. In Aalbeke, the samples were taken every 2.5 h during 24 h (total of 10 feeding/discharge events per day). In AZ Groeninge, there were 6 feeding/discharge events, the