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Application of a full-scale newly developed stacked constructed wetland and an assembled bio-filter for reducing phenolic endocrine disrupting chemicals from secondary effluent

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ABSTRACT

To mitigate the detrimental risk of residual phenolic endocrine disrupting chemicals (PEDCs) from the discharge of sewage treatment plants (STPs) and to increase the utilization of constructed wetlands (CWs), both a stacked CW (SCW), comprising vertical and horizontal flow CWs, and an assembled bio-filter (ABF) were developed and their capability to remove PEDCs from STP discharge at high hydraulic loading rates (HLRs) of 0.5–2.0 m/d evaluated. The SCW and ABF showed different performance on PEDCs removal because of process structural difference. The removal percentages of PEDCs were associated with their octanol-water partition coefficients (Log*Kow*), except 4-nonlyphenol (4-NP) whose apparent removal rate was affected by its precursors, and declined in the order of triclosan (TCS) at 80% and 69%, 4-*t*-octylphenol (4-*t*-OP) at 58% and 51%, estrone (E1) at 48% and 44%, and bisphenol-A (BPA) at 45% and 32% in the SCW and ABF systems on average, respectively. The efficiencies of SCW and ABF were superior to the single type of CWs, such as surface flow CW, vertical flow CW, and horizontal flow CW, in terms of removal rates or first-order removal rate constants. Recommended value of HLR for optimal utilization of SCW and ABF to ensure essential removal of PEDCs from STP discharge was between 1.5 and 2.0 m/d.

1. Introduction

Certain progress has been achieved by traditional sewage treatment plants (STPs) regarding the elimination of endocrine disrupting chemicals (EDCs). However, residual EDCs that have escaped the STPs remain in concentrations above environmental disturbance thresholds and therefore, they pose a potential risk to aquatic organisms (Sun et al., 2013). The estrogenic activity of secondary effluent is mainly derived from natural estrogens, contraceptives, and some industrial phenolic compounds such as bisphenol-A (BPA) and the degradation of intermediate products of alkylphenol ethoxylates (APEs) (Johnson and Sumpter, 2001; Sun et al., 2013). Triclosan (TCS) is widely used as a microbicide in personal care products, but it also has potential androgenic effects on fish (Foran et al., 2000). The above compounds are considered as pseudo-persistent pollutants because of their continuous entry into the environment through sewage/domestic wastewater, and they are called phenolic EDCs (PEDCs) because of their phenolic group (Johnson and Sumpter, 2001). It is therefore essential to improve their removal efficiency and to reduce the risk from PEDCs in secondary effluent.

Constructed wetlands (CWs) have been applied successfully to the removal of nutrients, chemical oxygen demand (COD), and extensive trace organic compounds in recent years (Barceló and Petrovic, 2008). Basic wetland types include surface flow CWs (SFCWs), vertical flow CWs (VFCWs), and horizontal flow CWs (HFCWs). It has been reported that the estrogens in secondary effluent could be removed rapidly by small-scale VFCWs and HFCWs (Song et al., 2009, 2011). Furthermore, it has been reported that full-scale SFCWs could reduce E2 and EE2 in river water (Gray and Sedlak, 2005), alkylphenols (APs) and BPA in river water (Gross et al., 2004; Yang et al., 2011), and TCS in secondary effluent (Park et al., 2009). However, treatment efficiencies vary considerably depending on variables such as system type and design, temperature, hydraulic loading rates (HLRs), nutrient mass-loading rates,

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vegetation, and microbial communities, of which the first three are the most important (Kuschk et al., 2003).

The environmental conditions provided by different types of CW for wastewater treatment vary; some can promote a specific redox condition, while others have a wide range of redox conditions within the wetland matrix, leading to the formation of microbial biofilms with functionally different respiration processes and pollution removal capacities (Barceló and Petrovic, 2008). The VFCW is generally considered a highly aerobic system, because wastewater drains vertically through the planted matrix, allowing for unsaturated conditions and excellent oxygen transfer that favors aerobic microbial processes (Barceló and Petrovic, 2008). The HFCW has both oxidized and reduced zones but overall, it is generally considered an anoxic system that favors anoxic microbial processes such as traditional denitrification (Barceló and Petrovic, 2008). Previous studies have indicated that a series combination of a VFCW and an HFCW obtains better performance than single type CWs, with regard to the comprehensive purification of COD and nutrients, because of the complementary properties of the various CWs (Melián et al., 2010). However, one of the major constraints of a full-scale CW is the requirement for a relatively large land area, which is often not readily available. In this context, a reformatory serial mode, in which a VFCW is stacked on an HFCW, might lead to a requirement for a smaller land area.

Another means by which to reduce the land requirement is to increase the HLR, but this will usually lead to lower nutrient removal efficiency. Therefore, the appropriate value of the HLR should depend on the influent properties. It has been noted that CWs can run at HLRs as high as 4.8–15.6 m/d in treating tertiary effluent for water reuse purposes (Ayaz, 2008). However, rather small HLRs of 0.009–0.5 m/d have been adopted in previous studies for the treatment of river water or secondary effluent for PEDC purification (Gross et al., 2004; Park et al., 2009; Yang et al., 2011; Matamoros et al., 2012; Chen et al., 2014). Hence, there is potential to increase the utilization efficiency of CWs in removing traditional pollutants and PEDCs from secondary effluent by increasing the HLRs within a certain range.

Different from CWs, the bio-filter (BF) technique has been derived from drinking-water production filters and it combines physical and biological purification processes. The benefits of BFs lie in their compactness (small site encumbrance), modularity (treatment procedure can be adapted to match the wastewater flow able to be accommodated by the plant), and intensiveness (short hydraulic retention time, HRT). Therefore, the technique is highly recommended and increasingly popular for wastewater treatment (Gasperi et al., 2010). However, currently available data on the efficiency of the removal of PEDCs through the application of a BF are limited.

In this study, for the first time, we obtained data on the elimination of PEDCs from STP discharge by an SCW at HLRs of 0.5–2.0 m/d. Furthermore, an assembled BF (ABF) was chosen as comparable technology because of its virtue of a small footprint. Here, the treatment kinetics of both the SCW and the ABF systems are calculated and the correlation between the PEDCs' removal kinetics and Log*Kow* discussed.

2. Materials and methods

2.1. Experimental setup and sampling strategy

The Huizhou Fourth STP that serves Huizhou in South China, discharges secondary effluent at a flow rate of 40,000 m^3/d into the East River, which is an important potable water source for Guangdong Province, Hong Kong, and Macao. The treatment processes used in this STP include screening, primary sedimentation and conventional activated sludge treatment. A full-scale SCW and an ABF were built as tertiary treatment systems, which commenced treatment of part of the STP discharge in September 2010. The overall layouts and cross sections of the SCW and ABF are shown in Fig. 1. The SCW comprised a 96-m^2 VFCW (24 × 4 × 0.7 m; length × width × depth) operating under unsaturated conditions, stacked above a 204-m^2 HFCW (24 × 8.5 × 0.8 m; length × width × depth) operating under saturated conditions, i.e., the SCW only occupied an area of 204 m^2. The influent flowed sequentially through the VFCW and HFCW. The areas for the distribution and collection of water in the wetland bed were filled with coarse gravel (3–5-cm diameter) to avoid clogging, but 1–3-cm-diameter gravel was used elsewhere. In order to ensure uniformity of the water distribution and flow in the SCW, both the VFCW and HFCW were each divided into four equal units. *Canna glauca, Thalia dealbata, Canna indica*, and *Typha angustifolia* were planted in the VFCW units, and *Cyperus alternifolius*, *Arundo donax*, *Acorus tatarinowii*, and *Desmodium styracifolium* were planted in the HFCW units. One single species was planted in each subunit having a different species in each subunit. The ABF (3 × 3 × 2.8 m; length × width × height), operating under unsaturated conditions, was divided into four layers of equal thickness (0.4 m per layer) and the air space between adjacent layers was 0.2 m. The upper two layers were filled with blast furnace slag, which has good capability in phosphorus removal (Wu et al., 2010), and the subsequent two layers were filled with gravel, which is cheap and plentiful. This design provides the optimum oxygen enrichment (Zhu et al., 2013). The ABF was kept in a shaded building with louver. The experiment was performed during May to June 2011 when the coverage of the well-developed vegetation was nearly 100%. The STP discharge was stored in a short-residence-time reservoir before being pumped into the SCW and ABF systems (Fig. 1). In the tests, flowmeters were used to produce four different HLRs: 0.5, 1.0, 1.5, and 2.0 m/d, for which the theoretical HRTs of the SCW were about 24, 12, 8, and 6 h, respectively. Under the same HLRs, the HRTs of the ABF, calculated according to an empirical formula from an ABF developed by Zhu et al. (2013), were about 60, 42, 34, and 29 min, respectively. Both the facilities worked 12 h every day.

Under each HLR, the system was operated for 10 days and then inlet and outlet samples were collected daily during the subsequent three days when the performances of the systems were considered stabilized under the particular loading rate (Matamoros et al., 2007). Each daily sample comprised a mixture of collections at 10:00 a.m. and 4:00 p.m. (n = 3 for each HLR), including two parallel samples, which were all collected in 1-L glass amber bottles for PEDC analysis. In the field, a volume of about 50 mL of methanol was added to the samples and the pH was adjusted to pH = 3 using 0.4 mL of 4 M H_2SO_4. Then, the samples were kept refrigerated until analysis (within two days). Conventional wastewater quality parameters of total suspended solids (TSS), COD, and ammonium (NH_4^+) were analyzed simultaneously (State Bureau of Environmental Protection, 2002). The pH was measured by a conventional pH electrode (Testr 20, EUTECH Corporation, Singapore), and dissolved oxygen (DO) and temperature were measured using an OxyGard DO meter (YSI ProPlus, YSI Corporation, USA).

2.2. Chemicals, standards, and materials

The reference standards for BPA, E2, EE2, E1, 4-*tert*-octylphenol (4-t-OP), 4-nonylphenol (4-NP), and TCS were obtained from Dr. Ehrenstorfer (Dr. E) (Germany). Internal standards for BPA-D16, E2-D4, E1-D4, 4-*n*-nonylphenol (4-*n*-NP), and $^{13}C_{12}$-TCS were obtained from C/D/N Isotopes Inc. (Canada), Cambridge Isotope Laboratories Inc. (CIL) (USA), Toronto Research Chemicals Inc. (Canada), Dr. E (Germany), and CIL (USA), respectively. Stock solutions of the stan-