



# Treatment of a sulfate-rich groundwater contaminated with perchloroethene in a hydroponic plant root mat filter and a horizontal subsurface flow constructed wetland at pilot-scale



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## HIGHLIGHTS

- Subsurface flow constructed wetland (SSF CW) removed perchloroethene (PCE) robustly.
- Hydroponic plant root mat filter (HPRMF) removed PCE less efficient than the SSF CW.
- The carcinogenic metabolite vinyl chloride was not detected in HPRMF but in SSF CW.
- Accumulation of PCE metabolites in SSF CW was somewhat reduced by tidal operation.
- Redox potential and dissolved oxygen concentration were higher in the HPRMF.

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## ABSTRACT

A hydroponic plant root mat filter (HPRMF) was compared over 7 months with a horizontal subsurface flow constructed wetland (HSSF CW) regarding the removal of perchloroethene (PCE) (about  $2 \text{ mg L}^{-1}$ ) from a sulfate- ( $850 \text{ mg L}^{-1}$ ) and ammonia-rich ( $50 \text{ mg L}^{-1}$ ) groundwater with a low TOC content. At a mean area specific inflow PCE load of  $56 \text{ mg m}^{-2} \text{ d}^{-1}$ , after 4 m from inlet, the mean PCE removal during summer time reached 97% in the HPRMF and almost 100% in the HSSF CW. Within the first 2 m in the HSSF CW metabolites like dichloroethenes, vinyl chloride and ethene accumulated, their concentrations decreased further along the flow path. Moreover, the tidal operation (a 7-d cycle) in the HSSF CW decreased the accumulation of PCE metabolites within the first 1 m of the bed. The carcinogenic degradation metabolite vinyl chloride was not detected in the HPRMF. The smaller accumulation of the degradation metabolites in the HPRMF correlated with its higher redox potential. It can be concluded from this study that HPRMF appears an interesting alternative for special water treatment tasks and that tidal operation will show some positive effects on the removal of the accumulated PCE metabolites in HSSF CW.

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## 1. Introduction

Perchloroethene (PCE) is one of the most widely used chlorinated solvent. The annual production of PCE in the EU and the USA reached 164,000 (1994) and 160,000 t (1998). PCE is mainly used for the dry cleaning of textiles and metal degreasing. Due to

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improper use and storage, PCE has been recognized as being among the most ubiquitous groundwater contaminants (Watts, 2006). PCE and its microbial metabolites trichloroethene (TCE), dichloroethenes (DCEs) and vinyl chloride (VC) are known to be toxic to humans. VC is even known as a human carcinogen, and is a USEPA priority pollutant with a maximum contaminant level (MCL) of  $2 \text{ } \mu\text{g L}^{-1}$  in drinking water (USEPA, 2002). The MCL value in drinking water for both PCE and TCE recommended by the USEPA is  $5 \text{ } \mu\text{g L}^{-1}$  (USEPA, 2002). PCE and its metabolites are often accumulated in the groundwater if anoxic conditions prevail. In general,

PCE can be metabolized by indigenous microorganisms through different degradation mechanisms. Anaerobic conditions are necessary for the first step of reductive dechlorination of PCE to TCE, which can be followed by a reductive dechlorination or oxidation of TCE, dichloroethenes (1,1-DCE, 1,2-*trans*-DCE, 1,2-*cis*-DCE), VC and ethene under anaerobic and/or aerobic conditions (Bradley, 2003; Mattes et al., 2010; Kalimuthu et al., 2011).

Constructed wetlands (CWs) are increasingly used for the treatment of various types of wastewater (Vymazal, 2009) due to their low commercial energy requirements, easy operation and maintenance compared to conventional wastewater treatment systems (García et al., 2010). However, the application of CWs for treating water contaminated with chlorinated hydrocarbons is scarce to date. The removal of chlorinated ethenes has been investigated in microcosms (Bankston et al., 2002; Kassenga et al., 2003), phytoremediation applications (poplar trees) (Ma and Burken, 2003; James et al., 2009), pilot and full scale CWs (Pardue et al., 1999; Amon et al., 2007; Braeckevelt et al., 2011; Kadlec et al., 2012). In general, the potential pathways for the removal of chlorinated ethenes in wetlands are microbial degradation (reductive dechlorination, oxidation and co-metabolism), volatilization, plant uptake, phytovolatilization, phytodegradation and sorption (Pardue et al., 1999; Kadlec and Wallace, 2009). Nevertheless, microorganisms play an important role in CWs for the removal of organic pollutants, and plants have been shown to improve the microbial activity (Faulwetter et al., 2009; García et al., 2010). Moreover, due to the presence of plants in CWs, releasing oxygen and carbon from their roots, both oxic and anoxic conditions coexist, which support the growth and activity of both aerobic and anaerobic microorganisms (García et al., 2010).

Due to the low oxygen transfer rate, which restricts the performance of horizontal subsurface-flow (HSSF) CWs, the discontinuous outflow flushing mode, the so called “tidal flow”, has attracted attention since the late 1990's (Green et al., 1997; Sun et al., 1999; Vymazal and Masa, 2003; Cui et al., 2012; Hu et al., 2014). During the fast outflow flushing period, the wastewater acts as a passive pump to draw air into the soil pores. Thus, the tidal flow operation has the potential of improving the treatment efficiency through an enhanced aerobic microbial decomposition and pollutant biofilm contact (Sun et al., 2006). The positive effect of the pulsing water level on the removal of organics was also detected in HSSF CWs (Vymazal and Masa, 2003; Chung et al., 2012; Cui et al., 2012).

Floating plant root mats are a variant of vegetated ponds, in which the plants are no longer rooted in soil but grow on floating rafts, or use the self-buoyancy of their dense interwoven roots and rhizomes which form a mat (Van de Moortel et al., 2010; Headley and Tanner, 2011; Tanner and Headley, 2011; Winston et al., 2013). When the water level is lowered to such an extent that the mat touches the root proof bottom of the pond or channel, and the hydraulic flow is forced to flow directly through the root mat, this system can be considered to function as a hydroponic plant root mat filter (HPRMF). Schematics of HPRMF and HSSF CW are shown in Fig. 1. Due to the lack of soil medium in the HPRMF, it can reduce the construction cost to some extent compared with the soil based CWs. However, both the floating plant root mats and the non-floating HPRMF are still not yet broadly applied technologies. Floating plant root mats have until now only been tested for cleaning selected polluted water, including domestic wastewater, combined sewer overflow water, agricultural effluents, river and lake water, storm water, acid mine drainage and contaminated groundwater (Smith and Kalin, 2000; Song et al., 2009; Van de Moortel et al., 2010; Tanner and Headley, 2011; Chen et al., 2012; Winston et al., 2013). However, no information is available concerning the treatment of water contaminated with chlorinated ethenes using HPRMF.

In the same systems as in this study, the HPRMF showed a higher removal of low-chlorinated benzenes (monochlorobenzene, 1,2- and 1,4-dichlorobenzene, and 2-chlorotoluene) than the HSSF CW (Chen et al., submitted for publication). The objectives of this study were (1) to test the feasibility of using the HPRMF to treat the highly chlorinated aliphatic hydrocarbon PCE; (2) to compare the treatment performance of HPRMF and HSSF CW for PCE and the associated metabolites; (3) to investigate the effect of a discontinuous outflow flushing operation of the HSSF CW on the treatment of water contaminated with PCE.

## 2. Materials and methods

### 2.1. Setup of the experimental pilot-scale constructed wetlands

The HSSF CW was established in March 2003 and the HPRMF system in March 2010 in Bitterfeld, Germany. Each system consisted of a container of 6 m in length, 1 m in width and 0.6 m in height. The HSSF CW was filled with local aquifer material to a height of 0.5 m and planted with common reed (*Phragmites australis*). The local aquifer material consisted of gravel (25–47%), sand (50–67%) and silt/clay (2–8%). Thus the system had a hydraulic conductivity ( $k_f$  value) of  $2.1 \times 10^{-3} \text{ m s}^{-1}$  and a porosity of 0.28. More details about this HSSF CW are described elsewhere (Braeckevelt et al., 2011). The HPRMF was set up with 3-yr old well-developed plant root mats from *P. australis*, and a porosity of 0.70. The height of the densely interwoven root mats was about 30 cm. Both systems were continuously supplied with contaminated groundwater with a flow rate of  $5.0 \text{ L h}^{-1}$ . The water levels were adjusted to 0.4 m in the HSSF CW and 0.2 m in the HPRMF by a float valve in an open water compartment (a perforated column with a diameter of 20 cm) at a flow path of 5.5 m. By this water level of 0.2 m, the HPRMF sat on the bottom of the container without floating. The effluent was pumped out and water volumes were recorded by flow meters. The calculated average hydraulic retention times from the estimated porosities in the HSSF CW and HPRMF were about 134 h and 168 h, respectively, without considering the varying water loss.

Beginning in March 2003, the HSSF CW was operated with local contaminated groundwater, the main organic pollutant being monochlorobenzene. In order to investigate the performance of the HSSF CW for treating high chlorinated aliphatic hydrocarbons, PCE was added to the inflow since May 2006 at a concentration of around  $2 \text{ mg L}^{-1}$ . To improve the nutrition of the plants,  $30 \text{ mg L}^{-1}$  ammonium was added to the inflow water since March 2008, it was increased to  $50 \text{ mg L}^{-1}$  in April 2009. The groundwater contained a high sulfate concentration of about  $850 \text{ mg L}^{-1}$  and a chloride content of  $330 \text{ mg L}^{-1}$ . The main pollutants concentration in the inflow are listed in Table 1, detailed information on mineral composition of the groundwater was described elsewhere (Braeckevelt et al., 2008).

With the intention to test the effect of different flow regimes on the removal of PCE and its metabolites in the HSSF CW, the flow mode was changed from a continuous flow to a discontinuous outflow flushing procedure on July 21, 2010. From this time on, the HSSF CW was operated in a weekly cycle regime. The fast outflow flushing lasted only 2 h causing a fast decrease in the water level from 40 cm to 15 cm. The subsequent refilling ( $5.0 \text{ L h}^{-1}$ ) to a water level of 40 cm lasted about 34 h, followed by a continuous inflow/outflow phase of 132 h until a new cycle was started.

### 2.2. Sampling and analysis

In total, 12 sampling campaigns were carried out from May 11 to November 8, 2010. Inflow water samples were taken directly

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