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Performance of pilot-scale horizontal subsurface flow constructed wetlands treating groundwater contaminated with phenols and petroleum derivatives

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ABSTRACT

Groundwater contaminated with various organic and inorganic pollutants is a major issue especially in historic industrial areas with chemical industries and refineries. In this study, Constructed Wetlands, as an ecological and environmentally sustainable alternative, were tested at pilot-scale for the removal of phenolic compounds and petroleum derivatives from contaminated groundwater in a pump-and-treat remediation research facility in Germany. Three horizontal subsurface-flow Constructed Wetlands (two planted, one unplanted) were fed with contaminated groundwater containing methyl *tert*-butyl ether (MTBE), benzene and ammonia. In two of the beds, a solution of phenol and *m*-cresol (15 and 2 mg/L or 314.5 and 45.5 $mg/m^2/d$, respectively) was injected to the groundwater inflow. Results showed a complete removal of the two phenolic compounds in the beds without any alteration in the MTBE and benzene removal rates. This indicated that Constructed Wetlands are versatile and can be used to effectively treat different pollutants simultaneously. Higher contaminant removal efficiency of planted systems confirms the positive role of plants presence and their ability to promote biodegradation. Spatial distribution analyses showed that the major portion of the removal took place in the first part of wetland length, which indicated that the systems could potentially deal with higher loads and can be used to optimize the system design.

1. Introduction

Contaminated water at brownfields and industrial sites may pose a major environmental threat for ecosystems and public health, due to historic operational practices, incidents and leakages during storage and transport (Langwaldt and Puhakka, 2000; Seeger et al., 2011; Levchuk et al., 2014). Various toxic pollutants can end up in groundwater bodies, causing a significant ecological risk and posing a potential threat for public health, especially when these bodies would be used as drinking water sources or if they get in contact with surface waters.

Among the various pollutants, fuel hydrocarbons like BTEX compounds (benzene, toluene, ethylbenzene, and xylenes), MTBE (methyl-*tert*-butyl-ether) and phenolic compounds are very common in contaminated groundwater (Wu et al., 2006; Seeger et al., 2011; Van Afferden et al., 2011; Stefanakis et al., 2014).

BTEX compounds and MTBE are highly soluble and mobile groundwater contaminants of great concern. Benzene, is considered as a human carcinogen (USEPA, 2015). Due to the related health risks, concentration limits have been regulated for both benzene (5 μg/L, respectively) in drinking water (USEPA, 2009).

Phenols are organic contaminants present in wastewaters of different origin (Stefanakis et al., 2014), such as oil refineries and petrochemical industry (Abdelwahab et al., 2009), tanneries (Costa et al., 2008), olive mills (Herouvim et al., 2011), cork producing industry (Silva et al., 2015) and pulp and paper mills (Abira et al., 2005), while the usage of pesticides and disinfectants as well as natural sources like forest fires also contribute to phenol contamination (Stottmeister et al., 2010). Phenol presence in water and wastewater could be toxic to plants in case this water is (re)used for irrigation, while it may be toxic for bacteria too (Farré et al., 2001; Nair et al., 2008; Stefanakis et al., 2014). Phenol is one of the most toxic pollutants in wastewater, even at low concentrations

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(Herouvim et al., 2011) and is a priority pollutant with a limit of 1 mg/L in wastewater (WHO, 2011)

A pump-and-treat approach is often applied for remediation of impacted groundwater. The groundwater is pumped up and treated above ground using various physical and chemical methods such as membrane separation, adsorption onto porous media (e.g., activated carbon, zeolites), advanced oxidation processes (e.g., H_2O_2/O_3, H_2O_2/UV), air stripping and vapour extraction (Levchuk et al., 2014). However, these techniques may require significant expertise and complex equipment, may be accompanied by operational safety risks and finally also have high construction, operation and maintenance costs which makes them technically or financially infeasible.

Constructed Wetlands (CWs) are considered today as an attractive sustainable alternative remediation technology with robust performance characteristics, reduced construction costs and minimum operation and maintenance costs (Kadlec and Wallace, 2009; Stefanakis et al., 2014). The technology of CWs is widely recognized as one of the "green" options for decentralized water/wastewater treatment. They have been effectively applied for the treatment of domestic and municipal wastewater, for wastewater produced from various industrial and agricultural installations, as also for sludge dewatering (Vymazal, 2009; Kadlec and Wallace, 2009; Stefanakis et al., 2009, 2011, 2014). The purification capacity of CWs relies on various naturally occurring physical, chemical and biological processes that take place within the system and degrade the various pollutants, as a result of the synergetic actions of the system components, i.e., substrate media, plant roots and microbial community (Stefanakis et al., 2014).

Until today, the application of CWs for the treatment of groundwater contaminated with fuel hydrocarbons is limited. Studies have shown promising results for the removal of benzene and MTBE using horizontal subsurface-flow (HSSF) CWs (Seeger et al., 2011; Chen et al., 2012) or vertical flow (VF) CWs (Van Afferden et al., 2011; De Biase et al., 2013). Moreover, wastewaters with high phenol concentration (up to 500 mg/L) have been treated with CWs, e.g., domestic wastewater spiked with phenols (Tee et al., 2009), wastewater from olive mills (Del Bubba et al., 2004; Herouvim et al., 2011; Kapellakis et al., 2012), cork industry (Silva et al., 2015) and refineries (Knight et al., 1999) but respective applications for phenol contaminated groundwater have not been widely tested (Bedessem et al., 2007). As it is obvious, for these wastewater types inflow concentrations are usually higher than the concentration typically found in contaminated groundwater, while treatment often takes place in multistage CW systems. However, these studies provide good indications that phenolic compounds can be effectively treated in CW systems.

The present study focuses on providing a better understanding of the role of the system parameters (e.g., plant presence, loading rate) for the overall phenolic compound removal efficiency, and investigating the response of the system when various contaminants like benzene, MTBE and phenolic compounds are simultaneously present in the water. Specific objectives of the study are to evaluate: (i) the efficiency of HSSF CWs in the treatment of groundwater contaminated with phenolic compounds, benzene and MTBE, (ii) the role of system parameters such as plant presence for system performance, (iii) the possible interactions and/or treatment limitations due to the simultaneous presence of these different compounds, and (iv) the operational and environmental parameters that affect the performance.

2. Materials and methods

2.1. Site description

The experimental facility is located near the town of Leuna, Saxony-Anhalt, Germany, next to former refinery facilities and an industrial area. There is an extensive contamination of local groundwater and soil due to the large-scale ammonia processing industry for the production of fertilizers and explosives and to petroleum refinement for more than 100 years. At this site a pump-and-treat approach is used to contain and remediate the groundwater. Part of this pumped water has been used over the past 10 years for a series of experimental studies with constructed wetlands (Jechalke et al., 2010; De Biase et al., 2013; Van Afferden et al., 2011; Seeger et al., 2011, 2013).

2.2. Project description

The project consisted of two phases during which phenol and *m*-cresol were added to the groundwater injected into defined pilot-scale units: the preliminary phase (P1), which lasted about 3 months (14 August–24 October 2012) and included the setup of the experimental and operational procedures, the feasibility demonstration of the project approach and the testing of the applied evaluation methods, and the main phase (P2), which lasted about 8 months (8 April–27 November 2013) and covered the entire growth and active season of the plants. Table 1 presents the full timetable of the project and the important dates and respective actions taken during the project lifetime.

During the preliminary phase, 11 field sampling campaigns were organized. The main phase included 14 sampling campaigns during the phenol/*m*-cresol injection period, one sampling campaign before the injections started and two campaigns after the end of the injections. Reed biomass was harvested and weighed during the first days of December 2013, almost one and a half month after the injections stopped.

2.3. Description of pilot-scale units

Three pilot-scale HSSF CWs referred to as A–C were used. Each HSSF CW bed consisted of a steel basin (length × width × depth = 5.9 × 1.1 × 1.2 m, surface 6.5 m^2; Fig. 1). All beds were filled with fine gravel (grain size 2–3.2 mm) up to

Table 1
Project timetable and experimental activities. Sampling took place simultaneously in all beds. Beds A (planted) and B (unplanted) received contaminated groundwater with injected phenol/*m*-cresol, bed C (planted) received contaminated groundwater without phenol/*m*-cresol.

Experimental phase	Date	Activity
Preliminary (P1)	06 Aug–24 Oct 2012	Phenol/*m*-cresol injection period 11 field sampling campaigns
Main (P2)	08 Apr 2013	Field sampling (before injections period)
P2a (Q = 11 L/h)	17 Apr–5 Aug 2013	Phenol/*m*-cresol injection period 14 field sampling campaigns
P2b (Q = A, C: 11 L/h, B: 22 L/h)	5 Aug–23 Oct 2013 30 Oct, 27 Nov 2013 11 Dec 2013	 2 field samplings (after injections period) Reed harvesting and weighing