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Evaluation of the hydrological flow paths in a gravel bed filter modeling a horizontal subsurface flow wetland by using a multi-tracer experiment

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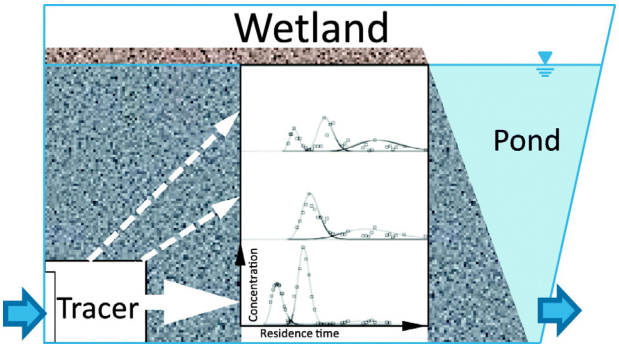
HIGHLIGHTS

- The three-dimensional hydrological flow path was studied in an active constructed wetland remediation system.
- A multiple flow system was inferred from a combination of tracer test and mathematical modeling.
- Main water flow was at the bottom of the wetland.
- An advanced interpretation of remediation efficiencies of such systems was proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

In recent years, constructed wetland systems have become into focus as means of cost-efficient organic contaminant management. Wetland systems provide a highly reactive environment in which several removal pathways of organic chemicals may be present at the same time; however, specific elimination processes and hydraulic conditions are usually separately investigated and thus not fully understood. The flow system in a three dimensional pilot-scale horizontal subsurface constructed wetland was investigated applying a multi-tracer test combined with a mathematical model to evaluate the flow and transport processes. The results indicate the existence of a multiple flow system with two distinct flow paths through the gravel bed and a preferential flow at the bottom transporting 68% of tracer mass resulting from the inflow design of the model wetland system. There the removal of main contaminant chlorobenzene was up to 52% based on different calculation approaches. Determined retention times in the range of 22 d to 32.5 d the wetland has a heterogeneous flow pattern. Differences between simulated and measured tracer concentrations in the upper sediment indicate diffusion dominated processes due to stagnant water zones. The tracer study combining experimental evaluation with mathematical modeling demonstrated the complexity of flow and transport processes in the constructed wetlands which need to be taken into account during interpretation of the determining attenuation processes.

1. Introduction

Constructed wetlands have become of increasing interest in recent years as an economical solution for treating wastewater. Subsurface

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flow constructed wetlands provide a heterogeneous filter and buffer system where abiotic and biotic processes, e.g. sorption, hydrolysis, photolysis, evaporation and biodegradation, may take place simultaneously, resulting in contaminant removal (Imfeld et al., 2009). Wetlands have been shown efficient in the removal of organic contaminants such as the chlorinated solvents, pharmaceuticals, personal health care products, pesticides as well as the immobilization of metals (Gambrell, 1994; Imfeld et al., 2009; Kidmose et al., 2010; Li et al., 2014; Onesios et al., 2009; Schmidt et al., 2014; Verlicchi and Zambello, 2014). However, the processes contributing to the removal are not fully understood and of high interest for future wetland applications. The currently developed applications for industrial and urban scale water treatment, however, require a detailed understanding of the intertwined processes occurring in the highly heterogeneous environment of wetland systems (Imfeld et al., 2009). In particular, a thorough understanding of the water flow characteristics determining the retention time and flow paths will be a requisite to run such systems efficiently (Zahraeifard and Deng, 2011) and before up-scaling of these approaches can be successful. Then the implementation can be considered as feasible alternative or supplement for conventional wastewater treatment or contaminated site management (Gearheart et al., 1989).

To date, several studies were published addressing constructed wetland processes in more detail. For example, the contributing processes to chloroethene (Imfeld et al., 2010), monochlorobenzene (Braeckevelt et al., 2007; Schmidt et al., 2014) as well as MTBE (Jechalke et al., 2010; Rakoczy et al., 2011), pharmaceuticals (Matamoros and Bayona, 2006) or metals (Weis and Weis, 2004) removal and immobilization and related microbial biomass (Tietz et al., 2007; Truu et al., 2009) was described (Imfeld et al., 2009). However, these processes also need an insight into the hydrology and the flow characteristics of such wetland systems (King et al., 1997; Machate et al., 1997; Małoszewski et al., 2006a; Małoszewski et al., 2006b; Ranieri et al., 2011). Indeed, the understanding of the flow characteristics in a wetland is of utmost importance for determining the overall residence time of the water in the system and thus the contact time between the reactive surfaces and contaminant of interest (Małoszewski et al., 2006b). Overall, the residence time of a contaminant in a treatment system needs to be longer to the time required for its degradation to reach a complete removal. Often, only the average residence time is determined as only samples at a single point without depth distinction within the gravel bed or at the outlet are considered in constructed wetlands (Guo et al., 2017; Małoszewski et al., 2006b; Ranieri et al., 2011). The theoretical retention time can be approximated as the ratio between pore water volume within the bed and the applied volumetric flow rate assuming homogeneous and plug flow conditions and that all water in the wetland is mobile. In reality, no such homogeneous systems exist and not only average residence times are of importance but the residence time distributions (RTD) covering the entire range of potential reaction times. Particularly heterogeneities like preferential flow or stagnant water zones are of importance as they decrease or increase the residence time, respectively. In surface water dominated wetlands (Holland et al., 2004; Lange et al., 2011) as well as in subsurface flow systems (Kidmose et al., 2010; Langergraber, 2008; Małoszewski et al., 2006b), the combined use of tracers and mathematical modeling have been adequate tools to identify and quantify heterogeneous flow paths and residence times which can be different from theoretical calculations. Most of these tracer applications measure the tracer breakthrough curves (BTCs) only at the outlet. The contribution of different process towards removal, e.g. aerobic, anaerobic or abiotic, are not clear as such systems are from high complexity in their flow path due to changes in flow, evapotranspiration, rain gains, heterogeneous distribution of plants, of load and so on which make each system almost unique (Imfeld et al., 2009). Therefore, the objective of this study was to identify complex horizontal and vertical transport processes in a constructed wetland. Specifically, we aimed to use a combined multiple tracer and mathematical modeling approach to quantify different flow paths, transport processes and residence times within the constructed wetland. The outcome of this study will contribute to a general understanding of heterogeneous hydrological conditions in similar systems which are crucial to understand observed removal of contaminants. As a model system, a constructed wetland fed with monochlorobenzene (MCB) contaminated groundwater was used, consisting of a planted and unplanted segment (Schmidt et al., 2014). For this study, only the unplanted segment was investigated. In the unplanted gravel bed filter of this horizontal subsurface-flow constructed wetland system, about 40% of the MCB was removed in the gravel bed while over the transition zone into the pond another 50% was eliminated leading to an overall reduction of MCB of 90% compared to the inflow. The removal was apparently linked to iron reduction, however, due to the uncertainties in flow paths and actual residence time, the rate of removal and active regions could not be assessed.

2. Material and methods

2.1. Model constructed wetland system

The horizontal subsurface flow wetland system within the SAFIRA project located directly at the contaminated field site in Bitterfeld, Germany consisted of a stainless steel basin with the dimension of 6 m (length) × 1 m (width) × 0.7 m (depth) (Fig. 1) with a gravel bed (5 m × 1 m × 0.6 m) and a free water pond (1 m × 1 m × 0.5 m) at the outflow side (Figs. S1–S3) (Kaschl et al., 2005; Schmidt et al., 2014). The grain size grading of the filter material was assessed following standard test DIN 18123 resulting in Gaussian distribution of the granular size between 0.63 mm and 6.3 mm. Total measured porosity was 41% ($\varepsilon = 0.41$). The water flow was generated through external pumps at the inflow and outflow. Inflow was continuous using a rotary piston pump from Ismatec (Wertheim, Germany) with a flow rate (q) held at 1 L h^{-1} representative for the conditions in the aquifer corresponding to a hydraulic loading rate (HLR) of the subsurface flow gravel bed and free water pond of 4.8 mm d^{-1} and 24 mm d^{-1}, respectively (Kadlec and Wallace, 2008). The water level at the outflow was held constant at 50 cm (10 cm under gravel bed surface) using a tubing pump from Ismatec (Wertheim, Germany) controlled by a float sensor from Kobold Messring (Hofheim, Germany). Water flow mass balances during the tracer test were calculated from the pump rates.

2.2. Tracer test

A multi tracer test was performed from April to May 2011 (16.3 °C mean air temperature). The gravel bed was covered with polyethylene foil to minimize processes like evaporation, dilution through rain or photodegradation of the tracer chemicals. A mixture of bromide (as KBr; 550 mg L^{-1}), uranine (120 μg L^{-1}) and deuterium oxide (0.14 at%; $\delta D = 8050‰$) in contaminated groundwater from the regional aquifer was injected. Three tracers were chosen which had different diffusion coefficients, thus allowing identifying stagnant water zones and diffusion dominated transport processes (Knorr et al., 2016). The tracers were added as a pulse injection over 20 h at the flux of 1 L h^{-1} from a separate tank which was connected to the inflow of the wetland. Afterwards, the inflow was reconnected directly to the groundwater feed keeping the same flow conditions. Samples were taken at 4 m and, as indicated, at 4.5 m from the inflow central in the gravel bed at three different depths (−27.5 cm; −37.5 cm; −47.5 cm from water surface level) simultaneously with a peristaltic pump from Ismatec (Wertheim, Germany) at a relatively low flow rate of 4 mL min^{-1} to minimize artificial influences to the flow. Polyethylene scintillation vials from VWR (Darmstadt, Germany) were filled with 20 mL pore water for each sampling point and time and were stored in 8 °C temperature in the dark prior to analysis. The sampling regime is listed in detail in SI.