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Adsorption and biodegradation of three selected endocrine disrupting chemicals in river-based artificial groundwater recharge with reclaimed municipal wastewater

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ABSTRACT

Endocrine disrupting chemical (EDC) pollution in river-based artificial groundwater recharge using reclaimed municipal wastewater poses a potential threat to groundwater-based drinking water supplies in Beijing, China. Lab-scale leaching column experiments simulating recharge were conducted to study the adsorption, biodegradation, and transport characteristics of three selected EDCs: 17β-estradiol (E2), 17α-ethinylestradiol (EE2) and bisphenol A (BPA). The three recharge columns were operated under the conditions of continual sterilization recharge (CSR), continual recharge (CR), and wetting and drying alternative recharge (WDAR). The results showed that the attenuation effect of the EDCs was in the order of WDAR > CR > CSR system and E2 > EE2 > BPA, which followed first-order kinetics. The EDC attenuation rate constants were 0.0783, 0.0505, and 0.0479 m^{-1} for E2, EE2 and BPA in the CR system, respectively. The removal rates of E2, EE2, and BPA in the CR system were 98%, 96% and 92%, which mainly depended on biodegradation and were affected by water temperature. In the CR system, the concentrations of BPA, EE2, and E2 in soil were 4, 6 and 10 times higher than in the WDAR system, respectively. According to the DGGE fingerprints, the bacterial community in the bottom layer was more diverse than in the upper layer, which was related to the EDC concentrations in the water-soil system. The dominant group was found to be proteobacteria, including Betaproteobacteria and Alphaproteobacteria, suggesting that these microbes might play an important role in EDC degradation.

Introduction

China has been suffering from water shortages, especially in northern arid areas. Artificial groundwater recharge with treated reclaimed wastewater provides many advantages, such as groundwater supplementation, mitigation of declining groundwater levels, and storage of reclaimed water for later usage (Wu et al., 2011). The artificial recharge of several aquifers has been successfully accomplished at many other locations around the world (Samadder et al., 2011; Vandenbohede et al.,

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2009; Greskowiak et al., 2005; Bouwer, 2002). The occurrence of endocrine-disrupting chemicals (EDCs) in river and groundwater has recently raised serious public concern. The release of EDCs from reclaimed wastewater treatment plants (RWWTPs) and the subsequent occurrence of EDCs in receiving rivers can impact groundwater quality. The concentrations of estrogens in the effluent of secondary wastewater treatment are generally at low levels (from 0.1 to 12.3 μg/L), and even tertiary-treated reclaimed municipal wastewater contains an appreciable concentration of EDCs, approximately 0.1–87.4 ng/L, which may alter the normal hormone functions and physiological status of humans and wildlife (Grovera et al., 2011). Various natural and synthetic chemicals that can induce estrogen-like responses have been identified, including natural steroids (17β-estradiol (E2)) and synthetic chemicals (17α-ethinyl estradiol (EE2) and bisphenol A (BPA)) (Stasinakis et al., 2010). Estrogen is normally found at a low level (nanograms per liter) with a high estrogenic activity and mainly includes natural steroidal estrogen, such as 17b-estradiol (E2), and the synthetic contraceptive 17a-ethinylestradiol (EE2) (Li et al., 2013a).

EDC accumulation and the free flow of water in the soil profile are pre-conditions for EDC leachage into groundwater (Ju et al., 2006). EDCs can move downward with the percolation of water and might lead to estrogen pollution in groundwater. Excessive EDC in groundwater is a serious public health problem in most areas of northern China because drinking water in this region comes from groundwater. EDCs in groundwater have been intensively studied in the following places: artificial aquifer recharge sites (Díaz-Cruz and Barceló, 2008), effluents of wastewater treatment plants (Qiang et al., 2013), and urban aquatic environments (Musolff et al., 2010). In the aforementioned sites, large amounts of reclaimed water and river water containing sewage effluents have infiltrated the groundwater. Based on the previous studies, it has been shown that this concentration of EDCs in groundwater would influence the normal hormone functions and physiological status of humans (Daniel et al., 2008). However, it is difficult to clarify their spatial and temperature variations, mainly due to the limited monitoring wells and the few sampling campaigns that have come from a single aquifer. Calculating the EDC balance is a potentially useful method for predicting the risk of EDCs leaching into groundwater.

In the vadose zone, EDCs can be adsorbed by soil, migrate with groundwater flow or be degraded by microorganisms. The predominant EDC form is determined by factors such as oxidation and reduction potentials (OPR), pH, dissolved oxygen (DO), temperature, and microbial activity. Therefore, EDC transformation and transport during artificial groundwater recharge are complicated processes. The bacterial community is able to dissipate EDCs, and biodegradation plays a major role in the elimination of BPA pollution in the environment, thus reducing the possible risk of EDCs (Zhang et al., 2013). Previous research works based on traditional culture-dependent approaches have provided a substantial amount of knowledge about the diversity of EDC degraders. However, culture approaches can typically select for microorganisms that are able to compete well under laboratory conditions (Zhang et al., 2012). Yang et al. (2014) reported the aerobic biodegradation of bisphenol A in river sediment and the associated bacterial community change. However, no report has been made available on the change of the microbial community structure in response to EDC biodegradation.

In this study, a lab-scale leaching column was designed to simulate aquifer recharge to further investigate the adsorption, biodegradation, transport and leaching characteristics of EDCs. The purpose of the present study was to simulate EDC transformation and transport, to investigate the microbial community structure in the recharge soil and to investigate its change with EDC biodegradation, thus supporting the development of EDC pollution prevention and control measures during river-based artificial groundwater recharge using reclaimed municipal wastewater in China.

1. Materials and methods

1.1. Vadose zone soil samples

Chaobai River vadose zone aquifer media were mainly composed of water-permeable gravel sand in the upper section, fine sand in the middle section and less-permeable silty clay in the aquiclude section in the Huabei plain of Beijing. Simulation columns numbered 1, 2, and 3 were filled with representative soil vadose zone media from two parts of the Chaobai River (40°10′N, 116°42′E), which is a typical streambed in north China.

The hydrogeological cross-sectional north-to-south view (Fig. 1) shows that the shallow aquifer's stratigraphic number of 80 m increased from east to west as the lithology of the aquifer changed from gradient gravel to fine sand. Table 1 shows the physical and chemical properties of the three representative soil vadose zone media. The physical and chemical properties of the three representative soil vadose zone media were different. Cation-exchange capacity (CEC) was determined following the procedures defined by Chapman (1965). Total organic carbon (TOC) was determined using a solid total organic carbon analyzer (SCSH, TOC-VCPH, Japan) (Gustafsson et al., 1997).

1.2. Raw water

Raw water used for lab-scale experiments was the tertiary effluent from the Yinwenjichao reclaimed water treatment plant in Beijing. The water treatment processes consist of ozone pre-oxidation, a membrane bioreactor, chemical phosphorus removal and disinfection. The water samples were collected and stored in the dark for a maximum of 3 days. The main characteristics of the artificial recharge water were COD 50 mg/L, NH_4^+ 1.0 mg/L, TN 6.7 mg/L, SO_4^{2-} 69 mg/L, and HCO_3^- 317 mg/L. COD was determined by a Hach reagent. The cations were measured by an ICP (X Series, Thermo) apparatus. The anions were measured by ion chromatography (ICS-2500, Thermo).

1.3. Lab-scale column recharge system

The lab-scale installations are shown schematically in Fig. 2. From the feed tank, the raw water was pumped into three aquifer treatment columns with the same flow rate. The three aquifer treatment columns were the same size: 0.20 m in diameter and 1.50 m in height, with a packed-bed height of 0.9 m and a supporting layer height of 0.15 m. The three