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The role of organic matter in the removal of emerging trace organic chemicals during managed aquifer recharge

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ABSTRACT

This study explored the effect of different bulk organic carbon matrices on the fate of trace organic chemicals (TOC) during managed aquifer recharge (MAR). Infiltration through porous media was simulated in biologically active column experiments under aerobic and anoxic recharge conditions. Wastewater effluent derived organic carbon types, differing in hydrophobicity and biodegradability (i. e., hydrophobic acids, hydrophilic carbon, organic colloids), were used as feed substrates in the column experiments. These carbon substrates while fed at the same concentration differed in their ability to support soil biomass growth during porous media infiltration. Removal of degradable TOC (with the exception of diclofenac and propylphenazone) was equal or better under aerobic versus anoxic porous media infiltration conditions. During the initial phase of infiltration, the presence of biodegradable organic carbon (BDOC) enhanced the decay of degradable TOC by promoting soil biomass growth, suggesting that BDOC served as a co-substrate in a co-metabolic transformation of these contaminants. However, unexpected high removal efficiencies were observed for all degradable TOC in the presence of low BDOC concentrations under well adopted oligotrophic conditions. It is hypothesized that removal under these conditions is caused by a specialized microbial community growing on refractory carbon substrates such as hydrophobic acids. Findings of this study reveal that the concentration and character of bulk organic carbon present in effluents affect the degradation efficiency for TOC during recharge operation. Specifically aerobic, oligotrophic microbiological soil environments present favorable conditions for the transformation of TOC, including rather recalcitrant compounds such as chlorinated flame retardants.

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1. Introduction

Managed aquifer recharge (MAR) systems, such as riverbank filtration (RBF) and soil aquifer treatment (SAT), are widely used natural processes for drinking water augmentation projects using source water that might be impaired by wastewater discharge. Previous studies have demonstrated that MAR systems are effective in dampening and reducing

the concentrations of dissolved organic carbon (DOC) as well as various trace organic contaminants (TOC) that might be present in impaired source waters (Drewes and Fox, 1999; Brauch et al., 2000; Grünheid et al., 2005). The presence of TOC has become a key concern for drinking water augmentation projects during the past decade (Kolpin et al., 2002; Heberer, 2002; Focazio et al., 2008). Although adverse human health effects caused by these compounds at concentrations

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commonly observed in impaired water resources are very unlikely (Schwab et al., 2005), minimizing exposure of wastewater derived contaminants in these projects is desired. Previous research on the fate of TO_{RC} during MAR has primarily focused on collecting anecdotal and site specific information on their occurrence and removal (Drewes et al., 2002; Montgomery-Brown et al., 2003; Grünheid et al., 2005; Dillon et al., 2008). Studies delineating the mechanisms and boundary conditions for the transformation of wastewater derived TO_{RC} during MAR are lacking.

Previous studies demonstrated that the type and bioavailability of effluent organic matter (EfOM) controls the extent of soil biomass growth in MAR systems (Rauch and Drewes, 2004; Rauch and Drewes, 2005). EfOM may consequently effect the degradation of TO_{RC} by serving as a co-substrate in microbiologically facilitated transformations (Stratton et al., 1983). The diversity and expression of the soil microbial community also depends on composition and concentration of the organic carbon substrate controlling trophic cycles in the subsurface (Preuß and Nehrkorn, 1996; Szewzyk et al., 1998). The composition of EfOM (i.e. in terms of its bioavailability) is primarily determined by the degree of wastewater treatment employed (Drewes and Fox, 1999), which can vary widely from primary to conventional to advanced wastewater treatment. As a result, effluent qualities fed to MAR systems can vary in biodegradable dissolved organic carbon (BDOC) concentrations from less than 1 up to 15 mg/L or more. As a consequence, soil microbial communities growing on different levels of BDOC can differ widely in total biomass and diversity.

The objectives of this research were to investigate the role of 1) abiotic vs. biotic conditions, 2) BDOC and 3) the type of organic carbon matrices on the removal of select TO_{RC}, such as pharmaceutical residues, personal care products, and household chemicals, during MAR.

2. Methodology

2.1. Target organic contaminants

Compounds selected for this study represent small molecular weight organic chemicals (180 to 360 Dalton) that are hydrophilic at neutral pH regimes as indicated by an octanol/water partition coefficient at pH 7 ($\log D_{\text{pH}=7}$ of less than 2.6). TO_{RC} with these properties have a high potential to migrate into groundwater and are not expected to be adsorbed onto porous media. The molecular structures and physicochemical properties of the target compounds chosen for this study are presented in Table 1. These compounds cover a wide range of biodegradability as previously reported for soil/water systems.

The anticonvulsants carbamazepine and primidone have been classified as recalcitrant during wastewater treatment and MAR in earlier studies (Heberer, 2002; Drewes et al., 2003; Clara et al., 2004). The chlorinated phosphate esters tris(1-chloroisopropyl)-phosphate (TCPP) and tris(2-chloroethyl)-phosphate (TCEP) are two widely used flame retardants and are persistent in the aquatic environment (Heberer et al., 2001; Fries and Püttmann, 2003). During bank filtration in Germany, however, TCPP and TCEP exhibited a significant reduction, which was attributed to biotransformation in the aquifer

(Heberer et al., 2003). Amy and Drewes (2006) also reported removal of TCEP to concentrations below the detection limit after two years of subsurface travel in an MAR facility supporting that chlorinated flame retardants can be biotransformed under anoxic conditions. Propyphenazone is a poorly biodegradable analgesic that persists during RBF (Heberer et al., 2003) and SAT (Drewes et al., 2003). For diclofenac, a popular analgesic drug, low removal due to biodegradation or adsorption was reported (Buser et al., 1998; Möhle et al., 1999) unless soils contain a high organic carbon content (Drillia et al., 2003). Several studies report a faster degradation of diclofenac under anoxic conditions as compared to aerobic conditions (Zwiener and Frimmel, 2003; Hua et al., 2003). Opposing results were reported by Schmidt et al. (2004) in that diclofenac was almost completely removed during aerobic bank filtration but recalcitrant during anaerobic recharge. Ibuprofen, ketoprofen, and naproxen are common analgesics that are well degradable during wastewater treatment (Buser et al., 1999; Zwiener and Frimmel, 2003; Carballa et al., 2004) and during soil infiltration (Sedlak and Pinkston, 2001; Drewes et al., 2003). Gemfibrozil is a commonly prescribed blood lipid regulator found in unconfined shallow aquifers impacted by wastewater infiltration in the low to moderate ng/L-concentration range (Heberer and Stan, 1997; Drewes and Shore, 2001; Heberer, 2002). Gemfibrozil was removed below the limit of detection within a few weeks during groundwater recharge using SAT (Drewes et al., 2003).

2.2. Analytical methods

2.2.1. GC/MS analysis

The following TO_{RC} were analyzed by gas chromatography coupled with mass spectrometry (GC/MS) using a HP 6890 gas chromatograph and a HP 5973 quadrupole mass spectrometer from Agilent Technologies (Waldbronn, Germany) adopting a method published by Reddersen and Heberer (2003): gemfibrozil, primidone, diclofenac sodium salt, carbamazepine, ketoprofen, naproxen, phenacetine, tris(2-chloroethyl)phosphate (TCEP) (Sigma Aldrich Chemicals), tris(chloropropyl)phosphate (TCPP), and propyphenazone (Pfaltz & Bauer, Inc.). Stock solutions were prepared by dissolving the compounds in milli-Q water adjusted to a pH of 10, during sonification and in the dark. A volume of 500 mL of sample was collected and filtered (0.45 μm , Whatman) prior to solid phase extraction. 10,11-dihydrocarbamazepine (Sigma Aldrich Chemicals) and 2-(*m*-chlorophenoxy) propionic acid (Sigma Aldrich Chemicals) were used as surrogate standards. The limits of detection (LoFD) and limit of quantification (LoQ) for the target compounds ranged from 5 to 10 ng/L and from 10 to 50 ng/L, respectively. Only parent target compounds were investigated in this study, metabolites and conjugates were not analyzed.

2.2.2. HPLC analysis

A high performance liquid chromatography (HPLC) HP 1100 (Agilent Technologies) combined with a UV diode array detector (DAD) was used to quantify concentrations of primidone, phenacetine, carbamazepine, naproxen, diclofenac, and ibuprofen in the lower $\mu\text{g/L}$ range during adsorption breakthrough tests. The samples were directly injected without filtration or other

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