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Modeling the fate of organic micropollutants during river bank filtration (Berlin, Germany)

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ABSTRACT

Emerging organic contaminants (EOCs) are frequently detected in urban surface water and the adjacent groundwater and are therefore an increasing problem for potable water quality. River bank filtration (RBF) is a beneficial pretreatment step to improve surface water quality for potable use. Removal is mainly caused by microbial degradation of micropollutants, while sorption retards the transport. The quantification of biodegradation and adsorption parameters for EOCs at field scale is still scarce. In this study, the fate and behavior of a range of organic compounds during RBF were investigated using a two dimensional numerical flow- and transport model. The data base used emanated from a project conducted in Berlin, Germany (NASRI: Natural and Artificial Systems for Recharge and Infiltration). Oxygen isotope signatures and hydraulic head data were used for model calibration. Afterwards, twelve organic micropollutants were simulated with a reactive transport model. Three compounds (primidone, EDTA, and AMDOPH) showed conservative behavior (no biodegradation or sorption). For the nine remaining compounds (1.5 NDSA, AOX, AOI, MTBE, carbamazepine, clindamycin, phenazone, diclofenac and sulfamethoxazole), degradation and/or sorption was observed. 1.5 NDSA and AOX were not sorbed, but slightly degraded with model results for $\lambda = 2.25e^{-3}$ 1/d and $2.4e^{-3}$ 1/d. For AOI a $\lambda = 0.0106$ 1/d and $R = 1$ were identified. MTBE could be characterized well assuming $R = 1$ and a low 1st order degradation rate constant ($\lambda = 0.0085$ 1/d). Carbamazepine degraded with a half life time of about 66 days after a threshold value of 0.2–0.3 μg/L was exceeded and retarded slightly ($R = 1.7$). Breakthrough curves of clindamycin, phenazone, diclofenac and sulfamethoxazole could be fitted less well, probably due to the dependency of degradation on temperature and redox conditions, which are highly transient at the RBF site. Conditions range from oxic to anoxic (up to iron-reducing), with the oxic and denitrifying zones moving spatially back and forth over time.

1. Introduction

The increasing pollution of the aqueous environment with organic micropollutants, sometimes referred to as emerging organic contaminants (EOCs), is a worldwide problem (Halling-Sorensen et al., 1998; Ziylan and Ince, 2011). In recent years, a large number of EOCs such as pharmaceutical residues, personal care products, adsorbing organic halogens or industrial chemicals have been increasingly detected in surface waters as well as in groundwater (Loeffler et al., 2005).

River bank filtration (RBF) as a pretreatment step to improve surface water quality for potable use has a long history, and is still in use in many parts of the world (Hiscock and Grischek, 2002; Tufenkji et al., 2002). However, in urban partly closed water cycles the quality of the bank filtrate can be affected by discharge of treated wastewater into the surface water upstream of the RBF system (Reemtsma et al., 2006). This has, for example, been demonstrated for the city of Berlin in Germany (e.g. Heberer et al., 1998, 2008; Ziegler,

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2001). Wastewater associated EOCs that are not or only partly removed during wastewater treatment may be retained or depleted during underground passage towards the RBF wells due to sorption and/or microbial degradation. Removal efficiencies thereby typically depend on the bioaccessibility of the compounds, prevailing redox conditions (Greskowiak et al., 2006; Gruenheid et al., 2005; Massmann et al., 2006), residence times and temperatures (Massmann et al., 2006) within the aquifer. The quantification of removal efficiencies (in terms of percent removal between river and RBF well) of EOCs in RBF systems has previously been addressed in a number of studies (e.g. Maeng et al., 2011; Massmann et al., 2008a; Wiese et al., 2011 and references therein). Certainly, a rigorous quantification of biodegradation kinetics (e.g., first-order rate constants or Monod-parameters) and adsorption parameters (e.g., linear distribution coefficients) for EOCs during RBF at field scale is still scarce. The few studies addressing this subject are limited to a small set of compounds (e.g., Greskowiak et al., 2006; Mueller et al., 2011).

Due to the highly transient nature of RBF and other managed aquifer recharge systems such as (deep) well injection and recovery or ponded infiltration, a thorough understanding and process-based analysis about the transport behavior of organic and inorganic contaminants typically requires detailed numerical groundwater flow and multi-component reactive transport modeling, as has been demonstrated in several previous studies (e.g., Greskowiak et al., 2005, 2006; Prommer and Stuyfzand, 2005; Saaltink et al., 2003; Sharma et al., 2012; Wallis et al., 2011). These rather detailed studies carried out multi-component reactive transport modeling for a rather limited number of contaminants, i.e., one or two contaminants.

The present study, therefore, aimed to quantify the attenuation behavior for a total number of twelve EOCs, which have been detected during RBF in Berlin, Germany in previous investigations (Grünheid et al., 2005; Heberer, 2002a; Heberer et al., 2002; Massmann et al., 2008a). Since quite a large number of EOCs was considered, a detailed analysis of the influencing processes on the transport behavior such as redox and temperature effects was omitted. Instead, the present study's objective was to estimate apparent field scale first-order rate constants and linear distribution coefficients for the studied EOCs through the application of a numerical groundwater flow and multi-species reactive transport model developed for the studied site.

Though first-order rate constants and linear distribution coefficients provide only a rather simplified description of the biodegradation and adsorption processes (since they neglect influences of physico-chemical conditions and their temporal and spatial variability) they are nevertheless useful to provide first estimates of the fate of EOCs at field scale in general and are already available for some RBF sites (Mueller et al. 2011, Schmidt, 2005). In contrast to the frequently given values in % removals at a specific site (e.g. Gruenheid et al., 2008), they allow for comparisons between sites.

The compounds investigated in the present study include the pharmaceutically active compounds (PhACs) diclofenac, phenazone, primidone, clindamycin, carbamazepine, sulfamethoxazole (SMOX) and the metabolite 1-acetyl-1-methyl-2-dimethyl-oxamoyl-2-phenylhydrazide (AMDOPH). Furthermore, the organic pollutants ethylenediaminetetraacetic acid (EDTA, complexing agent), methyl tert-butylether (MTBE, gasoline additive), 1.5 naphthalenedisulfonic acid (1.5 NDSA, intermediate product of dyestuff industry) and X-ray contrast media, adsorbable organic halogens (AOX) and adsorbable organic iodines (AOI) are considered. Each of these compounds is present in Berlin's surface and groundwater (Heberer, 2002a, 2002b; Heberer et al., 2002; Scheytt et al., 2005; Wiese et al., 2011) but only limited or no information about field scale biodegradation and adsorption parameters so far exists. Hence, this knowledge is of great importance for the site management at Berlin and comparable RBF systems worldwide.

2. Materials and methods

2.1. Lake Tegel RBF site and previous work

2.1.1. Study area and instrumentation

In Berlin's semi closed water cycle, water production is partially based on the indirect reuse of wastewater (Gruenheid et al., 2005; Ziegler et al., 2002). RBF is induced by abstraction wells placed around surface water courses all over Berlin. Secondary treated wastewater effluents are discharged into receiving surface waters which are the source of the bank filtrate.

The study area (Fig. 1) is located in the north west of Berlin, at the eastern shore of the Lake Tegel, where a well gallery of the Berlin Waterworks (Berliner Wasserbetriebe, BWB) is situated. The lake water quality is influenced by the Upper Havel, entering the lake from the south–west and the discharge of treated wastewater from the wastewater treatment plant (WWTP) Schönerlinde (Ziegler, 2001), passing by the Nordgraben (Fig. 1). The wastewater release is relatively constant over the year. According to Ziegler (2001), Lake Tegel contains 17–35% treated wastewater.

The present study focuses on a transect (transect Tegel) near the production well 13 (Figs. 1,2). The transect consists of a sequence of groundwater monitoring wells between the eastern shore of Lake Tegel and the inland observation well I (Fig. 2). Three shallow (wells A, B, and C), two intermediate (wells D and E), two deep (wells F and G) and one very deep (well H) monitoring wells that are oriented approximately in groundwater flow direction were considered. The filter screens reach depth between 5 and 38 m. Well locations and length of the filter screens are given in Fig. 2.

The lake and transect were intensely investigated within a large 3-year project named NASRI (Natural and Artificial Systems for Recharge and Infiltration; KompetenzZentrumWasser, Berlin, 2009, see data origin below) and subsequent studies. So far, a number of publications reported on investigations at the Lake Tegel RBF transect. Data of a range of pharmaceuticals and their residues were presented by Heberer and Adam (2004), Heberer et al. (2004) and Massmann et al. (2007, 2008, 2009). Gruenheid et al. (2005) presented results of bulk dissolved organic carbon (DOC) and some trace organic compounds. Environmental tracer data was evaluated by Massmann et al. (2008c). Massmann and Sueltenfuß (2008) assessed excess air data derived from noble gas analysis. Wiese and Nuetzmann (2009, 2011) performed hydraulic investigations. Sulfonamides and psychoactive compounds were investigated by Richter et al. (2008) and Hass et al. (2012) respectively. Finally, a statistical analysis of NASRI EOC data from the RBF transect Tegel was conducted by Wiese et al. (2011).