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Modeling the transport behavior of 16 emerging organic contaminants during soil aquifer treatment

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HIGHLIGHTS

- Attenuation potential for secondary treated effluents by pond recharge investigated
- Description of transport behavior for 16 organic micropollutants
- Field data evaluated with one-dimensional reactive transport model
- In-situ parameter of sorption and degradation derived from model
- Comparison of parameters with literature values

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ABSTRACT

In this study, four one-dimensional flow and transport models based on the data of a field scale experiment in Greece were constructed to investigate the transport behavior of sixteen organic trace pollutants during soil aquifer treatment. At the site, tap water and treated wastewater were intermittently infiltrated into a porous aquifer via a small pilot pond. Electrical conductivity data was used to calibrate the non-reactive transport models. Transport and attenuation of the organic trace pollutants were simulated assuming 1st order degradation and linear adsorption. Sorption was found to be largely insignificant at this site for the compounds under investigation. In contrast, flow path averaged first order degradation rate constants were mostly higher compared to the literature and lay between 0.036 d^{-1} for clofibric acid and 0.9 d^{-1} for ibuprofen, presumably owing to the high temperatures and a well adapted microbial community originating from the wastewater treatment process. The study highlights the necessity to obtain intrinsic attenuation parameters at each site, as findings cannot easily be transferred from one site to another.

1. Introduction

Soil-aquifer treatment (SAT) is a common application of managed aquifer recharge (MAR). It utilizes the natural filter and removal capacity of the soil and aquifer via ponded infiltration to enhance the quality of surface or (treated) wastewater for later extraction and supply (Drewes et al., 2003; Dillon, 2005). The increasing load of emerging organic contaminants (EOCs) such as pharmaceuticals in waste and surface waters, however, may pose a significant problem for MAR, since many of these substances are recalcitrant during subsurface passage. As a result, some of them have already been detected in production wells of riverbank filtration and ponded infiltration systems (Ternes, 1998; Laws et al., 2011). Even though the effect on human health has not yet been proven for the detected concentration range (ng L^{-1} to μg L^{-1}), the increasing detection of waste water originated compounds is seen to be problematic for a safe water supply also under ethical aspects (Daughton and Ternes, 1999; Clara et al., 2005).

Predicting the fate of emerging compounds during MAR does not only require a sound understanding of the physical flow and transport processes and MAR operation, but also about the fate of these substances as a result of biodegradation and sorption processes (e.g. Greskowiak et al., 2005; Maeng et al., 2011a). However, a detailed

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process identification at operational scale requires a comprehensive set of hydraulic and hydrogeochemical data (Greskowiak et al., 2006), which in practice, and thus outside focused research projects, is rarely available. For practical considerations during MAR operation simple 1st-order degradation rate constants and linear adsorption coefficients provide useful information to quantify the removal of EOCs for a first-pass assessment (Stuyfzand et al., 2007; Patterson et al., 2010; Henzler et al., 2014). However, to date, only limited information about field-scale biodegradation and sorption parameters is available, raising the urgent need for more field experiments and their quantitative evaluation.

For this purpose a model-based quantification of a field-scale experiment on the transport behavior of sixteen selected common pharmaceuticals during soil aquifer treatment was carried out. The field trial was conducted at a test site consisting of a pilot pond placed in an alluvial aquifer in Northern Greece into which conventionally treated effluent was infiltrated. The aim was to extract field-scale biodegradation and adsorption parameters with the help of transient flow and reactive transport modeling. As at field scale it is commonly difficult to separate processes due to the lack of known input functions and the lack of a simultaneous ideal reference tracer, the experimental design was adapted to allow for this aspect.

Sixteen EOCs with different transport properties were investigated in this study. They include X-ray contrast agents (iohexol, iomeprol, iopamidol, iopromide), antibiotics (sulfamethoxazol), anticonvulsants (primidone, carbamazepine, diazepam), lipid regulators (clofibric acid, bezafibrate, gemfibrozil), anti-inflammatory drugs (phenazone, naproxen, ibuprofen, diclofenac) and antihistamines (cetirizine) which have been detected in secondary treated effluents (STEs) from the local waste water treatment plant (WWTP) in the typically encountered concentration ranges (Loos et al., 2013; Verlicchi et al., 2012).

The results of the present study add to the presently only scarcely available sorption and biodegradation parameters of EOCs under specific environmental conditions. They are finally compared with those of other studies that were conducted under differing environmental conditions, thereby putting them in a wider scientific context.

2. Materials and methods

2.1. Site description and hydrogeology

The test site, located north of the WWTP of Thessaloniki (Greece), includes an experimental infiltration pond and a network of monitoring wells (Fig. 1). The infiltration pond as well as a pipeline from the WWTP was constructed in summer 2007 for the purpose of the infiltration experiment. The unconfined aquifer is mainly composed of Pleistocene and Holocene alluvial deposits (Schaffer et al., 2012b). Further information on the mineral composition of the alluvial material can be found in Niedbala et al. (2013). The thickness of the aquifer ranges from 30 m to 120 m with a relatively high hydraulic conductivity varying between 7×10^{-4} m s^{-1} and 3×10^{-3} m s^{-1}. The existing unsaturated zone extends to depths between 2 and 2.5 m directly beneath the bottom of the pond.

2.2. Experimental conditions

Recharge experiments were conducted from August 28 to September 28, 2008. The pond (Fig. 1) was alternately filled with tap water and secondary-treated wastewater. Tap water was applied in order to obtain uniform conditions for the whole research area and to enhance potential desorption of the target compounds from the aquifer material, whereas STE was used to investigate the behavior of these compounds during subsurface passage. The schedule for the alternating tap and waste water application is detailed in Table 1. During the experiments, the water level within the pond varied between 0 and 1.5 m above the pond basis depending on the supply rate to it, leading to varying infiltration rates (Fig. 2) and at least temporally an unsaturated zone. They, in turn, generated highly transient groundwater flow conditions in the vicinity of the pond.

The observed water temperatures were relatively high throughout the entire experiment. In the pond water, temperatures varied from 20 °C to around 29 °C, while temperatures in the monitoring wells lay between 20 and 24 °C. High concentrations of nitrate were observed in the pond (Fig. 3) and the strongly decreasing concentrations during

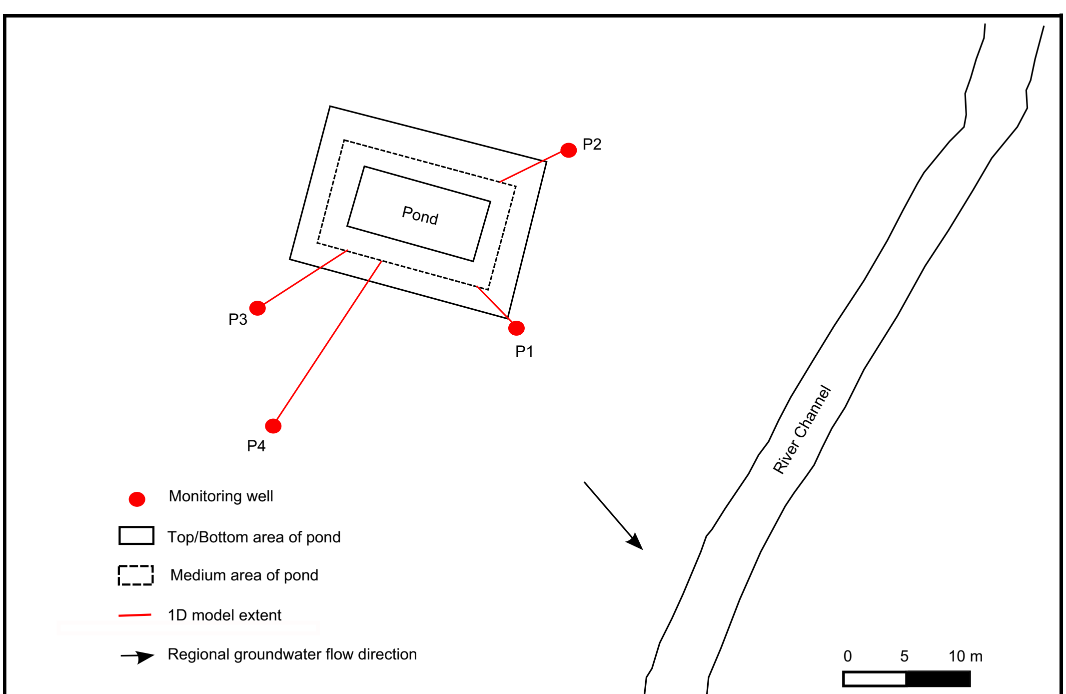

Fig. 1. Map of the investigation area showing the location of infiltration pond, surrounding observation wells as well as extension of the 1D models.