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Influence of a compost layer on the attenuation of 28 selected organic micropollutants under realistic soil aquifer treatment conditions: Insights from a large scale column experiment

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ABSTRACT

Soil aquifer treatment is widely applied to improve the quality of treated wastewater in its reuse as alternative source of water. To gain a deeper understanding of the fate of thereby introduced organic micropollutants, the attenuation of 28 compounds was investigated in column experiments using two large scale column systems in duplicate. The influence of increasing proportions of solid organic matter (0.04% vs. 0.17%) and decreasing redox potentials (denitrification vs. iron reduction) was studied by introducing a layer of compost. Secondary effluent from a wastewater treatment plant was used as water matrix for simulating soil aquifer treatment. For neutral and anionic compounds, sorption generally increases with the compound hydrophobicity and the solid organic matter in the column system. Organic cations showed the highest attenuation. Among them, breakthroughs were only registered for the cationic beta-blockers atenolol and metoprolol. An enhanced degradation in the columns with organic infiltration layer was observed for the majority of the compounds, suggesting an improved degradation for higher levels of biodegradable dissolved organic carbon. Solely the degradation of sulfamethoxazole could clearly be attributed to redox effects (when reaching iron reducing conditions). The study provides valuable insights into the attenuation potential for a wide spectrum of organic micropollutants under realistic soil aquifer treatment conditions. Furthermore, the introduction of the compost layer generally showed positive effects on the removal of compounds preferentially degraded under reducing conditions and also increases the residence times in the soil aquifer treatment system via sorption.

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1. Introduction

Due to their suspected but often still unknown toxic effects, over the last decade attention has been drawn to the occurrence and fate of organic micropollutants (OMPs), such as pharmaceuticals, personal care products, artificial sweeteners, and pesticides in the aquatic environment (Ferrari et al., 2003; Jones et al., 2004; Tran et al., 2013). Many OMPs are able to resist the treatment in wastewater treatment plants (WWTPs) (Reemtsma et al., 2010; Loos et al., 2013). As a consequence, WWTP treated effluents contain OMPs up to the mg L^{-1} range (Loos et al., 2013) and represent the major source of OMPs in the aquatic environment (Halling-Sørensen et al., 1998; Heberer, 2002).

Managed aquifer recharge, such as soil aquifer treatment (SAT), is a promising and widely applied post-treatment technology to improve the quality of the treated effluent and, thus, of the water released into the environment. The efficiency in terms of OMP removal from the aqueous phase during the passage of treated wastewater through different SAT systems has been studied by several authors (e.g., Drewes et al., 2003; Asano and Cotruvo, 2004; Amy and Drewes, 2007). Among others, Amy and Drewes (2007) demonstrated that SAT is an effective method to remove OMPs from WWTP effluents, although a few OMPs still persist. The recently reported occurrence of OMPs in groundwater, impacted by recharge with treated wastewater, highlights the importance of an improved understanding of the OMP behavior during their subsurface passage (Teijon et al., 2010; Huerta-Fontela et al., 2011; Jurado et al., 2012). Major attenuation processes in SAT systems are sorption at solid/water interfaces and (bio) degradation in the water phase or biofilms (Katayama et al., 2010; Maeng et al., 2011; Schaffer et al., 2012a). The hydrophobic sorption potential of OMPs to solid organic matter (SOM) is determined by the molecule polarity, which usually correlates well with the *n*-octanol/water partition coefficient log K_{OW} for neutral compounds and can be estimated by the K_{OC} concept (Karickhoff et al., 1979; Sabljić et al., 1995). However, many OMPs have dissociable functional groups (acidic/basic) and form ionic (non-hydrophobic) species within the common pH range of WWTP effluents (Schaffer and Licha, 2014). For organic anions (deprotonated acids), therefore, the pH-dependent *n*-octanol/water partition coefficient log *D* considering the acid dissociation constant pK_a should be used instead of log K_{OW} for predicting the sorption affinity (Kah and Brown, 2007; Franco and Trapp, 2008; Schaffer et al., 2012a). However, this does not apply for organic cations (protonated bases) where a stronger sorption is usually observed (Kah and Brown, 2007; Schaffer et al., 2012b; Niedbala et al., 2013; Kutzner et al., 2014). In contrast, biodegradation of OMPs is an even more complex field in environmental sciences and, compared to sorption processes, it is rather difficult to derive any prediction from the physico-chemical compound properties alone. Several authors observed a significant impact of redox conditions (Massmann et al., 2006, 2008; Maeng et al., 2010; Barbieri et al., 2011; Liu et al., 2011), the concentration of biodegradable dissolved organic carbon (BDOC) (Quintana et al., 2005; Maeng et al., 2011; Hoppe-Jones et al., 2012), and the presence of adapted microorganisms (Hoppe-Jones et al., 2012; Grenni et al., 2013). From these findings, technical strategies to specifically stimulate OMP elimination during SAT can be derived.

Therefore, the overall objective of this study was to examine and compare the fate of 28 widespread OMPs in systems with and without a DOC-releasing solid matrix under realistic SAT conditions. Column experiments were conducted in order to obtain defined hydraulic conditions (steady-state flow), full recovery of the injected water and, thus, to simplify the data evaluation and interpretation. In contrast to many other laboratory scale studies, the used columns have a comparatively large size (≈150 L) and were fed on-site with secondary effluent of a WWTP in order to simulate chemically and microbially realistic recharge conditions. Due to the use of a real wastewater matrix, the injected water also contained, e.g., nutrients and adapted microorganisms from the activated sludge treatment. Since the fate of OMPs highly depends on aquifer properties and redox conditions, two different column systems (both with two columns in duplicate) with varying contents of SOM were used. The applied setup with and without compost layer allows studying the behavior of OMPs with respect to different redox conditions, fractions of organic carbon (f_{OC}), and releases of biodegradable dissolved organic carbon (BDOC). Since the fate of organic substances further depends on their transport properties, a wide spectrum of OMPs (8 neutral, 12 anionic, 7 cationic compounds, and 1 zwitter-ion) was investigated. Eventually, the results provide insight into the retardation and degradation behavior of many OMPs under different aquifer conditions and can allow deriving further implications for the design and operation modes of SAT systems.

2. Materials and methods

2.1. Organic micropollutants

In this study, 28 substances were selected to represent a broad bandwidth of environmentally relevant OMPs with different structural properties. Table 1 provides relevant physico-chemical properties, charge states and the respective limits of quantification (LOQ) for the studied OMPs.

2.2. Secondary effluent as feed water containing the OMPs

The column systems were installed at the WWTP El Prat de Llobregat (Barcelona, Spain) and fed with its treated effluent (mechanical treatment followed by activated sludge and denitrification). The WWTP facilities receive the wastewater from Barcelona and other surrounding towns (≈420,000 m^3 d^{-1}). For the experiments, the secondary effluent was additionally spiked with 1 μg L^{-1} of DIA, ISO, PHE, DCL, MCP, SMX, CLA, FLX, HAL, and TMX, since these compounds were not detected or not present in sufficiently high concentrations in the original secondary effluent. The resulting inflow concentrations were constant and are shown in the Supplementary data (Figs. S10–S12).