Science of the Total Environment 612 (2018) 985–994

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Evaluation of EOC removal processes during artificial recharge through a reactive barrier

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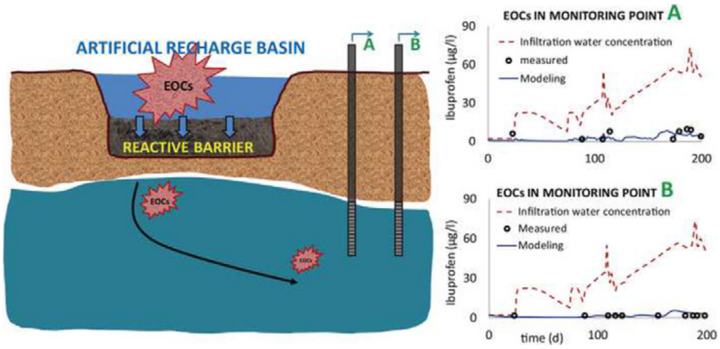
HIGHLIGHTS

- This study shows that a reactive barrier can enhance EOC removal.
- The barrier releases DOC to favor a broad range of redox states.
- Biotransformation and sorption were estimated with a numerical model for ten EOCs.
- The model was calibrated against an experimental database.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 20 June 2017
Received in revised form 5 August 2017
Accepted 5 August 2017
Available online 6 September 2017

Editor: D. Barcelo

Keywords:
Artificial recharge
Reactive barrier
Emerging organic compounds
Attenuation processes estimation

ABSTRACT

A reactive barrier that consisted of vegetable compost, iron oxide and clay was installed in an infiltration basin to enhance the removal of emerging organic compounds (EOCs) in the recharge water. First-order degradation rates and retardation factors were jointly estimated for 10 compounds using a multilayer reactive transport model, whose flow and conservative transport parameters were previously estimated using hydraulic head values and conservative tracer tests. Reactive transport parameters were automatically calibrated against the concentration of EOCs measured at nine monitoring points. The degradation rate of each compound was estimated for three zones defined according to the redox state, and retardation coefficients were estimated in two zones defined according to the organic matter content. The fastest degradation rates were obtained for the reactive barrier, and the estimated values were similar to or higher than those estimated in column and/or field experiments for most of the compounds (8/10). Estimated retardation coefficients in the reactive barrier were higher than in the rest of the aquifer in most cases (8/10) and higher than those values estimated in previous studies. Based on the results obtained in this study the reactive barrier seems to be able to enhance the removal of EOCs.

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http://dx.doi.org/10.1016/j.scitotenv.2017.08.054
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1. Introduction

Artificial recharge of water that is directly or indirectly affected by wastewater effluents is an option for improving the qualitative state of the recharge water and the quantitative state of groundwater bodies and associated surface water bodies. This option, however, may be limited by the presence of emerging organic compounds (EOCs), which are typically constituents of these effluents, and are detected in the groundwater, surface water, and even drinking water (Schwarzenbach et al., 2006). EOCs, such as pharmaceutical residues, are biologically active, but remain unregulated, which has raised concern. Many EOCs are not eliminated by conventional treatment plants, which were not designed to remove them (Chefetz et al., 2008). These contaminants may reach the aquatic environment through treatment plant effluents and, infiltration from livestock, industry, or farming areas (Lapworth et al., 2012; Rivera-Utrilla et al., 2013).

Several active treatment procedures are reported for removing EOCs, such as advanced oxidation, reverse osmosis, and nanofiltration (Zhang et al., 2011; Rivera-Utrilla et al., 2013). These procedures are generally expensive in terms of investment and energy (Rivera-Utrilla et al., 2013; Maeng et al., 2011b). In contrast, passive treatment systems, such as bank filtration and artificial recharge via infiltration basins, may be a low-cost alternative to advanced water treatment procedures (Drewes et al., 2003b; Shamrukh and Abdel-Wahab, 2008; Maeng et al., 2010; Valhondo et al., 2014; Alidina et al., 2014). However, the efficiency of passive systems depends on numerous factors and varies from site to site. Therefore, quantitative evaluations of efficiency and robustness at the field scale are required for these systems to became common practice.

The design of natural treatment methods requires an understanding of the processes that influence the fate of EOCs during subsurface flow. The main processes are sorption, generally to organic matter and clay minerals, and biological degradation or transformation (Lapworth et al., 2012). Sorption is generally viewed as a straightforward process, although its actual characterization can be complicated by natural heterogeneity and competition for sorption sites. Biotransformation is difficult to assess because EOC concentrations are typically low (ranging from a few nanograms per liter to a few hundred micro grams per liter). Therefore, it is very unlikely that EOCs would be used as a primary substrate, so it can be assumed that they are biotransformed by co-metabolism (Tran et al., 2013). Co-metabolic degradation implies the presence of a primary substrate, which provides enough energy to support microbial growth (Tran et al., 2013). The bioavailability of biodegradable DOC as the primary substrate affects the microbial community structure and consequently also affects EOC biotransformation (Alidina et al., 2014; Rauch-Williams et al., 2010; Li et al., 2013; Mazahirali et al., 2014; Regnery et al., 2015). Biodegradation of the primary carbon source, DOC, produces different enzymes depending on the electron acceptor (metabolic path). These enzymes are involved in the transformation of EOCs, and therefore the redox conditions affect the EOC biotransformation. In fact, several authors have described the redox-dependence of the biotransformation of diverse EOCs (Greskowiak et al., 2005; Greskowiak et al., 2006; Massmann et al., 2008; Barbieri et al., 2011; Storck et al., 2012; Liu et al., 2013; Valhondo et al., 2015) and how changing redox conditions might be effective for removing such contaminants (Maeng et al., 2011b). Other factors, such as temperature (Massmann et al., 2006), pH, nutrient availability, and residence time (Massmann et al., 2008) also affect EOC removal during subsurface passage.

Current research is yielding a body of evidence that supports the engineering of artificial recharge with methods to promote varying types of sorption sites and redox conditions. Most research, however, is performed at the laboratory scale (Alidina et al., 2014; Li et al., 2013; Bertelkamp et al., 2014; Regnery et al., 2015; Schaffer et al., 2015). While these work facilitate process understanding, they miss potential synergetic effects associated with the heterogeneous and complex hydrochemical conditions that are inherent in natural systems. Thus, sorption and biotransformation parameters derived from laboratory tests cannot be directly used at field sites due to the heterogeneity and complexity of natural systems. A lower number of studies have been performed at field scale where a natural or an added tracer was used to estimate recharge water residence time (Grünheid et al., 2005; Henzler et al., 2014; Laws et al., 2011; Nham et al., 2015; Wiese et al., 2011). The degradation rates were then estimated based on the residence time of each sample in the aquifer and the variation of each EOC concentration. In same cases complex reactive transport models accounted for mixing ratios among different water sources (Wiese et al., 2011).

As a novelty with respect the previously referred to studies, a reactive barrier at the bottom of an infiltration basin in an artificial recharge site was installed to promote the processes involved in the attenuation of EOCs (Valhondo et al., 2014, 2015). The barrier comprised vegetable compost, which releases DOC into the infiltrated water, favoring a broad range of redox conditions underneath. Very small fractions of clay and iron oxide were added to increase the sorption sites for cationic and anionic EOCs, respectively. The qualitative behavior of the system turned out to be satisfactory and EOC removal was indeed enhanced (Valhondo et al., 2014, 2015). However, a quantitative evaluation of the effective sorption and biodegradation parameters was needed in order to compare them to others obtained in laboratory and/or field experiments. The sorption and biodegradation rates of ten EOC were estimated using a multilayer 2D model which was previously calibrated against head and tracer concentrations (Valhondo et al., 2016).

Therefore, the objective of this work was two-fold: first, to describe a method for evaluating effective sorption and biotransformation parameters in a real-scale complex artificial recharge system; and, second, to evaluate these parameters and compare them to values available in the literature from laboratory and field conditions.

2. Materials and methods

2.1. Field site

The artificial recharge facility is located in Sant Vicenç dels Horts (Barcelona, Spain), close to the Llobregat River and approximately 15 km from the sea (Fig. 1A). The facility comprises a settlement basin and an infiltration basin (≈5000m^2 each) connected by a pipe. Llobregat River water, affected by numerous treatment plant effluents (Köck-Schulmeyer et al., 2011), was carried into the settlement basin (2–4 days residence time) before flowing to the infiltration basin. A CTD-Diver (Schlumberger Water Services, Delft, The Netherlands) and a flow meter (Teledyne Isco Inc., Lincoln, NE, USA) were installed in the connecting pipe to measure electrical conductivity, temperature, and flow rate of the water entering the infiltration basin. Infiltration rate averaged 1 m/d.

The aquifer beneath the facility, extended to 20–23 m deep with 12–14 m saturated thickness during the experiment time (2010–2014). It contained Quaternary alluvial sediments, mainly gravel and sand with a small fraction of clay (Barbieri et al., 2011), on top of Pliocene marl, which was assumed to be impervious. The varying percentages of gravel, sand, and clay of the sediments caused heterogeneous layering (Gámez et al., 2009). The groundwater flows naturally from NNW to SSE with a mean 2.3‰ gradient (Iribar et al., 1997), but the flow structure changes during artificial recharge periods when the gradient beneath the infiltration basin becomes nearly vertical (≈10%) (Valhondo et al., 2016).

Water was sampled at the infiltration and at eight piezometers and two suction cups to monitor native aquifer water and the evolution of water along the flow path (Fig. 1B). One piezometer, P1, was located far enough upstream to be seldom affected by recharge water and was