Science of the Total Environment 545–546 (2016) 629–640

Contents lists available at ScienceDirect

Science of the Total Environment

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

The fate of organic micropollutants during long-term/long-distance river bank filtration

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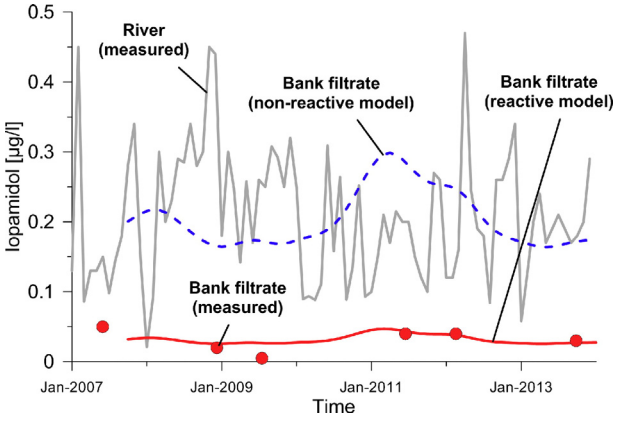
HIGHLIGHTS

- Long-term behavior of organic micropollutants at bank filtration investigated.
- Field data evaluated by numerical reactive transport modeling.
- Most compounds detected in the river completely removed in the bank filtrate.
- Removal parameters for 19 partially and fully removed compounds determined.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 27 October 2015
Received in revised form 13 December 2015
Accepted 13 December 2015
Available online xxxx

Editor: D. Barcelo

Keywords:
River bank filtration
Organic micropollutants
Numerical modeling
Long-term behavior
Removal
Degradation

ABSTRACT

The fate of organic micropollutants during long-term/long-distance river bank filtration (RBF) at a temporal scale of several years was investigated along a row of monitoring wells perpendicular to the Lek River (the Netherlands). Out of 247 compounds, which were irregularly analyzed in the period 1999–2013, only 15 were detected in both the river and river bank observation wells. Out of these, 10 compounds (1,4-dioxan, 1,5-naphthalene disulfonate (1,5-NDS), 2-amino-1,5-NDS, 3-amino-1,5-NDS, AOX, carbamazepine, EDTA, MTBE, toluene and triphenylphosphine oxide) showed fully persistent behavior (showing no concentration decrease at all), even after 3.6 years transit time. The remaining 5 compounds (1,3,5-naphthalene trisulfonate (1,3,5-NTS), 1,3,6-NTS, diglyme, iopamidol, triglyme) were partially removed. Their reactive transport parameters (removal rate constants/half-lives, retardation coefficients) were inferred from numerical modeling. In addition, maximum half-lives for 14 of the fully removed compounds, for which the data availability was sufficient to deduce 100% removal during sub-surface passage, were approximated based on travel times to the nearest well. The study is one of very few reporting on the long-term field-scale behavior of organic micropollutants. It highlights the

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http://dx.doi.org/10.1016/j.scitotenv.2015.12.057

efficiency of RBF for water quality improvement as a pre-treatment step for drinking water production. However, it also shows the very persistent behavior of various compounds in groundwater.

1. Introduction

Contamination of the aquatic environment in general and groundwater more specifically with organic micropollutants from agricultural, industrial and urban sources has received much attention in the past years as evidenced by various review articles (e.g., (Heberer, 2002; Jurado et al., 2012; Lapworth et al., 2012; Loos et al., 2010; Maeng et al., 2011b; Schwarzenbach et al., 2006; Stuart et al., 2012). Concerns arise in particular when the groundwater is exploited for drinking water purposes, especially when it originates from managed aquifer recharge (MAR) systems, for example via river bank filtration (RBF) along the more contaminated rivers. The subsurface has, however, the ability to considerably improve the water quality during RBF and generally acts as a pre-treatment step for drinking water production. According to Hiscock and Grischek (2002), the natural attenuation of pollutants during RBF is mainly caused by filtration, dilution, biodegradation and sorption, leading to the complete removal of suspended solids, bacteria, parasites, viruses and biodegradable compounds as well as the partial removal of adsorbable compounds and other water constituents. Besides other factors, one important criteria for the attenuation capacity of the subsurface is the residence time underground (i.e. the travel time from the river to a production well), which can vary from days to several months and occasionally up to several years between sites (see for example Grischek et al. (2002), Table 1). Various authors have so far investigated the attenuation of organic trace pollutants during RBF, but studies were mainly conducted at sites with relatively short travel times in the range of days to months only (e.g., Achten et al., 2002; Greskowiak et al., 2006; Heberer et al., 2008; Henzler et al., 2014; Massmann et al., 2006; Schmidt et al., 2007; Storck et al., 2012; Wiese et al., 2011), while few have studied the fate of organic micropollutants at sites with a residence time of several years (Stuyfzand et al., 2006; Stuyfzand et al., 2007; Stuyfzand, 1989).

Approaches with different complexities have been used to quantify the fate of organic micropollutants at MAR and RBF sites, including simple calculations of % removals (Heberer et al., 2008; Massmann et al., 2008a), sometimes dependent on the redox conditions (Grünheid et al., 2005; Storck et al., 2012; Stuyfzand, 1998; Wiese et al., 2011), 1st order degradation independent on redox conditions or temperatures (Henzler et al., 2014), 1st order degradation dependent on redox or temperature conditions (Burke et al., 2014a, 2014b; Stuyfzand et al., 2007), and Monod degradation dependent on redox and temperature conditions (Engelhardt et al., 2014; Greskowiak et al., 2006).

The objectives of this study were (i) to assess the long-term/long-distance behavior of a large number of organic micropollutants present in the aquatic environment, thereby categorizing them into very persistent, partially removable and fully removable compounds under the given time-scales, and (ii) to identify field-scale biodegradation and sorption parameters for those compounds that were only partially removed during RBF. For this purpose, a 1D flow and solute transport model for a RBF transect between the Lek River and the drinking water abstraction wells of the Rodenhuis well field in the Netherlands was set up. As the site has very long travel times in the order of years, the focus lies on compounds which have previously mostly been characterized as fully persistent, based on small-scale laboratory studies or field studies at MAR/RBF sites with shorter travel times. The results of this study add to the currently sparse quantitative data available for organic micropollutants at real field sites, in particular for large scales and long travel times.

2. Material and methods

2.1. Field site

The Rodenhuis RBF study site is located on the Lek River, a tributary of the Rhine River in the Netherlands, situated between the cities of Rotterdam and Utrecht (Fig. 1). The transect with observation wells w37, w38 and w39 is aligned along the flow direction between the river and the public supply well field at Rodenhuis, as ascertained by a numerical groundwater flow model (Oasen Drinking Water Company, personal communication).

The hydrogeological conditions at the site are shown in Fig. 2. The upper geological unit consists of interbedded Holocene sandy clay and peat deposits (Westland Formation), together forming the upper aquitard. These deposits are locally intersected by sandy infills of former river channels and the present river channel. The aquifer below consists of a 28 m thick sequence of mid to late Pleistocene fine to coarse sands. Middle Pleistocene fine sandy clays (Kedichem Formation) form an aquitard at the base of the aquifer. The production and observation wells are screened within the main aquifer. The upper aquitard (Westland Formation) acts as a confining layer for the main aquifer. However, it is uncertain where the sandy channels within the upper aquitard are located and whether or not they cut through the upper aquitard completely. In the study area, the river runs through a low-lying, drained landscape and is constrained by a dike (Fig. 2). The

Table 1
Groundwater travel times during river bank filtration at selected sites in Asia, Europe and USA.

Authors	Bank filtration site	Travel times
Sheets et al. (2002)	Ohio (USA)	20 h–3 months
Massmann et al. (2007, 2008b)	Tegel, Wannsee (Germany)	4–5 months Decades (deep bank filtration)
Vogt et al. (2009)	Thur (Switzerland)	<60 days
Hunt et al. (2005)	La Crosse, Wisconsin (USA)	<1 year
Grischek et al. (2002)	Summary of European sites	<1 year (14 sites), 1–2 years (3 sites), 2–3 years (6 sites, Danube), >11 years (1 site, Netherlands)
Boving et al. (2014)	Kali River (Karnataka, India)	45 days
Storck et al. (2012)	Missouri (USA)	<10–60 days
	Platte (USA)	7–28 days
	Great Miami River (USA)	1–65 days
	Rhine river(Germany)	7–60 days
	Elbe river(Germany)	4–150 days
	Ruhr river(Germany)	5–15 days
Stuyfzand et al. (2006, 2007)	Summary of Dutch sites	2 days ->36 years