



Temperature dependent redox zonation and attenuation of wastewater-derived organic micropollutants in the hyporheic zone



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HIGHLIGHTS

- Attenuation of organic micropollutants during infiltration is temperature influenced.
- Changes in temperature significantly impact the redox milieu.
- Temperature and redox dependencies were found for nearly all investigated compounds.
- The first meter of infiltration is highly reactive regarding micropollutant attenuation.

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ABSTRACT

The hyporheic zone - a spatially fluctuating ecotone connecting surface water and groundwater - is considered to be highly reactive with regard to the attenuation of organic micropollutants. In the course of the presented study an undisturbed sediment core was taken from the infiltration zone of a bank filtration site in Berlin and operated under controlled laboratory conditions with wastewater-influenced surface water at two different temperatures, simulating winter and summer conditions. The aim was to evaluate the fate of site-relevant micropollutants, namely metoprolol, iopromide, diclofenac, carbamazepine, acesulfame, tolyltriazole, benzotriazole, phenazone and two phenazone type metabolites, within the first meter of infiltration dependent on the prevailing temperature. A change in temperature resulted in a development of significantly distinct redox conditions. Both temperature dependencies and related redox dependencies were identified for all micropollutants except for benzotriazole and carbamazepine, which behaved persistent under all conditions. For the remaining compounds degradation rate constants generally decreased from warm and oxic/penoxic/suboxic over cold and oxic/penoxic to warm and manganese reducing (transition zone). Individual degradation rate constants ranged from 0 (e.g. diclofenac, acesulfame and tolyltriazole in the transition zone) to $1.4 \times 10^{-4} \text{ s}^{-1}$ for metoprolol under warm conditions within the oxic to suboxic zone.

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1. Introduction

The occurrence of organic micropollutants within the aquatic environment has been frequently addressed throughout the literature published in recent years and is the topic of numerous review articles (e.g. Burkhardt-Holm, 2010; Jones et al., 2001; Kümmerer, 2009; Schwarzenbach et al., 2006). The entry pathways are diverse. Within urban areas, organic micropollutants are mainly introduced into the water cycle via sewage systems. As contaminants such as pharmaceutical residues, disinfection products and industrial agents are constituents

of municipal and industrial sewage, and they are discharged into wastewater treatment plants where some of them are not or only incompletely removed (e.g. Vieno et al., 2007). In partly closed water cycles, treated wastewater containing residuals of these pollutants is discharged into the surface water and thus resupplied to the water cycle. Apart from treated wastewater as the primary source of organic micropollutants in urban surface waters, runoff from sealed areas may significantly contribute to their contamination as well (e.g. Gan et al., 2012; Meyer et al., 2011). By infiltration of (polluted) surface water into the subsurface, a process which is frequently induced during managed aquifer recharge (MAR), organic micropollutants may consequently be recharged to groundwater (e.g. Heberer, 2002).

The interface between the entities surface water and groundwater – also referred to as the hyporheic zone – is described as a spatially

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fluctuating ecotone, where important ecological processes are influenced by water movement, permeability, substrate, resident biota and physiochemical features (Boulton et al., 1998). As pointed out by Brunke and Gonser (1997) this transition zone plays a critical role regarding a large number of exchange processes between surface and groundwater. It is described as a domain with intensive processing of organic carbon and nutrients (e.g. Findlay et al., 1993; Triska et al., 1993), and also considered to be highly reactive with respect to the attenuation of organic (micro)pollutants (Lawrence et al., 2013). It has frequently been shown that the quality of infiltrating surface water changes significantly within the first meters of infiltration (Heberer et al., 2008; Jüttner, 1999; Schwarzenbach et al., 1983). At riverbank filtration settings, processes responsible for the attenuation of organic micropollutants during passage through the infiltration zone, such as advection, dispersion, sorption/desorption, redox processes and biological transformations (Schwarzenbach et al., 1983), are interdependent and highly transient. A quantitative understanding of the attenuation processes in this highly reactive zone is crucial for predicting contaminant behavior during riverbank filtration, as well as in surface water–groundwater interaction zones in general.

Attenuation processes lead to a significant decrease in contaminant concentrations and are to a large extent microbially mediated (e.g. Scow and Hicks, 2005). The biological activity is supposed to be highest immediately after infiltration, indicated by exponentially declining reaction rates with distance from the point of infiltration (Stuyfzand, 2011). Furthermore, it is well known that the biological activity is strongly influenced by the ambient temperature (e.g. Ratkowsky et al., 1982; Zwietering et al., 1991).

Several studies on surface water–groundwater interactions at different field scales, focusing on the fate and behavior of organic micropollutants, have recently been performed (e.g. Banzhaf et al., 2011; Greskowiak et al., 2006; Hoehn et al., 2007; Lewandowski et al., 2011; Massmann et al., 2008a). Results revealed that important factors determining their attenuation during infiltration are temperature (Massmann et al., 2006) and prevailing redox conditions (Greskowiak et al., 2006), which in turn are mutually dependent.

Nevertheless, as already stated by Lawrence et al. (2013), investigations under controlled conditions focusing on the hyporheic zone within urban settings are widely lacking. In order to partly fill this gap, the herein presented study comprises laboratory experiments investigating the attenuation of 10 wastewater-derived organic micropollutants within the hyporheic zone adjacent to an urban river. For this purpose, an undisturbed sediment core was sampled from the infiltration zone of a riverbank filtration site in Berlin, Germany and operated under laboratory conditions with surface water from the sampling site. In order to simulate winter and summer infiltration conditions, experimental investigations were carried out at 6.5 °C and repeated at 19.7 °C.

The study included wastewater-derived contaminants which were found to be present in trace concentrations (ng L^{-1} to $\mu\text{g L}^{-1}$) in the surface water at the sampling site. The organic micropollutants studied are the pharmaceutical residues diclofenac, carbamazepine, metoprolol and phenazone, the phenazone-type metabolites acetylaminoantipyrine (AAA) and formylaminoantipyrine (FAA), the X-ray contrast media iopromide, the artificial sweetener acesulfame as well as the agents tolyltriazole and benzotriazole. Tolyltriazole and benzotriazole are primary used as anticorrosive agents in many industrial fluids, but also in households in dishwashing agents (Reemtsma et al., 2010).

The aim of the study was to evaluate the fate of the mentioned micropollutants within the hyporheic zone in dependency of prevailing redox conditions and of two different temperatures, representing winter and summer conditions. The results of the present study expand the current knowledge with regard to the transport and fate of wastewater-derived organic micropollutants in the near-shore zone during riverbank filtration and provide temperature and redox

dependent 1st order degradation rate constants for each of the studied compounds.

2. Methods

2.1. Sampling location and core drilling

An undisturbed core was taken from a well investigated study site in Berlin, Germany. The riverbank filtration site at Lake Wannsee, located in the southwest of Berlin, has already been subject of several field studies on the occurrence and behavior of pharmaceutical residues during bank filtration (Heberer et al., 2008; Massmann et al., 2008a, 2008b). Along the shoreline of the lake, the local water supplier operates production wells in order to abstract raw water for drinking water purposes, permanently inducing bank filtration. The hydrochemistry within the infiltration zone at the sampling location is highly transient. It is characterized by strong annual temperature fluctuations, showing variations of about 25 °C over the year, and similarly variable redox conditions, ranging from aerobic to iron reducing conditions (Massmann et al., 2008b).

The core was drilled in the riparian zone at a water depth of approximately 0.3 m. An aluminum tube with an inner diameter of 0.08 m was introduced into the sediment by using a vibrocorer system. Subsequently, the upper opening was sealed with a cap in order to generate a vacuum, and tube and sediment core together were removed using a combination of tripod and pulley. Thus, an undisturbed sediment core of the length of 1.2 m was obtained and brought into laboratory, where it was incorporated into the experimental setup approximately 5 to 6 h after sampling. Temperature during transport was in the range of 5–8 °C.

2.2. Experimental setup and implementation

The core was positioned upside down so as to operate the column in an upward mode in order to avoid gas entrapment. After placement in the laboratory, the column was equipped with eight oxygen probes and eight rhizome samplers at different depths (see Fig. 1). The utilized oxygen probes were constructed according to Hecht and Koelling (2001), but based on 2 mm plastic optical fibers. In order to regularly monitor the oxygen distribution within the core, the probes were connected to a fiber optic oxygen transmitter (Fibox 3, PreSens). The installed rhizome samplers enabled a depth dependent sampling along the core triggered by suction using a syringe. Oxygen probes and sampling valves were additionally installed at the in- and outlet of the column. A schematic drawing of the experimental setup is given in Fig. 1.

The column influent, i.e., surface water originating from the sampling site, was injected by means of a peristaltic pump. The target compounds were found to be present in the utilized surface water with typical concentrations given in Table 1. Thus, there was no need to artificially add them to the inflow solution and the experiment was performed under conditions very similar to those prevalent at the field site. The effluent water was collected in a measuring beaker in order to regularly quantify the flow rate.

Values describing the transport properties within the column (effective porosity, mean pore water velocity and hydrodynamic dispersivity) were derived from a conservative tracer test conducted with bromide. Results revealed an effective porosity n_e of 0.3, a longitudinal dispersion coefficient D_l of $4.5 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$ and a mean pore water velocity v_a of $1.6 \times 10^{-5} \text{ m s}^{-1}$, yielding a hydraulic retention time of 21 h within the column. According to sediment analysis the core mainly consisted of medium sized sand with an organic matter content of 0.4% (w/w).

The column was conditioned for three months until steady state conditions along the monitored depth profile with regard to the redox relevant parameters oxygen (O_2), nitrate (NO_3^-), iron (Fe^{2+}), manganese (Mn^{2+}) and sulphate (SO_4^{2-}) as well as pH value were observed.

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