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Sorption and biodegradation of organic micropollutants during river bank filtration: A laboratory column study



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ABSTRACT

This study investigated sorption and biodegradation behaviour of 14 organic micropollutants (OMP) in soil columns representative of the first metre (oxic conditions) of the river bank filtration (RBF) process. Breakthrough curves were modelled to differentiate between OMP sorption and biodegradation. The main objective of this study was to investigate if the OMP biodegradation rate could be related to the physico-chemical properties (charge, hydrophobicity and molecular weight) or functional groups of the OMPs. Although trends were observed between charge or hydrophobicity and the biodegradation rate for charged compounds, a statistically significant linear relationship for the complete OMP mixture could not be obtained using these physico-chemical properties. However, a statistically significant relationship was obtained between biological degradation rates and the OMP functional groups. The presence of ethers and carbonyl groups will increase biodegradability, while the presence of amines, ring structures, aliphatic ethers and sulphur will decrease biodegradability. This predictive model based on functional groups can be used by drinking water companies to make a first estimate whether a newly detected compound will be biodegraded during the first metre of RBF or that additional treatment is required.

In addition, the influence of active and inactive biomass (biosorption), sand grains and the water matrix on OMP sorption was found to be negligible under the conditions investigated in this study. Retardation factors for most compounds were close to 1, indicating mobile behaviour of these compounds during soil passage. Adaptation of the

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biomass towards the dosed OMPs was not observed for a 6 month period, implying that new developed RBF sites might not be able to biodegrade compounds such as atrazine and sulfamethoxazole in the first few months of operation.

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1. Introduction

In the Netherlands, 6.5% of the total amount of produced drinking water is obtained from river bank filtrate (Geudens, 2012). Traditionally, river bank filtration (RBF) was used for the removal of pathogens, bacteria, protozoa, disinfection by-product pre-cursors, natural organic matter (NOM), etc., as well as dampening variations in water quality. Since organic micropollutants (OMPs) are increasingly being detected in Dutch surface waters (ng/L $- \mu$ g/L range) (Verliefde et al., 2007), interest has risen in the capability of RBF to provide an effective barrier for these compounds. In contrast to other water treatment processes used for OMP removal, such as membranes and advanced oxidation processes, RBF offers natural treatment, low cost, and no requirement for chemical supplementation. These aspects, in combination with the fact that several drinking water companies in the Netherlands already use RBF as a pretreatment step in their multi-barrier treatment trains, make RBF an attractive option for OMP removal. However, insight into the role that OMP characteristics (functional groups, physico-chemical properties) play in to why certain OMPs are removed during RBF while others, such as carbamazepine, show very persistent behaviour is lacking. This makes it very difficult for drinking water companies to assess what type of additional treatment is required to prevent OMPs from penetrating through the treatment trains ending up in the drinking water.

Field parameters influencing OMP removal are subject to large variation. Therefore, the general approach to investigate OMP removal during soil passage (and thus also RBF) is mimicking this process in controlled soil columns. Many laboratory column studies have already been performed to mimic OMP removal during soil passage. These studies focused on: OMP removal under specific redox conditions (Banzhaf et al., 2012; Baumgarten et al., 2011), the effect of temperature on OMP biodegradation (Gruenheid et al., 2008), the fate of OMPs under saturated or unsaturated conditions (Scheytt et al., 2004, 2006), the role that cationic exchange capacity of the soil plays in sorption of cationic OMPs (Schaffer et al., 2012b), the influence of pH on sorption of ionizable compounds (Schaffer et al., 2012a), the effect of the initial OMP concentration dosed (Baumgarten et al., 2011), the effect of the amount and type of organic carbon source present in the water (Baumgarten et al., 2011; Maeng et al., 2011a,b; Onesios and Bouwer, 2012; Rauch-Williams et al., 2010) and comparing OMP removal in a pilot column to full scale field conditions (Benotti et al., 2012). However, these studies do not differentiate between OMP removal via sorption and biodegradation. As such, the individual contribution of these underlying mechanisms on OMP removal is not well understood. More insight into these mechanisms is required to enable the development of predictive models for OMP removal during RBF. Such predictive models (e.g. Quantitative Structure Activity Relationship (QSAR) models) can be used by drinking water companies to assess whether a newly detected compound will be effectively removed during RBF or additional treatment is required. A first step towards this better understanding would be to distinguish between the two main removal mechanisms, sorption and biodegradation (Maeng et al., 2011a, b), to assess which mechanism is the dominant contributor to overall OMP removal. Distinguishing between the two removal mechanisms will also provide insight into the sorption and biodegradation potential of the compounds in relation to each other.

Few studies have attempted to determine OMP biodegradation by comparing a biotic (active biomass) sand column to an abiotic (inactive biomass) sand column at a certain time point and allocate the difference in OMP removal between these two systems to biodegradation (Maeng et al., 2011a, b; Onesios and Bouwer, 2012). However, this only gives a rough estimate of the percentage of OMP biodegraded and can lead to an underestimation when stable effluent OMP concentrations have not been obtained yet. In addition, a difference in OMP removal percentage at one time point does not elucidate how fast a compound is degrading, which makes it more difficult to quantitatively compare between compounds. To enable a fair comparison between OMP biodegradability a more accurate parameter would be the biodegradation rate that can be determined from the OMP breakthrough curve. In addition, modelling the OMP breakthrough curves takes into consideration OMP dispersion and allows determining the retardation factor which represents the extent of OMP sorption.

Most column studies on OMP sorption and biodegradation during soil passage involved only one to four compounds (Baumgarten et al., 2011; Gruenheid et al., 2008; Scheytt et al., 2004, 2006), making it impossible to link physico-chemical properties to OMP removal behaviour and observe trends. The behavior of a larger collection of OMPs in soil column systems has been investigated in a few studies (Maeng et al., 2011a, b; Onesios and Bouwer, 2012; Patterson et al., 2011), and was still mainly limited to negatively charged and neutral compounds, thus not covering a wide range of physicochemical properties. Moreover, many studies dose higher OMP concentrations (10–700 µg/L) (Onesios and Bouwer, 2012; Patterson et al., 2011) than the concentrations found for most compounds to be present in surface water (lower ng/L – μ g/L range) (Verliefde et al., 2007). Baumgarten et al. (2011) reported that increasing the sulfamethoxazole concentration in the influent by one order of magnitude showed significantly better removal of the compound. Thus, dosing higher OMP concentrations than present in the environment could significantly overestimate the biodegradation rates. Therefore it is very important to dose OMPs at concentrations representative of those detected in surface water.

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