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Removal of pharmaceuticals in Moving Bed Biofilm Reactors – The impact of design and operating conditions

Elena Torresi[1,2], Fabio Polesel[1], Barth F. Smets[1], Henrik R. Andersen[1], Magnus Christensson[2], Benedek Gy. Plósz[1,3]

[1]DTU Environment, Technical University of Denmark, Kongens Lyngby, Denmark (elto@env.dtu.dk, fabp@env.dtu.dk, bfsm@env.dtu.dk, hran@env.dtu.dk)
[2]Veolia Water Technologies AB, AnoxKaldnes, Lund, Sweden (magnus.christensson@anoxkaldnes.com)
[3]Department of Chemical Engineering, University of Bath, Claverton Down, Bath, UK (b.g.plosz@bath.ac.uk)

Abstract: Moving Bed Biofilm Reactors (MBBRs) have been recently proposed as a means to enhance the removal of organic micropollutants, e.g., pharmaceuticals, during biological wastewater treatment. This study presents a comparative assessment of three different types of MBBR in terms of removal of pharmaceuticals, including (i) pre-denitrification with single- and multi-stage configurations (MBBR1); (ii) nitrification with controlled biofilm thickness (MBBR2); (iii) and for post-denitrification with dosing of two different external readily degradable carbon sources. Results obtained in long- and short-term experiments revealed an enhancement of biotransformation rate constants – compared to activated sludge process - on average, for a major number of micropollutants of approximately ~60% in the post-denitrification MBBR, possibly due to cometabolism and the use of high readily degradable carbon sources (ethanol and methanol). Comparison of obtained biotransformation kinetics with values from relevant literature for conventional activated sludge suggested MBBRs as effective solution for increasing micropollutant removal in municipal wastewater.

Keywords: micropollutants; MBBR; nitrification; carbon source; wastewater.

Introduction

Numerous organic pollutants, such as pharmaceuticals and personal care products, undergo incomplete removal in conventional municipal wastewater treatment plants (WWTP) due to design and operational limitations, resulting in their discharge to receiving water bodies at trace level. Presently, research has been focusing on the improvement of conventional WWTP performance using novel biological treatment systems. Moving bed biofilm reactors (MBBRs), as alternative to conventional activated sludge, are based on biofilm growth on suspended plastic carriers. Enhanced removal of several pharmaceuticals was accordingly observed in aerobic MBBRs (Falås et al., 2012). However, no study has previously attempted to comprehensively elucidate micropollutant removal capabilities of MBBRs at different stages of the biological treatment train.

In this study, we investigated different optimization strategies using MBBRs towards the removal of 23 commonly detected pharmaceuticals in municipal wastewater under different operational conditions. We assessed the impact of (i) biofilm thickness in nitrifying MBBRs; and (ii) organic carbon quality and availability in anoxic pre- and post-denitrifying MBBRs. Assessed biotransformation kinetics of the targeted pharmaceuticals were compared to relevant literature for activated sludge, thus identifying potential benefits in the employment of MBBRs towards the removal of micropollutants

Material and Methods

Three laboratory-scale MBBR systems were operated with continuous-flow feeding of municipal wastewater from local WWTPs under conditions of pre-denitrification (MBBR1),

nitrification (MBBR2) and post-denitrification (MBBR3). Details of the operational conditions are presented in Table 1. In MBBR1, denitrification and biotransformation of micropollutants were evaluated in a single-stage and a three-stage MBBR configuration where the biofilm was exposed to a decreasing gradient of influent organic carbon loading and degradability. The influence of biofilm thickness (ranging between 50 and 500 µm) was studied in nitrifying MBBR2 using novel Z-carriers, which allowed for the development of biofilm of defined thickness. In MBBR 3, two commonly dosed carbon sources (methanol and ethanol) were compared in parallel post-denitrifying MBBRs. Biokinetics of (de)nitrification and micropollutant biotransformation rate constants (k_{Bio}, L g^{-1} d^{-1}) were estimated from batch experimental results using Activated Sludge Models (ASMs) and ASM for Xenobiotics (ASM-X) (Plósz et al., 2012), respectively.

Results and Conclusions

In MBBR1 study, the highest and lowest biotransformation kinetics were found in the first and the last stage MBBR, respectively (up to 4-fold decrease for selected compounds), which correlated with the estimated denitrification rates in the different stages. In MBBR2 study, the biotransformation of more than 60% of targeted compounds was enhanced in thicker biofilms (>200 µm), while thinner biofilms (~50 µm) resulted in higher biotransformation of the three sulfonamide antibiotics and diclofenac. The methanol-dosed MBBR of the MBBR3 study exhibited an enhancement of k_{Bio} (up to 2.5-fold) for a number of micropollutants (nine out 23) compared to the ethanol-dosed MBBR. Detailed results of the investigations on MBBR1–3 are presented in Torresi et al. (2016, 2017) and Polesel (2016). In Figure 1, the range of values of k_{Bio} obtained for each micropollutant under the conditions tested was reported. A general trend of enhanced biotransformation rate constant k_{Bio} for a major number of micropollutants (~60%) was observed in MBBR3. One of the possible reasons for this observation can be found in the addition of high readily degradable carbon sources (ethanol and methanol) commonly used in post-denitrification that could be beneficial for the removal of a number of micropollutants possibly due the cometabolic effect of the primary substrate. When comparing k_{Bio} estimated in the three MBBRs with values from relevant literature for conventional activated sludge, an improvement of several (but not all) targeted micropollutants was shown, e.g., for atenolol, carbamazepine, citalopram, clarithromycin, diclofenac. It can be thus concluded that MBBRs represents a suitable technology for the optimization of micropollutant removal in municipal wastewater under a range of operating conditions (nitrifying, pre- and post-denitrifying).

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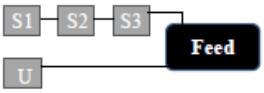
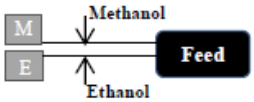
Table 1. Operational conditions of the three MBBR systems, in which pharmaceutical removal was investigated.

	MBBR 1	MBBR 2	MBBR 3
Conditions	Pre-denitrification (anoxic)	Nitrification (aerobic)	Post-denitrification (anoxic)
Configuration			
Carriers	K1	Z-carriers	K1
Conditions tested	Three-stages (S) vs single stage (U)	Different biofilm thicknesses	Two different carbon sources
Continuous-flow operation	3 MBBRs in series (S) 1 MBBR (U)	MBBR with Z200-Z500 carriers (R1) MBBR with Z50 (R2)	MBBR with methanol (M) MBBR with ethanol (E)
Batch experiments	**How**: Separated batch experiments for each MBBR (4 batches) **When**: ~at 3 and 15 months after start-up	**How**: Separated batch experiments for each Z-carrier (5 batches) **When**: ~at 5 and 9 months after start-up	**How**: Separated batch experiments for the 2 MBBRs (5 batches) **When**: ~at 3.5 months after start-up
Feed	Pre-clarified wastewater and addition of NO_3-N	Effluent wastewater and addition of NH_4-N	Nitrified wastewater and addition of methanol or ethanol
Hydraulic residence time	~8.9 h	~2 h	~2 h
Volume	6 L for each system	3 L for R1, 1.5 L for R2	1 L for each MBBR

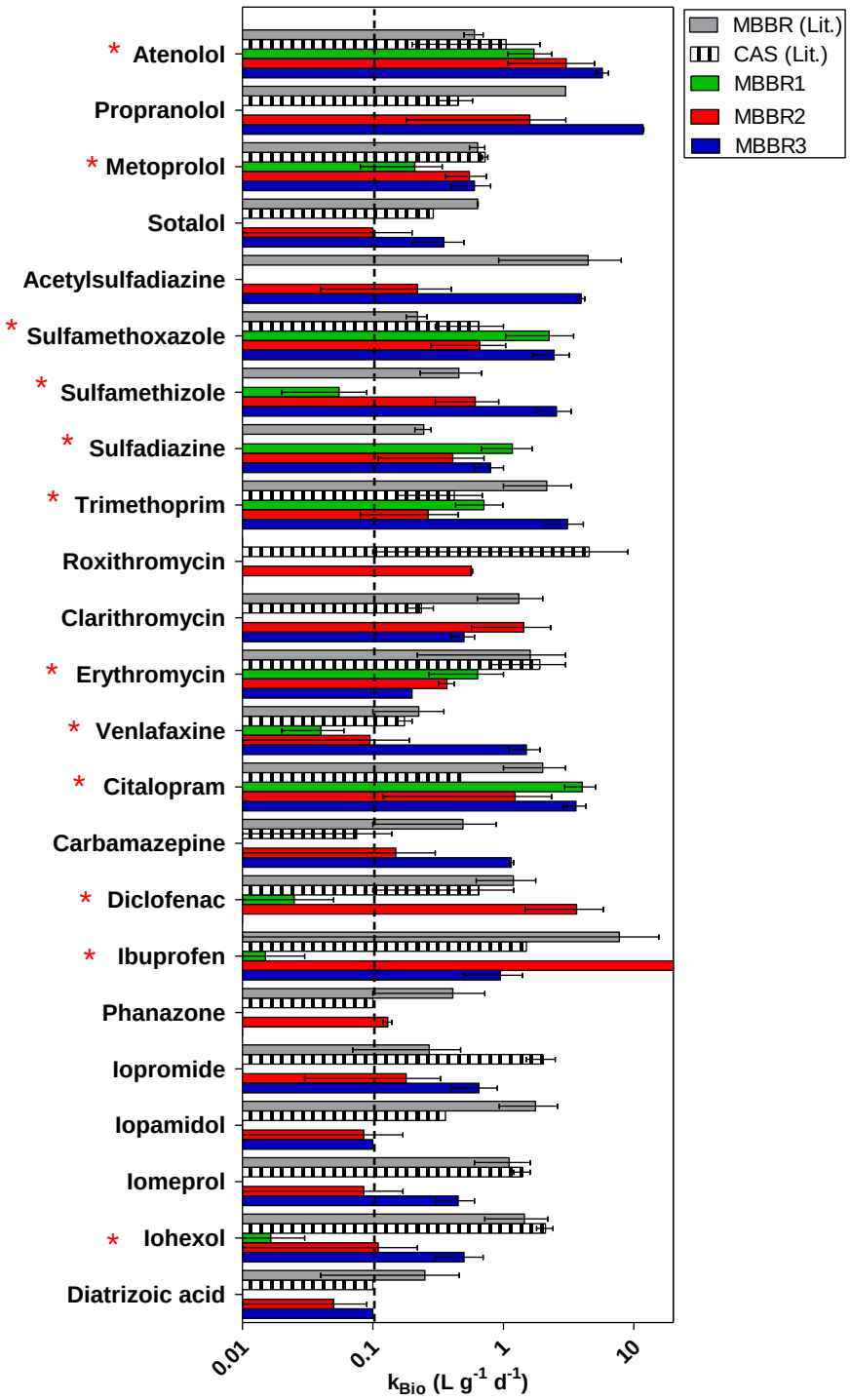


Figure 1. Summary of biotransformation rate constants (k_{Bio}) for targeted pharmaceuticals in MBBR1–3 and comparison with literature values from conventional activated sludge systems (CAS). Asterisks denote pharmaceuticals that were detected in all MBBRs. For a number of compounds, the comparison is reported only between MBBR2 and MBBR3 study, as only micropollutants naturally found in pre-clarified wastewater were studied in MBBR1.