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ELIMINATION OF PHARMACEUTICALS IN SINGLE- AND THREE-STAGE PRE-DENITRIFYING MBBR

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Summary: This study investigated the elimination of pharmaceuticals in pre-denitrifying moving bed biofilm reactors (MBBRs) in single- and three-stage configurations. Under batch conditions, biotransformation and retransformation of two pharmaceuticals (trimethoprim, sulfamethoxazole) occurred at comparable or higher rates than in denitrifying activated sludge. Based on estimated rate coefficients, concentrations in continuously operated MBBRs were predicted.

Keywords: pharmaceutical removal; moving bed biofilm reactor (MBBR); pre-denitrification

Introduction. Moving bed biofilm reactors (MBBRs) have been recently proposed as a means to enhance the elimination of pharmaceuticals during biological wastewater treatment. Falås et al. (2012) showed the enhancement of biological transformation kinetics by MBBR biofilm under aerobic conditions. To date, scarce results are available on the fate of pharmaceutical in denitrifying MBBR. In denitrifying activated sludge, biotransformation rates for pharmaceuticals were found either as significantly lower or comparable to rates obtained under aerobic conditions (Suarez et al., 2010; Plósz et al., 2010). Staging of activated sludge (Plósz, 2007) and biofilm reactors (Plósz and Vogelsang, 2012) induced specialization of heterotrophic biomass due to reaction kinetic principles and to the gradient in carbon source availability, thereby enhancing pre-denitrification. The objectives of this study were (i) to assess and compare pharmaceutical removal kinetics in single- and three-stage denitrifying MBBR configurations; and (ii) to evaluate the kinetic model developed by comparing predicted and measured concentrations in continuous-flow MBBR systems.

Materials and Methods. Two MBBR configurations, with single-stage (U) and three-stage (S1, S2, S3) bioreactors (total operating volume=6 L each, HRT=8.9 h), were operated in parallel, receiving pre-clarified wastewater with only indigenous pharmaceutical concentrations and external nitrate dosing (influent=103 mgN L^{-1}). After 100 d of continuous operation, 24-h batch experiments were performed using pre-clarified wastewater (initial adjusted nitrate=100 mgN L^{-1}) for each MBBR. Samples were analyzed with HPLC-MS/MS (Escolà Casas et al., *submitted*). Analytical solutions, derived from the Activated Sludge Model for Xenobiotics (ASM-X, Plósz et al., 2013), were used to estimate biotransformation (k_{Bio}) and retransformation rate coefficients (k_{Dec}, referring to the retransformation from, e.g., conjugated metabolites to parent chemicals). Values of k_{Bio} were corrected using literature K_d (Plósz et al., 2010; Göbel et al., 2005) to account for sorption. Parameters were estimated by minimizing mean average error (MAE) between measured and simulated batch concentrations. Estimated k_{Bio} and k_{Dec} were then used to predict concentrations in MBBRs during continuous operation, which were compared to measured concentrations. Results for trimethoprim (TMP) and sulfamethoxazole (SMX) have been selected and are presented here.

Results and Conclusions. TMP was effectively biotransformed during batch experiments (Fig. 1a; k_{Bio}=0.39–1.03 L $gTSS^{-1}$ d^{-1}). Previous investigations on denitrifying activated sludge did not reach conclusive results, and either significantly lower (Suarez et al., 2010) or comparable (Su et al., 2015) k_{Bio} values were reported for TMP. Concentration of SMX (Fig. 1b) increased in the first part of the batch experiment, indicating comparably fast retransformation from conjugated metabolites. Estimated k_{Dec} values (2.24–5.78 L $gTSS^{-1}$ d^{-1}) were consistently higher than k_{Bio} (0.64–1.90 L

$gTSS^{-1}$ d^{-1}), in agreement with findings for denitrifying activated sludge (Plósz et al., 2010). The lowest k_{Bio} and k_{Dec} values were obtained for S3, in which denitrifying biofilm was exposed to limiting electron donor availability during continuous operation. Predicted concentrations, based on batch estimations of k_{Bio} and k_{Dec}, were in good agreement with the measurements during continuous operation for both TMP (Fig. 1c) and SMX (Fig. 1d).

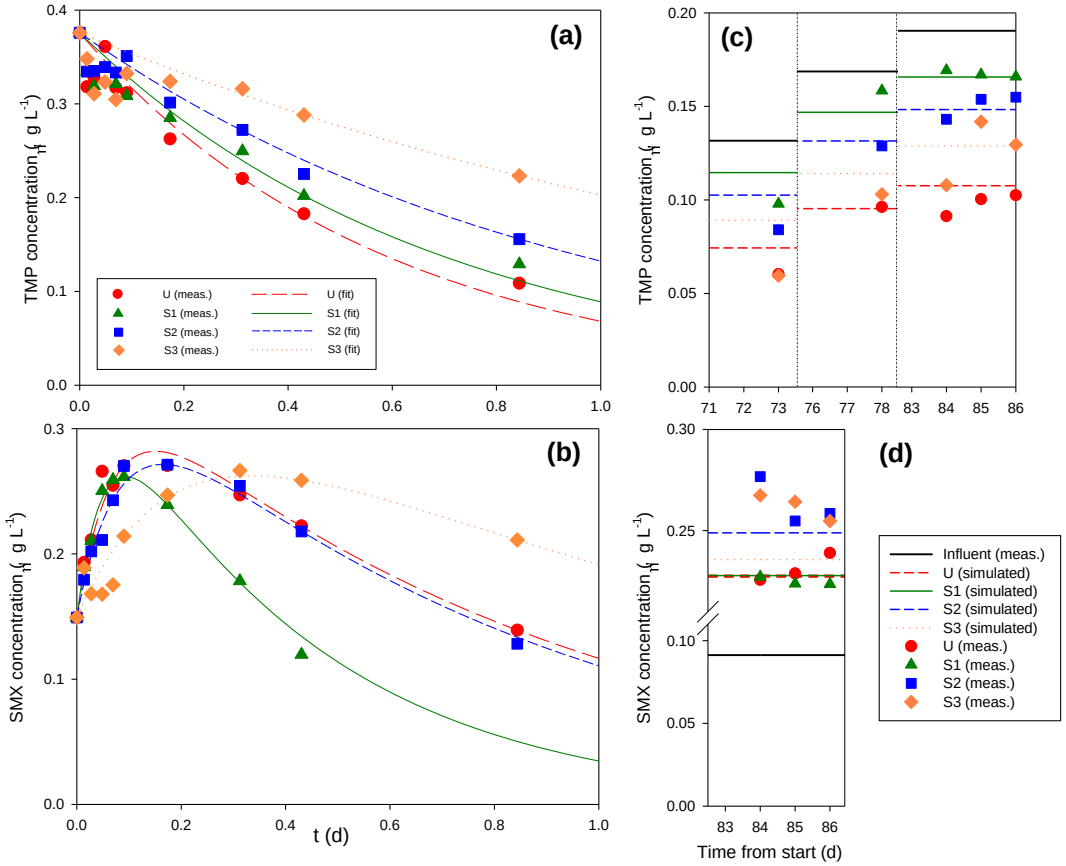

Figure 1. Measured (dots) and predicted concentrations (lines) during batch experiments and during continuous-flow operation of the reactors in the single-stage (U) and three-stage (S1, S2, S3) MBBR configuration for TMP (a, c) and SMX (b, d).

Our experimental and model-based observations suggest that: (i) TMP can be rather effectively removed in denitrifying MBBRs; (ii) retransformation to parent SMX can significantly impact its elimination; and (iii) electron donor availability in single- and three-stage MBBRs can shape microbial community functions in terms of secondary/co-metabolic processes.

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