Combined use of Anaerobic and Aerobic Moving Bed Biofilm Reactors for Micropollutants' Removal from Wastewater

Kora E.[1], Theodorelou D.[1], Herpich A.[2], Gatidou G.[1], Fountoulakis M.S.[1], Stasinakis A.[1,*],

1Department of Environment, University of the Aegean, 81100 Mytilene, Greece

2 Hochschule für Technik, Wirtschaft und Kultur Leipzig Postfach 30 11 66 04251 Leipzig, Germany

*corresponding author: e-mail: Stasinakis A., astas@env.aegean.gr

Abstract
A novel laboratory-scale continuous flow wastewater treatment system, consisting of an anaerobic moving bed biofilm reactor (AnMBBR) and an aerobic MBBR, was used for investigating the removal of hydroxybenzothiazole (OH-BTH) and selected benzotriazoles from municipal wastewater. The system was operated under three different Experimental Phases where different organic loadings were applied: 0.82 kg COD m^{-3} d^{-1} (Phase A), 0.2 kg COD m^{-3} d^{-1} (Phase B) and 2.1 kg COD m^{-3} d^{-1} (Phase C). The system achieved sufficient COD removal, nitrification and biogas production. All target micropollutants were partially removed during the experiments. Total removal efficiencies ranged from 32% for 5-methyl-1H-benzotriazole (5TTR) to 97% for OH-BTH. The contribution of the strictly anaerobic bioreactor was important for 5TTR, 5-chlorobenzotriazole (CBTR) and xylytriazole (XTR), while the use of aerobic bioreactors resulted to important increase of target compounds removal and it was exclusively responsible for the removal of OH-BTH and benzotriazole (BTR).

Keywords: MBBR; Anaerobic reactor; Benzothiazole; Benzotriazole

1. Introduction

Moving bed biofilm reactors (MBBR) technology has been extensively used for the removal of major pollutants from municipal and industrial wastewater (Odegaard, 2006; Vyrides et al., 2018). Recent studies have also shown the potential of such systems to remove organic micropollutants (Mazioti et al., 2015a; Polesel et al., 2017; Tang et al., 2019). Most of these studies have been conducted under aerobic conditions, while no information is available for the ability of a combined strictly anaerobic-aerobic MBBR system to remove organic micropollutants from domestic wastewater as well as for the contribution of different bioreactors on their elimination.

The main objective of the current study was to evaluate the ability of an anaerobic-aerobic MBBR system to remove organic micropollutants from municipal wastewater. As target compounds, some benzotriazoles and one benzothiazole were selected. These compounds are extensively used in industries as anticorrosion agents (Kloepfer et al., 2005; Salas et al., 2016; Li et al., 2017), as intermediate products (Voutsa, 2006; Salas et al., 2016) and as pigments (Li et al., 2017). They are frequently detected in municipal wastewater due to their high water solubility and biodegradation resistance (Voutsa, 2006; Kloepfer et al., 2005).

2. Methods

2.1. *Experimental Set up and Operation*

Two moving bed biofilm reactors were initially connected in series. The first reactor was an anaerobic MBBR (AnMBBR) and the second was an aerobic MBBR (AeMBBR1) (Table 1). Between the two reactors, a flask chamber was installed for the collection of the biogas produced by the AnMBBR (Figure 1). The System was fed with municipal wastewater from University Campus for seven months. At the last part of the study (Phase C), an additional aerobic MBBR was added to investigate further target compounds' removal (Table 1, Figure 1).

Table 1. Lab-scale continuous flow characteristics

Reactor	Volume (L)	HRT (h)	Filling Ratio (v/v)	Temp. (˚C)
AnMBBR	3.0	10	40%	32-34
AeMBBR1	3.8	13	33%	24-27
AeMBBR2	3.8	13	33%	22

The operation of the system was divided into three Phases where different organic loading rates were applied (Phase

A: 0.82 ± 0.40 kg COD m^{-3} d^{-1}, Phase B: 0.2 ± 0.1 kg COD m^{-3} d^{-1} and Phase C: 2.1 ± 0.2 kg COD m^{-3} d^{-1}). Samples were periodically taken to determine COD and NH_4/N concentrations, while the produced biogas was also monitored in a weekly basis. At the last part of each Phase, an aliquot of the target compounds was added at

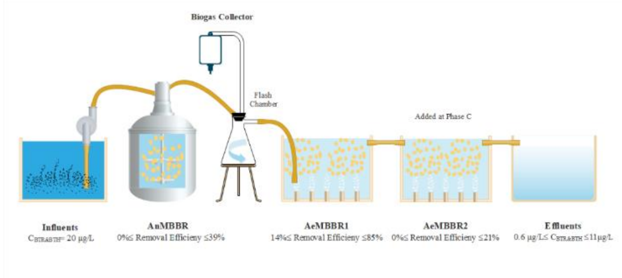

Figure 1. Schematic diagram of the lab-scale system.

3. Results

3.1. Major pollutants removal and biogas production

The system removed sufficiently COD (>84%) and NH_4-N (>96%), while the daily produced biogas ranged up to 172.7 mL L $reactor^{-1}$ d^{-1} (end of Phase C). Both AnMBBR and AeMBBR1 contributed equally to COD removal, while all ammonium nitrogen was removed via nitrification in aerobic MBBRs.

3.2. Target micropollutants removal

All target micropollutants were partially removed during the experiments. Among target compounds, the lowest total removal efficiencies were observed for 5TTR (32%), while the highest for OH-BTH (97%). The higher removal efficiency was observed during Phase C for the three out of five compounds (5TTR, CBTR and XTR), while OH-BTH was removed more in Phase B (Table 2).

Table 2. Micropollutants' removal efficiency (%) in each Experimental Phase.

	Target compounds removal efficiency (%)				
Exp. Phase	**BTR**	**5TTR**	**CBTR**	**XTR**	**OH-BTH**
A	50 (±10)	53 (±5)	41 (±13)	51 (±8)	80 (±9)
B	33 (±8)	32 (±10)	37 (±10)	43 (±14)	93 (±2)
C	60 (±6)	86 (±14)	56 (±17)	91 (±2)	97 (±1)

Concerning the role of different reactors on micropollutants' removal, the AnMBBR contributed signficantly to the removal of 5TTR, CBTR and XTR, whereas AeMBBR1 removed exclusively OH-BTH and BTR. The addition of AeMBBR2 in Phase C enhanced OH-BTH removal.

the influents for 7 days in order to achieve initial concentration of approximately 20 μg L^{-1} for each compound. The concentrations of target compounds were measured in the dissolved phase according to Mazioti et al. (2015b).

References

Kloepfer, A., Jekel, M., & Reemtsma, T. (2005). Occurrence, sources, and fate of benzothiazoles in municipal wastewater treatment plants. *Environmental Science and Technology*, **39**, 3792–3798.

Li, J., Zhao, H., Zhou, Y., Xu, S., & Cai, Z. (2017). Determination of benzotriazoles and benzothiazoles in human urine by. *Journal of Chromatography B*, **1070**, 70–75.

Mazioti A.A., Stasinakis A.S., Pantazi Y., Andersen H.R. (2015a) Biodegradation of benzotriazoles and hydroxy-benzothiazole in wastewater by activated sludge and moving bed biofilm reactor systems. *Bioresource Technology* **192**, 627-635

Mazioti, A. A., Stasinakis, A. S., Gatidou, G., Thomaidis, N. S., & Andersen, H. R. (2015b). Sorption and biodegradation of selected benzotriazoles and hydroxybenzothiazole in activated sludge and estimation of their fate during wastewater treatment. *Chemosphere*, **131**, 117–123.

Odegaard H. (2006). Innovations in wastewater treatment: the moving bed biofilm process. *Water Science and Technology* **53**, 17-33.

Polesel F., Torresi E., Loreggian L., Casas M.E., Christensson M., Bester K., Plósz B.G. (2017) Removal of pharmaceuticals in pre-denitrifying MBBR – Influence of organic substrate availability in single- and three-stage configurations, *Water Research* **123**, 408-419.

Salas, D., Borrull, F., Marcé, R. M., & Fontanals, N. (2016). Study of the retention of benzotriazoles, benzothiazoles and benzenesulfonamides in mixed-mode solid-phase extraction in environmental samples. *Journal of Chromatography A*, **1444**, 21–31.

Tang K., Spiliotopoulou A., Chhletri R.K., Ooi G.T.H., Kaarsholm K.M.S., Sundmark K., Florian B., Kragelund C., Bester K., Andersen H.R. (2019) Removal of pharmaceuticals, toxicity and natural fluorescence through the ozonation of biologically-treated hospital wastewater, with further polishing via a suspended biofilm, *Chemical Engineering Journal* **359**, 321-330.

Voutsa, D., Hartmann, P., Schaffner, C., & Giger, W. (2006). Benzotriazoles, alkylphenols and bisphenol A in municipal wastewaters and in the Glatt River, Switzerland. *Environmental Science and Pollution Research*, **13**, 333–341.

Vyrides I., Drakou E.M., Ioannou S., Michael F., Gatidou G., Stasinakis A. (2018) Biodegradation of bilge water: batch test under anaerobic and aerobic conditions and performance of three pilot Aerobic Moving Bed Biofilm Reactors (MBBRs) at different filling fractions, *Journal of Environmental Management* **217**, 356-362.