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Tracing the limits of organic micropollutant removal in biological wastewater treatment

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ABSTRACT

Removal of organic micropollutants was investigated in 15 diverse biological reactors through short and long-term experiments. Short-term batch experiments were performed with activated sludge from three parallel sequencing batch reactors (25, 40, and 80 d solid retention time, SRT) fed with synthetic wastewater without micropollutants for one year. Despite the minimal micropollutant exposure, the synthetic wastewater sludges were able to degrade several micropollutants present in municipal wastewater. The degradation occurred immediately after spiking (1–5 μg/L), showed no strong or systematic correlation to the sludge age, and proceeded at rates comparable to those of municipal wastewater sludges. Thus, the results from the batch experiments indicate that degradation of organic micropollutants in biological wastewater treatment is quite insensitive to SRT increases from 25 to 80 days, and not necessarily induced by exposure to micropollutants. Long-term experiments with municipal wastewater were performed to assess the potential for extended biological micropollutant removal under different redox conditions and substrate concentrations (carbon and nitrogen). A total of 31 organic micropollutants were monitored through influent-effluent sampling of twelve municipal wastewater reactors. In accordance with the results from the sludges grown on synthetic wastewater, several compounds such as bezafibrate, atenolol and acyclovir were significantly removed in the activated sludge processes fed with municipal wastewater. Complementary removal of two compounds, diuron and diclofenac, was achieved in an oxic biofilm treatment. A few aerobically persistent micropollutants such as venlafaxine, diatrizoate and tramadol were removed under anaerobic conditions, but a large number of micropollutants persisted in all biological treatments. Collectively, these results indicate that certain improvements in biological micropollutant removal can be achieved by combining different aerobic and anaerobic treatments, but that these improvements are restricted to a limited number of compounds.

1. Introduction

Discharge of organic micropollutants via treated wastewater is well documented (Dickenson et al., 2011; Loos et al., 2013), and may induce adverse environmental effects (Jobling et al., 1998; Brodin et al., 2013). Removal of these compounds in wastewater treatment plants (WWTPs) has been extensively investigated (Miège et al., 2009; Luo et al., 2014), and the biological treatment has often been identified as critical to the degree of micropollutant removal (Carballa et al., 2004; Zorita et al., 2009).

Several aspects of biological wastewater treatment have been discussed as relevant for micropollutant removal, including solid retention time (SRT) (Clara et al., 2005; Maeng et al., 2013; Petrie et al., 2014), hydraulic retention time (HRT) (Gros et al., 2010), nitrification (Tran et al., 2009; Helbling et al., 2012; Sathyamoorthy et al., 2013), heterotrophic activity (Majewsky et al., 2010), redox conditions (Suarez et al., 2010; Xue et al., 2010), pH (Gulde et al., 2014) and suspended/attached growth (Zupanc et al., 2013; Falås et al., 2013). A general consensus on the main drivers for the biological micropollutant removal at WWTPs is, however, lacking. This can be due either to some critical parameters being unknown or to that single parameters are unlikely to explain degradation of structurally different micropollutants in mixed microbial communities, such as biological wastewater systems.

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Organic micropollutants can generally be divided into easily, moderately, sporadically, and poorly degradable compounds in conventional biological wastewater treatment systems. These groups of micropollutants should most likely be targeted in different ways to reach or maintain low residual concentrations in biologically treated wastewater. Identification of positive and negative removal effects at typical treatment conditions could secure low discharge of easily degradable compounds and aid the development of new treatment strategies for moderately and sporadically degradable compounds. A profound change of biological treatment may, however, be required if the poorly degradable compounds in existing wastewater treatment systems are to be removed biologically.

Several organic micropollutants present in treated wastewater appear to be degraded in soil aquifer treatments (Amy and Drewes, 2007; Ternes et al., 2007), which suggests that further optimization of biological micropollutant removal at WWTPs is possible. These post-treatments are usually characterized by long HRT, low substrate availability and decreasing redox along the flow-path. Certain microbial reactions are most readily expressed at low redox and are sensitive to changes in electron acceptor composition. Reductive dehalogenation, for example, occurs mainly at low redox and is frequently involved in the anaerobic transformation of halogenated compounds (Mohn and Tiedje, 1992; Bhatt et al., 2007). Whether a combination of different redox conditions can enhance micropollutant removal at WWTPs has, however, not been fully elucidated.

The primary growth substrates in biological treatment systems can suppress micropollutant transformation rates (Su et al., 2015) and act as microbial selectors (Li et al., 2014). Changes in the composition of the biodegradable carbon in a synthetic feed solution have been reported to affect the microbial composition and the micropollutant degradation capacity of laboratory-scale soil columns over the long-term (Alidina et al., 2014; Li et al., 2014). However, the transformation of the six micropollutants investigated in these soil columns did not respond uniformly to the change in feed solution and the subsequent shift in the microbial community structure (Alidina et al., 2014). In short-term experiments, it has also been reported that transformation of different compounds respond differently to the presence of readily degradable carbon (Tan et al., 2013; Su et al., 2015). Moreover, it has been noted that associations between nitrification and transformation of specific compounds such as diclofenac and trimethoprim are difficult to reproduce in nitrifying systems treating similar synthetic wastewater with high ammonia and low organic carbon contents (Suarez et al., 2010; Fernandez-Fontaina et al., 2012). Thus, to reach a more comprehensive understanding of organic micropollutant removal in biological wastewater treatment, there is a need to expand single-system studies focusing on few micropollutants in one process to multi-system studies covering a wide array of organic micropollutants in different biological treatment processes.

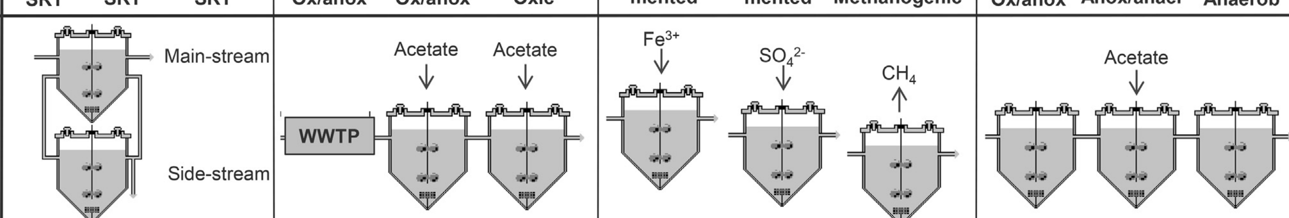
Therefore, the objective of this study was to investigate the removal of ≥20 organic micropollutants in a diverse set of isolated and combined aerobic and anaerobic process schemes (Table 1), and thereby assess the limits and potential of organic micropollutant removal in biological municipal wastewater treatment. The study comprised 15 biological reactors with a total operation time of >10 years, which allowed a direct comparison of treatment schemes and reproducibility.

2. Materials and methods

2.1. Reactor setups and sampling procedures

The reactor setups and sampling procedures applied in this study are presented in Table 1 and detailed in Section 2.1.1–2.1.6.

Table 1
Reactor setups and sampling procedures.

	Activated sludge (AS) *Synthetic wastewater* (*Number of reactors 3×2*)	Activated sludge (AS) with oxic post-treatment (*Number of reactors 3*)	Anaerobic stand-alone reactors (*Number of reactors 6, two different HRTs*)	Activated sludge (AS) with anaerobic post-treatment (*Number of reactors 3*)
	25 d SRT; 40 d SRT; 80 d SRT	AS Ox/anox; Post-tr. 1 Ox/anox; Post-tr. 2 Oxic	Iron supple-mented; Sulfate supple-mented; Methanogenic	AS Ox/anox; Post-tr. 1 Anox/anaer; Post-tr. 2 Anaerob
Configurations				
Retention times	**HRT:** Main-stream 15 h, Side-stream 12-20 d **SRT:** 25 d, 40 d, 80 d	**HRT:** 1 d; 1 d; 1 d **SRT:** ~25 d; Biofilm; Biofilm	**HRT:** 1 d or 12 d **SRT:** Biofilm (Short HRT), Hybrid biofilm (Long HRT)	**HRT:** 12 h; 7 d; 7 d **SRT:** 10 d; Biofilm; Biofilm
Sampling	**How:** Separate batch experiments **When:** 12 & 15 months after start-up	**How:** Flow proportional sampling over 3 weeks **When:** 6 months after startup	**Short HRT:** As for activated sludge with oxic post-tr. **Long HRT:** As for activated sludge with anaerobic post-tr.	**How:** Continues composite and grab sampling over 6 months **When:** 3 months after startup
Feed	**Synthetic wastewater**	**Municipal wastewater** Origin: Dübendorf	**Municipal wastewater** Origin: Dübendorf (Short HRT) Origin: Koblenz (Long HRT)	**Municipal wastewater** Origin: Koblenz

AS – Activated sludge; HRT – Hydraulic retention time; SRT– Solid retention time; Post-tr. – Post-treatment; WWTP – Full scale wastewater treatment plant.