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CHARACTERISING THE REMOVAL OF TRACE ORGANIC CHEMICALS IN WASTEWATER – ARE WE USING THE RIGHT TOOLS?

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Summary: Hypothesis tests posed on trace organics fate and removal in wastewater are often answered using approaches that can introduce significant bias in observations made on the system. Using non-representative sampling approaches in sewer and wastewater treatment plant studies is an example (Ort et al., 2010). This study provides a critical discussion of processes and methodologies used in laboratory- and full-scale wastewater experimental studies, and offers potential solutions to observed pitfalls with the support of a model-based assessment.

Keywords: xenobiotic trace chemical removal; retransformation; environmental representativeness

Introduction. In a comprehensive literature review (Polesel et al., *in preparation*) we critically discuss some of the factors significantly influencing observations made on xenobiotic trace chemicals (XTCs) fate in laboratory- and full-scale wastewater systems. The aim of this study is to discuss these factors and related pitfalls, specifically: (i) The underestimation of biotransformation rate coefficients (k_{Bio}) by monitoring influent and effluent of a CSTR or a CSTR series, thereby ignoring retransformation processes that can mask XTC removal by producing parent chemicals from e.g., human metabolites (Plósz et al., 2010, 2012)—a process often ignored also in batch experiments; (ii) The over- or underestimation of k_{Bio} from data inferred from experiments run in the absence of growth substrates, known to competitively inhibit or enhance XTC biotransformation; (iii) The over- or underestimation of representative k_{Bio} in batch tests at XTC concentration levels significantly higher than in the environment (Alexander, 1985; Collado et al., 2012); (iv) The underestimation of k_{Bio} by using synthetic growth medium in long-term experimental studies (Berg and Nyholm, 1996), thereby minimizing the possible induction of gene expression—responsible for an effective heterotrophic biotransformation—by indigenous chemicals structurally similar to XTC analytes; (v) The overestimation of sorption coefficients (K_D) by using XTC concentration in solids, additionally containing the irreversibly sequestered fraction (Wu et al., 2009; Plósz et al., 2012)—possibly propagating to k_{Bio} estimates; (vi) The over- or underestimation of community illicit drug use (sewage epidemiology) by ignoring in-sewer transformation of parent drugs and metabolites (Plósz et al., 2013; Ramin et al., 2014).

Results and Discussion: The case of Retransformation. As noted above, retransformation to parent XTCs can have a significant impact on their fate in wastewater systems—possibly not appreciated by experimental observations. A number of retransformation processes, overall contributing to the actual or apparent formation of XTCs in the aqueous phase, have been identified. **(i) From human metabolites.** Retransformation from human metabolites is of high relevance for selected XTCs, e.g., pharmaceuticals. Almost one quarter of excreted pharmaceutical metabolites are conjugated forms of the parent compound (Testa et al., 2012) and can undergo retransformation to the parent form through cleavage of the conjugated moiety. Deconjugation has been observed in biological wastewater treatment (Plósz et al., 2010, 2012) and in upstream sewers (Jelic et al., 2015). The impact of these processes on pharmaceutical removal can be quantified using operational diagrams presented by Plósz et al. (2012) based on the Activated Sludge Model for Xenobiotics (ASM-X, Plósz et al., 2010, 2012). Fig. 1 presents an application of this methodology for sulfamethoxazole (SMX) and its conjugate, N4-acetyl-SMX. Good agreement between predicted and literature removal efficiency (only parent SMX— η_{LI} —and parent plus conjugated

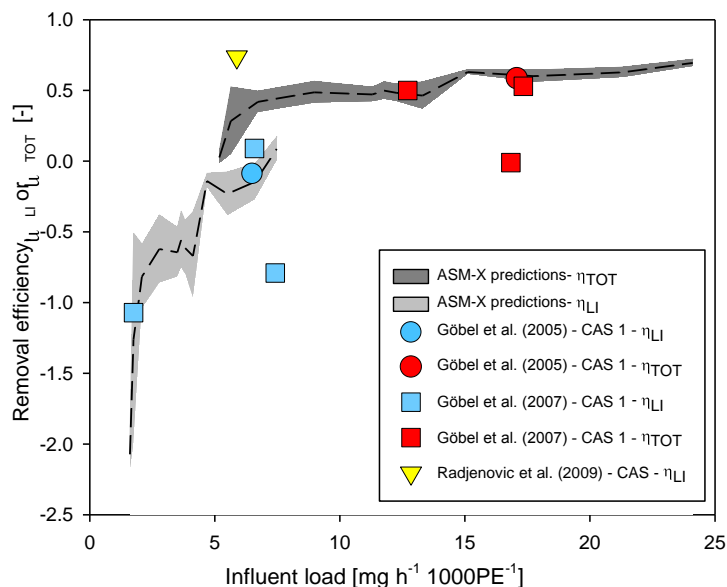


Figure 1. Comparison of predicted and full-scale measured parent-based (η_{LI}) and total removal efficiencies (η_{TOT}) for SMX.

Colloidal matter is subject to extracellular hydrolysis (Larsen and Harremoës, 1994), which may lead to the release of sorbed chemicals to the aqueous phase. Concentration increase of sorptive pharmaceuticals (fluoroquinolones, tetracyclines; Lindberg et al., 2006; Plósz et al., 2010) was accordingly registered during primary settling and activated sludge treatment. Similar findings for macrolides (Göbel et al., 2007) were attributed to the aqueous release of amounts excreted in faeces.

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SMX— η_{TOT}) shows that retransformation mostly occurred during wastewater treatment. Instead, underestimation of measured elimination indicates that significant retransformation may have occurred in upstream sewers (due to, e.g., high in-sewer residence time).

(ii) From commercial chemicals. XTCs can be formed from other commercial chemicals (excreted unchanged) via biotic transformation. For instance, fungal transformation of the antibiotic enrofloxacin results in the formation of ciprofloxacin (Wetzstein et al., 2006).

(iii) From particulate matter hydrolysis. In wastewater, XTCs can undergo sorption onto particulate and colloidal matter (Barret et al., 2010).