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Impacts and fate of triclosan and sulfamethoxazole in intensified re-circulating vertical flow constructed wetlands

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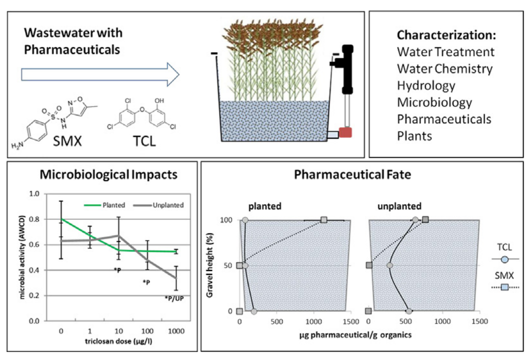
HIGHLIGHTS

- Triclosan and sulfamethoxazole quickly removed from water.
- Ex-situ assay showed pharmaceuticals to significantly impact microbial function.
- No major impacts to microbial communities during in-situ pharmaceutical exposures
- Constructed wetlands continued to treat wastewater during pharmaceutical exposures.
- Triclosan and sulfamethoxazole were found to remain in the gravel-associated biofilm.

GRAPHICAL ABSTRACT



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ABSTRACT

The impacts to microbial function, overall performance and eventual fate were assessed for triclosan (TCL) and sulfamethoxazole (SMX) in intensified (re-circulating) vertical subsurface flow (VSSF) constructed wetlands (CWs). The potential toxicity of each pharmaceutical to the intrinsic microbial communities was first assessed over a wide exposure range (0–1000 μg/l) via an ex-situ dose-response assay to estimate the concentration at which adverse effects were likely to occur. Based on these results an acute (7 day) in-situ exposures (500 μg/l) were then performed and impacts to the mesocosm systems monitored for 1 month via community-level physiological profiling (CLPP) alongside chemical oxygen demand (COD) removal rates and a range of water quality, and hydrological parameters. Despite the clear potential for negative impacts to microbial function from both compounds observed at 100 μg/l in the ex-situ dose-response test, no impacts were observed for the 500 μg/l in-situ exposure in the VSSF mesocosms. COD removal, water chemistry, plant health, and hydrological parameters did not significantly change in response to the in-situ exposure. In terms of fate, the removal efficiency for both TCL and SMX was high (>80%) after 1 h and complete removal (>99.7%) was observed after 168 h. Following the in-situ exposure, and subsequent one month effects-monitoring period, the mesocosms were decommissioned with the media biofilm spatially assessed for organic content as well as TCL and SMX concentrations. TCL and SMX were found to have persisted in the media and demonstrated spatial variation with an overall 2–20% and 5–6% recovered respectively. This suggests that biofilm bound TCL and SMX were biologically degraded in VSSF CWs, however may also accumulate in the biofilm if TCL and SMX are maintained in the influent. These results reinforce the robustness and potential of constructed wetlands for the treatment of pharmaceutical and personal care product (PPCP) contaminated wastewater.

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1. Introduction

The increasing presence and variety of pharmaceutical and personal care products (PPCPs) now detectable in the aquatic environment is an issue of widespread concern. The term PPCPs covers a broad class of unregulated products, including antibiotics and antimicrobials used by individuals for personal health or cosmetic reasons or used by agribusiness to enhance livestock productivity (EPA, 2012; Reyes-Contreras et al. 2011). Antibiotics and antimicrobials are, in broad terms, used to kill or prevent further growth of microbial populations. Antibiotic consumption worldwide was estimated to have increased by 36% between 2000 and 2010 (Van Boeckel et al. 2014). Following human ingestion or use, PPCPs are excreted or washed into the sewer system and transported to municipal waste water treatment plants (WWTPs). The fact that most PPCPs are designed specifically to have an antibiotic, antimicrobial and/or antibacterial effect poses risk to the biota of the receiving ecosystem. Whilst some PPCPs demonstrate acute toxicity in the aquatic environment others may lead to subtle yet significant effects across numerous species (Daughton and Ternes 1999). Other serious concerns over the fate of these compounds in the environment include the development of antibiotic resistance and endocrine disruption (Boxall et al. 2012; Gee et al. 2008; Kümmerer 2009; Raut and Angus 2010; Helt et al. 2012).

The two compounds selected for use in this study are ubiquitous and effective antimicrobials with triclosan (TCL) being a common antibacterial and sulfamethoxazole (SMX) being a widespread antibiotic. The presence of both of these compounds in the aquatic environment has been clearly demonstrated throughout the world (Perez et al. 2013; Straub 2016) and the need for cost effective and environmentally conscious removal technologies has led to the investigation of constructed wetlands for this purpose (Ávila and García 2015; Matamoros and Bayona 2013). Intrinsic microbial communities are essential to the ongoing health and function of constructed wetlands and as such exposure to antimicrobial compounds within the wastewater to be treated is undesirable as it could theoretically lead to reduced wetland functionality. Triclosan ($C_{12}H_7Cl_3O_2$; TCL), is a broad spectrum synthetic antimicrobial agent that is widely used in personal care products including deodorants, cleansers and hand sanitizers (Singer et al. 2002). The use of TCL has increased over the last 30 years due to its highly effective antimicrobial properties and ease of processing in both solids and liquids. Sulfamethoxazole ($C_{10}H_{11}N_3O_3S$; SMX), is a common sulfonamide bacteriostatic antibiotic used primarily to treat bacterial infections in human patients (Collado et al. 2013). Following oral ingestion, it is only partially metabolized in the human body with 45–70% of the administered dose reportedly excreted via urine within 24 h (Collado et al. 2013; Cribb and Spielberg 1992). Due to this excretion, small communities undergoing collective medical treatment for a bacterial infection can be a significant potential source of SMX to the environment. The main pathways for TCL and SMX to the environment are from municipal and hospital wastes (Berglund et al. 2014), both compounds have been frequently found in wastewater and drinking water (Collado et al. 2013; Singer et al. 2002) and many studies have demonstrated that wastewater treatment plants do not fully remove these compounds due to their low biodegradability (Ricart et al. 2010; Ternes et al. 2004). Treatment of grey water for individual buildings has become more popularized with many non-conventional technologies available however the treatment of PPCPs continue to be a challenge (De Gisi et al. 2016).

Constructed wetlands (CWs) have gained popularity due to their relative ease of operation and maintenance, sustainability, low energy consumption, and ability to operate as a functional habitat for wildlife (Ávila et al. 2014). They are now amongst the most widely used means of treating urban wastewater from individual homes and small communities (Matamoros and Bayona 2013) and are increasingly used to treat wastewater contaminated with a variety of PPCPs (Ávila and García 2015; Matamoros et al. 2012; Zhao et al. 2015). Mechanisms involved in the removal of PPCPs from CWs may include microbiological degradation, plant uptake and metabolization (Carvalho et al. 2014), adsorption to biofilm or substrate, volatilization, photocatalzyed oxidation, and other advanced reduction/oxidation reactions based on novel substrates or intensification schemes. Reported removal efficiencies vary depending on the contaminant and the configuration of the constructed wetland (Li et al. 2014) but overall constructed wetlands appear to offer an effective option with removal rates for many PPCPs similar or higher than those reported in conventional WWTPs (Li et al. 2014). For example, as reviewed by Wang and Wang (2016), some specific WWTP examples are the findings of Blair et al. (2015) who showed −35% removal of SMX, and 55.5% removal of TCL in a conventional activated sludge plant, and Roberts et al. (2016) who documented >99% removal of TCL also from a conventional activated sludge process. Reported removal rates for various CWs designs vary. Mean removal efficiencies for TCL are typically lower and more variable, in the range of 16–97% whereas SMX has been considered to be readily removed with efficiencies >70% (Li et al. 2014; Vymazal et al. 2017). However, these data are based on the results of surface flow (SF), horizontal sub-surface flow (HSSF) and hybrid systems. With respect to VSSF CWs less data is available however Nowrotek et al. (2015) observed removal efficiencies of 42–44% for SMX in lab scale VSSF mesocosms, and Zhao et al. (2015) previously found removal efficiencies of >90% for TCL in VSSF systems. No data has yet been generated for intensified VSSF systems (high oxygen content through recirculation, or aeration).

CWs are most commonly built for the treatment of secondary or tertiary domestic wastewater. A number of mechanisms are involved in the treatment of domestic wastewater in CWs, however microbial processes are seen to be the main driver for the removal of organics and the transformation/removal of nitrogen species (Weber 2016). Although CWs have shown great promise for the removal of PPCPs from water, the effects of PPCPs on the inherent microbial community, and subsequent overall operation of CWs, has only been directly studied three times. Weber et al. (2011) evaluated and observed the effects of ciprofloxacin on the development of vertical flow CW mesocosms. The microbial community was found to be initially affected by the 2 ppm ciprofloxacin exposure, however quickly recovered over a one-month period. Perhaps of more significance was the negative effect seen to the plants (*Phragmites australis*), and the developmental differences observed in dispersivity, porosity, and evapotranspiration between exposed and unexposed mesocosms. Nowrotek et al. (2015) observed no detrimental effects on ammonia oxidizing bacteria exposed to 6 ppm SMX and diclofenac in VF CWs treating synthetic domestic wastewater. Zhao et al. (2015) found surface flow CWs with different plant species to all show reduced bacterial abundance when exposed to 60 ppb TCL.

A recent review on PPCP removal at onsite wastewater treatment systems show measured maximum influent concentrations of TCL and SMX to be 80 μg/l and 26 μg/l respectively (Schaider et al., 2017). Although these measured concentrations are quite low, CWs treating wastewater from individual homes represents one of the most extreme scenarios for influent antibiotic concentrations at certain peak times. Influent SMX and TCL concentration data for CWs at individual homes has not yet been reported, therefore theoretical calculations are provided here. Prescribed SMX dosage for infections varies, however 800 mg every 12 h for adolescents and adults is common. If SMX is only 45% metabolized (Collado et al. 2013; Cribb and Spielberg 1992), for a home of four people all prescribed SMX for the same sickness over the same period, approximately 3.5 g of SMX could enter a single home CW (through wastewater) in a single day. In a reasonably water efficient four-person home, where perhaps only 500 l of water is used in a day, a daily-averaged SMX concentration of approximately 7 ppm could theoretically be expected to enter a single home CW. With respect to TCL, as much as 2% triclosan can be found in detergent/soap dispersions (Bhargava and Leonard, 1996). However, as shown by Jones et al. (2000), 1% dispersions are also found to be effective. Per Larson et al.