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Research article

Fate of triclosan in laboratory-scale activated sludge reactors - Effect of culture acclimation

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ABSTRACT

Triclosan (TCS); a widely used antimicrobial biocide, exists in several pharmaceutical and personal care products. Due to its wide usage, TCS is detected in wastewater at varying concentrations. Biological treatability of TCS and its effect on chemical oxygen demand (COD) removal efficiency were investigated running laboratory-scale pulse-fed sequencing batch reactors with acclimated and non-acclimated cultures. The culture was acclimatized to TCS by gradually increasing its concentration in the synthetic feed wastewater from 100 ng/L to 100 mg/L. There were no effects of TCS on COD removal efficiency up to the TCS concentration of 500 ng/L for both acclimatized and non-acclimatized cases. However, starting from a concentration of 1 mg/L, TCS affected the COD removal efficiency adversely. This effect was more pronounced with non-acclimatized culture. The decrease in the COD removal efficiency reached to 47% and 42% at the TCS concentration of 100 mg/L, under acclimation and non-acclimation conditions respectively. Adsorption of TCS into biomass was evidenced at higher TCS concentrations especially with non-acclimated cultures. 2,4-dichlorophenol and 2,4-dichloroanisole were identified as biodegradation by-products. The occurrence and distribution of these metabolites in the effluent and sludge matrices were found to be highly variable depending, especially, on the culture acclimation conditions.

1. Introduction

Triclosan (TCS) (5-chloro-2-[2,4-dichlorophenoxy]-phenol) is one of the most commonly used antimicrobial biocides in daily life and exists as an ingredient in several pharmaceutical and personal care products such as soaps, toothpaste, deodorants and detergents (Dann and Hontela, 2011).

The presence of TCS in broad spectrum of products has resulted in reach of this compound to wastewater treatment plants (WWTPs) and surface waters (Reiss et al., 2002). Therefore, the presence of TCS like biocides in environmental systems has become a prominent issue in recent years in parallel with the increased uses. TCS has been encountered in different environments at varying concentrations: 0.16–2300 ng/L in surface waters (Yavuz et al., 2015), 2.3–562 μg/L in the influent of WWTP's (Mezcua et al., 2004), 0.1–269 ng/L in the effluent of WWTP's (Mezcua et al., 2004), 24–434 ng/L in Austrian WWTPs' effluents (Ying and Kookana, 2007) and 28–37,189 μg/kg in sewage sludges (Kolpin et al., 2002).

TCS is known to have numerous adverse effects on aquatic and terrestrial organisms (Orvos et al., 2002; Wilson et al., 2003), bioaccumulate on the organisms due to its high hydrophobicity (Ying and Kookana, 2007; Samsoe-Petersen et al., 2003), act as an endocrine disrupting substance (Stoker et al., 2010), disrupt the microbial population of receiving soils (SCCS, 2011), cause bacteria to develop antibiotic resistance (Ying and Kookana, 2007; Fernandez-Fuentes et al., 2012; Walsh et al., 2003) and cause skin, eye and respiratory irritations, allergic diseases and inhalation toxicity on human beings (NICNAS, 2009) (Table S1, Supplementary Information).

Several studies were conducted on the removal of TCS in the WWTPs. Biological treatment is one of the alternatives and different removal efficiencies were reported depending on the process and operational conditions (Bester, 2003; Kanda et al., 2003; Mcavoy et al., 2002; Sabaliunas et al., 2003). Activated sludge process has the highest performance (i.e. 95–98%) for TCS removal compared to rotating biological contactors (58–96%) and trickling filters (86–97%) (Thompson et al., 2005). There are also supportive studies reporting significant TCS removals, around 95–98%, in activated sludge systems (Kanda et al., 2003; Mcavoy et al., 2002; Roberts et al., 2016; Wang and Wang, 2016). In a

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most recent study conducted by Petrie et al. (2014), on the other hand, lower TCS removal efficiencies have been reported (84% for activated sludge system; 74% for trickling filter). This variable nature is also indicated in a comprehensive review paper by Onesios et al. (2009) where a wide range (48%–98%) was reported for TCS removal efficiency in full scale WWTPs. While some studies indicate the adsorption as a significant mechanism for the substantial removal of TCS in activated sludge systems (Bester, 2003; Heidler and Halden, 2007; Thompson et al., 2005), some others determined the biological transformation as the dominant process (Federle et al., 2002). Thompson et al. (2005) stated that dissolved oxygen (DO) level is a determining factor for the removal mechanism in such a way that biodegradation becomes responsible at high DO levels whereas sorption becomes dominant when lower DO levels prevail. So, the picture is still not clear. Dhillon et al. (2015), in a comprehensive review paper, indicated that more efforts are needed for understanding the fate and distribution of TCS in various environmental compartments, in particular, WWTPs and sediments which are the final sinks.

The adaptation of microbial culture to inhibitory substances like TCS, may enhance the treatment performance and result in the reduction or complete elimination of the inhibitory effect (Sahinkaya and Dilek, 2007; Silvestre et al., 2011). None of the previous studies considered this issue, except the one by Stasinakis et al. (2007) who investigated the fate and toxicity of TCS in activated sludge systems. TCS effect on activated sludge process was found to be minor for acclimatized biomass. They observed deterioration in ammonia removal and nitrification capacity of the activated sludge system in case of non-acclimatized biomass exposed to 0.5 mg/L TCS. However, after acclimatization of biomass, nitrification was fully recovered and even 2 mg/L TCS did not affect nitrification. Although they considered the effect of acclimation, they did not investigate by-product formation in relation to acclimatization and also influent TCS concentration. In fact, several metabolites are formed during the biodegradation of TCS (Federle et al., 2002) and one of the well-known transformation products is the methyl triclosan which is more toxic than the parent (Bester, 2005; Chen et al., 2011; Lozano et al., 2013). Federle et al. (2002) indicated that TCS metabolites are very polar and less bioaccumulative than TCS, without identifying individually.

In order to fill in the gaps in literature, a comprehensive study was undertaken regarding the biological treatability of TCS bearing wastewaters using acclimated and non-acclimated microbial cultures. Specific objectives were to investigate the effects of culture adaptation on the fate and removal of TCS in activated sludge process. In accordance with this aim, a laboratory-scale activated sludge unit (pulse-fed sequencing batch reactor, SBR) receiving a synthetic wastewater was operated for acclimation and non-acclimation conditions and major mechanisms affecting its fate were clearly introduced. Possible biodegradation by-products of TCS were also designated.

2. Materials and methods

Prior to the reactor operations, synthetic wastewater and TCS stock solutions were prepared and seed microbial culture to be used in the experiments was taken. The analytical standards, methods and devices to be applied during the study were set accordingly. The pulse-fed SBRs were operated with TCS acclimated and non-acclimated microbial cultures the details of which are introduced in the following sub-sections as well. Samples taken from the reactors were then analyzed for their TCS, COD, mixed liquor suspended solids (MLSS), pH and sludge volume index (SVI) content in order to come up with a conclusion on the treatment performance of the activated sludge system.

2.1. Synthetic wastewater, source of microbial culture and sample preparation

The SBR was fed with a synthetic wastewater (Table 1) containing varying TCS concentrations (100 ng/L-100 mg/L), determined with the consideration of both TCS toxicity threshold values (EC_{50}: 0.7 μg/L for green algae; 390 μg/L for daphnia magna; LC_{50}: 0.26 mg/L for freshwater fish) reported in literature (Orvos et al., 2002; US EPA, 2008) and TCS concentrations (<0.00087–562 μg/L) likely to be encountered in wastewaters (BIOHYPO, 2010–2012; Mezcua et al., 2004). Synthetic wastewater was prepared according to the composition proposed by Dilek et al. (1998) in order to have a composition similar to municipal wastewater and study the treatment performance of the activated sludge system. In this way, the highly variable nature of the raw wastewaters making the laboratory investigations difficult is also avoided by using the same synthetic wastewater composition in all set of the experiments. The synthetic wastewater was spiked with stock TCS to obtain desired TCS levels. TCS concentration in the stock solution was measured in order to ensure the delivery of the desired concentration(s) to the test vessel(s). Moreover, TCS concentration in the reactors right after spiking the desired volumes of stock solution were measured as to report the initial feed concentrations. In all cases, there existed ±0.7% deviation in measured value from spiked theoretical TCS concentration.

Microbial culture used as a seed during the experiments was obtained from METU Technocity Bio-Membrane type WWTP located in Ankara, Turkey.

TCS stock solutions (i.e. 1 mg/L, 1000 mg/L and 10,000 mg/L) were prepared in ultra-pure water containing 0.04 M sodium hydroxide (NaOH) and stored in refrigerator at +4 °C in dark. Since microbial growth is truly affected from the carbon sources in the media, NaOH in which TCS is readily soluble (SCCS, 2011) was preferred as stock solution agent instead of methanol and acetone as also performed by Veldhoen et al. (2006).

All labware used in the studies were cleaned prior to experiments by means of washing with a detergent called Alconox, rinsing with hot tap water and ultra-pure water, and final rinsing with HPLC grade methanol. After the cleanup procedure, the labware were dried at 105 °C for 1 h before usage.

2.2. Pulse-fed sequencing batch reactor (SBR) operations

The SBR was fed with the synthetic wastewater containing varying TCS concentrations (100 ng/L-100 mg/L). Steady state condition was monitored through the daily COD and MLSS measurements after operation at least 3 sludge retention time (SRT) and the culture was considered to be in steady state when the last three measurements did not differ more than 5% as value. Following

Table 1
Composition of synthetic wastewater (Dilek et al., 1998).

Ingredient	Concentration (mg/L)
Proteous-peptone	470
NaCl	156.70
Na_2SO4	17.20
K_2HPO4	44.60
$MgCl_2 \cdot 6H_2O$	3.700
$FeCl_2 \cdot 4H_2O$	4.520
$CaCl_2$	2.794
$MnSO_4 \cdot H_2O$	0.0638
$ZnSO_4 \cdot 7H_2O$	0.0819
$CoCl_2 \cdot 6H_2O$	0.0753
$CuSO_4$	0.0760
$(NH_4)_6Mo_7O_{24} \cdot 4H_20$	0.0338

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