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Influence of thermal hydrolysis-anaerobic digestion treatment of wastewater solids on concentrations of triclosan, triclocarban, and their transformation products in biosolids

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HIGHLIGHTS

- Significant reduction of TCC concentrations occurred during thermal hydrolysis.
- TCS, MeTCS, and 2,4-DCP were concentrated due to anaerobic digestion.
- TCS, MeTCS, and 2,4-DCP were not impacted by thermal hydrolysis.

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ABSTRACT

The growing concern worldwide regarding the presence of emerging contaminants in biosolids calls for a better understanding of how different treatment technologies at water resource recovery facilities (WRRFs) can influence concentrations prior to biosolids land application. This study focuses on the influence of solids treatment via the Cambi Thermal Hydrolysis Process™ in conjunction with anaerobic digestion (TH-AD) on concentrations of triclosan (TCS), triclocarban (TCC), and their transformation products in biosolids and sludges. Concentrations of the target analytes in biosolids from the TH-AD process (Class A), sludges from the individual TH-AD treatment steps, and limed biosolids (Class B) from the same WRRF were compared. TCC concentrations were significantly lower in Class A biosolids than those in the Class B product - a removal that occurred during thermal hydrolysis. Concentrations of TCS, methyl triclosan, and 2,4-dichlorophenol, conversely, increased during anaerobic digestion, leading to significantly higher concentrations of these compounds in Class A biosolids when compared to Class B biosolids. Implementation of the TH-AD process had mixed effect on contaminant concentrations.

1. Introduction

Extensive use of pharmaceutical and personal care products (PPCPs) by society has led to their presence in the wastewater treatment (WWT) process, including wastewater effluent and biosolids (Lozano et al., 2013; Malmborg and Magnér, 2015). Two such compounds are the antimicrobials triclosan [5-chloro-2-(2,4-dichlorophenoxy)-phenol] (TCS) and triclocarban [*N*-(4-chlorophenyl)-*N*-(3,4-dichlorophenyl) urea] (TCC), both of which have been demonstrated to show endocrine disrupting capabilities (Ahn et al., 2008; J. Chen et al., 2008; Hinther et al., 2011; Zorrilla et al., 2008) and, due to concerns regarding their ecological impact, are currently under phase-out regulations in consumer antiseptic wash products in the United States (U.S.) (FDA, 2016). TCC and TCS have been detected in all stages of the WWT process and most notably concentrate in the solids fraction (Heidler et al., 2006; Lozano et al., 2013; Narumiya et al., 2013).

In the US, treated wastewater solids (biosolids) are commonly land-applied as a means of nutrient recovery/soil reclamation (Laturnus et al., 2006) allowing for this material to become a potential source of organic pollutants to the environment. TCC and TCS have been detected in biosolids from wastewater treatment facilities throughout the U.S. (Andrade et al., 2015; Pycke et al.,

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2014) and studies have shown the ability of TCC and TCS to persist in agricultural soils after the land-application of biosolids, with estimated half-lives of 191 and 107–258 d, respectively (F. Chen et al., 2014; Lozano et al., 2010). Furthermore, these antimicrobials can accumulate in the roots of plants grown in biosolids-amended soils (Pannu et al., 2012; Wu et al., 2013) and earthworms living in treated soils (Kinney et al., 2008; Macherius et al., 2014), indicating the potential for ecological risk.

TCC and TCS can also be partially transformed both biotically and abiotically during the WWT process. For instance, methyl triclosan (MeTCS) can form during aerobic treatment (Lozano et al., 2013), has higher endocrine disrupting capabilities (Hinther et al., 2011) than its parent compound TCS, and is more persistent in biosolids amended soils than TCS (Lozano et al., 2012). Furthermore, the carbanilide analogs of TCC associate primarily with solids within the WWT process (Pycke et al., 2014) and have also been linked to endocrine system disruption (Ahn et al., 2008). The amount of TCC and TCS degradation and the compounds formed is highly dependent on the treatment processes employed by the WWT plant (Pycke et al., 2014).

Currently, WWT facilities within the United States (US) are focusing efforts into the beneficial recovery of resources throughout the treatment process and many have begun changing their designation from WWT plant to water resource reclamation facility (WRRF) (WEF, 2014). This emphasis on resource recovery not only includes the land-application of biosolids for beneficial uses, but changes in treatment processes as well. One such process is the innovative Cambi Thermal Hydrolysis Process™ (CambiTHP™), a pretreatment for anaerobic digestion of wastewater sludge. Amongst other benefits, the CambiTHP™ allows for a reduction in the volume of final solids as well as increases the biodegrability of sludge – leading to an increase in biogas production during anaerobic digestion, which can be captured and beneficially used as an energy source. Limited studies have dealt with the fate of organic microconstituents in the CambiTHP™ process. Previous experiments on the fate of TCS, bisphenol-A, and nonylphenol ethoxylates in spiked water samples and nonlyphenol in spiked sludge treated via high temperature and pressure by a lab-scale chemical digestion bomb found that these compounds were not degraded during the treatment. Furthermore, the study found that laboratory-scale anaerobic digesters treating sludge with conventional mesophilic anaerobic digestion (MAD) more readily transformed nonylphenol ethoxylates to nonylphenol than sludge that had been pretreated with thermal hydrolysis prior to MAD (McNamara et al., 2012). The present study examines concentrations of TCC and TCS as well as three TCS and five TCC transformation products in biosolids from a single WRRF in the Mid Atlantic region of the US that has recently changed its solids handling process. In October 2014, the facility began transitioning from lime-stabilizing final solids (Class B biosolids) to treating solids via the CambiTHP™, in conjunction with anaerobic digestion (TH-AD) (Class A biosolids), the first of its kind in the US and currently the largest in the world. Class A biosolids have severely reduced pathogen levels as well as increased vector attraction reduction when compared to Class B biosolids, allowing for increased options for land application. The study goal was to expand upon a previous study of historical trends of TCC and TCS in limed biosolids from this WRRF (Andrade et al., 2015) by examining how advances in solids handling processes within this same facility can influence the concentrations of these antimicrobials as well as their degradation products (not examined in the previous study) in biosolids prior to their application onto agricultural soils. Additionally, the study examines the influence of individual stages of the TH-AD process itself on the concentrations of these antimicrobials and their transformation products.

2. Materials and methods

2.1. Target analytes

Class A and Class B biosolids, as well as sludge samples collected throughout the TH-AD process, were analyzed for the antimicrobials TCS and TCC. Additionally, all samples were analyzed for three TCS transformation products: MeTCS, 2,4-dichlorophenol (2,4-DCP), and triclosan-o-sulfate (TCS-O-Sulf); and five TCC degradation products: 4,4'-dichlorocarbanilide (DCC), 1-(3-chlorophenyl)-3-phenylurea (MCC), carbanilide (NCC), 4-chloroaniline (4-CA), and 3,4-dichloraniline (3,4-DCA). These compounds have been identified previously in the literature as TCS and TCC byproducts (X. Chen et al., 2015, 2011; Latch et al., 2004; Mulla et al., 2016; Pycke et al., 2014). Compound structures are provided in Fig. 1.

2.2. Wastewater treatment plant

The current study focuses on sludge and biosolids samples collected from an east coast U.S. municipal WRRF serving a highly

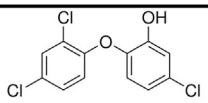
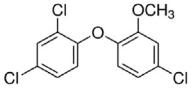
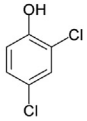
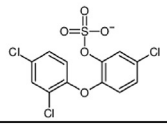
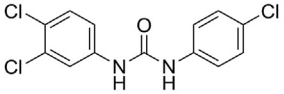
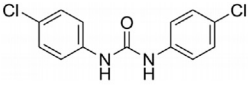
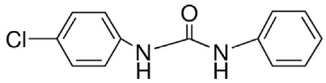
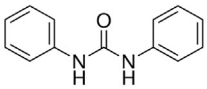
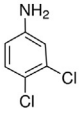
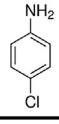
Compound	Structure
Triclosan	
Methyl Triclosan	
2,4-Dichlorophenol	
Triclosan-o-sulfate	
Triclocarban	
4,4'-Dichlorocarbanilide	
1-(3-Chlorophenyl)-3-phenylurea	
Carbanilide	
3,4-Dichloroaniline	
4-Chloroaniline	

Fig. 1. Structures of Triclosan, Triclocarban, and their Transformation Products.