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Fate and mass balance of triclosan and its degradation products: Comparison of three different types of wastewater treatments and aerobic/anaerobic sludge digestion

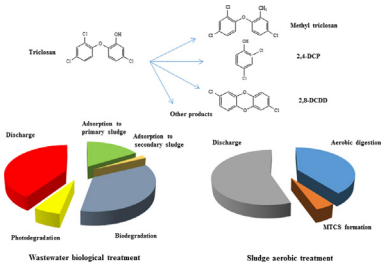
Fatemeh Tohidi [a], Zongwei Cai [a,b,*]

[a] State Key Laboratory of Environment and Biological Analysis, Department of Chemistry, Hong Kong Baptist University, Kowloon Tong, Hong Kong, China
[b] School of Environmental Science and Engineering, Guangdong University of Technology, Guangzhou, China

HIGHLIGHTS

- Fate of triclosan was investigated in different wastewater and sludge treatments.
- TCS was considerably biodegraded in aerobic/anaerobic sludge digestion process.
- TCS transformation products were observed based on the treatment practice.
- K_d values were estimated for TCS, 2,4-DCP and MTCS in primary and secondary sludge.
- TCS temporal variations as well as few relationships were investigated.

GRAPHICAL ABSTRACT



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ABSTRACT

Triclosan (TCS) as an antimicrobial agent has been ubiquitously found in wastewater and sewage sludge. TCS may undergo transformation/degradation during wastewater treatment. Some of the resulted products such as 2,4-dichlorophenol (2,4-DCP), 2,8-dichlorodibenzoparadioxin (2,8-DCDD) and methyl triclosan (MTCS) are presumed toxic/persistent compounds. In this study, fate of TCS and the probability of formation of important degradation products were investigated in three susceptible wastewater/sludge treatment practices. 24.1% and 27.2% of the loading TCS was adsorbed to the generated sludge, whereas up to 60% of the loading TCS was biotransformed. Up to 9.9% and 13.0% of TCS loss was attributed to the formation of 2,4-DCP and 2,8-DCDD in chlorination and UV disinfection, respectively. Anaerobic and aerobic sludge digestion processes eliminated up to 23.0% and 56.0% of TCS, respectively. About 7.4% of TCS in aerobic digestion was transformed to methyl triclosan (MTCS). Significant temporal variation of TCS was observed in primary sedimentations, except for chemically enhanced primary treatment that was suggested to be governed by chemical-forced sedimentation. Distribution coefficient (K_d) of TCS was directly correlated to the total organic carbon of the sludge (TOC). Moreover, strong correlation was observed between elimination efficiency in primary sedimentation and loading concentration.

1. Introduction

The production and use of triclosan (TCS) (Table 1S, Supplementary information) as a broad-spectrum anti-microbial agent in a wide range of health care products has extensively become perva-

* Corresponding author at: State Key Laboratory of Environment and Biological Analysis, Department of Chemistry, Kowloon Tong T1215, Hong Kong China.
E-mail address: zwcai@hkbu.edu.hk (Z. Cai).

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sive since its first introduction in 1972. Singer et al. [1] estimated that TCS is globally produced over 1500 tons per year by which Europe participates in 23.3% of the total production. As a result of widespread use of TCS, the chemical has been found in nearly every compartment of the environment such as surface water [2], wastewater [3], sediment [4], sludge [5], human plasma and milk [6], urine [7], fish [8] and etc. TCS has low acute and chronic toxicity on estuarine organisms [9,10]. However, there are some big concerns about this compound. Firstly, TCS has been reported to have adverse effect on the aquatic livings [11,12] as well as the microbial and algae community [13–15]. Secondly, endocrine disruption effect of this compound has been documented vastly [12,16]. More importantly, TCS can be the precursor of more toxic compounds. Yet, due to insufficient information about the safety and potential hazard of TCS, the chemical is still under review by the U.S. Food and Drug Administration (FDA) and has been banned as biocidal product in Europe.

Basically, TCS can be transferred to wastewater through industrial or domestic sewage flow, ranging from ng/l to μg/l in sewage treatment plants (STPs). Subjected to treatment, a considerable amount of TCS will adsorb to the sewage sludge [17] leaving a fraction to be discharged to the environment [18]. During aerobic treatment, TCS can be partially degraded through microbial activity [19]. Thus, elimination of TCS may be mainly physical sorption onto the sludge and biodegradation, and to less extent other transformations [20–22]. Even though the portion of TCS transformation is fairly low, it is still considered as the major concern because the products are known or presumed carcinogens/toxicants. It has been well documented that TCS can transform to chlorophenols, dioxins, methylated TCS, chlorinated TCSs and etc. in certain circumstances [3,4,8]. For instance 2,4-dichlorophenol (2,4-DCP) stemmed from TCS molecule breakdown, is merged in Group 2B, possibly carcinogenic to humans (World Health Organization 1999) and is considered as the priority toxic pollutant by US Environmental Protection Agency (USEPA). 2,8-dichlorodibenzoparadioxin (2,8-DCDD) originated from TCS transformation, is a member of dioxin family with a recently reported relative potency factors of 0.0001 compared to 2,3,7,8-tetrachlorodibenzoparadioxin (2,3,7,8-TCDD) [23]. 2,8-DCDD can transform to higher chlorinated dioxins with higher toxicity under certain conditions such as chlorination [4]. Lastly, microbial degradation of TCS under aerobic condition results in methylation of hydroxyl group to form methyl triclosan (MTCS). Although formation of MTCS mitigates the possibility of dioxin formation by shielding the OH group, it ends up with a more persistent compound than the parent TCS which tends to bioaccumulate in the sludge or living organisms after being discharged [24].

Although TCS occurrence in wastewater has been reported vastly, little attention has been paid to its significance at different STPs sites related to treatment practice. In addition, the behavior of compound in the sludge treatment has been barely noticed probably due to the complexity of sludge matrix that makes the analysis quite troublesome. TCS and its degradation products are prone to partition to the sludge due to their hydrophobic characteristic. If not treated appropriately, polluted sludge may increase public health risk severely due to accumulation of organic pollutants and ultimate transfer to the environment. Thus, finding out the behavior and fate of TCS along a STP without reliable knowledge about the occurrence in wastewater, partitioned fraction to the sludge, degradation and generated products is impossible. More important is that the products might exhibit higher toxicity and persistency than TCS. Therefore, current research was aimed to determine the fate of TCS during different wastewater and sludge treatment practices that were susceptible to affect TCS and investigate how each treatment may contribute to the fate of TCS. Besides, attempt was done to focus on TCS degradation during aerobic/anaerobic sludge digestion. The information obtained based on the specific treatment processes is very valuable for setting the correct strategies toward the improvement of treatment efficiency which in turn causes the reduction in introduction of organic compounds into the environment. For this purpose, analysis of raw and treated wastewater and sludge samples were carried out and the mass balance of TCS in three STPs were performed accordingly based on the experimental data. TCS fate was probed in real-based treatment plant during wastewater and sludge aerobic/anaerobic treatment condition. In addition, the occurrence of probable degradation products, 2,8-DCDD, 2,4-DCP and MTCS and the transformation rate were investigated. Finally, the adsorption behavior of TCS and some of the occurred degradation products was studied.

2. Materials and methods

2.1. STPs description

Three different STPs, STP (I), STP (II) and STP (III) were selected based on different treatment practices. STP (I) is a primary treatment plant serving a population of 23,000 people. The plant is designed to treat about 8600 cubic meters per day domestic wastewater. The treatment train is comprised of pretreatment (grit screening) and primary treatment. After passing the primary sedimentation tank with a hydraulic residence time of 2–3 h, the wastewater is directly discharged to the environment. Sludge treatment process consists of aerobic digestion and the treated sludge is dewatered to be disposed to landfill. STP (II) runs chemically enhanced primary treatment (CEPT) that is supposed to improve treatment process by enhanced flocculation. The plant is capable of handling daily 1.4 million cubic meters of sewage on average for serving a population of 3.5 million people. After contacting with chemicals and settling in primary sedimentation for about 2–3 h, the wastewater is conducted to disinfection chamber for treating with sodium hypochlorite and being discharged accordingly. Produced sludge is only centrifuged and dewatered for landfill disposal. STP (III) as a secondary treatment employs activated sludge treatment (biological treatment) with a daily capacity of 200,000 cubic meters per day for a population of 600,000 people. The influent passes grit removal and primary sedimentation to flow into biological tank where biologically degradable compounds are prone to degradation by the microorganisms. The hydraulic residence time of aerobic biological treatment is between 8 and 10 h. The effluent from the biological tank settles in secondary sedimentation and finally is exposed to UV radiation in UV chamber for disinfection purpose. The sludge originated from primary sedimentation and secondary sedimentation are blended and fed into an anaerobic digester where the sludge is anaerobically digested at 35–37 °C for an average period of 19 days. After treatment, the sludge is centrifuged and dewatered for final disposal. The description about STPs and wastewater quality is summarized in Tables 2S–4S, Supplementary information.

2.2. Sample collection and preparation

Five campaigns were conducted during one year to collect composite samples from wastewater and sludge. Campaigns 1, 4 and 5 were related to December to March (cold dry periods) while campaigns 2 and 3 were performed in May to September (warm rainy periods). Basically, the samples were taken from inlet, after each treatment unit and the outlet of wastewater and sludge. Average climate temperature ranged from 8 to 16 °C and 28–36 °C for cold and warm periods, respectively. Sampling points are presented

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