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Triclocarban enhances short-chain fatty acids production from anaerobic fermentation of waste activated sludge

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ABSTRACT

Triclocarban (TCC), one typical antibacterial agent being widely used in various applications, was found to be present in waste activated sludge at significant levels. To date, however, its effect on anaerobic fermentation of sludge has not been investigated. This work therefore aims to fill this knowledge gap. Experimental results showed that when TCC content in sludge increased from 26.7 ± 5.3 to 520.5 ± 12.6 mg per kilogram total suspended solids, the maximum concentration of short-chain fatty acids (SCFA) increased from 32.6 ± 2.5 to 228.2 ± 3.6 (without pH control) and from 211.7 ± 2.4 to 378.3 ± 3.2 mg COD/g VSS (initial pH 10), respectively. The large promotion of acetic acid was found to be the major reason for the enhancement of total SCFA production. Although a significant level of TCC was degraded in the fermentation process, SCFA was neither produced from TCC nor affected by its major intermediates at the relevant levels. It was found that TCC facilitated solubilization, acidogenesis, acetogenesis, and homoacetogenesis processes but inhibited methanogenesis process. Microbial analysis revealed that the increase of TCC increased the microbial community diversity, the abundances of SCFA (especially acetic acid) producers, and the activities of key enzymes relevant to acetic acid production.

1. Introduction

Triclocarban (TCC) is abundantly added into household consumables and other personal care products such as soap, cosmetics, and shampoo, due to its bacteriostatic properties (Halden and Paull, 2004; Souchier et al., 2015). It is reported that each person would consume 2.06 ± 0.68 mg of TCC daily, and its consumption amount is estimated to achieve at 227–454 tons in USA annually (Heidler et al., 2006). The extensive use of TCC inevitably leads to its environmental release. In fact, TCC is universally detected in terrestrial and aquatic ecosystems at ppb or even ppm level (Lozano et al., 2013; Subedi et al., 2014). TCC is reported to have capability of inhibiting the growth and metabolisms of organisms, thus its potential toxicity to the environment has recently attracted growing concerns.

Several efforts have been carried out to assess the toxicity of TCC to aquatic model organisms, terrestrial organisms, mammals, and humans (Brausch and Rand, 2011; Halden, 2014; Huang et al., 2014; Zarate et al., 2012). For example, Yang et al. (2008) investigated the toxicities of 12 antibacterial agents to the growth of alga *P. subcapitata* and found that TCC caused the highest toxicity among these antibacterial agents with the 50% inhibition concentration of 17.5 μg/L for algal growth. Chung et al. (2011) demonstrated that TCC stimulated the overexpression of aromatase, which converted androgens to estrogens, in early zebrafish embryos. As the last barriers before TCC entering into the natural environment, wastewater treatment plants (WWTPs) could remove most of the TCC present in wastewater via precipitation, biosorption, or other biomass mediated processes (Miller et al., 2008; Ying et al., 2007).

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Heidler and Halden (2007) found that 76% of the influent TCC was adsorbed into the sludge due to its hydrophobic mature through mass balance analysis. Lozano et al. (2013) demonstrated that the total TCC concentration in wasted sludge from WWTPs was approximately 4.15 ± 0.77 kg/d, whereas the wastewater effluent only contained 0.13 ± 0.11 kg/d. Heidler et al. (2006) further found that the TCC concentration could be up to 51 mg/kg in biosolids at the East Coast of U.S. Similar values were also measured in other areas, such as 39 mg/kg in Washington of U.S. and 25.2 ± 0.6 mg/kg in China (Zhu and Chen, 2014). According to our survey on available literature, sludge TCC content was found to be in the range of 20–60 mg/kg TSS in past years (Heidler et al., 2006; Verlicchi and Zambello, 2015; Zhu and Chen, 2014). Due to the increasing use of TCC-based products, sludge TCC content may increase to a much greater level in the future. The intercepted TCC in waste activated sludge might also bring risks to its subsequent treatment, requiring to be assessed urgently.

Generally, waste activated sludge is treated by anaerobic digestion process to produce methane, as this process can effectively reduce the amount of sludge, kill pathogenic microorganisms, and produce energy biogas methane (Wang et al., 2017b, 2017c). Recently, anaerobic fermentation of waste activated sludge to produce short chain fatty acids (SCFA) has attracted growing attention (Duan et al., 2016; Luo et al., 2016; Wang et al., 2016; Zhao et al., 2015a, 2016), because the produced SCFAs are value-added products that can be employed either as raw materials for microbial production of biodegradable plastics or as preferred carbon sources for biological nutrient removal (Jiang et al., 2009; Mino et al., 1998).

Anaerobic fermentation is mainly a biological process, with a variety of microorganisms such as *Bacillus* sp., *Clostridium* sp., and methanogenic Archaea involved. As an antibacterial agent, TCC might suppress the activities and growths of these microbes, which thereby affects the anaerobic fermentation process. Meanwhile, some of the microorganisms present in sludge anaerobic fermentation systems might have ability to bio-degrade TCC, resulting in the changes of its fate in the fermentation process. Although many studies were performed in terms of sludge anaerobic fermentation in the past years, almost all of them focused on promotion of SCFA yield through optimizing pretreatment methods (e.g., free nitrous acid, and ultrasonic), operational conditions (e.g., temperature and pH), or sludge composition (e.g., enhancement of carbohydrate and polyhydroxyalkanoates levels) (Feng et al., 2009; Li et al., 2016; Wang et al., 2013, 2015a; Yan et al., 2010; Zhao et al., 2015a, 2015b). To date, however, all the possibilities mentioned above have not been clarified before, remaining a gap between sludge anaerobic fermentation and emerging contaminants.

The aims of this work are therefore to investigate the behavior of TCC in sludge anaerobic fermentation systems and to reveal the details of whether and how TCC affect anaerobic fermentation of sludge. To achieve a comprehensive understanding, the effect of different TCC levels on SCFA production was first compared in both fermentation systems with or without alkaline pretreatment. Then, the degradation potential of TCC in anaerobic fermentation process was assessed by measuring the variation of TCC in the fermentation systems. As it was found that the increase of TCC content largely promoted SCFA (especially acetic acid) production, the facts of what happen in the TCC-present anaerobic fermentation were explored. To the best of our knowledge, this is the first work clarifying the effects of TCC on sludge anaerobic fermentation and the details of how TCC affects this process. The findings achieved fill the gap between sludge anaerobic fermentation and the emerging contaminant, TCC, and may have important implications to sludge treatment in the future.

2. Materials and methods

2.1. Sources of waste activated sludge and triclocarban

The sludge used in this study was withdrawn from the secondary sedimentation tank of a municipal wastewater treatment plant in Changsha, China. The raw sludge was filtrated by stainless steel mesh (2.0 mm) and was concentrated by setting at 4 °C for 24 h before use. The main characteristics of the concentrated sludge are as follows: pH 6.7 ± 0.1, total suspended soils (TSS) 15,850 ± 290 mg/L, volatile suspended soils (VSS) 12,650 ± 130 mg/L, soluble chemical oxygen demand (COD) 480 ± 10 mg/L, total COD 14240 ± 270 mg/L, total carbohydrate 1570 ± 250 mg COD/L, total protein 7910 ± 330 mg COD/L, lipid and oil 186 ± 25 mg COD/L, and TCC 26.7 ± 5.3 mg/kg TSS. The protein and carbohydrate are the top two organic compounds in sludge, accounting for approximately 67% of total COD. Extra addition of TCC used in this study was purchased from Chemical Industry Park, Shanghai, China, and the purity of this standard is over 98%.

2.2. Batch experiment for SCFA production from sludge anaerobic fermentation in the presence of TCC at different levels

In this experiment, eight reproductive reactors with working volume of 1 L each were conducted at 35 ± 2 °C for 15 d. These reactors were divided into two groups, with four reactors in each. The pH in Group-I was not controlled. Since alkaline condition was reported to be beneficial to SCFA production, alkaline pretreatment (initial pH 10 was used in this test) was applied to Group-II. In this experiment, two TCC concentrations (i.e., 26.7 ± 5.3 and 47.5 ± 6.7 mg/kg TSS) at environmentally relevant level were selected. As sludge TCC content might increase in the future due to its growing use, two higher concentrations of TCC (i.e., 145.3 ± 9.2, 520.5 ± 12.6 mg/kg TSS) were also carried out in this work. In each group, each reactor was fed with 600 mL of the concentrated sludge, and then different dosages of TCC were added into the four reactors at the beginning of the experiment, resulting in the initial sludge TCC level of 26.7 ± 5.3, 47.5 ± 6.7, 145.3 ± 9.2, 520.5 ± 12.6 mg/kg TSS, respectively. After that, all reactors was aerated with N_2 for 5 min, sealed, and placed in an air-bath shaker at stirring speed of 120 rpm.

2.3. Long-term operation of the semi-continuous reactors for the measurements of microbial community and key enzyme activities

In this test, two semi-continuous reactors, which were fed with 600 mL of alkaline pre-treated sludge either containing 26.7 ± 5.3 (Reactor-I) or containing 520.5 ± 12.6 (Reactor-II) mg/kg TSS TCC, were conducted. All the operational conditions were the same as described in the above batch test, excepting the semi-continuous operation described below. According to the above batch test results obtained in Group-II, the maximum SCFA production was measured at 6 d in the reactor fed with 26.7 ± 5.3 mg/kg TSS TCC sludge and 10 d in the reactor fed with 520.5 ± 12.6 mg/kg TSS TCC sludge, thus the sludge retention time in Reactor-I and Reactor-II should be controlled at 6 d and 10 d, respectively. On each day, 100 mL and 60 mL of fermentation mixtures were manually withdrawn from Reactor-I and Reactor-II, respectively. Then, the respective same volumes of raw alkaline pre-treated sludge were respectively added into the two reactors. It took about 2 months for SCFA production from the two reactors being relatively stable, and then the analysis of microbial community and key enzyme activities were made.