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Occurrence of trace organic contaminants in wastewater sludge and their removals by anaerobic digestion

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HIGHLIGHTS

- 18 out of 36 monitored TrOCs were consistently detected in raw primary sludge.
- These TrOCs occurred predominantly in the solid phase.
- TrOC removal by anaerobic digestion was governed by their molecular structure.
- An increase in SRT led to an increase in biogas production and VS removal.
- SRT increase did not lead to any discernible increase in TrOC removal.

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ABSTRACT

This study aims to evaluate the occurrence of trace organic contaminants (TrOCs) in wastewater sludge and their removal during anaerobic digestion. The significant occurrence of 18 TrOCs in primary sludge was observed. These TrOCs occurred predominantly in the solid phase. Some of these TrOCs (e.g. paracetamol, caffeine, ibuprofen and triclosan) were also found at high concentrations (>10,000 ng/L) in the aqueous phase. The overall removal of TrOCs (from both the aqueous and solid phase) by anaerobic digestion was governed by their molecular structure (e.g. the presence/absence of electron withdrawing/donating functional groups). While an increase in sludge retention time (SRT) of the digester resulted in a small but clearly discernible increase in basic biological performance (e.g. volatile solids removal and biogas production), the impact of SRT on TrOC removal was negligible. The lack of SRT influence on TrOC removal suggests that TrOCs were not the main substrate for anaerobic digestion.

1. Introduction

Wastewater treatment involves the settling of solid materials and transformation of dissolved and suspended organic matter to sludge. During wastewater treatment, a large volume of sludge is produced. The EU generates about 10 million tonnes of dry sludge each year (Fytili and Zabaniotou, 2008). In Australia, dry sludge production from wastewater treatment increased by about 3% each year from 0.3 million tonnes in 2010 to 0.33 million tonnes in 2013 (Semblante et al., 2014). Thus, the production of excess sludge from wastewater treatment is a vexing problem and necessitates effective management strategies.

Wastewater sludge has a high organic content and a host of pathogenic vectors. As a result, wastewater sludge must be treated or stabilised prior to environmental disposal. The organic content in wastewater sludge can be converted into energy through a range of technologies including anaerobic digestion (Karthikeyan and Visvanathan, 2013) and microbial fuel cell (Oh et al., 2014). Amongst them, anaerobic digestion is probably the most widely used technology for wastewater sludge treatment (Chernicharo et al., 2015; Kim et al., 2011).

During the anaerobic digestion process, a consortium of microbes metabolizes and converts organic substances into biogas in the absence of oxygen. Anaerobic digestion can achieve a sludge solid reduction of 40–60% (Malina and Pohland, 1992) and generate methane gas as a renewable fuel. The digested sludge from anaerobic digestion can be used as fertilizers and soil conditioners in agriculture (Elliott et al., 1990).

Land application of the digested sludge is a sustainable option because it enables the recovery of important nutrients and adds economic value to what is conventionally perceived as waste.

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Nevertheless, recent discovery of the widespread occurrence of trace organic contaminants (TrOCs) in municipal wastewater suggests that some of these compounds can be transferred to sludge during wastewater treatment (Citulski and Farahbakhsh, 2010; Semblante et al., 2015). These TrOCs include pesticides, industrial chemicals, components of consumer products, pharmaceuticals and personal care products, hormones, and other organic pollutants that are regularly released into municipal wastewater by anthropogenic activities (Luo et al., 2014).

TrOCs have been frequently found in municipal wastewater at very low concentrations (Verlicchi and Zambello, 2015). At a sufficient concentration, some of these TrOCs have the potential to cause chronic disorders in animals and humans. Several countries have already imposed controls on certain TrOCs such as nonylphenol and nonylphenol ethoxylates, polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins and dibenzo-p-furans. However, a clear approach to address TrOCs in digested sludge has not yet been developed (Smith, 2009).

Some TrOCs are lipophilic. In other words, they can be transferred to the solid phase during primary and secondary clarification (Clarke and Smith, 2011), resulting in significantly higher concentrations (several μg/kg dry weight or more) in sludge than wastewater. Persistent TrOCs have the potential to bioaccumulate during land application and, if left unchecked, may impose adverse risk to humans and the ecosystem.

Antibiotics and other pharmaceutically active compounds were amongst the most investigated TrOCs in digested sludge. Trimethoprim, sulfamethoxazole, ciprofloxacin and doxycycline were notable antibiotics detected at the low mg/kg dry weight range in digested sludge from Swedish wastewater treatment plants (Golet et al., 2003; Lindberg et al., 2005). Ciprofloxacin and diphenhydramine were also detected in more than 80 sludge samples across the USA (Grumbles, 2009). In Japan, Narumiya et al. (2013) reported the occurrence of 45 TrOCs in the digested sludge. Concentrations of several compounds (e.g. ofloxacin, triclosan and triclocarban) exceeded 1 mg/kg dry sludge (Narumiya et al., 2013). Several personal care products including triclosan and triclocarban have also been reported to accumulate in anaerobically digested sludge to a high concentration (Heidler and Halden, 2007; Heidler et al., 2006).

Most previous studies concerning anaerobic treatment have focused specifically on the removal of TrOC from the aqueous (water) phase. Thus, findings from these studies are not readily applicable to anaerobic digestion of wastewater sludge. Indeed, results from recent studies (Carballa et al., 2007; Hernandez-Raquet et al., 2007; Malmborg and Magner, 2015; Narumiya et al., 2013) examining the removal of TrOCs from both aqueous and solid phases by anaerobic digestion show that the overall removal efficiency could be lower compared to studies that only reported TrOC removal from the aqueous phase.

It is noteworthy that most previous studies involved the spiking (artificial addition) of TrOCs to the feed sludge at elevated concentrations. Malmborg and Magner (2015) studied the fate of 14 different TrOCs during anaerobic digestion by spiking each compound at 50 mg/L into the sludge. They showed that several compounds (e.g. trimethoprim, citalopram, and furosemide) were well removed by anaerobic digestion. However, several others including fluoxetine and carbamazepine were persistent to anaerobic digestion. Similar results were reported by Carballa et al. (2007) who added TrOCs to feed sludge at concentrations between 4 and 400 μg/L. Narumiya et al. (2013) was probably the only group of authors who have monitored the environmental concentrations of TrOCs in the feed sludge. They showed that 4 out of 26 compounds, namely, sulfamethoxazole, trimethoprim, caffeine and acetaminophen detected in the thickened sludge were well removed by anaerobic digestion while most of the remaining compounds were not significantly removed.

This study aims to reveal the occurrence and fate of TrOCs during anaerobic digestion of primary sludge. Basic biological performance of anaerobic digesters at a range of sludge retention time (SRT) is systematically examined. TrOCs concentrations in the aqueous and solid phase from both primary and digested sludge are quantified to examine their fate during anaerobic digestion.

2. Methods

2.1. Wastewater sludge

Anaerobically digested sludge and primary sludge were taken from a full scale wastewater treatment plant in New South Wales (Australia) as inoculum and feed, respectively. The primary sludge was stored at 4 °C for a maximum of 2 weeks before fresh sludge was collected again. The total solids (TS) content of this primary sludge was 25.7 ± 6.6 g/L (average ± standard deviation of eight samples). The ratio of volatile solids (VS) over TS (VS/TS) of this primary sludge was stable (0.89 ± 0.03) during the current study. pH value of the primary sludge was in the range of 5.35–5.59.

2.2. Anaerobic digester

Three identical anaerobic digesters were used. Each digester (Supplementary Data Fig. S1) consists of a 28 L conical shape stainless steel reactor, a peristaltic hose pump (DULCO® Flex from ProMinent Fluid Controls, Australia), a thermal couple with temperature gauge, a custom made gas counter, and a gas trap for biogas sampling. Hot water flowing inside a rubber hose wrapping around the digester was used for heating. The entire reactor was insulated by polystyrene foam. The temperature of the digester was maintained at 35.0 ± 0.5 °C by regulating the temperature inside the rubber hose using a temperature control unit (Neslab RTE 7, Thermo Fisher Scientific, Newington, USA). When necessary, biogas from the gas counter was directed to a gas trap for biogas composition analysis.

2.3. Experimental protocol

Each digester was seeded with anaerobically digested sludge at the beginning of the experiment. The peristaltic pump was operated continuously at the flow rate of 60 L/h to provide sufficient sludge mixing. The active volumes of all three digesters were maintained at 20 L throughout the experiment. The SRT of the three digesters were set at 15, 20 and 30 d, respectively, by withdrawing and feeding a predetermined volume of sludge each day. The digesters were first stabilized for 2 weeks. Digested sludge and feed samples were then collected for analysis over 12 weeks of continuous operation.

2.4. Analytical methods

2.4.1. Biogas production and composition

Biogas production was monitored using an online gas counter. Biogas composition analysis was conducted every week. Approximately 1 L of biogas was collected in the gas trap (Supplementary Data Fig. S1). A portable gas analyser (GA5000 Gas Analyser, Geotechnical Instruments (UK) Ltd, England) was then used for biogas composition analysis (Nghiem et al., 2014). Methane production activity (L-CH_4/g $VS_{removed}$) was calculated based on the methane composition in biogas and the biogas production rate.

2.4.2. Sludge characterisation

Sludge samples were taken weekly from each digester for analysis. Primary sludge samples were also characterised on a weekly