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Effect of microwave hydrolysis on transformation of steroidal hormones during anaerobic digestion of municipal sludge cake

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ABSTRACT

Fate and removal of 16 steroidal (estrogenic, androgenic and progestogenic) hormones were studied during advanced anaerobic digestion of sludge cake using microwave (MW) pretreatment. Effect of pretreatment temperature (80, 120, 160 °C), operating temperature (mesophilic at 35 ± 2 °C, thermophilic at 55 ± 2 °C) and sludge retention time (SRT: 20, 10, 5 days) were studied employing eight lab-scale semi-continuously fed digesters. To determine the potential effect of MW hydrolysis, hormones were quantified in total (sorbed + soluble) and supernatant (soluble) phases of the digester influent and effluent streams. Seven of 16 hormones were above the method reporting limit (RL) in one or more of the samples. Hormone concentrations in total phase of un-pretreated (control) and pretreated digester feeds ranged in <157–2491 ng/L and <157–749 ng/L, respectively. The three studied factors were found to be statistically significant (95% confidence level) in removal of one or more hormones from soluble and/or total phase. MW hydrolysis of the influent resulted in both release (from sludge matrix) and attenuation of hormones in the soluble phase. Accumulation of estrone (E1) as well as progesterone (Pr) and androstenedione (Ad) in most of the digesters indicated possible microbial transformations among the hormones. Compared to controls, all pretreated digesters had lower total hormone concentrations in their influent streams. At 20 days SRT, highest total removal (E1+E2+Ad +Pr) was observed for the thermophilic control digester (56%), followed by pretreated mesophilic digesters at 120 °C and 160 °C with around 48% efficiency. In terms of conventional performance parameters, relative (to control) improvements of MW pretreated digesters at a 5-d SRT ranged in 98–163% and 57–121%, for volatile solids removal and methane production, respectively.

Abbreviations list: Ad, androstenedione; An, androsterone; CAS, chemical abstract services; COD, chemical oxygen demand; E1, estrone; E2, 17β-Estradiol; E3, estriol; EE2, 17α-Ethinylestradiol; EDC, endocrine disrupting compound; K_d, partition coefficient; Ms, mestranol; MW, microwave; OLR, organic volumetric loading rate; Pr, progesterone; PS, primary sludge; RL, reporting limit; SDL, sample specific detection limit; SRT, solid retention time; Tr, testosterone; TS, total solids; TVFA, total volatile fatty acids; TWAS, thickened waste activated sludge; VS, volatile solids; WAS, waste activated sludge; WWTP, wastewater treatment plant.

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1. Introduction

In Canada, approximately 388,700 dry tones of biosolids are produced yearly; about 43% of it is land applied. This percentage is expected to increase as different municipalities are recognizing land application to be the most sustainable and environment friendly disposal option for biosolids, as opposed to landfilling and incineration (Apedaile, 2001). However, in addition to existing environmental and health concerns related to presence of coliforms and heavy metals, presence of some natural hormones, pharmaceutical and personal care products in biosolids are causing public opposition against the beneficial reuse of biosolids (Citulski and Farahbakhsh, 2010). A number of these compounds are known to interfere with the natural functioning of endocrine system in human and animals and are called endocrine disrupting compounds (EDCs) (Hansen et al., 1998; Tyler et al., 2005). Among these, natural steroidal hormones have been characterized to be the most potent of all EDCs (Legler et al., 1999, 2002a, 2002b). Some studies show that although steroidal hormones have the potential to undergo transformations in agricultural settings, they are likely to persist long enough to impact the water quality of runoff (Yang et al., 2012). Furthermore, others indicate that hormone leaching and runoff can result in after land application of manure (Kjaer et al., 2007; Jenkins et al., 2009).

Anaerobic digestion is a commonly used biosolids treatment method that converts the organic waste into methane-enriched biogas and fertilizer by combined action of a mixed community of microorganisms (Botheju and Bakke, 2011). Stabilization of high-strength organic waste, production of heat or electricity by recovered methane, and reduced greenhouse gas emissions have made anaerobic digestion more favorable in today's world. Recent developments in this field indicate that thermal (i.e., conventional and microwave (MW) heating) (Ge et al., 2010; Wett et al., 2010; Yu et al., 2010), mechanical (e.g., high pressure homogenizer, sonication, stirred ball mills) (Onyeche, 2006; Wood et al., 2009; Salsabil et al., 2010) and chemical (e.g., acidic and alkaline, ozonation) (Lin et al., 2009; Kim and Youn, 2011) pretreatments applied to complex organic waste (i.e. industrial and municipal secondary sludge, manure) prior to anaerobic digestion can substantially accelerate the hydrolysis and subsequent methane recovery in smaller digesters. Among these, thermal hydrolysis methods are reported to show improved pathogen destruction and dewaterability in addition to higher improvement in methane recovery compared to other methods (Pino-Jelcic et al., 2006; Carballa et al., 2009; Yu et al., 2009). Initially, MW hydrolysis studies at 2.45 GHz and below boiling point of water using kitchen type MW ovens have shown to effectively break the complex polymeric network of waste activated sludge (WAS) and disintegrate the organic macromolecules (e.g., protein, polysaccharides and nucleic acids) (Eskicioglu et al., 2007a). Later, pretreatment of thickened WAS (TWAS) with a bench-scale industrial MW unit and pressure sealed vessels resulted in 31% higher biogas production and 75% improvement in dewaterability at a temperature of 175 °C (Eskicioglu et al., 2009).

To date, conventional parameters, such as organics removal, methane recovery, pathogen removal, dewaterability, have been the focus of almost all the studies dealing with thermal hydrolysis in lab- or full-scale operations. A large research gap exists regarding the effects of thermal hydrolysis on fate and removal of micropollutants, e.g., steroidal hormones, during advanced anaerobic digestion. Only a few studies have reported the fate and removal of estrogenic hormones (e.g. estrone (E1), 17β-estradiol (E2), estriol (E3) and 17α-ethinylestradiol (EE2)) from full-scale and lab-scale conventional anaerobic digesters without advanced hydrolysis (Andersen et al., 2003; Furlong et al., 2010; Muller et al., 2010; Ifelebuegu, 2011; Paterakis et al., 2012). A single study using thermal pretreatment by conventional heating (Carballa et al., 2006) reported the removal efficiencies of estrogenic hormones (i.e., E1, E2 and EE2) in pilot-scale mesophilic and thermophilic anaerobic digesters utilizing mixed municipal waste sludge. Despite the high removal efficiency (>80%) of the natural estrogens, little or no effect of pretreatment was observed.

As pretreatments disintegrate the complex polymeric network in waste sludge samples, it is likely that some hormones, initially encapsulated within the polymeric network, may be released into the soluble phase and render themselves more or less biodegradable depending on the changes on their molecular structures at different pretreatment conditions (low or elevated temperatures, mechanical shear, or chemical dose). Furthermore, as sludge pretreatments solubilize organics (possibly hormones as well), it could be postulated that there is a potential to shift the ultimate disposal route of residual hormones in digestate from a landfill or agricultural land to main wastewater stream. As wastewater treatment plants (WWTPs) incorporating thermal hydrolysis prior to anaerobic digestion may contain higher solubilized hormones in their digestates, there may be higher concentration of hormones recycled back to the head of the treatment train with the digester centrate. Therefore, the objective of this work was to study the effect of MW hydrolysis on fate and removal of steroidal hormones (synthetic and natural displayed in Table 1, Fig. S1), that are already present (without spiking) in dewatered municipal sludge cake, during advanced anaerobic digestion. The effect of pretreatment and digestion temperatures as well as sludge retention time (SRT) have also been evaluated at the lab-scale.

2. Materials and methods

2.1. *Substrate and inocula*

Municipal sludge cake was collected bi-weekly from Kelowna Pollution Control Center (KPCC) (British Columbia (BC), Canada), currently serving more than 94,000 population. In this WWTP after preliminary treatment and primary sedimentation, wastewater flows into a modified Bardenpho unit designed to remove C, P and N. The secondary effluent then undergoes UV disinfection before being discharged into the Okanagan Lake. Fermented and gravity thickened primary sludge (PS) and WAS, thickened by a dissolved air flotation unit, are pumped separately and mixed (40:60, %v/v) before dewatering by centrifugation. The dewatered sludge cake with a total solids (TS) content of 17.5 ± 1% (w/w) was used for MW

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