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Occurrence and transformation of veterinary antibiotics and antibiotic resistance genes in dairy manure treated by advanced anaerobic digestion and conventional treatment methods☆

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A B S T R A C T

Manure treatment technologies are rapidly developing to minimize eutrophication of surrounding environments and potentially decrease the introduction of antibiotics and antibiotic resistant genes (ARGs) into the environment. While laboratory and pilot-scale manure treatment systems boast promising results, antibiotic and ARG removals in full-scale systems receiving continuous manure input have not been evaluated. The effect of treatment on ARGs is similarly lacking. This study examines the occurrence and transformation of sulfonamides, tetracyclines, tetracycline degradation products, and related ARGs throughout a full-scale advanced anaerobic digester (AAD) receiving continuous manure and antibiotic input. Manure samples were collected throughout the AAD system to evaluate baseline antibiotic and ARG input (raw manure), the effect of hygenization (post-pasteurized manure) and anaerobic digestion (post-digestion manure) on antibiotic and ARG levels. Antibiotics were analyzed by liquid chromatography-tandem mass spectrometry (LC-MS/MS), and the ARGs *tet*(O), *tet*(W), *sul*1 and *sul*2 were analyzed by quantitative polymerase chain reaction (Q-PCR). Significant reductions in the concentrations of chlortetracycline, oxytetracycline, tetracycline and their degradation products were observed in manure liquids following treatment ($p < 0.001$), concomitant to significant increases in manure solids ($p < 0.001$). These results suggest sorption is the major removal route for tetracyclines during AAD. Significant decreases in the epimer-to-total residue ratios for chlortetracycline and tetracycline in manure solids further indicate degradation is desorption-limited. Moreover, *sul*1 and *sul*2 copies decreased significantly ($p < 0.001$) following AAD in the absence of sulfonamide antibiotics, while tetracyclines-resistant genes remained unchanged. A cross-sectional study of dairy farms utilizing natural aeration and liquid-solid separation treatments was additionally performed to compare levels of antibiotics and ARGs found in AAD with the levels in common manure management systems. The concentration of antibiotics in raw manure varied greatly between farms while minimal differences in ARGs were observed. However, significant ($p < 0.01$) differences in the levels of antibiotics and ARGs (except *tet*(W)) were observed in the effluents from the three different manure management systems.
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1. Introduction

The release of antibiotics into the environment from animal waste has become a salient issue due to the growing links between antibiotic use and resistance (Ghosh and LaPara, 2007; Khachatourians, 1998; Pruden et al., 2006). Wastes resulting from animal feeding operations are considered important sources of antibiotics to surrounding ecosystems following their agronomic application as fertilizer (Hu et al., 2010; Martinez-Carballo et al., 2007). More than 15.6 million kg of antibiotics are sold for agricultural purposes in the US alone (USFDA, 2016), for disease control (therapeutic), prophylaxis, and growth promotion (sub-therapeutic) purposes. Following consumption, up to 90% of certain antibiotics pass through animals unmebatolized (Kim et al., 2011; Sukul et al., 2009), and some excreted metabolites convert back into the active drug upon introduction into the environment (Díaz-Cruz and

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Barceló, 2007; García-Galán et al., 2008). Once released, many antibiotics persist in the environment due to immobilization (Schmidt et al., 2007) or insufficient degradation (Chenxi et al., 2008).

Several studies have reported wide variations in antibiotic concentrations in manure due to vastly different use patterns for swine, calves, and adult cattle. Levels up to several mg/kg have been reported in animal excrement resulting from intensive sub-therapeutic antibiotic use in China (Pan et al., 2011; Zhao et al., 2010). However, levels are predicted to be minimal in United States' diary waste streams receiving predominantly adult cow manure due to stringent restrictions on antibiotic use in lactating animals (Davis et al., 2009; Payne et al., 1999; USFDA, 1996). Treatment of milk-producing animals with antibiotics is accompanied by lengthy market withdrawal periods for milk or meat. Use is therefore restricted to extreme cases of respiratory, reproductive, and hoof illness, with limited treatment for mastitis due to the associated economic disincentive of extended product disposal (Zwald et al., 2004).

Continued antibiotic release into the environment through land application of manure is considered closely linked to the global increases in antibiotic resistance and proliferation of antibiotic resistant genes (ARGs) (Heuer et al., 2011). The occurrence of antibiotics exerts selection pressure on native bacteria carrying ARGs and may instigate horizontal gene transfer (Martinez, 2009). The threat of antibiotic resistance is compounded by the mobile nature of ARGs, which can be transferred to organisms of different taxa, including potentially pathogenic bacteria (Bennett, 2008; Ochman et al., 2000). Additionally, ARGs may retain viability following host-cell death and can be recombined with genetic material of living bacteria through transformation (Aminov and Mackie, 2007).

Manure treatment has been identified as a promising method for reducing the nutrient and pathogen loading of cattle waste (Burke, 2001; Hubbard and Lowrance, 1998; Kearney et al., 1993), and decreasing agricultural output of antibiotics over traditional storage strategies (Alvarez et al., 2010; Arikan, 2008; Wu et al., 2011). Notable reductions in antibiotic concentrations have been reported in lab-scale anaerobic digesters (Alvarez et al., 2010; Arikan, 2008; Arikan et al., 2006) and composting systems (Dolliver et al., 2008; Qian et al., 2016; Wu et al., 2011). However, most studies are limited in analyses to specific antibiotics and do not consider all of those approved for agricultural use. Additionally, knowledge regarding the occurrence and degradation of antibiotics in full-scale manure treatment systems receiving continuous antibiotic input at varying concentrations is largely absent. Many recent studies investigating the occurrence of antibiotics in anaerobic digesters have focused on the inhibiting effect of antibiotics on the digestion process (Angenent et al., 2008; Shi et al., 2011; Stone et al., 2009), rather than on the occurrence or elimination of antibiotics in the waste stream. Similarly, the effects of new treatment technologies, such as hygenization by pasteurization, remain unstudied with respect to the potential for elimination of antibiotics and ARGs from manure.

Herein, an examination of the occurrence of tetracycline and sulfonamide antibiotics and selected ARGs in a full-scale advanced anaerobic digester (AAD) receiving dairy manure is presented. The system is equipped with pasteurization technologies to accelerate the hydrolysis of organic materials, protect methanogenic bacteria, and remove pathogens (Ariunbaatar et al., 2014). Sulfonamides and tetracyclines represent important classes of antibiotics, accounting for almost 50% of the total agricultural antibiotic consumption (USFDA, 2016), and thus, along with related ARGs, present substantial environmental concern. Although traditional anaerobic digesters are fairly common on dairy farms in the United States with 209 facilities operating nation-wide, there are only three operational AAD systems registered to dairy operations nation-wide (USEPA, 2016). As a result, little is known about the effect of AAD treatment on the occurrence and elimination of antibiotics and ARGs in diary manure.

Common management practices, including naturally-aerated (NA) lagoon storage and liquid-solid separation (LSS), are also examined in a single-season cross-sectional fashion to provide comparison with the persistence of antibiotics in AAD. Naturally-aerated lagoon storage is the most common manure management practice in the United States with over 90% of operations with greater than 200 head utilizing some sort of lagoon storage (USDA, 1996). Liquid-solid separation practices, in which the organic and inorganic solids are separated from manure slurry by gravitational or mechanical means for separate storage and processing, are less common (USDA, 2016). In general, farms utilizing LSS systems use the separated solids as a feed-stock for digestion processes, or as renewable bedding source (Garcia et al., 2009; USDA, 2016). While only 10% of farms utilize recycled manure products for bedding, nearly 50% of the dairy cows in the US are housed with bedding made from recycled manure solids (USDA, 2016).

This study marks the first attempt to examine the effect of a full-scale AAD on the abundance of antibiotics and ARGs in dairy manure. This work further examines both liquid and solid manure components to provide insight into the phase distribution of antibiotics and to minimize reporting error associated with variations in manure composition. Separation of liquids and solids further allows for a more precise examination of potential environmental impacts of manure disposal. Additional emphasis is placed upon the relative quantities of parent antibiotics to their degradation products (i.e. tetracycline to 4-epitetracycline) to examine variations in the degradation occurring under each treatment process.

2. Materials and methods

2.1. Sample collection, storage, and analysis

Duplicate samples were collected from a 2200-cow dairy farm utilizing an AAD facility for manure treatment. Raw manure is mixed with food-grade organic wastes at a weekly-determined ratio (35%–40%) prior to any treatment to improve methane output (Mata-Alvarez et al., 2014). The AAD facility employs pre-digestion pasteurization (67 °C, 1 h) for hygenization and pre-hydrolysis of the mixed input for enhanced methanogenic efficiency (Ariunbaatar et al., 2014; Ziemba and Peccia, 2011). The pasteurized mixture is then subjected to mesophilic anaerobic digestion with a 22-day hydraulic retention time. Samples were collected during winter and spring from (1) unmixed raw manure, (2) after mixing and pasteurization, and (3) following anaerobic digestion. Survey samples (winter) were collected during the same sampling event. Spring samples were collected during two consecutive days and again after 22-days to follow one batch of manure through the treatment process. A schematic of the AAD system is presented in Supplemental Fig. S1.

Conventional, naturally aerated (NA) manure samples were collected during the spring from the lagoon of a 1400-cow dairy farm containing two months' storage, into which manure is constantly added and removed for application as fertilizer. Raw and separated manures were also collected from a liquid-solid separation (LSS) system located on an 1800-cow dairy farm where raw manure is mechanically separated into liquids and solids by a screw-press followed by secondary sedimentation. Resultant liquids are subsequently stored and spread as fertilizer. Separated solids are dried and reused as animal bedding. Due to limited access to NA and LSS farms, only spring samples were collected and analyzed. Schematics of the NA and LSS systems are presented in Figs. S2 and S3, respectively.

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