

Research Paper

Available online at www.sciencedirect.com ScienceDirect

journal homepage: www.elsevier.com/locate/issn/15375110



Seasonal and daily emissions of methane and carbon dioxide from a pig wastewater storage system and the use of artificial vermiculite crusts

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ARTICLE INFO

Article history: Received 9 September 2014 Received in revised form 16 December 2014 Accepted 29 December 2014 Published online 16 January 2015

Keywords: Greenhouse gas emission Wastewater Storage Daily and seasonal dynamics Reduction performance Vermiculite There is insufficient knowledge of the seasonal and daily dynamics of greenhouse gas (GHG) emissions from preliminary storage tanks before anaerobic digestion as well as the reduction strategies required, particularly for full-scale cases. A full-scale three-stage wastewater storage tank system was investigated to quantify the dynamics of daily and seasonal GHG emission and the ability of vermiculite as artificial crust material to reduce GHG emissions. Overall CH₄ and CO₂ emissions in winter were approximately 8 g [CH₄] m⁻² h⁻¹ and 3 g [CO₂] m⁻² h⁻¹, respectively. These values were significantly lower than the 35 g [CH₄] m⁻² h⁻¹ and g [CO₂] m⁻² h⁻¹ values found in summer because of the lower temperatures. The daily fluctuations of CH₄ and CO₂ emission fluxes did not depend on the diurnal temperature changes, but on the barn wastewater flushing operation. The ability of vermiculite as artificial crust material to reduce GHG emissions on a full-scale pig wastewater storing system was verified. The overall reduction from the use of vermiculite, expressed as CO₂ equivalent per year, was calculated to be 36.9% for summer and 17.1% for winter, indicating a useful potential application.

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1. Introduction

Global climate has become warmer since the late 19th century and this is most likely to be caused by anthropogenic activity. Most of the observed increase in global average temperature is likely to be caused by the increase in greenhouse gas (GHG) concentrations found in the atmosphere (Stocker et al., 2013). Among the world's largest emitters of GHG, China is reported to have one of the largest total potential emission because of its rapid and continued industrial development over the last five decades (Birol, 2014; Guan, Liu, Geng, Lindner, & Hubacek, 2012). Faced with this situation, the Chinese government has committed to cut the carbon dioxide (CO_2) emission per unit of gross domestic product by 40 %–45 % by 2020 from the 2005 level of 7.467 billion tons of CO_2 equivalent (NDRCCC, 2013).

Based on the 2013 report of the Second National Communication on Climate Change of the People's Republic of China, GHG emission from agriculture contributed to 11% of China's total GHG emissions in 2005. This result indicates the importance of the potential GHG emission reduction from agricultural activities (NDRCCC, 2013). Livestock is a significant part

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http://dx.doi.org/10.1016/j.biosystemseng.2014.12.009

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Abbreviations	
GHG	Greenhouse Gas;
VS	Volatile Solid;
TSWST	Three-Stage Wastewater Storage Tank System;
SCOD	Soluble Chemical Oxygen Demand;
TOC	Total Organic Carbon
CH_4	Methane
CO ₂	Carbon Dioxide
O ₂	Oxygen
NH_4^+-N	Total Ammonia Nitrogen;
Symbols	
F	GHG Emission Flux (mg $h^{-1} m^{-2}$)
М	Molar Mass (g mol ⁻¹)
Р	Atmospheric Pressure (Pa)
R	Universal Gas Constant
	(8.3144 Pa $m^3 mol^{-1} K^{-1}$)
Т	Temperature (K)
dc/dt	Linear Fitting Value of the Gas and Time
h	Height (m)
ρ	Gas Density (mg m $^{-3}$).

of agriculture and it contributes large quantities of GHG emissions. Therefore, investigations of GHG emissions from livestock and corresponding reduction technologies have been increasingly gaining the attention of both scientists and engineers.

China has the world's largest pig livestock production; approximately 700 million pigs were slaughtered in 2012 (Bureau, 2012). In all pig breeding processes the two sources of GHG emissions, namely, pig house and manure composting, were traditionally recognised as being important in investigations of GHG emissions. In the case of pig house, GHG emission is mainly from enteric gas and manure fermentation. Research and practical work in this field has been comprehensively carried out in the last few years, and the results have indicated that emissions from this source can be influenced by different factors and can vary with animal species, feeding schemes, manure handling practices, and environmental conditions (Blanes-Vidal, Hansen, Pedersen, & Rom, 2008; Dong et al., 2007). In view of these results, various GHG reduction strategies have been developed including improved feeding practices, specific agents, and dietary additives as well as longer-term management changes and animal breeding (Smith et al., 2008). Manure composting in a livestock farm, which is organic degradation by anaerobic and aerobic microbial activities, is another source of GHG emission (Li, Li, & Li, 2003). GHG emissions from this source are closely related with oxygen (O₂) content inside a compost (Xie & Li, 2003). However, the emission behaviours and reduction strategies for the preliminary storage of wastewater before full treatment, particularly wastewater from the Ganqingfen manure cleaning system, have often neglected and have not gained enough attention so far.

In the Ganqingfen system, the overwhelming majority of the solid parts of faeces are cleaned using a shovel for composting; the remaining manure and urine are flushed by water and collected for further treatment (Jiang, Schuchardt, Li, Guo, & Luo, 2013). Anaerobic digestion for biogas is the most commonly used treatment in China because of its economic efficiency and sustainability. Anaerobic digestion in the biogas production process requires stored wastewater and/or feeding tank system for wastewater storing. The wastewater remains in these tanks for 1 or 2 d before entering the digester. Significant amounts of methane (CH₄) and CO₂ are produced by microbial activities during the storage of the wastewater in these tanks (Sommer, Petersen, Sørensen, Poulsen, & Møller, 2007). Some analogous studies that focused on GHG emission on different scenarios have been conducted. The production of CH_4 and CO_2 are influenced by a multitude of different factors, and complex interactions exist between different emitting sources (Amon, Kryvoruchko, Amon, & Zechmeister-Boltenstern, 2006) such as temperature, pH, and volatile solid (VS) content. A positive linear response from $\rm CH_4$ and $\rm CO_2$ emissions to increasing water temperature has been indicated in a pig waste holding tank (MacDonald et al., 1998; Sharpe & Harper, 1999). Beyond this observation, pH has been found to impact on the emission of CH_4 , and a laboratory-scale experiment that utilised stored pig slurry showed that lower pH reduces CH₄ emission (Berg, Brunsch, & Pazsiczki, 2006). However, few experimental data are available in relation to the investigation on GHG emission at a preliminary storage tank before anaerobic digestion under the Gangingfen system in China since the influencing factors have not been clarified.

Reduction of GHG emissions during preliminary storage treatment has become an urgent issue. Several studies have indicated that the natural crust can reduce CH₄ emissions from stored wastewater by acting as a sink for CH₄ through the use of bacterial oxidation (Husted, 1994; Petersen, Amon, & Gattinger, 2005; Sommer, Petersen, & Søgaard, 2000). Inspired by these studies, different kinds of materials have been tested as artificial crusts to reduce the emissions of GHG. Amon, Kryvoruchko, and Amon (2006, Amon, Kryvoruchko, Amon, et al., 2006) carried out a comparative investigation on the CH_4 reduction performance of a wooden lid and chopped straw placed on the untreated slurry tank (Amon, Kryvoruchko, Amon, 2006). The results indicated that the arrangement of a wooden lid on the surface of untreated slurry tank could effectively reduce CH₄ emission, whereas a layer of chopped straw has the potential to increase emission. Similarly, Berg et al. (2006) determined that a crust made of straw can increase emissions of CH4; a similar consequence was observed when perlite and lightweight expanded clay aggregate (Leca) were used (Berg et al., 2006). These results emphasised the importance of artificial crust materials on GHG reduction. Vermiculite, one of the world's lightest minerals, is a hydrous, silicate mineral that is classified as a phyllosilicate. It has high layer charge, which produces high cationic exchange capacity and a strong cation exchange adsorption capacity (Wang, Wu, Dang, & Xie, 2006). Moreover, it floats on a water surface, acts as a discharge barrier of gases, and provides a habitat for CH₄ oxidation (Ambus & Petersen, 2005). Despite these results, very limited research work has been conducted on vermiculite, and the experiments have been conducted have mainly been at laboratory scale.

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