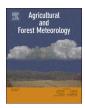
ARTICLE IN PRESS

Agricultural and Forest Meteorology xxx (xxxx) xxx-xxx



Contents lists available at ScienceDirect

Agricultural and Forest Meteorology



journal homepage: www.elsevier.com/locate/agrformet

Year-round methane emissions from liquid dairy manure in a cold climate reveal hysteretic pattern

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ARTICLE INFO

Keywords: Micrometeorological mass balance method Tunable diode laser trace gas analyzer Methane flux Stored liquid dairy manure Methane conversion factor Arrhenius equation Hysteresis of temperature response Volatile solids Acetic acid

ABSTRACT

Liquid dairy manure storage is favourable for methane (CH₄) production and a significant source of this greenhouse gas. Manure storage in cold climates faces large temperature variations over the course of a year, and added on-farm complexities in manure loading require year-round measurements which are currently lacking. The objectives of this study were to 1) quantify CH4 emissions from farm-scale liquid dairy manure storage in a cold climate, 2) investigate the effect of manure temperature on CH₄ emissions, and 3) compare measured CH₄ emissions with values derived from the USEPA temperature model. Methane fluxes were measured from Aug 2010 to early Nov 2011 on a commercial farm in Ontario, Canada, using a micrometeorological mass balance method which relied on 3 towers with air intakes at 4 heights placed around the storage tank. Manure temperature and volatile solid content (VS) were also measured. Monthly CH4 emissions scaled by VS decreased from 43.8 g CH4 kg^{-1} VS in Sep 2010 to 5.3 g CH₄ kg^{-1} VS in Jan 2011, and were correlated with temperature ($r^2 = 0.94$). Manure temperature increased starting in Feb 2011, but the increase in CH₄ emissions was delayed to Jul 2011. Hence, the response of CH₄ emissions to temperature showed a hysteretic pattern where emissions in the warming branch after the winter (Apr-Jun) were significantly lower than emissions in the cooling branch after the summer (Sep-Nov) despite similar temperatures. The temperature model predicted emissions during the cooling phase well, but overpredicted emissions during the warming phase, indicating other limiting factors for microbial CH₄ production. Thus, the annual predicted CH_4 emissions were about $2.3 \times$ larger than the measured values. The newly observed hysteresis effect should be considered when predicting CH₄ emissions from stored manure.

1. Introduction

Methane (CH₄) is an important greenhouse gas (GHG) with 28 times the global warming potential of carbon dioxide (CO₂) over a 100-year time horizon, contributing 18% of current anthropogenic global warming (Myhre et al., 2013). Agriculture is a significant source of CH₄, with manure management contributing 13 and 26% of Canada's and United States' total agricultural CH₄ emissions, respectively (Environment Canada, 2015; US EPA, 2016). Manure management in the dairy industry contributes 27% of the milk carbon footprint of 1.0 kg CO₂ eq kg⁻¹ fat and protein corrected milk, mainly as CH₄ (Verge et al., 2007; Jayasundara and Wagner-Riddle, 2014). However, these estimates are based on models derived from limited data that have not been widely verified for commercial farms.

Methane production from dairy manure results from microbial decomposition of organic materials under anaerobic conditions (Husted,

1994; Steed and Hashimoto, 1994; Khan et al., 1997) and is influenced by the quantity of manure produced (Safley and Westerman, 1989), manure composition (Hashimoto et al., 1981; Petersen et al., 1998), manure management system (Steed, 1994; Steed and Hashimoto, 1994; Dustan, 2002; Chadwick et al., 2011) and climatic conditions (Hashimoto et al., 1981; Safley and Westerman 1989; Sommer et al., 2004). Type of manure management system determines important CH₄ production regulating factors, including contact with oxygen, water content, pH and nutrient availability (IPCC, 2006). In liquid systems, manure is typically scraped from the barn floor, mixed with wash-water and stored as a liquid (< 10% dry matter) in a lagoon, pond or tank, until land application. These conditions are conducive to anaerobiosis, thus producing a significant amount of CH₄ compared to solid manure management systems (Husted 1994; Steed and Hashimoto, 1994; US EPA, 1999; Sneath et al., 2006; Amon et al., 2006). More than 50% of Canadian dairy farms manage manure as a liquid (Jayasundara et al.,

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https://doi.org/10.1016/j.agrformet.2017.12.185

Received 10 July 2017; Received in revised form 7 November 2017; Accepted 6 December 2017 0168-1923/ Crown Copyright © 2017 Published by Elsevier B.V. All rights reserved.

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2016). Increasing use of liquid systems to manage dairy manure is a general trend observed in Canada (based on data from Statistics Canada, 2003; VanderZaag et al., 2013; Jayasundara et al., 2016), the United States (US EPA, 2016; Wightman and Woodbury, 2016) and in many European countries (Eurostat, 2016).

Temperature is a key climatic factor that affects methanogenesis. In liquid manure systems, high temperatures result in rapid organic matter decomposition, which leads to increased CH₄ generation (Masse et al., 2003; Møller et al., 2004; Sommer et al., 2007; Umetsu et al., 2005; Masse et al., 2008). These studies were conducted in controlled environments with small manure volumes ($< 0.5 \text{ m}^3$) and/or using models to investigate the temperature effect on CH₄ production. For commercial farms in cold climates, liquid systems handle manure in large volumes (> 1000 m³; Hilborn, 2015) and face significant stratification and temperature variations throughout the year as they undergo seasonal changes such as summer to winter cooling and winter to summer warming. During winters, liquid manure in storage tanks can be covered with ice and/or snow and in early spring, surface thaw occurs. VanderZaag et al. (2011) conducted measurements at a Canadian dairy farm over 7-months through winter and spring and observed a large CH₄ flux event in spring due to surface thaw. Higher CH₄ emissions at two dairy farms in eastern Ontario, Canada, in fall compared to spring (673 g vs. 249 g CH₄ lactating $cow^{-1} d^{-1}$) were attributed to greater fall temperature (VanderZaag et al., 2014). In addition, physical changes such as snow accumulation and crust formation in liquid manure storage add complexity as to how temperature influences CH4 emissions from manure during different seasons.

Given the recognized importance of CH_4 emissions from liquid manure systems to the carbon footprint of milk, studies have measured CH_4 fluxes from dairy manure, stored as liquid on commercial farms in cold climates (Husted, 1994; Kaharabata et al., 1998; VanderZaag et al., 2011; Minato et al., 2013; VanderZaag et al., 2014). However, most studies were not conducted year-round but rather ranged in duration from six to eight months. Exceptions are the study by Husted (1994) who measured CH_4 emissions at two-week intervals over 1 year using a chamber method, and Baldé et al. (2016) who used the backward Lagrangian stochastic dispersion technique over a two-year period. Although the study by Baldé et al. (2016) was conducted on a commercial farm in a cold climate, the dairy manure studied was the liquid fraction after solid-liquid separation, and therefore there is a need for further study of emissions from farm-scale storage of unseparated (or raw) manure.

Year-round, continuous and farm-scale measurements are necessary to capture seasonal changes in environmental factors and the associated CH_4 emissions from stored liquid dairy manure. Additional measurements are also recommended by IPCC (2006) to reduce uncertainties in emission factors used for calculating GHG emissions. In the IPCC (2006)

WT - Wind tower

model the CH₄ emission factor is calculated based on volatile solids (VS) excreted per animal per year, maximum CH₄ producing capacity (B₀, m³ CH₄ kg⁻¹ VS excreted) and a prescribed CH₄ conversion factor, depending on region. For liquid dairy manure, the CH₄ conversion factor is based on the Arrhenius temperature response model ('f factor) and storage time (USEPA model by Mangino et al., 2001). Comparisons have shown the USEPA temperature model underestimated emissions from swine manure (Park et al., 2006), overestimated emissions from dairy manure in winter and spring (VanderZaag et al., 2011) and overpredicted CH₄ emissions in cool months but underpredicted CH₄ emissions in warm months from solid-separated liquid dairy manure (Baldé et al., 2016). To better understand the temperature response of CH₄ emissions it is important to further assess this model with yearround, continuous measurements for untreated dairy manure.

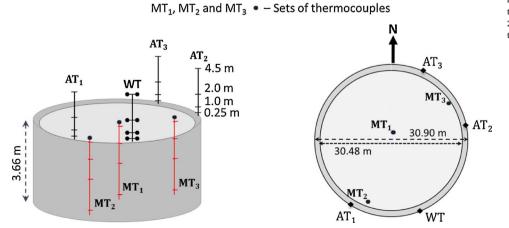
The objectives of this study were: 1) to quantify CH_4 emissions from liquid dairy manure stored in a circular-concrete tank on a commercial farm in Ontario, Canada (annual average temperature < 10° C) and capture its annual variability, 2) to investigate the interaction effect of time of year and temperature on CH_4 emissions, and 3) to compare predictions derived from the USEPA temperature model with measured CH_4 emissions. A micrometeorological mass balance method was used in measuring emissions as it provides continuous and integrated measurements from heterogeneous sources and does not interfere with gas exchange processes between the surface source and the atmosphere (Denmead et al., 1998).

2. Methodology

2.1. Experimental site and manure management

Methane fluxes from a manure storage tank at a commercial dairy farm in Drayton, Ontario, Canada (43° 45' 18.22" N 80° 40' 16.60" W) were measured from Aug 2010 to early Nov 2011. This storage tank received a portion of the manure from a Holstein dairy cow herd comprised by an average of 127 lactating cows, 44 dry and transition cows and 45 heifers (2-10 months). Liquid manure stored elsewhere on the farm was not monitored. The manure from 15 calves (0-2 months) was composted offsite, thus was not considered in this analysis. Details on production, feeding and manure management are given in Ngwabie et al. (2014). Briefly, manure collection was carried out mechanically every 4 h by scraping the concrete floor of the barn where cows and heifers were housed, into a covered pit at the end of the barn. The collected manure was periodically pumped into an outdoor-circular, concrete storage tank (diameter of 30.48 m and depth 3.66 m, Fig. 1). The tank was relatively full just before this study started and about 548 m³ of manure from the under-barn storage was added on July

> Fig. 1. Wind and air sampling tower positions around the manure tank and temperature profile measurement with depth at three locations. Manure temperature was measured at depths of 0.05, 1 and 2 m from the manure surface and 0.50 m from the tank bottom. Left – Side view; Right – Top view.



 AT_1 , AT_2 and AT_3 – Air sampling towers

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