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# Biofiltration of methane from cow barns: Effects of climatic conditions and packing bed media acclimatization



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# ABSTRACT

The performance of biofiltration to mitigate CH<sub>4</sub> emissions from cow barns was investigated in the laboratory using two flow-through columns constructed with an acclimatized packed bed media composed of inexpensive materials and readily available in an agricultural context. The biofilters were fed with artificial exhaust gas at a constant rate of 0.036 m<sup>3</sup> h<sup>-1</sup> and low inlet CH<sub>4</sub> concentration (0.22 g m<sup>-3</sup> = 300 ppm). The empty-bed residence time (EBRT) was equal to 0.21 h. Additionally, in order to simulate temperature changes under natural conditions and determine the influence of such cycles on CH<sub>4</sub> removal efficiency, the upper part of the biofilters were submitted to temperature oscillations over time. The maximum oxidation rate (1.68  $\mu$ g <sub>CH4</sub> g<sup>-1</sup><sub>dw</sub>h<sup>-1</sup>) was obtained with the commercial compost mixed with straw. Accordingly, it was considered as packing bed media for the biofilters. The CH<sub>4</sub> removal efficiency was affected by the temperature prevailing within the biofilters, by the way in which the cooling-warming cycles were applied and by the acclimatization process. The shorter the cooling-warming cycles, the more oxidation rates varied. With longer cycles, CH<sub>4</sub> removal rates stabilized and CH<sub>4</sub> removal efficiencies attained nearly 100% in both biofilters, and remained at this level for more than 100 days, irrespective of the temperature at the top of the biofilter, which was – at times – adverse for microbiological activity. The first order rate constant for  $CH_4$  oxidation kinetics of the entire system was estimated at 15 h<sup>-1</sup>. If such rate could be transposed to real field conditions in Canada, home to nearly 945,000 dairy cows, biofiltration may be applied to efficiently abate between  $2 \times 10^6$  and  $3 \times 10^6$  t yr<sup>-1</sup> of CO<sub>2</sub> equivalent (depending on how estimates are performed) from bovine enteric fermentation alone.

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### 1. Introduction

Global livestock agriculture was responsible for 12–18% (5.2– 7.9 Pg CO<sub>2</sub> eq year<sup>-1</sup>) of the anthropogenic greenhouse gas (GHG) emissions annually (Ecofys, 2016; IPCC, 2014; Shafer et al., 2011) and agricultural emissions of GHGs could increase to 7.9– 8.5 Pg CO<sub>2</sub> eq year<sup>-1</sup> by 2050 (Shafer et al., 2011). According to USEPA (2012), approximately 37% of global agricultural methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) arise from direct animal and manure emissions. Enteric CH<sub>4</sub> comprises 17 and 3.3% of global CH<sub>4</sub> and GHG emissions, respectively, and is largely derived from ruminant livestock (Ecofys, 2016; USEPA, 2012, 2010). CH<sub>4</sub> is a powerful GHG, with an estimated global warming potential of 28–36 times higher than that of carbon dioxide (CO<sub>2</sub>), over 100 years (Myhre et al., 2013; USEPA, 2017).

In Canada, emissions associated with the agriculture sector accounted for 8% of the country's total GHG emissions in 2015  $(0.06 \text{ Pg CO}_2 \text{ eq year}^{-1})$ , 28% of which were CH<sub>4</sub> emissions. Emissions from enteric fermentation accounted for 42% (0.025 Pg CO<sub>2</sub> eq year<sup>-1</sup>) of total GHG emissions associated with the agriculture sector in the country (Government of Canada, 2015). The dairy sector is the third most important farming sector in Canada (Government of Canada, 2017a). The dairy industry is concentrated in the central region of Canada, namely Quebec and Ontario, with 82% of Canada's dairy farms (Government of Canada, 2017a). Of the total GHG emissions of the Province of Quebec, 40.8% are attributed to bovine enteric fermentation (MDDEP, 2014).

On Quebec's dairy farms, cows remain confined to barns during the winter. Inside a typical barn, the air exchange occurs at a rate of 6–7 times every hour to maintain a high-quality environment for



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the animals. This leads to high air exchange rates, with exhaust gas containing very low CH<sub>4</sub> concentrations. Canada wants to reduce footprint emissions and is therefore looking for viable alternatives (Government of Canada, 2015; MDDEP, 2014).

The main CH<sub>4</sub> emissions reduction strategies studied within the animal husbandry context are: (1) robust ecologically-based management practices and technologies; (2) best feeding management and nutrition; (3) use of rumen modifiers; and (4) increasing animal production through genetics and other management approaches (Knapp et al., 2014; Shafer et al., 2011). As far as mitigation of GHG emissions is concerned, biofiltration is a technique that has been commonly applied in agricultural and industrial sectors (Akdeniz et al., 2011), but has received relatively less attention when it comes to abatement of CH<sub>4</sub> emissions in animal houses. Typical CH<sub>4</sub> concentrations inside animal houses range between 5 and  $100 \text{ mg m}^{-3}$  (milking cow). The average ventilation rate is 1000 m<sup>3</sup> h<sup>-1</sup> (Melse and Werf, 2005). One important difficulty in using biofilters for CH<sub>4</sub> biotic oxidation is related to such high air exhaust rates, because it requires long residence times and very large biofilters (Melse and Werf, 2005; Schmidt et al., 2004). Inside the biofilter, methanotrophs are able to oxidize the CH<sub>4</sub> under aerobic conditions, while generating oxidation by-products such as water (H<sub>2</sub>O) and carbon dioxide (CO<sub>2</sub>). Their activity depends on the presence of sufficient concentrations of both CH<sub>4</sub> and O<sub>2</sub>, and is therefore limited in their distribution inside of the biofilter by diffusion of CH<sub>4</sub> and O<sub>2</sub> (De Visscher et al., 1999; Scheutz and Kjeldsen, 2004, 2003). The biofilter internal bed temperature has a profound influence on the methanotrophic activity in oxidizing CH<sub>4</sub>. Most methanotrophs are mesophiles, whose optimum operating temperatures lie between 25 and 35 °C (Boeckx and Cleemput, 1996; Scheutz et al., 2009; Scheutz and Kjeldsen, 2004), although methanotrophic communities have the capability of adapting to temperatures varying between 0 and 55 °C (Einola et al., 2007; Humer and Lechner, 1999; Scheutz et al., 2009). Temperature influences not only biotic activity; it also affects CH<sub>4</sub> and O<sub>2</sub> diffusion coefficients (Delhoménie and Heitz, 2005; Gómez-Borraz et al., 2017).

One may expect that under severe climatic conditions, such as observed in Canadian winters,  $CH_4$  oxidation in biofilters constructed to treat cow barn exhaust gas, would come to a halt.

Considering the conditions prevailing in dairy cow barns in Canada (high exhaust rates and low  $CH_4$  concentrations), it is essential to assess the influence of temperature variation cycles in the efficiency of biofilters to mitigate  $CH_4$  emissions. The purpose of this study was to verify the validity of the hypothesis that, given proper acclimatization of the packing bed media, large biofilters constructed with common farm materials can sustain biotic  $CH_4$  oxidation under typical Canadian dairy farm conditions, even under adverse temperature conditions for biofiltration.

#### 2. Materials and methods

#### 2.1. Selection of packing bed media

Commercial compost (comm-comp), sawdust (swd), straw (stw), manure compost (man-comp) and woodchips (wd-chp) were tested. Laboratory experiments were performed by mixing these materials at different ratios: (a) comm-comp/swd/stw (1:1:1 v/v); (b) comm-comp/stw (1:1 v/v); (c) comm-comp/swd (1:1 v/v); (d) comm-comp/swd (1:2 v/v); (e) comm-comp/swd (2:1 v/v), (f) man-comp/wd-chp (1:1 v/v); and (g) man-comp/ wd-chp/stw (1:1:1 v/v).

The final selection of the packing bed media for the biofilter was based on  $CH_4$  oxidation rates obtained during short-term activation tests. The latter were carried out over a period of 6 weeks, in 18.9-L buckets filled with 5 L of the tested media.  $CH_4$  loading

(injection of 10 mL of pure CH<sub>4</sub>) was performed twice a week. For the determination of oxidation rates, gas samples were taken immediately after loading and 3 h later. The CH<sub>4</sub> concentrations were then obtained using a 3000A gas chromatograph (Agilent Technologies). Both CH<sub>4</sub> loading and samples collection were performed using syringes. The moisture content of the packing media tested ranged from 43% to 64% and the density ranged from 0.3 g cm<sup>-3</sup> to 0.5 g cm<sup>-3</sup>.

## 2.2. Acclimatization process

The same experimental set-up used for the packing bed media selection was adopted for the acclimatization process.  $CH_4$  was loaded periodically and the  $CH_4$  concentrations within the buckets were monitored over time to ensure that the samples were continually exposed to its presence. Acclimatization was performed in duplicate prior to each of the three subsequent biofilter tests (described below). The duo acclimatization biofilter test forms what is referred herein as a biofilter set.

For Sets A and B, the  $CH_4$  initial loading increased with time (from 200 mL to 3000 mL of pure  $CH_4$ ), while for Set C the  $CH_4$  loading remained constant (1000 mL of pure  $CH_4$ ). One important aspect of the acclimatization process is that from Sets A to B and B to C, 50% of the packing bed used in one set was reused to build the biofilters of the following set. In addition, the lids of the buckets were opened periodically to allow proper aeration of the samples. The acclimatization process lasted approximately one month for each set.

#### 2.3. Biofiltration tests

Flow-through column experiments were performed in duplicates to reproduce biofilters operating under the winter conditions of a typical cow barn containing 150 cows. As shown in Fig. 1, the 11.8-L Plexiglas<sup>®</sup> columns were filled with 7.6 L of the selected packing bed media. In the reduced scale of the laboratory, the modelled biofilters were fed with a constant exhaust gas rate equal to 0.036  $\text{m}^3 \text{h}^{-1}$  and the inlet CH<sub>4</sub> concentration equal to 0.22 g m<sup>-3</sup> (or 300 ppm; personal communication with Daniel Massé - Agriculture and Agri-Food Canada). This exhaust rate  $(0.036 \text{ m}^3 \text{ h}^{-1})$ was calculated based on the following premises: a minimum ventilation rate equal to 1000 m<sup>3</sup> day<sup>-1</sup> per cow (Turnbull and Huffman, 1988; Table 1) and a very large biofilter (1300 m<sup>3</sup>, was our preliminary design value). The latter premise was based on Melse and Werf (2005), who concluded that very large biofilters are necessary to abate CH<sub>4</sub> emissions from animal houses. Adopting these values resulted in an empty-bed residence time (EBRT) equal to 0.21 h. The responses of the biofilters were monitored during three relatively long testing periods. Set A was carried out from May to December 2013, Set B from March to November 2014 and Set C from March to July 2015.

To determine the influence of temperature cycles on the efficiency of the biofilters to abate  $CH_4$  emissions, we submitted the biofilters to temperature oscillations simulating natural cycles undergone by biofilters exposed to winter conditions. The cooling system consisted of copper tubing wrapped around the exterior of the Plexiglas<sup>®</sup> column and connected to a temperature-controlled bath (constant temperature circulator – Polystat<sup>®</sup>). Only the upper part of the biofilter was cooled to simulate a condition whereby frost penetrates to a certain depth. The temperature of the bath was controlled throughout the experimental period, leading to variable temperature gradients within the biofilters. Thermocouples allowed monitoring of the temperature of the packing bed media at three different heights, 5 cm, 20 cm and 40 cm from the base of the biofilter.

The CH<sub>4</sub> loading rate was controlled by a flow meter, whereas the inlet and outlet gas concentrations were monitored using an

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