

**Manitoba Sustainable Agriculture Practices Program (MSAPP)
Research & Development Projects
2009-2010**

Final Report

**Biofilters for Mitigating Methane Emission
from Covered Hog Manure Storage**

Submitted by

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April, 2010

Objective / Background

This project is the preliminary phase of designing and testing biofiltration systems for removing greenhouse gas (GHG) emissions from covered hog manure storage. The specific objectives of this phase are: (1) to conduct a thorough literature search to compile information on the use of biofilters in mitigating GHG emissions from livestock operations; and (2) to assess the technical feasibility of using biofilters in mitigating GHG emissions from covered hog manure storage based on the literature review; and (3) to conceptualize a biofiltration system design for covered hog manure storage typical in Manitoba.

Results & Observations

Part 1. Literature Review

Executive Summary

Liquid manure storage may contribute to methane (CH₄) emission and this emission can be greatly reduced if appropriate management practices are applied. Biofiltration has been used in other fields for mitigating GHG emission (e.g., landfill) and shown promise for mitigation CH₄ emission from liquid manure storage.

For covered liquid manure storage, the methane concentration in the headspace varied depending on the air-tightness of headspace. In theory, the headspace concentration equals to undiluted biogas that contains about 425 gm⁻³ (65% v/v) if the headspace is completely sealed (airtight). In practice, however, the cover is seldom completely airtight, which results in methane concentrations ranging from 0.1 to 20 gm⁻³. A large variation was observed among the published CH₄ emission rates, ranging from 0.0007 to 0.64 m³m⁻²day⁻¹ or 0.2 to 202.74 g m⁻²day⁻¹.

Biofiltration is one of the promising techniques that are capable of reducing 80% of GHG emissions from manure storage. The CH₄ removal efficiency is influenced by many factors, including CH₄ and O₂ concentrations, temperature, moisture, composition of the filter bed, nutrient, and empty bed residency time (EBRT).

Biological conversion of methane in a biofilter is a slow process due to the low water solubility of methane. The residence times (EBRT) between 5 min and 5 h have been used,

whereas a typical EBRT of 25 s is used for common biofilter applications. Temperature at which methanotrophic bacteria are active ranges from 10°C to 45°C. The maximum activity is found at around 30°C. The optimal filter bed water content depends on both the gas flow rate and the type of filter bed (soil, compost, etc.) and ranges from 30-70% of the water holding capacity. Compost is the best material for filter bed. The optimal pH for methanotrophic bacteria is neutral to slightly acidic. Copper and Nitrogen compounds especially nitrate are reported important nutrients to methanotrophic bacteria but their optimal concentrations have not been founded. Phosphorus and other elements such as potassium and manganese are reported to affect the performance of methanotrophic bacteria but need further confirmation.

GHG emissions in the agricultural sector

In Canada, the agricultural sector is responsible for about 10% of the production of Greenhouse Gases (GHG). Among those gases, the nitrous oxide (N₂O) contributes 61% of all N₂O emission in Canada, methane (CH₄) contributes 38% and carbon dioxide (CO₂) contributes less than 1%. In the agricultural sector, the main sources of emissions are from the bacterial activity in a ruminant's digestive system (55%), from soil (24%) and from manure (21%) (Daniel Massie 2006). Therefore, the manure management system is one of the primary sources of greenhouse gas (GHG) emissions from the agricultural sector (Patty et al. 2005; Cole et al. 1997). It is possible to reduce methane (CH₄) emission from manure storage systems by up to 25 to 80% using appropriate manure management and treatment practices (Cole et al. 1997). To achieve the goal of reducing the net GHG emission to 6% of 1990 level by 2008-2012, which was committed under Kyoto Protocol, substantial efforts have been made to establish and practice appropriate methods for manure management by local and federal governments in Canada.

About 14% of livestock operations in Canada use liquid manure storage systems. Among liquid manure storage facilities, various manure storage tanks and liquid manure storages are the primary sources (Statistics Canada 2003) and considered to be responsible for significant portions of the overall CH₄ emission from manure storages. In recent years, covering liquid manure storage or manure storage tanks is advocated to be an effective way of preventing odor and GHG emission. MSAPP BMP 1201 states: "Covering liquid manure storage facilities can be an effective way to reduce GHG emissions. Stored manure emits methane (CH₄), a potent

GHG. An impermeable storage cover traps CH₄ and prevents its release. The CH₄ can be flared off to produce carbon dioxide (CO₂), a less potent GHG, or used as a source of heat for the farm". Flaring CH₄ may require expensive equipment and skills to operate. Additionally the CH₄ content of the emitted gas needs to be high enough to allow effective combustion. An alternative is using biofiltration (biofilters). A biofilter uses microorganisms (bacteria) to break down various compounds in the air when the air passes the biofilter media. The bacterial oxidation of methane is a common phenomenon that occurs in nature, such as tropical forest; grasslands; landfills cover soil; peatlands, and rice paddy soils. Removing methane by biofiltration is basically aerobic conversion of methane to carbon dioxide (CO₂) and water by methanotrophic (methane consuming) bacteria grown in the biofilter media. Despite the vast body of literature detailing the oxidation of methane in natural environments, relatively little research has been conducted on the application of methane biofilters for manure storage emissions (Daniel Massé 2006). This report summarizes the information on the use of biofilters in mitigating GHG emissions, assesses the technical feasibility of using biofilters in mitigating GHG emissions from covered hog manure storage based on the literature review, and conceptualizes a biofiltration system design for covered hog manure storages typical in Manitoba.

The release rate of methane from liquid manure storage

For covered liquid manure storages, the methane concentration in the headspace depends on air exchanges between the headspace with outside air. In theory, at an air-exchange rate of zero, the headspace concentration equals to undiluted biogas that contains about 425 gm⁻³ (65% v/v) methane (Safley and Westerman 1988, 1989; DeSutter and ham 2005). In practice, however, the cover contains some openings that permit ventilation that results in a methane concentrations ranging from 0.1 to 20 gm⁻³ (0--3%, Roland et al. 2005).

The published gas fluxes were expressed in various units. For comparison, some published results were recalculated to a standard unit. For the data expressed in volumetric flux unit, the measured fluxes of CH₄ varied greatly and ranged from 0.0007 to 0.64 m³m⁻²day⁻¹. For studies that expressed flux results on weight-based units, the measured fluxes ranged from 0.2 to 202.74 g m⁻²day⁻¹ (Table 1). A large variation was observed among the published CH₄ emission rates. Conducting a statistical variance analysis was not possible due to the different

experimental conditions in which those data were generated. The measured fluxes usually have great spatial, daily, and seasonal variations (Safley and Westerman, 1988; DeSutter and Ham, 2005). Spatial variation was observed both within the same liquid manure storage and among storages and was closely related to substrate distribution (Safley and Westerman, 1988; Wagner-Riddle et al. 2006). Greater GHG flux was observed near inlet portion of storages, where more manure was distributed than other places. The flux variation observed among several storages in one area was a result of loading rate differences among storages (Safley and Westerman, 1988). Higher loading rates often resulted in higher biological activity and therefore higher gas flux.

The flux of biogas from storages showed both daily and seasonal variations. Higher flux rates were usually observed during the day and summer, while lower flux rates were observed during the night and winter (Sharpe and Harper, 1999). The daily and seasonal variations in biogas emission rates resulted from storage temperature and wind speed variations. Higher temperature and wind speed during the day and summer would result in higher emission rates, and lower temperature and wind speed during the night and winter would result in lower emission rates (Sharpe and Harper, 1999). However, the pattern of seasonal flux variation might be altered by substrate availability for methanogenesis. DeSutter and Ham (2005) found the highest emission rate in spring, which declined during summer due to substrate limitation. The peak emission rate occurred in early June and nearly 50% of annual gas losses occurred within 30 days.

Table 1. Gases fluxes from liquid manure storage facilities

Sources	Methods	Animals	Storage system	Emission rates		Units
				CH ₄	N ₂ O	
Safley and Westerman (1988)	Gas collection float	Swine,	Liquid manure storage	0.014 ^Z -		m ³ m ⁻² d ⁻¹
		poultry,		0.35 ^Z		
Humenik and Overcash (1976)		Dairy,	Pilot scale Liquid manure storage	0.15 ^Z		
Allen and Lowery (1976)		Swine	Liquid manure storage	0.004 -		
				0.039 ^Z		
Chandler et al. (1983)		Dairy	Liquid manure storage	0.0007 -		
		poultry		0.014 ^Z		
Safley and Westerman	Floating cover	Swine	liquid manure storage	0.46 -		
		Poultry		0.64 ^Z		
				0.13		

(1989)		Swine	liquid manure storage	0.081		
Massé et al. (2003)	wet-cup gas flow meters	Dairy	Lab-scale manure slurry container	0.00106 - 0.0148 ^Y		
		Swine		0.005-0.03 ^Y		
Khan et al. (1997)	MMB	Dairy cow	Liquid manure storage	0.2 -10 ^Y		g m ⁻² d ⁻¹
Kaharabata et al. (1998)	SF6	Swine	Opened manure slurry tankers	202.74 ^Y		
Sharp and Harper (1999)	Flux-gradient	Swine	liquid manure storage	5.23 ^Y		
Harper et al. (2000)	Flux gradient	Swine	liquid manure storage		0-0.036 ^Y	
Külling et al. (2003)	Non-steady chamber	dairy	Lab-scale manure slurry container	1.17 ^Y - 1.39 ^Y	0.017 - 0.034 ^Y	
DeSutter and ham (2005)	Floating gas collection dome	Swine	Liquid manure storage	12.02 ^Y		
Wagner-Riddle et al. (2006)	MMB	Swine	liquid manure storage	1.73 - 95.04 ^Y		
Park et al. (2006)	MMB	Swine	liquid manure storage	0.40 - 90.72 ^Y		
Husted (1994)	Chamber	Swine	Lab-scale manure slurry container	0.4-35.8		g m ⁻³ d ⁻¹
Patthey et al. (2005)	Non-steady chamber	Dairy Beef	Lab-scale manure slurry container	15.96 9.76	0.101 0.017	g kg ⁻¹ (DM) ^X (3 months) ⁻¹

^Z The data was obtained through biogas amounts x 0.7.

^Y Recalculated data to uniform units.

^XDM = dry matter.

The principles of biofiltration

Biofiltration uses microorganisms to degrade and oxidize gaseous pollutants to un-harmful gases. This technique has been used for reducing odors, hydrogen sulfide and ammonia emissions emanating from farms (Daniel Massé, 2006). It has also been employed to treat air streams contaminated with benzene, toluene, ethylbenzene, and o-xylene (BTEX) (Tahraoui and

Denis Rho, 1998, Li et al., 2002), volatile organic compounds (Yoon and Park, 2002), ethanol vapor (Arulneyam and Swaminathan, 2000), hydrogen sulfide and ammonia (Chitwood and Devanny, 1997). However, the application of biofiltration in the treatment of CH₄ is relatively new. The experiment conducted by Massé (2006) indicates that biofiltration is one of the promising techniques capable of reducing GHG emissions from livestock operations, particularly methane.

Methane biofilters use methanotrophs living in porous media to oxidize CH₄ to CO₂. Methanotrophic bacteria (or methanotrophs) use methane as their energy and carbon source whereby methane is degraded to carbon dioxide and water (Hanson and Hanson, 1996). Methane oxidation by methanotrophs occurs in natural environments and can be found in many natural aerobic - anaerobic interfaces. For example, methane oxidation has been reported in tropical forests, grasslands and meadows, landfill cover soils, deserts, and agricultural soils (Nikiema et al. 2007).

A man-made biofilter is designed and constructed with the goal of exerting maximum efficiency for gas treatment. A good biofilter is a three-phase bioreactor: the filter bed constitutes the solid phase, the biofilm the liquid phase and the gaseous pollutants the gas phase (Nikiema et al. 2007). The solid biofiltration media and the liquid phase offer microorganisms surface for immobilization, nutrients and water for growth, and the space for gas exchange. The gaseous phase offers microorganism the necessary gases (CH₄ and O₂ for methane biofilter) for their survival. Therefore, biofilters favor some specific microorganisms' activities.

According to the way that gases circulate in the biofilter, the biofilter can be classified as a closed system or open system (Nikiema et al. 2007). The majority of biofilters, as used in lab-scale experiments, are closed systems, in which air supply is ensured by a forced ventilation system. Gas circulation in the biofilter can be effected from either top to bottom or conversely. In a closed biofilter, maintaining steady operational parameters is also a relatively easy, resulting in good performance. Nikiema et al (2007) summarized the performance of biofilters used for mitigation of CH₄ emission from landfills and reported methane conversion values as high as 90%.

Open systems are mostly found in landfill sites. In this case, the flow of the polluted gas in the bed proceeds upwards, while the O₂ diffuses from the ambient air into the bed (passive ventilation) (Nikiema et al 2007). The main disadvantage of this process lies in the difficulty of

controlling the operational parameters, such as temperature and moisture levels. Moreover, transfer of O₂ to the bed's lowest layers is a very important limiting factor for the overall performance (Kjeldsen et al. 1997; Gebert et al. 2001). For example, removal efficiencies of up to 60% were obtained, when the empty bed residence times (EBRT) was at least an hour, with an open biofilter installed on a landfill site (Du Plessis et al. 2003; Gebert and Groengroeft 2006a, b).

The size of the biofilter should be at a scale of at least 1 m³ of filter bed for achieving flow rates of CH₄ in the range of 0.01–2.5 m³ h⁻¹ (Straka et al. 1999; Stresse and Stegmann 2003; Haubrichs and Widmann 2006). The height of the open biofilters with passive ventilation, used for CH₄ elimination, must also be lower than 1 m (Kjeldsen et al. 1997; Boeckx and Van Cleemput 2000; Stein and Hettiaratchi 2001; Stein et al. 2001; Park et al. 2002; Tagaris et al. 2003). Open systems are usually less expensive (by at least 15%) than closed systems.

Microorganisms

Methanotrophs

Three basic steps were identified in the process of decomposing CH₄ by methanotrophs. The first step is the oxidation of CH₄ to methanol through utilizing the enzyme methane monooxygenase, MMO (Hanson and Hanson 1996; Auman and Lidstrom 2002). Then the methanol is further transformed into formaldehyde. In the final step, formaldehyde produced from the previous step is used in a dissimilatory pathway (i.e. being oxidized to CO₂, with formate as an intermediate) or via several types of assimilatory pathways, leading to the synthesis of cell components, which are necessary for the growth of methanotrophs (Hanson and Hanson 1996).

Generally, the specific bacteria responsible for the decomposition of CH₄ are named as methanotrophs. However, depending on their roles in the CH₄ decomposition process, the genera of methanotrophs are grouped into three main types: type I, type II, and type X. Type I includes genera *Methylomonas*, *Methylomicrobium*, *Methylobacter*, *Methylocaldum*, *Methylophaga*, *Methylosarcina*, *Methylothermus*, *Methylohalobius* and *Methylosphaera*. They assimilate formaldehyde by the ribulose monophosphate pathway and their cellular membranes are mainly made up of fatty acids with 16, or sometimes 14 atoms of carbon (Nikiema et al

2007). Type II includes genera *Methylocystis*, *Methylocella*, *Methylocapsa* and *Methylosinus*. They assimilate formaldehyde through the serine pathway and their cellular membranes contain fatty acids of 18 carbons. Type X includes genera *Methylococcus*. It has both properties of types I and II. Its cellular membranes fatty acids have 16 carbons and the assimilation of formaldehyde is through both the ribulose monophosphate cycle and the serine pathway (Nikiema et al 2007).

The methanotrophic communities have the capability of adapting to various environmental conditions. In terms of temperature, methanotrophs are active between 0 and 55⁰C. Some type II genera such as *Methylocystis* and *Methylosinus*, are acidophilic and exhibit a maximum growth rate in acidic media with the pH range from 5 to 5.5. *Methylomicrobium*, a type I genera, was reported being at ease in saline media having sodium chloride concentrations ranging from 0.5 to 5.6% wt/wt, and pH ranging from 7.5 to 10 (Trotsenko and Khmelenina 2002).

Methane monooxygenase enzyme (MMO)

A specific enzyme known as methane monooxygenase or MMO was reported to be the key enzyme allowing methanotrophs to perform the decomposition of CH₄ (Hanson and Hanson 1996). This enzyme exists in two forms: particulate MMO (pMMO) and soluble MMO (sMMO). The pMMO enzyme can be found in and synthesized by all methanotrophs, except *Methylocella*, but the sMMO is almost always present in bacteria of type II and X. Methanotrophs containing pMMO grow more rapidly than those having the sMMO (type II and X) (Nikiema et al 2007).

Other bacteria

In fact, methane is mainly degraded by methanotrophs. In addition to methanotrophs, some other bacteria can also decompose CH₄ under some situations. For example, nitrifying bacteria, which are responsible for the decomposition of ammonia (NH₃), can also degrade CH₄. However, their performance rate is less than 5% of the pure methanotrophic populations (Hanson and Hanson 1996; Bodelier and Frenzel 1999). Also, some bacteria involved in the decomposition of methanol are capable of degrading CH₄ if the CH₄ concentrations remain below 10% v/v.

The performance of methane biofilters

The performance of a methane biofilter is measured by a removal efficiency parameter as defined in following equation:

$$\text{Removal Efficiency } RE = \frac{C_{in} - C_{out}}{C_{in}} \times 100$$

where, C_{in} is the methane concentration of the gas at the inlet of the biofilter in ppm, and C_{out} is the methane concentration of the gas at the outlet of the biofilter in ppm. This Removal Efficiency was sometimes given different names, Conversion (X), for example (Nikiema et al 2007). Elimination capacity (EC) is another parameter that can be used to access the performance of a biofilter. It was used to assess the performance of biofilters, which were used in the mitigation of CH_4 emission from landfill sites (Nikiema et al. 2007). It was calculated using the following equation:

$$EC = IL \times \frac{X}{100}$$

where EC is elimination capacity ($g\ m^{-2}\ d^{-1}$ or $g\ m^{-3}\ d^{-1}$), IL is the inlet load ($g\ m^{-2}\ d^{-1}$), and X is Conversion (%). The inlet load (IL) is calculated according to following equation:

$$IL = \frac{C_{in} \times Q}{S}$$

where C_{in} is the CH_4 concentration of the biogas flowing into the methane biofilter in $g\ m^{-3}$, Q is the flow rate of the biogas in $m^3\ d^{-1}$, and S is the biofilter bed cross section in m^2 .

The performance of methane biofilters depends on various factors, including the biofilter type, the empty bed residence time (EBRT), and operating conditions. In mitigating CH_4 emissions from landfill sites, the closed biofilter was reported to show good performance, with CH_4 X-values as high as 90% (Dammann et al. 1999; Streese et al. 2001; Gebert et al. 2001; Du Plessis et al. 2003; Nikiema et al. 2005), in contrast to a X-value of 60% that was obtained from the open methane biofilter with an EBRT of at least an hour (Du Plessis et al. 2003; Gebert and Groengroeft 2006a, b). The best EC obtained with a laboratory-scale closed biofilter was in the

range of 325 and 400 g m⁻² d⁻¹ (Hettiaratchi and Stein 2001). In general, the biofilter eliminates some 10–100% of the CH₄ escaping from the upper layers of landfills, depending on local climatic conditions (Nozhevnikova et al. 1993; Kightley et al. 1995; Czepiel et al. 1996; Chanton et al. 1999; Christophersen et al. 2000; Bajic and Zeiss 2001; EPA 2005; Stralis-Pavese et al. 2006).

Compared with many cases of biofilters used for mitigating CH₄ emission from landfill sites, few cases were reported for using biofilters for mitigating CH₄ emission from manure storages. Masse (2007) reported that the removal efficiency reached up to 80% for the four types of media material used in the biofilter. This technology was identified as a promising method for controlling methane emissions from manure storages. Venugopal et al (2003) conducted a lab-scale experiment using a methane biofilter for mitigating CH₄ emission from a gas meter station. They report that the conversion varied with temperature, reaching 90% during the summer and dropping to 20% during the winter. Melse et al. (2005) reported a CH₄ removal rate up to 85% in a lab-scale biofilter used in the mitigation of CH₄ emissions from a liquid manure storage.

Factors affecting the efficiency of a methane biofilter

Oxygen, methane, and carbon concentrations

Methane degradation by methanotrophs requires CH₄ and O₂ as substrates. In fact, methanotrophs can be found in small quantities in any environment exposed simultaneously to significant amounts of CH₄ and O₂ (Borjesson et al. 1998; Dammann et al. 1999). However, the optimal concentrations of CH₄ and O₂ for CH₄ degradation have not been determined. Literature showed a variation in CH₄ and O₂ optimal concentrations. It was reported that type I bacteria grew better in an environment with O₂ concentration of 21% v/v, associated with a CH₄ concentration less than 1,000 ppmv, while type II bacteria develop better in an environment with CH₄ concentration above 1% v/v and O₂ concentration at about 1% v/v (Hanson and Hanson 1996; Henckel et al. 2000). However, some type I bacteria have their growth stimulated only in the presence of an appreciable concentration of CH₄ (> 1% v/v), and correspondingly, a low amount of O₂ (< 1% v/v) (Henckel et al. 2000; Erwin et al. 2005). Bender and Conrad (1994), Czepiel et al. (1996) and Stein and Hettiaratchi (2001) have shown that, by increasing the O₂ concentration from 3 to 20% v/v in the gas mixture, the CH₄ conversion varies only slightly

(less than 10%). However, a decrease of O₂ concentrations from 3% to 1% causes a decrease of CH₄ oxidation of more than 50%. In the experiments of Stein and Hettiaratchi (2001), maximal CH₄ elimination was obtained at O₂ concentrations between 0.75% and 1.6%. The presence of CO₂ in a biofilter modifies the behavior of the microorganisms. Acha et al. (2002) reported that the activity of the methanotrophs, using the serine pathway for the assimilation of formaldehyde obtained during the decomposition process of CH₄, requires some CO₂ input (partial pressure of CO₂ around 11.6 kPa).

Temperature

The temperature in which methanotrophic bacteria are active ranges from 10°C to 45°C. The maximum activity was found at around 30°C (Bender and Conrad, 1994; Whalen et al., 1990; Boeckx and Cleemput, 1996). However Priemé and Christensen (1997) observed methane oxidation to be active in temperatures as low as 1 °C in the field and 2 °C in soil cores experiments. King and Adamsen (1992) observed methane consumption at -1 °C and they suggested that methane consumption might occur at low temperatures as long as the soil water remains liquid. Summerfield et al. (1993) showed that the soil microflora was active even when the soil was snow covered and near 0 °C, and that methane consumption was taking place under these conditions.

It was reported that the conversion (X) fell by around 50% when the temperature was reduced from 30 to 20 °C or from 29 to 24 °C (Dammann et al. 1999; Streese et al. 2001). Between -5°C and 10 °C of ambient temperature, the biological elimination of CH₄ in an open biofilter system (landfill cover soil) considerably decreased, i.e. more than 80%, compared to the value at 15°C (Christophersen et al. 2000; Le Mer and Roger 2001).

Moisture content

The optimum moisture content depends on the kind of biofilter material. It ranges from 30-70 % of the water holding capacity, which is clearly lower than the optimum for odor and trace gas treatment. (Whalen et al., 1990; Bender and Conrad, 1995; Boeckx and Cleemput, 1996). Very high soil water content may impede gas diffusion and thus restrict the supply of CH₄ to the methanotrophs. The low solubility of CH₄ in water enhances this effect especially at low CH₄ concentrations (Bender et Conrad, 1995).

The optimal filter bed water content depends on both the gas flow rate and the type of filter bed (soil, compost or other material employed) (Christophersen et al. 2000). Optimal moisture content of soil materials (from the upper layers of landfills) ideally lies between 13 and 15.5% wt/wt, on a dry basis (Whalen and Reeburgh 1996; Boeckx and Van Cleemput 1996; Chiemchaisri et al. 2001b; Stein and Hettiaratchi 2001; Jackel et al. 2001; Park et al. 2002, 2004). For composts or biological residues, optimal bed moisture lies between 25% and 50% wt/wt (Humer and Lechner 1999b). See Table 1 for a summary of these studies.

Table 2. Optimal water content of Biofilter beds used for methane elimination (Nikiema et al, 2007)

Filter bed	Water content (% wt/wt)	Sources
Compost	25–50	Humer and Lechner (1999b)
Landfill cover soil	13–30	Boeckx and Van Cleemput (1996), Park et al. (2002), Stein and Hettiaratchi (2001), Visvanathan et al. (1999), Giani et al. (2002)
Meadow soil	30–50	Mingxing and Jing (2002)
Woodland soil	18–33	Mingxing and Jing (2002)
Various soils	11–35	Bender and Conrad (1995), Christophersen et al. (2000)

pH

It is generally suggested that a neutral or slightly acidic medium be maintained for a methane biofilter (Bender and Conrad, 1995). Nikiema et al (2007) suggested that the pH of the filter bed is a parameter of lesser importance because the biodegradation of CH₄ does not generate intermediate or final products capable of significantly influencing the pH. The optimal

pH values for the oxidation of CH₄ are in fact the same as those promoting the growth of the majority of methanotrophic bacteria.

According to Hanson and Hanson (1996), methanotrophs are neutrophiles but can tolerate pH from 5.5 to 8.5. Bender and Conrad (1995) suggested that the optimum pH ranges between the values of 6.7 and 8.1 for the soil-based filter beds, while Le Mer and Roger (2001) suggest the range lies between 5 and 6.5 for peat.

Filter bed

The filter bed is the solid phase of the biofilter. A good filter bed provides sufficient space for the development of microorganisms and also has a texture providing a great moisture holding capacity, in addition to appropriate bacteriological and mechanical properties (Nikiema et al 2007). Various materials, which are generally classified as soils, composts, and other materials or combinations, have been used as the filter bed media and tested by researchers (Nikiema et al 2007). Compost, made from mature yard wastes yielded the best results with Elimination Capacity (EC) up to 590 g m⁻² d⁻¹ and at values for Conversion rate (X) of between 90 and 100%, during more than 100 days of continuous filter operation (Haubrichs and Widmann 2006). Compost, made from dead leaves, also yielded good results (Hettiaratchi and Stein 2001; Wilshusen et al. 2004). In addition, the time required to reach 100% conversion is less for the mature compost than that for freshly generated compost, 15 and 55 days respectively. This result suggests that mature compost is a preferred media for the biofiltration of CH₄ (Humer and Lechner 1999b). Agricultural soils, soils derived from mountains, forests and rice plantations, peat bogs and swamps, have also been tested for CH₄ biofiltration (Dobbie and Smith 1996; Hutsch 1998b; Del Grosso et al. 2000; Hettiaratchi et al. 2000; Cai and Mosier 2000; Nozhevnikova et al. 2001; Stein and Hettiaratchi 2001; Novikov and Stepanov 2002; Kravchenko 2002). All of these soils contain different proportions of sand, clay, silica and organic matter. The most effective soils for CH₄ elimination are those taken directly from the upper layers of landfill covers. An EC of 435 g m⁻² d⁻¹, corresponding to an X value of greater than 80%, has been reported in the literature (Park et al. 2002). The addition of organic residues to soil, such as vegetable residues (beet leaves, wheat straw), clarifier sludges or composts, can improve its CH₄ elimination. The EC values, reported from these modifications (100–200 g m⁻² d⁻¹), correspond to some 40–100% of CH₄ conversion, and remain below the EC obtained

during similar experiments with compost-based beds (Borjesson et al. 1998; De Visscher et al. 1999; Humer and Lechner 1999b; Park et al. 2002). The mean size of the soil particles must preferably lie between 0.5 and 2 mm (Bender and Conrad 1995; Kightley et al. 1995; Borjesson et al. 1998; Hettiaratchi et al. 2000; Min et al. 2002). Indeed, when particle sizes are less than 0.02 mm, the bed tends to become packed, preventing the effective diffusion of pollutants in the gas phase and then negatively affecting the conversion (Bender and Conrad 1995; Le Mer and Roger 2001; Min et al. 2002). With either synthetic or inert filter materials, a few interesting results were obtained during CH₄ biofiltration. In an experiment, involving biofiltration by percolation with glass particles (Sly et al. 1993), for a residence time of 20 min and an IL of around 200 g m⁻² d⁻¹, more than 95% of CH₄ conversion was achieved. The highest EC reported in the literature is 700 g m⁻² d⁻¹, obtained by Nikiema et al. (2004b) during their experiments with an inorganic-packed bed biofilter of 0.018 m³, the gas flow rate being 6 m³ d⁻¹ and the CH₄ concentration maintained at between 7,000 and 7,500 ppmv.

Nutrients

Nutrients such as copper, nitrogen and phosphorus are necessary for the growth of microorganisms and therefore are factors that affect the performance of a biofilter (Trotsenko and Khmelenina 2002). These nutrients are supplied to the microorganisms through a mix with water used to humidify the filter bed (Nikiema et al. 2005).

- ***Copper:*** It affects bacterial growth, however the threshold concentrations have not yet been determined. Hanson and Hanson (1996) demonstrated that while copper inhibits the sMMO enzyme at concentrations above 1 μmol L⁻¹, it supports the synthesis of the pMMO at concentrations between 1 and 5 μmol L⁻¹. Thus, by adjusting the bed copper concentration, it is possible, in various cases, to develop a medium rich in bacteria of types I or II (Wise et al. 1999; Erwin et al. 2005). It has also been noted that, in adding around 0.02 g of copper, in the form of CuCl₂, per kg of paddy soil, CH₄ oxidation is slightly stimulated (an increase of around 5%) (Mohanty et al. 2000).
- ***Nitrogen compounds:*** Nitrogen is usually provided to microorganisms in an inorganic form: e.g. nitrate (NO₃), ammonium (NH₄) or nitrite (NO₂) ions. Many research studies have been performed to determine the effect of each of these compounds on methanotrophs. The sources of NH₄ most frequently tested are ammonium chloride, ammonium sulfate and urea.

For NO_3^- , sodium nitrate and potassium nitrate are the most studied. On some occasions, ammonium nitrate was used as a nitrogen source (Kightley et al. 1995; Hettiaratchi et al. 2000).

The NH_4^+ was considered to have a competition with CH_4 when it was provided as a nitrogen source (Mancinelli 1995; Boeckx and Van Cleemput 1996; Humer and Lechner 1999b; Sitaula et al. 2000; Novikov and Stepanov 2002). In addition to oxidizing methane, methanotrophs can convert NH_4^+ to NO_2^- . Novikov and Stepanov (2002) reported that 12–28% of the methanotrophic population was dedicated to a nitrification step instead of the CH_4 oxidation. The inhibitory effect of NH_4^+ could be minimized if higher CH_4 concentrations were continuously provided to the filter media.

Table 3. Studies investigating the effect of N on the work of CH_4 biofilters (Nikiema et al 2007)

Sources	Filter beds	N forms & Concentration	effect
Hettiaratchi et al. (2000)	Soil	25 mg N per kg soil in the form of NH_4^+ or NO_3^- .	improvement of CH_4 elimination by 100%
Chiemchaisri et al. (2001a)	Soil	≥ 30 mg N per kg soil in the form of NH_4^+ or NO_3^-	inhibit CH_4 elimination
Bronson and Mosier 1994; Cai and Mosier 2000; Hettiaratchi et al. 2000; Novikov and Stepanov 2002; Park et al. 2002		10–200 mg N– NH_4^+ kg soil ⁻¹	inhibit CH_4 elimination, extends depends on the type of soil
Nikiema et al. (2005)	Inorganic filter material	sodium nitrate, from 0.14 to 0.75 g N l ⁻¹ sodium nitrate > 0.75 g N l ⁻¹	5 times increase in the EC (from 130 to 700 g m ⁻² d ⁻¹). decrease of the CH_4 oxidation conversion

Boeckx and Van Cleemput 1996; Park et al. 2002	soil	25 ---100 mg N-NO ₃ ⁻ kg soil ⁻¹	No CH ₄ elimination effect
Kumaraswamy et al. 2001	soil	2,500 mg N-NO ₃ ⁻ kg soil ⁻¹	Inhibit CH ₄ elimination

NO₃⁻, instead of NH₄⁺, is the preferred source of fixed nitrogen for methanotrophs (Mancinelli 1995) and can improve CH₄ elimination (Le Mer and Roger 2001).

Nitrite is well known as an inhibiting compound for methane elimination by methanotrophs (King and Schnell 1994; Mancinelli 1995; Boeckx and Van Cleemput 1996; Hanson and Hanson 1996). This compound can be generated when incomplete nitrification processes occur in the filter media (Dunfield and Knowles 1995; Kravchenko 2002).

- **Phosphorus:** Generally speaking, phosphorus is of universal importance in promoting the growth of bacteria. However, few documents have been found in clarifying P effect on CH₄ elimination. Several scientists (Kightley et al. 1995; Hettiaratchi et al. 2000; Le Mer and Roger) reported that the addition of 0.1 g P-K₂HPO₄ nutrient per kg soil did not result in any noticeable effect on promoting CH₄ elimination. Thus, the role of phosphorus in CH₄ elimination remains unclear and further investigations would be needed (Nikiema et al 2007).
- **Other elements:** Potassium sulfate or manganese oxide increases the oxidation of CH₄ (Kumaraswamy et al. 2001). Addition of lime provides a soil-based bed with a neutral pH and thus appears to be interesting for CH₄ biofiltration (Hilger et al. 2000b). Excessive concentrations of sodium chloride and potassium chloride are both CH₄ elimination inhibitors (Cai and Yan 1999; Kravchenko 2002; Gebert et al. 2003), probably due to their osmotic effects.

Inoculation and incubation

When contact is created between methanotrophs and CH₄ in a biofilter, an induction step, during which X is weak (0–10% of the steady state conversion), always precedes the optimal system functioning. This lag phase is due to the activation and growth of the

methanotrophic bacteria (Bender and Conrad 1995; Henckel et al. 2000) and its duration is determined by the operating conditions (CH_4 concentration, temperature and moisture of the filter bed). During the experiments carried out by Henckel et al. (2000), in which microcosms were maintained under a CH_4 continuous flow environment, some 6 and 19 days were required to reach steady X, for high (10,000 ppmv) and low (1,000 ppmv) CH_4 concentrations, respectively. In order to aid the establishment of the specific and competitive methanotrophic population in the filter bed, inoculation of the bed by selected methanotrophic bacteria is usually performed, even if the success of this practice is not guaranteed. At the laboratory scale, another common practice involves incubation, consisting of a prolonged exposure (several days or weeks) of the filter bed to significant CH_4 concentrations, ranging between 1,000 and 200,000 ppmv. The higher the CH_4 concentration, the more the growth of the methanotrophs is promoted. The consequence then is a rapid increase in the oxidation rate (Bender and Conrad 1995; Hanson and Hanson 1996; Henckel et al. 2000; Le Mer and Roger 2001; Mor et al. 2006). For example, the oxidation rate for CH_4 at initial concentration of 100,000 ppmv is around $0.8 \text{ g CH}_4 \text{ kg soil}^{-1} \text{ d}^{-1}$, which is 10 times higher than the value observed for initial CH_4 concentration of 10,000 ppmv (Bender and Conrad 1995). Since all bacteria do not develop within the same range of CH_4 concentrations, the choice of the incubation parameters must be made judiciously. At the end of the induction phase, a peak value in the conversion of up to 3 times compared to that obtained for a steady operation (e.g. $X = 64\%$) can be noted (Hettiaratchi and Stein 2001; Abichou et al. 2006a).

Empty Bed Residence Time (EBRT)

Biofilters have been developed and operated for landfill gas treatment at various methane inlet concentrations up to 260 g m^{-3} (40% v/v) at empty bed air residence times (EBRT) between 5 min and 5 h, whereas typical EBRT are 25 s to over a minute for common biofilter applications (Melse and Van De Werf 2005).

Biological conversion of methane in a biofilter is a slow process due to the low water solubility of methane (Henry's law constant is $1.5 \times 10^{-3} \text{ M atm}^{-1}$), and this is why such long EBRT are applied. Previous work by Streese and Stegmann (2003) showed first-order removal kinetics for methane inlet concentrations up to $16 \text{ g of CH}_4 \text{ m}^{-3}$ in an operated biofilter.

Summary and Conclusions

1. For covered liquid manure storage, the methane concentration in the headspace varied depending on the sealed condition of the headspace. In theory, the headspace concentration equals undiluted biogas that contains about 425 gm^{-3} (65% v/v) if the headspace is completely sealed. In practice, however, the cover seldom completely airtight, which results in methane concentrations ranging from 0.1 to 20 gm^{-3}
2. Biofiltration is one of the promising techniques that are capable of reducing 80% of GHG emissions from manure storage. The CH_4 removal efficiency is influenced by many factors, including CH_4 and O_2 concentrations, temperature, moisture, composition of the filter bed, nutrient, empty bed residency time (EBRT).
3. Biological conversion of methane in a biofilter is a slow process due to the low water solubility of methane. The residence times (EBRT) between 5 min and 5 h have been used, whereas a typical EBRT of 25 s is used for common biofilter applications.
4. The temperature in which methanotrophic bacteria are active ranges from 10°C to 45°C . The maximum activity was found at around 30°C .
5. The optimal filter bed water content depends on both the gas flow rate and the type of filter bed (soil, compost or other material employed) and ranges from 30-70 % of the water holding capacity.
6. Compost is the best material for filter bed. The optimal pH for methanotrophic bacteria is neutral to slightly acidic. Copper and Nitrogen compounds especially nitrate are reported a important nutrient to methanotrophic bacteria but their optimal concentration has not been founded. Phosphorus and other elements such as potassium and manganese were reported to affect the performance of methanotrophic bacteria but need to further confirmation.

Part 2. Conceptual Design of CH₄ Biofilter for Covered Manure Storage

A conceptual model was developed for applying a biofilter to mitigating GHG emission from two types of covered manure storage configurations that are used in Manitoba: negative air pressure cover (NAPC) and heavy-gage plastic cover which allow gas pressure to build up under the cover (fig. 1). In a NAPC system, small pumps (fans) are running continuously to create a negative pressure under the cover. But these pumps are not capable of delivery the required pressure to push air through the biofilter media. Therefore, a dedicated biofilter pump (fan) is necessary. Valve 2 may be opened to allow fresh air to be drawn into the system if the negative pressure becomes too high, or if additional oxygen is required for the biofilter. During winter months, there may not be any biogas production, and therefore, Valve 1 may be closed and Valve 2 is opened to bypass the biofilter.

When the system is used for a non NAPC cover, where positive pressure may build up under the cover, a pressure sensor is installed to monitor the pressure under the cover and controls the Valve 1, When there is sufficient pressure accumulated under the cover, Valve 1 is opened (activated by the pressure sensor) and biogas flows into biofilter. Otherwise, this valve remains closed and the biofilter pump is stopped, and no biogas flows out of manure storage.

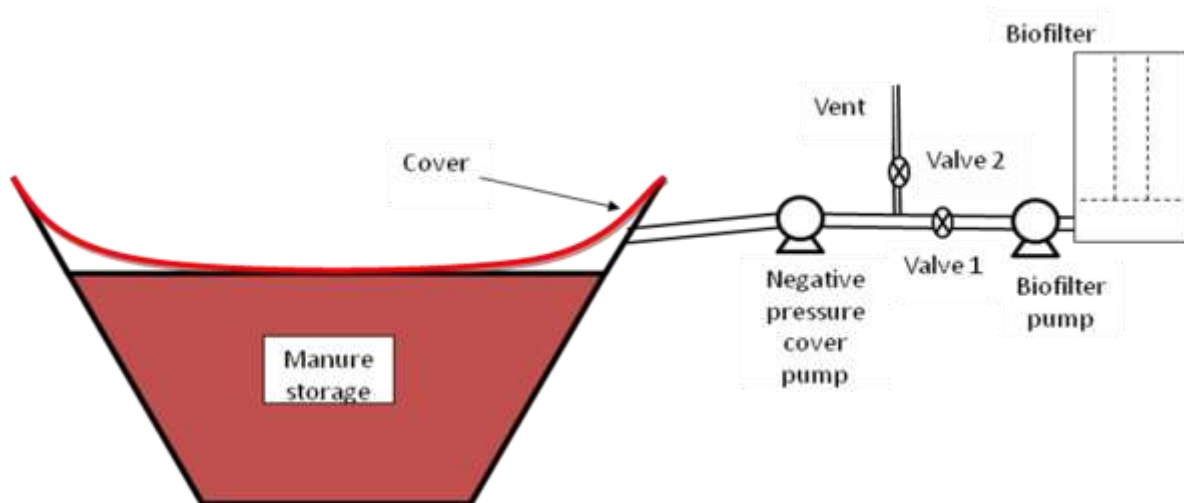


Figure1. The schematic of biofiltration system for covered manure storage

A horizontal airflow biofilter is proposed for its low airflow resistance (fig.2). The filter is a hollow heart cylinder. Biogas flows into the inner cylinder core and passes through the cylinder wall (filter media). The filter bed is composed of a mixture of compost and wood chips (50:50 % V). A perforated pipe network is embedded into the filter for supplying water, nutrient, and oxygen. Water content of the biofilter should be maintained at 30% by weight. Nutrients (N, P, Cu etc.) should be supplied with water. Whenever the pipe network is not used for supplying water and nutrients, it will be used for supplying oxygen through pumping fresh air into it. No heating system is applied to the designed biofilter.

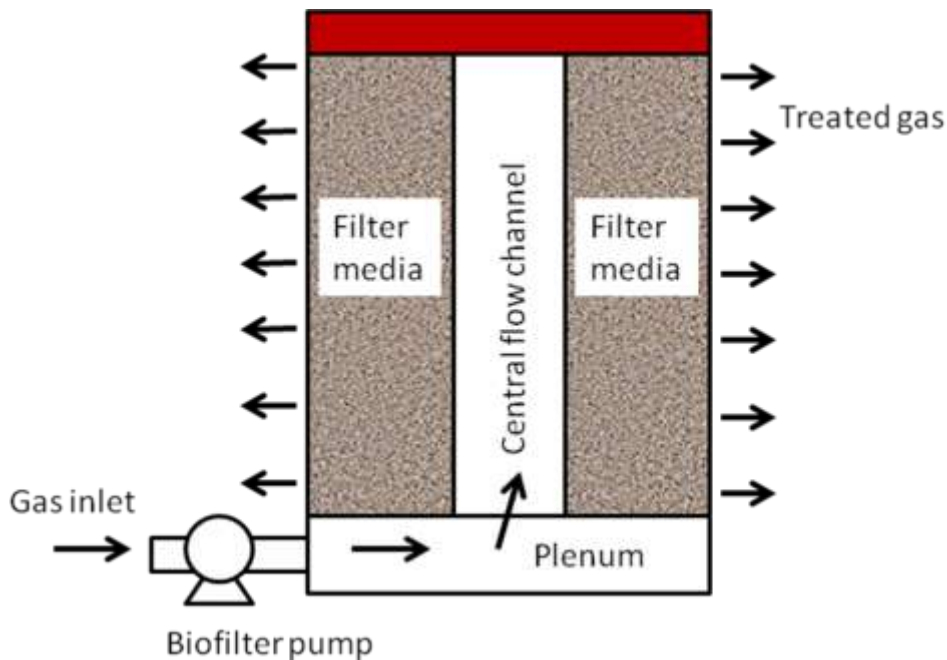


Figure 2. The schematic of the designed biofilter

The following is a sample calculation for a typical hog manure storage with a volume of 1000 m^3 and biogas emission rate of $0.05 \text{ m}^3 \text{ m}^{-3} \text{ day}^{-1}$ with CH_4 concentration of 62% (V) (Safley and Westerman 1988).

- Biogas production: $50 \text{ m}^3 \text{ day}^{-1}$
- CH_4 concentration: 62%(V)

- Biogas flow rate: $2.08 \text{ m}^3\text{h}^{-1}$
- Size of the biofilter: 2.08 m^3 (EBRT=1hr)
- Diameter of inner circle: 0.33 m
- Diameter of outer circle: 1.67 m
- Height of filter media: 1 m
- Filter media: Compost + wood chips (50/50 % V/V)
- Water content: 30% (WT)
- Pressure drop: 2 Pa/m

Next steps

A full-size biofiltration system should be designed, constructed, and tested on a commercial hog farm.

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