High-rate Methanotrophic Biofiltration (HMBF) Technology to Minimize Atmospheric Emission of Greenhouse Gas

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Abstract

Recent technology innovations associated with biological methane emission control and on-going laboratory- and field-based research on high-rate methanotrophic biofiltration (HMBF) systems are presented. Methanotrophic biofiltration, or the use of methanotrophic bacteria to oxidize CH_4, is a relatively recent method to control low volume methane (CH_4) emissions associated with landfills accepting biodegradable organic waste. Most operating methanotrophic biofilters are developed as passively aerated systems. A primary limitation of this type of biofilter is the reduction in CH_4 oxidation efficiency at relatively high flow rates because of the dependency on surface aeration to keep the biofilter bed aerobic and the inability to utilize the entire filter bed. This limitation prevents these biofilters from being used in applications such as controlling oil well vents and casing gas relief valve emissions where the gas volumes are relatively high. At these locations, HMBFs can be used because such systems could handle high flow rates. Recent research has shown that HMBFs are capable of operating at triple the methane elimination capacity of a passively aerated biofilter. HMBFs use media capable of supporting high growth of methanotrophic bacteria and are actively aerated using innovative gas and air feeding configurations to maximize CH_4 utilization. Results from on-going HMBF research, including the development of novel aeration configurations, show the possibility of using HMBFs for controlling solution gas emissions associated with oil production as well as point source emissions at medium-sized landfills with gas collection systems. Atmospheric emission of solution gas containing primarily CH_4, and small quantities of other volatile organic compounds, is a major contributor to overall CH_4 budget in the atmosphere. Since HMBFs are closed systems, they could be operated throughout the year even in cold climates such as Canada and northern Europe. Furthermore, HMBFs occupy a lower overall footprint compared to passively aerated biofilters. Therefore, this technology would be the preferred CH_4 emission control technology in many different conditions and environments.

Keywords: methane, biofiltration, greenhouse gas, methanotrophic bacteria, active-aeration

Introduction

Methane (CH_4) is the second most important greenhouse gas after carbon dioxide (CO_2), with a global warming potential (GWP) of 34 and average global concentration of 1800 ppb in 2014, a concentration unprecedented within the last 800,000 years [1]. About 60% of global CH_4 emissions are the result of human activities such as the oil and gas industry operations, livestock farming, and landfilling of biodegradable organic waste. In the oil and gas industry, flaring or venting often disposes of excess or unwanted waste gas rich in CH_4. Flaring is an environmental and human-health concern because of the production of highly toxic by-products and direct venting contributes to global warming. Recent research has shown that methane biofiltration, a biocatalysis-based method, could be used to control such emissions without producing by-products with serious environmental and health concerns. This approach is of significant interest because of the expected low capital and operational costs. Furthermore, the process of soil methanotrophy has been studied extensively in controlling fugitive CH_4 emissions from sanitary landfills [2,3]. However, the gas flow rates from point sources associated with the oil and gas industry are much higher than those encountered at typical sanitary landfills.

Oil and Gas Industry GHG Emissions and Control

In exploration and production of oil and natural gas, some of the gas produced is often vented directly into the atmosphere or flared, producing volatile organic compounds and other hazardous air pollutants that constitute negative environmental and human health consequences. In the province of Alberta, Canada, the oil and gas industry released 31.4 megatonnes of carbon dioxide equivalents in 2014 [4]. It is estimated that about 95% of these emissions are from engineered emissions or venting and fugitive emissions or leaks, with unburnt gas during flaring of sour gas accounting for the rest [4]. The Alberta government has pledged to reduce 45% of its methane emissions from the oil and gas industry by 2025. To do this, the industry must improve existing technology and/or adopt innovative methane reduction technologies. These technologies should be highly specific to the origin of the emissions.

There are many sources of CH_4 releases across the entire oil and gas supply chain. Some of the key emissions occur at the wellhead and at processing and storage locations. During oil production, the liquids release entrained gas (known as solution gas), which can be vented from the wellhead or from tanks unless captured [5]. Similar emission sources are associated with natural gas production. Since natural gas wells are often in distant locations without electricity, gas pressure is used to control and power a variety of control devices and on-site equipment, such as pumps [5]. These pneumatic devices characteristically bleed minor amounts of gas during their operation. Furthermore, during gas production, water is removed from the gas stream by glycol dehydrators, which vent some gas to the atmosphere. Such quantities are relatively small and difficult to collect in sufficient quantities in economical ways to allow collection and transportation to a gas processing plant.

Most solution gas, when available in sufficient quantities, is collected and transported by pipeline to a gas processing plant. The solution gas that cannot be directed to pipelines or used practically onsite due to safety or infrastructure reasons is vented or flared. Venting of solution gas containing more than 90% methane is a concern because of its high GWP. Flaring is the process of combusting the gas at a relatively low-temperature in a semi-enclosed burner. Although flaring will reduce the associated carbon released to the atmosphere, this practice has the negative impact of producing significant quantities of toxic by-products of human health concern. Although flaring is cost effective, the toxic by-product formation during the low-temperature burning of solution gas is a serious concern that would limit its application in most situations. Instead of flaring or venting, the two practices that have negative environmental or human health concerns, solution gas that may not serve any beneficial purpose could be directed to a biofilter that supports the growth of methnotrophic bacteria. Since the flow rates could be high, the biofilter should be operated as a HMBF.

High-rate methanotrophic biofiltration (HMBF) Technology

Classical biofilter operations typically use passive aeration, where atmospheric air interaction on the surface is the only O_2 source, and gas flow is controlled by the pressure difference between the biofilter and ambient air [6]. Oxidation rates in passively-aerated biofilters range between 5.3 to 152 $\frac{\mu g\ CH_4}{h\ g\ dw}$ [7-11]. Having O_2 enter in the opposite direction as the influent gas, leads to the reduction of system efficiency via formation of restricted CH_4 oxidation zones and not utilizing the full potential of the filter bed. Since methanotrophs are obligate aerophiles, preferring O_2 concentrations below atmospheric levels [12], performance may be better supported through active introduction of O_2 to the biofilter [6]. Streese and Stegmann [13] conducted laboratory experiments to determine the feasibility of treating landfill gas with actively-aerated columns by introducing a mixture of air and CH_4 with concentrations varying between 0.58-3.5% v/v. Their results indicated that performance declined for compost after five months of operation due to O_2 holdup by extracellular polymeric substances (EPS) accumulation. Haththotuwa et al., [14] studied the performance of actively-aerated columns subjected to high fluxes of CH_4 and air mixtures introduced at the bottom. Columns were packed with soil and fed at CH_4 fluxes ranging from 407 to 1212 $g/m^2/day$ in seven stages. The study suggests a maximum oxidation rate of 705 $g/m^2/day$. [15] also studied the performance of actively-aerated columns injecting air in 1/2, 1/3, and 1/6 proportions along the column depth. They observed good distribution of CH_4 oxidation throughout the columns and reported increase of oxidation by a factor of 5.5 compared to passive systems. However, they also observed O_2 transport disruptions after long-term operation. These research show that actively aerated systems perform better than passive systems and the use of multiple gas and air injection points may provide additional benefits.

This paper discusses results from our recent laboratory-based research and field investigations on the development of high-rate methanotrophic bifiltration (HMBF) systems developed specifically to control high volume and highly

concentrated CH_4 gas emissions associated with oil and gas industry operations. Some of the key results from laboratory experiments are presented together with some of the preliminary results from the field operations from the field-scale HMBF in operation in Hanna, Alberta.

Materials and Methods

Flow-through column experimental apparatus and test procedure

In laboratory experiments, flow-through columns with different air and gas feeding systems were tested (see Figure 1). Each column was made of rigid plexiglass tubes with inner diameter of 14 cm. The columns were closed at both ends with plexiglass end caps fitted with rubber O-rings. A perforated plate covered with a fine steel mesh at the base of the column supported the medium, which was packed to a depth of 70 cm**.** Sampling ports were drilled at 10 cm intervals and plugged with silicone septa to allow periodic sampling. The gas samples were analyzed for CH_4, O_2, CO_2, and N_2 using a HP Micro-Gas Chromatograph with Thermal Conductivity Detector. CH_4 (99% purity) was supplied through the bottom and flow rates increased (from 6 mL/min to 18 mL/min) in five stages. Columns were aerated at a flow rate 10 times higher than that of CH_4 at each stage, calculated based on stoichiometric demand.

The treatment gas was injected form the bottom and the air was injected from the bottom as well as along the column length.. The Column C1 was aerated at only one level with the air probe positioned at the bottom. The Column C2 received air at two levels; with one injection probe located at the bottom, and the other located 35 cm above. The Column C3 was subjected to air injection at three points positioned evenly along the 70 cm depth of the column. The columns were operated continuously for 195 days and gas concentrations were measured periodically to allow the determination of time dependent performance (as methane oxidation efficiency and rate). Details of the medium characteristics and the methods used to calculate oxidation rates and efficiencies are presented by Farrokhzadeh [16].

After 195 days of operation, the columns were dismantled, compost samples were collected from the top, middle, and bottom sections of each column and analyzed for oxidation kinetic parameters and microbial populations.

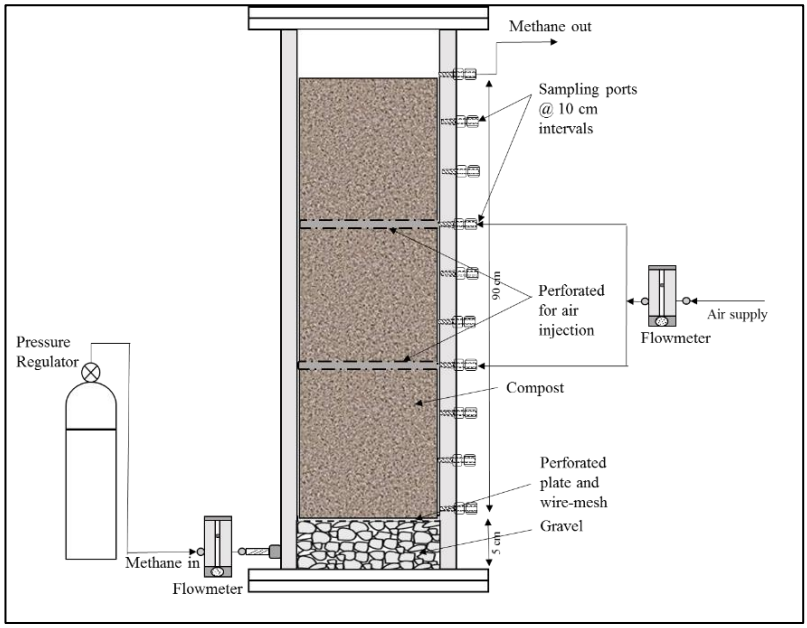

Figure 1: Schematic diagram of column configurations

Field-scale high-rate methanotrophic biofilter (HMBF) – design and operational details

Using the laboratory experimental results as guide, an active aeration field-scale high-rate methanotrophic biofilter (HMBF), was designed, constructed and installed at a single-well battery site in Hanna, Alberta. An oil battery site

is a facility where production fluid from a well is separated into gas, oil and water. A storage tank is used as a treater which separates the fluid into these three layers; salt water at the bottom, crude oil emulsions in the middle and natural gas at the top. The gas layer at the top may also contain solution gas that gets released when the fluid pressure in the treater is reduced. The HMBF was designed to receive a line of solution gas from the storage tank as the source of methane. The solution gas stream contained about 92% CH_4.

The schematic diagram of the field HMBF, with key dimensions, is presented in Figure 2. The HMBF consisted of a conical frustum shaped tank of $4m^3$ total volume and is designed to treat solution gas flow rates between 10 to $40m^3$/day. This would provide an empty bed retention time of upto 2.4 hours. The internal walls of the housing and the external walls of the tank were insulated using 5 cm closed cell spray foam insulation providing an R value of 12.5 to ensure there are no heat losses or gains during system operation. At highest design capacity, the HMBF would receive $40m^3$ of methane and $200m^3$ of air, adding up to a total of $240m^3$ of air-methane mixture per day. This gas is well mixed and uniformly distributed across the cross section of the HMBF. The gas distribution system consists of a piping network, gravel, and a geotextile to undertake this task. The distribution system was fabricated using 2" ABS pipes, with holes drilled into its surface to distribute the gas. The gas distribution system was placed on top of the 10 cm layer of 20 mm gravel. More gravel was filled on top of the distribution system to tightly secure it's positioning, and finally, the geotextile layer was placed on top to prevent the HMBF medium, compost, from clogging the gravel layer.

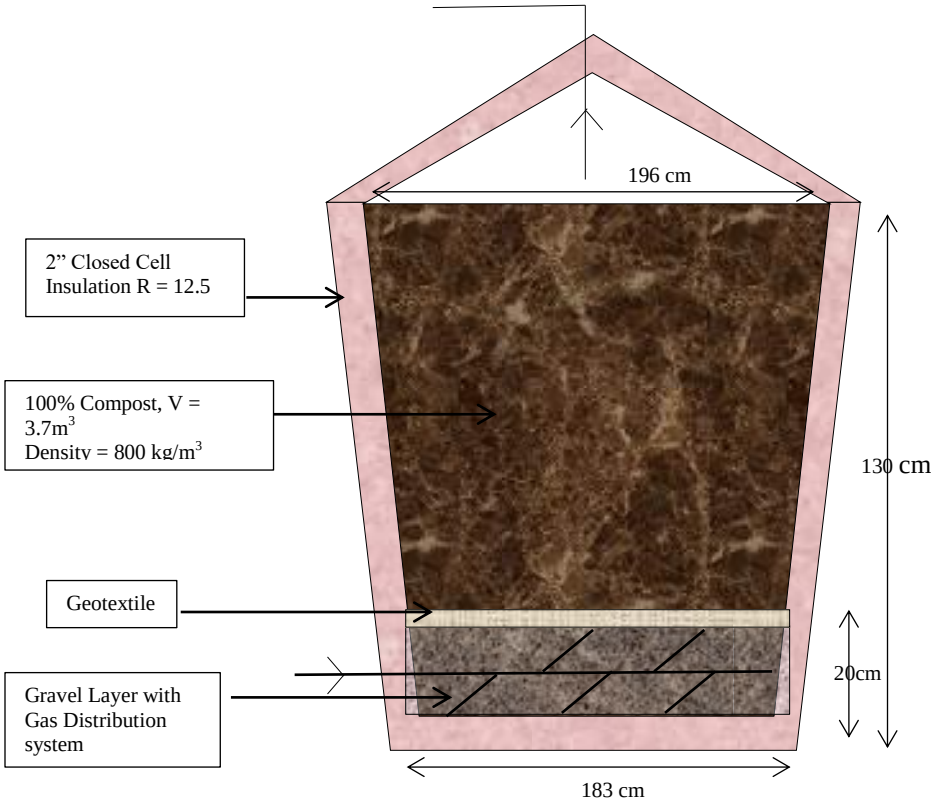

Figure 2: Schematic diagram of the field-scale HMBF

According to stoichiometry of the methane oxidation reaction, an air to methane ratio of 10:1 (v/v) is needed for methane oxidation. However, increasing the air flow would result in higher flowrates resulting in a reduced retention time and therefore an air to methane ratio of 5:1 was chosen. The compost selected as the HMBF medium had initial moisture content (MC) of 40% and field capacity (FC) of 50%. The compaction density of the compost medium was 800 kg/m^3. During operation, the performance of the HMBF was measured by its methane removal efficiency. Inlet and outlet methane flowrates and air flowrates and the inlet and outlet gas concentration and outlet gas concentrations were measured through sampling ports. The efficiency of the HMBF was calculated from:

$$Oxidation\ Efficiency = (CH_{4,in} - CH_{4,out} * (N_{2,in}/N_{2,out}))/CH_{4,in} * 100$$

This equation includes a N_2 component, which accounts for dilution that takes place through the addition of air.

Methane oxidation is an exothermic reaction and generates heat. Therefore, there is possible direct correlation between the temperature inside the HMBF and methane oxidation. A total of 15 temperature sensors were installed at three levels as shown within the HMBF to monitor the temperature profiles inside the HMBF. One sensor was placed outside to record the ambient temperature. The 15 sensors were connected to two data loggers, to ensure time-dependent temperature changes could be monitored on a continuous basis.

Results and Discussion

Effect of flow rates and air injection configurations on methane oxidation efficiencies and rates – Laboratory experimental results

The average methane oxidation efficiencies and oxidation rates over the 195 days of operating flow-through columns with five different methane flow rates are presented in Table 2. The column with two injection points (Column C2) exhibited the best performance in terms of methane oxidation rate and oxidation efficiency, 1309 $g/m^3/d$ (or 916.3 $g/m^2/d$) and 92%, respectively.

Table 1: Average oxidation rates and oxidation efficiencies of flow-through columns

Stage	Loading rate ($g/m^3/d$)	Oxidation rate ($g/m^3/d$)			Aeration efficiency (%)		
		C1	C2	C3	C1	C2	C3
I	529	420	423	501	78	77	95
II	794	519	671	704	65	84	89
III	1059	716	1025	931	65	**92**	84
IV	1324	600	1217	1083	45	89	80
V	1588	563	**1309**	633	35	82	40

The time-dependent oxidation rates and efficiencies for the three columns, C1, C2 and C3, over the 195 day time period are presented in Figures 3, 4 and 5, respectively.

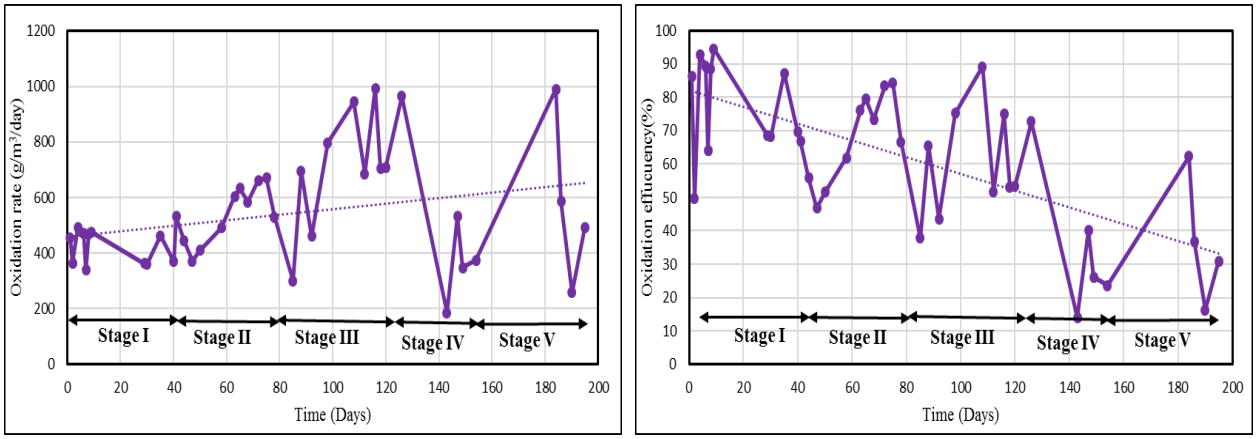

Figure 3: Oxidation rate ($g/m^3/day$) and oxidation efficiency (%) over time – Column C1

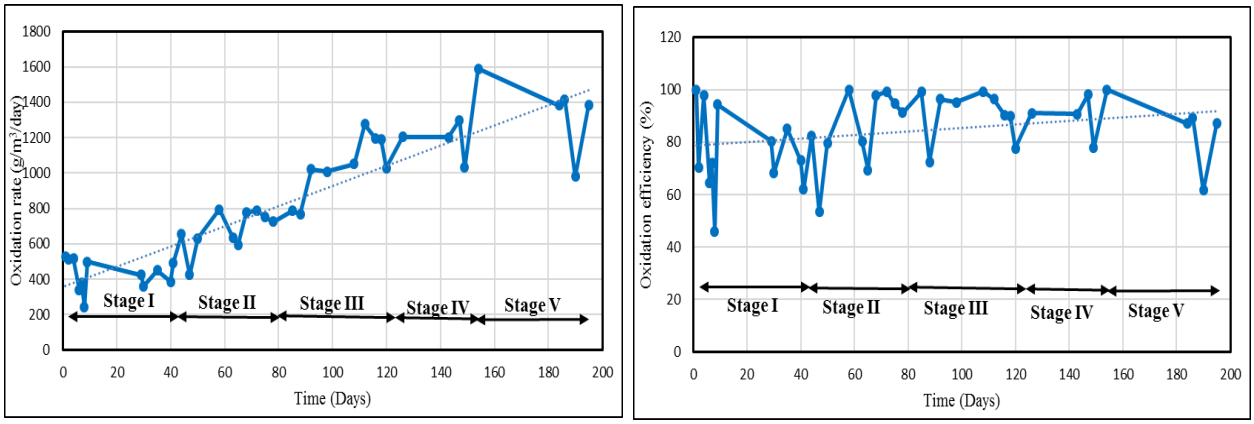

Figure 4: Oxidation rate (g/m^3/day) and oxidation efficiency (%) over time – Column C2

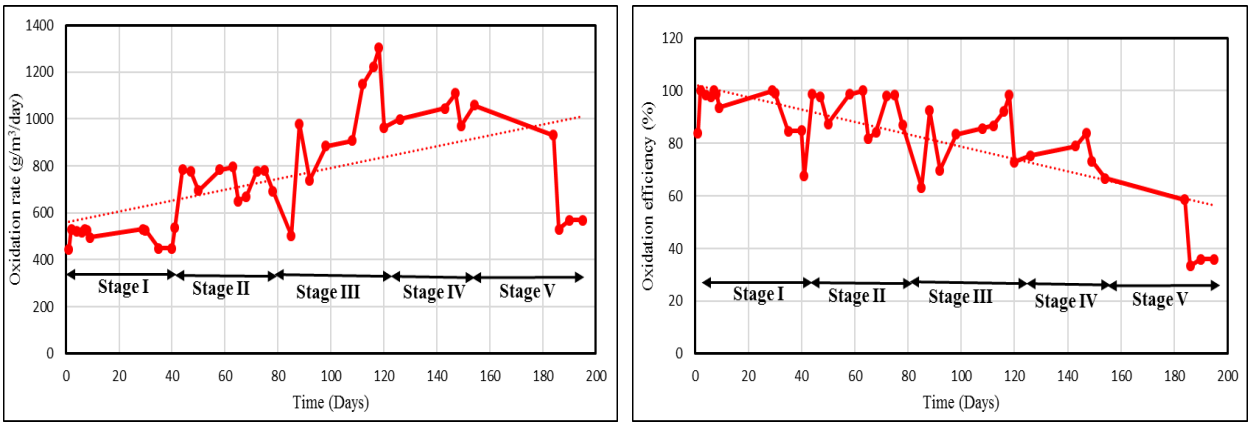

Figure 5: Oxidation rate (g/m3/day) and oxidation efficiency (%) over time – Column C3

The columns were operated in five stages, with increasing flow rates in each stage; 529, 794, 1059, 1324, and 1588 (g/m^3/d). When the columns operated at the lowest flow rate of 529 g/m^3/day, the performances of all three columns were similar. But, when the CH_4 flow rate increased to 794 g/m^3/day (555 g CH_4/m^2/day), there was marked difference between the Column C1 and the others with Column C1 operating at about 50% of the oxidation potential of Column C3. When the flow rate increased to 1058 g/m^3/day (741 g/m^2/day), the average oxidation efficiencies of columns C1, C2 and C3 were 65%, 92%, and 84%, respectively. Increasing the flow rate to 1323 g/m^3/day (926 g/m^2/day) during stage four, more divergence of performance was noticed. With an average oxidation rate of 1217 g/m^3/day (852 g/m^2/day) and a maximum of 1323 g/m^3/day (925 g/m^2/day), Column C2 had the best performance with an average oxidation rate twice of Column C1 with only 600 g/m^3/day (420 g/m^2/day). C3 operated at an average oxidation rate of 1083 g/m^3/day (758 g/m^2/day). With further increase of flow rate to 1588 g/m^3/day (1112 g/m^2/day), the performances of columns C1 and C3 decreased drastically. The oxidation rates decreased after day 184 until day 195 probably due to the effects of decreasing residence times.

Tim-dependent temperature variations and methane oxidation efficiencies – Early results from field investigation

The solution gas supply from the storage tank was connected to the MBF on 20th of September 2016. The contour plots in Figure 6 and Figure 7 show the distribution of temperature across the cross section of the HMBF at 60cm and 90cm on 1st of December, 2016.

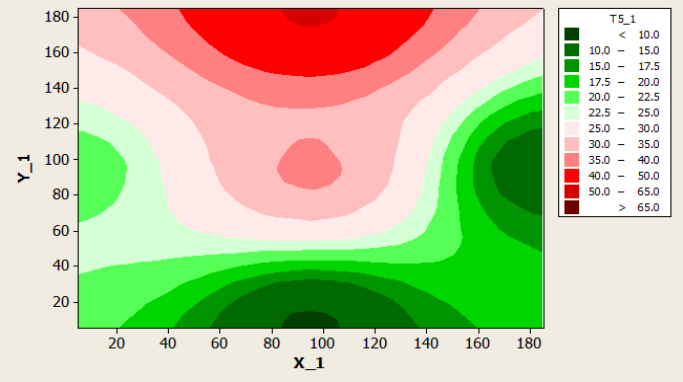

Figure 6: 60cm below surface level

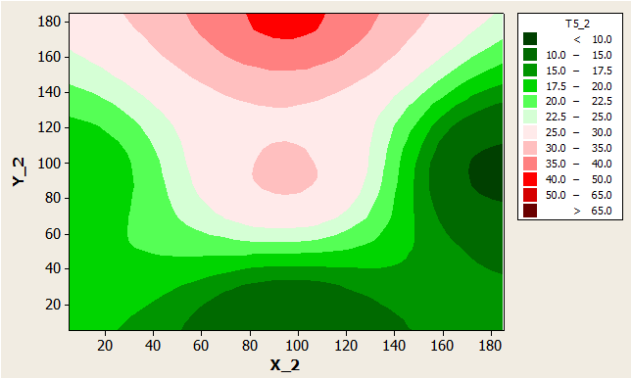

Figure 7: 90cm below surface level

As seen from the figures, the temperature inside the MBF was higher than 10ºC, which was much greater than the recorded average atmospheric temperature (-5ºC) on 1st of December indicating some methanotrophic activity within the HMBF. It is also evident that the temperature follows the exact pattern at both the surface levels although there is a slightly higher average temperature at 60cm. The temperature data shows that heat generation is not consistent within the HMBF, with certain areas of high heat generation or hot spots. This is quite common in field scale methane biofilter systems, either actively- or passively-aerated.

Table 3 presents the methane oxidation efficiencies during winter and early spring calculated from inlet and outlet gas flow rates and concentrations. Methane oxidation efficiencies increased over time indicating the presence of an extended lag phase before methanotrophic activity establishes in the HMBF. Since the HMBF was installed in fall, it appears that methanotrophic activity was low during late fall and winter. However, the rate of methane oxidation has seen a steady increase in late winter and early spring. Further data are being collected to establish the methane oxidation patterns throughout the year. Since Canadian weather shows extreme temperatures over the four seasons, the effect of weather extremes on the performance of the HMBF could be clearly established with year-round data.

Table 2: Hanna HMBF performance during winter and early spring

		Inlet Flowrate (m^3/day)		**Concentrations of Gas mix (v/v)**		**Efficiency (%)**
				Inlet	**Outlet**	
27/02/2017	Solution Gas	23	CH_4	11.1	7.2	38.73
	Air	121	O_2	18.6	18.8	
13/03/2017	Solution Gas	23	CH_4	9.5	6.5	34.13
	Air	171	O_2	20.8	20.9	
8/5/2017	Solution Gas	23.83	CH_4	13.2	7.3	48.93
	Air	130.55	O_2	18.4	19.5	
14/05/2017	Solution Gas	15.4	CH_4	6.6	3.5	48.90
	Air	137	O_2	19.9	20.3	

Conclusion

An actively aerated closed high-rate methane biofilter (HMBF) was designed using laboratory data and installed at a single well battery site in Hanna, AB during the fall of 2016. During laboratory-scale experimental program, three biofilter designs were tested changing aeration levels in one, two, and three levels. The oxidation rates/ efficiency plots over time at various flowrates suggest that the two-level aerated biofiltration system yields the best performance over the entire range of flow rates tested. In the field HMBF, initially the solution gas containing 92% methane was fed from one injection point at the bottom mixed with air. The HMBF was designed to accept upto 40 m^3/day of methane. The methane oxidation efficiency was low at the beginning but has increased considerably in early spring, or after six months of continuous operation. The current plan is to increase the flow rate gradually as the HMBF starts to oxidize methane at its full capacity and change the air injection configuration to be compatible with the two-level air injection system tested during laboratory studies. Temperature inside the HMBF is not uniform across its cross section, suggesting the presence of hot spots with high bacterial activity.

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