



Aerated biofilters with multiple-level air injection configurations to enhance biological treatment of methane emissions



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HIGHLIGHTS

- Methane biofilter performance was improved through multiple-level aeration design.
- A comprehensive set of statistically-designed lab experiments were conducted.
- 2-Level aerated biofilter showed a higher oxidation than the other studied biofilters.

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ABSTRACT

Aiming to improve conventional methane biofilter performance, a multiple-level aeration biofilter design is proposed. Laboratory flow-through column experiments were conducted to evaluate three actively-aerated methane biofilter configurations. Columns were aerated at one, two, and three levels of the bed depth, with air introduced at flow rates calculated from methane oxidation reaction stoichiometry. Inlet methane loading rates were increased in five stages between 6 and 18 mL/min. The effects of methane feeding rate, levels of aeration, and residence time on methane oxidation rates were determined. Samples collected after completion of flow-through experiments were used to determine methane oxidation kinetic parameters, V_{max} , K_m , and methanotrophic community distribution across biofilter columns. Results obtained from mixed variances analysis and response surfaces, as well as methanotrophic activity data, suggested that, biofilter column with two aeration levels has the most even performance over time, maintaining 85.1% average oxidation efficiency over 95 days of experiments.

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1. Introduction

With a global warming potential (GWP) of 28–34 over a period of 100 years, and 85 over a window of 20 years, methane (CH₄) contribution to heat trapping and global climate change is significant (Myhre et al., 2013; Caulton et al., 2014). Anthropogenic CH₄ has natural gas and petroleum systems as its largest contributors above landfills, wastewater treatment plants, rice paddy agriculture, and livestock farms. CH₄ accounts for approximately 13% of Canada's total emissions, and 14.3% of the global anthropogenic GHG emissions (Environment Canada, 2014; Chai et al., 2016).

A simple, cost-effective, and environment-friendly method available for CH₄ elimination is biological oxidation, a technology based on the use of CH₄-oxidising bacteria, known as methanotrophs. Methane biofilters (MBFs) have the potential to replace

flaring of waste gas containing CH₄ when flow rates are low (lower than 15–30 m³/h), intermittent, and concentrations are low (below 20–30% v/v) without producing toxic by-products. MBFs entail a layer of granular material on which the methanotrophs reside. CH₄ acts as the energy and carbon source for the methanotrophs and is oxidized to carbon dioxide (CO₂) and water (Mancebo et al., 2016). The mechanism is affected by a variety of parameters including CH₄ concentrations and flow rates, O₂ availability, temperature, moisture content, and the type of granular medium used.

Classical biofilter operations typically use passive aeration, where atmospheric air interaction on the surface is the only O₂ source, and the gas flow is controlled by the pressure difference between the biofilter and ambient air (Gebert and Groenroeft, 2006a). Oxidation rates in passively-aerated biofilters range between 5.3 and 152 $\frac{\mu\text{gCH}_4}{\text{gdw}}$ (Whalen et al., 1990; Jones and Nedwell, 1993; Kightley et al., 1995; Börjesson et al., 1998; Henckel et al., 2000; Gebert et al., 2003; He et al., 2008;

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Chiemchaisri et al., 2013). Having a countercurrent flow of O₂ as the influent gas in passive MBFs leads to formation of restricted CH₄ oxidation zones and reduction of system efficiency as a result of not utilizing the full potential of the filter bed. Moreover, since methanotrophs are obligate aerophiles (Mancinelli, 1995), performance may be better supported through active introduction of O₂ to the biofilter (Gebert and Gröngroft, 2006b).

Streese and Stegmann (2003) conducted laboratory experiments to determine the feasibility of treating landfill gas with actively-aerated columns packed with fine-grained compost by introducing a mixture of air and CH₄ with concentrations varying between 0.58 and 3.5% v/v. Their results indicated that performance declined for compost after five months of operation due to O₂ hold up by extracellular polymeric substances (EPS) accumulation. Although the decrease in performance could also be attributed to insufficient contact time, such effects were not accounted for in this study. Haththotuwa et al. (2012) studied the performance of up-flow actively-aerated columns subjected to high fluxes of CH₄ and air mixtures. Columns were packed with soil and fed at CH₄ fluxes ranging from 407 to 1212 g/m²/day in seven stages. The study suggests a maximum oxidation rate of 705 g/m²/day. Haubrichs and Widmann (2006) also studied the performance of actively-aerated columns filled with fine-grained compost material injecting air in 1/2, 1/3, and 1/6 proportions along the column depth. They observed the good distribution of CH₄ oxidation throughout the columns and reported an increase of oxidation by a factor of 5.5 compared to passive systems. However, they also observed O₂ transport disruptions after long-term operation. Concentrations tested were representative of poor landfill gas where the application of active aeration with variable airflow rates may not be feasible.

To assess the possibility of maintaining ideal O₂ concentrations for biological reactions throughout a biofilter column and ensuring that O₂ availability is no longer a limitation to CH₄ degradation, a multiple level air injection system is tested in this work. The long-term effects of multiple air injection points on biofilter performance are determined based on the resulting residence times, oxidation rates, microbial populations, and methanotrophic activities. Flow-through column experiments are carried out for the present study, as they provide a better representation of full-scale operation compared to batch experiments, to study the effects of the number of injection points and CH₄ loading rates. Statistically designed experiments and comprehensive statistical analyses conducted to evaluate the main effects and their interactions.

2. Materials and methods

2.1. Biofilter column packing medium

The packing medium used in the biofilter columns is leaf compost. Characteristics of the packing medium were estimated using the methodologies described in Farrokhzadeh (2016). The organic content, moisture content (dry weight), particle size, particle density, water holding capacity, dry bulk density and wet bulk density of the compost samples used in the experiments were 35%, 60%, <2.36 mm, 2.28 kg/m³, 71.4 %, 0.60 g/cm³ and 0.98 g/cm³, respectively.

2.2. Experimental protocol

Doehlert experimental design, also known as the uniform shell design, was chosen as the experimental protocol of choice with a number of air injection points and CH₄ flow rate as the independent variables and CH₄ oxidation rate as the response variable. A response surface, on which the optimum values of controlling vari-

ables are defined, was developed. The Doehlert design allows for the analysis of each of the factors at a different number of levels with fewer experiments compared to the central composite design (Doehlert, 1970; Ferreira et al., 2007). The factor with the highest number of levels was chosen as the most significant factor. The number of air injection points and flow rates was changed in three levels (one, two and three injection points) and five levels (6, 9, 12, 15 and 18 mL/min), respectively (see Table 1).

2.3. Analysis of variances (ANOVA)

Using regression analysis and a multiple factor ANOVA, the significance of each of the factors and their interactions was determined. All the calculations were performed using SPSS statistics software (IBM Corporation) and 3D graphs were plotted using SigmaPlot (Systat Software Inc.).

2.4. Reactor description

The experiments were conducted with flow-through biofilter columns made of rigid Plexiglas tubes with an inner diameter of 14 cm. The columns were closed at both ends with Plexiglas end caps fitted with rubber O-rings. The end caps were fastened to the columns with threaded rods that ran the length of the column. 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 (see Fig. 1). The gas samples were analyzed for CH₄, O₂, CO₂, and N₂ using an HP Micro-Gas Chromatograph with Thermal Conductivity Detector.

The columns were checked for leakage before experiments. CH₄ (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 ten times higher than that of CH₄ at each stage, calculated based on stoichiometric demand. Air was injected through brass-perforated probes located at desired levels. 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.

Oxidation efficiency and oxidation rate were used to compare the performances of the three flow-through biofilter columns. The oxidation efficiency was calculated from (Nikiema et al., 2007);

$$\text{oxidation efficiency (\%)} = \frac{Q_{in}C_{in} - Q_{out}C_{out}}{Q_{in}C_{in}} \times 100\% \quad (1)$$

where, C_{in} and C_{out} are inlet and outlet concentrations of CH₄ in g/m³, Q_{in} and Q_{out} are the flow rate of CH₄ entering at the column's base and flow rate of the column's effluent in m³/day, respectively.

The oxidation rate was calculated in g/m³/day from;

$$\text{oxidation rate} = \frac{1}{V} (Q_{in}C_{in} - Q_{out}C_{out}) \quad (2)$$

where, V is the volume of the filter bed in m³.

2.5. CH₄ oxidation kinetics parameters

After 195 days of continuous operation, the columns were dismantled, and compost samples were collected from the top, middle, and bottom sections of each column to determine the distribution of oxidation activities along the column depth. Batch microcosm experiments were conducted following the methods of Pokhrel et al. (2016) and Mancebo et al. (2014) and Mancebo and Hettiaratchi (2015) by incubating samples in airtight 1 L bot-

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