



Biofiltration of methane using hybrid mixtures of biochar, lava rock and compost

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ABSTRACT

Using hybrid packing materials in biofiltration systems takes advantage of both the inorganic and organic properties offered by the medium including structural stability and a source of available nutrients, respectively. In this study, hybrid mixtures of compost with either lava rock or biochar in four different mixture ratios were compared against 100% compost in a methane biofilter with active aeration at two ports along the height of the biofilter. Biochar outperformed lava rock as a packing material by providing the added benefit of participating in sorption reactions with CH₄. This study provides evidence that a 7:1 volumetric mixture of biochar and compost can successfully remove up to 877 g CH₄/m³·d with empty-bed residence times of 82.8 min. Low-affinity methanotrophs were responsible for the CH₄ removal in these systems ($K_{M(\text{app})}$ ranging from 5.7 to 42.7 μM CH₄). Sequencing of 16S rRNA gene amplicons indicated that Gammaproteobacteria methanotrophs, especially members of the genus *Methylobacter*, were responsible for most of the CH₄ removal. However, as the compost medium was replaced with more inert medium, there was a decline in CH₄ removal efficiency coinciding with an increased dominance of Alphaproteobacteria methanotrophs like *Methylocystis* and *Methylocella*. As a biologically-active material, compost served as the sole source of nutrients and inoculum for the biofilters which greatly simplified the operation of the system. Higher elimination capacities may be possible with higher compost content such as a 1:1 ratio of either biochar or lava rock, while maintaining the empty-bed residence time at 82.8 min.

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1. Introduction

Since the industrial revolution, methane (CH₄) has increased by 2.5 times and currently makes up 16% of all anthropogenic greenhouse gases (IPCC, 2014; NOAA/ESRL, 2018). Of all CH₄ emissions, more than 60% are anthropogenic and primarily originates from animal husbandry, landfilling, composting, wastewater anaerobic treatment, natural gas refineries, and coal mining (IPCC, 2013). Abatement of CH₄ emissions can be performed by collection and flaring at a minimum concentration of 20% v/v (Haubrichs and Widmann, 2006). As more than 55% of anthropogenic CH₄ emissions possess a concentration below the lower explosive limit of CH₄ in air of 5% v/v, they are incompatible for energy recovery or for

chemical oxidation processes for the removal of CH₄ (Estrada et al., 2014). In this regard, biotechnologies based on the activity of methanotrophic bacteria are considered a cost-efficient and environmentally-friendly treatment alternative to physical-chemical technologies. Among biotechnologies, biofilters have become popular within the past decades for the treatment of off-gases as they are considered cost-effective especially for the removal of diffuse emissions of CH₄ and, generally, result in lower environmental impacts as compared to physical-chemical treatments (Estrada et al., 2012).

Biofiltration relies on microorganisms for the transformation of air pollutants into less toxic and odorless chemicals. The packing medium, which can be biologically active or inactive, immobilizes the microorganisms that are responsible for the CH₄ removal. Biologically active organic material are typically low-priced and contain a natural source of nutrients, can maintain an adequate water content, and are a natural source of a diverse mixture of

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microorganisms which removes the challenge of obtaining a suitable inoculum for the system (Farrokhzadeh et al., 2017; Huber-Humer et al., 2011; Park et al., 2009; Stein and Hettiaratchi, 2001; Visvanathan et al., 1999; Wei et al., 2016). A negative aspect associated with using biologically active materials is that they are less structurally stable than inactive materials. Because biologically active materials, such as compost and soils, are more susceptible to compaction, they are prone to operational issues such as pressure-drops and channeling. On the other hand, when a biologically inactive material is used, primary nutrients such as nitrogen and phosphorus are typically not present and must be supplied using an external source, incorporated into the biofilter packing material, or manually supplied with a nutrient solution (Avalos Ramirez et al., 2012; Kim et al., 2014b; López et al., 2018; Nikiema et al., 2009).

A few studies have incorporated biologically inactive materials wholly or partially as hybrid material into biofilter beds in order to overcome issues related to filter bed plugging as a result of excessive biomass development and to promote homogeneous gas distribution by maintaining structural stability of the filter bed (Brandt et al., 2016; Melse and van der Werf, 2005; Perdikis et al., 2008; Syed et al., 2016; Syed et al., 2017). With the exception of biochar, or activated carbon, most biologically inactive materials have limited sorption capabilities and tend to be more expensive than biologically active materials (Gómez-Cuervo et al., 2017). The use of hybrid packing materials in biofilter systems takes advantage of both inorganic and organic properties and may improve the performance of the system (Dorado et al., 2012; Gómez-Cuervo et al., 2017).

The purpose of this study was to determine the minimum volume of compost, an organic and biologically-active material, in a hybrid mixture of biochar or lava rock, as inert filter bed materials, to achieve 100% CH₄ removal efficiencies at a preselected inlet CH₄ flow rate. Lava rock, typically used in landscaping, was chosen as a potential bulking agent due to its inherent porous nature and large particle size for the benefit of promoting gas flow (Delhoménie et al., 2002). The porosity of lava rock has been reported to be as high as 49% to 65% (Akdeniz et al., 2011; Chitwood et al., 1999). Porosity of biochar varies with feedstock, temperature and method of production and can range from 20% to more than 50%. Specifically, the objectives of this study consists of the following: (i) determine the effects of using different filter bed materials (compost, biochar, and lava rock) and four different volume mixtures of hybrid materials (1:1, 3:1, 7:1, and 15:1 of biologically-inactive:biologically-active) on CH₄ elimination capacities (ECs) and removal efficiencies (REs), (ii) investigate the effects of material composition and hybrid mixtures on CH₄ oxidation kinetics, (iii) examine the effect of material composition and hybrid mixtures on physical-chemical properties including pH, moisture content, and organic content, and (iv) determine the microbiological community structure across all treatments.

2. Materials and methods

2.1. Packing medium

Lava rock, of 3–20 mm diameter, (Burnco, Calgary, AB) and wood-based biochar (Diacarbon, Burnaby, BC) were used in combination with compost as packing medium for the biofilter columns. Sawdust is the primary feedstock for the biochar which is produced under fast pyrolysis conditions at 550 °C and a residence time of 20–30 min. The resulting biochar is similar in size and morphology to the untreated sawdust and ranges in size from 0.5 to

1 cm slivers. Compost was acquired from the East Calgary Waste Management Facility in Calgary, Alberta in July 2017 and sieved through a no. 8 sieve (2.38 mm) prior to conducting the experiments. Compost is an attractive medium as it is a low-cost nutrient source, high in organic matter content, and is a source of methanotrophs for the inoculation of the inert biochar and lava rock biofilters. The organic matter content (OC) (ASTM D2974-14; 2014), moisture content (MC) (dry weight) (ASTM D2216-10; 2010), bulk density, pH (ASTM D4972-13; 2013), and water holding capacity (WHC) (ASTM D2980-04, 2010) were determined for all three materials. Bulk density was determined by weighing the material in an 80-mL, 300-mL, and 1-L filled beaker and taking the average result.

2.2. Lab-scale biofilter description

Methane oxidation experiments were conducted through six biofilter columns made of Plexiglas[®] (inner diameter = 0.14 m; height = 0.88 m). The biofilters were enclosed at both ends with rubber O-rings and acrylic endcaps. The packing material was separated into 3 independent sections using stainless steel mesh plates in order to promote homogeneous gas distribution and reduce operational issues related to compaction and channeling. Prior to the experiments, each biofilter was tested for gas tightness using compressed air at 3 psi. Each biofilter was filled up to a height of 70 cm above a 9 cm gas distribution layer of 16/32 gravel. Sampling ports were drilled at 9 cm intervals and fitted with PTFE Tube Fitting, Male Connector (Swagelok[®], Calgary, AB) and silicone septa (SGE Analytical Science, Pflugerville, TX, USA). Air was distributed into the biofilter using perforated copper tubing of 10 cm in length that was sandwiched between two stainless steel mesh plates. The mesh plates also prevented the packing material from falling into and clogging the copper tubing. Total air flow was divided between two air injection ports at 2/3 and 1/3 at 31.5 cm and 58.5 cm, respectively, in order to take into account the decline in CH₄ concentration as it travels up the filter bed. Methane was fed at the lower port situated 4.5 cm from the bottom. An outlet port was connected at the top-most port at 88.5 cm and diverted the treated gas towards an exit stream.

The total oxygen flow rate was calculated in order to supply 2.5 times the CH₄ flow rate—providing a 25% margin of safety given the total oxidation of CH₄ requires the consumption of 2 moles of O₂ per mole of CH₄. With the assumption of no biodegradation nor CH₄ generation within the biofilter, the declining CH₄ concentration is a direct result of air input. Based on air dilution alone, with an inlet CH₄ concentration of 99% (Praxair, Calgary, AB), the CH₄ concentration is reduced to 11% at level 1 and 7.6% at level 2 and in the treated gas. A schematic of the biofiltration system is shown in Fig. 1. The method of aeration is similar to the set-up used by Haubrichs and Widmann (2006).

Gas measurements for O₂, N₂, CH₄, and CO₂ were made using a gas chromatograph (GC) with a TCD detector (see Section 2.6). All tests were performed at room temperature, 22 ± 3 °C. The performance of the 6 biofilters were determined by calculating the removal efficiency, RE, (%), elimination capacity, EC, (g CH₄/m³ · d), CO₂ production rate, PCO₂, (g CO₂/m³ · d), and the CO₂ yield coefficient, YCO₂, (g CO₂/g CH₄). Mathematical formulae used to calculate these parameters are provided in the Supporting Information section (Table S1).

2.3. Experimental method

The study was conducted under four different stages in

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