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Inhibitory effects of acidic pH and confounding effects of moisture content on methane biofiltration

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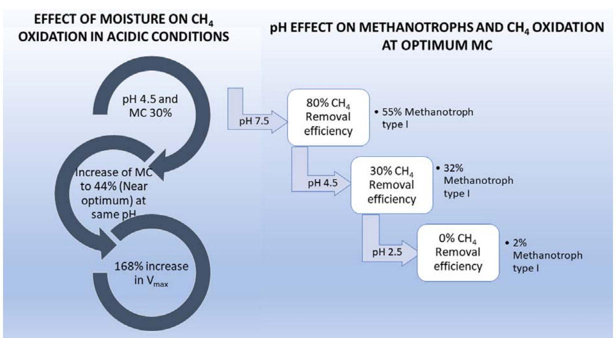
GRAPHICAL ABSTRACT



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ABSTRACT

This study focussed on evaluating the effect of hydrogen sulfide (H_2S) on biological oxidation of waste methane (CH_4) gas in compost biofilters, Batch experiments were conducted to determine the dependency of maximum methane oxidation rate (V_{max}) on two main factors; pH and moisture content, as well as their interaction effects. The maximum V_{max} was observed at a pH of 7.2 with decreasing V_{max} values observed with decreasing pH, irrespective of moisture content. Flow-through columns operated at a pH of 4.5 oxidized CH_4 at a flux rate of 53 g/m^2/d compared to 146 g/m^2/d in columns operated at neutral pH. No oxidation activity was observed for columns operated at pH 2.5, and DNA sequencing analysis of samples led to the conclusion that highly acidic conditions were responsible for inhibiting the ability of methanotrophs to oxidize CH_4. Biofilter columns operated at pH 2.5 contained only 2% methanotrophs (type I) out of the total microbial population, compared to 55% in columns operated at pH 7.5. Overall, changes in the population of methanotrophs with acidification within the biofilters compromised its capacity to oxidize CH_4 which demonstrated that a compost biofilter could not operate efficiently in the presence of high levels of H_2S.

1. Introduction

Methane (CH_4) is a potent greenhouse gas (GHG) with a global warming potential of 28–34 in the 100-year time horizon, and 85 over the 20-year time horizon (IPCC, 2013; Myhre et al., 2013; Caulton et al., 2014). Anthropogenic atmospheric CH_4 emission sources include oil and gas operations, landfills, rice paddy agriculture, and livestock farms, which account for more than 50% of total CH_4 emissions (Chai

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et al., 2016). Flaring and gas combustion for energy production are two common mitigation technologies that have been proven to be efficient in reducing such emissions, but these methods require CH_4 concentrations higher than 20% (v/v) and high volumes. Low-temperature combustion also produces toxic by-products (Johnson and Coderre, 2012). Biological methods are proven to be cost-effective for mitigating CH_4 emissions, with methane biofiltration offering the simplest approach in terms of implementation and operation, without producing toxic by-products (Lebrero et al., 2016; Farrokzadeh et al., 2017). A methane biofilter (MBF) typically consists of 3 phases i.e., the filter bed (solid phase), the biofilm (liquid phase), and CH_4 (the gas phase). Methanotrophic bacteria, or methanotrophs, use CH_4 as their carbon and energy source, and convert it to less-harmful carbon dioxide (CO_2), water and biomass (Mancebo et al., 2016). The mechanism is affected by a variety of factors including CH_4 concentration and flow rate, oxygen (O_2) content, temperature, moisture content (MC), pH, and type of the granular media used (Nikiema et al., 2007; Mancebo and Hettiaratchi, 2014).

In addition to CH_4, other gases are emitted from landfills, livestock facilities, oil and gas production and wastewater treatment plants (Yang and Allen, 1994; Syed et al., 2006; Kim et al., 2012). One of such gases is hydrogen sulphide (H_2S), which is a toxic air pollutant and could affect MBF operation by changing the behaviour of methanotrophs in the system (Caceres et al., 2014). Depending on its concentration, H_2S can have both positive and negative effects on CH_4 oxidation efficiency. At low concentrations, H_2S may provide sufficient nutrients for the growth of methanotrophs in the biofilm, whereas at high concentrations, it could inhibit bio-oxidation of CH_4 due to its toxicity and changing the pH of the system (Yu et al., 2009, Pratt et al., 2012; Caceres et al., 2014). In the presence of water and air, H_2S oxidizes to sulphuric acid resulting in pH reduction that can inhibit the growth and activity of methanotrophs (Nikiema et al., 2007; Pratt et al., 2012). In addition to growth inhibition, H_2S can also negatively impact the methanotroph's ability to oxidize CH_4 due to its high solubility in water at neutral pH, which has been found to be 3.85 g per kg of water at 1 atm and 20 °C (Caceres et al., 2014). H_2S also inhibits enzyme protein activities by binding to metal ions, which are cofactors of enzyme proteins in microbial cells (Yu et al., 2009). Furthermore, it has been reported that reduced sulphur compounds, such as methanethiol (CH_4S) and carbon disulfide (CS_2) have a significant inhibitory effect on the bio-oxidation of CH_4 in landfills, in which methanotrophs type I have been more affected than type II (Borjesson et al., 2004; Lee et al., 2012; Caceres et al., 2014).

Moisture content is a major factor affecting CH_4 oxidation capacity (Tate, 2015) as it is considered as a transport medium for residual compound removal and also nutrient supply. Considering the key roles they are playing, it is meaningful to investigate the potential inhibitory effects of H_2S and moisture content. However, only limited research has been conducted in the past to investigate the effect of H_2S on CH_4 oxidation when mixed with CH_4 (Caceres et al., 2014). Therefore, this study was conducted to investigate the primary effect of pH reduction on CH_4 oxidation due to H_2S, and the interaction effects due to moisture content. The direct toxicity effect of H_2S on methane oxidation is not reported here.

To study the inhibitory effect of low pH on CH_4 oxidation, batch microcosm experiments were conducted by changing pH and moisture content. One set of batch experiments were performed to assess the change in maximum CH_4 oxidation rate (V_{max}) at different pH values, and another conducted to determine the effect of moisture content on V_{max} at different pHs. A set of flow-through column experiments was also conducted to determine the CH_4 removal efficiency of compost biofilters under acidic conditions by simulating field conditions.

2. Materials and methods

2.1. Compost characteristics

Leaf compost obtained from the City of Calgary was used in batch incubation experiments and in the flow-through column experiments. Physical parameters of the compost were determined according to standard methods. Characteristics of the packing medium were estimated using the methodologies described in Barzgar (2017). The organic content, particle size, water holding capacity, porosity, particle density and bulk density of the compost samples used in the experiments were 38%, < 2.35 mm, 68.2%, 0.56, 1.24 g/cm^3 and 0.54 g/cm^3, respectively.

2.2. Batch experiments

2.2.1. Determination of kinetic parameters

Before conducting batch experiments to determine kinetic parameters, pre-incubation experiments were conducted to ensure that the compost samples had sufficient methanotrophic populations. About 100 g of compost was incubated in 1000 ml airtight glass bottles with 5–6% initial concentrations of CH_4 gas. Moisture content was maintained at 35% (Humer and Lechner, 1999), and temperature at 22 °C. The headspace gas concentrations were measured over time using the micro-gas chromatograph (micro-GC) VARIAN CP 4900 until reaching a steady oxidation rate.

To determine kinetic parameters, V_{max} (maximum reaction rate) and K_m (apparent saturation constant or Michaelis-Menten constant), samples were subjected to batch incubation experiments. Batch microcosm experiments were conducted following the method of Pokhrel et al., (2016) by incubating 10 g of media in 250 mL airtight glass (amber) bottles at room temperature of 22 °C. A headspace concentration of approximately 10% was achieved by injecting CH_4 into the bottles. Excessive pressure build-up inside the bottles was prevented by withdrawing the same volume of air from bottles prior to the addition of CH_4. The headspace of bottles was sampled 5 times during the experiments by removing 2 mL of gas, and CH_4 concentrations were quantified using the micro-GC with thermal conductivity detectors. The resulting data were used to plot substrate saturation curves and V_{max} values were calculated from a non-linear regression using SPSS IBM STATISTICS software.

2.2.2. Experimental design

One set of batch experiments were performed using the one-factor-at-a-time (OFAT) experimental design to determine the relationship between V_{max} and pH at moisture content (MC) of 30%. Different concentrations of sulphuric acid were added to the samples to maintain pH at the desired values. Other factors, such as temperature, nutrient content, CH_4 and oxygen concentrations were kept constant. In total, nine experiments in duplicate were conducted with pH values between 2.5 and 8.2 (note that 8.2 was the initial pH of compost). Then, the Central Composite Design (CCD) was used in the second set of batch experiments to investigate the effect of MC on CH_4 oxidation with varying pH. In this design, both pH and MC were varied simultaneously by changing the moisture content of compost along with the pre-determined amounts of sulphuric acid required to maintain the pH of compost at the desired values. Other factors, such as temperature, nutrients content, CH_4 and O_2 concentration were kept constant. CCD is commonly used to estimate first and second order effects efficiently and