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Research article

Steady state and dynamic behaviors of a methane biofilter under periodic addition of ethanol vapors

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ABSTRACT

Ethanol was added to a methane (CH_4) biofilter with inorganic packing materials over three cycles based on increasing the gas flow rates from 3 to 6 and finally to 12 L min^{-1} corresponding to empty bed residence times (EBRT) of 6, 3 and 1.5 min. The steady state performance of the CH_4 biofilter was studied for CH_4 inlet loads (ILs) of 33, 66 and 132 g_{CH4} m^{-3} h^{-1} prior and after each ethanol cycle. In addition, the steady state removal of a mixture of CH_4 and ethanol for a CH_4/ethanol mass ratio of around 7.5 g_{CH4} $g^{-1}_{ethanol}$ was evaluated over three cycles (EBRTs of 6, 3 and 1.5 min). In the absence of ethanol, the CH_4 removal efficiency (RE) dropped from 35 to 7% due to an EBRT decrease from 6 to 1.5 min. In addition, the presence of ethanol resulted in a CH_4 RE reduction at a constant EBRT in every cycle. The CH_4 REs dropped from 35 to 29%, 17 to 13% and 7 to 0% for corresponding ethanol ILs of 4.5, 9 and 18 $g_{ethanol}$ m^{-3} h^{-1} over the cycles. Moreover, the periodic presence of ethanol in the CH_4 biofilter allowed the study of transient behaviors of the biofilter during ethanol addition and the biofilter recovery after each cycle. The CH_4 RE reduction as a result of ethanol addition in each cycle was instantaneous. However, the CH_4 RE recovery after completion of ethanol addition took 10, 14 and 25 days for ethanol ILs of 4.5, 9 and 18 $g_{ethanol}$ m^{-3} h^{-1} respectively. The recovery time was related to the ethanol concentration in the leachate which were 1100 ± 200, 1100 ± 350 and 2500 ± 400 $g_{ethanol}$ $m^{-3}_{leachate}$ for corresponding ethanol ILs of 4.5, 9 and 18 $g_{ethanol}$ m^{-3} h^{-1}, respectively. Based on steady state and dynamic process conditions of the biofilter, the lowest gas flow rate of 3 L min^{-1} (EBRT of 6 min) produced the best performance when both pollutants were present (CH_4 IL of 33 g_{CH4} m^{-3} h^{-1} and ethanol IL of 4.5 $g_{ethanol}$ m^{-3} h^{-1}).

1. Introduction

Over the recent years, greenhouse gas (GHG) emissions of methane (CH_4) and carbon dioxide (CO_2) have been targeted for reduction due to their global warming effects (Schuur et al., 2015). At a recent climate change conference (COP 21, Paris 2015), more than 200 countries submitted an agreement to keep the global temperature from increasing more than 2 °C compared to pre-industrial levels (Rogelj et al., 2016). Methane, the second most important GHG, accounts for 16% of total GHG emissions in the world (IPCC, 2014). The impact of CH_4 on climate change is 25 times higher than CO_2 over a 100 years time frame (United States Environmental Protection Agency, 2016). Anthropogenic activities like landfills, energy sectors (e.g., natural gas refineries) and anaerobic wastewater treatment units contribute to 60% of the global CH_4 emissions worldwide (Ménard et al., 2012a). Methane elimination in biofilters is an appropriate technique for CH_4 concentrations below 3% (v/v) (Brandt et al., 2016). In a biofilter, CH_4 is transferred from gas to biofilm phase to be degraded into less hazardous components like CO_2, water and biomass through biological reactions (Kennes et al., 2009). An important future challenge for CH_4 biological elimination is the stability of CH_4 biofilters (Zamir et al., 2014). Factors such as sudden inlet load (IL)'s variations or periodic absence of CH_4 can disturb the stability of the biofilter and lead to poor performance during transient state (Ferdowsi et al., 2016). In this regard, the periodic addition of a second pollutant like ethanol vapors to a CH_4 biofilter may also disturb the stability of the biofilter. Few studies used biofilters for a

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mixture of the gaseous pollutants (Ménard et al., 2012a). However, emissions with multiple pollutants is a common situation in industries (Dixit et al., 2012). The CH_4 leakage from anaerobic wastewater treatment plants of food industries can include ethanol (United States Environmental Protection Agency, 2016). Ethanol is also considered a hazardous component for humans and targeted for removal in biofilters (Christen et al., 2002; Dastous et al., 2008). Unlike CH_4 which has poor solubility (dimensionless Henry's law constant of 28 at 25 °C, P = 1 atm) (Staudinger and Roberts, 1996), ethanol is completely miscible with water with a low dimensionless Henry's constant (0.002 at 25 °C, P = 1 atm). Therefore, ethanol is more readily bioavailable in the biofilm phase compared to CH_4 under similar conditions in the biofilters (Mackay et al., 2006). Ethanol biofilters are usually subjected to EBRTs shorter than 1 min (Dastous et al., 2008). In contrast, EBRTs longer than 4 min for CH_4 biofilters can provide sufficient contact time between CH_4 and the biofilm phase and can increase the bioavailability of CH_4 in biofilm (Hernández et al., 2015; Lebrero et al., 2016). Although a number of studies focused on the removal of CH_4 or ethanol in biofilters, to our knowledge no study has looked at steady state and dynamic behaviors of biofilters when the both pollutants are present. Therefore, the steady state performance of the biofilter should be studied in order to choose an appropriate EBRT when CH_4 and ethanol are fed simultaneously. The dynamic behaviors of a CH_4 biofilter under periodic presence of ethanol at different EBRTs gives a better understanding of the phenomena happening during pollutant removal. Because of the low ethanol dimensionless Henry's law constant of 0.002, a fraction of the inlet ethanol may dissolve in the biofilm phase and is subsequently drained as lixiviate (Morotti et al., 2011). If the ethanol absorption exceeds the ethanol biodegradation, a dynamic equilibrium based on the ethanol accumulation can occur in the biofilm phase during ethanol biofiltration. On the other hand, when the ethanol addition is completed, the residue of the accumulated ethanol likely delays the recovery of the biofilter.

The present study aimed to investigate the steady state performance and transient behavior of a biofilter for CH_4 removal under periodic ethanol loadings. The effect of gas flow rate on the biofilter performance was studied for individual CH_4 removal as well as during elimination of a vapor mixture of CH_4 and ethanol. The continuous loading of CH_4 under ethanol intermittent loading may cause unfavorable transient conditions for the biofilter. The biofilter dynamic responses during ethanol addition as well as the biofilter recovery when ethanol addition stopped were studied.

2. Materials and methods

2.1. Experimental setup

The biofilter was made of Plexiglas with a diameter of 0.15 m and a total height of 1 m. The biofilter included three equal sections to provide a total volume of 18×10^{-3} m^3. An inorganic material with an average diameter of 12×10^{-3} m and a specific surface area of 310 m^2 m^{-3} was used as support media. The exact nature of the packing materials cannot be disclosed due to a confidentiality agreement. The gas samples including CH_4, ethanol and CO_2 were collected from four gas sampling ports along the biofilter. The feed to the up-flow biofilter was a mixture of CH_4, humid air and ethanol. Methane stream was provided from a CH_4 cylinder (Praxair Inc., Canada) with a regulated pressure of 275 kPa. Humid air and ethanol vapors were produced by a humidification column and an ethanol bubbler respectively. The nutrient solution addition was fed for 1 min every day at a flow rate of 1.5 L min^{-1} in order to provide essential nutrients like nitrogen, phosphorous, potassium and copper for the biofilter's microbial culture. The characteristics of the nutrient solution were similar to the one used by Ménard et al. (2012b). Fig. S1 (Supplementary Materials) shows a schematic flow chart of the biofilter.

2.2. Microbial culture

The biofilter had been used for CH_4 elimination during four months (unpublished data). After a one week shutdown, the biofilter was restarted in order to begin the present study. Therefore, the microbial culture in the biofilter was already adapted to CH_4 removal. The initial source of inoculation was from the leachate of a CH_4 biofilter (Ferdowsi et al., 2016).

2.3. Analytical methods

Methane and ethanol vapors concentration were measured by a total hydrocarbon analyzer (FIA 510, Horiba, USA). To analyze the pollutant's mixture, after measuring the total hydrocarbon concentration, CH_4 was temporarily removed from the biofiter and the ethanol concentration was measured. The CH_4 concentration was considered as the difference between the total hydrocarbon and ethanol concentrations. The CO_2 concentrations were determined by a CO_2 gas analyzer (Ultramat 22P, Siemens, Germany). The ethanol concentrations in the leachate were analyzed using total organic carbon analyzer (TOC-V_E, Shimadzu, Japan).

2.4. Performance parameters

The performance of the biofilter was quantified by removal efficiency (RE), inlet load (IL), elimination capacity (EC) and CO_2 production rate (P_{CO_2}) as described below:

$$\text{Removal efficiency(RE)} \quad \frac{(C_{Gi} - C_{Go})}{C_{Gi}} \times 100 \quad (\%) \tag{1}$$

$$\text{Inlet load(IL)} \quad \frac{Q \times C_{Gi}}{V_{bf}} \quad \left(g\ m^{-3}\ h^{-1}\right) \tag{2}$$

$$\text{Elimination capacity(EC)} \quad \frac{(C_{Gi} - C_{Go}) \times Q}{V_{bf}} \quad \left(g\ m^{-3}\ h^{-1}\right) \tag{3}$$

$$CO_2\ \text{production rate}(P_{CO_2}) \quad \frac{(CO_{2out} - CO_{2in}) \times Q}{V_{bf}} \quad \left(g\ m^{-3}\ h^{-1}\right) \tag{4}$$

In the equations above, C_{Gi} and C_{Go} are the inlet and outlet pollutants concentration ($g_{CH_4} m^{-3}$ or $g_{ethanol}$ m^{-3}) respectively, V_{bf} is the biofilter volume (m^3), Q is the gas flow rate (m^3 h^{-1}), CO_{2in} and CO_{2out} are the concentrations of CO_2 ($g_{CO_2} m^{-3}$) regarding to inlet and outlet of the biofilter respectively.

2.5. Methodology and experimental conditions

Table 1 summarizes the operating conditions and experimental steps of the biofilter. The biofilter ran under three different EBRTs of 6, 3 and 1.5 min corresponding to gas flow rates of 3, 6 and 12 L min^{-1} respectively for a period of 281 days. The CH_4 ILs were 33, 66 and 132 g_{CH4} m^{-3} h^{-1} corresponding to the EBRTs. Ethanol with an average concentration of 0.45 $g_{ethanol}$ m^{-3} was introduced to the biofilter during three separate cycles based on EBRTs of 6, 3 and 1.5 min with corresponding ILs of 4.5, 9 and 18 $g_{ethanol}$ m^{-3} h^{-1}