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Methane oxidation and attenuation of sulphur compounds in landfill top cover systems: Lab-scale tests

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ABSTRACT

In this study, a top cover system is investigated as a control for emissions during the aftercare of new landfills and for old landfills where biogas energy production might not be profitable. Different materials were studied as landfill cover system in lab-scale columns: mechanical–biological pretreated municipal solid waste (MBP); mechanical–biological pretreated biowaste (PB); fine (PBS_f) and coarse (PBS_c) mechanical–biological pretreated mixtures of biowaste and sewage sludge, and natural soil (NS). The effectiveness of these materials in removing methane and sulphur compounds from a gas stream was tested, even coupled with activated carbon membranes. Concentrations of CO_2, CH_4, O_2, N_2, H_2S and mercaptans were analysed at different depths along the columns. Methane degradation was assessed using mass balance and the results were expressed in terms of methane oxidation rate (MOR). The highest maximum and mean MOR were observed for MBP (17.2 g $CH_4/m^2/hr$ and 10.3 g $CH_4/m^2/hr$, respectively). Similar values were obtained with PB and PBS_c. The lowest values of MOR were obtained for NS (6.7 g $CH_4/m^2/hr$) and PBS_f (3.6 g $CH_4/m^2/hr$), which may be due to their low organic content and void index, respectively. Activated membranes with high load capacity did not seem to have an influence on the methane oxidation process: MBP coupled with 220 g/m^2 and 360 g/m^2 membranes gave maximum MOR of 16.5 g $CH_4/m^2/hr$ and 17.4 g $CH_4/m^2/hr$, respectively. Activated carbon membranes proved to be very effective on H_2S adsorption. Furthermore, carbonyl sulphide, ethyl mercaptan and isopropyl mercaptan seemed to be easily absorbed by the filling materials.

Introduction

In recent years, there has been a large increase of levels of greenhouse gases (*i.e.*, CH_4, CO_2, N_2O) in the atmosphere (IPCC, 2013). CH_4 global warming potential is 28 times higher than CO_2 (IPCC, 2014), although its residence time in the atmosphere is relatively short in comparison with both CO_2 and N_2O (7–8 years) (Chanton and Liptay, 2000). Whilst methane can be naturally released into the atmosphere, 50%–60% of the total CH_4 emissions are due to anthropogenic activities (*e.g.*, fossil fuel extraction and use, rice paddy agriculture, ruminant livestock, landfills, man-made lakes and wetlands and waste treatment) (IPCC, 2013).

Methane emissions could be reduced through appropriate management of anthropogenic activities (Chanton et al., 2010). Landfills are one of the main anthropogenic biogenic emissions of CH_4 (Yang et al., 2015), and still one of the most common waste management option globally (Ziyang et al., 2015).

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Biologically breaking down processes of waste components in landfills generate gas (Widory et al., 2012). The production of landfill gas depends on the quantity and composition of the disposed waste (Chanton and Liptay, 2000), *i.e.*, the content of organic waste and its degradability. In general, landfill gas is mainly composed by CH_4 and CO_2 (50%–60% and 40%–45%, respectively), although N_2, O_2, H_2S, NH_3 and more than two hundred organic compounds are also present (Beylot et al., 2013). Moreno et al. (2014) directly measured landfill leaks with a portable detector, finding that they were characterised by high H_2S/NH_3 ratios. These researchers pointed to H_2S, NH_3 and organic compounds as responsible for landfill odours; H_2S being a major contributor (He et al., 2012). Landfill gas production also depends on waste age and other parameters, such as temperature, water content, nutrients, and inhibiting compounds (Scheutz et al., 2011). Young landfills (5–30 years) produce gas with a higher methane content (30%–60%) than older landfills (Karthikeyan et al., 2016).

Gas production in new landfills for pretreated municipal solid waste might be reduced by up to 90% of what expected in traditional landfills (Leikam and Stegmann, 1997). Although different alternatives are available to reduce landfill emissions (*e.g.*, waste pretreatment, aeration and flushing), not all residual emissions are avoided (Cossu et al., 2003; Raga et al., 2015). In large modern landfills, gas capture reduces methane emissions into the atmosphere (Chanton et al., 2010). Nevertheless, small and medium-sized landfills do not usually treat their emissions (Chiemchaisri et al., 2010). This might be due to several reasons. For instance, the low content of methane in the gas may not make profitable the capture and use of the gas for energy purposes (Einola et al., 2009).

Gas leaks must be efficiently controlled. According to Beylot et al. (2013), landfills without biogas management pose the highest potential impact on climate change, human toxicity and ecotoxicity. Soil cover makes it possible to control landfill emissions (He et al., 2012; Lü et al., 2012; Mei et al., 2011); CH_4 being degraded by naturally-occurring bacteria through a reaction with atmospheric O_2, to form CO_2 (Chiemchaisri et al., 2010). Moreover, cover materials in landfills support vegetation growth, optimise the water balance and reduce odours and water infiltration (Rachor et al., 2011; Ziyang et al., 2015). Landfill covers may be considered as a low-cost solution and be adapted to different cases in other types of landfills, so that it may be used as a unique gas treatment or combined with other methods (Einola et al., 2009).

Some materials, such as agricultural and horticultural soil, compost, sand and mechanically–biologically treated municipal solid waste have been studied as landfill cover soils (Hu and Long, 2016). Oxidation rate of methane and other pollutants in landfill gas depends on the type of cover material (Table 1); current research being mainly focused on methane (Beylot et al., 2013). Nonetheless, H_2S migration in landfill covers is complex and differs from that of CH_4 and CO_2 (Xu et al., 2014). Thus, further research is needed to study the attenuation of sulphur compounds by landfill covers.

The main aim of the present research was to investigate the behaviour of different kinds of materials as possible landfill cover system, even coupled with activated carbon membranes of different load capacities, concerning not only methane oxidation, but also the adsorption of H_2S and sulphur compounds.

Table 1 – Methane oxidation rate (MOR) of several landfill cover materials found in literature.

Type of material	MOR ($g\ CH_4/m^2/hr$)	Reference
Coarse sand	7.0	Kightley et al. (1995)
Fine sand	4.6	
Clay topsoil	4.6	
Municipal solid waste compost	5.7–18	Humer and Lechner (1999)
Sewage sludge compost	0.1–7.2	
Topsoil	2.9–3.6	
Landfill loam	8.8	Scheutz et al. (2003)
Mineral sand	1.5–2.3	Rachor et al. (2011)
Sediment (loamy sand)	0.2–0.6	
Screened garden waste c ompost	5	Pedersen et al. (2011)
Sewage sludge compost	4.7	
Unscreened 4-year-old garden waste compost	4.5	

1. Materials and methods

1.1. Experimental equipment

In this study, laboratory tests were carried out using plexiglas columns (height 100 cm, diameter 10 cm) installed in a climate chamber to ensure a constant temperature of 30°C (Fig. 1). The columns were filled with different materials as possible landfill cover systems (hereinafter referred to as filling material).

Hu and Long (2016) studied the effect of different cover thickness on methane on CH_4 oxidation; 90 cm being found as the optimum value in the initial 1- to 24-day stages and 60 cm in the 25- to 50-day stage. In the present study, the testing materials were filled into a depth of 60 cm and saturated in water to about 50% of the field capacity. Conditions in the top cover of a landfill were simulated by supplying biogas from the bottom of the columns and air from the top (Fig. 1). Thus, oxygen and methane from these two gas streams could penetrate the substrate as under natural landfill conditions. The biogas used in the tests was collected at a municipal solid waste (MSW) landfill and stored in Tedlar bags of 100 L capacity. The biogas was fed into the columns by means of N86 KT.18 membrane pumps, the flow rate being adjusted by means of 5850S digital flowmeters (Brooks Instruments). Air was blown using Air Professional 360 pumps (Prodac) and the flow rate was controlled by Sho-Rate GT1335 flowmeters (Brooks Instruments).

Sampling points were inserted at different depths along the lateral surface of the columns (Fig. 1). In these points, CO_2, CH_4, O_2 and N_2 concentrations were determined by means of a mobile landfill gas meter (LFG20, Eco-Control S.r.l.). H_2S and mercaptans (carbonyl sulphide, ethyl mercaptan and isopropyl mercaptan) were analysed on a HP5890 gas-chromatograph equipped with an HP-Plot Q column (Agilent Technologies). The temperature inside the column was also measured by means of Thermo Systems TS100 temperature probes.

Mass balance was the method applied to quantify and assess methane degradation process in the five different types of landfill covers. The results were expressed in terms of

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