Role of Landfill Cover Materials in Mitigating GHG Emissions in Biogeochemical Landfill Cover System

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ABSTRACT

Municipal solid waste (MSW) landfills are known to be one of the major sources of greenhouse gas (GHG) emissions into the atmosphere. In order to alleviate these emissions, an innovative biogeochemical cover system is proposed to mitigate both methane (CH_4) and carbon dioxide (CO_2) emissions, which are the predominant gases in landfill gas (LFG) emissions. This paper investigates four materials: soil, non-activated biochar, methanotrophic activated biochar, and basic oxygen furnace (BOF) slag for their CH_4 and CO_2 uptake capacity. First, the physical and chemical properties of the four materials were tested. Thereafter, several series of batch tests were conducted to determine CH_4 and CO_2 uptake by each material. The results demonstrate that the soil has the potential to oxidize CH_4 into CO_2 due to presence of CH_4 oxidizing (methanotrophic) bacteria, while the BOF steel slag has potential to sequester CO_2. The methanotrophic activated biochar showed enhanced biological activity due to high methanotrophic population, mitigating CH_4 efficiently. However, the non-activated biochar had little to no effect on the uptake of either CH_4 or CO_2. Finally, the combination of these cover materials at different proportions in different configurations is being investigated to optimize the biogeochemical cover system to mitigate both CH_4 and CO_2 emissions.

KEYWORDS: Activated biochar; biogeochemical cover; BOF steel slag; biochar; biocover; carbon dioxide sequestration; landfill gas; methanotrophs; methane oxidation

INTRODUCTION

Municipal solid waste (MSW) landfills are regarded as the third largest anthropogenic source of methane (CH_4) emissions in the United States. The landfill gas (LFG), generated due to anaerobic biodegradation of organic fraction in MSW, typically comprises of 50% CH_4 and 50% CO_2, both of which are greenhouse gases impacting global climate change. The CH_4 emissions from the landfills are known to be partially converted to CO_2 by the naturally available CH_4 oxidizing bacteria (methanotrophs) present in the cover soil. For nearly two decades, many investigators investigated the CH_4 oxidation capacity of the landfill cover soils based on batch tests and small-scale to near full-scale column studies tests to field-scale test plots (Sadasivam and Reddy 2014). To further improve CH_4 oxidation and mitigate CH_4 emissions from landfills, organic amendments to the cover soil have also been proposed and investigated in recent years (Stern et al. 2007, Scheutz et al. 2011, Sadasivam and Reddy 2014). Due to the degradation potential of organic-rich materials such as compost in the landfill cover soils, the use of alternative stable materials such as biochar, which is a stable and recalcitrant material to microbial degradation, is proposed for the long-term application (Yargicoglu and Reddy 2017a). Biochar is a solid product resulting from pyrolysis or gasification of organic wastes feed stocks such as waste wood, switchgrass, and corn stove, during bioenergy production. Recent studies have demonstrated that biochar derived from waste wood has great potential to oxidize CH_4 into CO_2 in the landfill covers (Reddy et al. 2014, Yargicoglu and Reddy 2017a). In spite of addressing CH_4 emissions, not much consideration has been given to control landfill CO_2 emissions, that typically range between 40 - 50% of the total landfill gas, and also the CH_4 oxidized CO_2 emissions.

The application of BOF slag as drainage material in landfill cover has been reported with respect to its geotechnical properties (Diener et al. 2010, Andreas et al. 2005). Furthermore, its application is widely being used in construction industry as an aggregate material and in environmental engineering applications as media for contaminant adsorption and CO_2 sequestration. It is investigated in treating heavy metals and TCE in soil and groundwater, phosphate removal from wastewater, and soil conditioner/fertilizer in agriculture as reviewed by Reddy et al. (2019). Recently, Reddy et al. (2018a) proposed the concept of an innovative biogeochemical cover to mitigate both CH_4 and CO_2 emissions from the landfills. Wherein, BOF slag, a byproduct from steel mills, is proposed for CO_2 sequestration due to the presence of various minerals such as CaO, portlandite ($Ca(OH)_2$) and larnite (Ca_2SiO_4) (Huijgen et al. 2005). The use of BOF slag as one of the landfill cover material in mitigating CO_2 emissions has not been considered to date. The BOF slag is proposed to be used along with other materials such as soil and biochar in the biogeochemical cover system in an optimal way to mitigate both CH_4 and CO_2 emissions.

This paper provides a brief overview of the concept of biogeochemical cover and then presents several series of batch experiments conducted to systematically evaluate the extent of CH_4 oxidation and CO_2 sequestration by the potential materials that could be used in it, specifically soil, BOF slag, biochar, and methanotrophic activated biochar.

BIOGEOCHEMICAL COVER CONCEPT

Biogeochemical cover is an innovative, low-cost landfill cover system consisting of steel slag in combination with soil and biochar (Reddy et al. 2018a). Steel slag is a co-product of steel making process, and basic oxygen furnace (BOF) slag is a type of steel slag, which is rich in alkaline minerals such as CaO, MgO, etc. The alkaline metal oxides present in the slag react with CO_2, forming stable carbonates. Many studies have explored the carbonation potential of steel slag for the mineral CO_2 sequestration for different industrial applications. Moreover, several past studies have shown promising potential of biochar-amended soil to mitigate CH_4 emissions by the enhanced methanotrophic oxidation of CH_4 (Reddy et al. 2014; Yargicoglu and Reddy 2017b).

The biogeochemical cover aims to combine the carbonation potential of BOF slag along with the methanotrophic CH_4 oxidation potential of biochar-amended soil to mitigate both CH_4 and CO_2 emissions from the MSW landfills, ultimately leading to “Zero Emissions Landfill Cover”. **Figure 1** shows the schematic of this steel slag and biochar-amended soil biogeochemical cover system. The proposed biogeochemical cover also has the potential to sequester hydrogen sulfide (H_2S) if present in the LFG as shown in **Figure 1**. The use of proposed biogeochemical cover in landfills will not only reduce the environmental concerns associated with the fugitive LFG emissions, but also provides new opportunity for the sustainable management of steel slags (especially finer slag) which are generally stockpiled in the steel industry or landfilled.

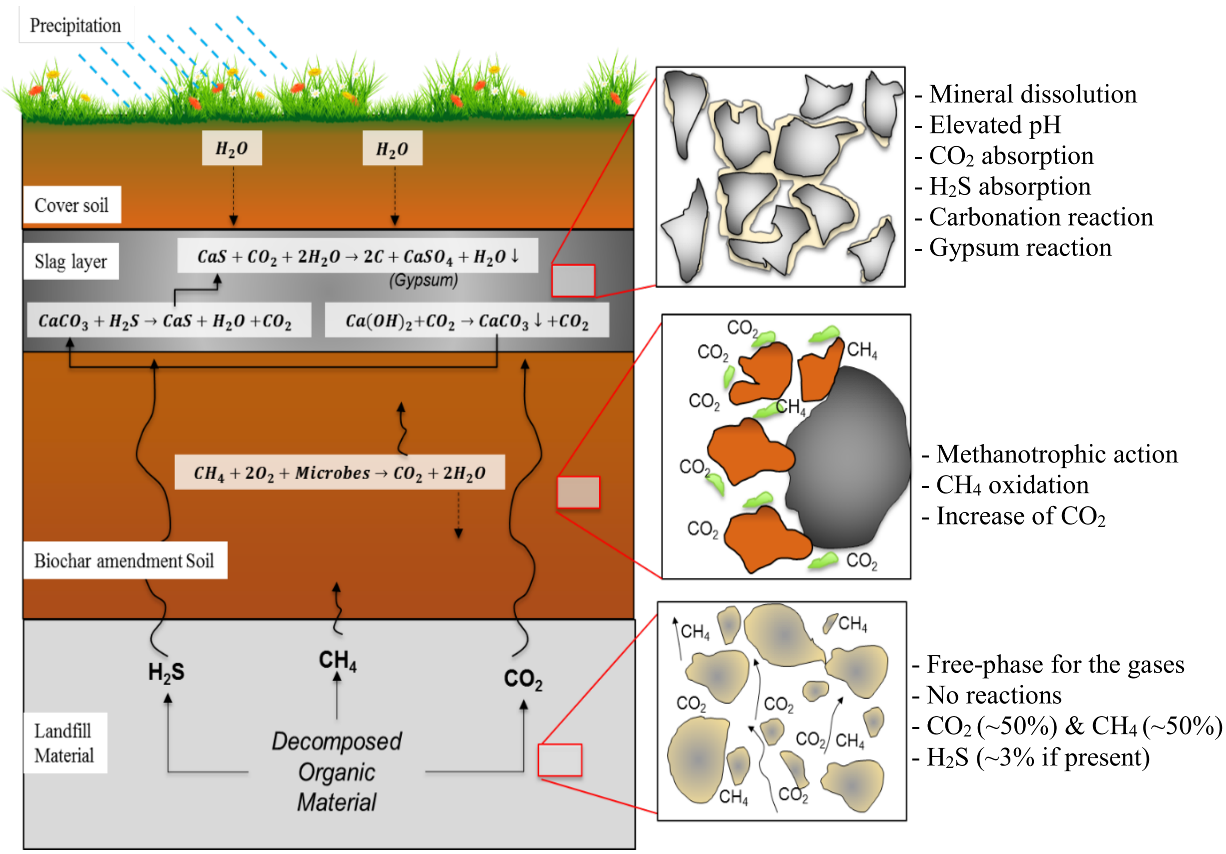

Figure 1: Schematic of biogeochemical cover system for zero emissions (Reddy et al. 2018a)

Although the proposed biogeochemical cover offers wide range of environmental as well as economic benefits, it is of utmost importance to analyze various system factors, which are crucial to the functioning of the coupled biogeochemical processes. A comprehensive laboratory testing program consisting of multiple tasks is undertaken for this purpose; this study presents the results from one of these tasks.

MATERIALS

Soil

Soil was collected from Zion landfill site, located in Zion, Illinois, USA. Soil samples were collected from an interim cover at a depth of ~1 to 2 feet and were shipped to the Geotechnical and Geoenvironmental Engineering Laboratory at the University of Illinois at Chicago (UIC) where it was stored at room temperature (23 ± 2°C). Soil samples were air dried (moisture content <0.5%), pulverized, and screened through a 2 mm sieve prior to conducting the experiments.

Biochar

Biochar was obtained from a commercial vendor in Illinois, USA. The biochar used in this study, designated as CE-WP2, was produced from waste pinewood subjected to gasification at a high temperature of ~520°C. In this study, biochar in pellet form was used with fines sieved and discarded. The biochar was oven-dried at 105°C to remove any moisture content before conducting the experiments.

BOF Slag

The BOF slag used in this experiment was obtained from Indiana Harbor East (IHE) of Arcelor Mittal steel plant, located in East Chicago, Indiana, USA. This slag, designated as IHE 9/17, is finer material being stockpiled at the plant; otherwise, requires landfill disposal. All the tests were performed using the bulk slag sample as obtained from the plant. The steel slag was also oven-dried at 105°C prior to conducting the experiments.

METHODOLOGY

Materials Properties Testing

All the physical and chemical properties of the cover materials selected for this study were tested as per ASTM standards. ASTM D422 and ASTM D854 were the testing protocol followed for characterizing particle size distribution and specific gravity, respectively. Dry density was determined based on weight of the dry material compacted in the permeameter and volume of the permeameter. Hydraulic conductivity was tested per the ASTM D2434. The water holding capacity (WHC) of the material was conducted by placing a known mass of sample in a funnel lined with Whatman filter paper and adding known amount of deionized water. The sample was allowed to soak for 2-3 hours and drain under gravity. The WHC of the material was determined by calculating the moisture content retained by the sample (Yargicoglu et al. 2015). For chemical characterization, 10 g of each material under investigation was soaked in 0.01M $CaCl_2$ solution (L/S of 1:1) for 2 hours and pH, ORP and electrical conductivity were measured as per ASTM D4972. The pH meter was calibrated with standard buffers of pH 4, 7 and 10 prior to measurement. Organic matter content was determined based on loss-on-ignition (LOI) method as per ASTM D2974. All tests on each material were conducted in triplicate, and the results were averaged.

Mixed Methanotrophic Culture Consortium

The mixed methanotrophic culture was cultivated in the laboratory using enrichments from the landfill cover soil as described in Rai et al. (2018). The biochar was activated by inoculating 5-7 g of biochar in 10 mL of the mixed culture in the presence of ~5 - 6% CH_4 (v/v) and ~5 - 6% CO_2 (v/v) balanced in air and incubated at room temperature of 23ºC.

Batch Tests

For the batch testing, 10 g of the selected material (soil, biochar, methanotrophic activated biochar, or BOF slag) was placed in 125 mL-serum vials and the moisture was adjusted to 20% (w/w) using deionized water, except methanotrophic activated biochar that was soaked in the culture. The vials were sealed airtight using butyl rubber septa followed by crimp cap. 20 mL of air from the headspace was replaced with equal volume of synthetic landfill gas comprising of 50% (v/v) CH_4 and 50% (v/v) CO_2 to achieve a headspace concentration of ~5 - 6% CH_4 (v/v), ~5 - 6% CO_2 (v/v) and a balance (~88 - 90%) of air. The change in the headspace concentration was determined by collecting and analyzing the gas samples on a regular basis using gas chromatography (GC) until the headspace concentration dropped to less than 1%. All the experiments were conducted in triplicate along with the controls (with synthetic landfill gas without any material). The controls using soil (sterilized for 2 hours using Napco Model 8000-DSE autoclave) were also tested to discern the effects of any microbial activity in the soil. The CH_4 oxidation rates were calculated from the linear regression analysis of CH_4 concentration versus elapsed time, based on the zero-order kinetics.

Gas Analysis

The gas samples were analyzed at regular time intervals and analyzed for CH_4 and CO_2 concentrations using an SRI 9300 GC equipped with a thermal conductivity detector (TCD) and CTR-1 column that separates N_2 and O_2 for simultaneous analysis of CO_2, CH_4, O_2 and N_2. Gas samples were withdrawn using 1 mL syringe where 0.5 mL of the sample was discarded and remaining 0.5 mL was injected into the GC to reduce any pressure effects due to sampling. A calibration curve for a minimum of three points was established using high purity standard gas mixtures ranging from 1% to 50% CH_4 and CO_2.

RESULTS AND DISCUSSION

Table 1 summarizes the physical and chemical properties of the cover materials tested. Based on sieve analysis, BOF slag and biochar consisted of 74% and 54% of sand-size fraction, respectively, and were classified as poorly graded sand (SP or SP-SM) equivalent as per the Unified Soil Classification System (USCS). Whereas, the cover soil consisted of more than 50% fines with plasticity index of 17%, and classified as silty clay (CL). The materials showed slightly acidic to highly alkaline pH, measuring 6.7, 7.6 and 12.4 for biochar, soil and BOF slag, respectively. The organic content was found to be 96.7% in biochar, 1.6% in BOF slag, and 5.8% in cover soil. The negative oxidation reduction potential (ORP) values indicates higher reduction potential in the order of BOF slag (-313.3 mV), soil (-53.8 mV), and biochar (-6.3 mV). The water holding capacity was found to be 51.6% for biochar, 43% for soil, and 20% for BOF slag. The hydraulic conductivity of the soil was 5.4×10^{-8} cm/s which qualifies as a low permeable material, whereas BOF slag and biochar possessed high hydraulic conductivity of 1.1×10^{-3} and 2×10^{-4} cm/s, respectively. The specific gravity of BOF slag was 3.5, high due to high iron oxide content, and for biochar was 0.6, and for the soil was 2.57. The relatively lower specific gravity of the soil as compared to typical inorganic soils is due to its organic content of 5.8%. The high organic content implies that the soil is rich in biomass and can sustain microbial activity.

Table 1: Physical and chemical characteristics of BOF slag, cover soil and biochar

Properties	ASTM Method	BOF Slag	Soil	Biochar
Grain Size Distribution:	D422			
Gravel (%)		20.8	3.7	45
Sand (%)		74.2	14.7	54
Fines (%)		4.9	81.9	1
D_{50} (mm)			0.009	4.3
C_c		0.7	-	0.82
C_u		18	-	2.42
Atterberg Limits:	D4318			
Liquid Limit (%)		Non-	39	Non-
Plastic Limit (%)		Plastic	22	Plastic
Plasticity Index (%)			17	
USCS Classification	D2487	SP-SM	CL	SP
Specific Gravity	D854	3.5	2.57	0.6
Dry Density (g/cm^3)		1.72	1.8	1.15
Hydraulic Conductivity (cm/s)	D2434	1.1×10^{-3}	5.4×10^{-8}	2×10^{-4}
Loss-on-Ignition (%)	D2974	1.6	5.8	96.71
pH (1:1)	D4972	12.4	7.6	6.5
Electrical Conductivity (mS/cm)	D4972	13.3	0.55	0.8
Redox Potential (mV)	D4972	-313.3	-53.8	-6.3

C_c=Coefficient of curvature; C_u=Coefficient of uniformity

Figure 2 shows the plot of CH_4 and CO_2 gas uptake with time in batch tests with landfill cover soil. An increase in the CH_4 uptake with time confirms CH_4 oxidation by the CH_4 oxidizing bacteria in the cover soil. This observation was further bolstered with the observed no significant changes in gas concentrations in the controls (sterilized soil and LFG), thus confirming the CH_4 oxidation by the naturally existing CH_4 oxidizing bacteria in the cover soil. A minimal CO_2 adsorption by the cover soil with an uptake of 12% was noticed, after which an increase in the CO_2 levels, as a result of CH_4 oxidation, was observed. The CH_4 oxidation rate calculated based on zero-order kinetics is found to be 4.1 µg CH_4/g/h. Overall, the results suggest that the landfill cover soil used in this study was rich in CH_4 oxidizing bacteria that were able to perform CH_4 oxidation.

Many reported studies, involving laboratory batch experiments, have also shown significant CH_4 oxidation in the landfill cover soils (Scheutz et al. 2009; Sadasivam and Reddy 2014). At 5% (v/v) CH_4 concentration, studies have shown that the CH_4 oxidation rates can range from 0.0096 µg CH_4/g/h (Bender and Conrad 1994) to 173 µg CH_4/g/h (Borjesson et al.1998a, b). The results obtained in this study were in agreement with the results from these studies; however, many other studies have reported the CH_4 oxidation rates to be as low as 0.0024 µg CH_4/g/h (Boeckx et al. 1996) and as high as 118 µg CH_4/g/h (Scheutz and Kjeldsen 2004), showing differences in the CH_4 oxidation rates mainly due to variances in the experimental and site-specific conditions.

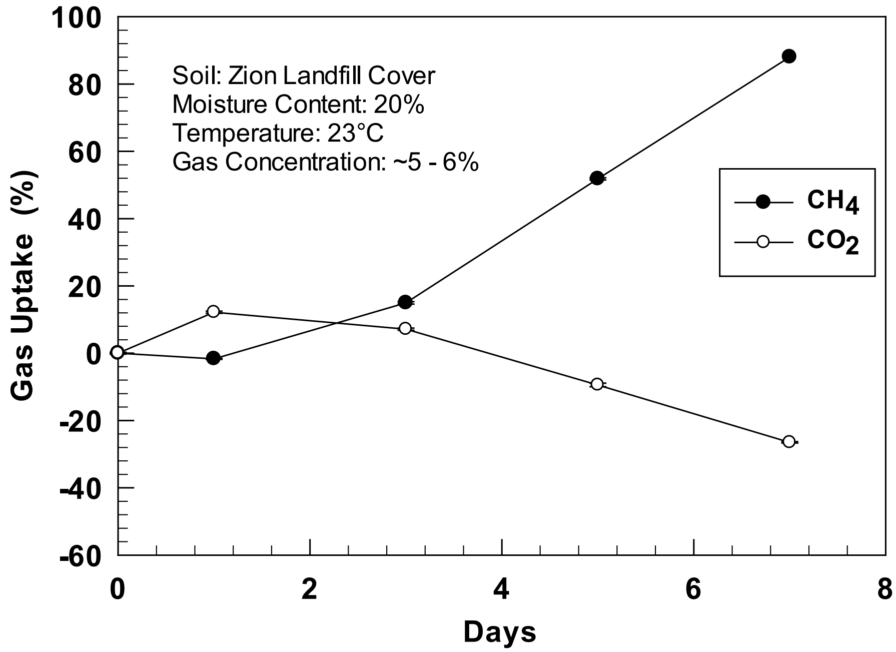

Figure 2: Methane and carbon dioxide uptake in landfill cover soil

Figure 3 shows the plot of CH_4 and CO_2 gas uptake with time in biochar. As biochar is free of any methanotrophs, the reduction in gas concentrations are presumed to be due to adsorption processes. The results show marginal adsorption of CH_4 on the biochar with a total CH_4 removal of 9.6%. The physical and chemical properties of the biochar are usually dictated by the feedstock and production processes (Yargicoglu et al. 2015). The CH_4 adsorption capacity also varies depending upon the type of biochar used. Sadasivam and Reddy (2015) reported differences in the CH_4 adsorption capacity in seven wood-derived biochars and granulated activated carbon (GAC) concluding minimal CH_4 adsorbing capacity in biochars (0.04 - 0.18 mol/kg) when compared to GAC. In contrary, a study by Sethupathi et al. (2017) demonstrated no adsorption of CH_4 in four different types of biochar studied, suggesting the adsorption capacity of the biochar is highly dependent on its feedstock and its physicochemical properties.

Furthermore, the results in **Figure 3** showed significant CO_2 removal in the first 24 hours with an uptake of 21%, but showed desorption in the consecutive days reaching an equilibrium after 5th day with an overall CO_2 removal of 5.3%. Biochar showed desorption followed by adsorption which could likely be due to the shaking of the vials before sampling or depressurization of the system due to sampling, resulting in the breakage of the weak intermolecular forces causing physisorption of CO_2 (Sethupathi et al. 2017).

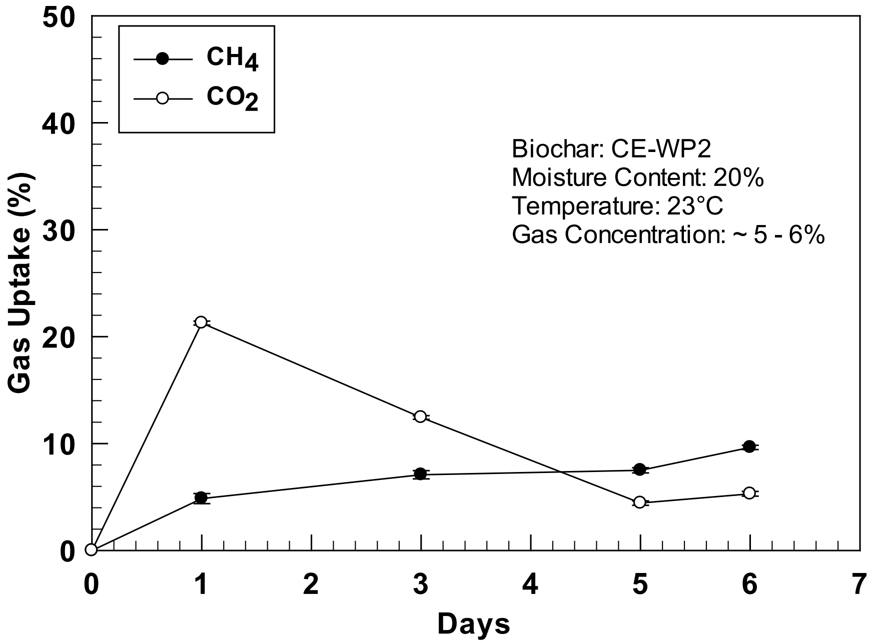

Figure 3: Methane and carbon dioxide uptake in biochar

Overall, these results demonstrate marginal adsorption of both CH_4 and CO_2 in the biochar. However, biochar is known to have potential in various agricultural and environmental applications due to its unique physicochemical properties such as water holding capacity, internal porosity, and surface area when amended with soil (Yargicoglu et al. 2015). It also has positive impacts on soil fertility including increasing soil pH, nutrient retention and cation exchange capacity (Chan et al. 2007). The effect of biochar amendment to the landfill cover soil is also under investigation by our research team, but it is beyond the scope of this paper.

Figure 4 shows the plot of CH_4 and CO_2 gas uptake with time in BOF slag. The trend in the CH_4 concentration shows minimal CH_4 adsorption capacity by the BOF slag with an uptake of 3.6%. However, significant removal of carbon dioxide with 75% of CO_2 uptake in 24 hours and 100% uptake in 5 days was observed. These results are consistent with the studies on CO_2 sequestration by BOF slag as discussed by Reddy et al. (2018b). The BOF slag is a highly reactive material due to the presence of high CaO (> 35%), making it conducive for CO_2 sequestration (Su et al. 2016). Due to the high alkaline nature of the BOF slag (pH 12.4), it is hypothesized that the BOF slag could induce negative impact on the CH_4 oxidation when amended with soil or biochar-amended soil. To confirm this hypothesis, the BOF slag in various combinations (mixed versus separated) with soil and biochar-amended soil are under investigation. The results from these investigations will be used for the design of a geochemical cover profile configuration for an effective CH_4 oxidation and simultaneous CO_2 sequestration.

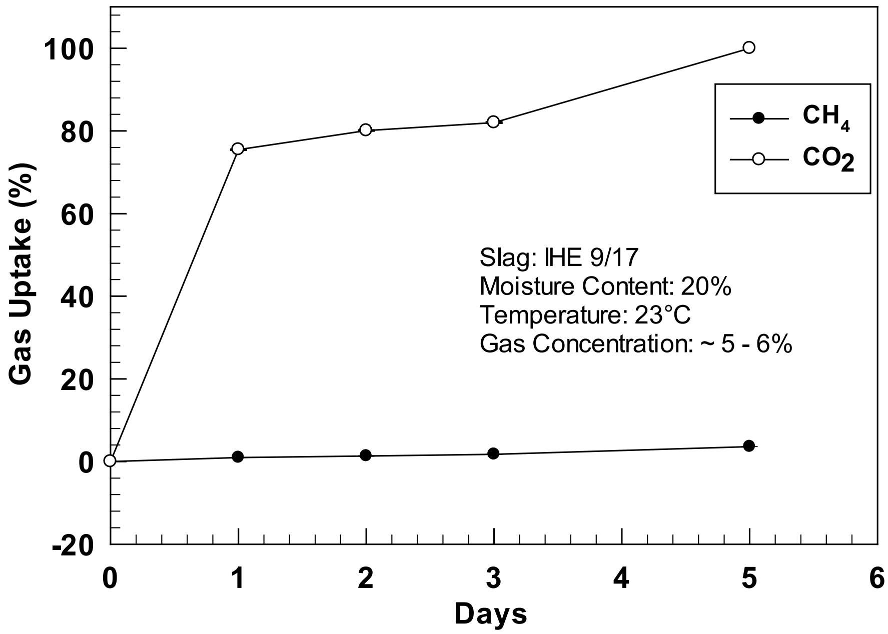

Figure 4: Methane and carbon dioxide uptake in BOF Slag

Biochar-amended soils have shown promising results in enhancing CH_4 oxidation in the landfills due to its favorable characteristics such as improved gas retention, water holding capacity, and habitable sites for proliferation of microbes as described by Reddy et al. (2014). However, the colonization of bacteria in the biochar takes longer time for acclimatizing and further improve oxidation rates. In this regard, this study evaluated the potential of methanotrophic activated biochar in the removal of CH_4. The plot of CH_4 and CO_2 gas uptake with time in methanotrophic activated biochar is shown in **Figure 5.**

An increase in the CH_4 uptake with time, as shown in **Figure 5**, is attributed to the performance of CH_4 oxidizing bacteria inoculated in the biochar. No significant CO_2 uptake by the methanotrophic activated biochar was detected, but increase in the CO_2 levels due to CH_4 oxidation by the CH_4 oxidizing bacteria was observed. Overall, the results suggest that the methanotrophic activated biochar had colonized in the highly porous, large surface area of biochar and were able to oxidize CH_4 without limitation to nutrients, showing CH_4 oxidation rate of 3.35 µg CH_4/g-biochar/h. We hypothesize that the methanotrophic activated biochar when amended with soil would mitigate CH_4 at faster rates when compared to non-activated biochar-amended soils. This study is being investigated by our research team and is beyond the scope of this paper.

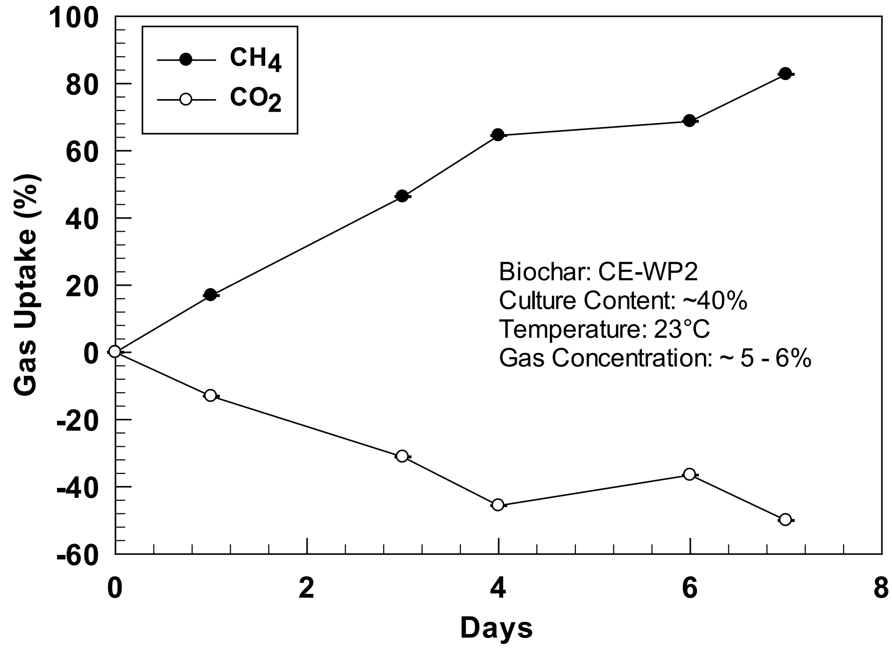

Figure 5: Methane and carbon dioxide uptake in methanotrophic activated biochar

CONCLUSIONS

Laboratory investigation on the landfill cover materials was conducted to evaluate the CH_4 and CO_2 uptake in order to determine their use in the newly proposed biogeochemical landfill cover system. The materials tested included: landfill cover soil, non-activated biochar, methanotrophic activated biochar, and BOF-slag. The results demonstrated that the landfill cover soil was dominated by the CH_4 oxidizing bacteria and were responsible for CH_4 oxidation. The non-activated biochar showed no CH_4 oxidation but showed low adsorption of CH_4 (9.6%) and CO_2 (5.3%). However, the methanotrophic activated biochar displayed substantial potential for mitigating CH_4, suggesting the use of biochar as a habitat for microbial community thus improving CH_4 oxidation. Furthermore, the BOF slag showed minimal uptake of CH_4 (3.6%) but demonstrated significant removal of CO_2 (100%), suggesting its use in the landfill cover system for CO_2 sequestration. Finally, the use of BOF slag in conjunction with soil, biochar-amended soil or methanotrophic activated biochar-amended soil, is under detailed investigation in order to develop a cover profile that best suited for effective CH_4 oxidation and simultaneous CO_2 sequestration.

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