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Methane Oxidation in Landfill Cover Soil: Case Study in Thailand

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Abstract— **The combination of flux measurements and gas profiles were used to calculate an estimate of the methane oxidation rate in the landfill cover soil as well as their oxidation factors. The calculation showed that the oxidation factors were between 0.10 and 0.58 which most observed OXs were greater than the default value in the IPCC Waste Model (OX=0.10). Based on this study, CH_4 emission from solid waste disposal sites could be significantly reduced by the utilization of appropriate cover soil type and condition with high CH_4 oxidation capacity.**

Keywords-methane oxidation; landfill; landfill cover soil; flux chamber

I. INTRODUCTION

Landfill gas (LFG) emissions are the great relevance to the global warming, particularly due to the methane content. Over a 100-year span, methane has a global warming potential of 21 because of its stronger molar absorption coefficient for infra red radiation and longer residence time in the atmosphere [1]. Methods for managing municipal solid waste (MSW) vary widely, ranging from open dumps, burning to sanitary landfills. Two main alternatives exist for managing CH_4 emission from landfill sites [2]. One option is to undertake landfill gas recovery with associated gas use, and is generally regarded as being the superior choice. The alternative option is that of encouraging CH_4 oxidation in the soil covering the landfill. Landfill cover soils provide a suitable environment where the transformation of methane to carbon dioxide, by means of methane oxidation can occur. This is a much cheaper and more effective option for reducing emissions in smaller and older landfills with lower amounts of CH_4 generation, compared with gas extracting, which becomes inefficient at low CH_4 contents. The top soil of a landfill is a dynamic mixing zone for air and landfill gas. Oxygen and nitrogen concentrations decrease with depth, while CH_4 and CO_2 increase with depth [3, 4].

Recent studies suggest that CH_4 oxidation by aerobic bacteria plays a significant role in the rate of CH_4 emissions from landfills. The rate of oxidation depends on both biochemical and physical processes in the soil [5]. In the presence of oxygen (O_2), CH_4 can be degraded by methanotrophic bacteria. Since methanotrophs produce an enzyme, methane mono-oxygenase (MMO), that has a very broad range of oxidation capacities [6]. The process leads to generation of CO_2 and water:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \quad (1)$$

This process leads to a substantial decrease in the CH_4/CO_2 ratio in the soil gas. The determination of CH_4/CO_2 ratio was determined with the distance from landfill by Ward et al. (1996) [7]. From this study, with increasing distance, the decrease in the ratio of CH_4 and CO_2 suggests CH_4 oxidation. CH_4 oxidation can range from negligible to 100 percent of internally produced CH_4. The thickness, physical properties and moisture content of cover soils directly affect CH_4 oxidation [8]. Boeckx et al., 1996 [3] examined the influences of moisture contents and soil temperature on the CH_4 uptake capacity of the neutral landfill cover soil. Soil moisture contents of 10 to 25% w/w gave a maximum CH_4 oxidation rate. When wetter, CH_4 consumption is slower because of limited gas diffusion. At lower soil moisture, microbial activity was reduced and consequently the oxidation capacity decreased. In addition, the optimum temperature was between 20°C and 30°C.

The "2006 IPCC Guidelines for National Greenhouse Gas Inventories" is the revised and newest guideline for estimating greenhouse gas (GHG) emissions from anthropogenic sources at the national level. GHG emissions from several sources, including solid waste disposal sites, can also be estimated using this guideline at each specific site. In this model, methane oxidation factor (OX) is one of parameters which can give a huge uncertainty to the estimation. Because it is difficult to measure, varies considerably with the thickness and nature of the cover material, atmospheric conditions and climate, the flux of CH_4, and the escape of CH_4 through cracks/fissures in the cover material. Field and laboratory studies which determine oxidation of CH_4 only through uniform and homogeneous soil layers may lead to over-estimations of oxidation in landfill cover soils [9].

It is important to note that percentage oxidation is a function of source strength of CH_4, or the flux rate at the

cover soil/waste interface. Temporal and spatial variations in flux rates and oxidation rates could further complicate total CH_4 oxidation observed at a given [10]. In order to minimize the uncertainty from using of OX in IPCC Waste Model for CH_4 emission inventory, development for a new methodology that will be used for CH_4 oxidation investigation at solid waste disposal sites is need. This method should be a practical, time saving and cost saving practice. In this study, the combination of flux measurements and soil gas profiles was conducted and evaluated at some landfills locate in Thailand.

II. METHODOLOGY

A. Calculation of in-situ methane oxidation

A combination of flux measurements and gas profiles can be used to calculate an estimate of the methane oxidation in the cover soil [11]. It is assumed that no carbon dioxide is dissolved in the infiltrating water and that the production of gas in the soil is negligible. Under stationary conditions the total flux of LFG at the surface is equal to that at the bottom of the cover soil. The total flux of landfill gas is:

$$\begin{aligned}\phi \text{LFG} &= \phi \text{CH}_4 \text{ top} + \phi \text{CO}_2 \text{ top} \\ &= \phi \text{CH}_4 \text{ bottom} + \phi \text{CO}_2 \text{ bottom}\end{aligned} \quad (2)$$

Where ϕ is the flux in mol m^{-2} –h. Knowing the total flux of landfill gas at the surface and the CH_4/CO_2 ratio beneath cover soil. The flux of methane beneath the cover soil can be calculated:

$$\phi \text{CH}_4 \text{ bottom} = (\phi \text{CH}_4 \text{ top} + \phi \text{CO}_2 \text{ top}) \cdot [C_{CH4bottom} /(C_{CH4bottom} + C_{CO2bottom})] \quad (3)$$

The difference between the methane flux at the top and at the bottom is the amount of methane, which is oxidized within the cover soil, methane oxidation rate (MO):

$$\text{MO} = \phi \text{CH}_4 \text{ bottom} - \phi \text{CH}_4 \text{ top} \quad (4)$$

The ratio between the spatial total MO and spatial total ϕ CH_4 bottom can be expressed as oxidation factor (OX):

$$\text{OX} = \text{Spatial Total MO} / \text{Spatial Total } \phi \text{CH}_4 \text{ bottom} \quad (5)$$

B. CH_4 and CO_2 emission measurements using closed flux chamber technique

CH_4 and CO_2 emission rates from the landfill site surface in this study were determined using the static chamber technique. The chamber that used in all study sites was constructed with ϕ0.30 m. - PVC pipe, 1.00 m. in height having PVC cap at the top of chamber. To protect air intrusion, the chambers was sealed to the ground by firming clayish around the outside.

The methane concentration in the chamber was measured by LMD - Anritsu SA3C15A (Anritsu Corporation). The concentration of methane was measured by laser beam that reflects from reflector in the chamber every 1 second period. The carbon dioxide concentration was also on-site measured by DX6210-01 gas sensor (RMT Ltd.) from sampling port every 30 second for 5 minute. The methane and carbon dioxide flux was determined by the following equation:

$$\phi = \text{V/A (dc/dt)} \quad (6)$$

where: ϕ = flux density of the gas (mol.$m^{-2}s^{-1}$), V = flux box volume (m^3), A = flux box footprint (m^2), dc/dt = rate of change of gas concentration in the chamber with time (mol.$m^{-3}s^{-1}$).

The positions of the measured points were determined using handheld global positioning system (GPS). In order to conduct the flux chamber measurements, numerous samples were collected across the landfill surface on a regular grid pattern at 30 – 40m intervals. Geospatial distributions of the methane emissions in this study were estimated by the Kriging method. This method offers the potential of calculating whole site emission estimates from limited point measurements, which could lead to improving overall emission and oxidation estimates.

C.Measurement of CH_4 and CO_2 concentration beneath cover soil

In order to investigate the waste degradation condition within the landfill by gas sampling without methane oxidation effect in cover soil layer, gas composition measurement were performed via the stainless tube that inserted to the landfill surface by using boring bar with 1m.-depth for hole making. At the top of cover soil, rubber and soil were used to seal against air intrusion. The landfill gas was analyzed using gas analyzer (GA2000PLUS) after stainless tube had been installed for 10 minutes.

D. Study site description

The field surveys for OX evaluation were conducted in August 2008 (wet season), August 2009, and February 2010 (dry season). The study sites located at Hua-Hin, Laemchabang, Pattaya, and Saraburi landfills. All study sites locate in central part of Thailand within 150 km. from Bangkok. The site condition at all study sites can categorize as managed landfill as following IPCC (2006) [9]. All of waste that placed in these landfills was only MSW. Most of cover soil condition was sandy loam which used at Laemchabang, Hua-Hin, and Saraburi landfills. But at the Pattaya, the cover soil was sandy clay.

III. RESULTS

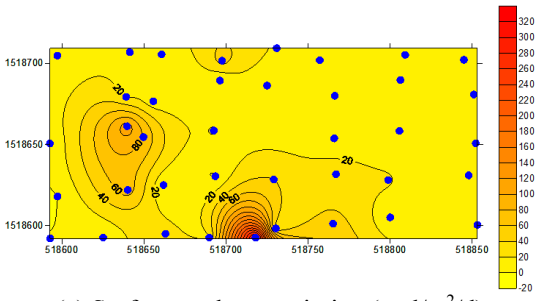
A. CH_4 oxidation

The summary of surface gas emissions, methane oxidation and oxidation factor at all study sites is showed in Table 1. The results showed that minimum and maximum of surface methane emissions were -0.81 and 321.86 mol/m^2/d, respectively. The means of surface methane emission varied between 2.33 and 22.41 mol/m^2/d. However, the average spatial surface methane emission were between 1.96 and 23.17 mol/m^2/d. For the surface carbon dioxide emission, it was found that the minimum and maximum were -4.64 and 3,009.46 mol/m^2/d. The means of surface carbon dioxide emission varied between 2.02 and 466.76 mol/m^2/d. The average spatial surface carbon dioxide emissions were

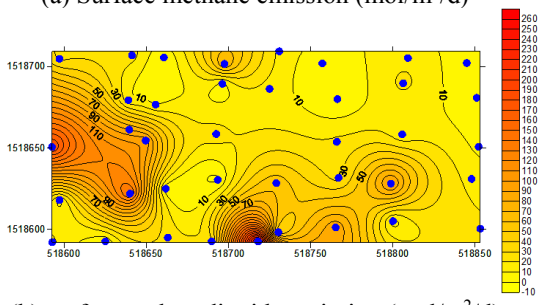
between 1.51 and 37.80 mol/m^2/d. The means of calculated bottom methane emission varied between 1.99 and 37.85 mol/m^2/d.

However, the average spatial bottom methane emissions were between 2.84 and 33.01 mol/m^2/d. From the calculation of MO from surface gas emission and gas composition beneath the soil cover at each sampling location, it was found that the minimum and maximum MO were 0.00 and 206.51 mol/m^2/d. The mean of MO varied between 0.32 and 12.11 mol/m^2/d. The average spatial MOs were between 0.27 and 12.82 mol/m^2/d. However, when calculated for OX, it was found that the OXs were between 0.10 and 0.58. This showed that the most OX values from this study in Thailand was higher than the default value (OX=0.10) from IPCC. The examples of the distribution of surface gas emission, and MO at Pattaya landfill in 2009 are shown in Figure 1.

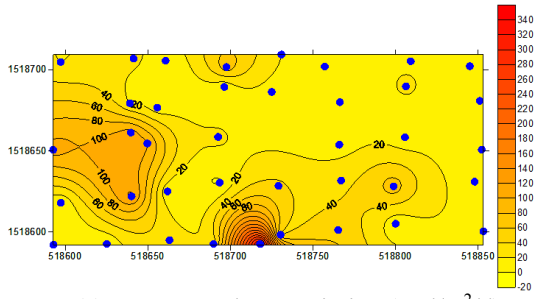
From this study, the results showed that OX varied from time to time that can make significant uncertainty for the methane emission inventory in the solid waste disposal sites. For the seasonal variation, the OXs in dry season were higher than OXs in wet season as occurred at Pattaya and Saraburi landfills. High porosity and low moisture content in soil can create some voids in daily cover that let oxygen transfer easily and create more oxidation zone compares to the soil condition in wet season that water might be blocked the oxygen path. However, at Laemchabang, the OXs in the wet season and dry season were not different because there was out of season – rain before the investigation. So the results of OX were not quite different.



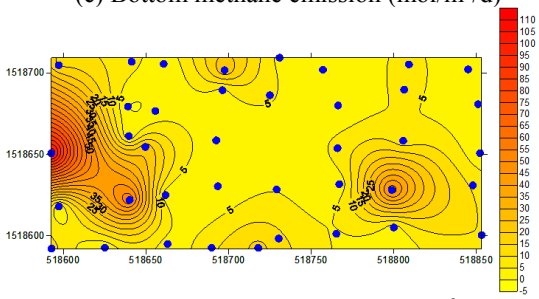
(a) Surface methane emission (mol/m^2/d)



(b) surface carbon dioxide emission (mol/m^2/d)



(c) Bottom methane emission (mol/m^2/d)



(d) Methane oxidation rate (mol/m^2/d)

Figure 1. The distribution of surface and bottom gas emission, and methane oxidation rate at Pattaya landfill in 2009

B. Relationship of bottom methane emission to methane oxidation rate and oxidation factor at each sampling point

From the relationship of calculated bottom methane emission to their methane oxidation rate and oxidation factor as shown in Figure 2 and 3, it was found that about 17% of the data was not oxidized. About 40% was oxidized with the rate of 2 mol/m^2/d. When consider to their oxidation factor, it was found that about 17% of the data had oxidation factor for 0.0. More than 76% of data, the oxidation factors varied between 0.10 and 0.9. The results also showed that 7% of investigation, all of bottom methane emission could be totally oxidized. From Figure 2, most of bottom methane emission is in the range of 0-30 mol/m^2/d. With this range, most of the methane oxidation rate was ranging between 0 and 10 mol/m^2/d.

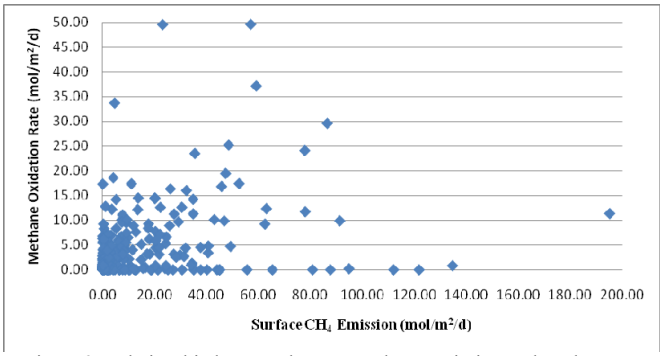


Figure 2. Relationship between bottom methane emission and methane oxidation rate in Asian countries

TABLE 1 Summary of surface gas emissions, methane oxidation and oxidation factor

			Study site											
			Hua-Hin 2008	Hua-Hin 2009	Laem chabang 2008	Laem chabang 2009	Laem chabang Area1 2010	Laem chabang Area2 2010	Pat taya 2008	Pat taya 2009	Pat taya 2010	Sara buri Area 2 2008	Sara buri Area 1 2009	Sara buri Area 1 2010
Surface CH_4 Emission	$mol/m^2/d$	Min	-0.08	0.03	0.01	-0.47	-0.03	-0.09	0.04	-0.81	-0.17	-0.02	-0.07	-0.33
		Max	12.41	40.64	134.79	91.33	55.67	24.56	35.12	321.86	102.2	121.88	40.93	17.32
		Average	2.45	6.81	17.50	12.35	14.36	6.43	9.45	25.60	17.69	22.41	2.54	2.33
		S.A.	2.89	7.38	14.70	10.90	15.36	8.05	10.63	21.47	17.15	23.17	4.46	1.96
Surface CO_2 Emission	$mol/m^2/d$	Min	-0.61	25.98	0.10	-0.58	-1.53	-0.51	1.49	-2.00	-4.64	0.16	0.03	-1.33
		Max	7.29	3,009.46	101.89	110.85	63.29	56.07	36.88	257.51	493.77	162.72	35.52	23.42
		Average	2.02	466.76	16.88	15.51	14.77	13.21	12.70	38.50	35.61	16.98	3.80	6.41
		S.A.	1.99	10.21	15.50	13.68	16.74	16.29	11.93	37.85	37.80	14.86	7.63	6.61
Bottom CH_4 Emission	$mol/m^2/d$	Min	0.00	0.50	0.15	-0.37	-0.95	-0.33	0.63	-1.64	-1.02	0.16	0.03	-0.70
		Max	10.63	59.05	135.61	116.12	70.00	34.50	33.41	338.90	308.71	106.62	45.94	21.43
		Average	2.59	10.11	19.70	16.35	17.69	10.79	13.18	36.20	28.99	21.83	3.80	4.69
		S.A.	2.84	10.29	17.30	14.43	19.67	12.40	13.82	33.01	29.56	21.19	7.25	4.58
CH_4 Oxidation (MO)	$mol/m^2/d$	Min	0.00	0.23	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
		Max	2.33	23.50	33.68	29.58	17.40	14.34	18.59	106.65	206.51	49.59	6.76	6.78
		Average	0.32	3.30	3.09	4.01	3.70	4.38	4.01	10.67	12.11	4.36	1.26	2.41
		S.A.	0.27	2.92	3.22	3.55	4.60	4.35	3.43	11.62	12.82	3.77	2.85	2.68
CH_4 Oxidation Factor (OX)	-	S.A.	0.10	0.28	0.19	0.25	0.23	0.35	0.25	0.35	0.43	0.18	0.39	0.58

*S.A.= Spatial Average

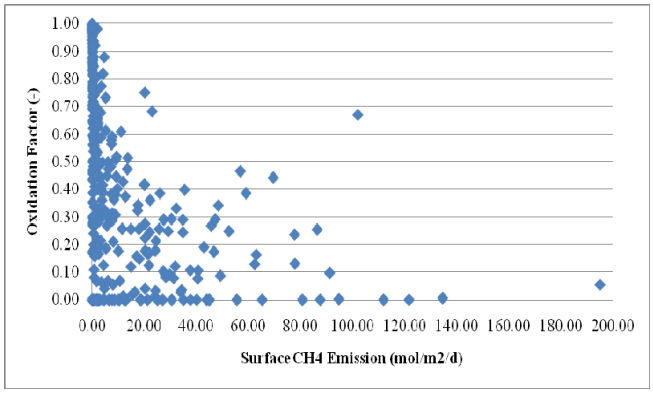


Figure 3. Relationship between bottom methane emission and oxidation factor in Asian countries

IV. CONCLUSIONS

Field investigations of surface methane and carbon dioxide emission as well as gas composition beneath the cover soil in Thailand had been conducted during 2008 – 2010. By using the closed flux chamber technique, high surface methane and carbon dioxide fluxes were observed. The combination of flux measurements and gas profiles were used to calculate an estimate of the methane oxidation rate in the cover soil as well as their oxidation factors. Methane oxidation was a very important process of landfill gas emissions because the methane emissions from active or closed landfill can be reduced naturally by the oxidation process. Knowing of methane oxidation rate and oxidation factor can reduce the uncertainty of the methane emission inventory from the solid waste disposal sites.

The calculation showed that about 17% of the data at the sampling point was not oxidized. About 40% could be oxidized with the rate not over 2 $mol/m^2/d$. When consider to their total oxidation factors, it was found that the oxidation factors were between 0.10 and 0.58 that most observed OX were greater than the default value in the IPCC Waste Model (OX=0.10).

The results also showed that the oxidation process properly active under the low methane emission range. Reducing methane generation and/or emission before it pass through the landfill cover soil by using landfill gas recovery process or changing the design to semi-aerobic landfill which produces lower methane can enhance the ability of methane oxidation process.

The future studies should consider the CH_4 oxidation in more type of landfilling as in open dumpsites that researchers rarely obtained this information as well as study in detail of soil cover type. Based on this study, CH_4 emission from solid waste disposal sites could be significantly reduced by the utilization of appropriate cover soil type and condition with high CH_4 oxidation capacity.

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