



Estimation of CH₄ oxidation efficiency in an interim landfill cover soil using CO₂/CH₄ ratios

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ABSTRACT

The first objective of this study was to discuss the applicability of the CO₂/CH₄ ratio method in order to assess CH₄ oxidation efficiency. To achieve this objective, a comparison between CO₂/CH₄ ratios and the mass balance method was conducted. The second objective of this study was to estimate the CH₄ oxidation efficiency in an interim landfill soil cover and assess how a CH₄ influx influences the CH₄ oxidation efficiency. The results showed that despite the CO₂ problems brought by respiration, the CH₄ oxidation efficiencies obtained by the CO₂/CH₄ ratio method led to similar results compared to the mass balance method. In this respect, the CO₂/CH₄ ratio method can be an indicator of the CH₄ oxidation efficiencies for landfill cover soils. The CH₄ oxidation efficiencies derived in this study through the CO₂/CH₄ ratio method ranged between 46% and 64%, and between 41% and 62% through the mass balance method. The results imply that the Intergovernmental Panel on Climate Change's (IPCC) default value of 10% for the CH₄ oxidation efficiency is an underestimation for landfill cover soils. CH₄ oxidation efficiency tends to be negatively correlated with CH₄ influx. Therefore, CH₄ influx reaching a landfill cover should be limited in order to increase the CH₄ oxidation efficiency.

Keywords: CH₄ oxidation, CO₂/CH₄ ratio, Landfill, Mass balance

1. Introduction

A significant source of CH₄ production landfills have historically been considered the largest atmospheric CH₄ emitter that represents 50-57% of global CH₄ emissions that come from the waste sector [1].

CH₄ oxidation is a natural process that takes place in soil systems without human interference, due to methanotrophic bacteria [2]. Replicating this natural process, landfill cover soil is used to facilitate the microbial oxidation of CH₄ in order to help mitigate CH₄ fluxes into the atmosphere. CH₄ oxidation is considered a cost-effective technology to reduce CH₄ emissions from landfills where a gas collection system has not been installed or is not economically feasible [3].

While much research has been conducted on CH₄ oxidation efficiency [4-7], there continues to be insufficient information due to the lack of a standard method for measuring the process [2].

Several field measurement methods are currently available to measure CH₄ oxidation efficiency including using carbon isotopes, mass balance, and CO₂/CH₄ methods. The carbon isotope measurement is currently the most precise method available for determining

CH₄ oxidation in landfill covers [8]. However, the carbon isotope method will underestimate the CH₄ oxidation efficiency in landfill cover soils as well as the high cost of analysis because the isotope method underestimates the importance of molecular diffusion during gas transport [9].

An alternative method that has been used recently to estimate the CH₄ oxidation is the mass balance, which could be used to estimate the CH₄ oxidation using the difference between the CH₄ flux at the top and at the bottom of landfill cover soil [10]. The bottom CH₄ fluxes could be calculated by using a ratio of CH₄ to CO₂ at the bottom of a landfill cover. The mass balance method was validated by previous literatures [10-12].

Gebert et al. [11] have recently employed an alternative CO₂/CH₄ ratio method to measure the CH₄ oxidation in landfill soils based on laboratory experiments. In addition, Pratt et al. [13] demonstrated a strong correlation between CH₄ oxidation efficiency determined by mass balance and the CO₂/CH₄ ratio for a pumice landfill cover soil. Therefore, the ease and low cost of the CO₂/CH₄ ratio method is an attractive approach for landfill operators [13].

While the CO₂/CH₄ ratio method is a simple and inexpensive tool, further study is needed to validate the effectiveness of the



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CO₂/CH₄ ratio method in actual landfill sites; so far these authors [11, 13] have only assessed the effectiveness of the CO₂/CH₄ ratio method based on laboratory experiments.

Thus, the first objective of this study was to discuss the applicability of the CO₂/CH₄ ratio method in order to estimate CH₄ oxidation efficiency. To achieve this objective, a comparison between the CO₂/CH₄ ratio and the mass balance methods was conducted. The second objective of this study was to estimate CH₄ oxidation efficiency in an interim landfill soil and assess how CH₄ influx influences the CH₄ oxidation efficiency.

2. Materials and Methods

2.1. Site Description

The study was conducted at a sanitary landfill in Anseong City, Korea (Fig. 1). The annual mean temperature at the city ranges from 12 to 13°C. The annual precipitation is 1,100 mm yr⁻¹. Since 2003, municipal waste, commercial waste, and bottom ash have been disposed of at this landfill. It covers an overall volume of about 568,200 m³ (38,200 m²), although only about 112,800 m³ is currently landfilled for municipal solid waste (MSW), which consist of approximately 10% paper, 10% plastic, 10% wood, 5% rubber, 2% others, and non-combustible 63% by wet weight. Currently, temporary landfill cover soil, consisting of 1 m sandy-clay loam material, limits surface CH₄ emissions. No final cover caps have been installed in any part of the landfill. Seventeen vertical wells in the landfill were installed in a grid pattern and the landfill gas (LFG) collected through the vertical wells was

emitted into the atmosphere.

In this study, three field campaigns to measure CH₄ oxidation efficiencies were performed: on October 2, 2014, October 17, 2014, and November 11, 2014. The average precipitation in October and November was 119.5 mm and 24.9 mm, respectively (Korea Meteorological Administration, 2014). Measurements have been conducted in the day time (10 a.m.-6 p.m.).

2.2. CH₄ Oxidation Efficiency

2.2.1. CO₂/CH₄ ratio method

CH₄ oxidation efficiency was determined using the CO₂/CH₄ ratio. The CO₂/CH₄ ratio is based on the comparison of the CO₂/CH₄ ratio in the surface soil with that in the LFG (Fig. 2). It was assumed that CO₂ is produced by oxidation of CH₄ only, and that 1 mol of CH₄ is converted to 1 mol of CO₂ as a result of the oxidation process [11]. Therefore, the CH₄ oxidation efficiencies could be calculated using the following equations [11, 13]:

$$\frac{CO_{2LFG} + X}{CH_{4LFG} - X} = \frac{CO_{2d}}{CH_{4d}} \quad (1)$$

$$CH_4(\text{oxidised; \%}) = \frac{X}{CH_{4LFG}} \times 100 \quad (2)$$

where X is the portion of oxidized CH₄ (vol.%), CH_{4LFG} is the CH₄ concentration of the LFG (vol.%), CO_{2LFG} is the CO₂ concentration of the LFG (vol.%), CH_{4d} is the CH₄ concentration on surface soil (vol.%) and CO_{2d} is the CO₂ concentration on surface soil (vol.%).

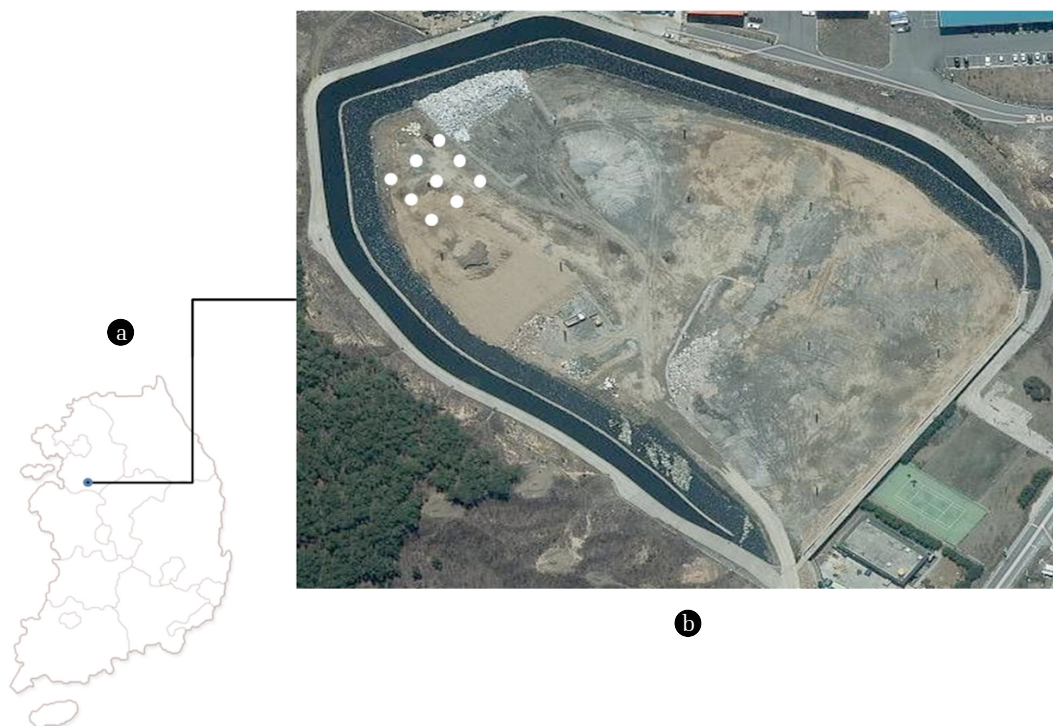


Fig. 1. Description of the study area. (a) Location of the Anseong landfill, (b) Sampling points.

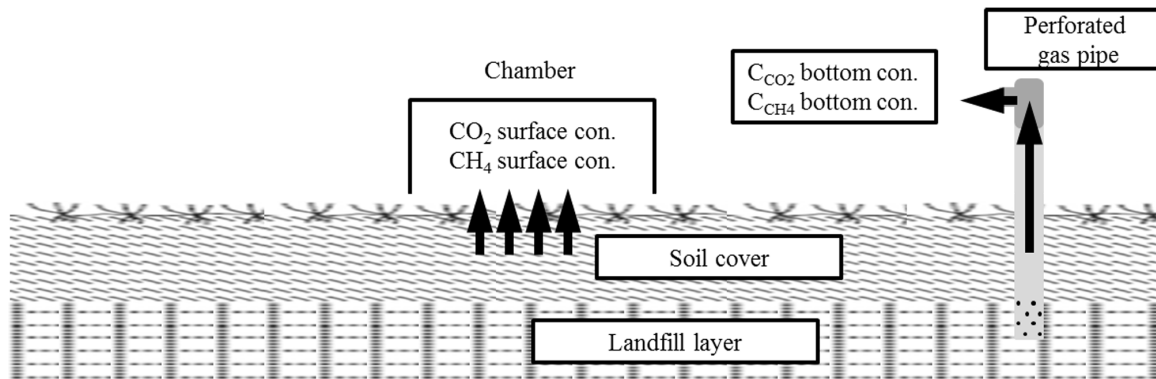


Fig. 2. Schematic representation of the CO₂/CH₄ ratio method.

In order to analyze CH₄ and CO₂ concentrations, gas samples were collected at the site using chambers (0.012 m³). The gas samples were collected after 1 hour so that the effects of air dilution were minimized [13]. CH₄ concentration was measured using a portable laser methane detector (LMD, Tokyo Gas Engineering). The LMD is a hand-held gas detector used for remote measurements of column density of methane-containing gases. The LMD can detect CH₄ concentrations between 10 and 10,000 ppmv and up to a distance of 150 m. Precision of analysis for LMD was checked against a gas chromatography (Younglin 6000M, Korea) using a TCD. The CH₄ concentration is calculated according to Eq. (3):

$$CH_4(\text{ppmv}) = \frac{M}{X} \quad (3)$$

where M is the path-integrated CH₄ concentration (ppmv·m), and X is the distance (m).

CO₂ concentrations were measured by GC/TCD (Younglin 6000M, Korea). GC analysis conditions included setting the oven temperature at 35°C and the TCD filaments at 120°C. The carrier gas was helium running at a flow rate of 50 mL/min.

2.2.2. Mass balance method

The CO₂/CH₄ ratio method was compared to the mass balance method. The calculation is based on the assumption that all the CO₂ is either already present in the system or results from CH₄ oxidation. CH₄ oxidation efficiency is defined by [10, 12]:

$$CH_4(\text{oxidised; \%}) = \frac{F_{CH_4 \text{ influx}} - F_{CH_4 \text{ surface flux}}}{F_{CH_4 \text{ influx}}} \times 100 \quad (4)$$

where $F_{CH_4 \text{ influx}}$ is the CH₄ flux under the cover, and $F_{CH_4 \text{ surface flux}}$ is the CH₄ flux measured at the ground level in the landfill. $F_{CH_4 \text{ influx}}$ is defined by:

$$F_{CH_4 \text{ under the cover}} = (F_{CH_4 \text{ ground level}} + F_{CO_2 \text{ ground level}}) \times \frac{CH_4 \text{ under the cover}}{CO_2 \text{ under the cover} + CH_4 \text{ under the cover}} \quad (5)$$

where $CH_4 \text{ under the cover}$ is the CH₄ concentration under the landfill cover soil, and $CO_2 \text{ under the cover}$ is the CO₂ concentration under

the landfill cover soil.

CH₄ and CO₂ surface fluxes were measured at the site using static chambers (with a diameter of 0.2 m and height of 0.4 m). Chambers were placed in a grid pattern; they were placed 10 m apart within a 20 m × 20 m section (Fig. 1). In order to collect gas samples for CO₂ concentration analysis, a sampling port was placed on each chamber. Gas samples (10 mL) were drawn from each chamber three times over the course of one hour. CH₄ concentrations were measured with the LMD. Linear increases in CH₄ and CO₂ concentrations in the chambers and CH₄ and CO₂ surface fluxes were calculated using Eq. (6) as follows:

$$F = V/A(dC/dt) \quad (6)$$

where V is the volume of the chamber, A represents the area (m²) covered by the chamber, and dC/dt is the change in headspace CH₄ and CO₂ concentration with time (mg m⁻³ day⁻¹).

3. Results and Discussion

3.1. Landfill Gas

Table 1 summarizes the change in CH₄ and CO₂ concentrations under the landfill soil cover. CH₄ concentrations fluctuated between 7.2% and 8.1% at the site of the landfill containing mainly non-combustible waste, including ash. Environmental factors (soil types, moisture, pH, temperature, and air) can impact the rate of CH₄ oxidation. Among these, methanotrophic activity is one of the crucial environmental factors in terms of dilution of CH₄ concentrations. If the landfill cover soil is exposed to high CH₄ concentration, it can develop a very high CH₄ oxidation capacity. According to Wang et al. [14], CH₄ oxidation rates in landfill soil were enhanced within a CH₄ concentration range of 0.01-10% (v/v), while they remained stable within a range of 10-30% (v/v). Chi et al. [15] showed that the rate of CH₄ oxidation efficiency sharply increases with the initial CH₄ concentration because CH₄ is the sole carbon substrate in all metabolic pathways.

Therefore, in the case of a landfill containing a high proportion of non-combustible waste, adjusting the gas wells may be a very effective way to increase the CH₄ concentration in the LFG, leading to enhanced CH₄ oxidation efficiency.

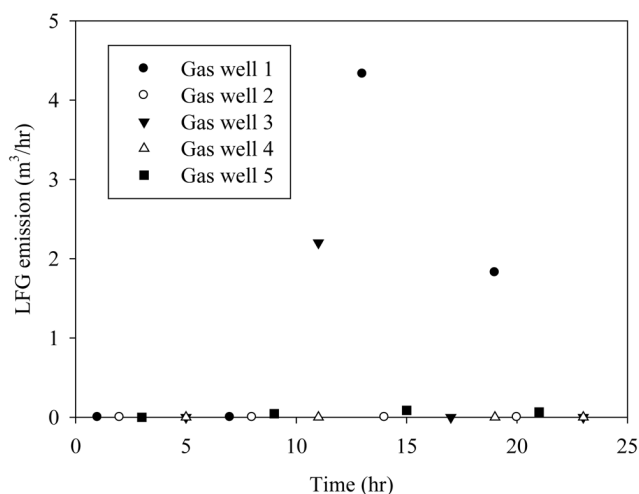


Fig. 3. Landfill gas emission through each gas well.

Seventeen vertical wells in the landfill were installed in a grid pattern. Fig. 3 presents LFG emission of five gas wells measured by an air velocity meter (TSI 8330-M-GB, USA) on October 2. The LFG emission at the gas wells was observed to vary from 0.0 to 4.3 m³ hr⁻¹. The LFG emission of gas wells 2 and 4 was measured at zero. CH₄ concentrations in the gas wells ranged from 0.0% to 2.2%. The occurrence of low LFG emission through the gas wells suggests that some gas wells in the landfill should be closed in order to increase CH₄ concentrations, resulting in an enhanced CH₄ oxidation efficiency.

Table 1. CH₄ and CO₂ Concentrations in Landfill Gas

Item	2nd October	17th October	11th November
CH ₄ (vol.%)	7.5	7.2	8.1
CO ₂ (vol.%)	5.4	5.3	5.9

3.2. Comparison Between CO₂/CH₄ Ratio and Mass Balance Methods

Fig. 4 shows a comparison between CH₄ oxidation efficiencies obtained from a mass balance and the CO₂/CH₄ ratio calculations. CH₄ oxidation efficiencies obtained through the CO₂/CH₄ ratio method are similar to those obtained through the mass balance method. However, the CO₂/CH₄ ratio method slightly overestimates the oxidation efficiencies compared to the mass balance method. It seems that the observed overestimation is affected by the respiration of landfill soil. The landfill cover soil respires, releasing CO₂, resulting in a higher CO₂/CH₄ ratio and an overestimation of oxidation [11]. Huber-Humer *et al.* [8] noted that the interference resulting from landfill cover soil respiration should be factored in the methods adapted from the CO₂/CH₄ ratios.

However, this factor is unlikely to be a concern because CO₂ generated by the oxidation of compounds is overshadowed by CH₄ oxidation in soils [11, 13]. The proportion of CO₂ produced by respiration is <10% at CH₄ influxes above 12 g CH₄ m⁻² d⁻¹ [13]. Moreover, in this study, the introduction of the correction for respiration in the vicinity of <10% led to a very close approximation

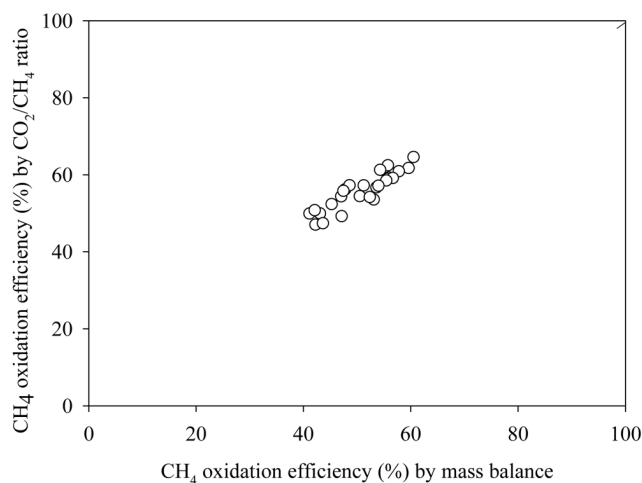


Fig. 4. Comparison of the CH₄ oxidation efficiencies determined by the mass balance and CO₂/CH₄ ratio methods.

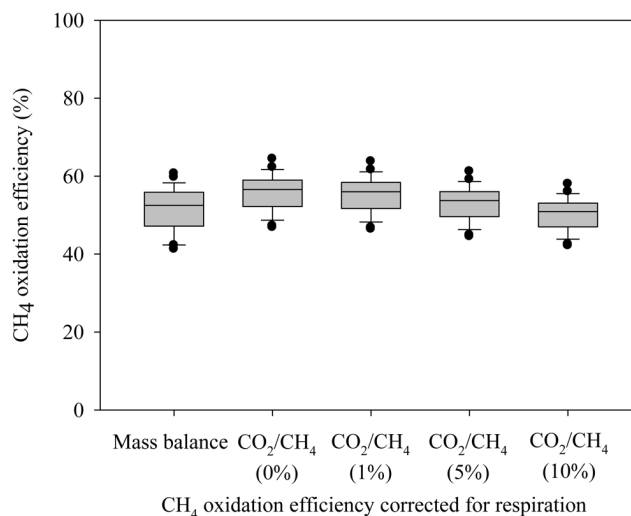


Fig. 5. CH₄ oxidation efficiency corrected for each respiration.

of the estimated oxidation efficiency (Fig. 5). The results indicate that the carbon balance is dominated by CH₄ oxidation. In this respect, the CO₂/CH₄ ratio method can be a reliable indicator of CH₄ oxidation efficiency for landfill cover soils.

3.3. CH₄ Oxidation Efficiency

As summarized in Table 2, CH₄ oxidation efficiencies ranged between 46% and 64% for the CO₂/CH₄ ratios, and for the mass balance, it ranged between 41% and 62%. This research supports the evidence that the Intergovernmental Panel on Climate Change's (IPCC) default value of 10% is an underestimation for landfill cover soils [16]. According to Börjesson *et al.* [6], the CH₄ oxidation efficiencies for active landfill sites were estimated to range from 1.1% to 25.4% and those of closed landfill sites were observed to vary from 30.8% to 46.9%. Liptay *et al.* [17] reported 24-35% CH₄ oxidation efficiency in the warm season. Chanton *et al.* [18] showed that CH₄ oxidation efficiency was 33%, with a CH₄ flux

Table 2. CH₄ Oxidation Efficiency Determined by the Mass Balance and CO₂/CH₄ Ratio Methods

Item	CO ₂ /CH ₄ ratio	Mass balance
2nd October	46-64%	41-61%
17th October	46-64%	47-62%
11th November	50-61%	42-57%

of 1 g m⁻² d⁻¹. In addition, Chanton et al. [19] measured the CH₄ oxidation efficiency of 50%, where the CH₄ flux was estimated at 20 g m⁻² d⁻¹. Therefore, further research is needed to estimate the CH₄ oxidation efficiencies of landfill cover soils in order to develop country-specific parameters for Korea.

The higher CH₄ oxidation efficiencies obtained in this study can be explained by the soil texture and layer thickness in addition to the low CH₄ surface flux. The landfill surfaces in this study had been covered by an interim landfill cover soil consisting of a 1 m sandy-clay loam composite (pH 6.4), which provided sufficient oxygen. CH₄ oxidation is positively linked to a sandy texture, indicating the dependence of CH₄ oxidation on gas permeability [20]. According to Kightley et al. [21], the CH₄ oxidation efficiencies for coarse sand and fine sand were 61% and 41%, respectively. Chanton et al. [18] summarized that the mean value for CH₄ oxidation efficiency for differing soil covers ranged from 22% for clay to 55% for sand.

Moreover, using a soil layer thickness of more than 50 cm for CH₄ oxidation can increase oxidation, particularly at low temperatures [22]. A thicker layer may also allow a higher CH₄ oxidation rate because temperature and moisture may be more suitable and remain stable than in a thin cover. According to Albanna et al. [23], increasing the soil layer thickness from 15 to 20 cm increased CH₄ oxidation efficiencies from 29% to 35%.

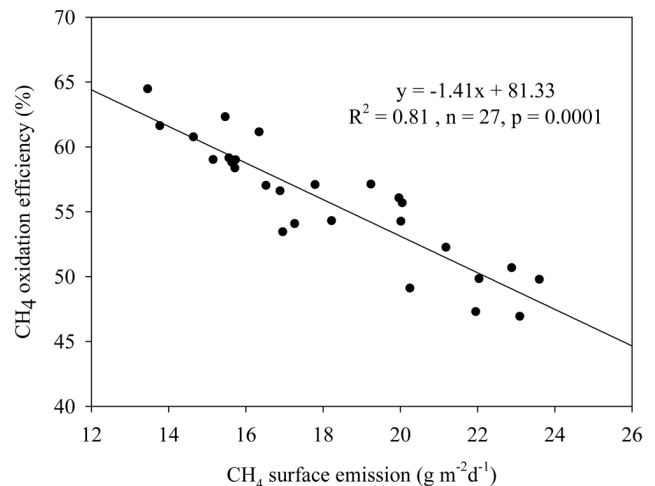
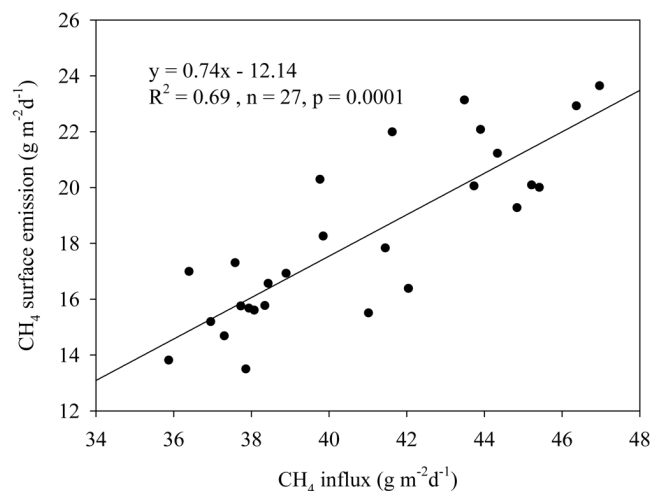
3.4. Influence of CH₄ Influx on CH₄ Oxidation Efficiency

CH₄ oxidation efficiency is a function of cover type, climatic conditions, and CH₄ influx to the bottom of the cover. The correlation between CH₄ surface emissions and CH₄ oxidation efficiency is shown in Fig. 6. The data were modeled using a linear function, and the resulting correlation coefficients (R²) of the linear regression were 0.81. The results show a negative linear relationship between the CH₄ surface emissions and the CH₄ oxidation efficiencies. Chanton et al. [19] have demonstrated that CH₄ oxidation efficiency would be greatest at sites of low CH₄ emissions and would decrease as CH₄ emissions increased. According to Visscher and Cleemput [24], a high CH₄ oxidation efficiency is obtained at CH₄ fluxes up to 50 g m⁻² d⁻¹.

The correlation between CH₄ surface emissions and CH₄ influxes is shown in Fig. 7. CH₄ surface emissions were observed to increase with increases in CH₄ influx. This result shows a positive linear relationship between CH₄ influxes and CH₄ surface emissions. Assuming that gas pressure below the landfill cover remains consistent, atmospheric pressure can lead to a variation in CH₄ surface emission. Czepiel et al. [25] reported that CH₄ surface emissions decrease with an increase in atmospheric pressure. When atmospheric pressure increases, CH₄ influx below the landfill cover decreases, resulting in oxygen diffusion from the upper boundary, thus increasing the amount of CH₄ that can be oxidized. According

Table 3. The Results of CH₄ Surface Fluxes in this Study

Item	2nd October	17th October	11th November
No. of measurements	9	9	9
Min (g/m ² /d)	13.5	13.8	15.2
Mean (g/m ² /d)	18.8	17.7	17.9
Max (g/m ² /d)	23.6	22.1	22.9
St. Dev (g/m ² /d)	3.2	3.1	2.3

**Fig. 6.** Correlation between CH₄ surface emission and CH₄ oxidation efficiency.**Fig. 7.** Correlation between CH₄ influx and CH₄ surface emission.

to Berger et al. [26], oxygen is one of the most important limiting factors in the CH₄ oxidation process and CH₄ oxidation efficiency is a decreasing exponential function of the CH₄ influx rate into the bottom of the cover. Thus, increasing CH₄ influx to a landfill soil cover can reduce the CH₄ oxidation efficiency. Pratt et al. [13] estimated that CH₄ oxidation efficiency was 84% with a CH₄ influx of 6 g m⁻² d⁻¹, whereas CH₄ oxidation efficiency was 55% with a CH₄ influx of 36 g m⁻² d⁻¹.

Therefore, CH₄ influx under a landfill cover soil should be limited, in order to enhance CH₄ oxidation efficiency. In the case of a landfill containing a high proportion of organic waste, including food waste, a gas collection system can be a very effective way to limit CH₄ influx reaching the cover [19]. Because CH₄ influxes in a landfill can be limited by a gas collection system, CH₄ oxidation efficiency will be greater in those landfills with gas collection than in those without [19].

4. Conclusions

Precise and reliable measurements of CH₄ oxidation efficiency in landfill soils are needed to develop country-specific parameters as used in the first order decay model of 2006 IPCC guidelines [16]. In addition, the reduction of LFG emissions could be applied to carbon emission cap and trading schemes, representing potential financial benefits for landfill operators [8].

In this study, CH₄ oxidation efficiencies were determined by the mass balance and CO₂/CH₄ ratio methods. Despite CO₂ respiration challenges, CH₄ oxidation efficiencies obtained by the CO₂/CH₄ ratio method led to similar results to the mass balance method. As such, the CO₂/CH₄ ratio method could be successfully used to establish CH₄ oxidation efficiency for landfill cover soil. Nevertheless, environmental factors (soil types, moisture, and CH₄ concentration) for CH₄ oxidation efficiencies still need to be studied for precise estimation.

The CH₄ oxidation efficiencies obtained through the CO₂/CH₄ ratio in this study ranged between 46% and 64%, and the ones obtained through the mass balance method ranged between 41% and 62%. The results support the fact that the IPCC default value of 10% is an underestimation for landfill cover soils.

As expected, CH₄ oxidation efficiency tended to be negatively correlated to CH₄ influx. When CH₄ influxes are high in a landfill, therefore, using a gas collection system can reduce the CH₄ influx that reaches the landfill cover soil, which can, thereby, improve CH₄ oxidation efficiency. In contrast, when CH₄ influxes and concentrations are too low in a landfill, adjusting the gas wells may be a very effective way to increase the CH₄ concentration in LFG, leading to enhanced CH₄ oxidation efficiency.

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