Methane and carbon dioxide emissions from closed landfill in Taiwan

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Abstract

The atmospheric concentrations and emission rates of CH₄ and CO₂ were studied at three sites of the Fu-Der-Kan closed landfill and after as the multi-use recreational park in northern Taiwan. Atmospheric CH₄ and CO₂ concentrations of closed landfill were 1.7–4.6 and 324–409 ppm, respectively. CH₄ and CO₂ emission rates ranged from 8.8 to 163 mg m⁻² h⁻¹ and from 495 to 1531 mg m⁻² h⁻¹, respectively. Diurnal variation was noted with higher values at night than those in daytime. After creation of the park, atmospheric CH₄ and CO₂ concentrations were 1.8–3.1 and 332–441 ppm, respectively. CH₄ and CO₂ emission rates ranged from 1.1 to 2.3 mg m⁻² h⁻¹ and from 135 to 301 mg m⁻² h⁻¹, respectively. There were no notable diurnal variations in either atmospheric concentrations or emission rates.

Keywords: Landfill; Greenhouse gas; FTIR; Flux; Diurnal variation

1. Introduction

Landfills are accounting for about 10–19% of annual global CH₄ emissions (Kumar et al., 2004; USEPA, 2006). The global warming potential of CH₄ is 23 times higher than that of CO₂ (USEPA, 2002). The atmospheric CH₄ concentration has been increasing at a rate of 0.2–1% y⁻¹ (Simpson et al., 2006). On a global scale, approximately 653 Tg y⁻¹ of waste is landfilled, and annual global CH₄ emissions from landfills range from 16 to 223 Tg (Bogner and Matthews, 2003; Simpson et al., 2006).

Landfilling and incineration are the major methods of waste disposal in Taiwan. 89% of waste was disposed by landfills in 1989. This percentage dropped to 11.2% in 2006 as the result of increased incineration and the resource recycling policy (EPA/Taiwan, 2007). Landfills function as bioreactors due to the controlled and managed burial of biodegradable organic materials. These organic materials decompose via a complex series of microbial reactions under anaerobic conditions. Eventually, they are converted into CO₂, CH₄, N₂O and H₂O. Landfill CH₄ and CO₂ generations are controlled by landfill management, cover soil, composition, temperature, pressure, moisture content, water table level, wind induced ebullition and pH of the refuse (Bogner and Matthews, 2003; Hegde et al., 2003). Methanogenesis in landfills occurs when the pH of organic waste is between 6.8 and 7.4 and it is stimulated by increasing moisture content (Jang and Yang, 2001).

The Fu-Der-Kan landfill site was closed in 1995, and a multi-use recreational park was constructed from 1999 to 2003. The soil surface is covered with grass and small trees. Although the CH₄ and CO₂ emission rates of active landfills are well documented (Bogner et al., 1999; Czepiel et al., 2003; Meraz et al., 2004), emissions from closed sites and sites which have been converted to park have not been investigated in detail.

Atmospheric CH₄ and CO₂ concentrations and emission rates from an active landfill in northern Taiwan were described in a previous study (Hegde et al., 2003). Two main methods are used for such measurements. Gas-type open-path Fourier-transform infrared (FTIR) spectroscopy has the advantages of good precision, rapid and
simultaneous measurement of different volatile compounds, and the ability to cover long-distance. Chamber techniques can be used to measure CH$_4$ and CO$_2$ emission rates from small areas, typically less than 1 m$^2$ (Yang and Chang, 1997). The advantages include simplicity, an appropriate scale for concurrent measurement of controlling variables, and the ability to determine the heterogeneity of surface emissions. Chamber techniques can also be easily compared to results from other studies because these methods have been widely employed in non-landfill settings for a variety of greenhouse gases (Rolston, 1986; Yang and Chang, 1998, 2001; Chang and Yang, 2003). In this study, FTIR was used successfully, along with gas chromatography (GC), to monitor atmospheric greenhouse gases. The atmospheric concentrations and emission rates of CH$_4$ and CO$_2$ were measured at Fu-Der-Kan closed landfill and after the construction of a multi-use recreational park in northern Taiwan.

2. Materials and methods

2.1. Site description

The Fu-Der-Kan landfill (25°01’53.9”N, 120°3’33.8”E) is located near Taipei City in northern Taiwan. This landfill was opened in 1985 for municipal solid waste (MSW) dumping, closed in 1995 and converted to a multi-use recreational park during 1999 to 2003. It covers 98 ha and 37 ha were used for landfilling of MSW. It received 3200 t of MSW every day during the operation period, with an average burial depth of 22.9 m. The landfill was covered by 1–2 m of waste landfill soil as surface material. Upon closing, the landfill was covered with another 1–1.5 m of loam-clay loam soil and reconstructed as a multi-use recreational park in 2001 (Fig. 1). The test areas cover about 32 ha of landfilling area including areas A, B, C, E, F, G and part of J around the testing areas. Site C is younger than sites A and B. Site C had received new MSW in 1999. During 1999-2000 testing period, site A (about 8 ha) included country activity area, lake, boating area and part of natural resource landscape; site B (about 12 ha) included Sun plaza, creation garden and part of natural resource landscape; site C (about 12 ha) included culture of indigenous people’s area, grass skiing field and part of natural resource landscape. The testing area covers about 90% of landfilling except management center and wastewater treatment factory. The vegetations in the sites A, B, and C were grass and small trees during the testing periods 1999–2000. The major vegetations were Centipede grass (Eremochlora aphio-roides) and Carpet grass (Axonopus affinis) in sites A, B, and C during testing period 2006. There is a setup for CH$_4$ gas extraction system and each year recovered 3442 t of CH$_4$, which should equivalent to 79.2 Gg of CO$_2$, while the recovered CH$_4$ was burned and produced 10.2 Gg of CO$_2$ only. Soil samples were collected at 0–20 cm depth in different burial locations. Randomly three soil cores in each site were collected, sieved to 2 mm and stored at 4 °C before analysis. Each soil core was analyzed separately for moisture content, pH and organic carbon.

2.2. Gas-type open-path FTIR spectroscopy measurement

FTIR spectroscopy was used to measure the concentrations of atmospheric greenhouse gases at a height of 197 cm above ground with a scan number of 100 for 7 min. Absorbance was read at wavenumber of 2999 cm$^{-1}$ for CH$_4$ and peak area from 2239 to 2393 cm$^{-1}$ for CO$_2$. The distance between the light source and the reflecting mirror was 35 m (for four chambers) or 100 m (for nine chambers). The details of the experimental set-up adopted for the FTIR spectroscopy method have been described in a previous paper (Chang et al., 2000). Because the interferometer characteristics are dominated by a high spectral resolution up to 0.06 cm$^{-1}$ and a configuration suitable for field measurements was needed, the MB-104 (BOMEM, Hartmann and Braun, Canada) was used for radiation emission as well as absorption measurements. The spectrum was observed on a PC screen and the mirror was adjusted to optimize the signal. The absorption peak areas and atmospheric concentrations of CH$_4$ and CO$_2$ have high correlation coefficients (0.998–0.999 and 0.996–0.997, respectively) with distance between 0 and 100 m. The accuracy of FTIR spectroscopy in CH$_4$ and CO$_2$ measurement is within 2% and 3%,
respectively. The atmospheric concentrations of greenhouse gases were determined from long-path measurements by differential absorption and a least squares fit of measurement, and were simulated by modeling air transmittances for various absorbed concentrations.

2.4. Analytical methods

Air and soil temperatures were determined on site either directly or under at 10 cm depth with a Hg-thermometer. Wind speed was measured with a weathercock (Weather link 4.0, USA). Light intensity was detected with a Toshiba SPI-5 photometer. Soil pH was determined using a pH meter directly in the soil or in a 1:1 (w/v) soil to water suspension. Moisture content was measured by drying the samples at 105 °C for 24 h until a constant weight was achieved. Total organic carbon (TOC) was estimated using a TOC-5000A analyzer (Code HI 8424C, Shimadzu, Japan). Experiments were carried out in quadruplicate; while nine experiments were used in the measurement of CH4 and CO2 emission rates on June 30, 2000. Data were subjected to coefficient of variance analysis and Duncan’s multiple range tests (p = 0.05) using the Statistical Analysis System (SAS Institute, 2002).

3. Results

3.1. Properties of cover soils

The properties of cover soil at different burial locations are shown in Table 1. The cover soils were acidic (pH 5.6–6.0) in 1999 and 2000, and they were neutral (pH 7.4–7.7) after the construction of a multi-use recreational park in 2006. There were significant pH differences between 1999–2000 and 2006 (p < 0.05). The soil moisture, soil temperature and TOC in 2006 were lower than those in 1999–2000.

3.2. CH4 and CO2 concentrations and emission rates at site A

Air temperature, atmospheric concentrations of CH4 and CO2 at site A are presented in Fig. 2. Atmospheric concentrations of CH4 and CO2 on October 30, 1999 ranged from 1.7 to 2.7 ppm and from 324 to 390 ppm, respectively. Average values were 2.2 ± 0.3 and 366 ± 24 ppm, respectively. On November 24, 1999, atmospheric concentrations of CH4 and CO2 ranged from 1.9 to 4.6 ppm and from 373 to 409 ppm, respectively, with average values of 3.1 ± 0.9 and 386 ± 11 ppm, respectively. Atmospheric concentrations of CH4 and CO2 on October 30 were lower than those on November 24 and it might be due to the high wind velocity on October 30 (4.2 ± 0.7 m s⁻¹) (Table 1). Further, the total organic C content of cover soil on November 24, 1999 in site A was about 25% lower than that on October 30, 1999 might be due to the construction as a multi-use recreation park during this period, and some new cover materials with low organic C content were layered on the surface. Therefore, both the organic C content of cover soil and the CH4 emission on November 24, 1999 were lower than those on October 30, 1999 because of high soil organic C and moisture content for active microbial reaction.
Site A was subsequently converted into a country activity area during the period from 1999 to 2003. Atmospheric concentrations of CH₄ on January 4, 2006 were between 1.8 and 3.1 ppm, with an average of 2.3 ± 0.5 ppm. CO₂ on that date ranged from 373 to 441 ppm, with an average of 396 ± 23 ppm. CH₄ and CO₂ emission rates were 0.4 ± 0.3 to 2.3 ± 0.6 mg m⁻² h⁻¹ and 72 ± 17 to 301 ± 5 mg m⁻² h⁻¹, respectively. Average CH₄ and CO₂ emission rate was 0.7 ± 0.4 and 95 ± 6 mg m⁻² h⁻¹, respectively (Fig. 3a–c). CH₄ and CO₂ emission rates in 2006 were lower than those in 1999 because of the more stable burial waste, low soil organic C and low moisture content.

### Table 1

Properties of cover soils in Fu-Der-Kan landfill and after as the multi-use recreational park

<table>
<thead>
<tr>
<th>Sampling date</th>
<th>pH</th>
<th>Moisture content (%)</th>
<th>Soil temp. (°C)</th>
<th>Atmos. pressure (Pa)</th>
<th>Humidity (%)</th>
<th>Wind velocity (m s⁻¹)</th>
<th>Organic carbon rate (mg m⁻² h⁻¹)</th>
<th>CH₄ emission rate (mg m⁻² h⁻¹)</th>
<th>CO₂ emission rate (mg m⁻² h⁻¹)</th>
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<tr>
<td><strong>Site A</strong></td>
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<tr>
<td>Oct. 30, 1999</td>
<td>5.5–6.1ᵇ</td>
<td>31.1 ± 1.7ᵃ</td>
<td>31.9 ± 1.0ᵃ</td>
<td>1015 ± 1ᵃ</td>
<td>74.4 ± 6.1ᵃ</td>
<td>4.2 ± 0.7ᵇ</td>
<td>20.8 ± 1.4ᵃ</td>
<td>21.7 ± 2.4ᵇ</td>
<td>1012 ± 14ᵃ</td>
</tr>
<tr>
<td>Nov. 24, 1999</td>
<td>5.5–6.2ᵇ</td>
<td>29.6 ± 1.8ᵃ</td>
<td>30.9 ± 1.0ᵃ</td>
<td>1014 ± 1ᵇ</td>
<td>79.3 ± 7.8ᵃ</td>
<td>2.3 ± 0.9ᵃ</td>
<td>15.4 ± 1.0ᵇ</td>
<td>8.8 ± 0.3ᵇ</td>
<td>495 ± 11ᵇ</td>
</tr>
<tr>
<td>Jan. 3, 2006</td>
<td>7.6–7.7ᵃ</td>
<td>19.6 ± 2.2ᵇ</td>
<td>20.5 ± 0.8ᵇ</td>
<td>1018 ± 4ᵇ</td>
<td>73.7 ± 6.4ᵃ</td>
<td>3.0 ± 1.6ᵃ</td>
<td>2.0 ± 0.2ᵇ</td>
<td>0.7 ± 0.4ᵇ</td>
<td>95 ± 6ᵇ</td>
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<tr>
<td><strong>Site B</strong></td>
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<tr>
<td>Oct. 30, 1999</td>
<td>5.1–6.2ᵇ</td>
<td>24.0 ± 0.3ᵇ</td>
<td>31.1 ± 1.0ᵃ</td>
<td>1015 ± 1ᵇ</td>
<td>74.4 ± 6.1ᵇ</td>
<td>4.2 ± 0.7ᵇ</td>
<td>14.7 ± 1.7ᵇ</td>
<td>12.8 ± 0.2ᵇ</td>
<td>901 ± 15ᵇ</td>
</tr>
<tr>
<td>Nov. 24, 1999</td>
<td>5.3–6.3ᵇ</td>
<td>29.3 ± 2.2ᵃ</td>
<td>26.9 ± 0.6ᵇ</td>
<td>1014 ± 1ᵇ</td>
<td>79.3 ± 7.8ᵃ</td>
<td>2.3 ± 0.9ᵃ</td>
<td>19.3 ± 2.2ᵇ</td>
<td>35.0 ± 3.1ᵇ</td>
<td>813 ± 17ᵇ</td>
</tr>
<tr>
<td>May 11-13, 2000</td>
<td>5.3–6.4ᵇ</td>
<td>26.5 ± 3.0ᵇ</td>
<td>29.5 ± 2.3ᵃ</td>
<td>1007 ± 1ᵇ</td>
<td>62.7 ± 6.5ᵇ</td>
<td>2.4 ± 1.2ᵃ</td>
<td>19.7 ± 3.0ᵇ</td>
<td>18.4 ± 2.2ᵇ</td>
<td>1531 ± 23ᵇ</td>
</tr>
<tr>
<td>Jan. 4, 2006</td>
<td>7.2–7.7ᵃ</td>
<td>17.5 ± 1.0ᵇ</td>
<td>20.6 ± 0.7ᵇ</td>
<td>1018 ± 4ᵇ</td>
<td>73.7 ± 6.4ᵃ</td>
<td>3.0 ± 1.6ᵃ</td>
<td>2.3 ± 0.2ᵇ</td>
<td>-0.1 ± 0.4ᵇ</td>
<td>20 ± 5ᵇ</td>
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<tr>
<td><strong>Site C</strong></td>
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<tr>
<td>June 30, 2000</td>
<td>5.6–6.3ᵇ</td>
<td>33.6 ± 2.9ᵃ</td>
<td>33.6 ± 2.1ᵃ</td>
<td>1007 ± 1ᵇ</td>
<td>71.1 ± 7.4ᵃ</td>
<td>1.5 ± 0.8ᵃ</td>
<td>20.7 ± 4.0ᵇ</td>
<td>163.3 ± 7.7ᵃ</td>
<td>1349 ± 16ᵃ</td>
</tr>
<tr>
<td>Jan. 5, 2006</td>
<td>7.3–7.6ᵃ</td>
<td>22.5 ± 2.0ᵇ</td>
<td>20.0 ± 0.9ᵃ</td>
<td>1018 ± 4ᵃ</td>
<td>73.7 ± 6.4ᵃ</td>
<td>3.0 ± 1.6ᵃ</td>
<td>2.6 ± 0.5ᵇ</td>
<td>0.2 ± 0.2ᵈ</td>
<td>-32 ± 7ᵈ</td>
</tr>
</tbody>
</table>

Means ± SD (n = 4, *n = 9). Emission rate was measured by chamber method. Values in the same column that do not share the same alphabetic superscript are significantly different at 5% level according to Duncan’s multiple range tests.

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**Fig. 2.** Air temperature and atmospheric concentrations of CO₂ and CH₄ measured with FTIR spectroscopy at site A of the Fu-Der-Kan closed landfill. Data are presented for (a–b) October 30, 1999 and (c–d) November 24, 1999: (a) and (c) air temperature; (b) and (d) atmospheric concentrations of CO₂ and CH₄.
Fig. 3. Air temperature, atmospheric concentrations and emission rates of CO\textsubscript{2} and CH\textsubscript{4} of the Fu-Der-Kan landfill after as the multi-use recreational park. (a–c) country activity area on January 4, 2006. (d–f) Sun plaza on January 5, 2006. (g–i) culture of indigenous people’s area on January 12, 2006. (a), (d) and (g) air temperature; (b), (e) and (h) atmospheric concentrations of CO\textsubscript{2} and CH\textsubscript{4}; (c), (f) and (i) CO\textsubscript{2} and CH\textsubscript{4} emission rates.

Fig. 4. Air temperature, and atmospheric concentrations of CO\textsubscript{2} and CH\textsubscript{4} measured with FTIR spectroscopy at site B of the Fu-Der-Kan closed landfill. Data are presented for (a–b) October 30, 1999 and (c–d) November 24, 1999: (a) and (c) air temperature; (b) and (d) atmospheric concentrations of CO\textsubscript{2} and CH\textsubscript{4}. 

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3.3. CH₄ and CO₂ concentrations and emission rates at site B

Air temperature, atmospheric concentrations of CH₄ and CO₂ at site B are shown in Fig. 4. Atmospheric CH₄ concentrations on October 30, 1999 ranged from 1.8 to 3.0 ppm, with an average of 2.4 ± 0.3 ppm. CO₂ concentrations were between 373 and 394 ppm, with an average of 388 ± 6 ppm. On November 24, 1999, atmospheric CH₄ and CO₂ concentrations were ranged from 2.6 to 4.2 ppm, and from 347 to 397 ppm, respectively, with an average of 3.6 ± 0.5 and 378 ± 15 ppm, respectively. CH₄ and CO₂ emission rates were 12.8 ± 0.2 to 35.0 ± 3.1 mg m⁻² h⁻¹ and 813 ± 17 to 901 ± 15 mg m⁻² h⁻¹, respectively (Table 1). Both the atmospheric concentration and emission rate of CH₄ were high on November 24, 1999, which may be partly attributed to high soil organic C and moisture content. Different situations, in contrast to site A, were found in site B on November 24, 1999. The new cover material used in site B on November 24, 1999 might contain high organic C. Therefore, both the organic C content of cover soil and the CH₄ emission in site B on November 24, 1999 were higher than those on October 30, 1999.

Sun plaza was constructed at site B during the years 1999 to 2003. Atmospheric CH₄ and CO₂ concentrations on January 5, 2006 ranged from 1.8 to 2.3 ppm and from 332 to 395 ppm, respectively. Average values were 1.9 ± 0.2 and 361 ± 12 ppm, respectively. CH₄ emission rate was −1.1 ± 0.5 to 0.6 ± 0.5 mg m⁻² h⁻¹ and CO₂ emission rate was −98 ± 11 to 224 ± 38 mg m⁻² h⁻¹. Average CH₄ and CO₂ emission rates were −0.1 ± 0.4 and 20 ± 3 mg m⁻² h⁻¹, respectively (Fig. 3). These values were lower than those in 1999 because of the more stable buried refuse, low soil organic C and moisture content.

3.4. CH₄ and CO₂ concentrations and emission rates at site C

The CH₄ emission rate (163.3 ± 7.7 mg m⁻² h⁻¹) and CO₂ emission rate (1349 ± 16 mg m⁻² h⁻¹) at site C were the highest of the three sites and the variation among the nine installed chambers was also large (Table 1). The elevated CH₄ and CO₂ emission rates were due to new MSW buried in this site during 1999.

Site C was converted into a culture of indigenous people’s area during the years 2001 to 2003. Atmospheric CH₄ concentrations were 1.9 to 2.3 ppm, with an average of 2.1 ± 0.2 ppm on January 12, 2006. CO₂ concentrations ranged from 343 to 422 ppm with an average of 397 ± 13 ppm. CH₄ and CO₂ emission rates were −0.8 ± 0.7 to 1.1 ± 0.5 mg m⁻² h⁻¹ and −135 ± 8 to 113 ± 15 mg m⁻² h⁻¹, respectively. Average CH₄ and CO₂ emission rates were 0.2 ± 0.2 and −32 ± 7 mg m⁻² h⁻¹, respectively (Figs. 3 g–i). CH₄ emission rates were in the same level as those in sites A and B.

3.5. Diurnal variation of atmospheric CH₄ and CO₂ concentrations and emission rates at site B

Fig. 5 shows the diurnal variations in air temperature, atmospheric concentrations and emission rates of CH₄ and CO₂ from May 11 to 13, 2000 at site B. The atmospheric CH₄ concentrations ranged from 1.3 to 3.8 ppm with an average of 2.7 ± 0.8 ppm. The concentration of
CH4 was high at night, which correlates with CH4 emission rate. The atmospheric CO2 concentration averaged at 374 ± 15 ppm and ranged from 350 to 399 ppm. The air temperature was the highest (31.7 °C) at noon on May 11 and the lowest (19.5 °C) at night on May 12.

The average CH4 emission rate at site B was 18.4 ± 3.3 mg m⁻² h⁻¹ (Table 1). The maximum rate was 54.1 mg m⁻² h⁻¹ at 3 a.m. on May 13, and the minimum rate was 2.5 mg m⁻² h⁻¹ at midnight on May 12. The average CO2 emission rate was 1531 ± 23 mg m⁻² h⁻¹. The maximum rate was 2302 mg m⁻² h⁻¹ at 9 a.m. on May 11, and the minimum rate was 893 mg m⁻² h⁻¹ at midnight on May 12.

The diurnal variation in CO2 emission was very consistent after completion of the recreational park for country activity area, Sun plaza and culture of indigenous people’s area in January 2006. The CO2 emission rates were high at night because of daytime photosynthetic activity by the grass and small trees. The diurnal variation of CH4 emission was insignificant due to the low organic matter content and more stable burial waste.

4. Discussion

Compared to other terrestrial ecosystems, landfills are characterized by a high rate of CH4 production and large CH4 gradients from the deeper production zones to the soil–atmosphere interface. Bogner et al. (1999) found that CH4 concentration was low at the surface and increased by about two orders of magnitude at 100 m depth. In paddy fields, the CH4 emission rate has been found to be by about two orders of magnitude at 100 m depth. In

In conclusion, both gas-type open-path FTIR spectroscopy and GC chamber method have been successfully used to measure atmospheric concentrations and emission rates of greenhouse gases in a closed and reconstructed landfill. The Fu-Der-Kan landfill closed in 1995 and was
reconstructed to form a multi-use recreational park during 1999 to 2003. Atmospheric concentrations and emission rates for CH4 were higher at night than the daytime during 1999 to 2000. The CH4 concentrations and emission rates in the closed landfill and reconstructed park were less than 10% and 1% of those in the active landfill, respectively. A systematic and continuous study of greenhouse gas emissions from closed landfills provides information about the field degradation of MSW, which in turn will help in the design of future landfills.

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