Estimation of Methane Emission Rate Changes Using Age-Defined Waste in a Landfill Site

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## Abstract

Long term methane emissions from landfill sites are often predicted by first-order decay (FOD) models, in which the default coefficients of the methane generation potential and the methane generation rate given by the Intergovernmental Panel on Climate Change (IPCC) are usually used. However, previous studies have demonstrated the large uncertainty in these coefficients because they are derived from a calibration procedure under ideal steady-state conditions, not actual landfill site conditions. In this study, the coefficients in the FOD model were estimated by a new approach to predict more precise long term methane generation by considering region-specific conditions. In the new approach, age-defined waste samples, which had been under the actual landfill site conditions, were collected in Hokkaido, Japan (in cold region), and the time series data on the age-defined waste sample's methane generation potential was used to estimate the coefficients in the FOD model. The degradation coefficients were 0.050 (1/y) and 0.062 (1/y) for paper and food waste, and the methane generation potentials were 214.4 (mL/g-wet waste) and 126.7 (mL/g-wet waste) for paper and food waste, respectively. These coefficients were compared with the default coefficients given by the IPCC. Although the degradation coefficient for food waste was smaller than the default value, the other coefficients were within the range of the default coefficients. With these new coefficients to calculate methane generation, the long term methane emissions from the landfill site was estimated at $1.35\times10^4$ $m^3$-$CH_4$, which corresponds to approximately 2.53% of the total carbon dioxide emissions in the city ($5.34\times10^5$ t-$CO_2$/y).

*Key words: Methane emission, age-defined wastes, change in methane generation rate, the first-order decay model, cold climate region*

## 1. Introduction

The landfilling of biodegradable organic materials, such as kitchen waste, papers and woods causes long term methane gas generation, which can contribute to global warming if not adequately collected by a gas-recovery system. Emissions from landfill sites account for 30% of the total anthropogenic methane emissions in Europe, 34% of those in the US, and 10% of anthropogenic methane emissions worldwide (Perdikea et al., 2008). The EU landfill directive (European Union, 1999) required EU member countries to reduce biodegradable organic waste going to landfill sites and set a goal of reducing the amount of biodegradable waste going to landfills by 65% by 2020 compared to 1995 levels. The evaluation and monitoring of methane emissions from landfill sites is meaningful for preventing global warming.

In general, there are two different approaches to estimate methane emissions from landfill sites: the direct approach, which is based on landfill gas (LFG) emission measurements from the landfill surface, and an indirect calculation based on a straightforward mass balance equation between methane production, recovery and oxidation at the landfill site (Bella et al., 2011).

In the direct approach, methane emissions can be estimated 1) from the soil gas concentration profiles of methane through the landfill cover materials or 2) by using the static and/or dynamic closed flux chamber method, which is applied to a relatively small part of the landfill surface (Bella et al., 2011). However, the direct approaches have limitations, because the measured values are only obtained at discrete points and for a limited short time of the measurement (Spokas et al., 2003). There are spatial and time variations in landfill methane emissions. To reduce the influence of these variations, some larger area measurement assessments, such as a dynamic plume measurement using CO and $N_2O$ as tracer gasses was proposed (Scheutz et al., 2011). However, scaling the direct measurement approach to long term methane emissions from landfill sites is difficult because the measurement is based on the limited temporal period of the measurement.

In contrast, for the indirect calculation, several models have been developed with different orders of kinetics, including zero-, first-, and second-order models, as well as some more complex models (Friedrich and Trois, 2011; Meima et al, 2008). Most of the models are based on the first-order decay (FOD) model to predict methane generation as a surrogate for landfill methane emissions, including the Intergovernmental Panel on Climate Change (IPCC) model (IPCC, 2006), TNO model (Oonk et al., 1994), GasSim Lite model (Golder Associates, 2010), LandGEM model (US-EPA, 2005), and Afvalzorg model (Scharff, 2010). However, the coefficients introduced in the FOD models typically overestimate LFG production because the coefficients are derived from a calibration procedure in ideal steady-state conditions (e.g., gas production factors, conversion coefficients for organic matter degradation) (Cossu et al., 1996). These ideal steady-state conditions differ significantly from actual landfill site conditions. Amini et al. (2011) evaluated the uncertainty in estimated landfill gas generation rates, including the default coefficients given by the IPCC. The uncertainty in the modeled LFG generation rate varied from ±11% to ±17% while the landfills were open, from ±9% to ±18% at the end of waste placement, and from ±16% to ±203% at 50 years after waste placement ended.

Other than the amount and composition of waste, such as the biodegradable organic fraction, there are a variety of factors influencing methane generation, including climate conditions, such as temperature and precipitation, landfilling methods, such as waste compaction, leachate recirculation, and anaerobic or semi-aerobic landfilling (Friedrich and Trois, 2011; Lou and Nair, 2003). Thus, the coefficients in the FOD model should be determined under more realistic landfill site conditions.

This study attempts to establish a new approach to estimate more accurate methane generation rate coefficients through investigation of a real landfill site that has been operated for over 20 years in a city of Hokkaido, Japan. In the new approach, age-defined wastes are sampled from the real landfill site. The time of year when each waste was landfilled is defined based on working records of the landfill. Furthermore, these wastes have been in the landfill under real circumstances for specific periods of time. Although the new approach requires some assumptions and prerequisites in terms of waste composition, the degradation rate, and sample representativeness, the new approach can be an improved method for estimating the amount of long term methane generation from landfill sites.

## 2. Materials and methods

### 2.1 Procedure of the new approach

This new approach consists of four steps, as shown in Figure 1. First, existing information on the history of municipal solid waste management in the study area and working reports of the landfill at the targeted site are investigated to predict the composition of the landfilled waste, the annual amount of landfilled waste, and the distribution of landfilled waste over space and time.

Second, sampling of age-defined wastes from the landfill site is conducted. The waste composition, moisture, volatile matter and ash contents, and methane generation potential are analyzed to define the main types of wastes contributing to long term methane emissions from the landfill site.

In the third step, the coefficients in the FOD model for each type of wastes are estimated using time series data on the methane generation potential. The long term methane emissions from the landfill site are predicted using the new coefficients and the amount and composition of landfilled waste predicted by the existing information.

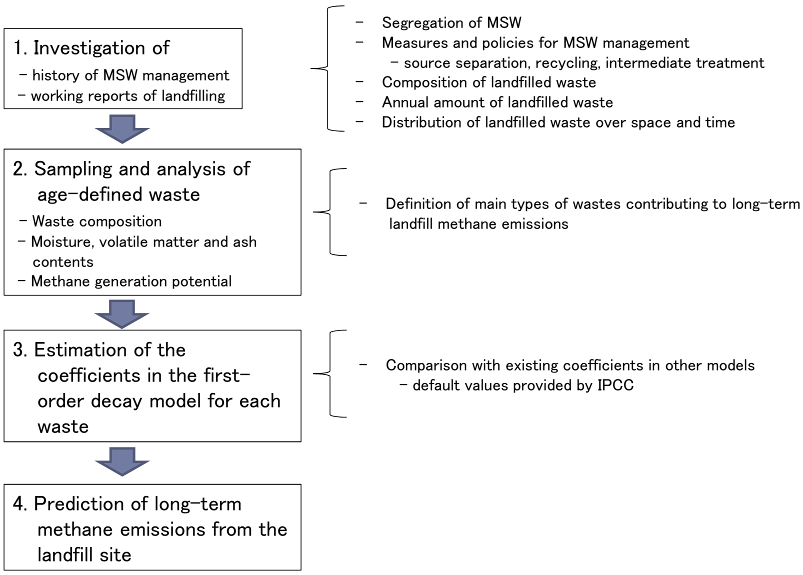


Figure 1 New approach for prediction of long term methane emissions from a landfill site

## 2.2 Prerequisites and assumptions

Some prerequisites and assumptions are required when the new approach is applied to predict long term methane emissions from a specific landfill site.

(A) Prerequisites for the landfill site

- Sampling of age-defined wastes that are clarified by working reports of the landfill is possible.
- The amount and composition of waste at the time when the waste was landfilled are predictable.

(B) Assumptions for landfilled waste

- The changes in the quality of each composition in the landfilled waste are negligible. For example, the quality of paper landfilled in the past is assumed to be the same as that of paper currently being landfilled.
- The biodegradable organic matter is converted to methane and carbon dioxide under anaerobic conditions. In Japan, semi-aerobic landfilling is mainly introduced where fresh air can enter the waste layer from leachate collection pipes. Although the landfilled waste near the pipes is under aerobic conditions, it is commonly recognized that the other large portion of the landfilled waste is under anaerobic conditions.
- Methane oxidation can be evaluated, at least using values from the literature.

**2.3 Specific landfill site investigated**

The landfill sites under consideration are in W city, which is located in the northern region of Hokkaido, Japan. The average temperature is 6.6 $^{o}$C, and the average annual precipitation is 1,058 mm. W city does not have waste incinerators, and all of the wastes except recyclables, such as steel, aluminum cans, and PET bottles, are directly landfilled. Table 1 provides a summary of the old and new landfill sites in W city. The new landfill site is a closed system disposal facility (CSDF) with a roof that prevents rainfall from infiltrating waste layers during operational and postclosure periods (Ishii and Furuichi, 2011). However, the waste degradation conditions in the new landfill site are assumed to be the same as those in the old landfill site because both landfill sites introduce semi-aerobic landfilling, and the artificial watering required for biodegradation is conducted in the new landfill site.

Table 1 Summary of the landfill sites under consideration

| | Old landfill | | New landfill | |
|---|---|---|---|---|
| | 1st section | 2nd section | 1st section | 2nd section |
| Period of landfilling | 1988 - 1998 | 1999- 2007 | 2007-2009 | 2010- |
| Landfilled waste | MSW (including kitchen waste) | | | |
| Current situation | Closure | Closure | Closure | Operating |
| Area | 205,700 $m^2$ | | 6,750 $m^2$ | 2,250 $m^{2*}$ |
| Volume | 1,200,542 $m^3$ | | 6,463 $m^3$ | 2,154 $m^{3*}$ |

* the amount of landfilled waste in 2010.

Table 2 shows the source separation of waste in 2010 and the year of commencement for the collection of recyclables. W city collects three types of waste: general waste for landfilling, bulky waste, and recyclables. In addition, the recyclables are separated into 15 types. W city started collecting recyclables step by step since starting the collection of cans, glass bottles, and PET bottles in 1997. In particular, the source separation and collection of paper, such as paper drink packs and newspapers, which can affect the methane emissions from landfill sites significantly, began in 2001.

Table 2 Source separation of waste and the year of commencement for the collection of recyclables

| Type of waste | | Year of commnecement |
|---|---|---|
| General waste for landfilling | | - |
| Bulky waste | | - |
| Recyclables | 1. Cans<br>2. Glass bottles<br>3. PET bottles | 1997 |
| | 4. Paper drink pack<br>5. Newspaper<br>6. Magazines<br>7. Cardboard<br>8. Other papers | 2001 |
| | 9. Clothes<br>10. Fluorescent<br>11. Dry battery | 2006 |
| | 12. Metals<br>13. Plastic containers and packages<br>14. White plastic tray | 2008 |
| | 15. Thermometer | 2009 |

Figure 2 shows a time series of the amount of landfilled waste (household and business wastes) according to the W city's landfilling records. The amount of landfilled waste dropped significantly in 2001 because the separate collection of paper began, as described before, and a charging system began for bulky waste, such as beds and bicycles. This study considered the change in both of the amount and composition of landfilled wastes to predict long term methane generation at the landfill sites in W city.

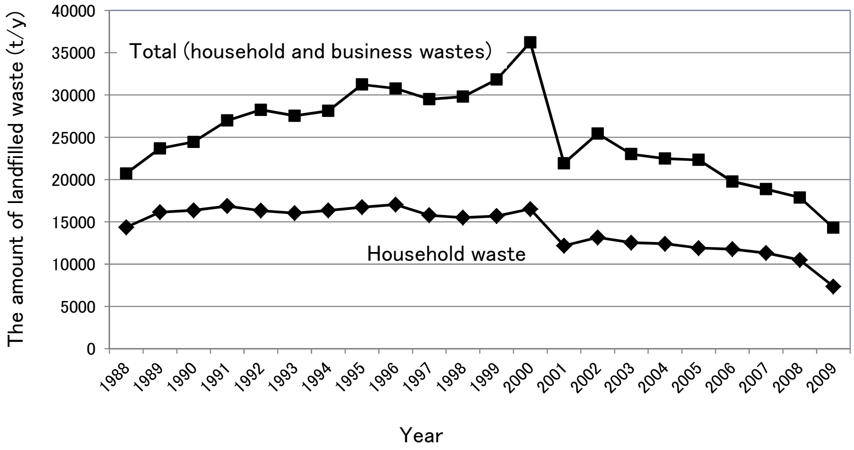


Figure 2 Time series of the amount of landfilled wastes in W city

### 2.4 Sampling of age-defined waste

At the landfill sites, waste was landfilled in the compartment spaces that were constructed next to the filled out compartment, filling one by one. This method of landfilling was advantageous for this study to define the age of landfilled wastes by analyzing W city's landfilling records. Considering the inhomogeneous nature of landfilled waste over space and time, the first section (landfilling from 1988 to 1998) of the old landfill site was separated into two areas, and three samples were collected from each area, as shown in Figure 3. The second section (from 1999 to 2007) of the old landfill site was also separated into two areas, and three samples and two samples were collected from each area. With regard to the first section (from 2008 to 2009) of the new landfill site, waste samples were impossible to be obtained because the final cover sheet was already implemented. Two samples were collected from the second section (from 2010) of the new landfill site.

In the sampling from the old landfill site, a loading shovel was used to remove the top cover soil (approximately 2 m in depth), and then approximately 0.4 $m^3$ of waste was excavated. Approximately 20 L of waste was collected from the excavated waste for laboratory analysis by fractional sampling, considering sample representativeness. In the sampling from the new landfill site, waste samples were collected twice with short interval in August and October 2010 because it was needed to estimate the biodegradation rate of certain waste types, such as kitchen waste, which can biodegrade easily and quickly.

Ideally, waste samples should be collected from deeper depths in the landfill site because the waste just below final cover might not be representative of the actual waste mass (e.g. affected by atmospheric temperature fluctuations). In addition, there are limitations of sampling in terms of machinery available. Since this study assumed that a long term change in waste degradation rate must be larger than a vertical change, wastes at 2 - 3 m in depth were collected.

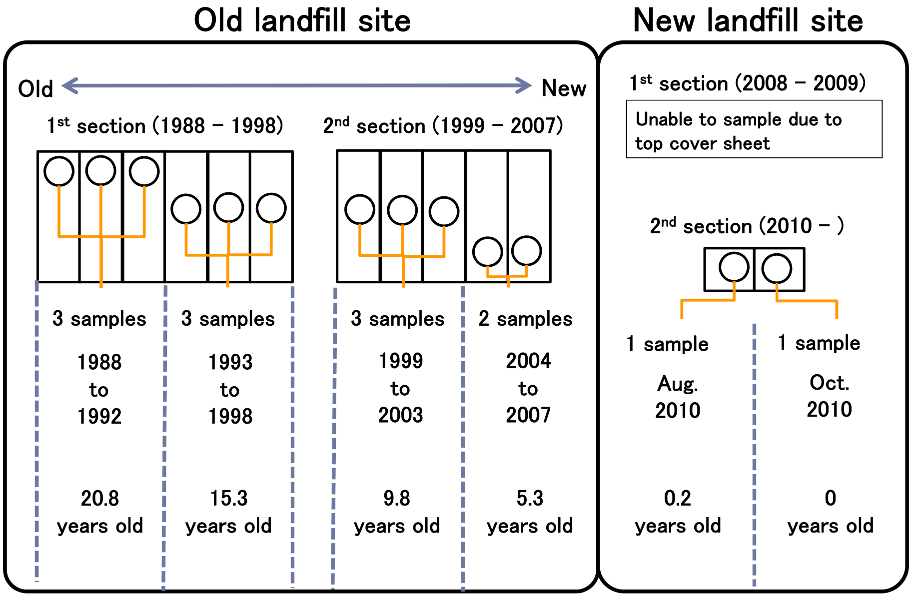


Figure 3 Sampling of age-defined waste from the landfill sites

## 2.5 Analysis of the age-defined waste

### 2.5.1 Waste composition

The 20 L of waste samples were sorted by hand into the following 12 types of wastes: (1) paper, (2) bamboo and straw, (3) grass and wood, (4) kitchen waste, (5) metal, (6) textiles, (7) plastic, (8) rubber, (9) glass and ceramics, (10) fecal waste of pets, (11) baby diapers, and (12) other types of waste. The other types of waste refer to all of the materials that were not categorized into (1) to (11) by visual examination.

### 2.5.2 Moisture, volatile matter and ash contents

The waste samples were dried at 105 °C until their weight stabilized, and the moisture content was determined by the change in the weight of the samples. The dried waste samples were then heated at 600 °C for 6 hours to determine the volatile matter (volatile solid, VS) and ash contents.

### 2.5.3 Methane generation potential

The methane generation potential was measured for each sample by batch testing. Three types of wastes (paper, kitchen waste, and other types of waste) were used. The procedure was as follows:

1) One gram of waste after drying and shredding, and 100 mL of sludge as inoculation were put into a 200-mL vial. The sludge for inoculation was obtained from a biogas plant for kitchen waste. Four duplicates and a control sample without waste were prepared.

2) The vial was sealed after headspace gas was replaced with nitrogen gas.

3) The vial was kept in a constant temperature reservoir (35 °C).

4) The amount of biogas generation was measured regularly; the methane and carbon dioxide concentrations were measured by gas chromatography for approximately 50 days, until the biogas generation becomes negligible.

### 2.6 Formula of the first-order decay model

Methane generation potential $P_i$ (t) (mL-$CH_4$/g-TS or $m^3$-$CH_4$/t-TS] at $x$ years after landfilling, which means remaining methane generation potential, can be described by the FOD model shown in eq. (1). The FOD model implies that a relatively large amount of landfill gas is formed immediately after landfilling, and that this amount gradually decreases with time.

$$P_i\ (x) = P_i^0 e^{-kx} \qquad (1)$$

where

$P_i\ (x)$ : the methane generation potential of waste $i$ at $x$ years after landfilling ($m^3$-$CH_4$/t-TS)

$P_i^0$ : the methane generation potential of waste $i$ at $0$ years after landfilling ($m^3$-$CH_4$/t-TS) ($= P_i\ (0)$)

$k_i$ : the degradation coefficient of waste $i$ (1/y)

$i$ : the type of waste

Using the amount of waste $i$ landfilled in year y, $W_i^y$, the total methane generation potential of waste $i$ landfilled in year y at $x$ years after landfilling, $M_i^{y,x}$, is presented in eq. (2).

$$M_i^{y,x} = W_i^y \times P_i\ (x) \qquad (2)$$

$W_i^y$ : the amount of waste $i$ landfilled in year y (t)

$M_i^{y,x}$ : the total methane generation potential of waste $i$ landfilled in year y at $x$ years after landfilling ($m^3$-$CH_4$)

Using the difference between the total methane generation potential according to eq. (2) for waste $i$ landfilled in year y, the total methane generation from $x$ years to $x+z$ years after landfilling is calculated with eq. (3).

$$M_i^{y,x} - M_i^{y,x+z} = W_i^y \times (P_i\ (x) - P_i\ (x+z)) \qquad (3)$$

For example, for waste $i$ landfilled in 1988 at 23 years after landfilling, the total methane generation for 1 year between 2010 and 2011 is calculated as follows:

$$M_i^{1988,23} - M_i^{1988,23+1} = W_i^{1988} \times (P_i\ (23) - P_i\ (23+1)) \qquad (4)$$

For the waste $i$ landfilled in other years, the total methane generation was calculated for each year in the same manner. The time series in the total methane generation for all types of waste was obtained by summing the total methane generation for the same year, as shown in Figure 4.

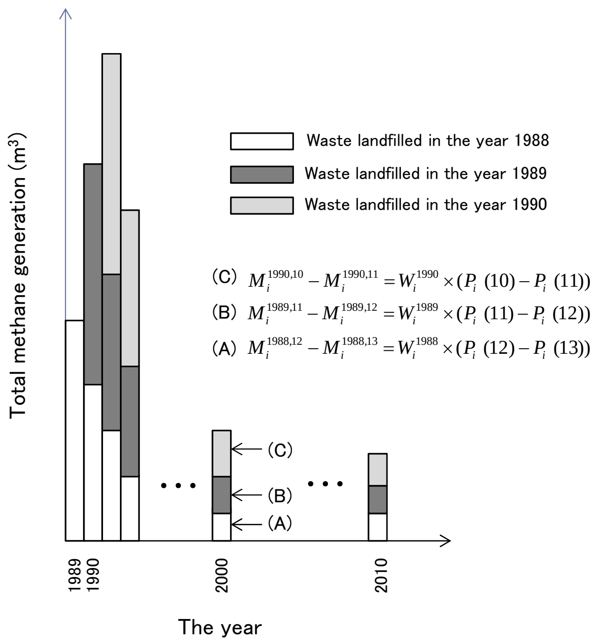


Figure 4 Example of calculation of the total methane emissions for waste $i$

# 3. Results and discussion

## 3.1 Prediction of the history of waste composition by the record of landfill

Information on both of the amount and composition of landfilled waste are required in this study. The data on the annual amount of waste landfilled were obtained for each year from 1988 to 2010, as already described in Figure 2. However, only limited data on the waste composition were available during this time period. In W city, the collection of recyclable paper began in 2001, as mentioned previously. Thus, by assuming that the waste composition did not change over time after 2001, the waste composition from 2001 to 2010 was determined using the result of a waste composition survey that was conducted in 2004 by W city. In contrast, the waste composition before 2000 was estimated using that of other cities without the collection of paper as recyclables. Table 3 shows the results of the prediction of the ratio of paper and kitchen waste landfilled in each time period. The ratio of paper ranged from 0.166 to 0.284 for household waste and from 0.308 to 0.483 for business waste. The ratio of kitchen waste ranged from 0.385 to 0.474 for household waste and from 0.192 to 0.352 for business waste.

Table 3 Prediction of the ratio of landfilled paper and kitchen waste

| Type of waste | | 1988 to 1993 | 1994 to 2000 | 2001 to 2010 |
|---|---|---|---|---|
| Household waste | Paper | 0.166 | 0.284 | 0.266 |
| | Kitchen waste | 0.385 | 0.440 | 0.474 |
| Business waste | Paper | 0.483 | 0.352 | 0.308 |
| | Kitchen waste | 0.192 | 0.201 | 0.352 |

### 3.2 Composition of age-defined waste samples

The average waste composition of each age-defined waste sample was determined, as shown in Table 4. Although there seemed to be variations in the elapsed time, the amount of (1) paper, (4) kitchen waste, plastic, and (11) other types of waste accounted for 80-90% of the total waste. The amounts of (2) bamboo and straw, and (3) grass and wood were small or zero. The amount of kitchen waste disappeared between 0.2 years and 5.3 years after landfilling, indicating that after a few years of landfilling, the kitchen waste degraded to the level at which it was not recognized by visual examination. Although paper has a tendency to decrease with time, approximately 20% of the paper remained even after 20 years of landfilling, as shown in Table 4.

Because biodegradable organic matter, such as paper and kitchen waste, degraded over time, the fraction of nonbiodegradable materials, such as plastic, tended to increase over time. In addition, the proportion of other types of waste increased with time, which indicates that the kitchen waste and paper were transformed into the other types of waste. Because the paper was extracted definitely from the other types of waste during hand sorting, the kitchen waste is mainly assumed to be converted to other types of waste. In other words, the portion of kitchen waste that is not still degraded can be categorized as other types of waste.

From the results of waste composition, papers, kitchen waste and other types of waste were extracted and defined as materials which can cause long term methane emissions from the landfill sites.

Table 4 Composition of age-defined waste samples

| | Time elapsed from landfilling (y) | | | | | |
|---|---|---|---|---|---|---|
| | 0 | 0.2 | 5.3 | 9.8 | 15.3 | 20.8 |
| (1) Paper | 30% | 35% | 18% | 19% | 26% | 21% |
| (2) Bamboo and straw | 0% | 0% | 0% | 0% | 0% | 0% |
| (3) Grass and wood | 1% | 6% | 3% | 2% | 4% | 2% |
| (4) Kitchen waste | 33% | 44% | 0% | 1% | 0% | 0% |
| (5) Metal | 5% | 0% | 5% | 2% | 10% | 2% |
| (6) Textiles | 7% | 4% | 6% | 6% | 1% | 1% |
| (7) Plastic | 16% | 11% | 52% | 49% | 34% | 45% |
| (8) Rubber | 0% | 0% | 0% | 0% | 0% | 0% |
| (9) Glass and ceramics | 0% | 0% | 0% | 2% | 3% | 5% |
| (10) Fecal waste of pets | 0% | 0% | 0% | 0% | 0% | 0% |
| (11) Baby diapers | 7% | 1% | 0% | 0% | 0% | 0% |
| (12) Other types of waste | 0% | 0% | 16% | 19% | 23% | 23% |

### 3.3 Moisture, volatile matter and ash contents

The results are shown in Figure 5 and 6. The results for kitchen waste and other types of waste are shown in the same graph because of the aforementioned assumption.

The average moisture contents were 65.7% for paper and 60.8% for kitchen waste and other types of waste, and the VS ranges were 21.4% to 36.9% for paper and 17.9% to 27.3% for kitchen waste and other types of waste.

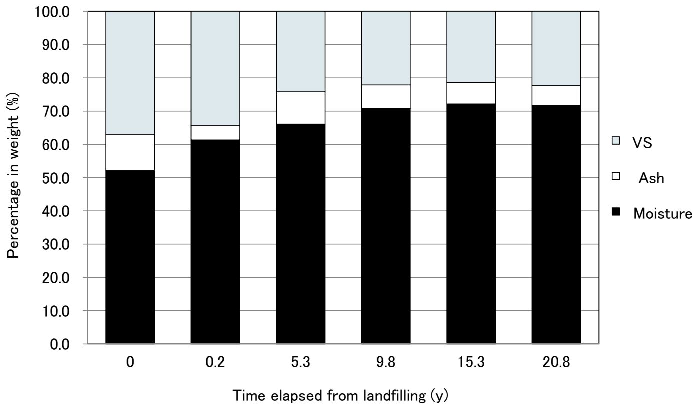


Figure 5 Moisture, volatile matter and ash contents in the paper

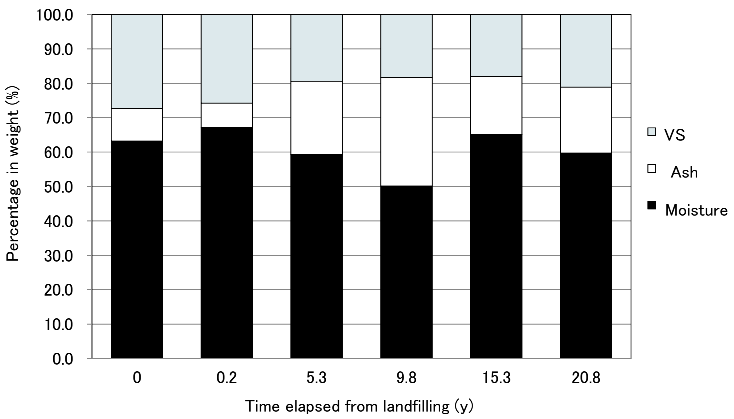


Figure 6 Moisture, volatile matter and ash contents in the kitchen waste (0 and 0.2 years) and other types of waste (5.3 to 20.8 years)

### 3.4 Methane generation potential

Figure 7 shows the amount of methane generated per weight of dried waste (total solid, TS = VS + ash) as a function of time, which were calculated by deducing the methane generation of the control sample with no waste. With the aforementioned assumption, the methane generation potentials of the kitchen waste and other types of waste were plotted together.

The methane generation of paper just after landfilling was 320 mL-$CH_4$/g-TS and it decreased to 114 mL-$CH_4$/g-TS after 21 years (60% of the biodegradable organic matter in the paper was degraded). In contrast, the methane generation of kitchen waste and other types of waste just after landfilling was the 250 mL-$CH_4$/g-TS, and it decreased to 65 mL-$CH_4$/g-TS (74% of the biodegradable organic matter degraded) after 21 years. These facts suggest that, even after more than 20 years of landfilling, 40% of the methane generation potential in paper and 26% of that in food waste and other types of waste still remained, with paper contributing more to future methane emissions than kitchen waste and other types of waste. Therefore, the measured methane generation in Figure 7 is able to be considered as the remaining methane generation potential, from which methane gas can be generated from the remaining decomposable organic matter in each waste.

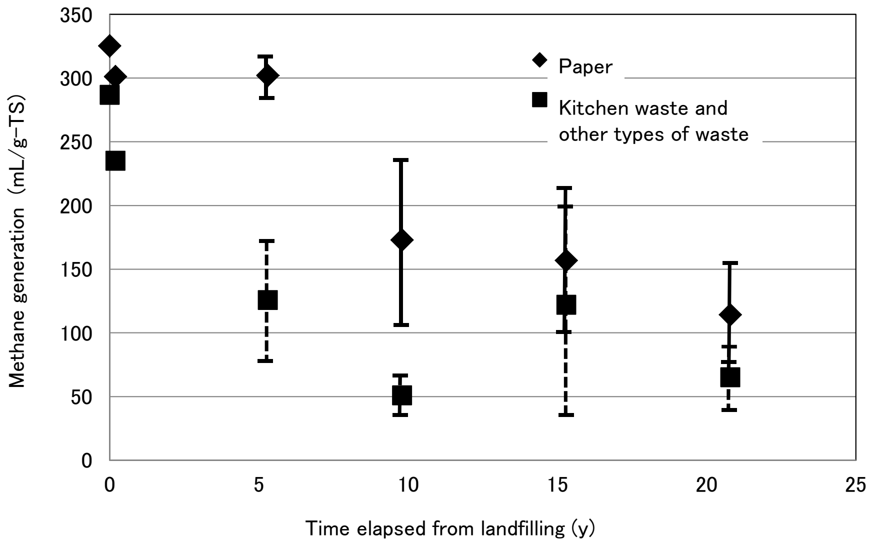


Figure 7 Changes in the methane generation potential

### 3.5 Determination of coefficients in the FOD model

The two lines in Figure 8 show the curve-fitted lines using the first-order decay model in eq. (1) to determine $P_i^0$ and $k_i$. The results are presented in eq. (5) for paper and eq. (6) for kitchen waste and other types of waste.

$$p_s(x) = 326.4e^{-0.050x} \quad (5)$$

$$p_r(x) = 208.2e^{-0.062x} \quad (6)$$

The correlation coefficients were 0.934 for paper and 0.578 for kitchen waste and other types of waste.

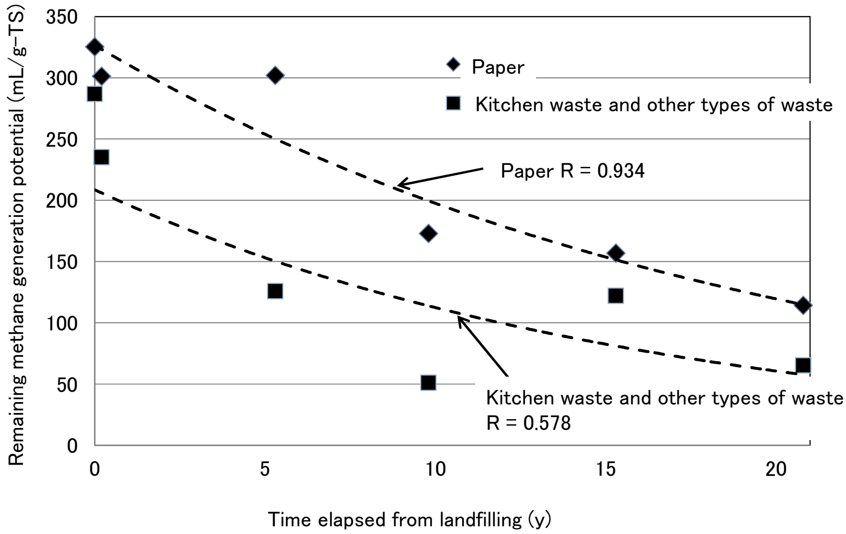


Figure 8 Curve fittings for the methane generation potential

The IPCC (2006) provides the default methane generation rate, which is the same as the degradation coefficient in this study, and methane generation potential. W city in Hokkaido, Japan belongs to the boreal-to-temperate climate zone. The ratio of the mean annual precipitation (MAP) to the potential evapotranspiration (PET) is greater than 1. In this case, as shown in Table 5, the degradation coefficients are 0.06 (1/y) (range: 0.05 (1/y) to 0.07 (1/y)) for paper and 0.185 (1/y) (0.1 (1/y) to 0.2 (1/y)) for food waste. In this study, the degradation coefficient for paper was 0.050 (1/y), which is within the range reported by the IPCC, but that for kitchen waste and other types of waste was 0.062 (1/y), which was one-third of the value reported by the IPCC, perhaps because the average temperature in W city is low (the average temperature is 6.6 $^{o}$C as described before). Because of the assumption that the kitchen waste would convert to other types of waste, which were not distinguished by visual examination, the degradation coefficient was calculated using the time series data on the methane generation potentials for both kitchen waste and other types of waste. Thus, the degradation coefficient was smaller than the default value given by the IPCC. Although the assumption that the kitchen waste would convert to other types of waste must be reevaluated, the degradation coefficient for kitchen waste and other types of waste includes certain influences of materials with methane generation potential, which is obtained from real field data. This fact was obtained by sampling age-defined wastes from real landfill sites, which has not been considered before, and suggests that the default values given by the IPCC may be too large to predict long term methane emissions from landfill sites.

With regard to the methane generation potential, the IPCC provides the following equation:

$$L_0 = DOC \cdot DOC_f \cdot MCF \cdot F \cdot 16/12 \qquad (7)$$

where,

$L_0$ : the methane generation potential (mL-$CH_4$/g-wet)

$DOC$ : the degradable organic carbon in the deposition year (-)

$DOC_f$ : the fraction of DOC that can decompose (assumed to be 1)

$MCF$ : the methane correction factor for aerobic decomposition in the deposition year (-) (assumed to be 1 for anaerobic management)

$F$ : the fraction of methane in generated landfill gas (volume fraction) (assumed to be 0.5)

16/12 : the molecular weight ratio $CH_4$/C (ratio)

The IPCC (2006) lists the DOC values for paper and kitchen waste as 36 -45% and 8 – 20%, respectively. Using these values and the assumed values in eq. (7), the values of $L_0$ were calculated as 336 – 420 mL/g-wet waste for paper, and 75 – 187 mL/g-wet waste for kitchen waste, as shown in Table 5. It should be noted that research into $DOC_f$ has progressed. Recently, Bareither et al. (2012) reported $DOC_f$ of 0.49 for paper. Using this value, the values of $L_0$ were also calculated in Table 5. Meanwhile in this study, when using the average moisture content of the waste samples as described in the section 3.3, $P_i^0$ in eqs. (5) and (6) was corrected to 214.4 mL/g-wet waste for paper and 126.7 mL/g-wet waste for kitchen waste and other types of waste. In the parameters, the methane generation potential of paper in this study was smaller than the IPCC value, perhaps because the VS value was relatively small compared to the default DOC value, although the VS and DOC values cannot be compared directly. However, comparing the methane generation potential calculated by using $DOC_f$ of 0.49, the methane generation potential in this study was almost the same.

Table 5 Comparison of the existing parameters

| | Degradation coefficient (1/y) | | | Methane generation potential | | | | |
|---|---|---|---|---|---|---|---|---|
| | IPCC* | | | IPCC | | This study | | |
| | Default | Range | This study | DOC (%) | $L_0$ (mL/g-wet waste) | VS (%) | Average of Moisture content (%) | $P^0_i$ (mL/g-wet waste) |
| Paper | 0.06 | 0.05-0.07 | 0.050 | 36-45 | 336-420 (165-206)** | 21.4-36.9 | 65.7 | 214.4 |
| Food waste | 0.185 | 0.1-0.2 | 0.062 | 8-20 | 75-187 | 17.9-27.3 | 60.8 | 126.7 |

* The reference vaues are for boreal and temperate and wet (MAP>PET) in the climate zone

** $DOC_f$ value is changed from 1 to 0.49 for paper (Bareither et al., 2012)

Regarding to further research to refine the new approach, the assumption of this study that kitchen waste would change to other types of waste, which was recognized by visual examination, as degradation progressed, should be reviewed. However, it is hard to find the food waste residue in real landfill sites. Therefore, food waste samples can be placed in a real landfill site so that the each sample be collected in different time. For example, each food waste sample can be put in plastic mesh bags, where water and gas can migrate through, and be buried in waste until they were dug out after a designated time.

### 3.6 Prediction of the change in the landfill methane emissions

The amount of methane generation from the old and new landfill sites from the year 2010 was predicted for the next 46 years (that is two times of the period from the beginning of landfilling in 1988 to 2010) using the method described in the section 2.6, as shown in Figures 9 and 10, where the new coefficients determined in the section 3.6, the predicted data on the amount of landfilled waste and the ratios of paper and kitchen waste to the amount of landfilled waste were used.

The methane generation will continue for at least 40 years from now, even in the old landfill site, which started operation 23 years ago. The amounts of methane generation over 46 years from the old and new landfill sites were estimated to be approximately $1.2 \times 10^7$ $m^3$-$CH_4$ and $2.9 \times 10^6$ $m^3$-$CH_4$, respectively, as shown in Figure 11. These generation amounts are close to the total methane generation potential calculated by $\sum_y \sum_i W_i^y \times P_i^0$. Contribution of landfilling paper to the methane emissions was large.

In particular, the amount of methane generation over the 1 year period of 2011 was estimated to be equivalent to $8.5 \times 10^5$ $m^3$-$CH_4$ and $1.7 \times 10^5$ $m^3$-$CH_4$ in the old and new landfill sites, respectively, and the sum reached $1.0 \times 10^6$ $m^3$-$CH_4$. The generated amount is equivalent to $1.5 \times 10^4$ t-$CO_2$ using a value of 21 for the equivalent factor of methane to carbon dioxide. In addition, assuming that the ratio of the methane oxidation process in the final soil cover is 0.1 (IPCC, 2006), up to $1.35 \times 10^4$ t-$CO_2$ of methane may be emitted into the atmosphere. This value corresponds to approximately 2.53% of the total carbon dioxide emissions in W city ($5.34 \times 10^5$ t-$CO_2$/y). The emission control of methane from both landfill sites should be conducted to reduce the total carbon dioxide emissions in W city.

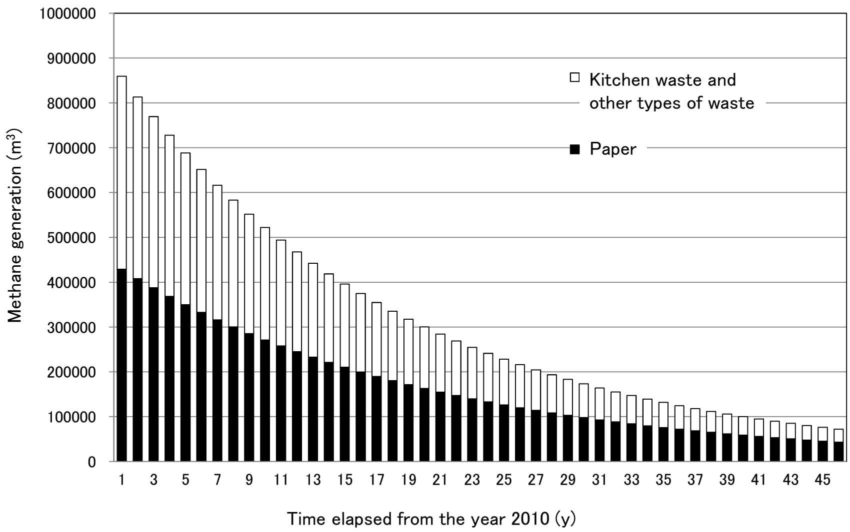


Figure 9 Prediction of long term methane generation from the old landfill sites

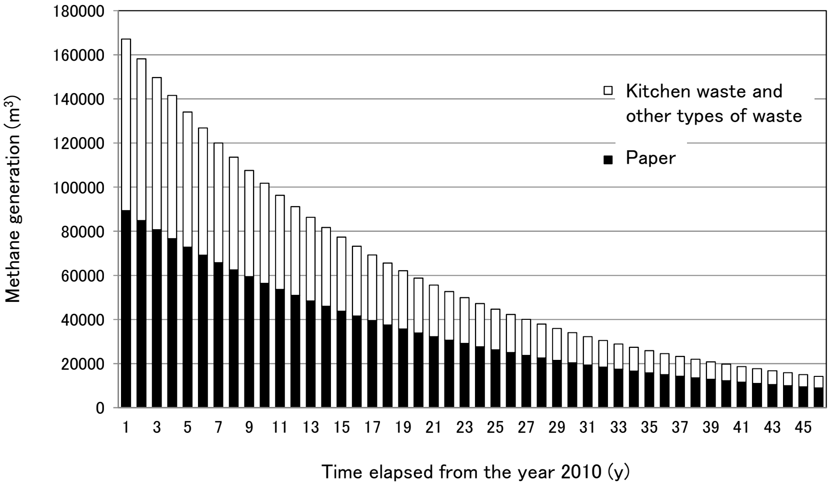


Figure10 Prediction of long term methane generation from the new landfill sites

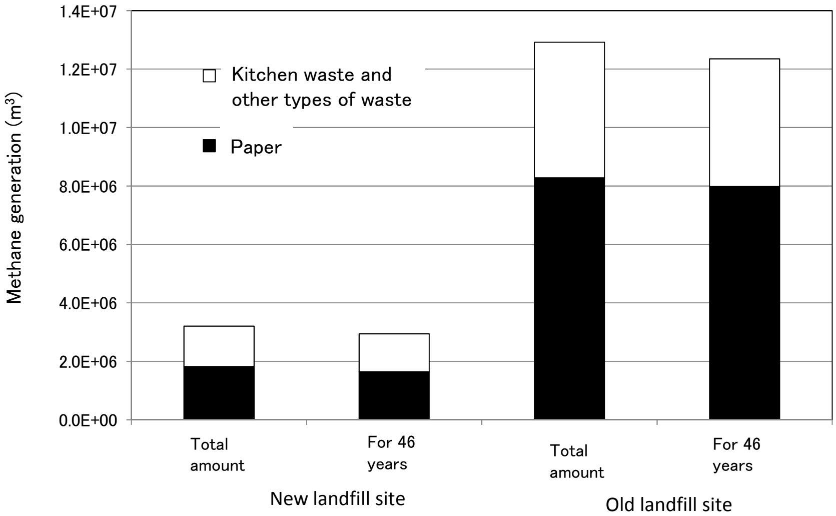


Figure 11 Comparison of the methane generation over 46 years with the total methane generation potential

## 4. Conclusion

This study predicted long term methane emissions from the landfill sites using a new approach, which estimated the coefficients in the first-order decay (FOD) model using age-defined waste samples obtained from a real landfill site. The new approach differs significantly from the conventional approaches. Although assumptions and prerequisites are required to apply the new approach to predict long term methane emissions from landfill sites, the new approach can be an improved method to predict methane generation at landfill sites more precisely. The following conclusions were obtained from this study.

(1) Even after more than 20 years since the start of landfilling, the amount of waste (paper and other types of waste) causing long term methane emissions was found to remain large. According to the results of the landfilled waste composition, the percentage of paper was approximately 20%, and the percentage of other types of waste, which were not categorized by visual examination, was also approximately 20%. Thus, a considerable amount of biodegradable organic matter still remained at landfill sites even after more than 20 years, including kitchen waste and paper.

(2) More realistic degradation coefficients in the FOD model, especially under cold

climate, were determined by using the obtained age-defined waste samples and were comparable to those established by the IPCC.

(3) In W city, the contribution of methane emissions from old and new landfill sites to the total carbon dioxide emissions was relatively high, with a value of approximately 2.53 %, because a large amount biodegradable waste, such as kitchen waste and paper, was landfilled. Thus, methane emissions from the landfill sites must be controlled.

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