LANDFILL GAS EMISSION MEASUREMENTS USING A MASS-BALANCE METHOD

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ABSTRACT: Methane and carbon dioxide emissions were measured at about 20 Dutch landfills in the years 1993 and 1994 using the mass-balance method. Measured emissions are compared with emissions, predicted using the first order decay model. The mass-balance proved to be well suited for this purpose and the results of the measurement confirmed the accepted landfill gas formation model. Ratio in methane and carbon dioxide emission suggest mean methane oxidation in the top-layer to be about 30%; this last conclusion is rather uncertain.

KEYWORDS: landfill, landfill gas emissions, methane emissions, emission measurement

1. INTRODUCTION

When organic wastes are deposited, the organic fraction is converted to landfill gas: a mixture of methane and carbon dioxide. Methane emissions from landfills are a major source of greenhouse gas emissions, and landfill gas extraction and utilisation is recognised as a cost-efficient method to reduce these emissions. Two reasons exist to improve current knowledge of landfill gas formation and emissions: (i) annex 1 countries under the Kyoto protocol have to carefully monitor and report emissions to UN-FCCC and this requires knowledge of methane formation, oxidation and emission. This is in particular the case for countries where significant reduction in emissions are expected (as in many EU-countries where waste policy aims at the mitigation of landfilling of organic materials); (ii) in the design and operation of projects for landfill gas extraction and utilisation reliable forecasts of expected amounts of landfill gas are of utmost importance.

Several models exist that describe methane formation in waste. Most of these models are assume first-order decay of organic material. In 1993 results from Dutch landfill gas extraction projects were used to validate landfill gas formation models and to determine the model-parameters (Coops et al., 1995). Conclusion was that first-order decay models, using the carbon contents in waste in table 1, a dissimilation factor of 58% and a half-time of biodegradation of 7 years described landfill gas formation on individual Dutch landfills about 25% accurate.

Table 1 – carbon content in Dutch waste, assumed in gas formation models

fraction	carbon content (kg tonne^{-1})
household waste	136
industrial waste	111
office, shop and services waste	140
sweeping waste	129
demolition waste	11
agricultural waste	135
sludges	90

Simultaneously emission measurements were performed in order to evaluate these findings. For this purpose methane and carbon dioxide emissions were measured on about 20 Dutch landfills in the years 1993 and 1994 and compared with emissions, predicted by model described above. The measurement results provide a relative large data-base of methane and carbon dioxide fluxes from landfills in a North-western European country shortly after 1990 (the reference year in the Kyoto Protocol) and just before waste composition drastically changed due to Dutch and European waste policy. This paper describes the measurement strategy and its results.

2. SELECTION OF MEASUREMENT METHOD

In literature various methodologies are described to measure methane emissions from landfills: (i) emissions can be calculated from concentration profiles in soil cores (Bogner and Scott, 1995); (ii) static and dynamic closed chambers can be used to sample a relative small part of the landfill surface (e.g., Bogner and Scott, 1995; Maurice and Lagerkvist, 1997; Perrera et al., 1999) (iii) mass-balance or micrometeorological methods give concentration profiles on top of the landfill from which emissions can be obtained form a larger part of the landfill (this study; Savanne et al., 1997) or (iv) away from the landfill plumes can be determined to obtain emissions from the entire landfill (Czepiel et al., 1996a; Scharff and Hensen, 1999).

Landfill gas emissions are highly variable. The heterogeneity of the material and the top-layer results in high spatial variability (e.g. Nozhevnikova et al., 1993; Mosher et al., 1996). Atmospheric influences (rainfall, wind, increases and drops in ambient pressure) result in variations of emissions in time (Czepiel et al., 1996a). When selecting a method to measure mean annual emissions from an

entire landfill, this spatial and temporal variation has to be taken into consideration. Besides it was considered extremely useful to measure both methane and carbon dioxide emissions in order to draw more accurate conclusions about landfill gas formation and methane oxidation. A last requirement was to measure emissions from a larger number of landfill sites, in order for conclusions to be statistically relevant. This also meant that the costs of a measurement at a single site should be acceptable.

At three sites the dynamic closed chamber and the mass-balance method were tested. The conclusions of the tests are given in table 2, and the dynamic closed chambers were considered not applicable, notably because the high spatial variations observed (requiring a lot of sample points thus rendering the method labour-intensive and costly) and because carbon dioxide emissions cannot be measured. The latter is because the chamber blocks solar irradiation and interferes with assimilation/dissimilation of the vegetation on top of the landfill, thus masking carbon dioxide emissions from the waste package.

Table 2 - comparison of mass-balance method and closed chambers for methane emission measurements

	advantages	Disadvantages
closed chambers	▪ easy interpretation ▪ useful to asses spatial and temporal variations in emissions	▪ small-sampling area ▪ unable to measure CO_2-emissions ▪ labour-intensive ▪ measurements normally performed only for shorter times
mass balance method	▪ large sampling area (> 2000 m2) ▪ labour-extensive ▪ measurement can be prolonged for longer times ▪ useful to assess mean emissions ▪ can be used for CO_2-emissions	▪ interpretation less straightforward

3. METHODOLOGY AND EQUIPMENT

Methane and carbon dioxide emissions are obtained in an interpretation of concentration and wind-velocity profiles. The method of interpretation often is referred to as the mass-balance method (Fowler and Duyzer, 1989) and methane emissions from the region upstream of the pole is obtained from:

$$J = \frac{\int_{z=0}^{z=l} u_z (c_z - c_l) dz}{x}$$

In which J (in g m^{-2} s^{-1}) is the methane flux through the landfill surface; u_z (in m s^{-1}) is the wind velocity at height z; c_z (in g m^{-3}) is the concentration at height z; l (in m) is the length of the pole; x (in m) is the fetch (the upstream length from the pole to the landfill slopes). In this calculation it is assumed that at the top of the pole background concentrations of CH_4 and CO_2 are measured.

Wind-velocity was measured at 0.65; 1.1; 1.5; 2.8 and 10 m using Lambsbrecht 1457552 cup-anemometers with a minimum wind-velocity of 1.0 m s^{-1}; wind direction was measured using a wind-master, Mark III. Precipitation (Lambsbrecht, type 15188), net irradiation (Campbell-Q6 net radiometer) and ambient temperatures (thermocouple) were registered as well. Air samples were taken at 0.65; 1.1; 1.5; 2.0; 2.8; 5.0; 7.0 and 10 meter, pumped through eight solenoid valves and fed to IR-analysing equipment. Carbon dioxide and methane concentrations were analysed using a Bruel & Kjaer 1302 multi-gas monitor. Methane was determined using the filter UA0987 with a detection limit of 0.1 ppm; carbon dioxide was measured using the filter UA0983 with a detection limit of 50 ppm; water was measured using the filter SB0527. Every sampling point was analysed for 7.5 minutes and every hour a concentration profile was obtained. The IR-output signals, the wind-direction and the

signals of the cup-anemometers were stored on disk. Sampling and analysis was completely automated and could operate autonomously for periods in excess of one week (see figure 1).

Figure 1 – Automated sampling and analysis of concentration profiles

4. MEASUREMENT LOCATIONS AND RESULTS

Measurements were performed in the years 1993 and 1994 on 21 landfills. Some of them were at that time still in exploitation; other ones were already closed and covered with soil (in most cases consisting of sand). Two landfills were capped with clay or a HDPE-liner. Table 3 gives an overview of the measurement locations and the methane and carbon dioxide fluxes obtained.

5. DISCUSSION

Applicability of the method

The method is well suited for measuring methane emissions. The difference in concentration between the lower sampling points and the atmospheric background concentration of about 2 ppm was generally larger than 10 ppm. Carbon dioxide emissions are somewhat less reliable due to the relative high background concentrations (390 ppm) compared to concentration differences over the pole. Besides, interpretation of carbon dioxide concentrations is complicated due to assimilation/dissimilation of the landfills vegetation. Figure 2 gives the variations methane and carbon dioxide fluxes throughout the day (N.B. results of several measuring days are compiled). In this figure the effect of vegetation on carbon dioxide emissions is clearly visible.

Table 3 - overview of measurement location and results

landfill	exploitation period	amount of waste (tonne)	area (ha)	gas extraction (m^3 hr^{-1})	top-layer	CH_4-flux	CO_2-flux	total emission (m^3 h^{-1})
in exploitation[1]								
1	1990 -	280 000	4	58	soil	5.03	6.73	470
2	1989 -	1 000 000	11	441	soil	3.60	5.52	1000
3	1988 -	630 000	5	-	soil	3.35	4.78	400
4	1987 -	166 000	2	-	soil	-	-	-
5	1985 -	7 000 000	20	-	soil	5.69	5.44	2300
6	1985 -	7 000 000	20	1080	soil	2.30	2.64	1000
7	1970 -	1 730 000	9	-	soil	15.52	14.61	2700
8	1969 -	7 880 000	26	-	soil	4.81	6.81	2800
closed								
9	1976-1988	477 000	9	100	soil	1.04	2.12	280
10	1987-1993	480 000	7	-	soil	2.12	2.11	300
11	1988-1993	1 760 000	27	150	soil	0.79	0.70	400
12	1986-1991	1 000 000	14	5	soil	n.d.	n.d.	n.d.
13	1987-1990	460 000	2	125	soil	3.07	3.38	130
14	1981-1989	390 000	8	105	soil	1.96	3.53	440
15	1985-1988	850 000	12	300	soil	0.35	0.49	100
16	1965-1988	450 000	6	55	soil	0.68	2.46	190
17	1974-1988	380 000	4	-	soil	0.22	0.59	30
18	1978-1984	150 000	11	20	soil	0.42	0.42	90
19	1972-1980	230 000	6	17	soil	1.25	1.26	150
capped								
20	1970-1986	2 320 000	30	580	clay	0.07	n.d.	21
21	1986-1991	830 000	8	170	HDPE	n.d.	n.d.	n.d.

1) at time of measurement, i.e. 1993/1994

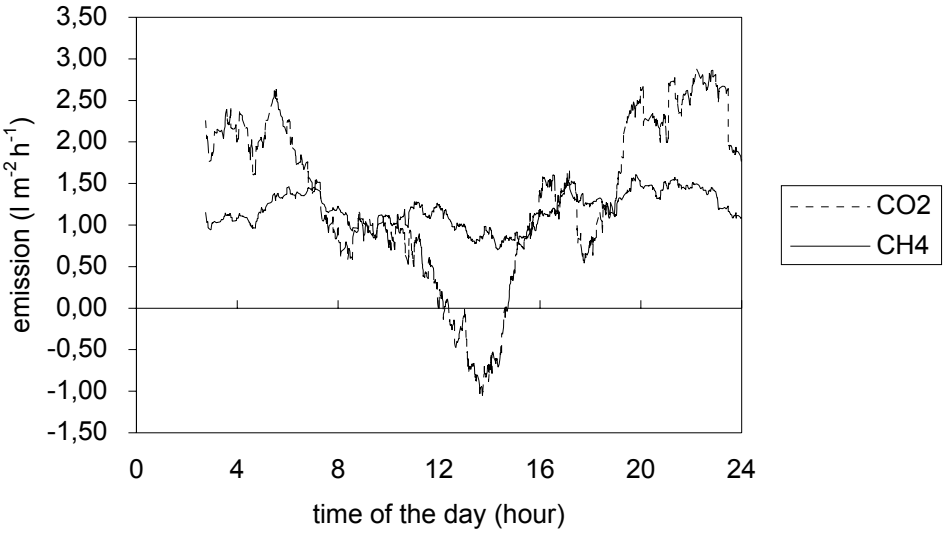

Figure 2 - daily fluctuations in methane and carbon dioxide emissions

There are some limitations to the measurement method. The applicability using this 10 m high pole is restricted to situations where the distance to the sides of the landfill is less than 150 m. When distances increase, the sample point in the top does not give a reliable indication of background concentrations anymore and larger poles have to be applied. Reliable measurements also require a minimum wind-speed of 1 m s^{-1}; measurement days with rainfall in excess of 25 mm day^{-1} are not taken into consideration.

Experience with the method indicated that about three weeks time is required to obtain consistent average emissions. When this prerequisite is fulfilled the accuracy of the method for methane emissions is estimated to be about 25%; the accuracy of carbon dioxide emissions is somewhat less.

Expected and measured emission – applicability of formation models

For each individual landfill, landfill gas formation can be calculated from the emissions through

landfill gas formation = (methane flux + carbon dioxide flux) * landfill surface + landfill gas extraction

In figure 3, the measured landfill gas formation is compared with the expected landfill gas formation, calculated from amounts of waste, waste age and composition, using the first-order decay model described by Coops et al. (1995). Observed formation is in fair agreement with expected formation and the results of the measurement confirm this model.

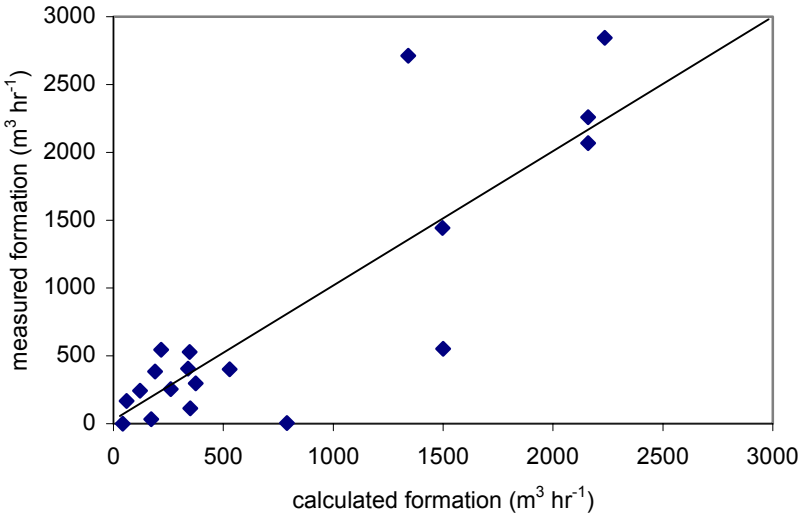

Figure 3: calculated vs. observed landfill gas formation at the 21 landfills

Extraction, oxidation and emissions

A number of landfills considered have a system for landfill gas extraction and utilisation. Comparison of landfill gas extraction and total emission gives an indication of recovery efficiencies of the landfill gas extraction systems. About half of the projects have efficiencies less than 30%. In these projects the amount of gas extracted is most likely determined by the capacity of landfill gas utilisation and no incentive exists to extract larger amounts of gas, although the technological possibility does exist. The other projects have efficiencies of 50% or higher, indicating that emissions can effectively be reduced by landfill gas extraction.

An indication of the extent of methane oxidation can be obtained by comparing the ratio of methane and carbon dioxide emitted and the ratio of methane and carbon dioxide extracted, which is about 57:43. Ratios of methane and carbon dioxide emitted are in many cases roundabout 60:40, suggesting about 30% oxidation, which is higher than the mean 10% oxidation proposed by IPCC and assumed in the Dutch national communication to UN-FCCC. However, taking into consideration the difficulties in assessing carbon dioxide emissions, the 30% oxidation is rather uncertain. Besides, all emission measurements were performed during spring, summer and autumn and no measurements are performed in winter: a period when methane emissions are known to be less (e.g. Czepiel et al., 1996b; Maurice and Lagerkvist, 1997), so it is not completely representative for mean annual oxidation. All

attempts to correlate observed oxidation with various parameters, such as fluxes through the top-soil, ambient temperature and others, yielded no result.

6. CONCLUSIONS

The mass-balance method is applicable to measure emissions from landfills. The method has advantages over closed chambers and possibly also some other methods: it has a large temporal and spatial resolution, can be automated and is relatively cheap and besides methane emissions, carbon dioxide emissions can be measured as well, although less reliable.

Using the mass-balance method emissions from 21 Dutch landfills are measured in the period 1993-1994. Observed emissions and the landfill gas formation from the emissions are in fair agreement with expected formation on basis of landfill gas projects. The results of the measurement confirm the landfill gas formation model by Coops et al. (1995).

Extraction efficiencies at about half of the landfill gas projects is in excess of 50%, indicating the technological feasibility to reduce significant part of methane emissions through landfill gas extraction.

Ratios of methane and carbon dioxide emitted suggest about 30% oxidation, which is higher than the mean 10% oxidation used in national emission inventories. However this 30% has to be considered rather uncertain.

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