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# Evaluation of Landfill Surface Emissions

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This study was started with the aim of evaluating different sampling techniques in order to develop a suitable and scientifically sound methodology for measuring the emissions of LFG from landfill surfaces. For this purpose, the work involved a bibliographical and theoretical study aiming to design new sampling equipment, as well as an experimental phase consisting in repeated measurement campaigns for the evaluation and comparison of such equipment on a sample site, i.e. a MSW landfill located in Northern Italy. Two sampling methods were designed and realized: a "flux chamber" and a "static box". The results of the first sampling campaigns show a good agreement between the two sampling techniques, but further studies will be needed in order to validate those results and study the influence of the meteorological conditions on the measured LFG fluxes.

## 1. Introduction

Landfills are known to be an important source of environmental pollution. Landfill gas (LFG) represents the major source of atmospheric pollution related to municipal solid waste (MSW) landfills (US EPA, 2008). LFG is the gaseous product of the anaerobic microbial decomposition of the organic matter of the waste, and it is composed for over 99% by CH<sub>4</sub> (typically in the range of 40-70%) and CO<sub>2</sub> (30-60%) (EI Fadel et al., 1997), thus being classified as a greenhouse gas. Indeed, LFG is estimated to account for approximately 3-19% of annual anthropogenic emissions of CH<sub>4</sub>, although there remains significant uncertainty associated with these estimates (Park and Shin, 2001). CH<sub>4</sub> and CO<sub>2</sub> are odourless, however, the presence of trace compounds (typically below 1%) having low odour thresholds, such as H<sub>2</sub>S, organic sulphur compounds (Kim et al., 2004) and VOCs (Davoli et al., 2003) gives the LFG a characteristic, highly concentrated and unpleasant odour. As a matter of fact, in many cases, offensive odours represent the major cause of populations' worries and complaints against landfills, and are therefore often the limiting factor to their exercise, or to the design and realization of new plants.

For these reasons, it is extremely important to be able to quantify LFG emissions into the atmosphere, in order to evaluate their environmental impact on the territory, and to prevent citizens from the exposure to odours or to potentially harmful pollutants by applying suitable control strategies (Palmiotto et al., 2014). A critical aspect associated with LFG emissions into the atmosphere is that they are variable, depending on different parameters, such as mainly atmospheric pressure, terrain humidity, but also temperature, wind speed and precipitations. These phenomenon is well known to field experts, but there are still few studies on that, and none of them up to now defines a clear cause-effect correlation (Czepiel et al., 2003; Rachor et al., 2013).

The first step towards the evaluation of landfills environmental impact is the quantification of the LFG emitted. Two ways are available for this purpose: either by using mathematical models or by conducting specific sampling campaigns on site.

Different models exist which allow to estimate the LFG produced by the landfill body as a function of the amount of yearly landfilled waste, its biodegradability, and rainfall. The LFG emitted may then be calculated as the difference between the LFG produced and the LFG extracted. The main drawback of such models is that they are generally very sensitive to the input parameters, thus making them unsuitable for environmental impact assessment purposes.

For this reason, it is preferable to base such studies on periodical site-specific sampling campaigns, which are more congruent with the real emissions. Unfortunately, despite the existence of guidelines and scientific studies indicating different methods for measuring the landfill gas emitted from a landfill surface (Mosher et al., 1999; Park and Shin, 2001; Sironi et al., 2005; Rachor et al., 2013), such methods are disagreeing and give not reproducible results.

The aim of this study was to evaluate different sampling techniques in order to develop a suitable and scientifically sound methodology for measuring the emissions of LFG from landfill surfaces. For this purpose, the work was organized in two phases. The first phase included a bibliographical and theoretical study aiming to design new sampling equipment; whereas the second, experimental phase consisted in repeated measurement campaigns for the evaluation and comparison of such equipment on a sample site.

## 2. Materials and methods

## 2.1 The studied site

The site where the experimental campaigns were conducted is a MSW landfill located in Northern Italy, active since 1993, and with a surface of about  $250'000 \text{ m}^2$ . The landfill is divided in 6 parcels: 5 are exhausted and currently in post-management, while the cultivation of the 6<sup>th</sup> parcel started in 2006.

## 2.2 Preliminary study of the landfill historical emission data

The studied landfill is very active from the point of view of environmental control: as far as diffuse emissions of LFG are concerned, these are measured regularly every 6 months by Emendo S.r.l.

The technique adopted by Emendo S.r.l. is the one described in the "Guidance on monitoring landfill gas surface emission" by the UK EA. This method is based on the use of a so called "flux box", which, despite its name, is a closed, non-fluxed (i.e. static) box at whose interior it is possible to measure an increase of the CH<sub>4</sub> concentration over time until a stationary condition is reached. From this trend it is then possible to calculate the CH<sub>4</sub> flux in mg/m<sup>2</sup>/s. Measurements shall be repeated on a sufficient number of points, which is determined by the following equation:

 $n = 6 + 0.15\sqrt{S}$ 

Where *n* is the number of points and *S* the monitored surface  $(m^2)$ . The emission of the whole landfill is calculated as the average of the flux data measured on the different sampling points, multiplied by the landfill surface. Hotspots have to be considered, as well. The advantage of this method is that it is simple and repeatable, although the fact that the sampling box used is closed may create some overpressures, thereby altering the CH<sub>4</sub> measurement (Rachor et al., 2013).

The flux box used in this case is in polypropylene, it has a volume of 0.1104  $m^3$  and a base surface of 0.01104  $m^2$ . CH<sub>4</sub> concentration was measured by means of a portable FID (GasTech by Crowcon).

As indicated by the English guideline, the landfill was divided in areas with similar characteristics, from which the LFG emissions should be comparable. The area considered for this study was named DEA-1 (Diffuse Emission Area) and includes all the areas of the landfill having a final cover, giving a surface of 102'277 m<sup>2</sup>, and 54 sampling points. Based on the analysis of the historical data measured by Emendo S.r.l. it was possible to identify some particularly significant emission points of the studied landfill.

### 2.3 Application of LandGem model

Before starting the experimental campaigns on the landfill surface, it was decided to make a preliminary evaluation by applying a model for the evaluation of the CH<sub>4</sub> generation. The model used was the LandGem by US EPA, which can be downloaded free from their website (www.epa.gov) as an Excel file. The model is based on a first order decay model:

$$Q_{CH4} = 1.3L_0 R(e^{-kc} - e^{-kt})$$

Where  $Q_{CH4}$  is the methane generation rate at time *t* (m<sup>3</sup>/y),  $L_0$  the methane generation potential (m<sup>3</sup>/Mg), *R* the annual refuse acceptance during active life (Mg/y), *k* the methane generation constant (1/y), *c* the time since landfill closure (y), and *t* the time since initial refuse placement (y).

Year	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003
Refuse acceptance (Mg/y)	272886	332873	374407	479973	430213	374061	340583	318513	273783	274977	250028
Year	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
Refuse acceptance (Mg/y)	240814	240814	101044	101044	290103	193552	151624	173000	82274	153915	0

Table 1: Annual refuse acceptance relevant to the studied landfill

Based on the site-specific data,  $L_0$  and k were set equal to 100 m<sup>3</sup>/Mg and 0.03 1/y, respectively. These values were then varied in order to analyse the model sensitivity to the input parameters. The annual refuse acceptance is reported in Table 1.

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#### 2.4 Field tests with Static Box by Polimi

The design of our "static box" was inspired by a study by Rachor et al. (2013), who used a specific kind of static chamber altered with respect to traditional chambers by addition of a 3-m open tube in order to prevent pressure differences with the atmosphere. The chamber by Rachor et al. was an aluminium cylinder with a height of 50 cm and a base area of  $0.12 \text{ m}^2$ . Because of the low CH<sub>4</sub> flux expected based on literature studies (< 1 L/h/m<sup>2</sup>) and the CH<sub>4</sub> diffusion coefficient in air, which makes the CH<sub>4</sub> concentration likely to be not uniform along the height of the chamber, it was decided to reduce our static box height to 10 cm. Moreover, in order to have a sufficient volume to measure the CH<sub>4</sub> concentration with the portable FID GasTech by Crowcon, which sucks about 0.85 L/min, and to cover a representative portion of the landfill surface, our "static chamber" was designed as a parallelepiped with a quadratic base (50 cm x 50 cm), thus having a total volume of 25 L. The chamber was realized in steel and it was equipped with a 3-m open Teflon<sup>TM</sup> tube (Figure 1, left).

Once the chamber is positioned on the landfill surface, the  $CH_4$  is measured inside the chamber at regular time intervals (about 3 mins): The  $CH_4$  flux relevant to the sampled point is then calculated as follows:

$$Q_{CH4} = \frac{V_{sb}}{S_{sb}} \left(\frac{dc}{dt}\right)$$

Where  $Q_{CH4}$  is the CH<sub>4</sub> flux (mg/m<sup>2</sup>/s),  $V_{sb}$  the volume of the static box (m<sup>3</sup>),  $S_{sb}$  the base surface of the static box (m<sup>2</sup>) and *dc/dt* the CH<sub>4</sub> concentration variation over time (mg/m<sup>3</sup>/s). *dc/dt* can be calculated as: *dc*  $n \cdot \sum t \cdot c - (\sum t) \cdot (\sum c)$ 

$$\frac{dt}{dt} = \frac{n \cdot \Sigma t^2 - (\Sigma t)^2}{n \cdot \Sigma t^2 - (\Sigma t)^2}$$

Where *n* is the number of measurements, *t* the times of the measurements (s) and *c* the measured CH<sub>4</sub> concentrations (mg/m<sup>3</sup>).



Figure 1: Detail of the "static box" with the Teflon<sup>TM</sup> open tube to prevent pressure differences with the atmosphere (left), and experimental apparatus for "flux chamber" sampling (right)

#### 2.5 Field tests with Flux Chamber by Polimi

The second sampling equipment that was tested is a "flux chamber", which was designed based on the flux chamber sampling system described in the US EPA guideline "Measurement of gaseous emission rates from landfill surfaces using an emission isolation flux chamber" (1986). This chamber was designed with the logic of being fluxed with neutral air. It consists of a semi-sphere in plexiglass, having a 50-cm diameter and a volume of about 30 L. The top of the semi-sphere is provided with two Teflon<sup>TM</sup> tubes: one is connected to an air bottle for the neutral air inlet, whereas the other one is connected to the FID when performing the measurement. The air from the air bottle passes through a flow-meter before entering the flux chamber, which allows to regulate the inlet neutral air flow. The inlet neutral air flow was varied in a rather wide range, comprised between 40 L/h and 300 L/h, in order to verify how it affects the measured CH<sub>4</sub> concentration (Figure 1, right). Based on the theory, the product between measured CH<sub>4</sub> concentration and inlet air flow should be constant.

Indeed, the mass balance on the flux chamber gives:

$$F_{in} + F_{CH4} = F_{out}$$

Which can be also expressed also as:

 $Q_{air} \cdot 0 + Q_{LFG} \cdot C_{CH4} = Q_{out} \cdot C_{CH4,out}$ 

Where  $Q_{air}$  is the neutral air inlet flow coming from the bottle and regulated by the flow-meter,  $Q_{LFG}$  is the LFG flow from the landfill surface, and  $Q_{out}$  is the flux chamber outlet flow, which can be considered equal to the inlet air flow, given that  $Q_{LFG}$  is much lower (by about two orders of magnitude). In this case, a CH<sub>4</sub> concentration ( $C_{CH4}$ ) equal to 50% was considered.

Once the flux chamber is positioned on the landfill surface, it is necessary to wait a time at least equal to one chamber exchange time ( $\tau = V_{fc}/Q_{air}$ ). After this time, the FID is connected to the flux chamber outlet tube and the CH<sub>4</sub> concentration is measured. The advantage of this sampling method is that the measured CH<sub>4</sub> concentration is constant, being a function of the inlet air flow and the LFG flow from the landfill surface. The drawback is that it requires longer sampling times (from 10 to 50 mins depending on the inlet air flow) and the usage of neutral air bottles.

## 3. Results and discussion

#### 3.1 Results of the LandGem application

The result obtained by the application of the LandGem model, by considering the actual refuse acceptance (Table 1) and a CH<sub>4</sub> concentration in LFG of 50%, and by setting the input parameters  $L_0$  and k equal to 100 m<sup>3</sup>/Mg and 0.03 1/y, respectively, gives a LFG generation in 2014 of 28530 Mg/y (Figure 2), corresponding to 2608 m<sup>3</sup>/h.



Figure 2: Results of the application of the LandGem model (in Mg/y) setting  $L_0$  and k equal to 100 m<sup>3</sup>/Mg and 0.03 1/y, respectively.

In order to evaluate the model sensitivity towards the input parameters  $L_0$  and k, these were varied between 80 m<sup>3</sup>/Mg and 120 m<sup>3</sup>/Mg, and 0.02 1/y and 0.04 1/y, respectively, which are all reasonable values for the studied landfill (Table 2).

k (1/y)	L₀ (m³/Mg)	LFG generation rate (10 <sup>6</sup> m <sup>3</sup> /y)	LFG generation rate (m <sup>3</sup> /h)	CH₄ generation rate (Mg/y)
0.02	80	13.69	1562	4565
0.02	100	17.11	1953	5706
0.02	120	20.53	2343	6848
0.03	80	18.27	2086	6096
0.03	100	22.84	2608	7620
0.03	120	27.41	3129	9144
0.04	80	21.76	2484	7260
0.04	100	27.21	3106	9075
0.04	120	32.65	3727	10890

Table 2: Results of the LandGem model obtained by varying the input parameters L<sub>0</sub> and k

It is possible to observe that, despite the input parameters  $L_0$  and k were varied within a reasonable range if considering the studied landfill site-specific data relevant to the rainfall of the area and the organic content of the accepted waste, which affect the values of k and  $L_0$ , respectively, significantly different results were obtained. Indeed, the LFG generation rate estimated by the model varies from a minimum of 1562 m<sup>3</sup>/h to a maximum 3727 m<sup>3</sup>/h.

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Such variations may be acceptable if the model is used as an instrument for previsional estimations for the design of the LFG collection system, but it may lead to unacceptable errors in the case of environmental impact assessment purposes. Indeed, the quantity of LFG emitted into the atmosphere can be calculated as the difference of the LFG generated and the LFG collected. In the case of the studied landfill, where the LFG is burned in co-generation motors for the production of electricity, the amount of collected LFG is easily obtained from the motors datasheets, and it is equal to 2200 m<sup>3</sup>/h. According to the values reported in Table 2, this means that, for the studied landfill, the collection efficiency would range from a minimum of 59% to a maximum of 140%, thus giving that the amount of LFG emitted is between +1500 m<sup>3</sup>/h and -700 m<sup>3</sup>/h, with an average emission rate of 400 m<sup>3</sup>/h.

#### 3.2 Field tests with flux chamber and static box

Table 3 shows the results of the measurements conducted on the studied landfill by means of our "flux chamber". It is possible to observe that, within the same day, the LFG emission rates measured at different inlet air flow rates are comparable to each other, as expected by theory. It is also possible to observe that there are significant differences in the LFG fluxes measured in different days: for instance, the 21 May extremely low CH<sub>4</sub> concentration were measured (always <10 ppm) with respect to the other days. From a first analysis of the meteorological data, the LFG fluxes seem to be related to the trend of the atmospheric pressure of the 48 h prior to sampling, giving that low LFG fluxes are measured when the atmospheric pressure is higher than the average pressure relevant to the studied area, but more measurements will have to be carried out in order to allow to have sufficient data to study such correlations.

Date	Inlet air flow (L/h)	С <sub>СН4</sub> (ppm)	Q <sub>CH4</sub> (m³/m²/s)	Q <sub>LFG</sub> (L/m²/h)	Q <sub>LFG</sub> (Mg/y)
05/03/2014	100	280	3.96E-08	2.85E-01	342.23
	250	110	3.89E-08	2.80E-01	336.12
	300	130	5.52E-08	3.97E-01	476.68
	500	60	4.24E-08	3.06E-01	366.68
	60	110	9.34E-09	6.72E-02	80.67
18/04/2014	150	70	70 1.49E-08 1.07E-		128.34
	300	40	1.70E-08	1.22E-01	146.67
	60	19	1.61E-09	1.16E-02	13.93
	100	20	2.83E-09	2.04E-02	24.45
24/04/2014	150	25	5.31E-09	3.82E-02	45.83
	200	33	9.34E-09	6.72E-02	80.67
	400	23	1.30E-08	9.37E-02	112.45
12/05/2014	30	26	1.10E-09	7.95E-03	9.53
	80	20	2.26E-09	1.63E-02	19.56
	300	14	5.94E-09	4.28E-02	51.33
19/05/2014	80	10	1.13E-09	8.15E-03	9.78
	100	7	9.90E-10	7.13E-03	8.56
21/05/2014	60	7	5.94E-10	4.28E-03	5.13
	80	5	5.66E-10	4.07E-03	4.89
	100	2	2.83E-10	2.04E-03	2.44
	200	1	2.83E-10	2.04E-03	2.44
10/06/2014	60	27	2.29E-09	1.65E-02	19.8
	80	21	2.38E-09	1.71E-02	20.53
	100	19	2.69E-09	1.94E-02	23.22
	200	11	3.11E-09	2.24E-02	26.89

Table 3: Results of the measurements with "flux chamber"

Table 4 compares the LFG specific fluxes  $(L/m^2/h)$  measured with the two different sampling methods tested: "flux chamber" vs. "static box". Of course, these are only preliminary results that will have to be verified and confirmed by the execution of other sampling campaigns, but it seems that there is a good agreement between the two sampling methods.

Date	Q <sub>LFG</sub> "flux chamber" (L/m²/h)	Q <sub>LFG</sub> "static box" (L/m²/h)
05/03/2014	3,17E-01	2,80E-01
18/04/2014	9,88E-02	7,95E-01
24/04/2014	4,62E-02	_
12/05/2014	2,23E-02	
19/05/2014	7,64E-03	_
21/05/2014	3,11E-03	_
10/06/2014	1,88E-02	3,51E-02

Table 4: Comparison of LFG specific fluxes measured by means of "flux chamber" and "static box"

#### 4. Conclusions

A "flux chamber" and a "static box" were designed and developed based on literature studies and other considerations deriving from our Laboratory's experience in the field of environmental sampling for measuring LFG fluxes from landfill surfaces.

The preliminary results of the measurement campaigns show a good agreement between those two measurement methods, but more data is needed in order to verify these results.

Future studies should focus on the correlation between the measured LFG fluxes and the meteorological conditions, such as atmospheric pressure that seems to have a direct influence on the LFG emission rate. This study also proves that models for the estimation of LFG generation are unsuitable for environmental impact assessment purposes, because they may lead to unacceptable errors due to their sensitivity to the input parameters.

#### References

- Czepiel P.M., Shorter J.H., Mosher B., Allwine E., McManus J.B., Harriss R.C., Kolb C.E., Lamb B.K., 2003, The influence of atmospheric pressure on landfill methane emissions, Waste Manage. 23, 593-598.
- Davoli E., Gangai M.L., Morselli L., Tonelli D., 2003, Characterisation of odorants emissions from landfills by SPME and GC/MS. Chemosphere 51, 357-368.
- El-Fadel M., Findikakis A.N., Leckie J.O., 1997, Environmental impacts of solid waste landfilling, J. Environ. Manage., 50, 1-25.
- Kim K.-H., Choia Y.J., Jeona E.C., Sunwoo Y., 2004, Characterization of malodorous sulfur compounds in landfill gas, Atmos. Environ. 39, 1103-1112.
- Mosher B.W., Czepiel P.M., Harriss R.C., Shorter J.H., Kolb C.E., McManus J.B., Allwine E., Lamb B.K., 1999, Methane emissions at nine landfill sites in the Northeastern United States, Environ. Sci. Technol. 33, 2088-2094.
- Palmiotto M., Fattore E., Paiano V., Celeste G., Colombo A., Davoli E., 2014, Influence of a municipal solid waste landfill in the surrounding environment: Toxicological risk and odor nuisance effects, Environment International 68, 16-24.
- Park J.-W., Shin H.-C., 2001, Surface emissions of landfill gas from solid waste landfill, Atmos. Environ. 35, 3445-3451.
- Rachor I.M., Gebert J., Gröngröft A., Pfeiffer E.-M., 2013, Variability of methane emissions from an old landfill over different time-scales, Eur. J. of Soil Sci. 64, 16-26.
- Sironi S., Capelli L., Céntola P., Del Rosso R., Il Grande M., 2005, Odour Emission Factors for the Assessment and Prediction of Italian MSW Landfills Odour Impact, Atmos. Environ. 39, 5387-5394.
- US EPA (US Environmental Protection Agency), 2008, AP 42, Fifth Edition, Compilation of Air Pollutant Emission Factors, Volume 1, Chapter 2.4 (draft): Municipal Solid Waste Landfills <a href="http://www.epa.gov/ttn/chief/ap42/ch02/draft/d02s04.pdf">http://www.epa.gov/ttn/chief/ap42/ch02/draft/d02s04.pdf</a>> accessed 31.01.2014