TESTING A SIMPLE AND LOW COST METHANE EMISSION MEASUREMENT METHOD

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SUMMARY: From 2007 onwards the E-PRTR regulation will be in effect. According to the E-PRTR guidelines methane emissions occurring during landfill activities should either be measured in situ, calculated by methane emission models or estimated by field experts. Models have shown large deviation in methane emissions and can therefore not be solely depended on. Quantification of methane emission by measurement is currently considered too expensive. Development of simple and cost effective measurement methods could enhance accuracy of estimations. The paper discusses the development of a simple and cost effective methane emission methodology performed on two European landfills. Furthermore the results obtained and costs associated with the new and simple method are evaluated and compared to dynamic plume methods.

1. INTRODUCTION

According to the E-PRTR Regulation (EC) 166/2006 (CEC, 2006), facilities undertaking an activity listed in Annex I of the IPPC Directive and exceeding the specific threshold values laid down in Annex I of the E-PRTR Regulation have to report their emissions to air and water. Emissions can be determined either by measurement, calculation or estimation. Landfills receiving more than 10 tonnes per day or with a total capacity exceeding 25,000 tonnes, have to report both emissions to water and emissions to air. According to the E-PRTR guidelines methane emissions occurring during landfill activities should either be measured in situ, calculated by methane emission models or estimated by field experts.

Recent studies have shown the huge differences in methane emission estimates of different models (Scharff et al., 2006; Ogor, 2005). The highest estimates obtained with the models can be five to seven times higher than the lowest estimates. This huge variation in results cannot be considered acceptable for E-PRTR reporting purposes. Current models are highly uncertain, mutually incomparable and inconsistent. The minimum accuracy that might be considered necessary for comparison between different models and countries is not met. Harmonisation of models may not necessarily solve the uncertainty.

Measurement of annual methane emissions at landfills with existing methods is considered too expensive. Moreover, single measurements should be considered as a ‘snapshot’ due to spatial (Nozhevnikova et al 1993) and temporal variation (Boeckx et al., 1996; Czepiel et al., 1996). This illustrates the need for development of simple and low cost annual methane emission

quantification methods. Such methods should enhance our understanding of methane emissions at landfills, improve model parameters over time and consequently enlarge comparability, consistency, accuracy and certainty of emission data within E-PRTR databases.

A simple and low cost version of the static plume method (SPM) (Jacobs, 2006) is in development by ECN and Afvalzorg. This straightforward approach was tested during a measurement campaign at Nauerna landfill, the Netherlands and two campaigns at Fakse landfill, Denmark. The measured methane emissions of these campaigns with the simple and low cost method were compared with those obtained by dynamic plume measurements (DPM).

2. MATERIALS AND METHOD

2.1. Quantifying methane emissions

The static and dynamic plume measurements are evaluated using meteorological data in combination with an atmospheric dispersion model (Scharff et al., 2003) . The model relates the measured concentration levels to the actual emission from the site. However if a reference release system is in place, use of both meteorological data and the dispersion model can be avoided using the following approach. For the methane and the reference gas emission (e.g. N_2O) the following can be said:

$$\text{Concentration } [CH_4] = \text{Dispersion} * Q[CH_4] \quad (1)$$

$$\text{Concentration [Reference]} = \text{Dispersion} * Q[\text{Reference}] \quad (2)$$

$$\text{So: } Q\left[CH_4\right] = Q[\text{Reference}] * \frac{\left[CH_4\right]}{[\text{Reference}]} \quad (3)$$

The data obtained during the methane emission measurement campaigns in this case study was compared to the Afvalzorg multi phase methane production and emission model (Scharff et al., 2006; Biocover, 2005) and to the results obtained by dynamic plume measurements (Scheutz et al. 2006).

2.2. Nauerna landfill

In January 2006 a single measurement campaign was performed at the Nauerna landfill situated west of Amsterdam. Nauerna has a total surface area of 76 hectares and is in operation since 1985. In total over 9 million tons of waste is landfilled at Nauerna. Waste composition can be considered predominantly inorganic.

The methane plume downwind of the landfill was measured on a transect perpendicular to the wind direction at distances of 400-1000 m downwind of the site. Along this transect 10 vacumated gas bottles were placed for a 6 hour sampling period (Figure 1). One gas bottles were located upwind of the landfill to obtain background levels. The gas bottles were 2,5 or 10 litres in size and all filled up to 0.5 bar absolute pressure using capillaries. The flow rates through the capillaries were evaluated in the lab. On the landfill a release system for a reference (tracer) emission was used to emit N_2O at an average rate of 2.2 g/s during the experiment. In order to evaluate the performance of the low cost gas bottle sampling technique, concentration measurements were performed on the transect along the gas bottle positions using a quantum cascade laser spectrometer (QCL, Aerodyne Research Inc.) mounted in a van. The setup of this

mobile QCL is very similar to the mobile tunable diode laser spectrometer used before at the Nauerna site (Hensen et al., 2000). The difference is an improved stability and a higher accuracy with the new laser spectrometer. Concentration measurements with a resolution of 3 ppb for CH_4 and 2 ppb for N_2O were obtained at 10 Hz. GPS data was obtained using a Garmin GPS76. During the measurements both wind speed and direction were measured and data was transmitted (1Hz) to the mobile van using a radio modem. Finally, after 6 hours of sampling each gas bottle was closed and CH_4 and N_2O concentrations in the gas bottles were analysed by the QCL system immediately after sampling.

Figure 1: Measurement setup at Nauerna landfill during January 2006 campaign and an example of the first type of gas bottle used for emission measurements.

2.3. Fakse landfill

In October 2006 and February 2007 two measurement campaigns were performed at Fakse Landfill. Fakse is situated in Fakse Municipality in south-eastern Sjæland, Denmark. The landfill has been in operation since November 1981 and comprises a total area of 12.1 ha. In total 740.000 tons of waste is landfilled at Fakse.

New types of gas bottles were manufactured before the Fakse campaign started. The new type gas bottles are standard steel refillable LP gas bottles (Primus cylinder no. 2012) and cheaper than analytical standard bottles. Cost effects are not presented in detail in this paper. An indication is given in the conclusions. Prior to the measurement all gas bottles were completely vacumated by a vacuum pump (Series Laboport N840 FT1.8, KNF Lab) (Figure 1). The gas bottles are 4.8 litres in size and all filled up to approximately 0.5 bar absolute pressure using capillaries (Alltech, internal diameter 0.13 mm). The flow rates through the capillaries were evaluated in the lab.

The methane plume downwind of the landfill was measured on a transect perpendicular to the

wind direction at distances of 100-500 m. downwind of the site. Along a transect 14 to 15 evacuated gas bottles were placed for a 4 hour sampling period (see figure 2). One gas bottle was located upwind of the landfill to obtain background CH_4 and N_2O levels. On the landfill 4 release systems for a reference emission were used to emit N_2O at an average rate of 2.7 and 2.5 g/s respectively during the two measurement campaigns. In order to evaluate the performance of the gas bottle sampling technique, concentration measurements were performed on the transect along the gas bottle positions using a mobile FTIR (Bomen MB100) mounted in a van. The FTIR is capable of measuring CH_4, CO_2, CO and N_2O in ppb range, due to its pathway of 56 m. (Galle, 2001). Finally, after 4 hours of sampling gas bottles were closed and CH_4 and N_2O concentrations in the gas bottles were analysed by the QCL system in the Netherlands between two to ten days after sampling.

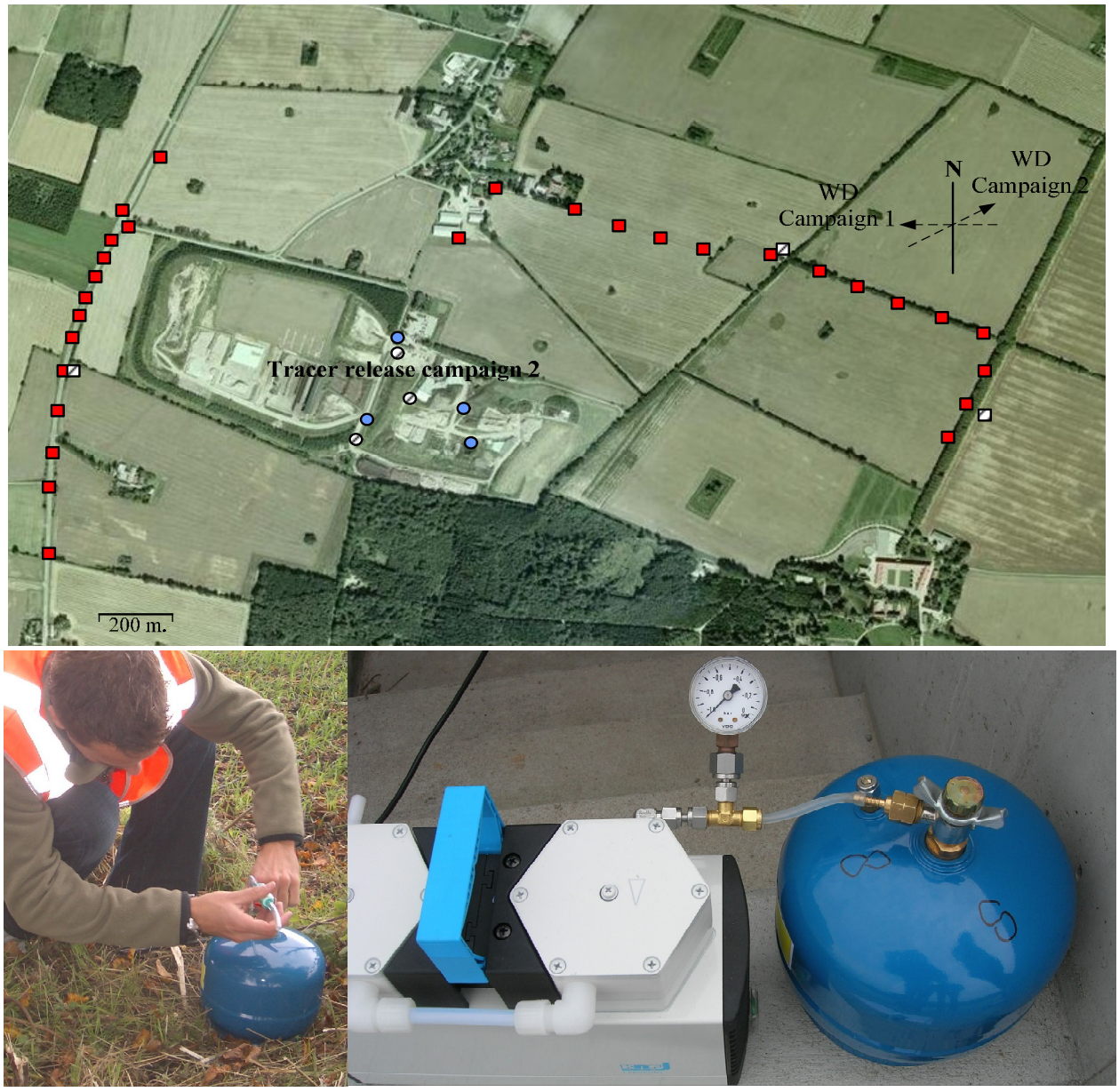

Figure 1: Measurement setup at Fakse landfill during October 2006 (campaign 1) and February 2007 (campaign 2).

3. RESULTS AND DISCUSSION

3.1. Nauerna

Figure 3 shows the CH_4 and N_2O concentration patterns, corrected for background conditions, obtained with the low cost method at Nauerna landfill. Both CH_4 and N_2O concentrations in the gas bottles were analysed three times with the QCL. The N_2O pattern originated from a point source on the landfill and therefore shows a narrower peak as compared to CH_4. The methane pattern is the result of the diffuse CH_4 source and various point sources within the landfill area. Dynamic plume measurements were performed 13 times throughout the day and provided similar methane and N_2O patterns.

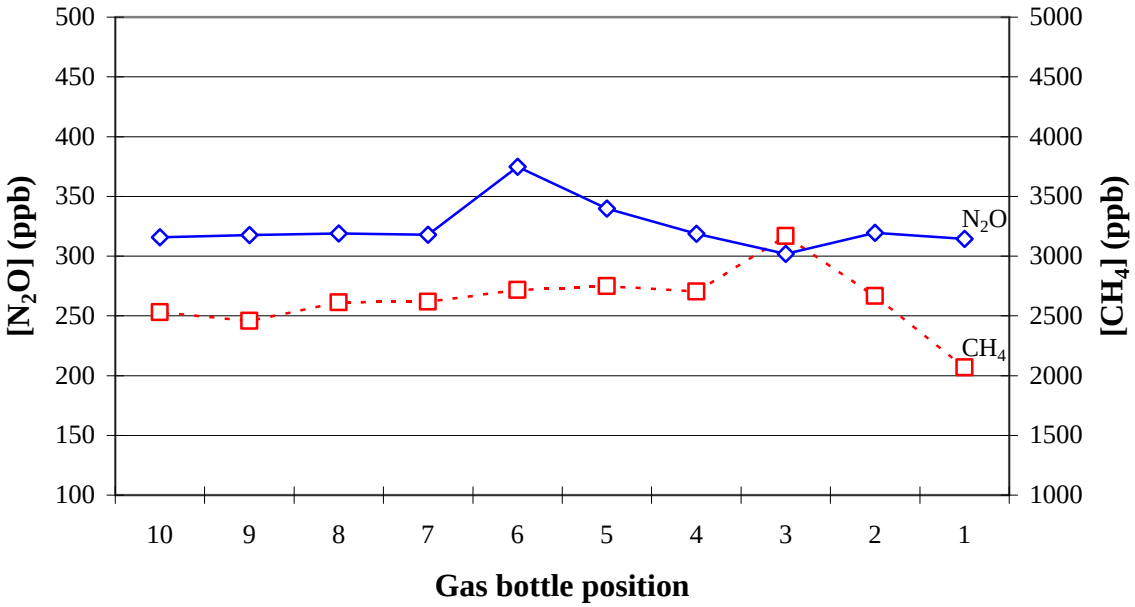

Figure 3: Obtained CH_4 and N_2O concentrations during measurement campaign at Nauerna landfill in each gas bottle.

3.2. Fakse

Figure 4 shows CH_4 and N_2O concentration patterns, corrected for background levels, obtained with the low cost method at Fakse landfill. Both the CH_4 and N_2O plumes are similar in shape.

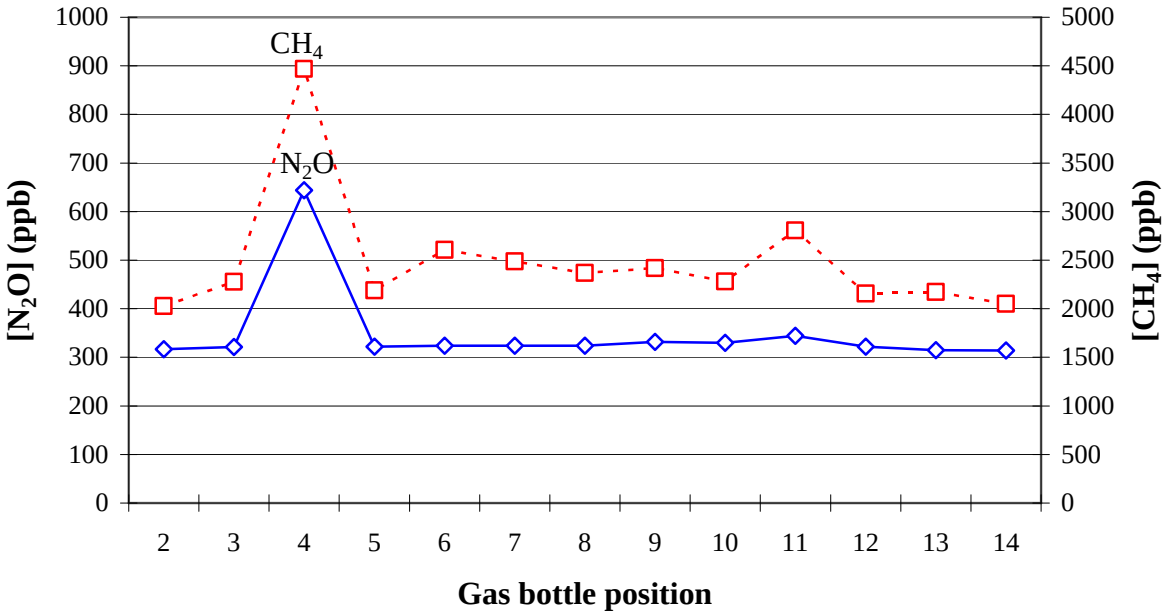

Figure 4: Obtained CH_4 and N_2O concentrations during first measurement campaign at Fakse landfill in each gas bottle.

Both plumes show a remarkable high peak at position 4. The reason for these high concentrations is not clear. A source at the landfill site (e.g. composting facility) can not explain this kind of sharp peak in the concentration pattern. Only a contamination in the gas bottle or a source at short distance from the road can produce such a high concentration that is not found in the adjacent gas bottles. The FTIR did not record similar concentrations at this position. For the emission evaluation the gas bottle at position 4 is considered an outlier and was therefore disregarded. During the second measurement campaign at Fakse wind direction shifted just before the measurements commenced. It was decided to anticipate on this wind shift and gas bottles were replaced. This resulted in differences in distance in relation to the landfill between the gas bottles. As dilution of methane and N_2O depends strongly on path length, individual gas bottle results have to be corrected for difference in path length. Calculations showed equal results. Either the direct or the perpendicular distance can be used here. Figure 5 shows the CH_4 and N_2O concentrations, corrected for background levels, obtained with the low cost method at Fakse landfill during the second campaign.

Again both CH_4 and N_2O plumes show a remarkable high peak, this time at position 5. The reason for these high concentrations is not clear. Extra dispersion modelling has been carried out in order to determine the width of the tracer plume. The hypothesis is that if the width of the tracer plume is very wide adjacent gas bottles should be affected in their CH_4/N_2O ratio. If latter is indeed true also released methane should disperse into a wide plume again affecting the ratio. Modelling results showed a very wide plume pointing out that the high concentration peaks at gas bottle position 5 can not be explained by landfill processes. For the emission evaluation the gas bottle at position 5 is considered an outlier and was therefore disregarded. It was the same gas bottle that produced the high peak in the first campaign. There seems to be a problem with this gas bottle. As it is a plumber's gas bottle it may have contained pure propane. Propane residues could have interfered with the methane measurement. This does however not explain the high N_2O concentration.

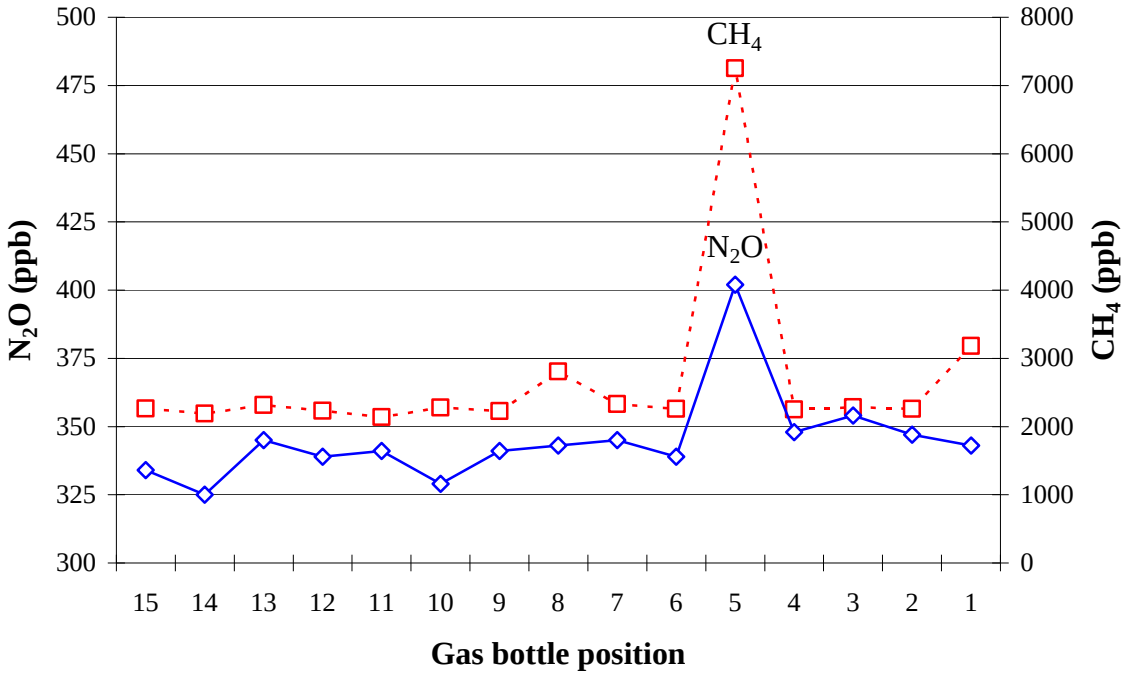

Figure 5: Obtained CH_4 and N_2O concentrations during second measurement campaign at Fakse landfill in each gas bottle.

3.3. Discussion

The results of all measurement campaigns are summarized in Table 1. Methane emissions obtained with different methods compare rather well. Remarkable is the agreement between methane emissions obtained by the QCL dynamic plume and the low cost method.

The Fakse campaign in October shows the importance of local landfill knowledge. During the campaign it be became apparent that an unknown source on the western part of Fakse landfill contributed significantly to the total methane emission. Up to that point is was unknown that the hill in the west was build up by domestic waste. The problem then arises, whether the tracer releases were positioned in such a manner that they could represent the emission situation at that moment in time. As the gas bottle method strongly depends on CH_4/N_2O ratios, one can imagine that suboptimal tracer positioning also strongly affects the obtained estimate. During this first campaign also a CO tracer study was performed. This enabled the FTIR-DPM method to determine a more representative whole site methane emission. The CO tracer could not be used for the gas bottle method and therefore results differ at first glance (Table 1). In addition to this so called best estimate obtained by the DPM, an ‘erroneous’ estimate was calculated. This estimate assumes that all methane was emitted from the eastern side of the landfill where the N_2O tracer was released. This is how during this first campaign the initial result of the gas bottle method was determined. Although not representative for the actual emission, both ‘erroneous’ estimates may therefore be compared. Comparison of both estimates indicates similar emissions obtained with both methods, but remain ‘erroneous’.

Being aware of the local landfill situation, optimal tracer positioning was anticipated during the second campaign in February. Results show that both techniques now produce similar methane emissions like the Nauerna campaign.

Table 1: Overview of both simple and cost effective-, DPM methane emission measurements and model estimations performed at Nauerna and Fakse landfill.

Method	Nauerna methane emission (g/s)	Fakse methane emission 10/06 (g/s)	Fakse methane emission 02/07 (g/s)	Meteo	Remarks
Afvalzorg Multiphase Production Model	48 ± 9	11º	10º	0	Emission = production – extraction – 10% oxidation
Dynamic plumes with model & N_2O	48 ± 9	-	-	√	No correction for N_2O
Dynamic plumes corrected for N_2O plumes (DPM – QCL)	44 ± 8	-	-	√	Reference estimate for Nauerna measurement campaign
Dynamic plumes with model (DPM - FTIR)	-	12 ± 2 *29 ± 13* *	10 ± 2	√	Reference estimate for Fakse measurement campaign
Gas bottle plumes vs. reference plume	48 ± 8	21 ± 2 *28 ± 3*	11 ± 2	0	Dilution only

º Biocover, 2005

*Results given in *Italic* are erroneous estimates

4. CONCLUSIONS

4.1. Suitability

Methane emission estimates generated with the three methods overall show good agreement. The low cost method confirms its potential, but still requires further verification. Aspects like consistency, suitability and accuracy of this technique need to be explored more intensively preferably at different landfills in different geographical and climatological settings.

At first glance the required equipment and handling of the simple and low cost method seems simple indeed. At the moment the analysis of the gas bottles can only be performed by a QCL, TDL or FTIR equipped with a rather sophisticated vacuum pump and a small volume sampling cell with a long pathway. Only a few of these machines are running throughout the globe and they demand very specifically trained professionals to operate. Another complicating requirement is knowledge and expertise of landfill gas emission in general and the local situation of the landfill in particular. As the first Fakse campaign has shown simply positioning gas bottles out in the field, releasing tracer and determining ratio's will not provide representative emission data. Especially the location of tracer release in relation to gas bottle positions is very important. It directly influences CH_4/N_2O ratios within the different gas bottles and thereby the estimated whole site methane emission. Although very obvious one also must keep in mind that quantifying methane emissions is very dynamic and complex. During the Fakse campaign the FTIR-DPM was available. This gave direct feedback about the emission dynamics at that specific time. The simple and low cost method does not provide methane insights directly. Proper setup and sampling procedures for the low cost method are extremely important. At this point such procedures are not available yet.

The accuracy of the simple and low cost method could be enhanced if more gas bottles are

placed along the transects, especially along the transect where the reference plume is found. The necessity of more gas bottles will lead to higher costs for analysis. The measurement campaigns at Fakse showed that a very good tracer simulation of the current methane emission as well as full coverage of the plume by the gas bottles is crucial to obtain accurate results. Furthermore meteorological data like wind speed and -direction will lead to better understanding of the plume shape observed by the gas bottles. It will provide an important quality control for the methane emission estimate. Again meteo equipment pushes the annual costs for the simple and low cost method, however only to a minor extend.

4.2. Cost efficiency

A comparison of costs between the DPM-QCL and the low cost method is given in Table 2. Table 2 focuses solely on measurement techniques. Costs for modelling are not included in this comparison. Hensen (2007) states that at least 20 measurement campaigns should be performed to obtain methane emission estimates with less than 5% uncertainty. In comparison to emission models the accuracy obtained with this frequency is much larger. However performing measurement campaigns at these frequencies also increases the costs way above applying models. By performing 5 measurements per year it is possible to obtain an annual emission of the landfill with less than 12% uncertainty (Hensen, 2007). Three measurement campaigns provide an uncertainty of less than 15%. This is considered sufficient to enable 'tuning' of landfill gas production and emission modelling.

Table 2: "Guestimate" of total annual costs in case of 10 measurements per year and combined with an overview of investments in research institute and client perspectives.

"Guestimate"	DPM (QCL)	SPM + Meteorological data	SPM – Meteorological data
Investment research institute	€ 300,000	-	-
Investment client*	-	€ 11,000	°€ 8,000
Material consumption. campaign^{-1}	€ 500	€ 250	€ 250
Laboratory costs.campaign^{-1}**		€ 1,000	€ 1000
Client costs.campaign^{-1}°°	€ 7,000	€ 600	€ 500
Total costs client. campaign^{-1}	€ 7,500	€ 1,850	€ 1,750
Total annual costs in case of 5 campaigns.a^{-1}	€ 37,500	€ 9,250	€ 8,750
Total annual costs in case of 20 campaigns.a^{-1}	€ 150,000	€ 37,000	€ 35,000

* Cost estimate for 16 gas bottles and meteo station on site

**Depreciation for analysers, analysis and measurement set up costs

° Cost estimate for 16 gas bottles only

°° Depreciation for gas bottles and/or meteo station included

Clearly the guestimated costs of the DPM-QCL compared to the gas bottle method are 4 times higher even with meteo equipment, when conducting at least 5 measurements per year.

4.3. Improvement

Further improvement will be expected for the type of tracer used during a measurement. In this

study a vast amount of N_2O was released, which has a global warming potential (GWP) of 296. The possibilities of propane as a tracer will be explored. The advantages of the use of propane as a tracer are that it has a GWP of 10 and it enables the use of GC-FID analysers. A proper GC-FID set up calibrated for atmospheric measurements is more complex than the average GC. Also a solution must be found to transfer samples from the vacumated gas bottles to the GC. Still the GC-FID is rather simple compared to the set up of a QCL. Switching to a GC-FID should lead to a situation that concentrations of methane and propane can be obtained from any respectable laboratory. This hopefully also results in a further cost reduction. A disadvantage of propane is the low explosion level when released in air. In order to release propane certain conditions must be met and safety measures will have to be taken. Propane release tests have been carried out and indicate hese drawbacks can be overcome.

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