ASSESSMENT OF THE CONTRIBUTION OF LOW EMISSIVE AREAS TO THE AVERAGE GLOBAL FLUX OF BIOGAS DIFFUSING THROUGH LANDFILL COVERS IN WALLONIA

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SUMMARY: ISSeP, in partnership with other experts (Ineris, Ephesia Consult), constantly develops acquisition methods for field measurements and (geo-)statistical tools for the estimation and cartography of diffusive methane fluxes on MSW landfills in Wallonia. In order to assess the contribution of areas displaying low methane emission rates to the estimated average global flux, measurements were performed using a dynamic flux chamber connected to two non-destructive detectors having different detection limits: Ecoprobe5® (detection limit of 300 ppm according our laboratory experiments) and Inspectra® Laser (full range of methane detection). Results showed that accounting for values measured with the Inspectra® Laser that are below the detection limit of the Ecoprobe5® does not lead to a significant increase of the estimated average global flux, nor to a decrease of its estimation uncertainty. Additional results revealed that the impact of emissions around gas wells is much more important to take into account since they contribute to an increase of 60% of the average global flux.

1. INTRODUCTION

In 1998, Walloon environmental authorities set up an ambitious monitoring network of sanitary landfills. The network includes 12 municipal solid waste (MSW) and non-hazardous waste landfills located on the Walloon territory (southern part of Belgium). Each site undergoes periodic environmental investigations on potential receptors around the landfill, i.e. surface water and groundwater, ambient air and residents located close to the landfill. More than 40 global or targeted campaigns on both water and air have been conducted since the establishment of the network (see corresponding technical reports on http://environnement.wallonie.be/data/dechets/cet). Air quality monitoring over landfills is usually conducted with respect to 4 key aspects: control of engine/flare emissions, evaluation of odour annoyance, assessment of ambient air quality and quantification and cartography of landfill gas (LFG) fugitive emissions through the surface of the landfill. The work presented here focuses on the last part of the surveillance. Specifically, methodological advances recently gained by ISSeP following the acquisition of new measuring devices are presented.

Proceedings Sardinia 2013, Fourteenth International Waste Management and Landfill Symposium
S. Margherita di Pula, Cagliari, Italy; 30 September – 4 October 2013

2. CONTEXT

Assessment of risks and nuisances related to gaseous emissions through the surface of MSW landfills in which waste with a residual organic fraction was dumped is a common issue in environmental management and monitoring. Many methodologies exist to measure and quantify methane (or LFG) emissions diffusing through the landfill cover. The most commonly used methods are those using traditional static flux chambers (Christophersen *et al.*, 2001), dynamic flux chambers (Taramini *et al*., 2003, Tregoures *et al.*, 1999), tracer dilution (Fredenslund *et al*., 2010, Gale *et al*., 2001, Scharff *et al.*, 2009, Scheutz *et al.*, 2011) or micrometeorological methods (Lohila *et al*., 2007), the vertical plume mapping (Babilotte *et al.*, 2010) and the differential aborption LiDAR method (Babilotte *et al*., 2008, 2010). Kjelden and Scheutz (2011) performed a thorough comparison of all of these methods. They came to the conclusion that methods relying on measurements at the landfill surface (i.e. with flux chambers) or close to the landfill have a tendency to underestimate global landfill gas emissions due to the often heterogeneous nature of emission patterns. However, while more sophisticated methods such as the tracer dilution method require expensive equipments and highly skilled operators, the dynamic flux chamber methodology has the advantage of being simple, fast and with a reasonable cost. It also allows an accurate localization of emissive areas. Insofar flux measurements are performed in areas suspected to be more emissive (over slopes, landslides, altered vegetation zones, around gas wells...), the probability of missing more emissive zones is relatively low. This is probably the reason why this method is still so popular among operators responsible for monitoring surface emissions from landfills. ISSeP thus uses it for flux estimation of diffusive biogas emissions over Walloon landfills.

On-site flux measurements are conducted using an adapted apparatus consisting of a closed dynamic chamber system (Ineris Patent “Mesure de flux surfacique de gaz”, French patent F96 05996 and European Patent Application EP 0 807 822 B1) connected to a multichannel non-destructive detector based on the infrared (IR) technology (Ecoprobe5®, RS Dynamics). The current methodology has been applied for 4 years and has shown to be reliable for the estimation of methane, carbon dioxide and total petroleum fluxes over the landfill surface. Part of these methodological developments have already been presented at the Sardinia 2009 conference (Kheffi *et al*, 2009). However, the detection limit of the Ecoprobe5® for methane, experimentally established at about 300 ppm, was deemed problematic for flux estimation in areas displaying very low emission rate (mainly rehabilitated zones).The contribution of such low emissive areas to the the global flux was an open question. In order to solve this problem, ISSeP acquired a new apparatus, the Inspectra® Laser detector, fully selective to methane (no false measurements due to the presence of hydrocarbons or other pollutants) with an extended measurement range from 1 ppm to 100% gas volume. This paper presents preliminary results of the comparison between the Ecoprobe5® sensor and the Inspectra® Laser for methane fluxes detection and quantification over landfills. The impact of including lower fluxes measured with the Laser detector in low-emissive areas on the global surface-average flux is evaluated and discussed.

Figure 1. Mont-Saint-Guibert landfill map – location of facilities and delimitation of the investigated areas in 2012.

3. DATA ACQUISITION

3.1 Investigated landfill

The study was conducted at the Mont-Saint-Guibert (MSG) landfill, located about 30 km south of Brussels. It is authorized as a "Class 2" landfill: only municipal solid waste and non hazardous waste are allowed. Total landfill capacity is 5.3 million cubic meters. It opened in 1958 and is environmentally monitored by ISSeP since 1998. Figure 1 shows the map of the landfill and its facilities (gas engines, leachate treatment plant, equipment rooms) as well as the location of the working area (pouring waste area) when ISSeP performed its measurements on LFG emissions in 2012. The total area of the landfill is approximately 443 000 m² (red contour, Figure 1) but the area covered by the study (tumulus) is estimated at 364 100 m² (pink area, Figure 1). The pouring area (about 13 000 m²) is excluded from the analysis because the measurements could not be carried out in satisfactory conditions due to the heterogeneity of the soil surface (apparent waste of different nature, size of the waste higher than the measurement system…). Most of the sampling points are usually located on temporary or permanent covers (soil, compost, sand or even grass).

3.2 Apparatus and sampling procedure

The detection and the quantification of methane present in the LFG diffusing through the cover of the landfill is carried out using both the Ecoprobe5® detector and the Inspectra® Laser detector. Table 1 shows their main technical features.

For on-site flux measurements, those two non-destructive detectors are simultaneously connected to the closed dynamic chamber equipped with two separate external recirculation systems (Figure 2). The footprint of the chamber is 0.25 m² and its internal volume is 0.029 m³. The pressure is kept constant (atmospheric pressure) in the room thanks to the recirculation of the air once analyzed while a propeller ensures air homogeneisation in the closed chamber.

Table 1. Technical characteristics of Ecoprobe5® and Inspectra® Laser.

	Ecoprobe5®	Inspectra® Laser
Detection technology	Infrared (IR)	Laser
Destructive/non destructive	Non destructive	Non destructive
Concentration range CH_4	300 ppm – 500 000 ppm (50%v/v)	1 ppm – 1 000 000 (100% v/v)
Theoretical sensitivity	1 ppm	1 ppm
Acquisition rate	0.5 sec	0.1 sec
Pump flow	1 l/min	1.2 l/min
Other detectable parameters	CO_2, Total Petroleum, O_2	-
Other probes	PID, temperature	-

The sampling strategy broadly follows the procedure established by ISSeP in 2009 (D'Or, 2009), based on the UK-EA standards (UK-EA, 2010). Emissive and non emissive areas are defined at the landfill surface on the basis of previous results (2009 campaign). Flux measurements are only performed in emissive zones ([CH_4]>500 ppm). The number of sampling points in emissive areas is established according to the size of the area and their position is defined randomly using a mapping software. When emissions of LFG are detected, measurements are densified so as to define the limit of the emissive area as precisely as possible. Within non emissive areas, a transect (predefined path on the area with methane concentration measurements every 5 m) is conducted with the Inspectra® Laser detector connected to a bell probe sampling system in order to confirm the absence of emissions. All points are located by means of a topographic GPS. As already mentioned, the pouring area of waste is not investigated.

Figure 2. Closed dynamic chamber system connected to Ecoprobe5® and Inspectra® Laser.

4. RESULTS AND DISCUSSION

4.1 Laboratory tests

The Ecoprobe5® and Inspectra® Laser detectors were first compared on a series of laboratory tests with the aim of establishing their intrinsic sensitivity threshold to methane and evaluating the correlation between the responses of both devices (Bietlot, 2012). Synthetic gas (syngas) samples with methane concentrations ranging from 1 ppm to 50% (v/v) were directly connected to the infrared and laser detectors for concentration measurements. Figure 3 separately shows detector responses in the lower range of CH_4 concentrations (on the left) and in the range 500-100 000 ppm (on the right). These laboratory experiments led to the following conclusions:

- Concentrations measured with the Ecoprobe5® and the Inspectra® Laser detectors show a linear response with respect to syngas theoretical concentrations.
- Both devices slightly underestimate the theoretical concentrations of methane in the lower range of concentrations.
- The announced reliability of the Laser detector is verified: the measurements of very low concentrations of methane (in the order of a few ppm or tens of ppm) are accurate.
- Ecoprobe5® is not reliable for lower CH_4 content detection and confirms its higher detection limit (300 ppm). Its use seems optimal for measuring methane concentrations above 1 000 ppm, but with a tendency to overestimate concentrations in high methane level syngas samples.
- The tests aiming at establishing the repeatability of the measurements (two days after) with both detectors on a selection of syngas samples are entirely satisfactory.

4.2 Field measurements over the MSG landfill

Field measurements of methane fluxes were performed over the MSW landfill of Mont-Saint-Guibert (MSG) in September and October 2012 (D'Or, 2013) using the apparatus presented in Figure 2 and applying the procedure described before (D'Or, 2009, see 3.2). According to this, 6 investigation zones were identified (areas delimited by the blue dashed line on Figure 1). The rest of the landfill was considered as non-emissive, as confirmed by the absence of methane detection during the transect control with the laser detector (path of the transect is represented by a green line on Figure 1).

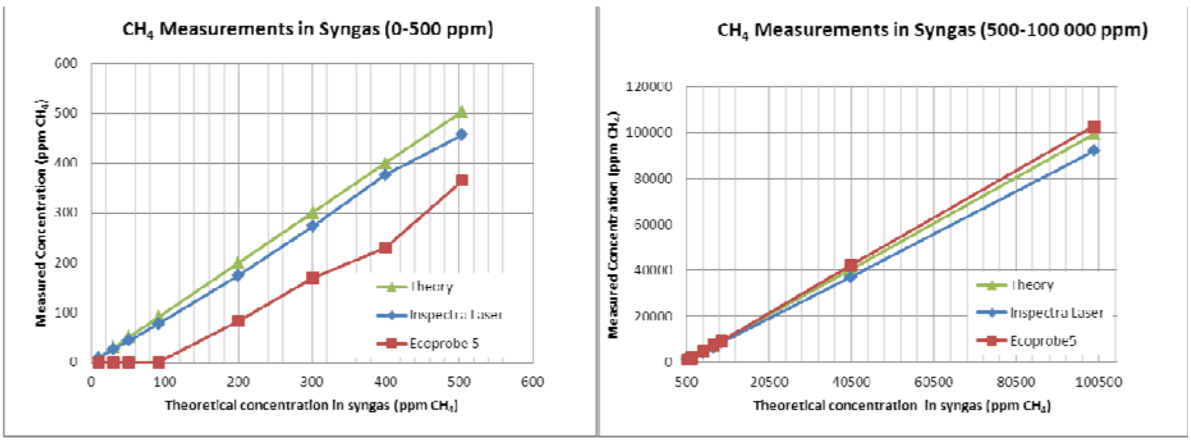

Figure 3. Response of Ecoprobe5® and Inspectra® Laser to 0-500 ppm and 500-100 000 ppm methane concentration ranges in syngas.

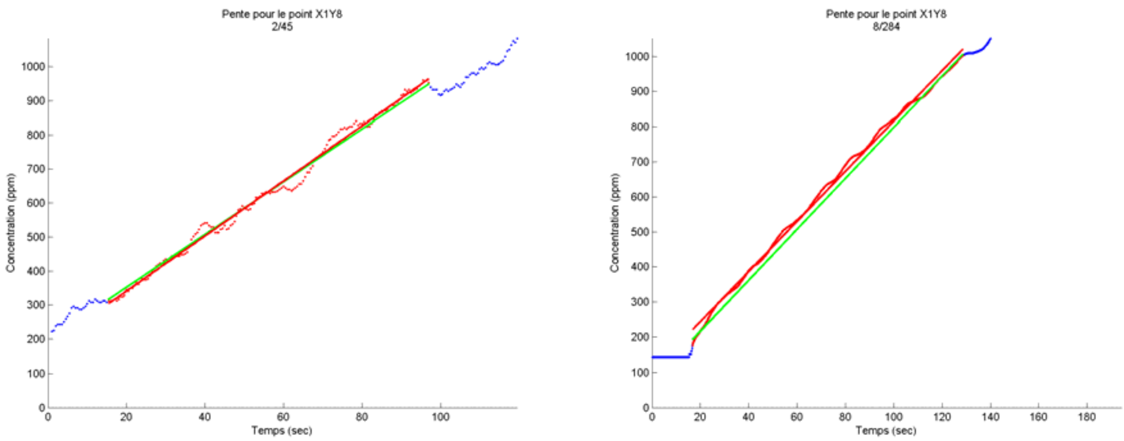

Figure 4. Time-series concentration curves and regression obtained with Ecoprobe5® (left picture) and Inspectra® Laser (right picture) at sampling point X1Y8.

During the campaign, a total of 283 measurements were obtained with the closed dynamic chamber equiped with both detectors. 244 sampling points were located in historically emissive zones and 39 measurements in the close vicinity of gas wells.

Surface gas fluxes are estimated based on the accumulation of LFG in the closed dynamic chamber using a semi-automatic method, as depicted in Figure 4. The temporal evolution of methane concentration in the chamber is monitored over a period of 2 minutes (blue line). The green line is a manually drawn regression curve and the red line is the estimated regression on a time interval choosen by the expert. The flux is computed from the slope of one of these lines, preferably that obtained by manual regression (red line). After this pre-treatment step, the following numbers of flux measurements are available:

- Ecoprobe5®: 283 flux measurements including 24 non-zero,
- Inspectra® Laser: 283 flux measurements including 158 non-zero.

4.3 Exploratory data analysis

4.3.1 Comparison of time-series of concentration measurements

Time-series of concentration measurments obtained with both devices were superimposed for comparison. The operation was carried for 45 points of CH_4 measurements available in both data sets. Some examples are shown in Figure 5. The corresponding estimated flux values (expressed in ml $CH_4/m^2/s$) are given in the legend. The last picture confirms that, at methane concentrations below 300 ppm, the Ecoprobe5® response can not be used for flux estimation while the Inspectra® Laser provides a response that can be representative of methane accumulation in the closed dynamic chamber.

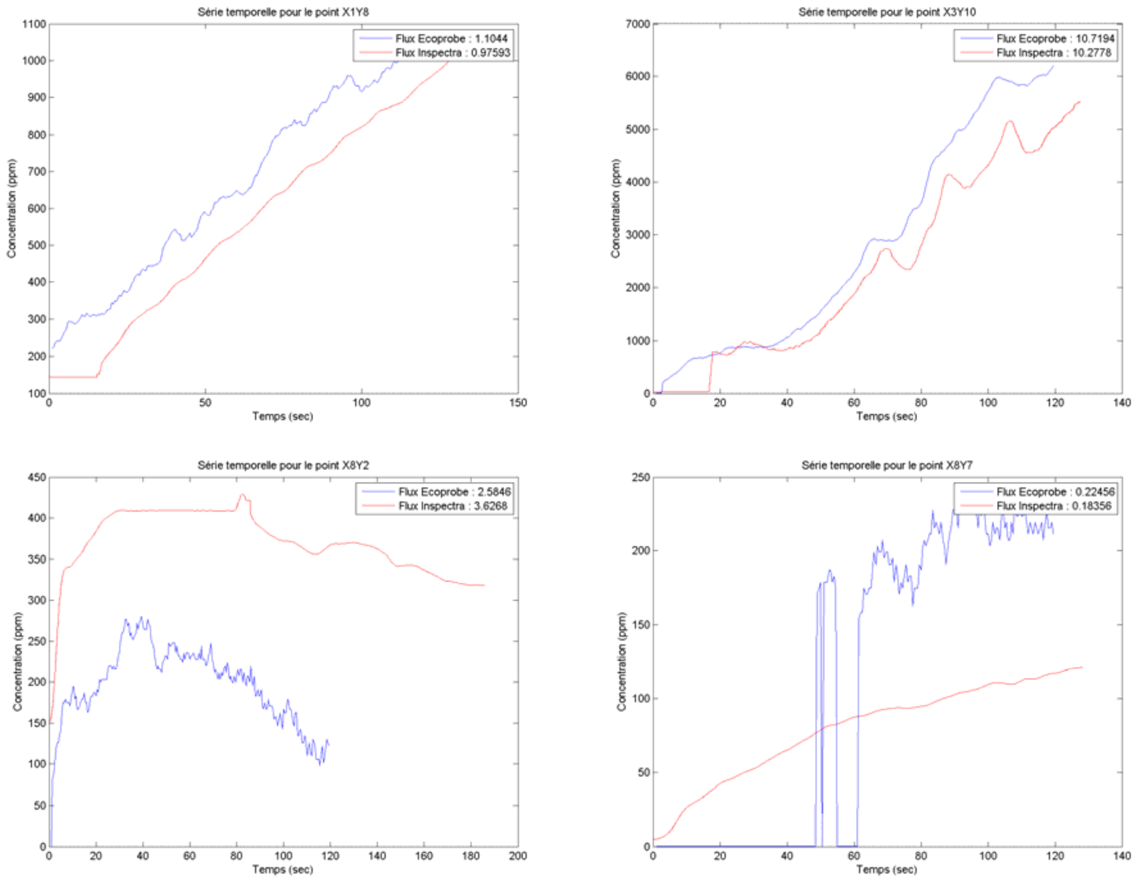

Figure 5. Comparison of time-series of CH_4 concentration data obtained with Ecoprobe5® and Inspectra® Laser at a selection of sampling points.

4.3.2 Comparison of estimated fluxes

Figure 6 shows the correlation between flux measurements performed with both detectors (left picture) over the full range of methane fluxes. The graph on the right focuses on values lower than 5 ml/m²/s. The correlation is very high, with a correlation coefficient equal to 0.9894, indicating a lack of bias between both methods. At the origin of the graph, a series of zero values measured with the Ecoprobe5® correspond to values higher than 0.5 ml/m²/s measured with the Inspectra® Laser. As a result of a higher sensitivity of the laser detector, so called “zero flux” measurements using the Ecoprobe5® sensor can correspond to flux values up to 1.5 ml/m²/s obtained with the Inspectra® Laser.

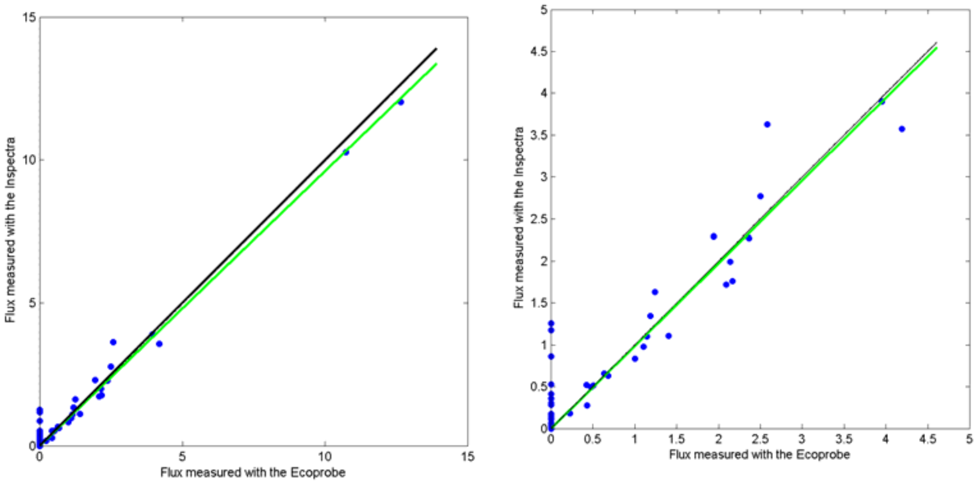

Figure 6. Relationships between CH_4 flux measurements with Ecoprobe5® and Inspectra® Laser over the whole range of values (left) and focusing on the low values only (right). The black line represents the bisector and the green line represents the regression of flux measurements with both detectors.

Statistics calculated on both data sets are shown in Table 2. They are calculated excluding the data collected in the neighborhood of the gas wells (i.e. using 244 out of 283 points). Average flux, standard deviation, percentiles and maximum values are very similar. The Inspectra® Laser has a lower proportion of zero values (35% *vs* 90%), confirming its ability to measure lower flux values.

4.4 Simulation maps and methane flux cartography

In order to estimate the global flux over the landfill, which corresponds to a surface-average gas flux that is globalized to the entire landfill (i.e. including non emissive areas), sequential Gaussian simulations (SGS, Goovaerts, 1997) are performed using the normal scores of the 244 flux measurements as well as a set of fictitious zero data added on a regular grid in the non-emissive areas. Hundred realizations were generated for each variable using the same variogram model for both variables, involving a nugget effect and two isotropic exponential structures with respective ranges equal to 65 and 2500 m and contributions equal to 0.3 and 1 respectively (Equation 1).

Table 2. Statistics on data flux (ml $CH_4/m^2/s$) measured by Ecoprobe5® and Inspectra® Laser.

	Ecoprobe5®	Inspectra® Laser
Number of measurements	244	244
Average	0.24	0.26
Standard deviation	1.18	1.14
Percentile 0.500	0.00	0.001
Percentile 0.750	0.00	0.02
Percentile 0.950	1.29	1.43
Percentile 0.975	2.42	2.48
Maximum	12.64	12.02

$$\gamma(h) = 0.41Nug(0) + 0.3Exp(65) + 1Exp(2500) \quad (1)$$

Figure 7 and Figure 8 show the average and the variance of the spatial distribution of methane fluxes at the landfill surface (i.e. the average and the variance over the 100 realizations). In this case, because of the presence of a very important nugget effect, the maps of the realizations are very pixelated, indicating a substantial variability over very short distances. The maps built using Ecoprobe5® measurements are presented in Figure 7 and those build with Inspectra® Laser measurements in Figure 8. The variance is an indicator of local uncertainty. As expected, the variance is more important in areas where high values are close to zero values, i.e. in the direct vicinity of areas identified as emissive. Estimated fluxes are generally very low. Several spots with slightly higher values are identified in each of the investigated areas. The largest is located in the southwest part of the landfill.

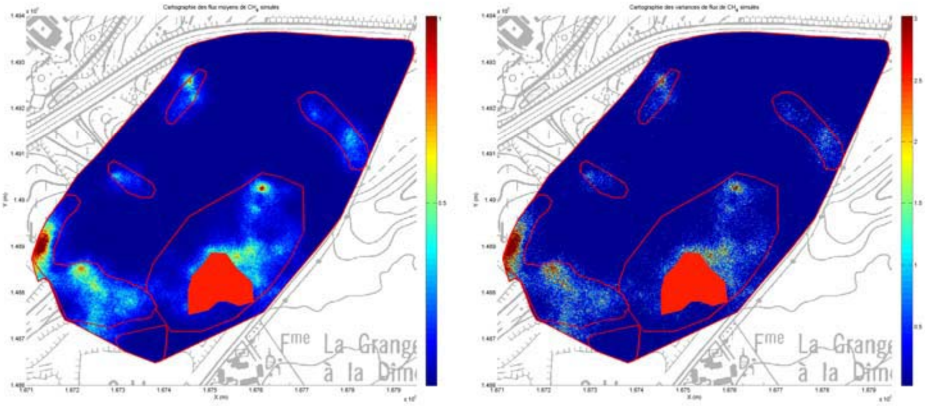

Figure 7. Cartography of methane fluxes measured with Ecoprobe5® and estimated by colocalized cosimulations. On the left, average value (flux in ml/m²/s) ; on the right, variance (variance in (ml/m²/s)²).

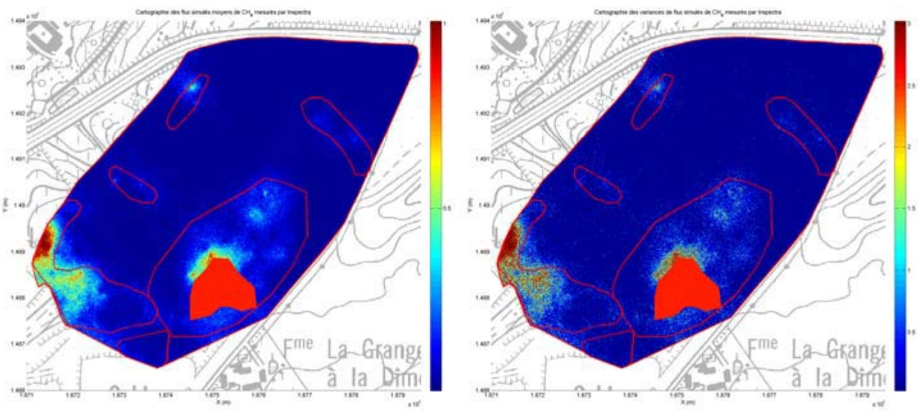

Figure 8. Cartography of methane flux measured with Inspectra® Laser. On the left, average value (flux in ml/m²/s) ; on the right, variance (variance in (ml/m²/s)²).

4.5 Comparison of estimated average global fluxes

Table 3 compares the statistics distribution of the average global flux of methane obtained by SGS using measurements collected with the Ecoprobe5® and the Inspectra® Laser. These results show that using the more sensitive laser detector rather than the Ecoprobe5® has a moderate impact on the estimation of the average global flux, both of them being in a very similar range of values (0.0595 ml/m²/s for Ecoprobe5® and 0.0578 ml/m²/s for Inspectra® Laser). The 95% confidence intervals defined by the 0.025 and 0.975 percentiles are also very similar. Therefore, taking into account the lower values measured by the laser detector instead of zero values measured by the IR detector does not lead to an increase of the average global flux, nor to a decrease of its estimation uncertainty.

Table 3. Statistics related to the average global flux of methane (ml/m²/s) measured with the Ecoprobe5® and Inspectra ® Laser detectors.

	Ecoprobe5®	Inspectra® Laser
verage global flux	0.0595	0.0578
Standard deviation	0.0086	0.0092
Percentile 0.025	0.0432	0.0422
Percentile 0.975	0.0774	0.0808
Maximum	0.0801	0.0850

The average values indicated in Table 3 were estimated by considering zero fluxes in the non-emitting areas (dark blue areas on Figure 7 and Figure 8). By replacing those fictitious null data by the minimum flux value ever recorded with the less sensitive IR detector during this survey, i.e. 0.2246 ml/m²/s, the average global CH_4 flux has been recomputed, yielding 0.0665 ml/m²/s. Compared to the average global flux given in Table 3 (0.0595 ml/m²/s), it corresponds to a 12% increase, which is actually in the same order of magnitude as the uncertainty related to the measurement of the average global flux estimated from Ecoprobe5® measurements. As the Inspectra® Laser confirmed that the values lower than the detection limit of the Ecorpobe5® can be neglected, using the detection limit values in a conservative attitude thus results in a 12% overestimation of the average global flux. At the Mont-Saint-Guibert landfill, it is thus recommended to use zero values to replace the Ecoprobe5® values lower than the detection limit.

4.6 Assessment of the contribution of gas wells to the total flux

As the results of the flux estimates obtained with the Inspectra® Laser and Ecoprobe5® did show any significant difference, ISSeP focused on the contribution to the total emission rate of the landfill of flux measurements performed at a representative selection of gas wells. To achieve this, 39 wells were investigated on a total of more than 200. In this section, flux measurements conducted close to gas wells were incorporated to previous results. Flux maps and statistics of estimated of average global flux were updated. Flux values measured close to gas wells were assigned to a surface covering a 3 meter radius area around the gas well. A value corresponding to the average of available measurements at investigated wells was assigned to non investigated wells. Table 4 compares the estimated average global flux calculated respectively including and excluding the gas well data set. The relative impact of the wells, expressed as the percentage increase between both average global fluxes, is calculated according to Equation 2.

$$I_{rel} = 100.\frac{\left(Flux_{including\_gas\_wells} - Flux_{excluding\_gas\_wells}\right)}{Flux_{excluding\_gas\_wells}} \quad (2)$$

Table 4. Comparison of average global flux estimations (ml/m²/s) without or with gas well contribution.

	Ecoprobe5®	Inspectra® Laser
Without gas wells contribution	0.0595	0.0578
With gas wells contribution	0.0970	0.1027
Relative Impact of gas wells (I_{rel}) (%)	63.0	77.7

This impact is important, being around 63% for methane fluxes measured with the IR detector and around 78% for fluxes measured with the laser detector. The impact of the fluxes around the wells is thus much more important than that linked to the sensitivity of the measurement device to low fluxes.

5. CONCLUSIONS

With the aim of improving LFG global flux estimations over Walloon MSW landfills, ISSeP conducted a campaign of measurements over the Mont-Saint-Guibert landfill using a closed dynamic flux chamber equipped with two non destructive detectors connected with separate external recirculation systems: Ecoprobe5® and Inspectra® Laser. These measurements were achieved to assess the contribution of areas displaying low methane emission rate to the average global flux of the landfill. The starting point of this study was the assumption that the use of an Ecoprobe5® detector, with a detection limit of about 300 ppm, led to an underestimation of the LFG fluxes in the less emissive areas (mainly rehabilitated ones). In order to verify this hypothesis, ISSeP conducted flux measurements simulatenously using Ecoprobe5® and Inspectra® Laser allowing methane measurements over an extended range of concentrations, i.e. from 1 ppm to 100% gas volume.

First, laboratory experiments confirmed the improved sensitivity at low methane concentration of the Inspectra® Laser with respect to the Ecoprobe5®. Above 1000 ppm, both devices performed equally well.

After field data collection and exploratory data analysis, simulation maps obtained for methane fluxes measured with both detectors were compared, as well estimations of average global flux. The results showed that using the more sensitive Inspectra® Laser rather than the Ecoprobe5® had a moderate impact on the estimation of the average global flux, both of them being in a very similar range of values (0.0595 ml/m²/s for Ecoprobe5® and 0.0578 ml/m²/s for Inspectra® Laser). Therefore, taking into account the low values measured by the laser detector instead of the zeros measured by the IR detector did not lead to an increase of the estimated average global flux, nor to a decrease of its estimation uncertainty.

ISSeP then focused on the contribution of flux measurements performed at a representative selection of gas wells to the global emission rate of the landfill. In this case, on the Mont-Saint-Guibert landfill, taking into account the local fluxes in such emissive zones led to a 60% increase of the estimated average global flux.

This state of thing led us to conclude that rather than focusing on the quantification of diffusive fluxes in low emissivity areas and their impact on the estimated average global flux, the areas of interest that must be investigated in priority are the annular zones around gas wells.

AKNOWLEDGEMENTS

The Authors wish to thank the Walloon Government for financial support and especially Mr. P. Nemry for giving ISSeP the opportunity to present the research and results. The Authors also thank Mr. R. Fafchamp for his valuable help in the laboratory tests.

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