

LANDFILL GAS (LFG) FUGITIVE EMISSIONS ON LANDFILL SURFACE - COMPARATIVE TEST OF ON SITE ANALYSIS DEVICES

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SUMMARY: ISSeP has been controlling for ten years the gaseous and liquid emissions of main landfills in Wallonia. The monitoring of landfill surface gas-emissions is usually performed with portable FID analysers. Such devices offer high sensitiveness but measuring ranges are sometimes too short for high methane concentration. It is why another portable device, based on IR-analyzer technology (ECOPROBE), has been tested and compared to FID in several case studies. Synthetic air/methane gas mixture has been used to verify the exactness of both portable analysers. In a second time, the linearity of IR-device has been verified in its large measuring range from high-rated LFGes. Finally, performances of FID and IR technologies have been compared on “on-field sampled” surface landfill gas (LFG). The tests show good proportionality and low detection limit of FID devices, but with low exactness and low upper saturation limit. The IR device shows good exactness and linearity on a wide measuring range, but his minimum detection value is higher than FID systems. This gap increases when analysing complex high-rated gas mixture as LFG. Despite this limitation, IR probe produces other simultaneous measurements such as carbon dioxide and oxygen concentrations. These data are complementary to methane values and useful for landfill surface emissions monitoring.

1. INTRODUCTION

Landfills take part to the worldwide warming effect with large emissions of greenhouse gas as CH₄ and CO₂, the main components of LFG (IPPC, 2001). Current landfill management has to take into account many impacts or harmful effect over local environment and to prevent them (Féd. Oublic Service, 2006). Modern landfill technologies, such as upper liner over completed landfill cells and LFG extraction system contribute to reduce drastically those greenhouse gas emissions. But the integrity of landfill capping remains the weakest element and LFG could escape through leakages (Kjeldsen, 1996). Surface monitoring of LFG emission allows leakage detection and help landfill managers with solving the problem, reducing the volume of lost LFG, and so, optimising LFG management system. As shown in the papers of famous scientific organisations, gas measurements on polluted soils or landfills are not only environmental or

economic matter, but can also be used to determine risks on human health.

Since 1998, the Walloon Scientific Institute (Institut Scientifique de Service Public) assesses the environmental impacts of main waste landfills in Wallonia (Godfroid and Dengis, 1999). Landfill control scheme investigates both water quality (landfill emissions, groundwater quality and surface water quality) and air quality (ambient air quality, combustion smokes from LFG valorisation/elimination systems, surface emissions and odour nuisance characterisations).

In landfill control scheme, surface monitoring operation is divided into two stages. In the first one, methane concentrations are measured with gas analyser all along the landfill cap. The analysed gas is directly sampled on the surface of the landfill cap with a sampling bell put on the ground. In the second stage, the results are georeferenced and processed by geostatistics softwares. Methane emissions can be estimated on each point of the whole site and isoconcentration maps are created (Awonoi & al, 2005; Bogne & al, 1997 and 2005)

Traditionally, methane concentrations are measured with flame ionisation detectors (FID), which gives semi-qualitative values in a concentration range from 0 to 10 000 ppm. This sensitive device is appropriate for measurements of surface emissions through landfill top-liner even at low intensities. Unfortunately, the upper saturation limit of the probe is too low. It implies that many values, higher than 10 000 ppm of methane, can not be measured.

Up to now no portable device is able to analyse methane concentrations both with high sensibility and high saturation rate. Available devices only offer one of these requirements, and the good balance is hardly reached. This is one of the main difficulties in measuring methane concentrations of landfill surface emissions (BRGM, 2000).

ISSeP had the opportunity to test another type of portable device, with embedded IR multichannel probe (IR-MP). It was mainly designed for on site measurements of VOC concentrations in vapours of contaminated soils. However, its announced technical characteristics would be, if verified, sufficient to measure the concentration of LFG components in landfill surface fugitive emissions. Potentially, IR-MP could be a very interesting complement to the FID device, and could even replace it, for the detection of methane emitting areas on landfills. In order to verify the potential of this new device, it is necessary to check its measuring sensibility and range have to compare its performance with alternative FID solution.

This paper presents the results of comparative tests performed by ISSeP on several portable analyser devices using different kind of LFG samples.

2. DEVICES AND METHODOLOGY

2.1 Portable analyser devices

2.1.1 FID devices

FID detectors measure hydrocarbons concentrations, including methane, in gaseous phase. The sampled gas is injected into a combustion chamber, settled inside the device, and burnt in an air-hydrogen flame. Burnt hydrocarbons produce high levels of ionisation, proportional to the hydrocarbons concentrations of the sampled gas.

ISSeP owns two successive versions of the SEWERIN PortaFID. The PortaFID M2 detector is fully analogical and is used since 1997. The PortaFID M3K detector is an improved version of M2, with analogical and digital LCD displays. Both of them are used for years in the surface monitoring of landfill control scheme. Theoretically, they both offer the same measuring range from 0 to 10 000 ppm. Linearity calibration is performed on two points by measuring first “free of methane” gas for “zero point” and then air-methane mixture calibrated at known concentration. The factory-set concentration is 10 ppm (measuring range) but it can be set on

other values: 100, 1 000 or 10 000 ppm.

2.1.2. IR-MP device

The ECOPROBE 5 is a portable IR-MP device built by RS DYNAMICS. It includes three autonomous analysers, with simultaneous outputs:

- The photo-ionisation detector (PID) measuring total concentration of volatile organic compounds (VOC). The PID analyser is equipped with an ultraviolet lamp of 10,6 eV and can detect many volatile organic compounds (ionisable below 10,6 eV) in a range from 0,1 to 3000 ppm, except methane.
- The infra-red analyzer (IR) measuring on separated channels the concentrations of CH₄, “total petroleum hydrocarbons” (TP) and CO₂. A fourth channel is used as reference to take into account the influence of dust, moisture or other disturbing factors in the correction of the other channels. Manufacturer working range of each channel is announced to cover gas from 50 ppm up to 50 % methane rate.
- A paramagnetic analyser, analysing the concentration of oxygen in a range from 0 to 100 %.

This multidetector-device is completed with atmospheric pressure sensor, temperature probe and GPS receiver. The tested material was a “trying specimen” without recent detection adjustment, and lent by the Belgian dealer.

2.2 Devices used for validation and comparison measurements

Two reference apparatuses were used for verifying on-site devices:

- HC51 M: This laboratory FID apparatus is designed for precise determination of low methane concentration in gas samples stored, for example, in TEDLAR bag. All other hydrocarbons are removed before analysing CH₄. Its measuring range is 50 ppb - 1000 ppm.
- GA 2000: This device is devoted to the LFG analysis and is used as gas monitoring in the landfill control scheme. His IR-analyser measures the concentrations of methane, carbon dioxide and oxygen. The used wavelengths are 3,41 µm for methane and 4,29 µm for carbon dioxide. Outputs are deteriorated when sampled gas contains others hydrocarbons.

2.3 Laboratory operations and measurements

All gas samples were prepared for the test series in the Institute’s air laboratory. They were obtained by progressive dilutions of various gas references. Nitrogen or ambient air is used as dilution gas with the massic diluter. All the gas samples have been diluted at 0,1 % accuracy.

Some gas samples have been analysed in laboratory by gas chromatography coupled to mass spectrometry. These analyses aim at determining VOC’s concentrations in tested samples. It also requires the suppression of all other gas compounds because of high accuracy of laboratory analyser devices. This is performed by an adsorption technique of VOC’s with adsorbent trap tubes, followed by desorption and final analyse.

2.4 Tests protocols

Specific tests protocols have been applied to three portable gas analysers: the ECOPROBE device, the PortaFID M2 and M3K detectors. The protocols consist in analysing three sample batches created with the aim of testing the apparatuses in varied measuring conditions.

2.4.1. Batch formed from 50% CH₄ standard gas

The first sample batch was formed from a synthetic air-methane gas mixture (standard 50%).

Manufacturer of this standard sample certifies no water and concentrations of others hydrocarbons lower than 1 %. This reference gas was successively diluted to obtain a series of samples covering a wide range of methane concentrations. The interest of testing devices on this first batch is to measure the exactness and accuracy of the analysing devices with an objective method, without any perturbing parameters. The disadvantage is precisely to be far from practical measuring conditions where methane is mixed with many other gases creating interferences.

2.4.2. Batch formed from high rated "natural" LFG

The second batch was obtained by diluting high-rated landfill LFG. The not-diluted gas was collected in TEDLAR bag at the entrance of the LFG treatment plants where the methane concentration reaches about 50 %. The reference concentration of VOC's, contained in the sampled gas, was measured with the HC51 M device. Outputs are expressed in isobutylene equivalents. These values were completed by laboratory GC-MS analysis. Again, growing dilution rates allowed obtaining a batch of samples with a varied range of LFG concentrations. This series of measurements aims at evaluating exactness and accuracy of analysing devices with high-rated complex gas mixtures. Comparing results on the two first batches helps to apprehend interferences of several gas compounds, as VOC's or water vapour, over the analysing outputs.

2.4.3. Batch of surface emitted LFG sampled on site

The last batch has been realised by on-site collecting samples in TEDLAR plastic bag on points emitting LFG at various intensity on the surface of a landfill. Sampling has been selected by a first FID measure in order to allow covering a concentration range as wide as possible (from 10 to more than 10 000 ppm). TEDLAR bags were filled with volumetric air pump (Personal air sampler GilAir 3) connected to the sampling bell that is used for FID surface measurements. The whole batch has been used to test the three portable analysers and the highest-rated sample were analysed with laboratory GC-MS to determine the VOC's concentration. The samples of this batch can be considered as low-rated LFG obtained by "natural dilution" during its transfer from source point in the waste mass to the surface of the landfill. The result allows comparing the efficiency of portable analysers in "real using conditions" of landfill surface emissions.

3. RÉSULTS

3.1 batch 1

Table 1 presents batch 1 results. The first column contains theoretical methane concentrations computed by weighting the original 50% proportion by the decreasing dilution rates successively applied. Only PortaFIDM3K was tested on batch 1. The GA 2000 is used as reference analysing device from 10 000 ppm of methane. HC51M reference tool were not used for measuring high dilution rates samples. Original concentration was certified at a very good accuracy and dilutions were achieved enough carefully to be confident on computed values.

Batch 1 results allow making the following observations:

- PortaFID M3K outputs are linear in the whole announced working range but with a bad exactness. Given CH₄ values are systematically half of theoretical concentrations.
- Announced detection limit of IR-MP should be 50 ppm whereas it is, in reality, 500 ppm for methane channel and 100 ppm for TP channel.
- Above these limit values, and up to 50% CH₄, both CH₄ and TP channels give linear response and good exactness. TP channel shows slightly better exactness than CH₄.

- PID analyser gives constant zero response, which is normal as gas sample of batch 1 are pure CH₄/air mix without any other hydrocarbons.
- On the contrary, CO₂ output is not zero, which is strange: samples should not contain CO₂.

Table 1 : output values of reference and portable analysing devices on batch 1 samples (standard mix air-CH₄ 50% diluted at decreasing rates)

Theoretical Values	Reference device GA 2000	Portable devices				
		FID M3K	IR multichannel probe			
CH ₄ (ppm)	CH ₄ (%)	éq CH ₄ (ppm)	PID (ppm)	CH ₄ (ppm)	TP (ppm)	CO ₂ (ppm)
50	< DL	17	0	0	0	0
250	< DL	140	0	0	275	35,2
500	< DL	240	0	0	452	45,3
1 250	< DL	670	0	1 261	1 305	36
2 500	< DL	1 400	0	2 378	2 521	37,8
5 000	< DL	3 200	0	4 482	4 802	49,8
50 000	53 000	> 11 000	0	42 123	43 361	28,5
500 000	500 000	> 11 000	0	469 823	483 958	0

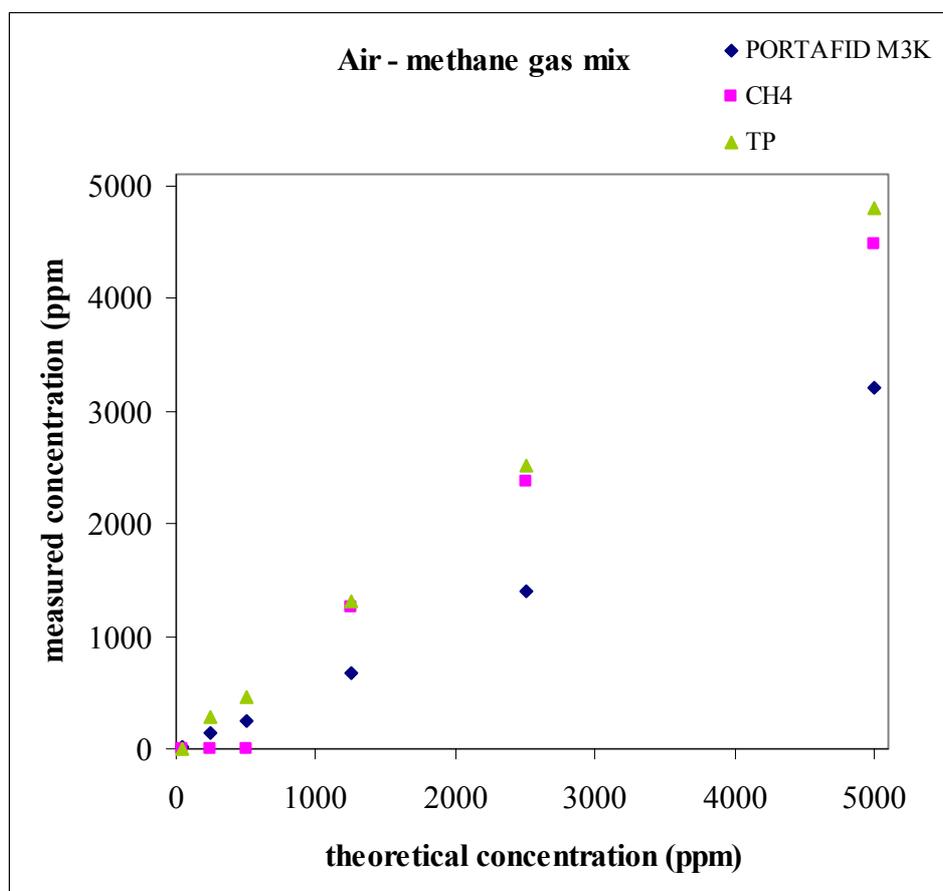


Figure 1. Evolution of the output values of portable analysing devices analysing air-methane gas mix versus theoretical methane concentration.

3.2 Batch 2

Test protocol used for on batch 2 is the same as for batch 1. Results are given on Table 2. The original reference gas is a high-rated landfill LFG. Theoretical methane concentrations are again calculated from the decreasing dilution rate applied to this reference. Opposite to batch 1, the original CH₄ concentration is not known. It has to be measured. In order to use the most precise device for this reference measure, one has measured the concentration of a diluted sample with HC51M device. From this result (bold in Table 2) and known dilution factors, it is possible to recover the concentration in original and less diluted samples. Reference devices used for batch 2 are HC51 M from 0 to 1 000 ppm of methane and GA 2000 from 10 000 ppm.

Table 2: output values of reference and portable analysing devices on batch 2 samples (high rated LFG diluted at decreasing rates)

Theoretical values		Reference devices			Portable devices					
		HC51 M		GA 2000	FID M2	FID M3K	ECOPROBE			
CH ₄	VOC's	CH ₄	VOC's	CH ₄	CH ₄ eq	CH ₄ eq	PID	CH ₄	TP	CO ₂
ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
22	0,05	25	0,1 ± 0,1	< DL	38	9	0	0	0	62
44	0,1	46	0,1 ± 0,1	< DL	72	18	0	0	0	69
88	0,2	62	0,1 ± 0,1	< DL	110	19	0	0	0	81
220	0,5	167	-	< DL	220	60	0	448	100	176
439	1	447	1,0 ± 1,0	< DL	480	210	0	735	388	371
878	2	878	2,0 ± 1,0	0	710	490	0	1 358	913	718
1 316	3	> 1 000	-	0	1 100	810	0	1 744	1 370	1 045
6 605	15	> 1 000	-	10 000	2 000	6 300	0	7 303	7 227	5 763
176 127	401	> 1 000	-	200 000	> 11 000	> 11 000	0	205 791	212 995	168 041
440 317	1003	> 1 000	-	520 000	> 11 000	> 11 000	0	485 681	520 163	401 769

The results show that:

- The PortaFID M2 is accurate from 0 to 1000 ppm of methane only. Saturation begins just above this value, making the use of this device hazardous.
- For highly diluted sample (up to 2000 ppm CH₄), PortaFID M3k has similar behaviour as on batch 1: it gives linear signal but 2 time lower that real concentration. Exactness is better for less diluted samples (from 2000 to 10000 ppm).
- The ECOPROBE gives accurate values and linear response for a wide range of samples. Measurements are even better than with GA2000 for very concentrated samples. However, detection limits is again higher than announced: there is no response below 200 ppm CH₄ for both methane and TP channels.
- For methane concentration lower than 10000 ppm, TP channel gives better results (more accurate measurement) than CH₄ channel. In this low concentration range, output values are strangely higher on CH₄ channel than on TP channel. It is a strange behaviour as methane is a part of hydrocarbons, TP channel output values should be always higher than CH₄ output.
- The PID analyser gives no signal, even for the most concentrated sampled gas in hydrocarbons. Seeing HC51M COV's measurements (semi-quantitative), the detection limit of PID probe seems to be higher than 1000 ppm of hydrocarbons.
- The ratio between the carbon dioxide channel and the methane channel stays strictly similar for all the LFG samples.

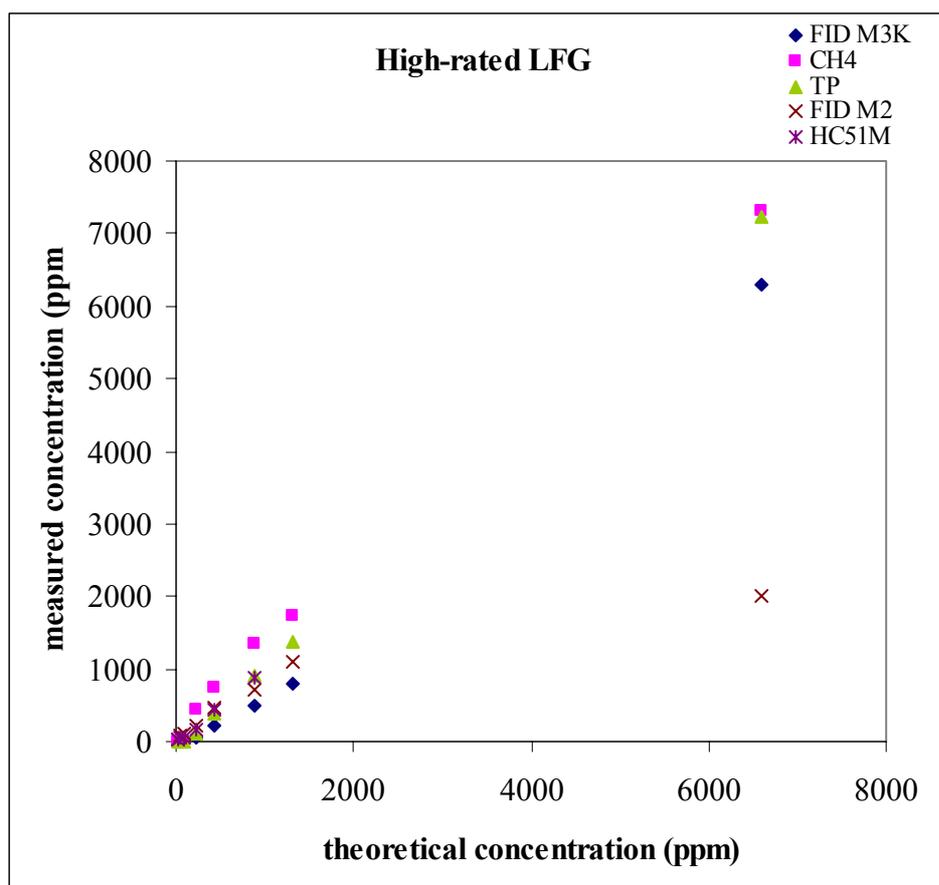


Figure 2: Evolution of the output values of portable analysing devices analysing LFG versus theoretical methane concentration.

Table 3: output values of reference and portable analysing devices on batch 3 samples (LFG emissions collected at the surface of a landfill).

Reference devices			Portable devices			
HC51 M	GA 2000	FID M3K	ECOPROBE			
CH ₄ (ppm)	CH ₄ (%)	éq CH ₄ (ppm)	PID (ppm)	CH ₄ (ppm)	TP (ppm)	CO ₂ (ppm)
20	< DL	1	0	0	0	576
22	< DL	7	0	0	0	855
34	< DL	3	0,01	0	0	646
324	< DL	140	0	0	0	1 447
862	< DL	540	0	1 037	60	1 378
> 1000	< DL	990	0	1 897	1 392	5 376
> 1000	< DL	1 100	0	2 415	1 446	9 268
> 1000	< DL	9 800	0	4 890	3 949	9 698
> 1000	16000	> 11 000	0	13 271	11 361	13 073
> 1000	47000	> 11 000	0	36 651	35 110	31 860

3.3 Batch 3

For the third batch protocol (see Table 3), there is no dilution in lab, thus no “theoretical values”. For low concentrations, HC51M must be taken as reference. For high concentrations, the only “points of comparison” are the GA2000 measurements, even if one has observed that he was not more accurate than IR-MP. As a result,

The results, presented on Table 3, show that:

- FID detector is much more sensible than both CH₄ and TP channels of IR-MP.
- Detection limit of IR-MP on landfill surface emitted gases is higher than for previous batches reaching a value situated between 300 and 1000 ppm.
- CO₂ output of IR-MP probe preserve a response below this limit but with more noise (at least 500 ppm) than on batches 1 and 2.

But the most important observation that one can make on Table 3 is probably that there is no reference lab device able to give an accurate reference measurement for the working range between 1000 ppm and 10000 ppm.

4. DISCUSSION

4.1 General discussion about measuring reliability of CH₄ concentration on landfill surface

The measurements of landfill surface emissions are not easy because of their uncertain geographical distribution over landfill cover. The landfill LFG is escaping through landfill cover using preferential ways such as fissures or cracks in the cover materials. Climatic conditions, as moisture and atmospheric pressure variations (Czepiel & al, 2003; Héroux and Guy, 2005), modify their permeability and imply movings of surface emission areas.

The performed tests show that measurement errors due to portable devices intrinsic lack of exactness or to the way of using them contribute to increase the uncertainty of the methods. This contribution will depend, in particular, of:

- the way of placing the sampling bell and the nature of surface on which it is placed;
- the analytical methods embedded in the analyser (IR, FID);
- interferences with other hydrocarbons contained in the emitted LFG;
- response range of linearity, type, quality and durability of calibration;
- the optional numerical correction and correction software applied to the signal.

As a result, in normal conditions of use, values read by operator on the screen of field analyser are not equivalent to the real concentration of methane in the emitted air. It only gives a rough order of magnitude of this absolute concentration and only allows relative comparison between measuring points.

4.2 Potential interesting contributions of IR-MP devices

Originally, Ecoprobe was a more simple IR-based CH₄ analyser. The actual version, tested in this work, is an IR-MP recently improved, updated and optimised for analysing soil gas in polluted areas and for soil remediation monitorings (Malherbe & al, 2001). Many upgrades have been implemented for increasing its capacity to analyse wide concentration ranges of many hydrocarbon components. These evolutions to a more universal detector make its CH₄ sensibility to decrease.

However, for characterizing fugitive emissions on landfill surface, the IR-MP keeps many advantages in comparison with portable FID:

- to give a surprising measurement accuracy on a very wide range of CH₄ concentrations;
- to furnish simultaneous measurements of oxygen and carbon dioxide thanks to its multichannel and multiprobe technology;
- to store output data into an internal memory that can be downloaded on personal computer.
- to be usable as “fix measuring station” with programmable differed or regular analyses, which is useful for temporal monitoring ;
- to be, according to the manufacturer, insensitive to moisture variations

This last assertion was not tested during this study. It is envisaged to do it in the future.

Nevertheless, the tested IR-MP also present a weak point: the detection limit value of methane is higher than the announced 50 ppm. In case of composite gas, such as LFG, the sensitivity of the probe is even worse: 500 ppm. It is too much for replacing FID sensor.

4.3 PID analyser of the IR-MP

This PID analyser is waited to be complementary to the IR-probe of the IR-MP device: it should measure the concentrations in gas of hydrocarbons, except methane.

But PID tested on the batches never gave any interpretable signal, even for rich LFG. It is well known assumed that high methane concentrations are seriously affecting the performances of PID sensor. This is called “quenching”. The PID sensor embedded on the tested probe is equipped with “anti-quenching system”, which put the PID output to zero when CH₄ concentration exceeds 4000 ppm. And it is possible that, in samples with less than 4000 ppm CH₄, other hydrocarbon concentrations are already lower than PID detection limit. So the signal could pass from a zero value caused by too low concentrations of other hydrocarbons to another zero value, but consecutive to quenching protection.

If this interpretation is correct, one has to conclude that PID sensors are completely unsuited for analyzing hydrocarbons in methane rich LFG.

5. CONCLUSIONS AND FURTHER INVESTIGATIONS

Three tests sequences have been performed on gas samples to compare the performances of portable CH₄ analysers. The results show that classical devices (FID and mono channel IR-probe (GA2000)) are totally unable to cover the full useful range of CH₄ concentrations encountered in biogaz surface emissions (from 50 to 30 000 ppm).

The new IR-MP technology tested, which combine multiple detectors with multichannel IR-analyser has been developed for many years with the objective of becoming able to detect a very large spectrum of hydrocarbon contaminants. This development has been achieved to the detriment of its initial CH₄ sensitivity. Its apparent CH₄ detection limit varies from 4 to 10 times the announced 50 ppm value. It implies that IR-MP can not replace FID device.

Anyway, IR-MP device presents a lot of advantage that are explained above giving to this device a real added value for research teams, landfill owners and authorities implied in the characterisation and semi-quantification of landfill surface fugitive emissions. Further investigations and test are already in hand including:

- testing IR-MP on its announced insensitiveness to gas moisture;
- development of on-field gas dilution coupled to FID device;
- modifying the signal treatment software of the IR-MP technology and going back to a less multicomponent but more CH₄-sensible version;
- in collaboration with INERIS research team, the test of these devices coupled with flux chambers on various landfills of the wallonian control network.

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