THE FLUX-METER: IMPLEMENTATION OF A PORTABLE INTEGRATED INSTRUMENTATION FOR THE MEASUREMENT OF CO_2 AND CH_4 DIFFUSE FLUX FROM LANDFILL SOIL COVER.

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SUMMARY: The municipal solid waste landfills are often considered as important sources of atmosphere contamination due to diffusive emissions of landfill gas (LFG) from the cover. As known LFG is composed by greenhouse gases, i.e. CH_4 and CO_2, and other compounds present in trace with toxic and malodorous characteristics. In order to evaluate the health consequences and the contribution to the greenhouse effect, the D.Lgs. 152/2006, acknowledging the EU Directive 96/61/CE, imposes to monitor and to evaluate the LFG diffused from landfill soil cover. In this work a new portable integrated instrumentation based on the method of accumulation chamber, named FLUX-meter, is presented. This new instrumentation allows to measure the CO_2, CH_4, H_2S, VOC flux at the air-soil interface and permits to quantify the total amount of LFG (as the total flux of the gas species listed above) emitted into the atmosphere and thus lost by the captured system.

1. INTRODUCTION

The municipal solid waste landfills represent significant sources of atmospheric contamination due to uncontrolled emissions of LFG from the landfill cover, which are present even when there is a system for the capture and combustion of LFG. LFG is mainly composed by methane and carbon dioxide - the two main gases responsible for the greenhouse effect - with a percentage of approximately 55% and 45% respectively. In particular landfill CH_4 accounts for about 22% (USEPA, 2001) to the total methane dispersed into the atmosphere. Moreover, the presence of other gaseous species in the LFG, such as volatile organic compounds (VOC) is an additional problem cause of the impact on human health. The Italian and European legislation provides that landfill emissions have to be evaluated to assess both the contribution to the greenhouse effect and the direct effects on the population. Consequently appropriate methodologies should be taken to limit them when necessary.

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There are many techniques available to quantify the flux released from landfill, i.e. gas survey, eddy correlation and flow chambers, but the technology here presented, known as "accumulation chamber method", is a static not-stationary method based on the determination of concentration gradient of CO_2 and CH_4 measured inside the accumulation chamber laid down on the soil.

Starting from the 70's the accumulation chamber has been used to evaluate soil respiration rate (Witkamp 1969; Kucera and Kirkman 1971; Kanemasu et al., 1974; Parkinson, 1981) and since 90's it has been employed to measure the NO_2 and CO_2 fluxes (Chiodini et al., 1996; Chiodini et al., 1998; Frondini et al., 2004; Werner e Cardellini, 2006) from volcanic and geothermal areas. Furthermore the collaborations between West Systems, IGG-CNR and Perugia University have allowed implementing a portable and simpler instrument (Virgili, 2008). This instrument is able to obtain flux measurements directly on the field in a very short time (each measurement takes about 2-3 minute). The main difference from the other methodologies is that soil flux measures carried out with the accumulation chamber are independent of flux regime (advective or diffusive) and of soil characteristics (porosity, diffusive coefficient, etc) (Tonani and Miele, 1991). This characteristic with the advantages of manageability, simple use and fast flux measurements of the instrument has allowed extending its application also to evaluate diffusive emissions from the cover of municipal solid waste landfills (Cossu et al., 1997; Cioni et al., 2002; 2003; Capaccioni et al., 2005; Raco et al., 2010).

This paper describes a portable integrated instrumentation named FLUX-meter, and in particular the high performance of the newer CH_4 detector based on Tunable Diode Laser Absorption Spectroscopy (TDLAS) combined with a Herriot multi-pass cell. In addition, a case of study is presented: a measurement survey was performed in Legoli landfill in 2012 in order to verify the integrated instrumentation directly on the field. The data acquired has also been used to estimate the efficiency of landfill gas collection system.

2. THE ACCUMULATION CHAMBER

Among the numerous techniques available to measure fluxes of a specific gas at soil air interface, the technique of static non-stationary accumulation chamber is considered as the least affected by environmental and meteorological parameters, as showed by Trègourés et al. (1999) comparing seven different methods for the fluxes measurement.

As known, the influence of meteorological condition on soil concentration and fluxes is generally high, so, in order to evaluate the influence of atmospheric pressure, repeated measurements were carried out in some sites located in the landfill area. Moreover, in order to minimize the influence of meteorological condition, the measure should be carried out in the shorter time during dry seasons.

The integrated instrument realized by West Systems consists of (Figure 1):

- an accumulation chamber with specific shape;
- a Non-Dispersive Infrared spectrometer for CO2 measurement;
- a tunable laser spectrometer combined with a Herriot multi-pass cell for the measure of methane;
- an electrochemical detector for H_2S measurement;
- a photoionization detector for VOC measurement;
- pneumatic system;
- battery;

- Palmtop computer (PDA) with integrated GPS antenna.

The sensitivity of each detector is reported in Table 1.

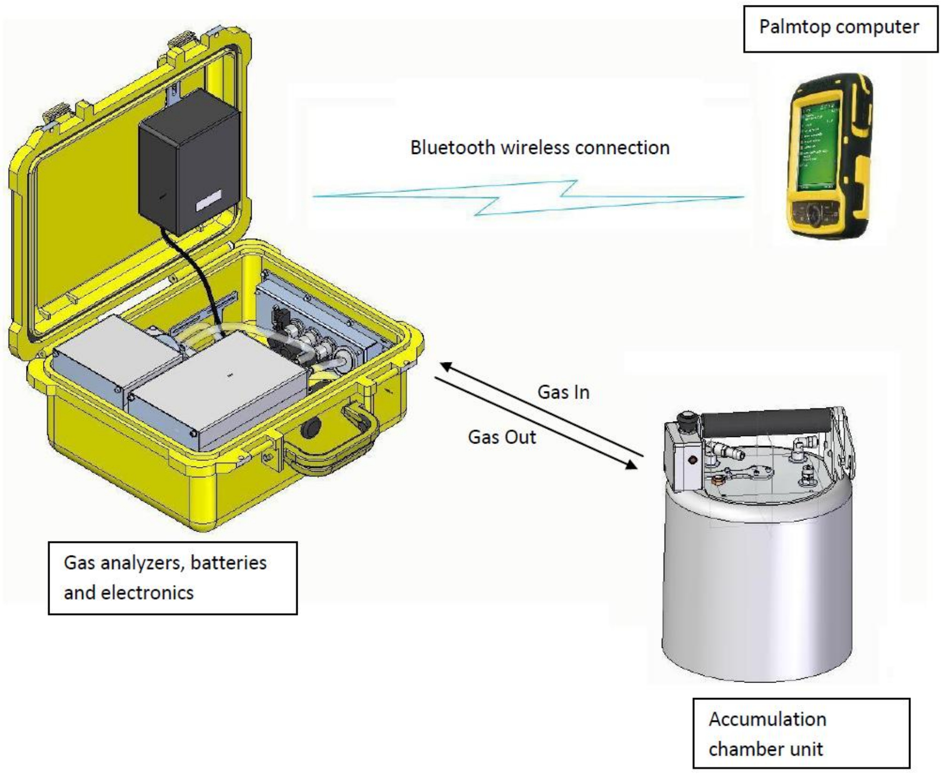

Figure 1: Sketch of the instrumentation

Detector	*Measurement Range*	*Resolution*	*Accuracy*
CO_2	0-20,000 ppm	RMS Noise at 370 ppm with 1 sec signal filtering: <1 ppm	3% of reading
CH_4	0-100,000 ppm	0.1 ppm	10% of reading
H_2S	0-20 ppm	RMS noise (ppm equivalent): < 0.02 ppm	10% of reading
VOC	0-10 ppm	1 ppb	(Isobutylene) 10% of reading

Table 1: Detectors specifications

A dedicated software package gives the operator the evaluation of the initial slope of the gas concentration – time line. The initial slope is proportional to the gas emission from soil. The speed of this instrument is approximately 2-3 minutes per measurement.

To meet the needs of the market, WEST Systems has developed a specific CH_4 detector, with the sensitivity, selectivity and linearity necessary to measure diffuse flux down to extremely low levels. In fact, while the classical instrumentation, based on near infrared absorption, is typically used for the measurement of uncontrolled biogas emissions from landfills, the new methane detector – based on Infrared Tunable Diode Laser Absorption Spectroscopy (TDLAS) – allows extending the instrument application also to evaluate diffusive emissions from landfill soil cover.

To increase the detector sensitivity, the tunable laser diode has been integrated with a Herriot multi pass cell in order to increase the length of the optical path (Figure 2). Therefore very low detection limits are achieved, i.e. 0.1 ppm, allowing to measure low CH_4 concentration close to atmospheric levels (around 1.7 ppm). The detector has two full scale values of 150 ppm and 10%, and the most suitable one is automatically selected. Moreover a very low power supply of 2 Watt is required.

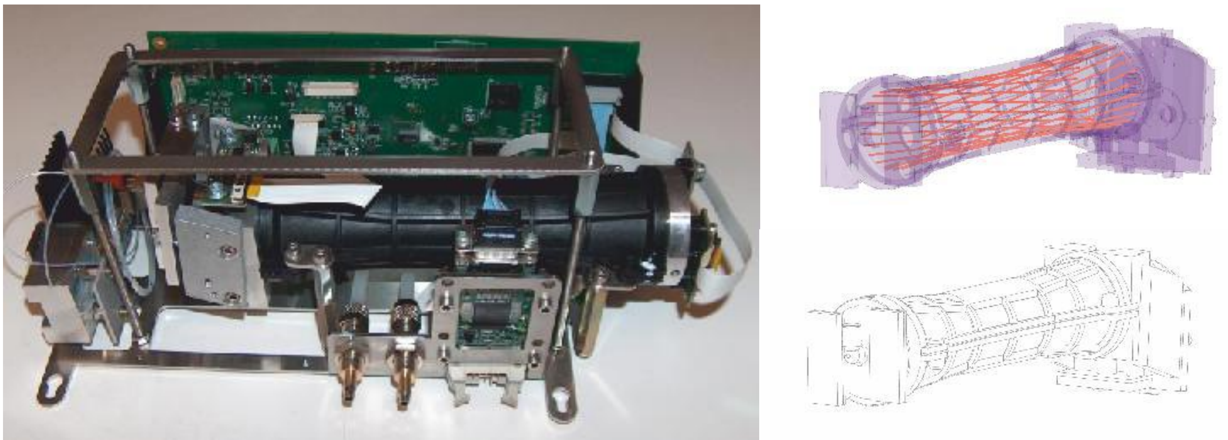

Figure 2. Methane detector, laser diode with Herriot multi-pass cell.

In order to perform a reliable measurement, the chamber edge must be well attached to the ground in order to avoid the input of atmospheric air; inside the chamber a fan ensures the mixing of the gases (Figure 3). In order to perform the measurement, the gas is continuously extracted from the chamber, sent to the analysers with a diaphragm pump, and then injected again inside the chamber. In the same time the operator can visualized on the PDA the increase of gas concentration vs. time and the software computes the flux in real-time.

Figure 3. Steps of a field measurement with the FLUX-meter.

The sensors with electronic and communication interface, pump, pneumatic filters and light NiMH battery are installed in the backpack. In that way the instrument is easily transportable; its total weight, including accumulation chamber, is less than 10 Kg.

The entire instrument is managed by a small handheld computer, a PDA based on Microsoft Windows Mobile operating system, which receives and exchanges information with the instrument via a Bluetooth wireless connection. A specific software allows the simultaneous acquisition of all the gas species (CO_2, CH_4, H_2S, VOC) concentration at 1 hz frequency; Moreover, the software acquires the GPS position and other data, such as pressure and temperature of the measuring cells of the detectors and the voltage of the battery of the system. Therefore the concentration data are represented in function of time, and the acquired flux curves are visualized in real time and can be checked and validated by the operator, interacting with the PDA touch screen. Moreover the GPS antenna integrated in the PDA allows the automatic geo-referencing of the flux measure; an icon in the top of the screen indicates by changing color, the status and the quality of the GPS signal. At the end of the measurement, the data is simultaneously saved into the PDA memory and on the on-board Micro-SD card to be available for further processing.

Detector	*Flux range*	*Accuracy*
Methane TDLAS	0.5 – 1,500 millimoles/m^2·day	± 25%
	1.5 – 1,000 moles/ m^2·day	± 10%
Carbon dioxide NDIR	1.0 – 1500 millimoles/m^2·day	± 25%
	1.5 – 300 moles/ m^2·day	± 10%

Table 2: Accuracy of measurements in function of the flux

2.1 Accumulation chamber - transient method

To understand the relationship between the flux and the initial slope of the gas concentration – time line, the transient method should be considered. The transient or static not-stationary method of accumulation chamber consists in measuring - during the time (t) - the gas concentration (C) under investigation within the chamber only opened on the side laid on the ground. When a diffusive flux from the soil occurs, a gradient concentration (dC/dt, expressed as ppm/sec) is measured within the chamber. Then the flux of a specific gas from the soil surface can be estimated as follows (Natale et al., 2000):

$$Flux = \frac{P \cdot V}{R \cdot T} * \frac{dC}{dt} * \frac{1}{A}, \qquad [E1]$$

where P and T are the environment pressure and temperature respectively, R (with a value of 8.314472 $m^3 \cdot Pa \cdot K^{-1} \cdot moles^{-1}$) is the gas constant, V is the volume and A is the area of the chamber.

From the equation [E1] it follows that the flux (expressed as $moles \cdot m^{-2} \cdot day^{-1}$) is proportional to the gradient concentration (expressed as $ppm \cdot sec^{-1}$) measured (Chiodini et al., 1998), while the proportionality constant is a function of the height of the accumulation chamber (H=V/A if the chamber has a cylindrical or parallelogram shape) and of the air pressure (P_{air}) and air temperature (T_{air}). Since the shape of the accumulation chamber is fixed and P_{air} and T_{air} are measurable, the flux of the specific gas is calculated directly by the gradient dC/dt.

However, during the measure, the gas concentration (C) rises within the chamber and, consequently, the gradient diffusion of the specific gas inside the chamber decreases over time leading to a possible underestimation of the flux (Figure 4). Therefore it is important to measure the gradient in few minutes after the chamber is laid down to the soil, as demonstrated also by Chiodini et al. (1998). In the example in Figure 4 the gradient is calculated through a linear interpolation of data highlighted in red. The slope (the gradient dC/dt) is yellow and the value is indicated in “slope (ppm/sec)” in the box named “Regression” at the top right in the Figure 4. The value in “Flux (moles/m^2/day)” is calculated automatically by the software using the data recorded i.e. “Barometric Pressure” and “Air Temperature”, indicated as P and T in the formula [E1]. The ratio V/A of the chamber is given by the type of chamber used. As showed in Figure 4, choosing a larger time interval, the slope of the line would have been lower causing an underestimation of the flow.

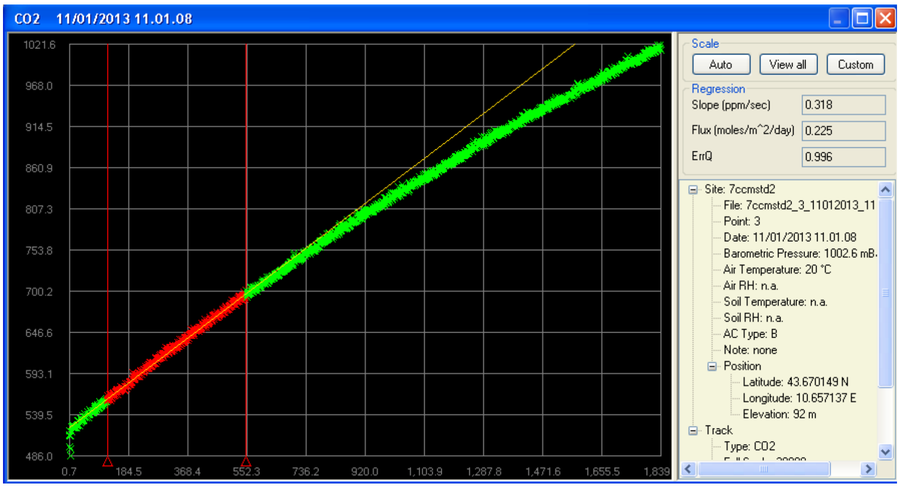

Figure 4. Graphical representation of the increasing concentration value vs time inside accumulation chamber.

A pneumatic system connects the gas sampled in the accumulation chamber and the gas analyser located in the backpack. Therefore this system implies a delay between the concentration in the chamber and value measured by the analyser and could produce an artefact in the first seconds of the measurement, as showed in Figure 5. In this case the determination of the gradient dC/dt is always performed in the range highlighted in red waiting about 80 seconds.

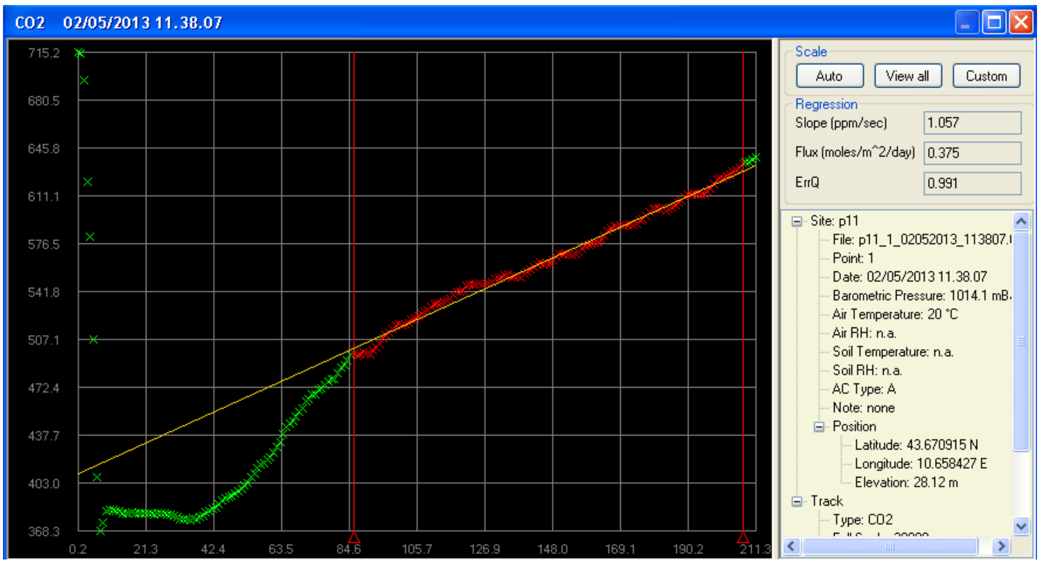

Figure 5. Graphical representation of a typical field measurement.

The most important advantage of FLUX-meter is given by the integration of high performance detectors, able to measure very low CH_4, CO_2, H_2S and VOC concentrations, in a portable instrumentation characterized by manageability, simple use and fast flux measurements. All these characteristics respond to the needs of scientists and engineers engaged in the determination of gas exchange at the soil-air interface linked to the gas escape from the landfill cover, from agricultural soil, from volcanic area and from geothermal exploration sites.

3 EXAMPLE OF DATA PROCESSING

The integrated instrumentation here presented has been used on a municipal solid waste (MSW) landfill covering a surface area of about 140,000 square meter by means of 226 flux measurements.

In order to quantify the total flux released from the landfill cover it is necessary to carry out a series of measurement in specific points inside the whole investigated area located in a grid as regular as possible. The size of the sampling grid is a critical point, due to the great spatial variability of gas flux values; generally a mesh size of 20x20 meters generates a reliable estimation when at least 100 measurements have been obtained.

In order to quantify a specific gas released from the investigated area and to recognize the presence of zones characterized by anomalous fluxes, statistical and geostatistical approach are adopted.

The estimation of the total amount of biogas discharged into the atmosphere has been carried out by the methodology based on partitioning the flux data by means of cumulative probability plots, formalized by Sinclair (Sinclair, 1970 and 1991). For each identified population the arithmetic mean of raw data and its 95% confidence interval has been calculated using the Sichel estimator (Sichel, 1966). An implementation of the Sichel method has been developed by West Systems. The software, which runs on personal computer, calculates the cumulative probability plot, using the data recorded by the PDA during the sampling activity and calculates the total output of LFG taking into account the extension of the investigated area (Raco *et al.*, 2010). By way of example, in Figure 6 the cumulative probability plots of CH_4 flux data is reported and each data subset is recognizable by different colours, while in Table 3 the main statistical characteristics of each population and the estimation of total amount of LFG and its 95% confidence interval are shown.

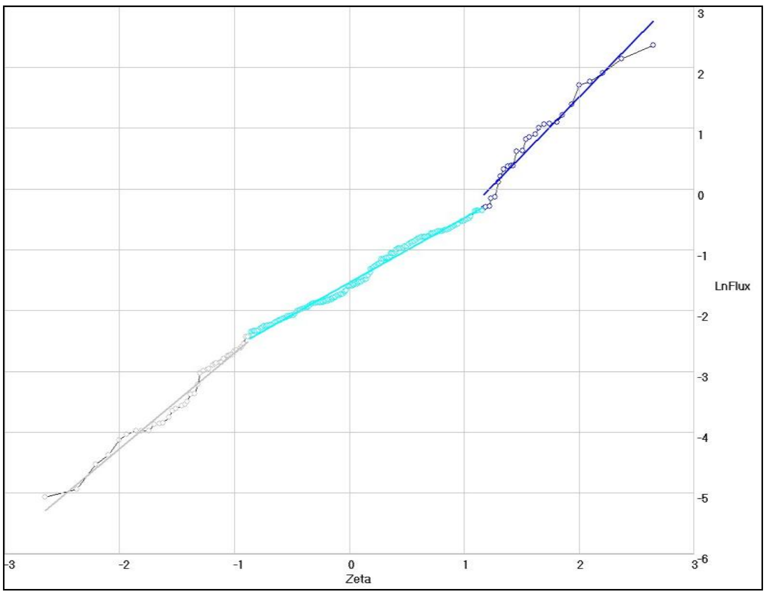

Figure 6. Cumulative probability plots of CH_4 flux.

Subset	*N. measures*	*Mean*	*Variance*	*Sichel-V*	*Flux (Nm^3/h)*	*95% confidence interval (Nm^3/h)*	
1	12	2.19	0.846	1.506	95.4	217.2	61.3
2	131	-0.964	0.889	1.557	46.0	56.4	38.9
3	45	-3.303	0.342	1.185	1.2	1.4	1.0
Tot CH_4					142.5	275.0	101.3

Table 3: Main statistical parameter of each subset of CH_4 flux data.

The Geostatistical approach is used to identify anomalous fluxes from the landfill cover; the data have been mapped using kriging method starting from the study of the experimental variogram. Experimental variogram, relative to the natural logarithm of the LFG flux, and respective variogram model are showed in Figure 7, while in Figure 8 and Figure 9 isoflux and standard deviation maps are presented.

Figure 7. Experimental variogram and variogram model. The green dots represent the experimental omnidirectional semivariogram relative to the natural logarithm of the measurements of LFG flux, while the solid blue line represents the variogram model selected.

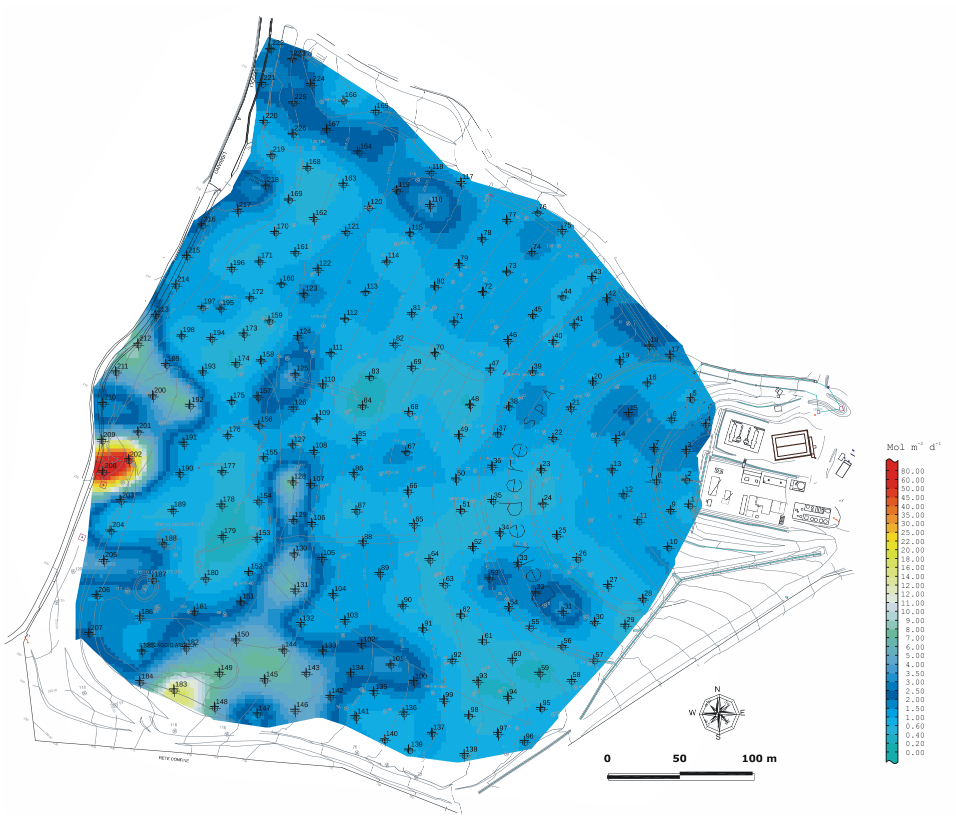

Figure 8. Isoflux map of LFG released from the cover of the MSW landfill. The black dots represent the locations of flux measurements performed with FLUX-meter.

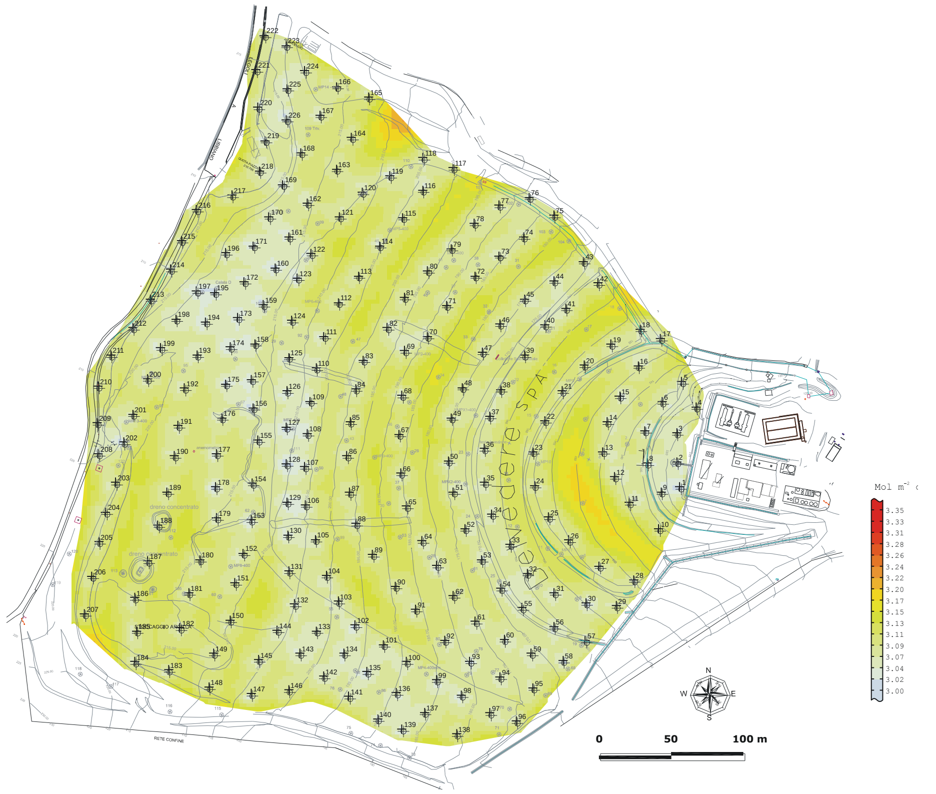

Figure 9. Standard deviation map associated to the isoflux map of LFG released from the cover of the MSW landfill. The black dots represent the locations of flux measurements performed with FLUX-meter.

5. CONCLUSIONS

The integrated equipment set up by WEST Systems presented in this paper allows the measurements of the real amount of CO_2, CH_4, H_2S, and VOC discharged into the atmosphere. Moreover the portable FLUX-meter permits to carry out quick and reliable punctual measurements, which can be processed by an implementation of the Sichel method, developed by WEST Systems, in order to estimate the total amount of LFG discharged into the atmosphere and consequently lost by the capture system. Besides, the elaboration of isoflux maps allow an easy visualization of zone of high fluxes.

REFERENCES

Capaccioni B., Didero M., Lucci P., Pirillo M., Scartoni P., Tatano F. (2005) Camera di accumulo portatile per il monitoraggio di emissioni di biogas da discarica: risultati sperimentali ed elaborazioni preliminari a scala provinciale, RS Rifiuti Solidi, Vol. XIX, n. 2, 96-110.

Chiodini G., Frondini F., Raco B. (1996) Diffuse emission of CO2 from the Fossa crater, Vulcano Island (Italy), Bull. Volcanol., 58, 41-50.

Chiodini G., Cioni R., Guidi M., Marini L., Raco B. (1998) Soil CO2 flux measurements in volcanic and geothermal areas, Applied Geochemistry, 13, 543-552.

Cioni R., Guidi M., Raco B., Guercio M., Corsi R. (2002) CO2 flux from soil: a methodology to estimate the diffuse biogas, Proc. of the 7th Int. Symp. on Environmental Issue and Waste Management in Energy and Mineral Production, SWEMP pp. 163-174.

Cioni R., Guidi M., Raco B., Giamberini S., and Daddi P. (2003) Measurement of biogas emissions from air-soil interface in the MSW landfill of Legoli (Pisa, Italy), Proc. of the Ninth Int. Waste Management and Landfill Symposium. SARDINIA 2003

Cossu R., Muntoni A., Chiarantini L., Massacci G., Serra P., Scolletta A., Sterzi G. (1997) Biogas emissions measurements using static and dynamic flux chambers and infrared method, Proceedings Sardinia 97, sixth International Landfill Symposium, S. Margherita di Pula, Cagliari 13-17 October, CISA.

Frondini F., Chiodini G., Caliro S., Cardellini C., Granieri D., Ventura G. (2004). Diffuse CO2 degassing at Vesuvio, Italy, Bullettin of Volcanology, 642-651, Vol.66.

G.Natale, P.Hernandez, T. Mori, K.Notsu (2000). Pressure gradient measurements in volcanic diffuse gas emanations, Geophysical research letters, 3985-3987vol.l27.

Kanemasu E.T. Power W.L., Sij J.W. (1974) Field chamber measurements of CO2 flux from soil surface, Soil Science, 118,4, 233-237.

Kucera C., Kirkham D. R., (1971) Soil respiration studies in tall grass Prairie in Missouri, Ecology, 52, 912 – 915.

Parkinson K.J. (1981) An improved method for measuring soil respiration in the field, J. Appl. Ecology, 18, 221-228.

Raco B., Cioni R., Guidi M., Scozzari A., Lelli M., Lippo G. (2005) Monitoraggio del biogas diffuso dal suolo da discariche RSU: il caso di Legoli, Peccioli, Pisa. Rifiuti Solidi 2005

Raco B., Battaglini R., Lelli M. (2010) Gas emission into the atmosphere from controller landfills: an example from Legoli landfill (Tuscany Italy). Environ. Sci. Pollut. Res. 17, 1197-1206.

Sichel H.S., (1966) The estimation of means and associated confidence limits for small samples for lognormal population, Proc. 1966 Symp. South African Institute of Mining and Metallurgy.

Sinclair A.J., (1974) Selection of threshold values in geochemical data using probability graphs, Journal of Geochemical Explorat., 3, 129-149.

Sinclair A.J., (1991) A fundamental approach to threshold estimation in exploration geochemistry: probability plots revisited, Journal of Geochemical Explorat., 41, 1-22.

Tonani F., Miele G. (1991) Methods for measuring flow of carbon dioxide through soils in volcanic settimg, Napoli '91. International Conference on Active Volcanoes and Risk Mitigation. Napoli, 27 August-1 September. (abstract).

Trégourès A. et al., (1999) Comparison of seven methods for measuring methane flux at a municipal solid waste landfill site. Waste Management Research, 17, 453-458.

Virgili G., Continanza D., Coppo L. (2008) Il FLUX-meter: implementazione di una strumentazione integrata portatile per la misura dei flussi di CO2 e CH4 diffusi dal suolo, Giornale di Geologia Applicata, 9 (1), 73-84.

Werner C., Cardellini C. (2006). Comparison of carbon dioxide emissions with fluid upflow, chemistry, and geologic structures at the Rotorua geothermal system, New Zealand. Geothermics vol.35 221-238.

Witkamp M., (1969) Cycles of temperature and carbon dioxide evolution from litter and soil, Ecology, 50, 922 – 924.