DEVELOPMENT OF A METHOD FOR THE MEASUREMENT OF NET METHANE EMISSIONS FROM MSW LANDFILLS

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SUMMARY: Field measurements of landfill methane emissions show natural variability. The determination of an average emission rate for a given field site requires sampling designs and statistical techniques which consider spatial and temporal variability. But such an approach necessitates a lot of sampling points using a dynamic enclosure method (dynamic flux chamber). To mitigate this problem, correlations between methane concentrations on the ground, measured with a funnel or by the instantaneous surface monitoring (ISM) method, and the surface methane emission measured with a flux chamber have been developed in order to minimize the number of samplings with flux chamber. For wind speed lower than 16 km/h, correlations are quite good with 154 enclosure measurements performed on a regular grid and on local zones with high methane emission rate which were located using the ISM method. For field measurements 30 sampling points are statistically enough to set up the correlation for the range emission of the characterized zone.

1. INTRODUCTION

Methane (CH_4) is the main component of landfill gas (LFG); it can be selected as a tracer. Thus, a simple and profitable method to appreciate the LFG surface emissions is the systematic screening of the site with a high sensitivity methane detector; measurements are made a few centimeters from the ground (Cooper et al., 1997). But this method does not quantify emissions. In order to measure the net CH_4 surface emissions, several methods exist such as flux chambers, micrometeorological methods, infrared thermography, etc. Flux chambers, in dynamic mode, are often used to quantify methane flows. It makes possible to locally characterize CH_4 surface emission (Kienbusch, 1986; Savanne et al., 1997). This technique offers many advantages, however it is only possible to characterize surfaces smaller than one square meter, whereas landfills surfaces are considerable as well. As a solution to this problem, the use of mathematical methods (geometrical, parametric, geostatistic) associated with measurements in flux chambers carried out at preset points on a regular grid allows to characterize the surface distribution of the CH_4 emissions (Bogner et al., 1997; Czepiel et al., 1996).

However, the number of measurements to be carried out with a flux chamber remains considerable (n 100), in order to obtain unbiased descriptive statistics of spatial data. The significant number of measurements needed with a flux chamber requires time, intrc[illegible] a possibility of significant variations of the sampling conditions or a very heavy [illegible]s. Moreover, the surface LFG emissions are characterized by a great spatial and temporal variability. These variations are due to many factors like the heterogeneous nature of waste, methane oxidation in the final cover (methanotrophic oxidation), the evolution waste permeability over time, the atmospheric conditions, etc. (Cossu et al., 1997). To carry out a strong density of measures in a short lapse of time could better characterize the surface LFG emissions. As example, (Bogner et al., 1997) specified the need to take measurements in a period of one or two days, with a weak variation of the atmospheric pressure. In order to carry out the maximum number of samplings in a short lapse of time and with a simple logistics, a method is proposed in this study. The method combines three techniques. Experimentation was carried out at the City of Montreal's sanitary landfill site, named “le Complexe Environnemental de Saint Michel (C.E.S.M)”, during several months in 2001 and 2002. This paper presents the methodology and the results obtained.

2. LANDFILL SITE DESCRIPTION AND ZONES OF MEASUREMENTS

2.1 The C.E.S.M Landfill

This landfill site is located in the north-central part of the city. It covers 75 ha with an average refuse depth of 70 m. Approximately 36 million tons of waste materials have been landfilled there between 1968 and 2000. Since May 2000, municipal solid wastes are no longer landfilled at this site which is almost filled up. Today, the final covering is in progress and achieved on about 10 percent landfill’s surface. To minimize atmospheric emissions, LFG is recuperated by an extensive collecting system of 350 vertical wells and over 18 km of pipes. This network converges at a pumping facility, where the landfill gas is compressed and burned in a power plant (boiler, steam turbine and alternator) to produce 23 MW of electricity.

2.2 Selection and description of studied zones

Three sectors with different characteristics, named A, B and C for the experiments, were selected to carry out experiments (Table 1). On each one of those sectors, a regular grid was established after a first screening with a portable flame ionization detector (FID) according to the method suggested by the US Environmental Protection Agency (US EPA) (Cooper et al., 1997). Those grids were used to locate each sampling points.

Table 1 - Experimental area characteristics

Area	Surface	Final cover information	Waste in place (metric tons)	Well number	Exploitation information
A	1,6	Covered	1112447	11	1973-1985

B	1,6	Not covered	668160	13	Since 1991
		Approximately 1,5 m of soil with de - inking material			
C	0,36	Not covered	150336	2	Since 1991

3. MATERIALS AND METHODS

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3.1 Sampling methods

3.1.1 Dynamic flux chamber

Many methods were developed in order to quantify surface methane emissions, largely described and compared in the literature (Bogner et al., 1997; Cossu et al., 1997; Czepiel et al., 1996; Hutchinson et al., 1993; Savanne et al., 1997). Among those, the dynamic flux chamber appears to offer many advantages. For this reason, E.P.A suggests this technique for the local measurement of methane surface emissions (Kienbusch, 1986) (see Figure 1). In the current study, the flux chamber used is out of plexiglass. It has a volume of 50 liters and covers a surface on the ground of 0,2 square meters.

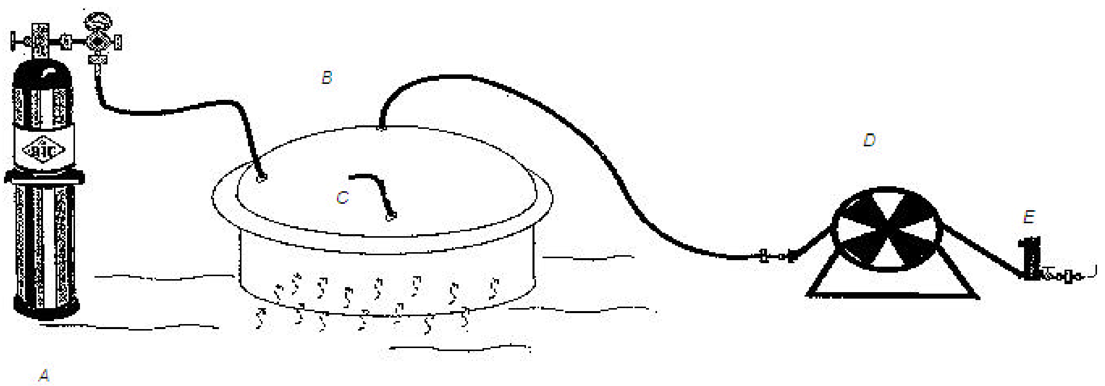

Figure 1. Schematic of flux chamber experiment

A: Pure air
B: Flux chamber
C: Measurement place of CH_4 concentration in the flux chamber
D: Pump
E: Rota meter

Measurements with the flux chamber were carried out at a wind speed lower than 16 km/h (4,5 m/s) to minimize uncertainties (Pokryszka et al., 1995; Savanne et al., 1997). The surface methane flow is determined by measurement of the methane concentration ($[CH_4]$) in the chamber, sealed on the ground with bentonite, after one duration equivalent to three times the residence time of the gas mixture in the chamber. The methane concentration was measured directly in the chamber with a portable FID assuming that it behaves as a completely mixed reactor (Gowing et al., 1997). Residence Time Distribution measurements performed on the flux chamber with a radioactive argon tracer and a set of radioactivity detectors have confirmed this hypothesis. The temperature was also measured in the chamber with a thermometer. The pure air flow (Q_{air}) in the flux chamber was fixed at 10 liters per minute. The methane flow (F) value is given by the following relation:

$$F = \frac{[CH_4] \times Q_{air}}{Surface\ area\ enclosed\ by\ the\ flux\ chamber}$$

3.1.2 Funnel

The funnel is made of polyvinyl chloride (PVC) (see Figure 2). Its volume is 4 liters and i[illegible] 3 a 0.05 square meters area. Methane concentration measure was made directly by inser[illegible] FID into the hole at the top of the funnel. The funnel is used to minimize the wind effects on methane concentration as they appear when measurements are made following the instantaneous surface monitoring method.

The methane concentration evolution in the funnel was followed during laboratory measurements on an experimental set-up simulating a landfill in order to determine the most suitable duration for measurements in funnel. For various emission rates the CH_4 concentrations change proportionally in the funnel with r^2 greater than 0,92. For practical reasons, sampling time with this method was fixed at 2 minutes for field measurements.

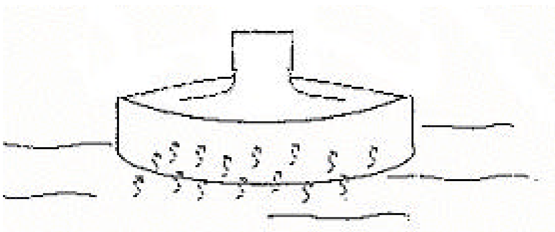

Figure 2. Schematic of funnel experiment

3.1.3 Instantaneous surface monitoring method (ISM)

There are two principal methods utilized for the detection of landfill gas emissions at the surface of the landfill; the Integrated Surface Sampling (ISS) method and the Instantaneous Surface Monitoring (ISM) method. In the ISM method a portable FID is used to instantaneously measure the concentration of total organic compounds (TOCs measured as methane) at the landfill surface divided into grids. Measurements are carried out for wind speeds lower than 16 km/h (4,5 m/s) at 5-7 cm of the ground. (Cooper et al., 1997) give all the monitoring procedures and considerations for the ISM method.

3.2 Meteorological data

Meteorological data regarding relative humidity, wind speed, daily temperature and barometric pressure were measured on an hourly basis by the meteorological station located directly on the

C.E.S.M. property.

3.3 Method description

The method consists to set up correlations between methane concentrations on the ground by the ISM or funnel method and methane flow as measured with the dynamic flux chamber. To establish these correlations, the methane flow range is covered using the results of the first screening by the ISM method. The correlations are set up with a minimum of 30 points to obtain unbiased correlation. The measurements are carried out during various days where the experimental conditions are appreciably the same ones.

The methane concentration is measured after 15 minutes with a portable FID in the chamber. Each measurement in the chamber is immediately followed by measurements using the ISM method and the funnel at the same location. In order to characterize the emissions on a surface, measurements are carried out at the nodes of a grid and at different points with strong emissions as identified during the screening phase, with the ISM method. All measurements are carried out whereas meteorological data (temperature, pressure, relative humidity, speed and direction of the wind) are recorded by the C.E.S.M meteorological station.

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4. RESULTS AND DISCUSSION

4.1 Correlations between measurements

According to sampling conditions presented in section 3, the correlations between methane surface emissions and methane concentrations in the funnel or on the ground by the ISM method were established for each day of measurements. Figure 3 and Figure 4 show respectively the correlations obtained on October 8th, 2002 and October 9th, 2002.

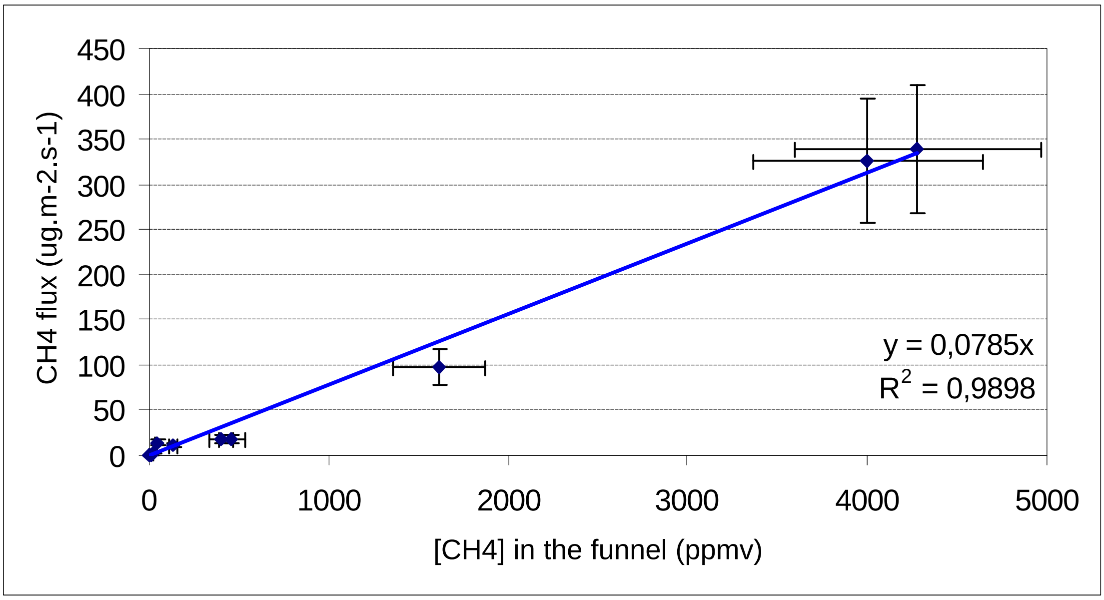

Figure 3. Correlation between $[CH_4]$ in the funnel and CH_4 flux

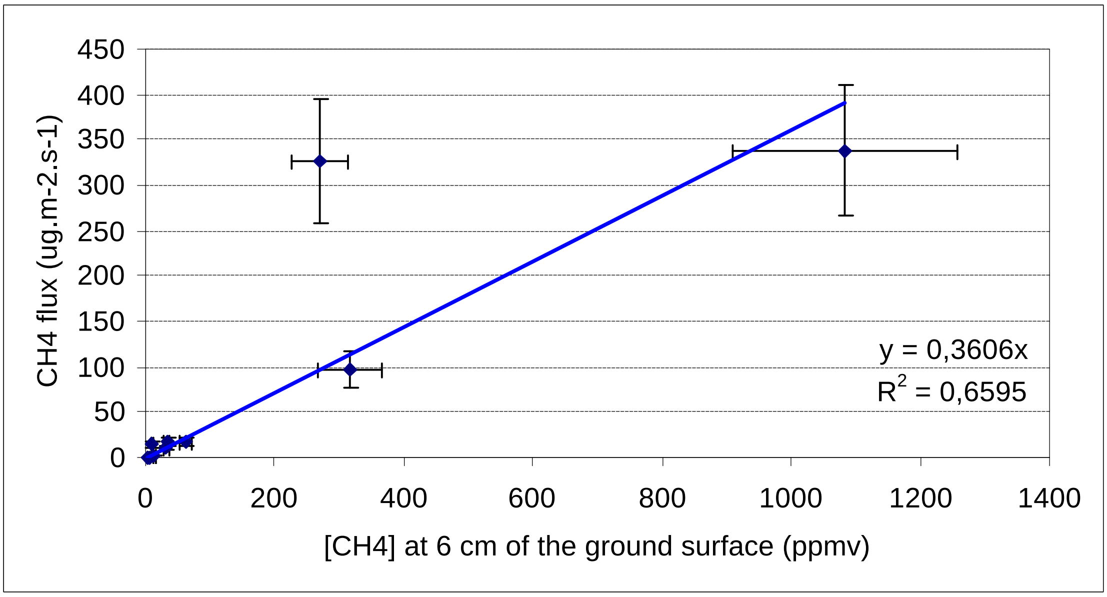

Figure 4. Correlation between [CH_4] on the ground surface and CH_4 flux

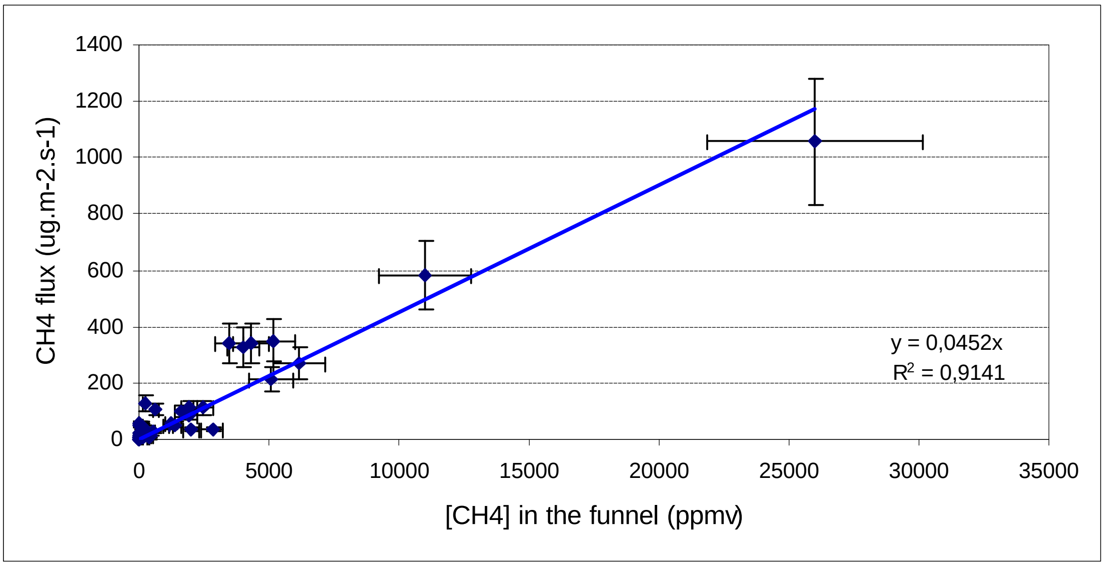

Figure 5. Correlation between [CH_4] in the funnel and CH_4 flux

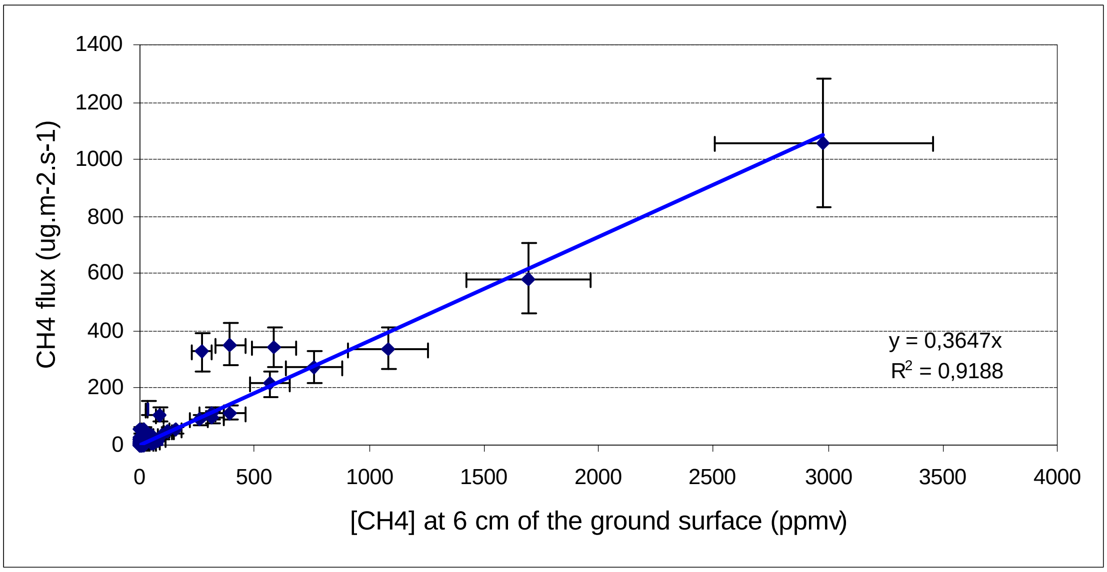

Figure 6. Correlation between [CH_4] on the ground surface and CH_4 flux

For each sampling day and for wind velocities lower than 16 km/h, a linear correlation was found for the two types of technique used (see Figures 3 and Figure 4).

Correlations were studied for various days during two years (see Figures 5 and 6). These correlations were made for different ground characteristics (dry, wet), for different atmospheric pressures, for cloudy sky or not, for different outside temperatures and humidities. The methane concentrations in the funnel (x_1) after 2 minutes were correlated with methane surface emission (Y). The equation $Y = 0{,}0452\ x_1$ was obtained with a determination coefficient $r^2 = 0{,}9141$ (see Figure 5). In the same way, the methane concentrations on the ground measured according to ISM method (x_2) were correlated with surface methane emission with the followed equation $Y = 0{,}3647\ x_2$ and $r^2 = 0{,}9188$ (see Figure 6).

Table 2 shows statistical analysis of correlations presented in Figures 5 and 6. The t and p values show that there is a good correlation between the couples of variables (Y, x_1) and (Y, x_2) (Bowker et al, 1965).

Table 2 - Statistical analysis of the correlations presented at Figures 5 and 6

X, Y	Mean	Std. Dv.	r(X,Y)	r^2	t	p	n
[CH_4] in the funnel (ppmv)	567,90	2449,70	0,956	0,914	40,45	0	154
CH_4 flux ($ug.m^{-2}.s^{-1}$)	35,41	113,01					
[CH_4] on the ground (ppmv)	82,19	309,43	0,959	0,919	40,81	0	154
CH_4 flux ($ug.m^{-2}.s^{-1}$)	35,41	113,01					

4.2 Applications

In order to set up correlations, statistics define a minimal number of 30 observations to be realized. In order to satisfy this requirement, the correlations will be established over several days.

For standard days, the use of correlations makes it possible to reduce the number of measurements with the dynamic flux chamber by correlating a measurement of methane concentration on the ground (measurement in funnel or by the ISM method) to a surface methane emission. From measurements with the methods carried out on the nodes of the grid and for highest methane emission points or zones determined using the ISM method during the screening step, emissions for the studied zones were estimated for various days by the kriging method with the Surfer 7 software (Figure 7). The variogram models retained for kriging were chosen to minimize errors between the measured and modeled values for all sampled points on the zone. The surface integration allows the estimation of the quantity of methane escaping from the zone per unit of time. The two correlations give roughly the same whole emission for the same surface and the same day (Table 3).

Table 3 presents some methane emissions determined for studied zones of the C.E.S.M site. For example, methane whole emission was estimated at (35,88 ± 7,54) $*10^{-3}$ g/s for area A. The emission is lower for this area. This zone with a final cover is older and generates less methane than other zones of the site. For area B, methane emission is 3,4 times greater than methane emission on area A. The measurements carried out on area C allowed to appreciate and estimate the methane emission reduction for various year periods, whereas the methane generation rate and emission decrease with the cold seasons (Cooper et al., 1997; Park et al., 2001).

Table 3 - Methane flows for studied zones

Method	Sampling day	Area	Net CH_4 emission ($ug.s^{-1}$)	Error ($ug.s^{-1}$)
Funnel	27-08-2002	A	35885	7536
Funnel	04-09-2002	B	121623	25541
Funnel	18-09-2002	C	74151	15572
ISM	18-09-2002	C	76731	16113
Funnel	08-10-2002	C	22630	4752
Funnel	09-10-2002	C	22337	4691
ISM	12-11-2002	C	29659	6228

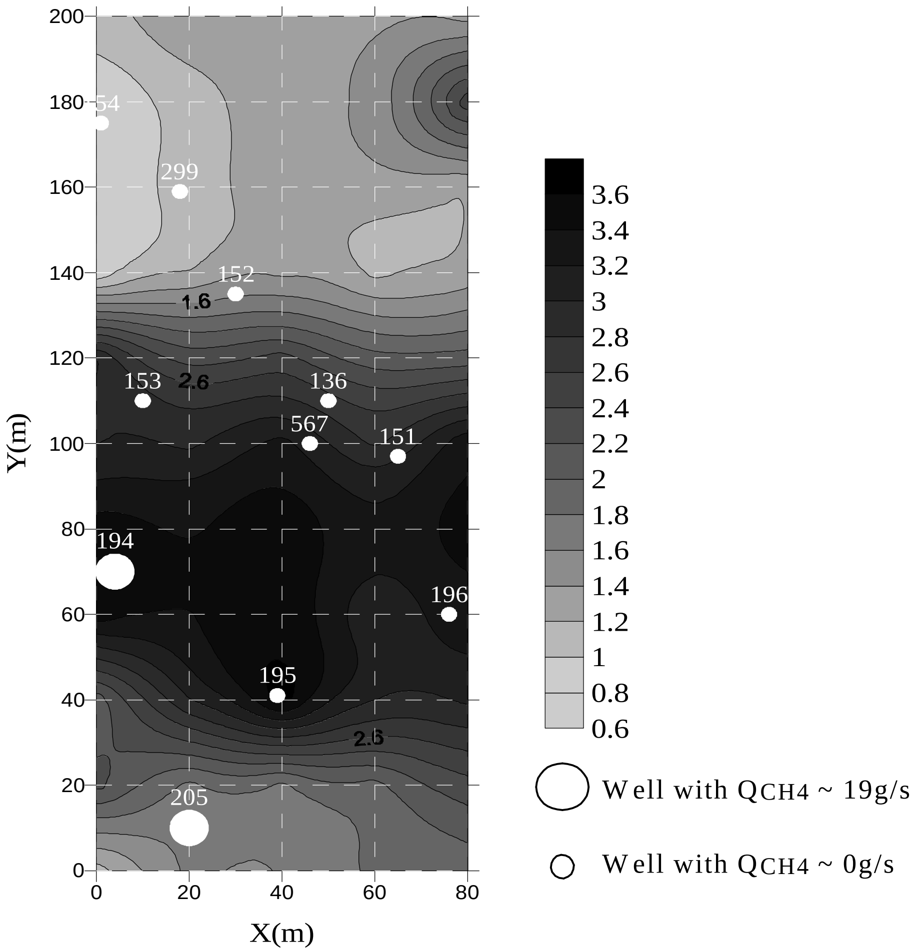

Figure 7. Methane flux chart for area A (ug.m^{-2}.s^{-1})

5. CONCLUSION

The use of the dynamic flux chamber, the funnel and the ISM method can make it possible to characterize net surface methane emissions for wind speeds lower than 16 km/h. By characterizing the range of methane emission for a surface by the dynamic flux chamber, correlations with methane concentrations on the ground measured with the funnel or by the ISM method were established. The correlations were relatively good and allowed to have rapidly enough measurements to draw up the methane emission chart with the kriging method. The two correlations give the same results, even if measurements with the funnel show a higher accuracy because this method minimizes the wind effects. For field measurements, 30 sampling points are shown to be statistically enough to set up the correlation. The choice between the two methods of measurement methane concentration on the ground to set up the final correlation depends on several elements; for sites with low methane emission (old sites or with aggressive LFG collecting system), the funnel method is suggested; whereas for younger sites, where methane emissions are higher, the ISM method is suggested.

The simple logistics and the low sampling time requirements of this method allow undertaking frequent campaigns and multiple measurements over an extended surface, thus enabling a better characterization under various weather conditions of the methane sites emissions. This technique decreases significantly time and labor investment and provides an adequate estimation of large area emissions with the added benefit of characterizing the spatial distribution of these emissions.

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