LANDFILL BIOGAS EMISSIONS MEASUREMENTS: QUALIFICATION AND QUANTIFICATION OF THE PATHWAYS OF EMITTED METHANE

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Abstract

One tonne of municipal solid waste stored in a landfill produces from 120 to 150 m^3 landfill gas within several years. Without any recovery system, the landfill gas can migrate toward the atmosphere and contribute to the greenhouse effect. Alternatively, due to its methane content, it can be recovered and used as an attractive source of energy from waste. The research program METAN has been developed in the Waste and Energy Research Centre (CR*ee*D, VIVENDI Group) to identify and quantify the various methane flows in landfills. This work will allow exact determination the contribution of landfills to global greenhouse gas emission as a function of the different landfill configurations and on-site management. The first application has been made on the experimental Montreuil/Barse MSW Landfill (CGEA-ONYX, France). A field campaign was completed in February 2000 in winter conditions. The quantification of the fraction of methane collected, methane emitted toward the atmosphere and methane oxidised through the cover has been investigated on a broad range of configurations (covered, temporary covered and non covered cells, with and without biogas recovery). The methane flow emitted by the clay covered cell represents 2% of the methane flow emitted by the operating cell. When the gas recovery system was turned off, the flux of methane measured on the same cell increased by a factor of two but stayed negligible compared to the operating zone. The emissions of methane are not distributed in a homogeneous way on all the cells. The chamber measurements show that the methane is emitted very unevenly: the surface biogas fluxes are mainly localised near discontinuities such as leachate wells or cells' borders. No oxidation was measured in the winter conditions except in the temporary cover.

   Key-words- Emissions, field measurements, greenhouse effect, landfill, methane, oxidation.

1. Introduction

Methane, which is a more potent greenhouse gas than CO_2, has increased in atmospheric concentration by a factor of 2 in the past century. About 70% of the current emissions are from human related activities (US EPA, 1990). Methane generated in landfills is a direct result of the natural decomposition of solid waste. The organic component of landfilled waste is broken down by bacteria in a complex biological process with gaseous products consisting mainly of methane and carbon dioxide.

Estimates of global landfill methane emissions to the atmosphere have ranged from 9 to 70 $Tg.y^{-1}$, differing mainly in assumed methane yields from estimated quantities of landfilled refuse and assumed methane oxidation.

The most recent estimates indicate an annual atmospheric contribution of about 20-30 $Tg.y^{-1}$. For France, national estimates prepared for the United Nations Framework Convention on Climate Change indicate that about 25% of the annual methane emissions are from waste disposal, constituting the second largest source after agriculture (United Nations, 1995).

But none of the existing national and global estimates are based on field measurements of emissions; rather they rely on estimated rates of methane production applied to national statistics for landfilled refuse. That is why this study was undertaken to quantify the various biogas flows on a landfill. At individual site, a methane balance provides a useful framework to describe landfill methane processes and net emissions.

Such a mass balance approach should be applied to the partitioning of methane production into methane emissions, methane oxidation, methane recovery, lateral methane migration and the change in internal methane storage (Bogner and Spokas, 1993).

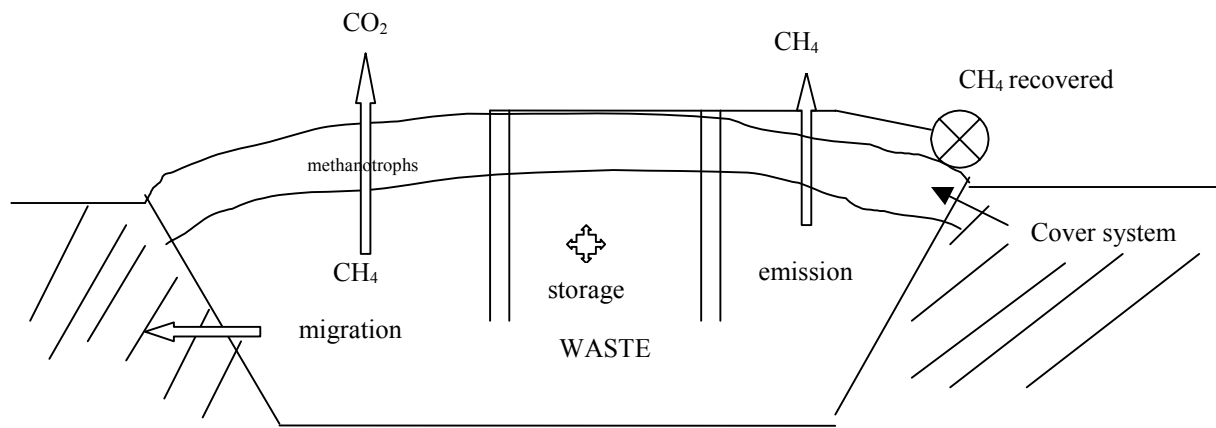Methane production = Σ(emission + lateral migration + recovery + oxidation + Δ storage)

Fig. 1. Schematic representation of the landfill methane balance (from Bogner and Spokas, 1993).

The research program METAN began in February 2000 with field measurement campaigns of (i) recovered fluxes (ii) surface fluxes of methane on the landfill and (iii) of methane oxidation through the landfill cover.

The investigated landfill presents several types of cell configurations. The adopted strategy to quantify the emitted biogas was to use complementary approaches: (i) Infrared thermography to locate thermal anomalies of the site, potentially due to emission zones, (ii) Direct measurement of methane emissions using both a tracer method and a static chamber method, (iii) A stable carbon isotopic study to determine the portion of methane oxidised during transit from the landfill surface.

2. Materials and Method

2.1. Study areas

The CGEA-Onyx landfill "Montreuil sur Barse" situated near Troyes in France has been operating since 1986, and has received ISO 14001 certification in 1999.

This municipal solid waste landfill (MSW) consists of three different parts (Fig. 2) : (i) a former area where MSW was disposed of from 1986 to 1993, (ii) an experimental area where MSW was disposed of from 1994 to 1999 (iii) and a new operating zone.
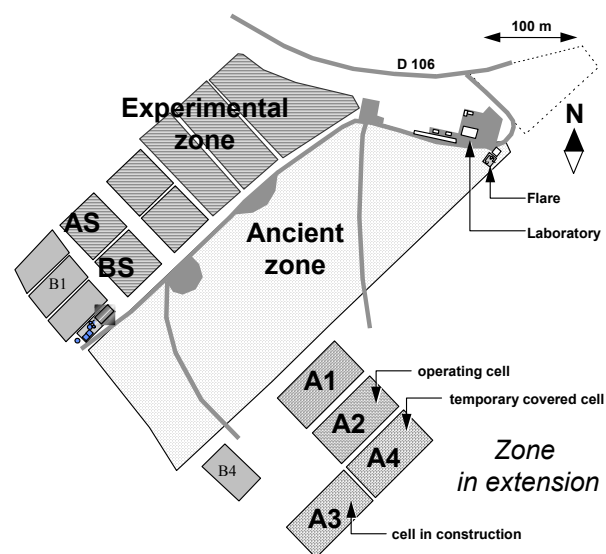

Fig. 2. Schematic map of the investigated areas

It has the advantage of a broad range of cell configurations: (i) an active cell (A2), (ii) a temporary covered cell (A4), (iii) two experimental cells made of different top cover configurations (AS with a clay cover and BS with a synthetic bentonitic geocomposite), and (iv) areas with and without gas recovery (AS and BS). The amount of waste landfilled in these several areas are presented in tab.1.

All of the measurements took place in the same two weeks so that it occurred approximately in the same conditions. The Infra Red Thermography (to establish the map of the thermal anomalies of the site and to locate the areas to be investigated) and the local flux measurements were the first campaigns to occur.

Year	Cells (amount of waste in tonnes)			
	As	Bs	A2	A4
1993	180,00	0	0	0
1994	8 556,96	0	0	0
1995	0	8 114,46	0	0
1996	801,42	1 054,64	684,08	0
1997	0	0	35 246,21	0
1998	0	0	0	0
1999	0	0	2 816,60	36 603,58
2000	0	0	25 926,24	26 017,00
TOTAL	9 538,38	9 169,10	64 673,13	62 620,58

Tab. 1. Amount of waste landfilled in the different cells.

Then the special fluxes measurements (according to the IR thermal anomalies map) and oxidation measurements campaign begun. The measures were first realised on the two cells As and Bs with the recovery of the biogas (pressure at the well of −2.5hPa for As and −1.4hPa for Bs). To evaluate the influence of the recovery of the biogas on the emission of methane through the cover, the recovery was stopped for two days (the pressure at the biogas well increased until 1.8hPa for As and 7hPa for Bs) before the fluxes were measured again. During these two days, the other areas were investigated.

2.2. Infra Red Thermography measurements

The IR thermography consists in visualising the spatial distribution of discernible surface temperatures, in the form of a thermograph. This visualisation is achieved by measuring the energy in a given spectral bandwidth radiating from an object. The sensor used is a thermal line scanner. The entire system is installed on an aeroplane, which, for this project, made transects once during the day and once at night.

The use of the IR Thermography allowed the establishment of a map of the thermal anomalies of the landfill. On a landfill, these anomalies may have several origins, among them: wet areas, zone of LFG emissions, areas where the microbial activity is important under the cover. The map of the thermal anomalies has been used to determine local chamber measurements location and to identify a possible correlation between the IR and the emissions of biogas.

2.3. Large area measurements of the fluxes of biogas by the tracer method

A first set of surface fluxes have been used to determine large area methane emissions. This method is based on tracer measurements and has already been described in Trégourès *et al*,
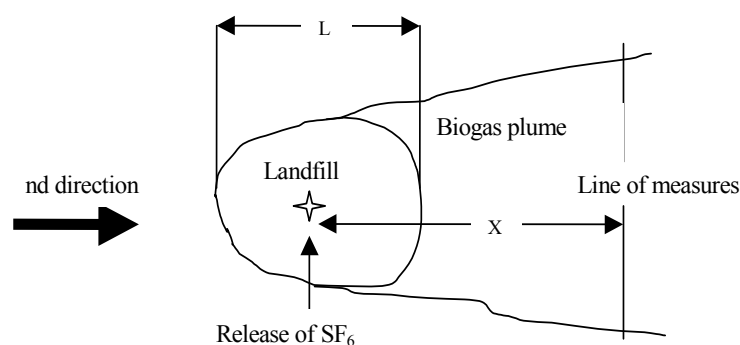

Fig. 3. Schematic principle of the downwind method.

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1999. This method relies on concurrent concentration measurements for the methane and an inert tracer gas (here SF_6) released at a known rate upwind the landfill (Fig.3.).

The methane flow is derived from the atmospheric transfer coefficient K measured by the tracer gas. From a methane source to a given point, K is the relationship between the concentration of methane integrated at that point and the mass released. When the methane flow rate Q is constant, K equals to C/Q where C is the concentration of methane at the point of measurement. While K is a function of the position of the point of measurement in relation to the source and weather conditions, it is independent of the type of effluent released. The flow rate Q is derived from K, which is determined (i) by the emission of a known mass flow of SF_6, and (ii) by measurements of tracer gas and methane concentrations at the same point.

The tracer method consists in injecting the SF_6 above the landfill and measuring the integrated concentrations of SF_6 and methane on a line situated downwind, based on the average wind direction. The readings were made using two gas phase chromatography devices equipped with a flame ionisation detector for methane and an electron capture detector for SF_6. (Trégoures et al., 1999). The error rate is inversely proportional to the X/L ratio. For the Montreuil measurement campaign, the uncertainty is on the order of 50%.

2.4. Local measurements of the fluxes of biogas by the static chamber method

The static chamber method n°1 has been used to measure methane fluxes from small areas of the landfill surface (0.25 m²). This portable method is static, without outside circulation of the gas. Its design revolves around a Figaro TGS-813C sensor, which operates within the chamber itself, downstream of a desiccant.

This device is equipped with a sensor voltage regulator, a pump, a fan and two autonomous loggers, one for the temperature and the other for relative humidity. The measure involves the placement of the static accumulation chamber on top of the ground surface for short measuring periods.

The flux is calculated from the change in chamber methane concentration with time and the chamber volume/area ratio: the concentration of CH_4 is measured continuously (and analysed on site) which allows to calculate the flow of methane in the chamber and then the flux of methane emitted by the area covered. The uncertainty of this method is on the order of 50%.

Another type of static chamber, n°2, has also been used. This chamber is constructed of a stainless steel hemisphere, which covered a surface area of 0.11 m². Two replicates were used for one point of measurement. During measurement times, each chamber was placed on top of a semi-permanent iron collar which was pushed into the cover materials to a depth of 5 cm at least. Each collar included a trough welded onto the top of the collar that exactly fitted the base of the chamber. Water was placed in this trough to provide a gas-tight seal with the chamber; hand clamps were also used to secure the chamber to the trough (Fig.4.).
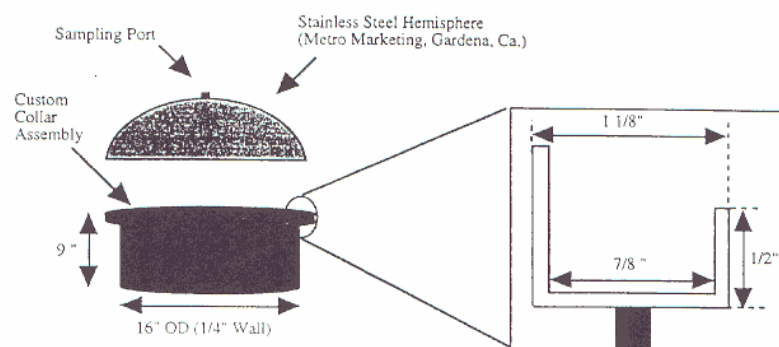

Fig. 4. Scheme of the static chamber n°2 (J. Bogner et al., 1994).

Gas samples were taken every 10 minutes during an hour, using syringes. Following gas analysis in the laboratory, a plot of concentration vs time was made for each flux and the slope of the best fit linear regression was taken as dC/dt in equation 1 (after Rolston, 1986).

$$J = (dC/dt)(V/A)$$ where
J = flux $(g.cm^{-2}.s^{-1})$
dC/dt = change in concentration over time $(g.cm^{-2}.s^{-1})$
V = volume of chamber (cm^3)
A = surface area under chamber (cm^2)

Equation 1. Calculation of the gas flow in the static chamber.

2.5. Quantification of the methane oxidised trough the cover.

Microbial methane oxidation is carried out by methanotrophs. Methanotrophs are a class of methylotrophs which have the specific enzyme (methane monooxygenase) that allows them to oxidise methane to methanol (Anthony, 1982). Methylotrophs are microorganisms capable of gaining energy from the oxidation of reduced carbon compounds.

Many factors can seasonally affect the mass of methane oxidised through landfill cover soil. These include all the controlling variables for soil microbial processes: temperature, moisture, nutrients, substrate form and availability, and presence/absence of toxins (Bogner, 2000).

A major uncertainty in estimating CH_4 flux from landfills is determining the attenuation of CH_4 emissions by methanotrophic bacteria in the aerobic outer portions of the cover soil. These bacteria intercept the gas as it migrates toward the atmosphere.

To estimate cover soil oxidation, measurements of the difference in the $\delta^{13}C$ of CH_4 within the anoxic zone and CH_4 released from landfills and captured downwind on the landfill.

Diagrams (Fig.5.) shows CH4 escape from landfills through (i) fissures and vents, which is measured through downwind plume sampling and (ii) transport through the soil cap, which is measured utilising the chamber technique and the downwind plume sampling method (Chanton *et al*., 1999).

This stable isotopic study was conducted to determine the major pathways of methane emission from the landfill and the portion of methane oxidised during transit from the landfill surface. Samples of methane were collected from the plume of air downwind of the landfill. Methane within these samples integrates the total emission for methane from the landfill. Specific samples were also collected from all major sites of potential

*Fig. 5. Plot to illustrate carbon isotopic variations for landfill methane (*Chanton *et al., 1999).*

emission including from ventilation pipes, from pipes harvesting methane from the subsurface and from bubbles of landfill gas effusing from cracks and borders of landfill cells.

Stable isotopes are useful for determining CH_4 oxidation because, as it occurs, the remaining CH_4 becomes ^{13}C enriched due to preferential utilization of the lighter ^{12}C isotope by bacteria. The difference in the isotopic signature of these two pools of methane is directly attributable to the fraction of methane oxidised $[f_0]$.

Oxidation percentage is determined by equation 2 (Chanton *et al*., 1999), which describes isotopic fractionation in an open system.
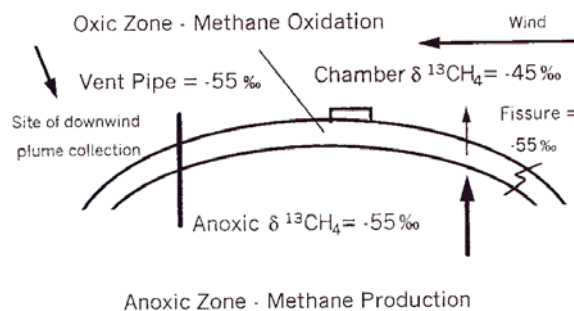
$$f_0 = [(\delta E - \delta A)/(\alpha_{ox} - \alpha_{trans}) * 1000] * 100$$

where f_0 is the % of CH_4 oxidised in transit trough the cover soil cap

$\delta E = \delta^{13}C$ value of emitted CH_4

$\delta A = \delta^{13}C$ value of anoxic zone CH_4

α_{ox} is the isotopic fractionation factor for bacterial oxidation

α_{trans} is the isotopic fractionation factor associated with gas transport.

Equation 2. Expression of the isotopic carbon fractionation.

Liptay *et al.* (1998) and Bergamashi *et al.* (1998) have argued that gas transport across the soil cap is dominated by advection. Therefore, it could be assumed that $\alpha_{trans}=1$.

The bacterial fractionation factor associated with methanotrophy was determined by incubating cover soil samples at *in situ* temperatures. The fractionation factor α can be determined with equation 3 (Chanton *et al.*, 2000):

$$\delta^{13}C_t = 1000 * (1/\alpha - 1)\ln(n/n_0) + \delta^{13}C_{t=0}$$

where n/n_0 is the fraction of methane remaining at time t

$\delta^{13}C_{t=0}$ is the $\delta^{13}C$ value of the methane at the initial time

Equation 3. Equation for determining the fractionation factor α

A time-series of analysis was performed to determine the fractionation factor α : cover soil samples were placed in closed flasks and a known concentration of methane was added. These flasks were incubated at outside temperatures, and two gas samples were taken every day during seven days. The determination of the isotopic composition of these samples permits calculation of, as in equation (2), the fractionation factor inherent to the soil and to its specific microbial flora.

3. Results and discussion

3.1 The IR thermography

Figure 6 presents the IR thermography performed in Montreuil/barse Landfill in February 2000. Thermal anomalies (in white) clearly show the site configuration. The saturated zone, like leachate lagoons and surface water collection network define the landfill contour. Heat emitted from composting platforms, operating zones, surface landfill gas collection systems and flare are also clearly detectable on the IR picture.
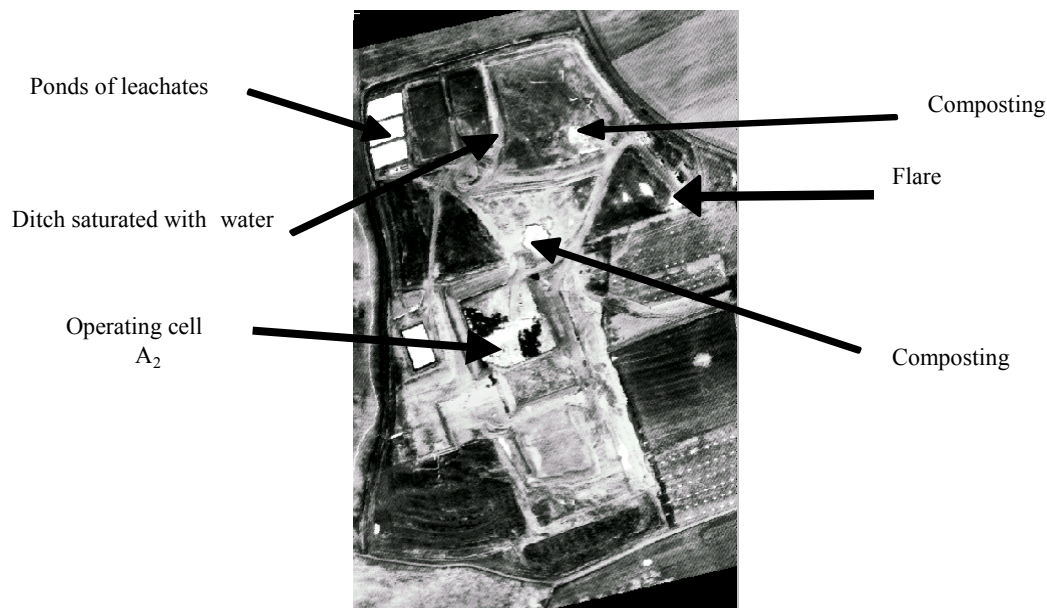

Fig. 6. Black and white map of the thermal anomalies of the site.

Unexplained thermal anomalies localised on different areas on the landfill have been investigated in order to determine if they correspond to LFG emissions zones. Figure 7 presents thermal anomalies detected on the experimental Cell GS (with a geosynthetic bentonitique cover and a 10% slope), and the associated surface flux measurement position.
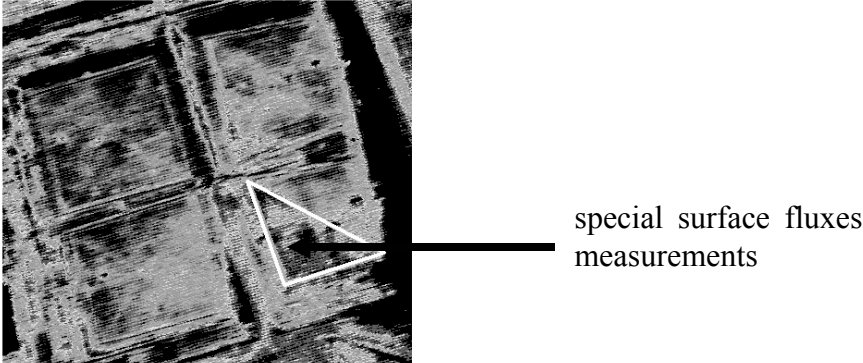

special surface fluxes measurements

Fig. 7. Map of the thermal anomalies of the experimental cells.

The measured flux in those areas present the same order of magnitude than the flux of cell As and Bs. The thermal anomaly is not directly linked to a higher LFG emission. The possible explanation to that thermal difference would be either the presence of saturated zone in the cover, or a weaker thickness of the cover layers in these areas. Among the 26 measurements performed in areas with thermal anomalies, 7 measurements have presented higher flux and were mainly linked to a zone with a different nature of the cover.

3.2. Results of the tracer method

The tracer method gives the global flux of methane emitted by an entire cell. First results have shown high fluxes on the different landfill areas. The values have not been correlated with the fluxes measured by the two chamber methods. In addition, fluxes measured by the tracer method on the operating cell is four times greater than the value given by the modelling of the LFG production. Complementary global fluxes will be performed in order to check these first results. The results can nevertheless be used relatively to compare the emission of methane on the landfill.

Figure 8 presents the relative emission of methane on the different covered cells expressed in percentage of the emission of methane on the operating one. Surface fluxes are reduced and represent from 9% to 2% of the operating zone emissions depending on the nature of the cover and the LFG
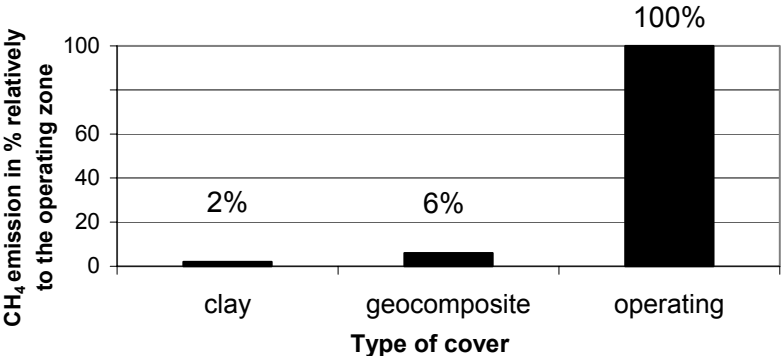

Fig. 8. CH_4 emission fluxes from the clay, geocomposite and operating zone with biogas recovery (except for the operating zone).

collection system. The lowest flux occurs above the clay-covered cell when LFG is collected and it corresponds to 2% of the flux on the operating cell. This measured flux is similar to the flux measured on the geocomposite cover (6%).
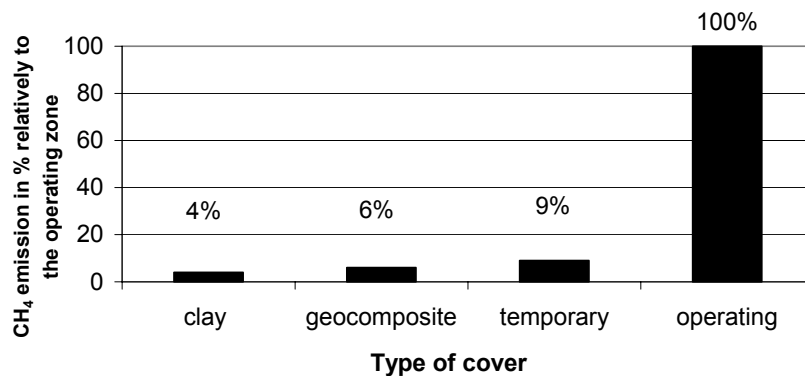

Fig. 9. CH_4 emission fluxes from the clay, geocomposite, temporary and operating cells without recovery.

In the configuration where LFG is not collected (Fig 9), surface fluxes have been multiplied by 2 but remain low when compared to the operating zone. The temporary cover is made of 30 cm of clay material placed without any special care. The temporary cover appeared very efficient as it emitted a flux corresponding to 9% comparatively to the operating zone and presents similar characteristics to the clay and bentonite cover.

3.4. Localisation of the points of emission.

The first chamber method (n°1) used in this study presents high uncertainties. Data obtained by the chamber method differ depending on the techniques. As an example, on cell Bs, fluxes given by the chamber n°1 (with a fan and on site analysis of the CH_4 concentration) range from 198.64 to 214 830 mg.m^{-2}.d^{-1}, and fluxes measured by chamber n°2 (without fan and further analysis in a lab) give values ranging from –2.47 to 33.8 mg.m^{-2}.d^{-1}. On top of that, the n°1 chamber method seems not to be precise for little fluxes. Due to the high uncertainties, the chamber measurements n°1 have been used only to locate the points of methane emissions.

Fluxes measurements' distribution is presented in figure 10 and 11. Each cell presents a broad range of values for the emission of methane. It appears clearly that only few limited areas are emitting methane (in black). Those areas are mainly localised near discontinuities such as the leachate or biogas wells or the cell's borders which represent preferential pathways for gas transfer. Apart these local zones, emissions through the cover are insignificant.
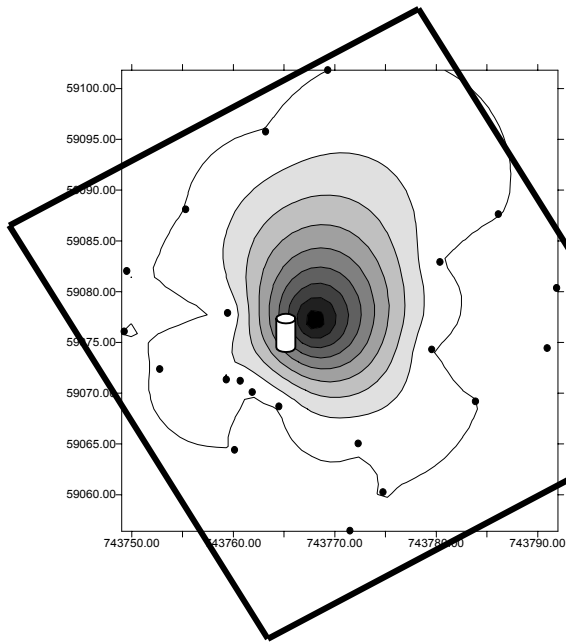
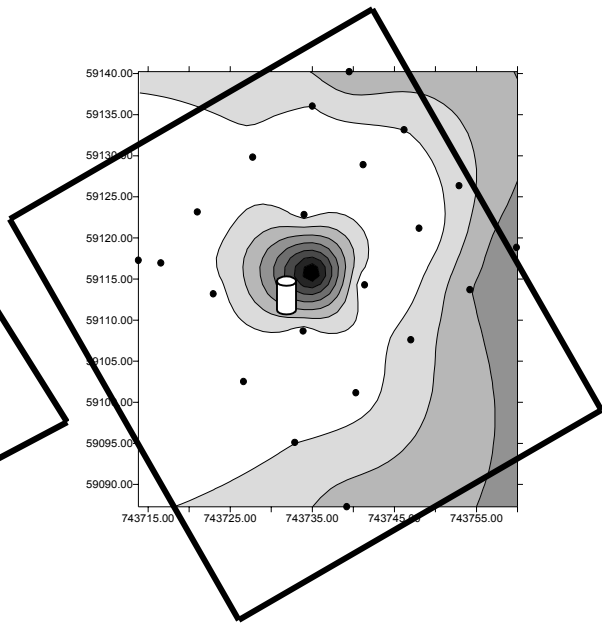*Fig. 10. Schematic localisation of the emission of methane on the geocomposite covered cell (Bs)*

Fig. 11. Schematic localisation of the emission of methane on the clay covered cell (As)

 biogas well   border of the cell

Often in the experimental zone, where strong suction was being applied in the process of methane recovery, negative fluxes were measured (Fig.12.), indicating that atmospheric methane was being taken up.

Apparently, no methane was supplied to the chamber from below, probably as a result of the negative pressure at depth.
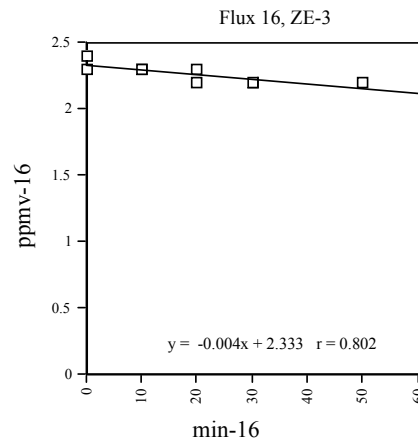
Fig. 12. Evolution of the CH$_4$ concentration in the chamber as a function of time

3.2. The oxidation of the methane throughout the cover

To calculate the fraction of methane oxidised in the soil cover, three factors were determined: (i) α the fractionation factor, which expresses the more rapid consumption of the lighter isotope of methane, $^{12}CH_4$ relative to $^{13}CH_4$, (ii) the isotopic composition of methane in the anoxic zone where it is produced and exists prior to exposure to oxidation (δA, equation 2) and (iii) the emitted methane (δE, equation 2) which has been exposed to oxidation. Emitted methane was sampled in two ways, by collecting it in chambers and by collecting air samples, particularly during a calm period at night at the site, and downwind of the site.

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The fractionation factor (α) was determined from the results of fig.13., to be 1.01615 ± 0.00005. This was determined by incubating landfill cover soil at ambient temperature and sampling the closed system incubations at several time periods. Alpha was calculated from the slope of the line fit to the data using equation 3.

Anoxic zone methane was sampled from vent pipes, gas extraction pipes and from bubbles effusing from cracks and along the sides of the landfill cells. Values for anoxic zone methane varied from $\delta^{13}C=$-56.1 ± 0.43 in the Ancient zone, $\delta^{13}C=$-55.62 ± 0.42 in the experimental zone,

Fig. 13. $\delta^{13}C$ (‰) vs ln (M/M$_o$).

$\delta^{13}C=$-61.72 ± 0.40 in the temporary cover zone and $\delta^{13}C=$-59.09 ± 0.23 in the fresh garbage zone. The overall mean for anoxic zone methane, determined by weighing each area equally, was $\delta^{13}C=$-57.99 ± 2.28 ‰. This value was used for δA in equation 2 for all atmospheric samples while δA values specific for each area were used in the calculation for chamber values.
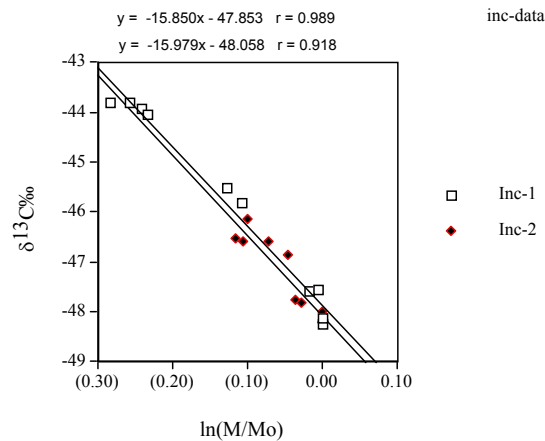
The percent oxidation values obtained from the air sample transects ranged from 0 (including negative numbers as 0) to 35%. Since the largest negative flux were around 10%, one might regard this as the accuracy of the method, ± 10%. If so, then landfill cover soil oxidation, as determined by this method was essentially 0%, except in samples Transect 4-7 and 4-8 which were collected near the temporary covered area. Here oxidation values of 20 to 35% were obtained. The percent oxidation as determined from chamber emplacements varied from 0 to 58%. Again, the only significant oxidation was measured at the temporary covered area, consistent with results obtained from the atmospheric sampling.

It appears that in winter, at this landfill, oxidation is confined to the temporary soil cover and is absent in the other zones.

In comparison with other landfills, it has generally been observed that the occurrence of CH_4 oxidation varies seasonally, being lower or absent in winter and greater in summer (Chanton et al., 1999, Chanton and Liptay, 2000, Börjesson et al., submitted, Börjesson and Svensson, 1997, Boeckx et al., 1996). Summer values suggest ratios between oxidized and produced CH_4 in landfills at around 25-30 % in the northeastern USA (Liptay et al., 1998), 25 to 35% in Florida, USA (Chanton and Liptay, 2000), and 39-53 % in Germany (Bergamaschi et al., 1998). This has been interpreted to indicate that oxidation takes place only in the surface soils which are temperature sensitive and not deep down in the profile. The lack of CH_4 oxidation at low temperatures has implications for budgets of CH_4 turnover in landfills, which on a global scale should include a latitudinal factor.

4. Conclusions

The results clearly show that landfill cover and gas recovery reduce drastically the net emissions of methane toward the atmosphere. The ratio net emissions on the cover zone with LFG collection/net emissions on the operating zone at the Montreuil/Barse landfill is 98%. The temporary shut-down of the biogas recovery doesn't lead to more major gas leaks but did

double the flux. The use of the static chambers permitted to locate the leakage zones. More attention should be paid then to the cracks, fissures, vents and cells' borders, which are the major pathways of emitted methane. Apart these special areas, fluxes of methane are negligible, and even atmospheric methane take up has been observed. The thermal anomalies detected didn't systematically correspond to emissions of methane but they are mainly linked to water saturated zones or different thermal transfers following cover configurations. The oxidation rates have been calculated using the fractionation factor measured with the site configuration ($\alpha = 1.01615$). It appears that in this winter conditions, a zero oxidation rate occurs in the areas with LFG collection. In the temporary zone, where there is no LFG recovery, the oxidation rate calculated was 35%. That means that in the covered zone, the soil has the bacterial potential to oxidise the methane but the *in situ* conditions (not enough methane coming through the cover, a water saturated soil or too cold temperatures) were not optimised.

There exist some management policies that could lead to reduce the emission of methane such as limiting the size of the cell, setting up a temporary cover as quick as possible even for a short period, making the discontinuities more water and airtight or recovering the biogas even to flare it.

These first results permitted to obtain a set of original data, which shows that present landfill management with cover and LFG recovery reduce the emissions of methane toward the atmosphere. This first measurement campaign will be completed by measurements under drier and warmer conditions and on the same configurations plus on a cell with geomembrane.

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