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ESTIMATING METHANE EMISSION AND OXIDATION FROM TWO TEMPORARY COVERS ON LANDFILLED MBT TREATED WASTE

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SUMMARY: Surface gaseous emissions and upper layer waste gas composition were measured on two French MBT plants with aerobic pre-treatment process. The goals were to characterize the gaseous emissions, and to assess the efficiency of the upper layer to oxidize the methane flux coming from the residual organic fraction. The first plant was operated without recovery of organic fraction and with concentration of the fine fraction in a cell. The methane fluxes were high and the oxidized methane fraction was moderate (40%) for selected point measurements. On the contrary, the second plant organic fraction was recovered. The residual methane emissions were low with a high fraction of oxidized methane (up to 80%). These results were obtained with favourable humidity and temperature conditions.

1. INTRODUCTION

Landfill methane emissions must be reduced in order to limit the global warming impact of a sustainable landfill. The decline of the quantity and quality of the landfill biogas (LFG) produced after the final phase of landfill capping needs to consider natural oxidation processes in order to mitigate the residual methane emissions. The results presented in the past Sardinia Symposia showed that mature compost could be used for the building of the biocover. Moreover, high methane oxidation efficiency (nearly 100% for a methane supply of 4 - 4.5 l/m²/h) was measured with column tests using old municipal solid waste material (Huber-Humer & al, 2007, 2008).

Another result of these studies is the possible improvement of the methane oxidation efficiency with the addition of structural compounds and waste sieving/homogenisation. MBT processes produce waste with high porosity homogeneity and therefore the MBT processed wastes are good candidate for the building of a biocover.

Due to the lack of data concerning the real fugitive methane emissions of this type of waste in the windrows and on the temporary and final covers, INERIS conducted from 2006 several surveys on different French landfills. The first part (2006 - 2007) of this study focused on the temporary covers, which generally produce a significant amount of fugitive methane emissions for landfills without waste pre-treatment (Tarimini & al, 2003; Ackermann & al, 2006, 2007).

The second part on this program deals more specifically about the efficiency and building materials of biowindows and is represented by on-going research.

The results presented in this paper are a part of a research program dealing with the comparison of gaseous emissions (Zdanevitch & al, 2009) and assessment of methane oxidizing potential of different types of covers and waste storage.

The results concern two MBT plants without methane recovery (plant A and plant B). The surveys were realized during the end of spring of 2006 and 2007.

2. MATERIAL AND METHODS

2.1 Methodology

The surveys conducted on the two MBT plants combine methane and carbon dioxide surface fluxes, methane and dioxide concentration measurements of gas collected in the shallow waste layer, and humidity measurements. The assessments of the oxidized methane fluxes used a LFG mass balance method considering the surface fluxes and the ratio between the methane and LFG concentrations measured in the waste (Christophersen M. & Kjeldsen P., 2000).

The waste LFG concentration measurements were realized every two surface fluxes measurements in a second step, after the surface flux measurement. The gas probe was inserted in the waste and allowed to sample gas at a depth of approximately 75 cm (Figure 1). The methane load before oxidation was determined by the methane fraction in the gas sample pumped from the gas probe and by the LFG (methane and carbon dioxide) surface fluxes and can be described by the equation (1) from Christophersen & al (2000) :

$$J_{CH4inf} = (J_{CH4sup} + J_{CO2sup}) * \left\{ \frac{C_{CH4inf}}{C_{CH4inf} + C_{CO2inf}} \right\} \quad (1)$$

where J_{CH4inf} and J_{CO2inf} are the methane and carbon dioxide loads before oxidation, respectively, and J_{CH4sup} and J_{CO2sup} are the methane and carbon dioxide surface fluxes, respectively. From the equation (1), methane oxidized flux J_{ox} is calculated by the difference between J_{CH4inf} and J_{CH4sup} .

This method supposes that the ratio between the surface gas concentration in the chamber and the gas pumped from the probe is only influenced by the methane oxidation. The repartition of the carbon dioxide fluxes must be checked in order to verify that the major source of carbon dioxide is linked with the LFG sources in the shallow waste deposit located near the surface.

In order to maximize the accuracy of this method, a selection of the measurements points based on the assessment of the relative error was made. The filtering of the data provided by this method reduces the local and global uncertainty of the methane oxidized surfaces fluxes determination.

The assessment of the relative error concerning the oxidized methane flux was determined by the equation (2)

$$\delta J_{ox}/J_{ox} = ((\delta J_{CH4inf}/J_{CH4inf}) * J_{CH4inf} + (\delta J_{CH4sup}/J_{CH4sup}) * J_{CH4sup}) / (J_{CH4inf} - J_{CH4sup}) \quad (2)$$

where $\delta J_{ox}/J_{ox}$ is the relative error of methane oxidized flux J_{ox} , and $\delta J_{CH4inf}/J_{CH4inf}$ and $\delta J_{CH4sup}/J_{CH4sup}$ are the relative errors of the methane load charge and methane surface flux, respectively.

2.2 Surface fluxes measurements and gas sampling

The surface fluxes measurements were performed with a static chamber with recirculation. The upper part of the chamber has a fan to mix the gas, and two sampling tubes. The base of the

chamber was sealed into the waste. The sampling gas was pumped into two analysers, a portable flame ionization detector (Telegon FID, AutoFIM II) and an infra red (IR) absorption detector (RSDynamics, Ecoprobe 5). The probe was hammered to the depth of 750 mm, in order to avoid the dilution with atmospheric gas. Then, the gas was pumped and analysed by the IR detector.

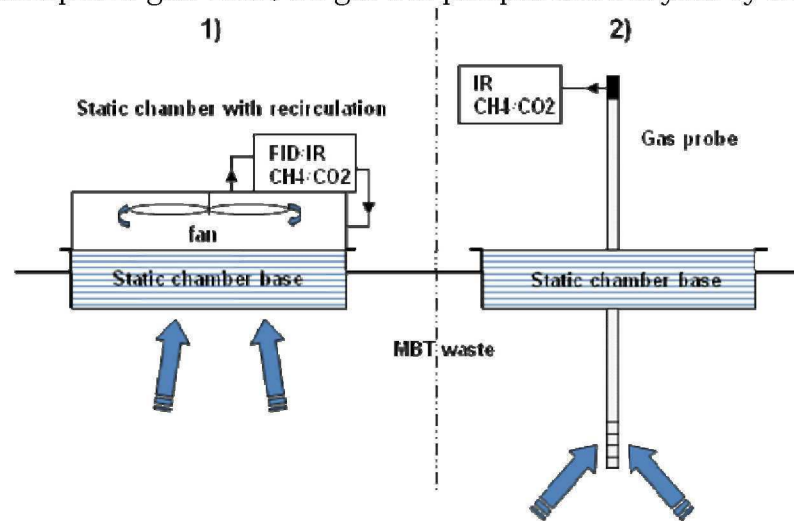


Figure 1. Two steps of the measurements of the methane oxidation in the upper layer of the MBT processed waste storages

3. EXPERIMENTAL RESULTS

3.1 MBT plant without organic fraction recovery

For the MBT plant A, the fine fraction which was sieved at a 70 mm mesh and the processed fraction which was sieved after pretreatment at a 50 mm mesh join aerated windrows for 6 weeks. After a passive maturation of 3 - 7 months, the compost was stored in the final storage cells. Each 6 months, a new 3 meters layer of composted waste was deposited on the storage cell.

3.1.1 Methane and carbon dioxide surface flux and concentration measurements

Two surveys were conducted on the first MBT plant A, totalizing 71 surface fluxes measurements. Ordinary kriging was used to interpolate the global flux from the punctual measurements. The global methane flux observed on the surface was high ($18 \text{ m}^3 \text{ CH}_4/\text{h}$) during the first survey and could be explained by the storage of a fraction which contains more organic matter than raw waste, and because aerobic treatment of waste leads to a more rapid onset of methanogenic conditions (Robinson & al, 2005).

The global carbon dioxide flux density was lower than the global methane surface fluxes ($12 \text{ m}^3 \text{ CH}_4/\text{h}$). This high ratio (1.5) between the methane and carbon dioxide global fluxes was even higher in the locality of the more recent deposit where this ratio reached 1.67. The more recent deposit locality (delineated by the isovalues 200 - 500 ml methane/min/m² in Figure 2) concentrates the methane fluxes (approximately $14 \text{ m}^3 \text{ CH}_4/\text{h}$). There is a clear relation between the locality of the high methane and high carbon dioxide surface fluxes.

This relation between methane and carbon dioxide emission location could also be observed on the samples gas from the probe (Figure 3). For each gaseous compound (carbon dioxide and methane) the concentrations generally reach 30% - 40% at a depth of 750 mm. The low concentration levels were essentially observed on the perimeter of the storage cell and could also be linked with local sealing problems of the probe in the waste and dilution of LFG collected from the gas probe.

The meteorological conditions of the first survey (June 2006) were rather good during the surface emissions measurements, on contrary of the second survey in May 2007. For the first survey, there was little rainfall before the surveys. Surface fluxes were measured on a regular grid with a grid spacing of approximately 5 m. Measurements of moisture content could be done by the sampling of only 3 liters of waste in the shallow layer on 3 different locations, due to the good homogeneity of the MBT processed fine fraction.

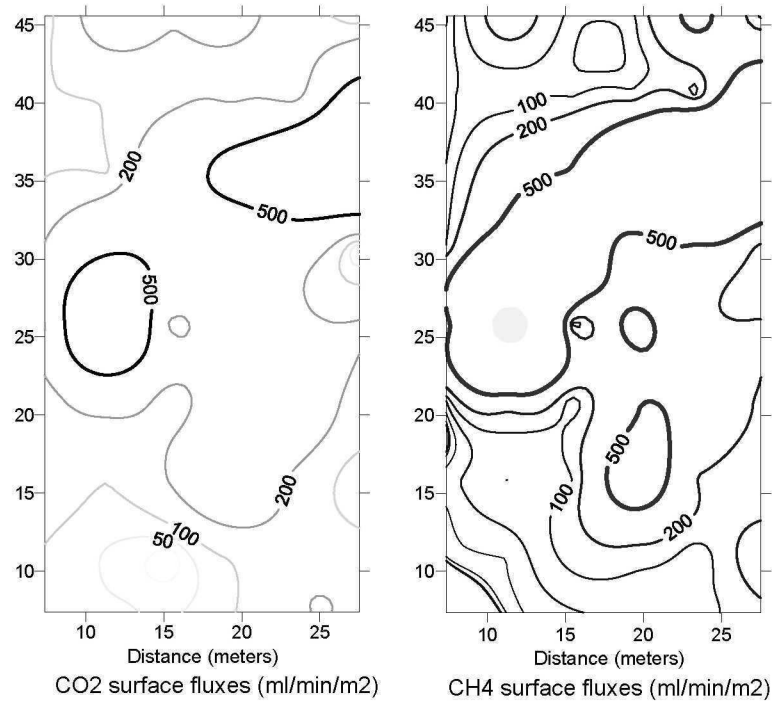


Figure 2. Methane and carbon dioxide surface fluxes for the site A

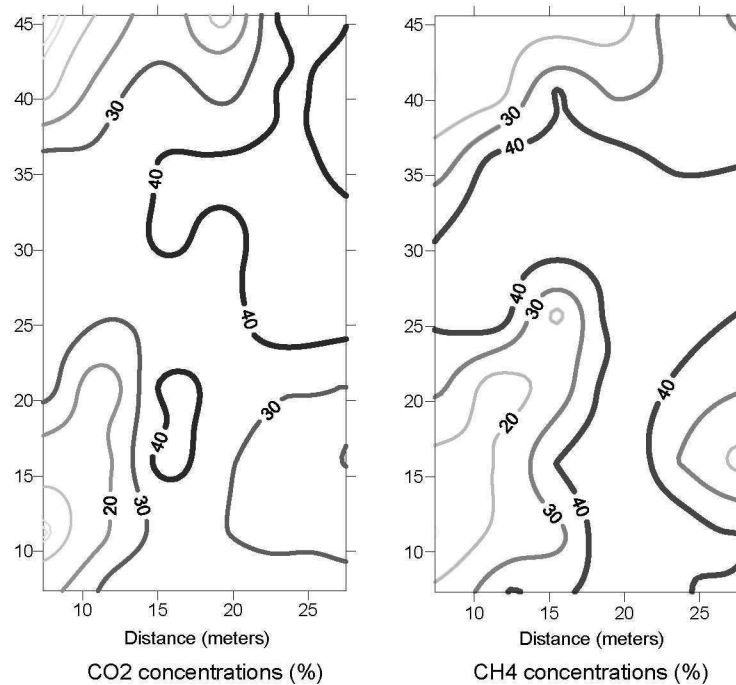


Figure 3. Methane and carbon dioxide concentrations of the gas probe for the site A

The average moisture content of this superficial layer collected from 0 – 100 mm depth, 16% (v/v), was sufficient to allow favourable conditions for methane oxidation and was also not too high to promote gas migration only in preferential flow paths.

The barometric pressure gradient was limited to 0.5 – 1 hPa/h and therefore did not interfere with the surface flux measurements. During the second survey (May 2007), the porosity of the waste fine fraction was almost saturated due to the rainy periods before the survey.

3.1.2 Assessment of the methane oxidized fraction

An average oxidative rate of 30 - 40% for the selected measurements points (18 points) was reached for the two surveys. The selected points allowed the observation of an oxidation rate for rather high (more than 200 L CH₄/m²d⁻¹) to low methane charge. This method cannot be applied for the high methane charges values of the hotspots and obviously for the near zero methane fluxes. Comparisons with similar MBT treated wastes (Einola & al., 2007) show that the oxidizing rate could reach 80% with the same methane charge for columns laboratory tests. The observed in situ methane oxidation potential seems to be limited to a value of approximately 70 L CH₄/m²d⁻¹ on this site for favourable temperature (25°C) and humidity conditions (Figure 4).

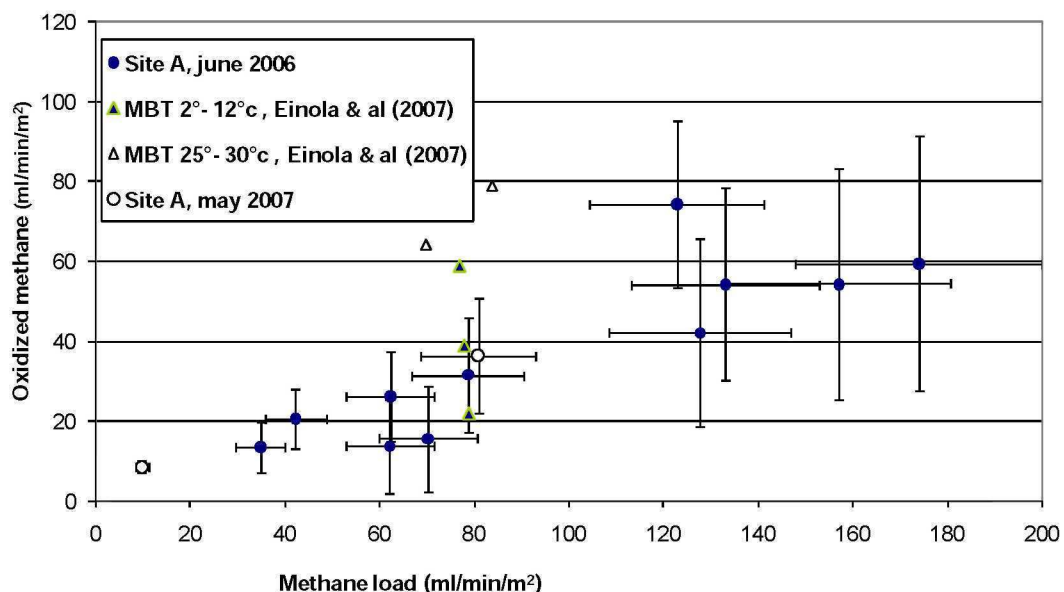


Figure 4. Comparison of oxidized methane fluxes for MBT processed waste from laboratory (Einola & al., 2007) and in situ measurements with relative errors bars

3.2 MBT site B with organic fraction recovery

The second MBT plant stores only the refuses of sorting before the process, and therefore the organic content is strongly reduced in the landfill cell. The treatment combines an aerobic pre-treatment (3-4 days) and a sorting process, with a final separation done at a 10 mm mesh. The fine biodegradable fraction is separated and goes to composting windrows.

3.2.1 Methane and carbon dioxide surface flux and concentration measurements

Only one survey could be conducted on the MBT plant B, before the completion of the final cover of the refuses storage. Surface flux measurements were done on the surface of the refuses which received also crushed wood coming from the refuses of the green waste composting process. The global methane flux observed on the surface was low (0.5 m³/h) and was only

observed in a few measurements points, on contrary of the carbon dioxide surfaces fluxes which were observed on a large set of the measurements points (Figure 5). Global carbon dioxide surface flux was much higher than global methane surface flux and reached $4.5 \text{ m}^3/\text{h}$. We could see a link between the locality of the high methane and carbon dioxide surfaces fluxes, and the methane surfaces fluxes was only observed in the center of the large area delimited by the isovalues $50 - 100 \text{ ml CO}_2/\text{min}/\text{m}^2$.

Gas probe concentrations measurements showed also a correlation between the locality of the high levels of methane and carbon dioxide concentrations at the depth of 750 mm . Gas migration could also occur in this shallow waste layer, with high content of plastic bags. The high spot of methane surface flux observed on the surface was at a minimal distance of 5 to 10 meters far from the methane isovalue of $5 \text{ ml CO}_2/\text{min}/\text{m}^2$. Due to the recovery of the fine fraction (organic matter) the carbon dioxide surface fluxes coming from the aerobic degradation were expected to be low.

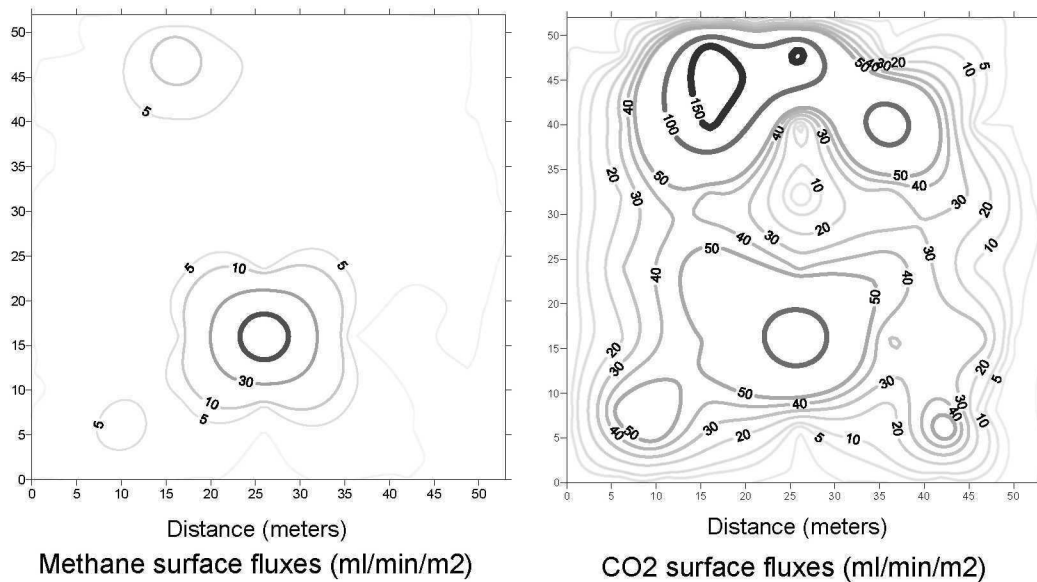


Figure 5. Methane and carbon dioxide surface fluxes for the site B

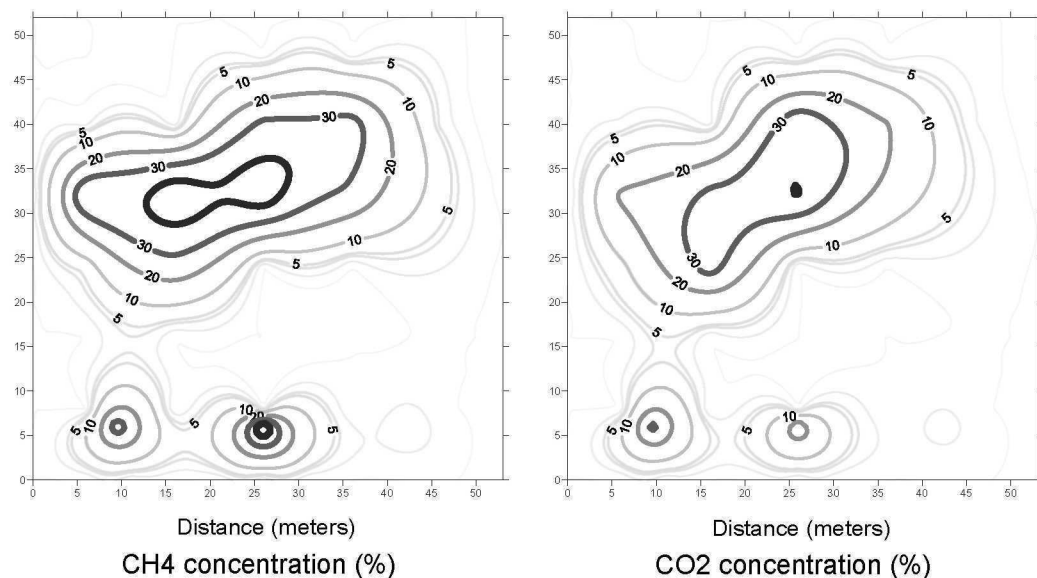


Figure 6. Methane and carbon dioxide concentrations of the gas probe for the site B

For the plant B, crushed wood was used for the completion of the final waste surface layer. Also the moisture content was only checked by a water balance calculation with the use of daily rainfall and evaporation instead of in situ or laboratory moisture measurements. Water reserves of 100 and 150 mm were used in order to estimate the free porosity for the LFG migration and diffusion in the shallow waste layer of the refuses deposit (Figure 7). The survey was done during the last week of May in 2007. The water reserve seemed to be almost full in this period but we did not observe any indications of water saturation during the survey.

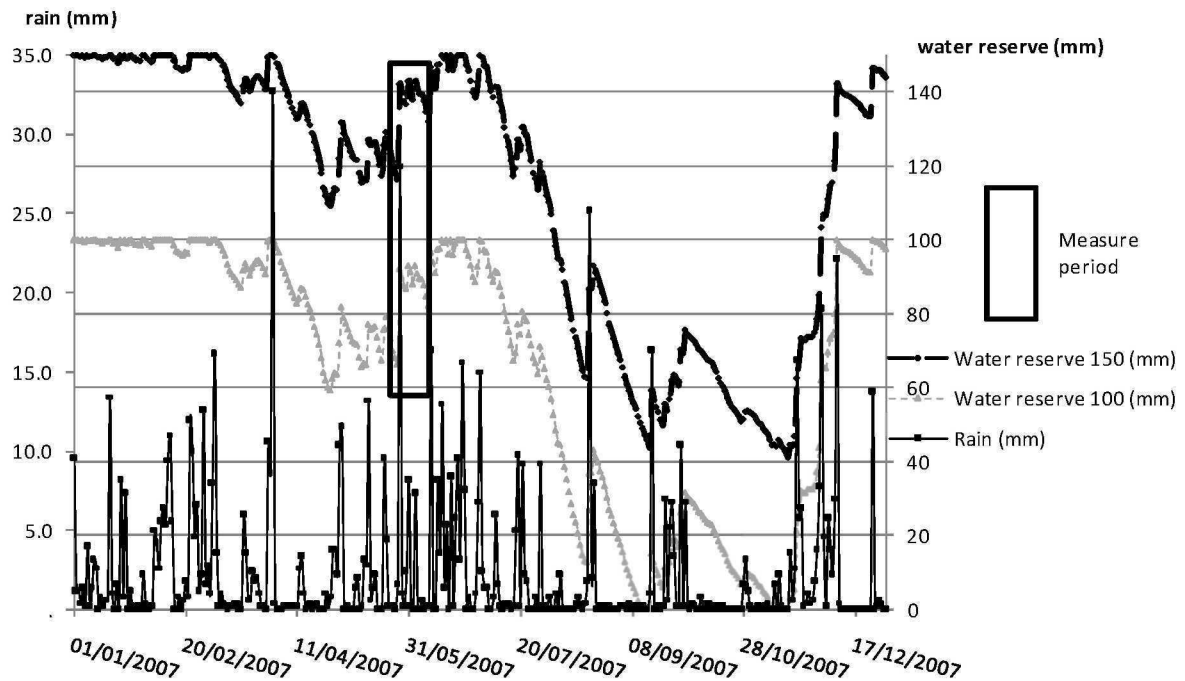


Figure 7. Water reserve assessment for the upper layer of the site B

3.2.2 Assessment of the methane oxidized fraction

The same point selection procedure as for plant A was applied to the measurements points (32 points). This method allowed estimating an average oxidizing rate of more than 80% for the temporary cover, but with average oxidized methane fluxes of only $35 \text{ L CH}_4 / \text{m}^2 \text{d}^{-1}$ on this site.

The lower load of methane and the presence of crushed wood (which seems to have similar structural properties as wood chips which are generally used in laboratory columns) probably explained this good oxidizing rate.

The filtration of the methane oxidized fluxes by the relative error calculation limits the dispersion of the results but also eliminates the low oxidized fluxes coming from larger methane loads. The selection method shifts the average of the methane and carbon dioxide fluxes observed on the whole data set. The average methane oxidized fluxes coming from the punctual measurements (Figure 8) and selection method is similar to the average oxidation rate estimation coming from an assumption of a LFG load with 60% methane and 40% carbon dioxide. Also this shift causes a slight overestimate of CH_4 oxidation.

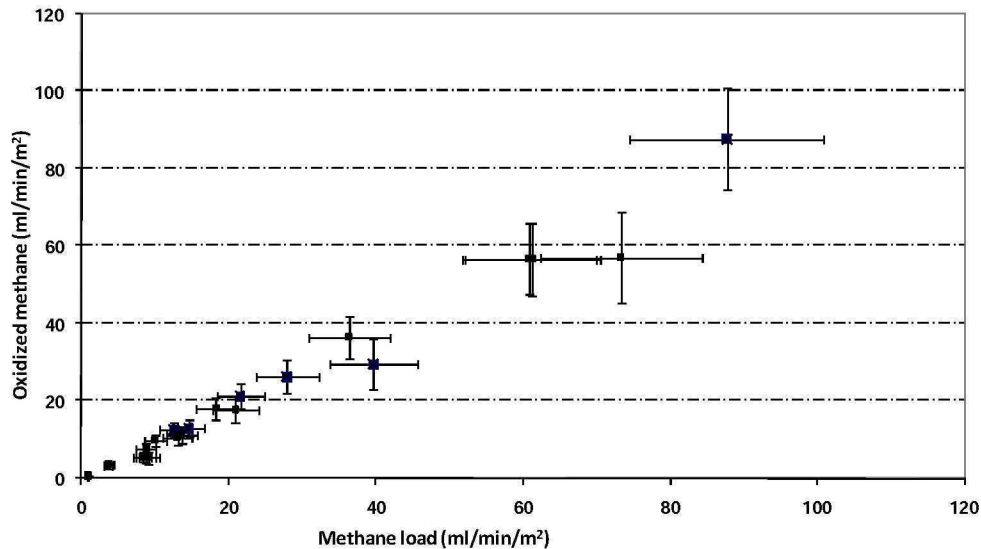


Figure 8. Oxidized methane fluxes versus methane loads for the site B with relative errors bars

4. RESULTS AND DISCUSSION

Different behaviours (Figure 9) can be observed on the two data sets provided by the methane oxidized fluxes measurements and data filtration. For the plant A, the methane load limits the methane oxidizing fraction. An asymptotic tendency observed with only 3 measurements points must be confirmed (asymptotic value of approximately 60 ml/min/m² of oxidized methane for the higher methane load). We do not observe the same limitation of the punctual methane oxidized fluxes for the plant B. Plant B cell, receiving refuses, may produce a particular longer channelling flow path in the macro pores which could explain a higher length of gas migration.

This migration seems to be observed between the centre of the areas of high methane concentration in the waste at a 750 mm depth and the surfaces fluxes hot spots. The temporary layer of the plant B was modified in the late 2007 by a final cover with a clay layer and a compost layer.

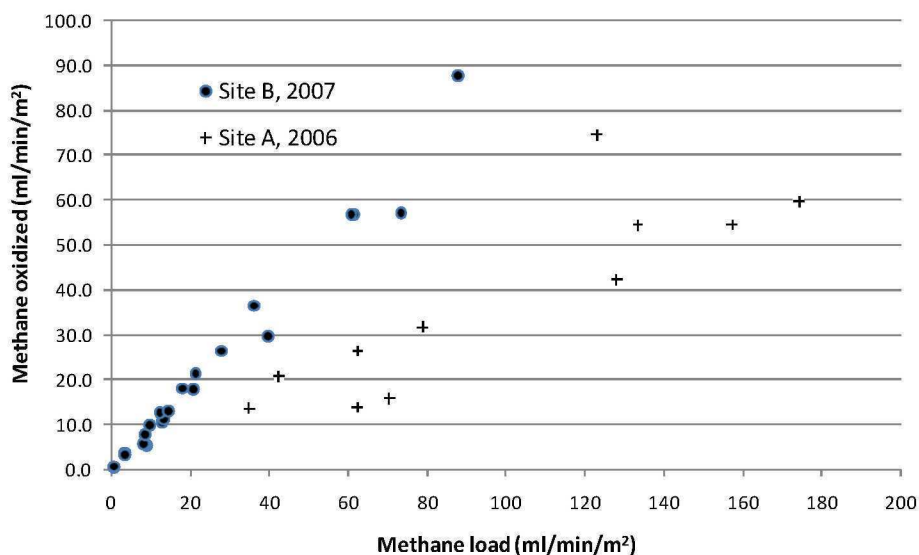


Figure 9. Comparison of the oxidized methane fluxes for the sites A and B

Improvements of the methods could be done by the measurements of the gas concentration at different depths but the survey period was limited by the time period of stable weather conditions. The requirements of the surface fluxes measurements limit the survey weather conditions (no rainfall during the survey and the days before, limited barometric gradient pressure) and therefore also the possibility to obtain new data set.

5. CONCLUSIONS

Oxidized methane fluxes measured on the site B were lower than those coming from site A, and the oxidating rate reached 80% and 40 % for a selection of the measurement points of the site B and respectively the site A. The methane load, higher for the site A, was the principal factor of the oxidation rate difference. These oxidation rates measured on the temporary cover highlight the role of methane oxidation, even for the landfilling phase before the final capping. The combination of waste aerobic pre-treatment, organic fraction recovery and methane fluxes oxidation produces very low residual methane fluxes. These results obtained for only three surveys need to be confirmed but allow extrapolation of a residual methane emission level below the value of 5 m³/ha/h for the MBT plant refuses storages with improved cover and MBT plants with organic fraction recovery. The methane emission levels of 5 and 10 m³/ha/h were proposed as guidance values for the use of biotic systems (Stegmann & al, 2003; Bour & al, 2005).

These results have been obtained in favourable conditions (medium temperature and humidity) and need to be checked for the winter weather conditions and especially different moisture conditions. The limitations of the methane oxidation were observed for humid conditions then the porosity of the superficial layer was filled with water. The very punctual LFG emissions under these conditions could hardly be measured. The accumulation of LFG below this water saturated layer probably leads to a drastic reduction of the oxidation potential. The ongoing research will focus on the optimizing of the bio covers and biowindows for the different climatic conditions with the participation of INERIS in the METHALIX program funded by the French agency for the energy and the environment.

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