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▶ To cite this version:

Olivier Bour, Isabelle Zdanevitch, Zbigniew Pokryszka, A. Lalet, A. Akerman. Temporal variation of LFG emission from different types of landfill cover. 14. International Waste Management and Landfill Symposium (Sardinia 2013), Sep 2013, Cagliari, Italy. pp.NC. ineris-00976248

HAL Id: ineris-00976248 https://ineris.hal.science/ineris-00976248

Submitted on 9 Apr 2014

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TEMPORAL VARIATION OF LFG EMISSION FROM DIFFERENT TYPES OF LANDFILL COVER

O. BOUR*, I. ZDANEVITCH*, Z. POKRYSZKA*, A. LALET** AND A. AKERMAN**

* INERIS, Parc ALATA, 60550 Verneuil-en-Halatte, France ** SITA FRANCE, Tour CB21, 16 place de l'Iris, Paris La Défense, France

SUMMARY: A research project "OSSIMED" funded by the French Environment and Energy Management Agency (ADEME) started in 2011. The main goal was to identify the amplitude and importance of the methane and carbon dioxide emissions variations observed on a landfill site and to give hints for a more robust assessment of the global greenhouse gas emission of a landfill site in operation. Nine monitoring areas were selected on 3 types of cover (daily, intermediate and final cover) of a pilot site and different time scales were considered in order to allow the LFG emission monitoring on an hour, day, week and seasonal basis. The whole methane flux assessment conducted with the LFG surface fluxes variations monitoring for each area allows to precise the diffusive and convective fraction of the methane fluxes for each type of cover. These results explain the difference between the high oxidation rates observed for the diffusive fluxes and the medium oxidation rate observed for the whole methane emissions.

1. INTRODUCTION

Control and reduction of fugitive emissions of methane is one of the challenges of the landfilling of Municipal Solid Waste (MSW). This reduction of the landfill gas (LFG) emissions requires the ability to measure low methane emissions and to use the results in order to evaluate these diffuse emissions on an annual basis. Moreover, the experience of low emission measurements made at INERIS demonstrates that the variations of methane fugitive emissions are even more pronounced than these emissions are low. The uncertainty of the measurements thus tends to increase with the reduction of emissions, and this difficulty is observed for all measurement methods. Factors such as the duration of the lag between the maintenance of the LFG network and the emissions measurement periods, as well as changes in climatic parameters also strongly influence methane emissions.

A research project "OSSIMED" funded by the French Environment and Energy Management Agency (ADEME) began in the year 2011. The main goal was to identify the importance and the amplitude of the LFG emissions variations from a landfill site in operation and to give hints for a more robust assessment of the global greenhouse gas emission from MSW landfill sites. The goals were also to observe the LFG fluxes variations for the principal types of cover used during landfill operations.

The study was conducted on a pilot site which uses a temporary cover for the exploitation area. Nine monitoring areas were selected on 3 types of cover (daily, intermediate and final cover) and the measurements were made with different time frequencies in order to allow the monitoring of the emissions on an hour, day, week and seasonal basis.

2. METHODOLOGY

2.1 Methodes used for localisation and determination of the gas fluxes.

In this study, we use a combination of two methods for localisation and determination of the gas fluxes, in order to have a better coverage of the surface.

The methane concentration measurements were made with FID (Flame Ionisation Detector) and Inspectra® methane analysor (laser spectroscopy technology using a laser diode).

The scanning of the methane emissions has two goals. The first goal was to give a first view of the distribution of the methane emissions and also to give the hints to sort the convective or diffusive emissions. The presence of the cracks was generally associated with convective fluxes or rather thin clay covers. In the case of the pilot site, the convective methane emissions were observed only near the landfill gas (LFG) drainage wells. These locations could be explained by a sufficient humidity of the cover.

2.1.1 INERIS static chamber method

The INERIS static closed chamber was used in order to measure precisely gas flux and to give reference values for the modified ISM method. The INERIS static chamber method consists in setting a box of $0,25 \text{ m}^2$ ground surface over a typical representative surface of the site with a recirculation of the gas. This method gives local flux values that need to be extrapoled to estimate the whole emissions.

In the closed chamber, the increase of methane and carbon dioxide was monitored by two analysors (Inspectra® and Ecoprobe®). The slopes of the curves give the flow rate of methane and carbon dioxide coming in the box. This method gives on the laboratory a precision and a repeativity better than 5%. In the field, the quality of the sealing of the chamber on the surface and the relative low velocity of the winds are the major parameters for a good reproductibility of the measurements.

The procedures involved in this method are protected by a French patent (No. 96-05996,filed on May 14th 1996) and an European invention patent (No. EP0807822B1) under the heading "Measurement of gas flows through surfaces". In practice, this method has been used for over 15 years for landfills, polluted soils and closed mines to measure the flux of gaseous hydrocarbons (Pokryszka and Tauziède, 2000) and CO₂ emissions (Pokryszka & *al.*, 2010).

A specific design of the INERIS accumulation chamber and of the ventilation was tested in order to allow high frequency flux measurements (more than 2/hour). This design was used to monitor fluxes from an hour to a week time scale, in order to observe the influence of different physicals parameters.

Due to the sealing of the chamber on the ground surface, the measurements were not as fast as the use of a simple chamber. Also, this type of measurement was only used for the locations with methane fluxes.

2.1.2 Modified ISM method (Instantaneous Flux Measurement)

The methane emissions measurements used a modified instantaneous surface measurements

(ISM) methodology in order to evaluate the whole gas fluxes of each cover type.

A gas probe was used with a simple closed chamber in order to give a fast measurement of the instantaneous flux (20 s) in order to give an Instantaneous Flux Measurement (IFM).

The correlation between the IFM fluxes and sealed static chamber with recirculation fluxes was significantly better than the correlation between ISM concentrations and the sealed static chamber with recirculation fluxes.

Contrary to the reference method (static chamber sealed with recirculation), the ISM and IFM results need a specific calibration and are more sensible to the variations of the wind velocities.

The correlation between the IFM and the INERIS chamber reached a value of 0,95 (r^2) for a mixed diffuse/hot spot emissions with good surface flatness (for final cover). In the case of the pilot site and monitoring of the 3 types of cover (temporary, intermediate, final), the coefficient of correlation was in the range 0.8 – 0.87.

The density of the measurements was adapted to the type of emission: the base density was 100 IFM points/ha with a refinement of the measurement mesh near the convective fluxes. In the specific case of the LFG drainage well, the distance between the measurement points could be as low as 50 cm, with a number of IFM points for each LFG drainage well which could reachs 30 IFM points.

As we can see later, the convective fluxes associated with the LFG drainage well could be more important than the diffusive fluxes: the duration of the measurements was finally a function of the type of cover and the number of LFG drainage wells.

The same two analysors (Ecoprobe[®] and inspectra[®]) were used with the IFM method and the static chamber sealed with recirculation for a selection of 10 to 20 points for each cover and survey.

2.2 Distinction between convective and diffusive fluxes

During the last years, different authors have presented the relation between the methane load of the cover and the rate of methane oxidation. Methanotrophic bacteria are ubiquitous: we can use the assumption that all the diffusive fluxes exhibit a significant methane oxidation if the methane load generates a methane surface flux lower than 150 g/m²/day.

For the diffusive fluxes measurements, the CH₄/CO₂ ratio was significantly lower than 0.6 Finally two criterias help to distinguish the methane diffusive fluxes:

- ratio CH₄/CO₂ << 0.6;
- methane fluxes $< 150 \text{ g/m}^2/\text{day}$.

3. RESULTS

3.1 Assessment of the convective/diffusive emissions

3.1.1 Temporary cover

This area received the "fresh" waste and have been covered each end of week by a layer of sand (30 cm). Due to the landfill exploitation, the duration of this type of cover was limited (half a week). The spatial and statistical distribution of the LFG emissions were without large heterogeneities: the maximum of the methane flux (61 g/m²/day) do not reachs a value of one order of magnitude more than the methane flux arithmetic mean (8 g/m²/day). The whole methane flux reach a value of 1 m³/h for the 2 200 m² of this cover surface.

3.1.2 Intermediate cover

Two cells were investigated. The cover texture of these two cells was similar to a sandy clay soil. The thickness of the cover was approximately 50 cm. The duration of this cover is several months to one year. Due to the presence of clay, the gas fluxes were observed only on 10% of the measurement points. We observed "hot spots" points with a methane maximum flux of $180 \text{ g/m}^2/\text{day}$ and an average methane flux of $4 \text{ g/m}^2/\text{day}$. The diffusive fluxes were highly reduced, but the convective fluxes represent an important part of the fluxes. The mixing of the diffuse and convective fluxes was observed on each cell. The whole methane flux reach 1 m^3 /hour (for a surface of 5 300 m²).

3.1.3 Final cover

Three parts of the pilot site have a final cover. In one part, the thickness of the clay layer of the cover was very high (7 m). In this part no diffuse emissions occurred. We could only observe low methane emissions on one side of this area and near a LFG well. Only one part of this particular cell was investigated. If we extrapolated the convective flux observed for the whole area (12 ha), we estimate a convective flux of approximately 6.3 m³ methane/hour for this area. The others two parts have a cover with a clay layer which thickness vary between 50 cm and one

meter. The average diffusive fluxes were very low (approximately 1 to 1.4 g CH₄/m²/day). The average convective fluxes were in the range 1.7 to 4.7 g CH₄/m²/day. The whole methane fluxes of these two parts reach a value of 15 m³/hour.

3.1.4 Emissions of methane of the pilot site

The pilote site has also an area with a geomembrane. In this particular area the fluxes were convective near the gas collection system but low (< 0.5 m^3 /hour). The whole pilot site methane emissions reach 24 m³/hour, for a surface of 28 ha. The fraction of the convective fluxes of the pilot site was high and reached approximately 13 m³/hour. We could observe that one half of the whole methane emissions was emitted by convective fluxes, with very low methane oxidation percentage. On the contrary, with assumptions concerning the quality of the LFG, the oxidation rate of the methane diffusive emissions was higher than the value of 10% to 25% generally used for the assessment of the methane emission on the annual basis.

The oxidation rate could not be assessed precisely for each cover without assumptions concerning the LFG composition for each area of the pilot site.

3.2 Variation of the LFG emissions

The LFG emissions mean values and variations observed on individual monitoring points were compared with the assessment of the methane emission for each type of cover (IFM methane measurements).

The LFG flux variations could be observed at different time scales. The temporal scale of a few days or a one-day basis represents the time scale of a flux survey. The flux variations observed during this time scale could modify the primary data for the fluxes assessment.

3.2.1 Temporary cover

The variations of the fluxes were observed for three points (H1, H2 and H3) located on new emissions points selected during the methane emission scanning of each new temporary cover. For a time scale of a few hours, we observe in the Figure 1 more variations for the higher fluxes. The CH_4/CO_2 ratio was low and the fluxes could be considered as diffusive fluxes for this criteria. Due to the fresh waste deposit, the stage of the stabilized methanogenesis could not be

reached, also the criteria of CH_4/CO_2 ratio was more difficult to use for this particular case. Nevertheless, methane emission fluxes were low for the entire period of monitoring.



Figure 1. variations of the methane and carbon dioxide fluxes on a few hours time scale.



Figure 2. comparisons between the average fluxes of the selected points and the methane IFM average flux for the specific period (26/9/11 12H - 18H)

The average methane flux coming from the IFM scan (8 g/m²/day) was in the range of the monitoring points methane fluxes $(2.3 - 22 \text{ g/m}^2/\text{day})$.

If we observe the emission variations for a period of severals days, the flux variations are generally in the range of one order of magnitude from the average value.

3.2.2 Intermediate cover

The selected three points (I1, I2, I3) could be monitored for a longer period for this type of cover. If we observe the variation of the methane and carbon dioxide fluxes (Figure 3), we can see that the punctual methane fluxes variations reach more than one order of magnitude for a time scale of a few days. The variations of the carbon dioxide flux were lower.

Due to the mixing of diffuse and convective fluxes, the average fluxes of the selected points were much higher than the IFM average fluxes. The CH_4/CO_2 ratio was also generally too high for diffusive fluxes.

If we consider the variations of the particular point I3, we could observe a correlation between





Figure 3. variations of the methane and carbon dioxide fluxes on a few days time scale.



Figure 4. comparisons between the average fluxes of the selected points and the methane IFM average flux for the specific period (27/9/11 - 30/9/11)

3.2.2 Final cover

In this particular case, the three selected points (F1, F2, F3) were located on two area with a clay layers of approximately 50 to 100 cm thickness. These monitoring points exhibit very high flux amplitude, which could reach more than 4 orders of magnitude for one point (F1).

We can observe a mixing of diffuse flux for the F2 and F3 points with high convective fluxes on point F1. Methane was only release on the point F1: The spatial concentration of the methane fluxes ("hot spots", point F1) explains the difficulties to extrapolate the results. Methane diffuse emissions were very low (F2, F3 points), but a few hot spots modify significantly the whole methane emissions.



Figure 5. variations of the methane and carbon dioxide fluxes on a few days time scale.

4. CONCLUSIONS

One goal of this study was to distinguish the real diffusive emission from the semi convective emission coming from the cover cracks and from the surroundings of the LFG drainage wells. If methane diffuse emissions could be evaluated by a 1D diffusion model (Bogner. J. et al, 2010) and could also be evaluated with surface fluxes measurements and geostatistical tools, the fraction of local convective emission is more difficult to assess. On the pilot site the methane convective fraction is higher than observed in previous studies, and reachs more than 50% of the whole site methane emissions. The presence of low permeability engineered covers is the main reason of the predominance of this type of emissions. Also one of the challenges of this ongoing "OSSIMED" project is to study this more important convective fraction of the methane emissions by high density IFM measurements.

AKNOWLEDGEMENTS

To Fabrice Richez, Francis Guillot, Gaetan Bentivegna who have conducted the flux measurements in two teams, in order to allow the monitoring of 4 flux locations in the same time.

We thank ADEME and the French Ministry of Environment for the funding of the project (OSSIMED) and SITA France for the use of their landfill sites.

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