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SOFTWARE DEVELOPMENT OF A MULTISTEP WORKFLOW FOR ASSESSING LANDFILL GAS SURFACE EMISSIONS FROM SAMPLING DESIGN TO GEOSTATISTICAL MODELING

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SUMMARY: INERIS and ISSeP have been working together on methodological aspects related to the assessment and the cartography of fugitive landfill biogas fluxes, in collaboration with KIDOVA and Ephesia who provided expertise on geostatistics. In order to obtain more reliable assessment of global and local landfill gas (LFG) surface emissions through landfill covers, Instantaneous Flux Measurement (IFM) as well as a two step sampling strategy have been used. This sampling strategy has been implemented by ISSeP and INERIS and compared favorably with other strategies. A multistep approach has been devised by ISSeP, INERIS and KIDOVA as a workflow to design sampling plans, to carry out data processing and to estimate local and global LFG emissions using statistical and geostatistical methods. Data coming from different steps of a case study were compared in order to provide the necessary information about methane surface emissions.

1. INTRODUCTION

Quantifying GHG emissions of landfills and identifying local sources can be done by using different approaches that involve different sampling strategies, different types of data and flux measurement devices, and different estimation methods. Each approach has advantages but also drawbacks.

In order to better understand the different approaches and the performance of different flux measurement devices to quantify LFG emissions, Field comparison tests have been made in the past (Savanne and al, 1995, Babilotte and al, 2010). Especially, tracer technics and flux box methods have been used in a large number of projects, but with the feedback that difficulties are faced for handling hot spots and, more generally, addressing sampling design.

These reasearch works, associated with field experience, led to the conclusion that the major difficulty results from the low footprint of point measurements and the risk of flux underestimation

(Börjersson and al, 2000). Such a bias is well known and mainly explains the poor precision of estimations only based on flux measurement data.

In order to reduce the uncertainty and to increase the chance of capturing all LFG sources, different approaches have been studied to obtain more reliable assessment of global and local LFG surface emissions through landfill covers. These include fast direct or indirect emission measurements using appropriate sampling strategies.

The use of faster methane measurements in air compared with time consuming surface flux measurements has been proposed, in order to overcome the low footprint of point measurements by performing a higher number of concentration measurements.

An increased number of measurements has been proposed by different authors, some of them using others parameters correlated with LFG fluxes like soil moisture (Spokas and al, 2003).

High-density Instantaneous Surface Monitoring (ISM) was used and correlated with dynamic flux chamber measurements (Fécil and al, 2003, Franzidis and al, 2008) and funnel flux chamber measurements (Franzidis and al, 2008).

Another significant improvement is about the sampling strategy that includes specific local sampling patterns to increase the density of measurement points in high LFG emission areas.

Specific contributions of cover discontinuities to the local and global emission assessment have been observed in the past. The fugitive emissions coming from LFG collection wells can largely contribute to overall emissions, either because of the difficulty to ensure an efficient sealing at the pipe connections or between the well itself and the covers in its close vicinity. Previous studies showed that the contribution of local fluxes coming from the vicinity of wells could reach 30 to 100 % of the total flux of the landfill (Bietlot and al, 2013, Bour and al, 2013). Consequently, the identification of emission sources must necessarily be performed before to measure fluxes in order to decrease the risk of underestimating fluxes.

The identification of such very local and timely LFG flux sources must be performed in the most integrated media, i.e. the air just above the surface of the landfill, through simple concentration measurements. In this sense, methane leaks in the surroundings of gas wells must necessarily be reduced to avoid masking weakly diffusive areas on the surface of the landfill.

About data processing, the main difficulty lies in the large proportion of low flux values and consequently in the highly skewed distribution of flux data.

A combination of appropriate measurement technics and data processes seems also be the way to better assess CO₂ and CH₄ fluxes coming from the cap of a landfill.

This article presents the multistep approach that has been implemented as a software workflow to address the sampling design and estimation issues related to landfill gas emission assessment. After a brief review of the measuring devices used to collect the different types of data required by the approach, the three parts of the workflow are presented. The application of the workflow is then illustrated on an actual landfill case study and the results are discussed.

2. BIOGAS EMISSION DATA AND ON-SITE MEASURING DEVICES

The accuracy of local and global landfill gas emission assessment strongly depends on the available data and the way they are spatially distributed. Deciding about the number, type and location of data is the purpose of the sampling design part of the workflow.

The recommended sampling strategy involves a two-phase approach. In the first step, a LFG concentration emission survey is carried out in order to identify the areas where LFG is emitted. The concentrations are generally measured close to the soil surface, between 5 and 7 cm from it (Rosevear and al, 2004). In the second step, high density Instantaneous Flux Measurements (IFM) are performed, using both a simple bell probe and a surface flux measurement chamber equipped with a gas recirculation system. At each survey step, one or two concentration measuring devices

may be required to cover the possible range of concentration values. ISSeP and INERIS generally use two analyzers: Inspectra® laser device (TD williamson) and Ecoprobe ® infrared analyzer (RSdynamics). The linearity of the response of the Inspectra and Ecoprobe analyzers has been successfully controlled (for methane concentration > 5 ppmv (Inspectra) and > 300 ppmv (Ecoprobe)).

The INERIS static flux chamber allows precise gas flux measurements and to provide reference values that are used to calibrate IFM measurements. The INERIS static chamber method consists in recording gas accumulation in air within a 0.25 m² area box which is equipped with a gas recirculation system and is directly put on the ground surface.

Methane and carbon dioxide concentrations are measured every second or half a second during a few minutes. The resulting concentration time-series are used for flux calculation. Fluxes are obtained from the slopes of the curves providing a linear relationship between concentration and time. Tests in laboratory show that the precision and repeatability of measurement is better than 5%.

Practically, the repetability of on-site measurements strongly depends on the tightness of the system once the flux chamber is placed on the landfill cover. Changes of soil parameters (moisture, temperature) or atmospheric conditions strongly affect flux measurements.

This procedure of flux measurements is protected by a French patent (No. 96-05996, filed on May 14th 1996) and a European invention patent (No. EP0807822B1) under the heading “Measurement of gas flows through surfaces”. This method has been used for more than 15 years to measure fluxes of volatile organic compounds (Pokryszka and Tauziède, 2000), CO₂ and methane on landfills, contaminated sites and closed mine sites.

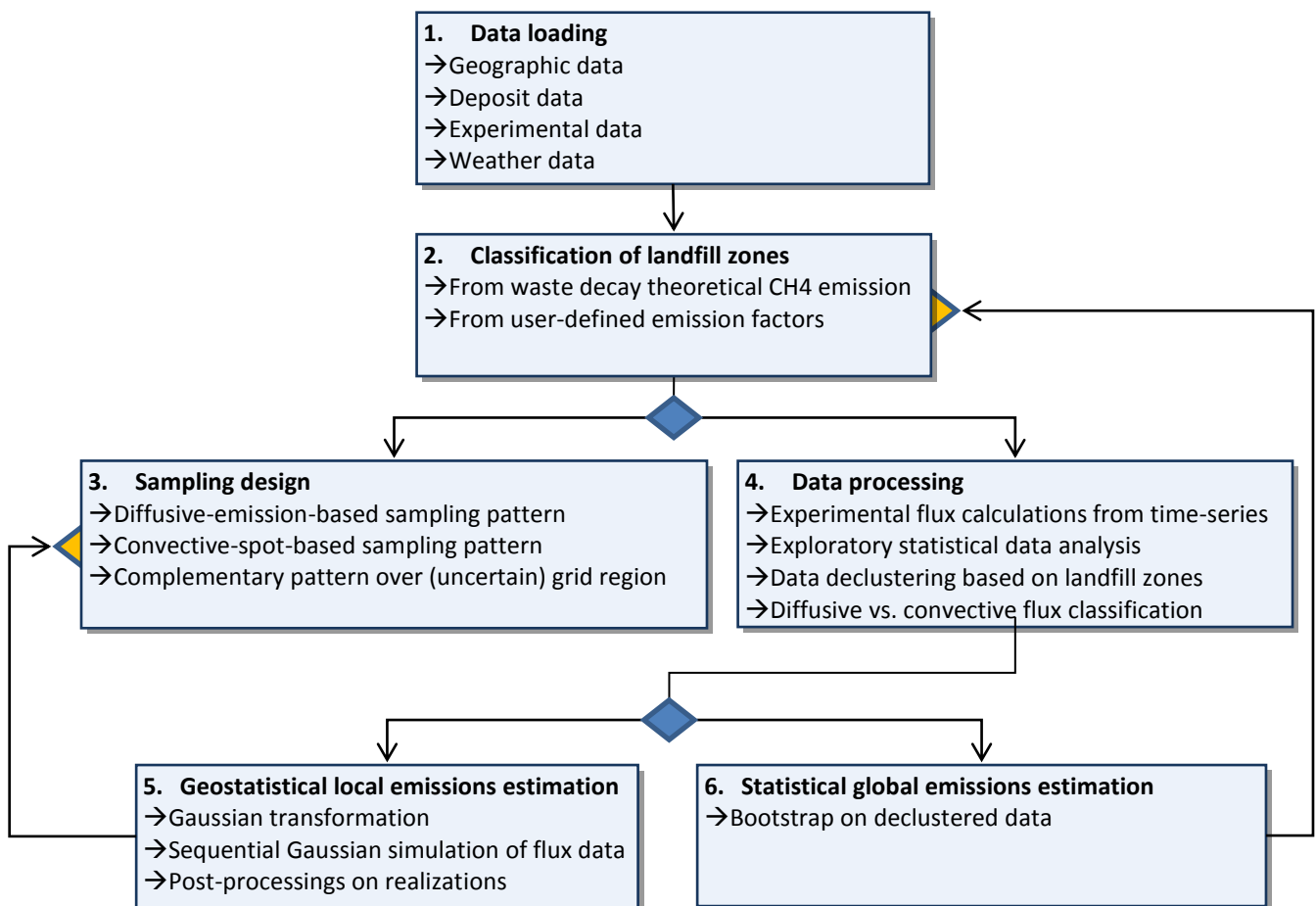


Figure 1. Schematic of the workflow.

3. PRESENTATION OF THE WORKFLOW

The workflow is divided into three parts as depicted in Figure 1. At the beginning of the workflow, a general part allows to import and manage all the data (landfill cell boundaries and other features, nature and amounts of landfilled wastes, landfill gas management...), to calculate theoretical fluxes based on the waste contents of landfill cells, and to classify the cells according to theoretical or experimental (estimated from data) fluxes. At the end of the general part, the user must decide whether she or he wants to continue with the sampling design part of the workflow, or with the one dedicated to flux calculation and global or local emission estimation.

The aim of the workflow is to assist the user at every step of the study, whether the objective is to develop a sampling strategy, or to estimate local fluxes, or overall biogas emissions, associated with uncertainty quantification. The workflow also allows to save and keep track of all the choices made by the user about data, options (methods) and calculation parameters. The workflow has been tested and the results validated on an actual case study (§4).

This workflow has been implemented as linked to the SoilRemediation® Suite developed by KIDOVA in partnership with Paradigm to provide cost-effective environmental software solutions integrating a limited version of the powerful GOCAD-SKUA® geological modeling software.

The three workflow parts are further detailed below.

3.1 General part of the workflow

In the general part of the workflow, beside the import and management of data, the primary objectives are to calculate theoretical (raw) emission potentials, associated with landfill cells, and to classify the cells according to either such theoretical emission potentials, or to global biogas emissions previously estimated from flux data.

The screenshot shows a software interface with four main steps:

- 1 Select or create a theoretical flux calculation parameters object**: A dropdown menu shows 'ADEME'.
- 2 Define the zone methane production potential**: Two radio buttons are present: 'From waste methanogenic potential (PCH4)' (unselected) and 'From waste Total Organic Carbon (TOC)' (selected). Below, a dropdown menu shows 'TOC - Total organic carbon - Contamination Soil - Grade - kg/t' and a text input field for 'Degradation temperature (°C)' contains '30'.
- 3 Define the waste Bioaccessible Organic Carbon (BOC) degradation kinetic(s)**: A table with columns 'Waste BOC fraction' and 'Ki'.

Waste BOC fraction	Ki
1 ME - High kinetic (biodegradable) ...	0.5
2 ME - Medium kinetic (biodegrada...	0.1
3 IE - Low kinetic (biodegradable) o...	0.04

Buttons for 'Add', 'Remove', and 'Edit' are to the right of the table.
- 4 Select the type of emission estimation parameters**: Two checked checkboxes are shown: 'Collection factor' and 'Oxydation factor'.

Figure 2. Theoretical flux calculation steps.

Two methods are available to calculate theoretical emission potentials. One relies on emission factors (Taramini and al, 2003). The other one consists in LFG production modeling, based on any number (generally three) of user-defined waste bioaccessible organic carbon (BOC) fractions (Figure 2). The user can specify correction factors to account for oxidation and LFG collection over the survey period. Different oxidation percentages, resulting from the modeling of LFG transport and methane oxidation in soils using CALMIM (Bogner and al, 2010) for each type of covers, can modify the default (10%) methane oxidation parameter.

Using CALMIM to estimate oxidation percentages is a convenient way to proceed, the oxidation capacity of landfill covers being generally not investigated because of the high spatial variability of methanotrophic activity (Rower and al, 2011). Local mass balance of LFG gas can also provide a distribution of methane oxidation percentage (Christophersen and al, 2000), provided the LFG concentration below the surface is well known.

3.2 Sampling design part

The aim of sampling design is to decide about the number of data to collect in each landfill cell, the types of data (screening concentration data, fast IFM data using a simple bell, precise flux data using a flux chamber) and possibly their locations (sampling pattern).

It is necessarily related to survey or assessment objectives and must take advantage of the available data. A multistep approach is proposed to manage all situations from designing a screening survey based on concentration measurements in the air, to the sampling design of flux measurements relying on theoretical or estimated global emissions (see §3.2), exploitation data (cell boundaries, gas well layout), and possibly (geostatistical) maps of estimated screening methane concentrations or methane fluxes as obtained from a previous survey (see §3.4).

The number and types of data to collect can be user-defined based on time and budget constraints. They can be calculated using the recommendations of environmental protection agencies based on landfill cell areas (USEPA, 1986). It can also be taken into account the theoretical or estimated global emissions to calculate cell-based measurement point densities, the higher the expected fluxes, the higher the density of points.

Cell boundaries and gas well layout being known as potentially associated with cover defects, they can be used to define local sampling pattern refinements from which potential convective gas flows are expected to be detected.

The maps of estimated screening methane concentrations or methane fluxes are another way to control the point densities or to define additional measurement points in zones where fluxes are likely to be higher (hot spots, convection-dominated flows). Zones of particular interest are identified by applying an appropriate threshold value to estimated concentrations or fluxes. Uncertainty issues can also be addressed, provided a geostatistical model of methane fluxes has already been generated (see §3.4). In terms of sampling design, additional sampling points are expected where uncertainty is too high.

3.3 Flux and emission estimation part

As a first step before to estimate local fluxes or global emissions, biogas fluxes are calculated from IFM data that have been measured using a simple bell or a flux chamber (§2). The calculation consists in estimating average gradients from measured concentration vs. time curves. The estimated gradients must be representative of stabilized flux measurement conditions, i.e., conditions in which the gradients are not affected by the measures themselves or by adverse weather effects. The fluxes are derived from gradients by multiplying them by a factor depending on bell or chamber parameters.

Two gradient calculation methods are proposed. The standard one is based on least-square fitting

of a straight line that is calculated on part of the curve (i.e. time interval) where the following conditions must be met: concentration and time are linearly correlated, the correlation is good enough, the number of concentration data is enough, and the slope of the regression line, which defines the average gradient, is higher than in other parts of the curve. The quality of estimated gradient is evaluated from a QC indicator that measures the correlation between concentration and time on the selected time interval (Figure 3). This calculation method has been used and validated on several Walloon landfill sites managed by ISSEP (Awono and al., 2005, Bietlot and al., 2013), and within the frame of research projects of INERIS (Akerman and al., 2007, Bour and al., 2013). The other method is statistical. The principle is to identify modes (populations) in the distribution of gradients, calculated along a concentration curve, and to estimate the average gradient from the mode that corresponds to satisfactory measurement conditions. This method is the purpose of ongoing tests.

About the estimation steps, three options are available, corresponding to different objectives.

If the objective is to carry out sampling design based on screening concentration data (see §3.3), maps of biogas concentrations can be estimated using a geostatistical method. It can be noted that the use of screening concentration data has been made possible thanks to the progress of analyzers and global positioning system (GPS) that allow to conduct air methane concentration surveys with high frequency measurements (see Scharff and al, 2003, and Babilotte and al, 2010, for applications to global methane emission assessment over vertical or horizontal planes).

If the objective is to estimate global gas emissions, two methods are proposed to assess from previously calculated flux data. One is statistical, the other one geostatistical. The statistical method is faster and only requires that the available flux data be weighted (declustered) to account for sampling bias and landfill cell boundaries. Polygonal, cell and kriging-based declustering methods are available (Mathieu and al., 2014). A bootstrap technique (Mathieu and al., 2014) is used to quantify the uncertainty about cell-based mean fluxes, from which it can be derived the uncertainty about global emissions at landfill cell scale.

The geostatistical method relies on stochastic simulation techniques (Sequential Gaussian Simulation, see Goovaerts, 1997, or GeoSiPol, 2005) to generate realistic maps (called realizations) of the spatial distribution of methane fluxes within each landfill cell. The maps are realistic in the sense that they honor all flux data, at data locations, and reproduce their spatial continuity as established from variographic analysis (Bietlot and al., 2013). They are not unique, however, many of them having to be simulated (100 or more), to allow quantifying local (spatial) and global uncertainty. Each flux simulation provides a possible rate value of methane emission at the landfill cell scale (emission rate = average simulated flux within the cell times cell area). By doing so, as many emission rates as there are simulations can be calculated, from which the uncertainty on global emission rates is estimated. If concentration or other flux data are also available and correlated with methane fluxes (e.g. CO₂ fluxes, less precise CH₄ fluxes measured with a bell), they can be taken into account into the simulation algorithm as secondary (complementary) information to reduce uncertainty.

It can be noted that none of the available estimation method takes into account theoretical emission potentials, methane oxidation information, and methane production rates from gas wells, to try better estimating global emission rates. Integrating such data in the estimation process would require to establish mass balance equations, which is right now beyond the scope of the workflow.

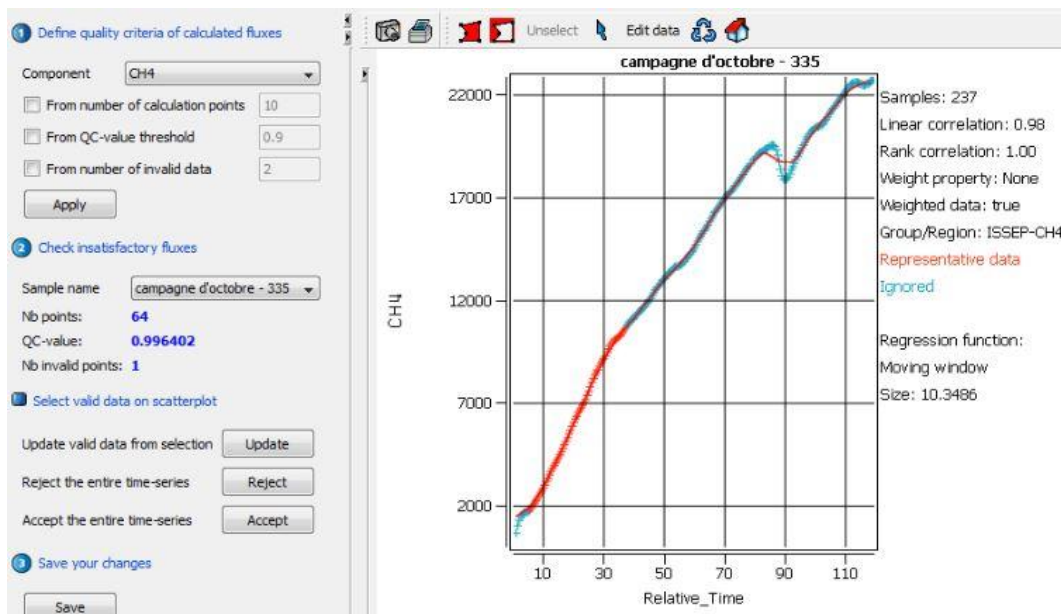


Figure 3. Quality control of biogas fluxes estimated by using the standard linear regression-based method.

4. APPLICATION OF THE WORKFLOW TO AN ACTUAL LANDFILL CASE STUDY

4.1 Presentation of the landfill site

The site is located in Wallonia, south part of Belgium. This landfill belongs to the monitoring network of ISSEP which includes 12 municipal solid waste (MSW) landfills. Air quality monitoring includes the quantification and the cartography of LFG surface emissions from the cover of the landfill.

The area of the site is 25 ha. Waste information is available for 6 cells which have a total area of 19 ha (see Table 1).

Table 1. Characteristics of the six landfill cells for which waste information is available.

	Cell 2	Cell 3	Cell 4	Cell 5	Cell 6	Cell 7
Area (ha)	4.15	3.55	3.6	3.0	2.8	2.0
Deposit (ktons)	1356	1292	1239	1148	910	957
Start of exploitation	1989	1994	1997	2000	2003	2008
End of exploitation	1994	2000	2000	2003	2007	-
Cover type	liner	silt	silt	silt	silt	daily cover

The first cell (Cell 1, 3 ha), which only contains inert wastes, has been ruled out of the study. Figure 4 displays the location of Cell 2 to Cell 7 where municipal solid wastes (MSW) and non hazardous wastes are landfilled. The facilities are also depicted (leachate ponds, collection wells). Biogas is collected through a gathering system including more than 100 vertical wells and horizontal pipes. Many LFG surface monitoring surveys have been conducted on this site since 2002.

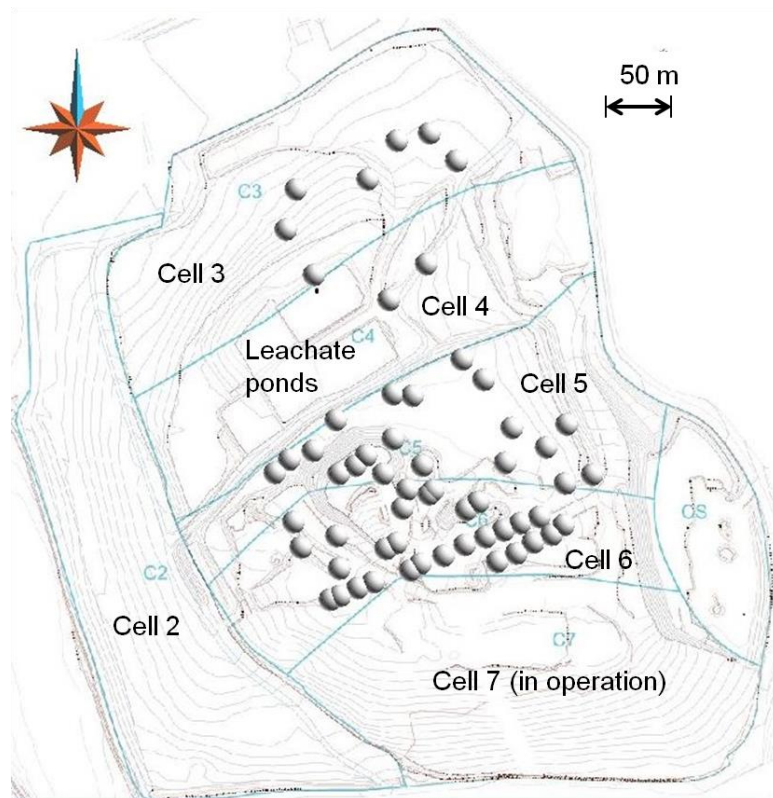


Figure 4. Top view of the landfill site where cell 2 to cell 7 are shown with collection wells (spheres)

4.2 Results from the sampling design part of the workflow

Three types of measurements have been collected and compared. The primary goal of the first ISSEP surveys (from 2002 to 2007) was to estimate maps of methane fluxes. In 2010, a flux chamber was used to assess global methane and carbon dioxide emissions at the whole landfill site scale.

IFM data, providing flux measurements without control of wind, were compared with precise flux data obtained by using a static chamber with recirculation and controlled weather conditions. Both types of flux measurement were successively performed at different sampling point locations to obtain more than 50 pairs of associated IFM and chamber flux data. These data were processed and compared.

The best correlation is observed between chamber methane flux and CO₂ IFM. It is explained by the higher methane detection threshold of the Ecoprobe used in 2010 (> 300 ppm) and the better spatial continuity of CO₂ fluxes (no zero values to manage).

Part of the work was also to compare the emission rates, estimated for each cell from the available flux data, with theoretical methane production potentials based on latest waste storage figures. The theoretical methane production potentials were calculated for all cells and show that cell 6 is the one with highest potential.

In 2015, a screening methane concentration survey has been carried out as the first phase of a two-phase sampling strategy approach. High frequency measurements were performed (2 measures / s) using the responsive Inspectra Laser detector. At the same time, the path followed by the operator was recorded using a GPS mobile device (1 measure / s). The synchronized concentration and GPS location data were used together at data import to calculate the locations of concentration measurement points (locations interpolated from the recorded path). Air concentration

measurements being highly sensitive to wind conditions, the wind speed was also measured to check that the conditions were good enough (wind speed < 3 m/s).

The methane concentration data were expected to provide information about methane emission sources, i.e. the location of cracks and others discontinuities that could be missed without having a high enough density of measurement points.

An estimation map of methane concentration was created using a geostatistical approach. The estimation was carried out on normal score transform (Gaussian transformation) of concentration data, then backtransformed to get estimates of local concentration statistics (see Goovaerts, 1997, or GeoSiPol, 2005, for details about the method). The Gaussian transformation was required to highlight the spatial correlation of measured concentrations (Figure 5). The estimated concentration map was used as providing information about surface emissions to decide about the density and sampling pattern of flux measurement points (ongoing second phase of the two-phase sampling strategy approach).

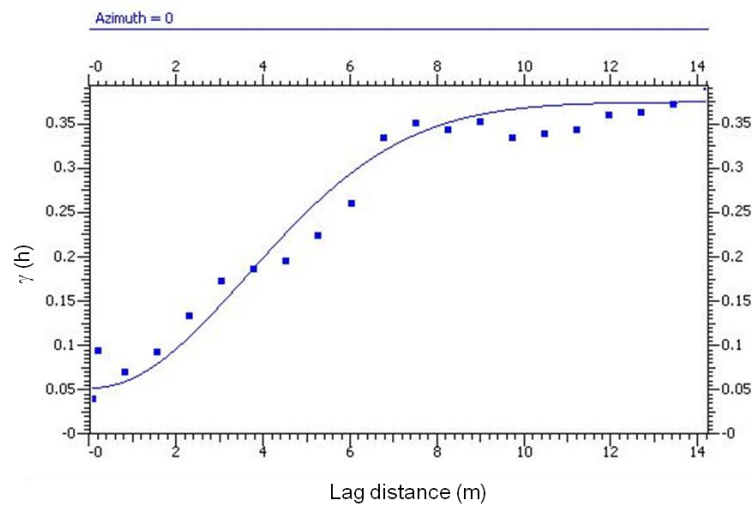


Figure 5. Omnidirectional variogram of the normal score transform of screening methane concentration (2015 survey). The experimental variogram (points in the plot) is fitted by an isotropic Gaussian variogram model (range = 8.8 m, sill = 0.38) associated with a nugget effect of 0.05 (line in the plot).

The estimated methane concentrations were compared with the IFM and chamber flux measurement data that were estimated from the 2010 survey. Figure 6 shows the estimated concentration map and the 2010 IFM and chamber flux measurement data. An attempt was made to interpret the estimated concentration values in terms of diffusive vs. convective fluxes. Threshold concentration values were defined and probability maps that the concentration be higher than the thresholds were calculated. At this stage, it was not possible to identify thresholds that would allow distinguishing convective fluxes from diffusive ones.

Practically, the screening concentration measurement survey was fast (< 2 days) and allowed to rationalize the combined IFM and chamber flux measurement survey to investigate the 19 ha area cover by the 6 cells.

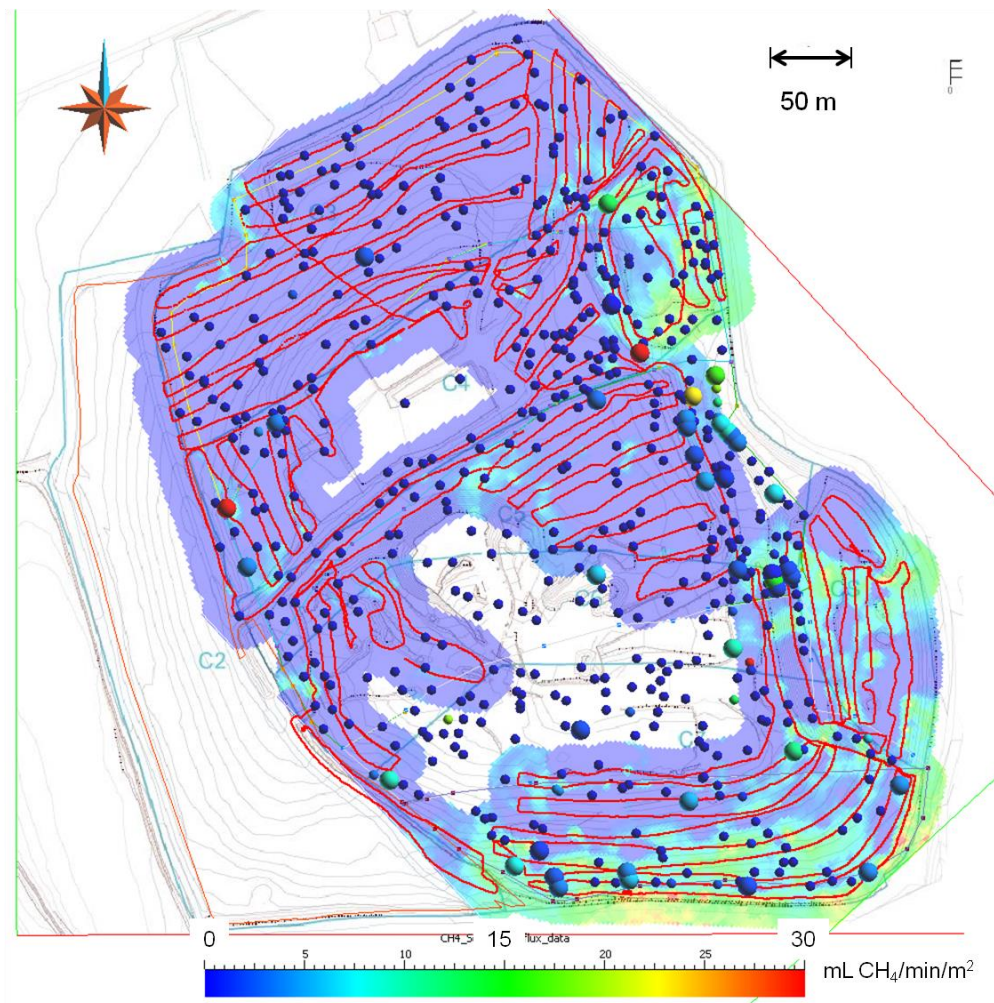


Figure 6. Geostatistical estimation map of screening methane concentration (2015 survey). The displayed concentration values are median values backtransformed from estimated normal score transform concentration. This estimation map of concentration can be compared with methane IFM data (small spheres) and chamber methane flux data (medium spheres) dating from 2010. The colorscale is related to flux values.

4.3 Results from the estimation part of the workflow

The global emission starts to be estimated by using bootstrap on weighted methane flux data (see §3.3). The 2015 IFM and chamber flux measurement data have just been collected. The results from this part of work are not yet available and will be completed in an updated version of this article.

5. CONCLUSIONS

A new multistep workflow involving theoretical emission potential assessment, two-phase sampling strategy, and local vs. global estimation aspects is proposed and started to be tested to better estimate global methane surface emissions from landfilled methanogenic wastes. The application of the workflow is illustrated with results from a Belgium landfill site. Among the particularities of the workflow, methods are provided to control the density of flux measurement points based on different criteria, and local or global estimation results from previous surveys can be used to

improve sampling design. The calculation of fluxes from concentration time-series is facilitated by QC indicators and interactive tools. Different user-friendly statistical and geostatistical estimation methods are also provided to take advantage of the different types of available data, whether local estimations of concentrations or fluxes, or global methane emission estimations at the cell or site scale must be performed.

Though the workflow covers all steps from waste characterization to emission estimations, a number of sampling design and estimation challenges still have to be addressed. One of them is about the way major emission sources may be identified. Gas well locations and cell boundaries can already been taken into account with local sampling pattern refinements, but using numerous screening concentration data would be a more general way to define targets where (convective) emission sources are more likely to be found to perform flux measurements.

Estimating the contribution of areas with low methane surface emissions is another challenge, especially if the aim is also to assess the methane oxidation contribution of these areas. Though methane oxidation efficiency can simply be modeled using methane oxidation potential parameters, the heterogeneity of soils and the need to account for moisture parameters of the final cover (sludge dredged with different slope angles) complicate the modeling task.

The variation of methane emissions with time is another challenge for sampling design and data processing: all fluxes data result from short duration concentration measurements that are representative of a particular date and time. They are known to change with time, from one day to another but also on the same day, thus leading to spatial distributions of fluxes that also change with time. Diffusive and convective fluxes must be distinguished, however, the latter being directly governed by surface air pressure. The assesment of global annual diffusive surface emissions may be achieved by simply knowing the soil parameters and carrying out two flux measurement surveys to calibrate a model (Bogner and al, 2010). To estimate the fraction of pressure driven convective emissions, a large number of measurements is required to characterize the changes over a defined timescale.

More research work is still required about the integration of near soil methane concentration in the air to better assess the temporal representativeness of convective fluxes.

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