INVERSE DISPERSION MODELLING FOR A QUICK SCAN SERVICE TO ASSESS FUGITIVE EMISSIONS FROM LANDFILLS

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Abstract: In the framework of the Climate-KIC, the WASTE-MITI² project (waste management solutions to address climate change impact), a method assessing the fugitive emissions from a landfill as a low-cost service has been developed. The objective is to be able to use on-board sensors, measuring methane concentration alongside a road located inside (or nearby) a landfill, to estimate the location and the rate of the methane emissions. In this paper the inversion methods developed by ARIA Technologies to achieve this goal is presented and real-world cases are documented. The experimental protocol, which relied on using a mobile cavity ring-down analyser, will be also described. From a set of methane measurements provided by LSCE and collected during two different measurements campaigns (November, 17th and December, 5th, 2016) on a landfill operated by SUEZ, two different inverse modelling methods were applied and their results compared. The first method is based on retro-plume technique considering several methane sources whose locations are supposed known. The target is to calculate the emission rates of these different sources. The second one is an Adaptive Multiple Importance Sampling (AMIS) algorithm. It is an adaptive Bayesian approach for source term estimation (Rajoana et al, 2016). It aims at estimating the location and the emission rate of a single and pointwise equivalent source minimizing iterations. The emission results will be discussed considering the comparison with other approaches like the use of a tracer gas (LSCE) or direct predictive landfill emission model output (SUEZ).

Key words: inverse modelling, GHG, methane emission, fugitive emission, landfill, Quick Scan

INTRODUCTION

Just after the carbon dioxide (CO₂), the methane (CH₄) is the most important anthropogenic GHG: releasing 1 kg of CH₄ into the atmosphere gives the same impact on global warming than releasing 28 kg of CO₂ on a 100 years scale (IPCC 2013). Though the main anthropogenic methane emissions are due to the production and transport of coal, natural gas, and oil, methane emissions also result from livestock, agricultural practices and by the decay of organic waste in solid waste landfills. One of the challenges for landfill operators as SUEZ is to collect and to valorise fugitive emissions from such landfills. Solid waste management in Europe and beyond varies widely between countries. In recent years the practice of landfilling has almost been abandoned for example in DEU, NL, DK and the organic content of the remaining landfilled waste is almost zero. Nevertheless, landfill (LF) emissions remain the dominant source of GHG emissions from the waste sector in Europe (Eurostat, 2011). Management options that minimize the GHG emissions from LF’s exist and can be cost-effective or can be implemented at relatively low costs per ton of CO₂ eq mitigated. A vast experience on effectiveness, robustness of measures and associated costs is present and was partly developed by WASTE MITI² (WM2) partners.

The aim of this project is to develop a transparent decision support tool that can be applied by landfill operators, including an advisory feedback loop to confirm the results are realistic. The decision support tool will be tested on a selected set of landfills managed by partner SUEZ. Previous Climate KIC initiatives during recent years investigated novel quantification methods and development of monitoring techniques for fugitive CH₄ and CO₂ emissions at site- or city-wide scale (FUME & CarboCount City, respectively). Previous research (Oonk, 2012; Monster et al., 2015) shows that LFG capture efficiencies can vary between 10 to 80%. The proposed Quick Scan Tool will support
identification of poor efficiency sites in combination with measures to improve biogas collection (e.g. better capture) or mitigate emissions (e.g. oxidative layers over hotspots). Eventually, such a service could become part of a regulatory framework or permit management.

SITE AND MEASUREMENT DATA SETS
For the base case as defined in the WASTE MITI² project, SUEZ selected a landfill dedicated to municipal waste where methane emissions are known as active (Figure 1). A closed landfill is located at the west part of the landfill (Figure 2). If measurements are made with a westerly wind, it will therefore be necessary to have sensors located between the two places, allowing to separate the emissions that might be coming from this closed landfill. According to SUEZ experts, the landfill could be divided in four different basins with different level of emissions and the total CH₄ emissions are expected to be between 3.8 and 7.6 tons per day. The exercise is to do an independent emission assessment using air concentration measurement campaign.

![Figure 1: landfill selected for the base case.](image1)

![Figure 2: A closed landfill is located at the west part of the landfill. It can present CH₄ emissions.](image2)

The concentration data set, provided by LSCE, consists of CH₄ concentrations measured by a Picarro G2203 detector, loaded on a moving vehicle. While the vehicle is travelling on the roads inside the landfill, the sensor can make one measure of the methane concentration every second, and store it with the corresponding GPS coordinates. The itinerary of the vehicle while collecting the data on November, 17th is represented on Figure 3, and the concentrations on Figure 4.

![Figure 3: Red line shows the all data set collected (here on November, 17th)](image3)

![Figure 4: Corresponding CH₄ air concentration from Picarro G2203 (mobile inside the car)](image4)

The inverse methods that we will be using to perform the reverse modelling are based on a reverse atmospheric model starting from a sensor network. We commonly use data from 30 to 40 well localised sensors. As a first step, we have aggregated the measured concentration (~10 000 points) and create a new set of sensors, appropriate to perform the inversion. Another important aspect of the modelling is the time discretization of the problem. In the provided data, only instantaneous measures are available as the vehicle moves, following a complex path and it is therefore not possible to have regularly discretized data at each location as a reduced set. The total duration of the measurements is about 3 hours, which is very short compared to the expected variations of the emissions rate of a landfill (which has an order of a month). Considering these points and the relative stability of the wind (speed and direction), we made the assumption of a stationary problem.
To reduce the number of sensors while keeping as much information as possible, the set of sensors is built to include:

- the noticeable CH$_4$ peaks detected along the path of the tour;
- but also, the low CH$_4$ values in-between these peaks to be able to separate one plume from another.

As it can be seen on the data collected on that day, a wide area around the landfill was investigated. However, in our case, only concentrations in the close vicinity of the landfill are useful (central part with high methane concentrations of the graphic on Figure 4). From this part of the data, we extracted the interval from 13:35 to 13:45, during which the wind was quite steady (Figure 5).

The optimization of the sensors location from this lap includes the following steps:

1. filtering of the concentrations and detection of the peaks;
2. selection of the main peaks at which to place sensors by enforcing a threshold distance along the itinerary between two sensors;
3. placing equally spaced sensors between these main peaks;
4. gathering nearby sensors in case the itinerary overlaps.

The optimized location of the sensors and the corresponding concentrations are displayed on Figure 6.

**METHODS AND RESULTS**

**Case A: Source locations are considered as known**

In this case, we take benefit of the linearity between concentration and emission.

\[
\begin{bmatrix}
A_{11} & \cdots & A_{1n} \\
\vdots & \ddots & \vdots \\
A_{m1} & \cdots & A_{mn}
\end{bmatrix}
\begin{bmatrix}
Q_1 \\
\vdots \\
Q_n
\end{bmatrix}
+ \begin{bmatrix}
B_1 \\
\vdots \\
B_m
\end{bmatrix} = \begin{bmatrix}
C_1 \\
\vdots \\
C_m
\end{bmatrix} \quad (1)
\]

$A_{ij}$ is the atmospheric transfer coefficient from source $i$ to sensor $j$.

$Q_j$ is the emission rate of the source $j$ from 1 to n.

$C_i$ is the concentration measured by the source $i$ from 1 to m.

In practice, most computations are performed with errors. We need to solve a system $AQ = C$, where the data (the elements of A and b) are not known exactly. Therefore, it is important to understand how the data errors can affect the solution $x$. Data perturbations. If $x$ is the exact solution of $AQ = C$, and $Q + \delta Q$ is the exact solution of a perturbed problem $(A + \delta A)(Q + \delta Q) = (C + \delta C)$, then this estimate, given up to linear terms of perturbations, holds:

\[
\frac{\|\delta Q\|}{\|Q\|} \leq \kappa(A) \left( \frac{\|\delta A\|}{\|A\|} + \frac{\|\delta C\|}{\|C\|} \right) \quad (2)
\]

Relative errors in A or C may be amplified in the solution vector Q by a factor $\kappa(A) = \|A\| \|A^{-1}\|$ called the “condition number of A”.

The generic implementation of the retro-plume inverse modelling technique in the ARIA View framework is the following in general case of m receptors and n sources:

- ARIA View is using PMSS as stochastic Lagrangian dispersion model PMSS that was validated in complex geometry (Armand P., 2007). PMSS can perform direct or inverse modelling run. Starting
from the m receptors, m retro-plumes are followed “backward in time” through the domain. Retro-concentrations at sources locations are averaged over 5 minutes and stored each 5 minutes. The matrix A is built, with n columns and m rows;

- Measurements at receptors locations are averaged in time to get 5-minutes averaged concentrations. Then these averaged concentrations are used to build the vector of measurement C.

- C is an input of the receptors oriented pre-processor. The main function of this pre-processor is to identify invalid data, to remove them from C, and to remove the corresponding row of the matrix A. If a whole column of A is equal to zero, it means that the source related to this column was not impacted by any retro-plume. Then this column is removed and the dimension of the Q vector is reduced.

- Singular value decomposition is applied to the matrix A and the condition number is computed.
  - If the condition number is small enough, the least squares problem related to system (1) is solved. The vector Q is obtained, together with the residual error ;
  - If the value of the condition number does not allow an accurate estimation of the emission rates, previous emission rates are kept.

- Once the vector Q is known, a forward simulation can be performed.

Several mitigation actions have been undertaken on this landfill. To measurement campaigns were performed one before and one after the mitigation actions. The main results of two on field campaigns, 17 Nov 2016 and 5 Dec 2016 are given on Figure 7 and Figure 8. That could be coherent with emission decreasing

![Figure 7: Calculated Emissions for the November 17th](3.85 tons/day)

![Figure 8: Calculated Emissions for the December 5th](2.92 tons/day)

**Case B: Source locations are not known**
The ARIAMIS software is using AMIS (Adaptive Multiple Importance Sampling) algorithm. AMIS is an adaptive Bayesian approach for source term estimation (Rajoana, 2016, and 2015). ARIAMIS was designed to assess the location and the emission rate of a single and pointwise source and uses a reverse atmospheric simulation from the sets of optimized sensors. ARIAMIS is also done to deal with time-independent data. We use it here as time-dependent but with constant measurements in time as input.

ARIAMIS has been run with the measurements of the November, 17th. The results are displayed below: the particles sample the probability distribution describing the location of the pointwise source. Thus, the convergence of the sampled particles to a single point shows the convergence of the algorithm to a single solution (black dots on Figure 9).

As can be seen on Figure 9, there is a convergence of the algorithm to a single point located inside the landfill (the blue cross represents the location of the sensors in the landfill). However an important sensitivity to the input parameters has been observed, as close input parameters can give very different locations of the source: this is mainly due to the fact that, as can be noticed from the Retro Plume results, the methane emissions take place on a wide area inside the landfill. The ARIAMIS method only works with pointwise solution and is therefore not adapted to such a case: the source location given by the algorithm in the best case is a “mean location” and the associated emissions rate can only give an idea of
the magnitude of the emissions but cannot be accurate. Taking into account the conclusions from the simulation on day 1, we decided not to model the second day with this algorithm.

Emissions rate \( 4.0 \times 10^7 \mu g/s = 3.46 \text{ tons/day} \)

**Figure 9**: Particles sampled at iterations 1, 2, 3, 4, 6, and 8

**COMPARISON WITH THE DUAL TRACER METHOD**
The tracer release method consists in releasing a tracer gas, here acetylene (C\(_2\)H\(_2\)), at a known rate from a location which is co-located with the unknown emission of a trace gas to be determined, here CH\(_4\). Concentrations of the tracer as well as the gas of interest are simultaneously measured using the mobile instrument downwind in the co-propagating plumes. The ratio of the area of the two plumes signals is proportional to the emission rate. Thus, knowing the emission rate of the released gas and the concentrations of both gases, we can calculate the emission rate of the gases of interest.

In this method, uncertainties are coming from the concentrations, the tracer flux and the correlation between the plumes. CH\(_4\) and C\(_2\)H\(_2\) errors are small, less than 2% and 10% respectively for one-minute average. Once the gas cylinder is installed and regulated, the flow of tracer gas is steady and well-known, and this error is negligible. The main uncertainties come from the analysis of the plumes, especially background determination for CH\(_4\). To get a better result, a first cartography is done to estimate the possible location of hotspots. The acetylene bottle is placed here close to the source of the studied gas. The flowrate of acetylene is approximately 0.2 kg/h measured with a flowmeter. The bottle is weighted before and after the release to confirm the average flow rate. Methane emissions are evaluated if concentration variations of C\(_2\)H\(_2\) and CH\(_4\) are well correlated. During the dual tracer campaign on September 19th, multiple crossings were performed through the plume of the acetylene, deliberately released from the main methane source region. Clear correlation between methane and acetylene enhancements were found for most events (2 event did not show sufficient co-variation). The flux estimates range from 3.3 t/d to 7.8 t/d with a mean source estimate of 5.8 t/d +/- 1.3 t/d (i.e. 22% variability from the mean). The data analysis was also conducted using peak height rather than peak area, which shows the results to be robust. The ambient air temperature was about 28°C on September 19th. This can explain higher CH\(_4\) emission values. Considering a unique source instead of large area source could also be a reason of overestimation.

**CONCLUSION**
The retro plumes method gives coherent values in comparison of the current knowledge of the emission of the landfill. Nevertheless, it has the disadvantage to force the pre determination of the emission zones. In the case of a wasteland, this requirement is still easy to satisfy. The other limitations of the method are to have enough sensors /pseudo-sensors considering the number of emissions zones and the location of this measurement point is also sensitive. Trying to multiply the number of emissions zones to have a “better description” may lead to a system of equations with high condition number and a divergent
solution. When the configuration of the model respects these limitations, the determination of emitted quantities leads to global realistic results. ARIAMIS is adapted to pointwise sources, for instance a leakage in a well or a barrel, whereas the retro Plume algorithm gives a useful insight when the emissions are located in a wide area. If the Retro Plume algorithm is enough to quantify the emissions, the two methods can be complementary for specific cases where a leakage is suspected. The dual tracer method is also a convenient way to assess the emission rate the limitation are typically the necessity to know where is the source and the source must be punctual. The use of a high quality mobile sensor is a good alternative and present many advantages but it seems difficult to substitute mobile measurement to long term on site network. The question of how is representative the days that have been modelled is completely pending. SUEZ and ARIA are working to make the methodology more and more operational and try to define the best economic business model for MRV GHG emissions supervision. The same methodologies as described in this paper have already been used to estimate mitigation effects on landfill.

REFERENCES
Monster J., Samuelsson J. Kjeldsen P., Scheutz C., 2015 ” Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method” Waste Management Volume 35, January 2015, Pages 177-186

ACKNOWLEDGMENT: The authors want to thanks the Climate-KIC for their support in this work. This work is an abstract of the “Waste –Miti2” project. SUEZ as PI, partners and all contributors are warmly thanks.