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FUGITIVE GHG FLUX ESTIMATON USING REVERSE MODELLING AND MOBILE ONSITE MEASUREMENTS

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Abstract: Methane (CH_4) and nitrous oxide (N_2O) are among the most potent greenhouse gases and therefore need to be monitored closely. As much as 40% of global N_2O emissions originate in human activity and that number even reaches 60% for CH4. Landfills and wastewater treatment plants are two important sources of these gases [Overview of Greenhouse Gases, United States Environmental Protection Agency], and their total emissions can sometimes be difficult to quantify without accepting high uncertainties. In 2015, Aria Technologies started developing with SUEZ a methodology called QuickScan now renamed as SCAN360. The goal was to quantify a site's CH_4 emissions using on-field measurements and inverse dispersion modelling. The concentration mapping was to be performed with one mobile sensor only, thus avoiding additional costs. The dispersion model used is PMSS, which considers the weather conditions recorded on-site, as well as the topography and surrounding buildings. Following an initial modelling step, each source's emission is automatically adapted using a regression coefficient. This coefficient results from the average difference between modelled and measured concentrations at a set of locations. Nearly a decade after its first launch, this method has continuously evolved and has found an important place in SUEZ Air &Climate's product portfolio. The number of wastewater treatment plants and landfills studied has increased rapidly, leading to a significant gain in experience and hindsight. Thanks to exchanges with clients as well as between the measuring and the modelling teams, rapid improvements in the method were made. This resulted in better flexibility and more efficient analyses. Additional gases have since been studied (mainly N_2O) and a significant return on experience has been gained. This short paper reports the main lessons learned on the application of SCAN360 for GHG quantification for industrial sites.

Key words: *GHG, reverse modelling, landfill, WWTP, Scan360, Quickscan*

INTRODUCTION

Methane (CH_4) and nitrous oxide (N_2O) are Green House Gases (GHG) that are emitted by human activities, among them two infrastructure types that are managed by SUEZ group: waste water treatment plants (WWTP) and landfills. The estimations of the emissions are generally done thanks to emission factors or analytical approaches based on process data, waste amount and types. The SimCET (Mauret, 2008) software for example is based on the model of the Solid Waste Association of North America (SWANA) referenced by US-EPA and is often used by the SUEZ group.

Direct measurement of surface emissions is not possible, as it would be for a stack, as these emissions are spread, numerous and not channeled. The available sensors are typically measuring the concentration of the gas in the air, but not the emissions. If these concentration measurements are performed downwind of the sources, then the atmospheric transport and diffusion has dispersed the emitted mass. This atmospheric effect between emissions and concentrations could be quantified thanks to an atmospheric dispersion model.

For the specific application of CH_4/N_2O emission estimation for landfills and WWTP, here are the main approaches that we have identified:

- Remote observations from space with satellite (Maasakkers *et al.* 2022): even if it allows the service to be easily applied on a new site, it is currently only sensitive enough for big emitting sites.
- Fluxes on 2D cross sections with drones: a concentration sensor is set up onboard a drone that flies across the downwind plume of the site at different vertical levels (Yong *et al.* 2024). If the whole plume is caught including observations of wind speed and direction, the GHG flux across the section can be computed by integration on this section. It gives access to the global emission of the site.
- Ground observations + reverse modelling. Two types of observation strategies can be distinguished: monitoring gas in atmosphere is usually done with fix points, thus building a network. It requires several sensors because only one point might not be enough to feed the reverse modelling. In the studied context, the required accuracy with the typical emission range and background values (~2 ppm for CH_4 and ~330-340 ppb for N_2O) implies to use expensive sensors. To use a limited number of sensors and provide a cost-efficient service for many sites, the second strategy is to use mobile measurements with only one sensor. In the framework of this mobile approach, two strategies could be identified:
    - Only far field measurements across the plume: it gives access to the global emission of the site. This strategy is not presented here but is detailed in Kumar *et al.* 2022)
    - Far field measurements across the plume and in-site measurements: it gives access to the contribution of each source but potentially requires a more detailed atmospheric dispersion model.

This last strategy is layed out in this document. It was initiated in 2015 during the WASTE MITI project (Albergel *et al.* 2017) on 2 landfills. The method has since then been industrialized among the SUEZ Air &Climate team and branded as Scan360 service. It has been applied on more than 40 cases (13 landfills (CH_4), 11 WWTP (CH_4 and N_2O), several repetitions for some sites).

The objective of this document is to summarize the lessons learnt by applying the methodology on this significant number of cases, confronting the theorical framework to the real life constrains.

INVERSION METHODS

The inversion problem that is met here is defined as following:

- Thanks to a first "sniffing" step with the mobile sensor, the sources are identified and located
- On WWTP their geometries are well known because the bounds of each structure are well defined. The spread leaks and diffuse emissions on landfills are such that only approximate polygons can be drawn to describe the different sources.
- The target is to quantify the global emission rate of the site but also the contribution of each source
- For some of the sources, the emission rate is already known because they are channeled. The inversion method should not change these rates.
- The emission rates are considered as constant during the observation period.
- Background concentration is significant.
- Potential parasite sources near the site are possible.
- The local meteorological data is well known because a station is temporarily installed.

Based on these elements, we have tested several inversion methods. They are all based on the comparison of the observed and computed concentrations at sensor locations. Assuming both CH_4 and N_2O are passive tracers, the hypothesis of linearity between concentration and emission rate is done. The correction factors for concentration can then be applied also on the emission rates.

The methods are also all based on the establishment of an initial first estimation of the emission of each source, taking advantage of the knowledge of the site thanks to process experts and thanks to the sniffing step. Emission rates from channeled sources can directly be defined and locked in through this step. For the other sources, this first estimation is computed by multiplying the average concentration observed over or very close to the source by the area of the source and an arbitrary (but identical for all the sources) velocity. This way, the first estimation already includes a preliminary ranking.

The first inversion method is the less accurate but quite robust: a linear regression is performed based on a Quantile-Quantile scatter plot (Q-Q plot). Only one correction factor is applied onto all the emission rates, except for the sources that have whose emissions have already been locked in. For the rest of the sources the initial ranking is conserved.

The second method is a single linear regression on a point-to-point comparison. Again, the initial ranking of the sources is conserved but not the emission rates that are potentially already known. This method should be more accurate than the first, but could be more sensitive to outliers.

The third method, that is described in (Albergel *et al.* 2017), consisting in solving a linear system provided by the dispersion model when computing the contribution of each source at each measurement point. This is easy to perform by just adding more scalars to be dispersed. This method doesn't imply to start with an initial first estimation. Solving this linear system is not always possible. As mentioned in Albergel *et al.* 2017, it can only be done when the condition number of the system is lower than 20. Qualitatively, this reflects the fact that the available concentration points might not be well located to be able to distinguish the contribution of the difference sources for the given wind direction.
Because of the constrains that are listed in a dedicated section below, it appears after several years of service application that the linear system is often not well conditioned. In this case, we apply an iterative adjustment for each source or group of sources to minimize computed/observed errors. Full automatization of this iterative process is the next challenge of the service.

Dealing with mobile observations

This section briefly explains how the data of the mobile sensor is preprocessed before feeding the inversion. The modelling inversion is based on steady state hypothesis: emissions and wind are supposed to be constant in time. Only a sub period of the concentration observations is kept, corresponding to a period during which the variation of wind speed and direction is limited.
During this sub period, the high frequency (1 Hz) observations are aggregated in order to limit the number of points and to slightly smoothen the signal. This aggregation step aims to average measurements around a set of local concentration peaks (Albergel *et al.* 2017). Our observation trajectory protocol implies to follow a same path several times, if possible. Then, the aggregation step should also include a spatial aggregation, which is similar to computing a time average of several paths.
With trees and buildings that are often present in WWTP and landfills, we have learnt that the spatial location of the mobile sensor that is given through a GPS equipment can include outliers. The path of the raw concentration data and the aggregated points should be double checked by the operators of the field campaign to minimize the use of such outliers as input of the inversion.

Dispersion model :

To include both the far field and the near field (*i.e.* inside the site) observed concentrations in the inversion, using a dispersion model that considers local effects of buildings and topography (such as wakes, speed up, turn around) is preferable. As full blown CFD might be too expensive to setup for the Scan360 service, the intermediate solution PMSS is used (Moussafir *et al.* 2013). It includes both explicit effects of buildings and topography. Figure 1 shows an example of a site (composting site which similar to a WWTP in terms dispersion modelling) with simplified geometry and 3D wind field computed by PMSS.

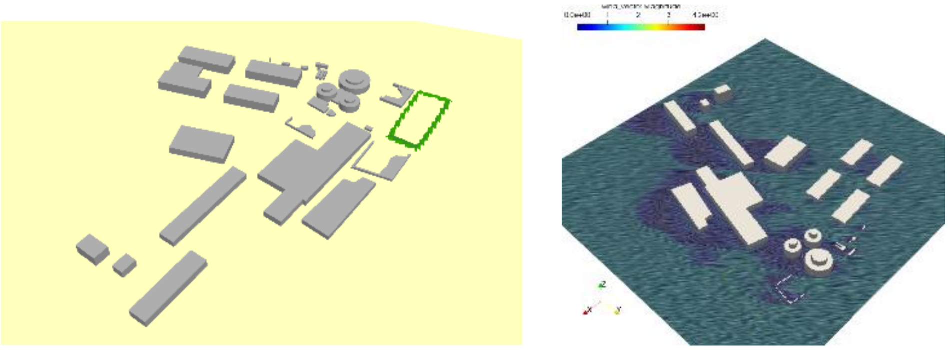


Figure 1. Left : Example of a modelled site with simplified buildings in grey and topography (here mainly an embankment) in green – Right : computed wind speed and stream lines on a horizontal slice near ground level

SITES CONSTRAINTS

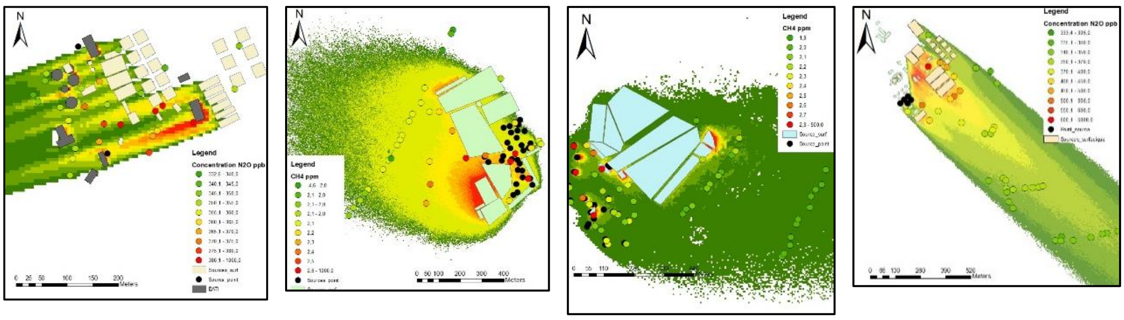
The equation system mentioned previously is often not well conditioned because the observation paths performed by the operators are highly constrained by different elements on sites. After 40 cases being treated, it is worth listing these different elements that we have met. Inside the site, measuring a concentration both to downwind and to upwind of each source might be impossible because of physical obstacles, explosive zones (ATEX area), or because of too steep terrain (especially on landfills). Outside the site, the operators are often constrained to walk or drive with the mobile sensor on roads or trails because of surrounding private properties or fields.

The emission intensities are often such that the CH_4 and/or N_2O signal is too low compared to background in the very far field. The network of roads/trails are then often just dense enough to have one downwind cross-section.

Moreover, the risk of unknown parasite sources falsifying the far field measurements becomes more and more important with increasing distance to the source. Around landfills, CH_4 emissions from cattle field have been met several times.

Analyzing the 40 treated cases, it appears that a limited number of configurations of available points' spatial repartition is seen: only inside site points; Inside points + 1 downwind cross section; Inside points + 2 cross sections at different downwind distance; Inside points + 2 sections aligned with wind direction; Inside points + 3 sections (mixed crossing and aligned)

Some examples are given on the next figure.



a. *b.* *c.* *d.*

Figure 2. Examples of different configuration of available observations compared to plume direction: a. : Only in-site (WWTP) – b. : 1 cross section (landfill) – c. : 2 cross sections (Landfill) – d. : 3 cross sections (WWTP)

MODEL FITTING PERFORMANCE

As the emission rates are unknown, the error on the inversion cannot be computed. Some exercises with controlled acetylene release have been recently performed on one of the sites and will be used for validation with the limitation that controlled releases are often point sources and not a group of diffuse sources.

In the framework of the Scan360 service, in order to quantify the performance of the modelling inversion, we have systematically computed a fitting error as the average of the absolute relative error between observation and computed concentration (with removal of background concentration). This average is computed with all the points used for the regression or can also be computed with a sub list of points that should be influenced by a given source. Then, the fitting error can be given globally, or source by source. By linearity hypothesis between concentrations and mass rates, the relative error on the concentration can be applied on the emission rates.

When analyzing the 40 treated cases, the following feedback can be given:

- The average error over the site is similar for CH_4 and N_2O: 43.3% for CH_4 and 40.4% for N_2O
- The average error for CH_4 comparing WWTP and landfills are similar (respectively 43.2% and 43.4%)
- The cases with fitting errors below 40% are the "easy" cases: the different observation zones were consistent with the modelled plume patterns. The cases with fitting errors above 50% are the ones, where a satisfactory fitting score was not obtained for all observation zones.
- The fitting error is globally improving with the number of available measurement sections, even if a larger number of sections can also be more constraining (see figure 3).

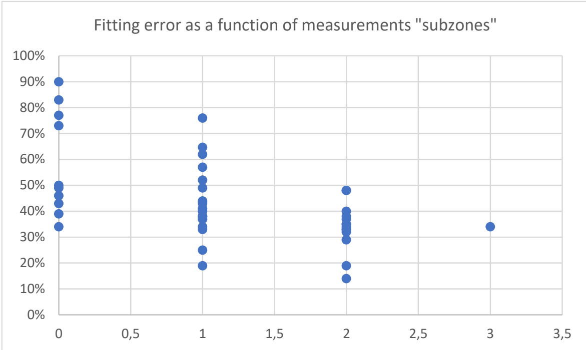


Figure 3. Fitting errors of the 40 cases sorted as a function the number of available observation subzones (in the site and/or sections outside of the site)

The fitting error is not the uncertainty of the estimation. It is only an indicator of whether the model fitting is satisfying are not. Computing in an exhaustive way the uncertainties would require performing more computations with their associated CPU cost. The different sources of uncertainties might be:

- Wind speed measurement
- Wind direction measurement
- CH_4/N_20 concentration measurement
- GPS localization of CH_4/N_2O mobile sensor
- Wind speed steady state hypothesis for modelling
- Wind direction steady state hypothesis for modelling
- Aggregation of CH_4/N_2O points
- Model fitting error (regression error, atmospheric turbulence estimation, internal dispersion model error)

PERSPECTIVES

To better track the performance of the fitting, it would be better to systematically compute the usual statistics given by Chang & Hanna 2003: FAC2, NMSE, R, FB.

To compute the contribution of each source, it appears that the linear system is often not well conditioned in practice and that the knowledge of sources type/activity from landfills and WWTP experts is an important added value. Therefore, a regression based on a nonlinear algorithm and that could learn from past cases and sources experts would be a greater improvement. However, it needs to be evaluated whether there is enough data yet to set up such an algorithm.

The full uncertainty of the estimation is not computed yet. By introducing more automatization into the process, ensemble modelling could be added without increasing to much the price of the service.

A comparison with a gaussian approach for the dispersion modelling has already been done on some sites in partnership with LSCE team, but using slightly different input data due to different preprocessing methods. We plan to do this comparison by just switching the dispersion model in order to quantify the gap in the estimation, especially for cases without significant terrain and building effects.

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REFERENCES

Albergel Armand, Denis Morin, Didier Buty, Maxime Nibart, Laurent Makke, Robin Goix, Robert Kelly, Caroline Bouchet and Felix Vogel, Inverse dispersion modelling for a quick scan service to assess fugitive emissions from landfills, 18th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes 9-12 October 2017, Bologna, Italy

Chang, Joseph & Hanna, Steven. (2004). Air quality model performance evaluation. Meteorology and Atmospheric Physics. 87. 167-196. 10.1007/s00703-003-0070-7.

Kumar Pramod, Grégoire Broquet, Christopher Caldow, Olivier Laurent, Susan Gichuki, et al.. Near-field atmospheric inversions for the localization and quantification of controlled methane releases using stationary and mobile measurements. Quarterly Journal of the Royal Meteorological Society, 2022, 148 (745), pp.1886-1912. ⟨10.1002/qj.4283⟩. ⟨hal-03665710⟩

Mauret M., INSA Toulouse, Ingénierie de Systèmes Biologiques et des Procédés (LISBP), Génie des Procédés Technologies Environnementales, France, Mars 2008, 54p.

Maasakkers Joannes D. *et al.*, Using satellites to uncover large methane emissions from landfills.Sci. Adv.8, eabn9683(2022).DOI:10.1126/sciadv.abn9683

Moussafir, J., Olry, C., Nibart, M., and Albergel, A., 2013. Aircity, a very High-resolution 3D Atmospheric Dispersion Modeling System for Paris. In 15th Int. Conf. on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes.

Yong Han, Grant Allen, Jamie Mcquilkin, Hugo Ricketts, Jacob T Shaw, Lessons learned from a UAVsurvey and methane emissions calculation at a UK landfill, Waste Management, Volume 180, 2024, Pages 47-54, ISSN 0956-053X, https://doi.org/10.1016/j.wasman.2024.03.025.