Statistical atmospheric inversion of small-scale gas emissions by coupling the tracer release technique and Gaussian plume modeling: a test case with controlled methane emissions

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Abstract

This study presents a new method for estimating the pollutant emission rates of a site and its main facilities using a series of atmospheric measurements across the pollutant plumes. This approach is based on a combination of the tracer release method, a Gaussian atmospheric transport model and a statistical atmospheric inversion approach. The conversion between the tracer controlled emission and the measured atmospheric concentrations across the plume provides knowledge on the atmospheric transport. The concept of the method consists of using this knowledge to optimize the configuration of the Gaussian model parameters and the model uncertainty statistics in the inversion system. The pollutant rates of each source are inverted to optimize the match between the concentrations simulated with the Gaussian model and the pollutants' measured atmospheric concentrations, accounting for the Gaussian model uncertainty. This new approach is evaluated with a series of inversions of controlled methane point sources using acetylene as a tracer gas. In these experiments, different configurations of methane and acetylene point source locations are tested to assess the efficiency of this method in comparison with the classic tracer release technique to cope with the distances between the different methane and acetylene sources. The results from these controlled experiments demonstrate that when the targeted and tracer gases are not well collocated, this new approach provides a better estimate of the emission rates than the tracer release technique. As an example, the relative error between the estimated and actual emission rates is reduced from 29% with the tracer release technique to 8% with the combined approach in the case of a tracer located 60 metres upwind of a methane source. This method also enables an estimate of different sources within the same site to be provided.

1 Introduction

Atmospheric pollution due to anthropogenic activities is a major issue for air quality and for climate change because of the increase in the atmospheric concentrations of greenhouse gases. Industrial sites are known to emit a significant part of the pollutants and greenhouse gases. For instance in France, industrial emissions represent between 10 and 30% of major air pollutants, such as carbon and nitrous oxides (Bort and Langeron, 2016). Currently, industries must list their emissions through national inventory reports, and some of them commit to reducing these emissions. However, the choice of an appropriate mitigation policy and the verification of its results require a good understanding of the emitting processes and a precise quantification of the emission rates. Industrial emissions are difficult to model and quantify

because of the diversity and the time variability of the emitting processes. Many emitting industrial sites have a typical size of 100–500 m^2, and they emit pollutants from very specific locations within this area. The transport of these pollutants in the atmosphere over distances from 0.1 to several kilometres from such sites can be viewed as a plume from point sources. One approach developed to quantify the surface emissions from such sites involves atmospheric concentration measurements around the site, particularly across these emission plumes, and a proxy of the atmospheric transport. This proxy is used to estimate emission rates from such observed concentrations, providing information about the structure and the amplitude of the plume. Among the different techniques to estimate emissions from concentrations is the tracer release method, which is used to provide estimates of the transitory emissions of a site (typically for a few hours during a given day). This method is based on mobile continuous measurements across the emission plumes of the studied pollutant and of a tracer purposely emitted as close as possible to the pollutant source with a known rate (Lamb et al., 1995). Because the atmospheric transport can be considered as linear for most of the pollutants for short distances (since they can be assumed to act as passive species over very short time scales), the plumes of the targeted gas and of the tracer should have the same structure if both gases are perfectly co-emitted at the same location and with the same time variations. In such a configuration, the knowledge of the ratio between the tracer controlled emission rate and the tracer plume magnitude can be used as an estimate of the ratio between the targeted gas emission rate and the targeted gas plume magnitude such that the measurement of the targeted gas plume magnitude can be converted into an estimate of the targeted gas emission rate.

This approach is relatively simple to implement and enables punctual estimations for a large number of sites. Nevertheless, this technique encounters some limitations, particularly when it is difficult to position the tracer emission close to the sources, as well as when the sources are spread over a significant area compared with the distance between the sources and the location of the measured concentrations, or when targeting individual estimates of the different emission rates from multiple sources whose plumes overlap over a given site. Typically, in industrial sites, pollutant sources may be sporadic and diffusive over a large area and their locations are not always precisely known. In these cases, the tracer release method can induce errors in the flux estimation since the tracer plume by itself cannot be used as an accurate proxy of the local transport from the targeted gas sources to the measurement locations. Moreover, this approach can hardly be used to provide an estimate of the different sources within a site. Even with the use of different tracer release points, the technique in itself does not provide solutions to separate the overlapping tracer and/or targeted gas plumes associated with different point sources.

Other techniques exploit atmospheric measurements using local atmospheric dispersion models to simulate the transport of the targeted gas from its sources to the measurement locations (Lushi and Stockie, 2010). Such models are driven by data on the meteorological conditions and are based on mass conservation to predict how a given emission rate for a given source impacts the 3D atmospheric concentration field of a pollutant. This can be used to infer the linear relationship between a given emission rate at a source and the concentration at a given location, the amplitude, or the integral of a measured transect through the gas plume. An estimate of the emission rate can thus be inferred from the measurements and from simple mathematical inversion using this linear relationship. In theory, the model and the inversion can be applied for a point source or for a source whose spread is known. The model can also be applied to multiple sources, and the same number of concentration measurements or measurement integrals appropriate to dissociate these different sources could be exploited. In such cases, a simple mathematical inversion of the invertible linear relationship between the emission rates and measurement indices should allow for estimation of the different emission rates associated with the different sources. The principle of this technique is relatively simple, but the transport model, the representation of the emission spread in this model, and the separation of the different plumes associated with different sources present some uncertainties. In particular, the transport over short distances is characterized by complex turbulent structures whose processes are difficult to model and can hardly be matched. Moreover, the simple mathematical inversion artificially requires extending or limiting the number of data extracted from the measurements to the number of sources to be quantified, which can represent a loss of information or can hide the fact that the problem is underconstrained.

Accounting for uncertainties in the model and addressing under- or overconstrained mathematical problems when using the correct number of data that corresponds to the complementary pieces of information in the measurements can be addressed using a statistical inversion framework, which can be viewed as a generalized inversion technique. In such a framework, a statistical estimate of the emission rates for the different targeted sources is derived to optimize the fit to the measurements, accounting for

the statistical uncertainties in the source and transport modelling, in the measurements and in the prior knowledge about the source location and magnitude (Goyal et al., 2005). Statistical inversions using atmospheric transport models and atmospheric concentration measurements have been used for decades to infer surface sinks and/or sources of pollutants and greenhouse gases at the continental to the city scales (Gurney et al., 2003; Bréon et al., 2015). However, the skill of such approaches strongly relies on the transport and source modelling accuracy and on the ability to quantify this accuracy.

This study aims at combining the tracer release technique, Gaussian plume modelling and a statistical inversion framework to develop a robust technique to improve the estimation of gas emissions from one or several point sources in an industrial site-scale configuration. The concept consists of using the knowledge on the transport given by the tracer controlled emission and concentration measurements to optimize the calibration of the Gaussian model parameters and to assess the statistics of the model errors for the configuration of the inversion system.

This method is tested for the quantification of transient methane emissions (for a given time window of several hours) using acetylene as a tracer gas and mobile measurements across the methane plumes for several hours. Methane is an important greenhouse gas with largely unknown point source emissions (Saunois et al., 2016). Typical methane emitting sites due to anthropogenic activities include waste processing plants (wastewater treatment plants and landfills), oil and gas extraction and compressing sites and farms (Czepiel et al., 1996; Yver Kwok et al., 2015; Marik and Levin, 1996). A precise and easy-to-implement method for estimating methane emissions could help operators of such sites in their local action plans to mitigate their emissions in the context of climate change. While a continuous monitoring of such emissions would help characterize the dependence of such emissions on meteorological conditions and on the change in the site processes through time, punctual estimates through a dedicated measurement campaign can help to detect and provide a useful order of magnitude for such sources that are generally poorly known (Yver Kwok et al., 2015).

We conduct a series of controlled experiments with known emissions of methane from one or two sources and of acetylene from one source and with concentration measurements through the methane and acetylene plumes at an appropriate distance from the source, as described below. The known emission of methane is used to validate the inversion results and thus to assess the efficiency of our new approach. In particular, the fit between these results and the actual emissions is compared with the one obtained with the more traditional computation associated with the tracer release technique to demonstrate the asset of the statistical inverse modelling framework. In section 2, we detail the theory and specific framework for the monitoring of methane sources with the tracer release technique, of the local dispersion modelling and statistical inversion exploited here, and of our method that combines these different techniques and tools. Then, we describe the specific experimental set-up and the inversion configuration used in this study (section 3), and finally, we discuss the results of these controlled experiments (section 4).

2 Methods

2.1 Framework to monitor of transient pollutant sources using mobile measurements across the atmospheric plumes

The methods described below correspond to a specific configuration of generic techniques for the atmospheric monitoring of gas emissions, focusing on the estimation of transient emission rates. They are based on the principle of measuring the concentrations across the emission plume of a targeted gas emitted by an industrial site at an appropriate distance several times over the course of a few hours. Crossing the emission plume allows the increase of its concentration above a background concentration to be measured, which corresponds to the concentration upwind of the industrial site that has not been affected by the sources within the site. Ideally, the background concentration is relatively smooth since other major gas emitters would be located away from this location and because atmospheric diffusion should cause their plume to vanish when air reaches the targeted site. The methods apply to inert non-reactive gases, and thus, the representation of atmospheric transport, linking emissions to concentrations, can be considered to be linear.

The choice of the measurement distances should follow several criteria. The distance has to be high enough such that the transport from the source to the measurement is correctly characterized with a transport model or the proxy from the tracer release (which depends on the spread of the single or multiple

targeted sources and thus indirectly on the size of the industrial site). However, the distance should be short enough such that the amplitude of the measured concentrations is high enough compared to the measurement and model precision. Finally, the distances should be adapted to the need for conducting measurements on roads located downwind of the point sources (depending on the specific wind directions during the measurement campaigns) when using instruments onboard cars as in this study.

The conversion of the concentration measurements through the plumes into emission rates is performed through proxies of the source spread and atmospheric transport, which links gas emission rates from one or several sources to the atmospheric concentrations through the observation equation:

$$p = Hf + \varepsilon_0 \tag{1}$$

where f is the control vector that contains the targeted emission rates of the methane sources, p is the observation vector, H is the observation operator (i.e. the combination of an operator representing the source spread and an atmospheric transport operator), and ε_0 represents the sum of errors in the observation operator and in the measurements.

The observation vector is constructed from the gas concentrations measured for each cross-section of the pollutant plume. The atmospheric transport operator is constructed using either the relationship between known collocated tracer emission and concentrations in the tracer release technique (section 2.2) or using a physical atmospheric transport model (section 2.3). While the model allows for addressing multiple sources or sources with a significant spread far better than proxies based on collocated tracers, such models have limited ability for simulating local transport dispersion. The sources are generally considered to be small enough so that they can be reduced to point sources, but the transport models used here allow for their spread over significant areas within the observation operator to be taken into consideration. Inferring gas emissions from gas concentrations implies inverting the atmospheric transport to express f as a function of p. If the size of f is the same as that of p (i.e. if the number of data points derived from the concentration measurements is set equal to the number of targeted sources), then the atmospheric transport matrix is invertible in a mathematical sense, and this inversion is straightforward (sections 2 and 2.3). As discussed in the introduction, such an inversion can hardly account for the amount of useful information provided by the measurements (typically, missing the shape of the emission plume when targeting a single source) and for uncertainties in the measurements and observation operator. To overcome these issues, one can perform a statistical inversion (section 2.4). Here, we propose a new method (section 2.5), combining the tracer release method, a local-scale Gaussian model, and a statistical inversion framework to overcome the issues associated with these different approaches and tools as discussed above.

This method is tested for the quantification of methane sources using acetylene as a tracer gas. Both of these gases are inert and can be considered non-reactive at the time scale and over the space scales corresponding to the time and distance between the release of molecules at the source and the measurement of concentrations downwind in the plume, with the lifetimes of methane and acetylene being approximately 10 years and 2–4 weeks, respectively (Logan et al., 1981)). In this study, the methane and acetylene concentrations are measured in a continuous manner along a line crossing the emission plume using an accurate analyser placed in a car. Our preliminary analysis shows that we obtain satisfying results when concentrations are typically measured at a distance of 100 to 1000 metres from methane sources of 1500 to 100000 gCH$_4$.h^{-1} and spread within an area of 100×100 m^2 to 500×500 m^2.

2.2 The tracer release method

The tracer release method was developed to quantify pollutant emissions and has already been used in a wide range of studies to estimate the sources of various types of gases such as methane (Babilotte et al., 2010), carbon monoxide (Möllmann-Coers et al., 2002) and isoprene (Lamb et al., 1986). This method consists of releasing a tracer gas with a known rate close to the targeted gas source when this source is clearly localized or in the middle of a site when numerous sources, diffuse or not, are present on the emitting site.

When both the released tracer and targeted sources are perfectly collocated and constant in time, they have the same spatial and temporal relative variations of their concentrations in the atmosphere (i.e. the same plumes ignoring a multiplication factor). By measuring their concentrations in a section of the downwind emission plumes and knowing the released tracer emission rate, the targeted emission

rate can be estimated using the following formula:

$$f_m = f_t \times \frac{p_m}{p_t} \qquad (2)$$

where f_m is the targeted emission rate, f_t is the measured tracer release rate, and p_m and p_t are indices of the amplitude of the measured targeted gas concentrations and the measured released tracer concentrations above the background, respectively. Various indices of the ratio between the targeted gas and the released tracer increased concentrations can be used. Typically on the one hand, it can be calculated using the maximum concentrations (peak heights of the signals) of both gases, and on the other hand, it can be established using the areas under both plume signals and above the background concentrations (Mønster et al., 2014). If the sources of the released and targeted gases are perfectly collocated and if their emission rates are constant, both of these approaches provide the same result given that both emission plumes are identical. However, if the collocation of both sources is not perfect or if the targeted emissions vary in time, then the shapes of the emission plumes of the released tracer and of the targeted gas can differ. To minimize the impact of this difference, the ratio of the integrated plumes is generally chosen because this index is less sensitive to the impact of thin turbulent structures than the peak height ratio (Mønster et al., 2014).

Such a computation is generally repeated several times (approximately 10–15 times in general), which means that the emission plumes are crossed several times, and equation 2 is applied for each crossing. The mean and standard deviation of the different results are used as the best estimate and uncertainty assessment for the source quantification. Plume crossings with a low correlation between the variations of the targeted gas and the released tracer along the measurement line are generally ignored in the computation of these statistics. Such statistics allow to account, at least partly, for the potential temporal variations of the emissions, for the measurement errors, and for the potential impact of the non-perfect collocation of the sources.

However, a mislocation of the tracer source too far from the targeted source can generate biases in the series of computations, which would impact the average estimate of the source without being reflected in the standard deviation of the individual emissions computations. Moreover, this technique provides an overall estimate of the emissions of a site but, when the site has several sources located quite close to each other, it cannot provide individual estimates of these sources.

2.3 Local scale transport simulation using a Gaussian plume model

Many types of transport models are used to simulate the dispersion of pollutants at the local scale (over distances from a few metres to 1 or 2 kilometres) (Baklanov and Nuterman, 2009).

While LES and CFD models allow for turbulent patterns over such spatial scales to be generated and for changes in the terrain topography and for buildings to be accounted for (Letzel et al., 2008; Britter and Hanna, 2003), they can hardly be set-up or controlled to perfectly match the turbulent patterns at a given time and location downwind of a source. Gaussian plume models provide a stationary and average view of the pollutant plumes driven by meteorological conditions that are stationary in time and homogeneous in space within the study period and area. This is a decent approximation for the modelling of dispersion over 1–2 minutes (i.e. the typical timescale associated with our experiments) and an area of approximately 1 km^2 when the wind speed is relatively high. These models cannot precisely account for the local topography and buildings. However, this type of model is suitable for many configurations of industrial sites located in nearly flat suburban to rural areas, and it is easily set up and applied for the simulation of local-scale transport.

The Polyphemus air quality modelling system, developed by the CEREA (Mallet et al., 2007), gathers several types of transport models allowing the representation of the dispersion in the atmosphere of diverse pollutants, such as passive gases, radionuclides or aerosols, from the local to continental scale. In this study, the Gaussian model of Polyphemus is used because it has been proven to be adapted for estimating gas emissions from local sites (Korsakissok and Mallet, 2009).

Gaussian models are based on a simple formula that provides the concentration of the pollutant at a location generated by a point source depending on the weather conditions. The Gaussian plume formula

is expressed as:

$$C(x, y, z) = \frac{Y}{2\pi\sigma_y\sigma_z\bar{u}} exp\left(-\frac{(y-y_s)^2}{2\sigma_y^2}\right)$$
$$\times \left[exp\left(-\frac{(z-z_s)^2}{2\sigma_z^2}\right) + exp\left(-\frac{(z+z_s)^2}{2\sigma_z^2}\right)\right] \quad (3)$$

where C is the concentration of the pollutant at a location of coordinates (x,y,z), Y is the source emission rate, and $\bar{u}$ is the wind speed. In this formula, the x axis corresponds to the the wind direction, y_s is the pollutant source ordinate and z_s is the release height above the ground. Instead of being deposited, the emission plume rebounds when it reaches the ground, which is a decent approximation regarding the studied gases. σ_y and σ_z are the horizontal and vertical Gaussian plume standard deviations and characterize the atmospheric conditions during the measurements. The modelled concentrations are strongly dependent on these two parameters. Polyphemus proposed several ways to parameterize these constants: the Doury formulas (Doury, 1976), the Pasquill-Turner formulas (Pasquill, 1961) and the Briggs formulas (Briggs, 1973). The Gaussian model and its parameterizations have been described by Korsakissok and Mallet (2009).

Briggs parameterization is the most adjustable parameterization of Polyphemus: not only does this parameterization consider the stability of the atmosphere via six classes from A (extremely unstable) to F (extremely stable) but it also takes into account the type of environment where the emissions occurred with an urban mode when the site is surrounded by buildings and a rural mode for the isolated sites. The standard deviations with Briggs parameterization are given by the following equations:

$$\sigma_y = \frac{\alpha x}{\sqrt{1 + \beta x}} \quad \text{and} \quad \sigma_z = \alpha x (1 + \beta x)^\gamma \quad (4)$$

where x is the downwind distance from the source and α, β and γ are coefficients that are dependent on the stability classes. All these coefficients can be found in Arya (1999).

Different source spatial extensions can be taken into account in this model. However, its configuration imposes the emission f_i of a given source to be spread homogeneously over such an extension. Simulations with the model for each individual source (ignoring the other one) can be used to compute each column of the H matrix in equation 1 and thus the full matrix and its inverse H^{-1}. Consequently, H^{-1} can be directly used for the inversion of the emission of the different sources as a function of the same number of indices on the measurements through their plumes:

$$f = H^{-1}p \quad (5)$$

2.4 Statistical inversion

The Bayesian principle of statistical inversion is to update a prior statistical knowledge (i.e. a prior estimate f^b and the uncertainties in it) of the emission rates f with statistical information from observations p. This update accounts for the statistical uncertainties in the observations, in the source, and in the transport model H, which is used to connect the emissions to the concentrations (Tarantola, 2005).

Since this theoretical framework allows for a control vector f and an observation vector p with different sizes to be taken into account, it can be used to assimilate the data from all plume crossings to compute the emission rate at once rather than repeating the computation and deriving statistics for the emission estimates out of the ensemble of computations as for the other techniques presented above. Furthermore, the previous techniques require a selection of the cases when the confidence in the tracer proxy or in the model is good enough to strengthen the robustness of the average (since the model and the tracer proxy skills highly depend on the shape and location of the plume during a given crossing). By assigning model and measurement uncertainties as a function of the plume crossing, this method allows the information from each crossing to be weighted differently according to its uncertainty when deriving the best estimate of the emissions.

The prior estimate of the emission f^b has to be independent of the atmospheric observations and can be provided by expert knowledge, emission inventories or process-based models. In practice, it is generally assumed that the uncertainties in f^b, in the observations and in the model have unbiased and Gaussian distributions. The prior uncertainty and the sum (henceforth called observation error) of the uncertainties in the measurement based on data p and on the observation operator H are thus

characterized by their covariance matrices B and R, respectively. Following these assumptions, the "posterior" statistical distribution of the emission rate knowing f^b and p is Gaussian and is characterized by its optimal estimate f^a and its covariance matrix A (which thus characterizes the unbiased and Gaussian uncertainty in f^a) (Bocquet, 2012) given by equations:

$$f^a = f^b + BH^T(R + HBH^T)^{-1}(p - Hf^b) \qquad (6)$$

$$A = (B^{-1} + H^T R^{-1} H)^{-1} \qquad (7)$$

2.5 A statistical inversion based on tracer release and Gaussian transport modeling

This study aims to develop a new method to estimate the emission rate of a pollutant by merging the previously described methods to overcome the issues associated with their individual usage. This new method is based on the statistical inversion framework described above, where the H matrix is derived from Gaussian model simulations for each point or spread source.

The main concept is to use the very accurate information on the atmospheric transport in the area of interest from the tracer release method and the Gaussian model simulations of the tracer plume to optimize the configuration of the Briggs parameters in the Gaussian model and to assess this model uncertainty for the configuration of the observation errors in the statistical inversion framework. Different configurations of the Gaussian model are forced with the known tracer emission rate, and the configuration whose simulation of the tracer concentrations best fits the tracer concentration measurements is taken as the optimal one. In practice, this fit is primarily checked for the indices chosen for the definition of p (i.e. the integration of the plume concentrations above the background for a given plume crossing), but it is also checked in a qualitative way by analysing the shape of the modelled and measured signals.

The knowledge of the tracer emission and the measured tracer concentration is also used to correct for part of the measurement uncertainty. As will be explained in section 3.2, there is a type of spatial offset between the measured plume and the actual plumes due to the lag between the air intake and the concentration measurement, which can be significant in the measurement framework discussed in this study. The spatial offset between the modelled and measured tracer plume is thus applied to the modelled plume of the targeted gas. This offset does not impact the computation of the areas under the emission plumes and thus the tracer release technique. However, it can impact some statistical inversions using more complex indices for the observation vector, typically when attempting to individually invert several sources based on the identification of their overlapping plumes (see section 3.5).

The statistics of the misfits between the tracer measurements and the model-based indices when using the optimal model configuration are used to set-up the covariances of the observation (measurement and model) error R. This optimization of the model parameters and/or characterization of the observation errors can be performed for each individual crossing of the plume or for all plume crossings together. If the wind conditions evolve rapidly or if there is a weak confidence in the fact that the Briggs parameters are the main source of uncertainty in the model (such that optimizing these parameters would only compensate for other sources of errors), the use of a specific optimization of the model for each plume crossing may be preferable. Using general statistics of the tracer model data misfits from all plume crossings would prevent weighting of the observation error and thus the information for each plume crossing depending on the modelling skills. Deriving different observation errors for each plume crossing requires the extrapolation of the single set of tracer model misfits into statistics for each plume crossing. These different options need to be chosen depending on the experimental case (see section 3.5 for the options taken for the specific test case of this study).

We apply this new combined approach to the specific framework described in section 3.1 for the quantification of methane sources using acetylene as the tracer gas. The following sections describe the experiments under controlled conditions for both acetylene and methane that are used to evaluate this method.

3   Description of the set up

3.1   General principle of controlled experiments

The following sections describe the experiments under controlled conditions for both acetylene and methane used to evaluate the approach detailed in section 2.5. A campaign was organized during two days of March 2016 at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE) in France (longitude: 48.708831°, latitude: 2.147613°, altitude asl: 163 m). One or two methane sources and one acetylene point release were generated with cylinders in the parking lot of the LSCE, which is located in a rural area in the southern region of Paris. The topography of this area is very flat, and only few buildings of small size can influence the atmospheric transport from the parking lot to the road where the concentrations are measured. This road is located approximately 150 metres away from the controlled sources. No major methane or acetylene sources in the vicinity of the LSCE could disturb the measurements. During this campaign, the methane and acetylene sources were dispersed in four different configurations to estimate the accuracy of the proposed method and the uncertainties linked to the misplacement of the tracer gas regarding the methane source. For each configuration, the methane and acetylene emission plumes were crossed 20–40 times (see table 1), and each serie of crossings were performed on the same day on a timescale of 1-2 hours. The usual increases in the acetylene and methane concentrations are 3–15 ppb and 50–500 ppb, respectively. Each measurement day was selected by taking the weather forecast into account and choosing days with a strong wind coming from the north to be able to measure the emissions from the parking lot on the measurement road. The average weather conditions of each campaign are summarized in table 1. The following sections describe the different components of the experimental and modelling systems used for the inversion of the methane sources.

3.2   Analytical equipments

Downwind gas concentrations were measured using a G2203 cavity ring-down spectrometer (Picarro, Inc., Santa Clara CA), which continuously measures acetylene (C_2H_2), methane (CH_4) and water vapor (H_2O). Based on infrared spectroscopy, the high precision of the system (precisions of 3 ppb and < 600 ppt for methane and acetylene, respectively, on 2 second interval) is due to its very long path length ($\simeq 20$ km) and the small size of its measurement cell (< 35 mL). Mobile measurements with such an instrument have already been successfully performed and published in previous studies (Mønster et al., 2014; Yver Kwok et al., 2015), thus demonstrating the potential of this method.

During the field campaigns that we organized for this study, the system was set-up in a car and powered by the car's battery. The air sampler was placed on the roof at approximately 2 metres above the ground with a GPS (Hemisphere A21 Antenna) to provide the location of the measurements. The sampled air was sent into the instrument by an external pump system allowing a short response time between the sample inlet and the measurements (less than 30 seconds). Despite the relatively fast response time of this system, the direct comparison of the measured and modelled tracer concentrations introduced a time shift that was more or less constant (see section 2.5). The corresponding spatial offset is well characterized by the comparison between the modelled and measured acetylene plumes, and it is accounted for when comparing the modelled and simulated methane plumes (see section 3.5).

3.3   Tracer and target gas release

Acetylene is commonly used as a tracer. Due to its low concentration in the atmosphere ($\simeq 0.1$–0.3 ppb), any release is easily detected. Acetylene also presents the benefit of being inert, and thus, negligible loss during the transport process is expected (Whitby and Altwicker, 1977). Other gases are suitable as tracers, such as SF_6, but acetylene is preferred because it is not a greenhouse gas. However, due to its flammability, its use requires specific precautions.

An acetylene cylinder (20 L) containing acetylene with a purity $> 99.6\%$ was used as the tracer source. A methane cylinder (50 L) with a purity of 99.5% was used for controlled methane release. The flows of both gases were controlled by a 150 mm flow metre (Sho-rate, Brooks) able to measure fluxes between 0 and 1500 L.min^{-1}. The different acetylene and methane emission rates were checked by weighing the cylinders before and after each test and timing the release duration. The amount of acetylene emitted was adjusted such that its emission plume can be detected on the roads where the

measurements were performed while keeping it at the lowest rate possible to limit the risks associated with its flammability. In this study, we used emission rates from 65 to 90 g.h^{-1} for acetylene. During the measurement campaigns, the cylinders were attached with straps to a fixed frame to avoid any accidents.

3.4 Tested configurations

This section details the four configurations utilized during this campaign for estimating the accuracy of the proposed method and the uncertainties linked to the misplacement of the tracer gas regarding the methane source (figure 1). The first configuration consisted of a collocated emission of acetylene and methane. This configuration enabled us to estimate the accuracy of the method and our system under optimal conditions. One cylinder of methane and one cylinder of acetylene were placed on a parking lot and connected together by a tube with a length of a few metres. This system aimed at ensuring the mixing of both gases and was designed to be as close as possible to the ideal situation in which methane and acetylene are emitted at the same location and under the same conditions. In principle, under such conditions, the tracer release experiment should provide a perfect proxy of the methane transport and should provide better estimates with the classic tracer release method than with the statistical inversion that relies on an imperfect, although optimized, modelling of the methane plume.

In reality, in industrial sites, methane source locations are not always well known, or it may be difficult to access these sources and place a tracer cylinder next to them. The second and third configurations tested the impact of non-collocated emissions. To represent this situation, one cylinder of methane and one cylinder of acetylene were used, and the methane cylinder was moved i) approximately 60 metres downwind from the acetylene bottle location (second configuration) and ii) approximately 35 metres laterally compared with the wind direction (third configuration).

Finally, during measurements on real industrial sites, several sources of methane may be encountered within the same site. The fourth configuration tested the influence of several methane sources on the estimation of their fluxes when one tracer source is used. For this purpose, a system of two tubes was connected to the methane cylinder, splitting its exhaust into two locations approximately 35 metres apart. The acetylene cylinder was collocated with one of the exhausts.

The advantage of the combined method proposed in section 2.5 over the traditional tracer release technique (which relies on the collocation of the target and the tracer gas sources) should be revealed in these last three experimental configurations. In the fourth case, the ability to quantify the total emissions from different sources (which cannot be achieved with the tracer release technique in our experimental framework due to the strong overlapping of the plumes from individual sources) is evaluated.

3.5 Gaussian model and inversion settings

In this section, we provide details on how the parameters of the Gaussian model are adjusted and how we set-up the different matrices and vectors corresponding to the application of equation 6 for the statistical inversion.

As explained in section 2.1, f is the control vector, which contains the targeted emission rates of the methane sources, and p is the observation vector. In this study, two different possibilities of calculating this vector are used. On the one hand, when we want to estimate the emission rate of a single source of methane (such as in configurations 1, 2 and 3), p corresponds to the integration of the entire methane plume concentrations above the background and thereby called p^{ent}. On the other hand, for estimating the emission rates of several sources of methane, the observed methane signal is divided into five slices of equal time, with each slice p_i being integrated and gathered into the observation vector p^{slc}.

Regarding the observation operator H, which describes the atmospheric transport from the point sources to the measurement transects simulated with the Gaussian model, the model responses are simulated for the methane sources with a unit rate and the specific wind conditions of each cross-section. In a way that is consistent with the definition of p, depending on the number of methane sources, the modelled methane concentrations above the background are either entirely integrated for a single source or split into five parts equal in time, with each part being integrated for several methane sources. These model responses are simulated with the Gaussian model parameters selected based on the acetylene data. Indeed, for each transect, the measured tracer concentrations are compared with the modelled tracer concentrations forced with the known tracer emission rate for different Briggs stability classes. The selected Briggs parameterization corresponds to the parameterization for which the integration of the

modelled tracer concentrations above the background is the closest to the value of the measured tracer concentration integration. The background concentration for each plume is defined as the value of the 5th percentile of the transect concentrations. Figure 2 illustrates this model parameterization selection. In this example, the concentrations modelled with the stability class B best fit the measured concentrations, which are represented in black. In some cases, the model cannot "reasonably" reproduce the observations due to the presence of large turbulent structures or transport conditions that are extremely unfavorable for the model (swift wind change and/or low wind conditions). When the relative error between the modelled integrated concentrations and the measured integrated concentrations is higher than 70%, the transect is removed from the analysis. This value of 70% is an empirical choice that has been defined with our dataset. In theory, the strategy of computing the statistics of the model error as a function of such misfits should ensure that the weight given to these transects in the inversion is low. However, in practice, we conservatively prefer to remove transects for which the confidence in the model is extremely low.

As explained in section 2.5, the modelling of the methane plume is shifted in time for comparison with the methane measurements using the offset between the tracer modelled and measured plumes. This offset is defined as the time between the maximum modelled tracer concentration and the maximum measured tracer concentration.

The modelled vs. measured tracer concentrations when using the optimized configuration of the transport model are used to set-up the variances of the observation error in the inversion configuration (i.e. the diagonal of the covariance matrix R). In the case of a unique methane source, we use the absolute value of the difference between the modelled vs. measured integration of the plume concentrations above the background to set-up the standard deviation of the observation error for the corresponding observation (i.e. for the corresponding transect). When there are several methane sources within a site, we use the absolute value of the difference between the modelled vs. measured integration of each slice of the divided plume concentrations above the background to set-up the standard deviation of the observation error for the corresponding observation (i.e. for the corresponding slice of a given transect). We assign a minimum value for these standard deviations to prevent one transect or slice of a transect to dominate too much over the others in the inversion process. In the least squares minimization process associated with the statistical inversion, a data assimilated with a considerably lower observation error than the others may fully drive the inversion results. However, for some transects, an excellent fit may occur between the model and the measurements in terms of integration of the emission plumes whereas, the shapes of the modelled and measured tracer plumes can be significantly different, revealing some significant model and/or measurement errors. Applying a threshold to the observation errors limits the impact of their underestimation through the objective comparison between the modelled and measured integrated tracer concentrations within the emission plumes or slices of plumes. We make the assumption that there is no correlation of the errors when modelling or measuring plume slices from one slice to the other slice of a given transect or from one transect to another one such that the R matrix is set up diagonally.

As explained in section 2.4, the Bayesian principle of statistical inversion exploits the statistical prior knowledge on the emission rate, i.e. their estimate f^b and the associated uncertainties. When monitoring methane emissions from waste treatment sites, farms, or gas extraction or compression sites, the typical prior knowledge of the emissions (from process models, typical national- to regional-scale factors) can bear more than 100% uncertainty and for many of these sites, the order of magnitude of the uncertainty in the emissions is not known. Despite working in the framework of a controlled release experiment, we attempt to set-up the inversion system to have the same conditions as when monitoring the emissions from such sites, and we thus set the standard deviation of the prior uncertainty in the targeted (even though well known) methane emission rate to 80% of their prior estimate, which is taken as its actual value in configurations 1, 2, and 3 and 288 g.h^{-1} for each methane in configuration 4. We also assumed that there is no correlation between the uncertainties in the methane emissions from different targeted sources within a site since they generally correspond to different processes (e.g. the aeration process and the clarification process in wastewater treatment plants (Yver Kwok et al., 2015).

3.6 Using the Gaussian model and a theoretical experiment with synthetic data to estimate the errors due to the mislocation of the tracer emission when using the traditional tracer release technique

The Gaussian model is used to quantify the error induced by spatial offsets between the tracer and methane sources via a type of observing system simulation experiment (OSSE) with synthetic data (Rayner et al., 1996; Chevallier et al., 2007). Different sets of locations and thus spatial offsets between the tracer and methane sources are tested. Initially, the different configurations mentioned above are tested to provide estimates of this error for the actual computations with real data, except for the first configuration, where the tracer and methane sources are perfectly collocated. In homogeneous meteorological conditions, if the measurement road is orthogonal to the wind direction, a lateral offset between the methane and tracer sources (i.e. orthogonal to the wind direction and parallel to the road) has in principle no impact on the estimate calculated with the tracer release technique, unlike a downwind or an upwind shift. Their emission plumes are simply shifted without any impact on the integral of the concentrations. However, in our configuration 3, the meteorological conditions are not homogeneous even if homogeneous meteorological conditions are assumed in the OSSEs, and the road is curvilinear, and thus, the measurements are not perfectly orthogonal to the wind direction, which induces an error due to the mislocation of the tracer. This is the reason for why this error is estimated with the OSSEs for configuration 3.

Then, the theoretical upwind and downwind offsets (from 20 to 200 metres) between the methane and tracer sources are tested to obtain more insights about the evolution of this error as a function of the spatial offset. In the corresponding OSSEs, we assume that the true methane and acetylene emission rates are those used for the experiments with real data. The synthetic methane and acetylene concentrations are simulated with the Gaussian model forced with these emission rates and similar weather conditions as during the campaign. The corresponding emission plume transects for both gases are integrated along the same paths as during the campaign as well as for hypothetical orthogonal paths across the plumes at different distances (from 100 to 2750 metres) from the methane source to provide a general assessment of the evolution of the error due to spatial offsets between the methane and acetylene sources as a function of such a distance. The plume integrations are used to estimate the methane emission rate using the same formula as the tracer release technique (equation 2). The resulting methane emission rates are compared to the "true" one. The comparison provides a direct estimate of the error associated with the fact that the location of the tracer emission does not accurately fit with the location of the methane emission since in these computations, the same model configuration is used to simulate the acetylene and methane concentrations, and we ignore measurement errors.

4 Results

We now present and discuss the methane emission estimates from controlled point sources calculated with the tracer release technique (section 4.2) and with the combined statistical approach detailed in the previous sections 2 and 3 (section 4.3). These results are compared with the known methane emission rate to illustrate the ability of each method to derive a good estimate of the emissions. We also analyse the covariance matrix A of the theoretical uncertainty in the emission estimates when using the statistical approach (equation 7), which provides a complementary assessment, according to the inversion of our own diagnostics, of the reliability of the results and of the level of separability between the two methane sources in configuration 4 of the experiments.

Standard deviations from this A matrix should provide the total uncertainty in the emission estimates, accounting for all sources of error when using statistical inversion, assuming that they are random and unbiased and have a Gaussian distribution. When using the tracer release technique, one way to estimate the uncertainty in the average estimate is to analyse the standard deviation STD_{tr} of the individual inversions based on each measurement transect and to derive it as $\frac{STD_{tr}}{\sqrt{n}}$, where n is the number of transects. This would assume that the sources of errors have the same statistical distribution for each transect, whereas the statistical inversion is set-up based on transect-specific diagnostics of the model error. Furthermore, the error associated with the mislocation of the tracer emission is assumed to generate a bias in the computation rather than a random error since the measurements are taken in a

relatively narrow range of downwind positions. We thus use estimates of this error from the OSSEs (see section 3.6) to complement the assessment of uncertainty in the results from the tracer technique, with the standard deviation of the total uncertainty being taken as the root sum square of the bias due to the mislocation of the tracer estimate and of the standard deviation of the random uncertainty derived from the variability of the results from one transect to the other one (i.e. $\frac{STD_{tr}}{\sqrt{n}}$). Therefore the error bars associated with the results from the tracer release technique and from the statistical inversions rely on different assumptions depending on the approach but they can still be compared in the sense that they should in principle both cover all sources of uncertainties.

4.1 Observing system simulation experiments to estimate errors due to a mislocation of the tracer

This section presents the results from the OSSEs described in section 3.6. The bias in the estimates of the methane source with the tracer release technique due to the mislocation of the tracer regarding the methane sources is estimated to be 69.2% for configuration 2, 36.3% for configuration 3 and 73.0% for configuration 4 when the measurements are taken along the transects corresponding to the actual experiments with real data. Considering the amplitude of these errors, we can expect that our combined statistical approach has a high potential for providing better estimates than the tracer release approach for configurations 2, 3 and 4.

OSSEs are also used to examine the evolution of this error as a function of the spatial offset and to provide more insights about the influence of the distance between the sources and the measurements. The impact of several theoretical offsets along the wind direction (upwind and downwind) between the methane and tracer (from 20 to 200 metres) is evaluated for different configurations of the measurements with transects through the emission plumes at 100 to 2750 metres from the methane source. The corresponding estimates of errors are presented in figure 3 (with the results for the downwind and upwind shifts provided in figures 3a and 3b, respectively). The OSSEs confirm the importance of the collocation between the tracer and methane sources for the tracer release technique when the offset between the tracer and methane sources is along the wind direction.

When the tracer is released upwind of the methane source, the emission rate is overestimated because of the atmospheric diffusion, which makes the integral of the released tracer concentrations through the emission plume lower than if both sources were collocated. The opposite occurs if the released tracer is placed downwind of the methane emission location. In the following, we characterize the biases by their absolute value and the fraction of the actual source that they represent.

When the tracer source is either upwind or downwind of the methane source by more than 100 metres and the measurements are taken at less than 300 metres, the bias exceeds 40% of the actual value of the source. The errors due to upwind shifts are generally similar to the errors due to downwind shifts over the same distances. The more distant the measurements are performed, and the less impact the shift distance has. When the measurements are taken at more than 1200 metres, the bias due to the mislocation of the tracer becomes less than 20% of the actual value of the emissions but at such distances, the signal to measurement noise ratio would likely be too small to derive precise estimates of the emissions.

4.2 Tracer release method estimates

The time series of acetylene and methane measurements for each tracer release experiment are shown in figure 4. Figure 5 presents one example of the measured acetylene and methane cross-sections used for calculating the methane emission rate for each campaign. For the first campaign, both the acetylene and methane profiles are similar due to the collocation and the mixing of the sources, but we can still observe a significant difference between both emission plumes. The shift between the sources is reflected by a smaller relative amplitude and a higher relative width of the acetylene plume compared to the methane plume in configuration 2 than in configuration 1 and by a lateral shift of the acetylene plume compared to the methane plume in configuration 3. At first sight, we distinguish only one methane emission plume in the measurements for configuration 4 because the plumes of the two methane sources strongly overlap, which does not strongly support the separation of these sources when analysing such a signal but which should help the tracer release technique to derive the total estimate of the emissions from the two sources. The emission rates calculated using the tracer release technique in this section

are the averages of the rates calculated with the different crossings, and the corresponding estimates of uncertainty are described in the introduction of this section 4. Table 2 lists the estimated methane emission rates and the methane emission rates actually released for each configuration tested.

In the first configuration, the average methane emission rate is estimated to be 454 ± 50 g.h^{-1} using the plume integration approach. The set of the methane emission estimates for each cross-section follows a Gaussian distribution. The actual controlled methane release rate was 382 ± 7 g.h^{-1}, which is 19% lower than the averaged emission estimate and slightly more than one standard deviation of the assumed uncertainty in this average estimate. This misfit is associated with the measurement error because we are working with low emission sources, which can be challenging to quantify and which have an important influence on the measurement error.

The second configuration with the released tracer emitted 60 m upwind regarding the methane source provides an estimation of 551 ± 299 g.h^{-1} of methane using the plume integration approach, whereas the actual amount of methane released was 428 ± 7 g.s^{-1}. The relative difference between the estimated and actual values of the emissions is thus 29% and is logically larger than that in configuration 1. The uncertainty in the estimate encompasses the actual value of the emissions, mainly due to the bias associated with the mislocation of the tracer release.

In the third configuration, where acetylene is emitted with a lateral shift of 35 metres from the methane emission location, the estimated rate is 421 ± 158 g.h^{-1} for an actual methane emission rate of 360 ± 7 g.h^{-1} (plume integration approach). The relative difference between the values is 17% and is on the same order of magnitude as when the acetylene and methane sources are collocated, which can be explained by the weak impact, in theory, of the lateral shifts on the tracer (see section 4.1) and by the fact that given the short distance between the two emission points, both emission plumes are subject to similar meteorological conditions. The uncertainty in the estimate of the emission rate again well encompasses the actual value of the emissions.

In the fourth configuration, we attempted to use the tracer release technique to retrieve the total emission rate of two methane sources separated by 35 metres with the tracer collocated with one of the methane sources. The total actual methane emission rate was 482 ± 7 g.h^{-1}, and this flux was equally split into 241 ± 7 g.h^{-1} at each location. Using the tracer release method and the plume integration approach, the total emission rate is estimated to be 760 ± 358 g.h^{-1}. The relative difference between this estimation and the actual estimation is thus 58%, which is considerably larger than in the previous configurations. However, the actual emissions are within the range of one standard deviation of the uncertainty in the emission estimate for which the bias due to the tracer mislocation should be very large.

4.3 Combined approach

Figure 6 presents examples of results obtained using the combined statistical approach with one or several methane sources. The behaviour of the inversion system and the values in the concentration and observation space are illustrated for one transect only. The graphs on the left side show the prior and the posterior fluxes in blue and green, respectively, which are compared with the real emission rate in white with one methane source in the upper case and two sources in the lower case. The graphs in the centre present modelled concentrations with the prior and the posterior fluxes in comparison with the measured concentrations. The graphs on the right side present the indices used in the observation vector, i.e. integrations of the atmospheric emission plume above the background. In the upper case with one methane source, the entire plume is integrated, and in the lower case with two methane sources, the plume is divided into five slices equal in time and the observations correspond to the integrals over each of these slices (see section 3.5). These graphs illustrate that the posterior estimates of the fluxes have a better fit to the actual emission rate than the prior estimate. They also show that the simulated concentrations fit the measurements far better after inversion than when considering the prior simulation.

For the first configuration, the methane emission is estimated to be 472 ± 2 g.h^{-1}. Although this estimate is on a good order of magnitude, as expected, the tracer release method provides a better estimate (table 2). Indeed in this configuration, where the tracer and the methane are collocated and well mixed, the tracer is a better proxy of the atmospheric transport than the model. Again, this case is rare in real industrial cases.

For configuration 2, the statistical inversion provides an estimate of the total methane emission of 464 ± 1 g.h^{-1}, which is a better estimate than with the tracer release method. The combined approach can

clearly reduce the estimate error linked to a downwind or an upwind mislocation of the tracer. A similar improvement can be observed for configuration 3, where the statistical inversion provides an estimate of 360 ± 2 g.h^{-1}, and for configuration 4 (with two methane sources, one located next to the tracer) where the estimate of the total emitted methane is 482 ± 25 g.h^{-1}. In all these cases, the statistical inversion predicts a very low standard deviation of the posterior uncertainty in the emission estimates. This could appear optimistic (the diagnostics of these errors strongly rely on our set-up of the prior uncertainty and in our derivation of the model uncertainty), and in practice, for configurations 1 and 2, this standard deviation does not encompass the actual emissions. However, this weak estimate of the posterior uncertainty still reflects the very good fit between the inverted and actual total emissions in configurations 3 and 4. In general, the lower bars of uncertainties that we derived for the statistical inversion than for the tracer release are supported by the better results obtained with the combined approach.

For the partitioning of the two sources in configuration 4, the combined approach provides an estimate of 173 ± 14 g.h^{-1} and 310 ± 25 g.h^{-1} for the two methane sources when both sources actually emitted 241 ± 7 g.h^{-1}. This weak ability to separate the signal from each source because of their close location is confirmed by the strongly negative correlations (-0.41) of the posterior uncertainties in these sources (diagnosed from matrix A).

5 Conclusions

We propose a new atmospheric concentration measurement-based method for estimating the transient emissions of gas from point sources or more generally from industrial sites. This method is based on a combination of the tracer release technique, Gaussian plume modelling and a statistical inversion framework. The concept is to optimize the model parameters based on the knowledge provided by the tracer release and concentration measurement and to exploit tracer model – measurement misfits to prescribe the statistics of the modelling error in the statistical inversion framework. Compared to the traditional tracer release technique, the method has the advantage of exploiting the knowledge on the atmospheric transport provided by the known tracer known release and measured concentration without relying on the collocation of the tracer emission and of the targeted gas emission (which can hardly occur in real study cases). The statistical framework accounts for the different sources of uncertainties in the source estimate and for solving different targeted sources together and to consider any valuable number of pieces of information in the measurement of the targeted gas for such an inversion.

The advantage of this approach compared to the traditional tracer release technique is evaluated with field experiments close to our laboratory of controlled emissions of tracer gas (acetylene) and targeted gas (methane) under four spatial configurations and of mobile measurements of the acetylene and methane concentrations across their emission plumes. This set of experiments allowed us to prove that both approaches can provide consistent estimates of the transient targeted emission rates for each configuration. We showed that in the simplest case with one source of methane collocated with the tracer and well mixed, the tracer release method is the best approach for estimating the targeted emission rate since the tracer is a better proxy of the atmospheric transport than the Gaussian model, even if the configuration of the latter is optimized based on the tracer release and concentration measurements. However, this case is the most unlikely for real industrial sites, where the released tracer may be located away from the targeted sources which are generally multiple and spread over significant surfaces. We demonstrated that the mislocation of the released tracer induces large errors that depend not only on the distance between the tracer and the targeted sources but also on the distance between the measurements and the sources based on OSSEs. In these cases, we illustrated that the calibration of a Gaussian model using the tracer release method and the integration of the calibrated model in a statistical inversion framework help to reduce this error and to provide a better estimate of the point total emissions. We also illustrated the potential ability of the statistical inversion to separately estimate the different emission rates from multiple sources even if the two targeted sources of methane were too close such that their plumes were hardly separated by the inversion in the experiments conducted for this study.

During measurement campaigns on actual industrial sites, the locations of the methane sources are not exactly known as in our tests. This lack of information could induce additional uncertainties to our estimates. Another source of uncertainty is the fact that in the tested configurations, methane point sources were used whereas during field campaigns, spread and fugitive sources may be encountered.

The methodology proposed herein should thus now be evaluated and potentially refined for the monitoring of sources in actual industrial sites. Finally, this study and the tracer release technique target transient estimates of the emissions due to the inability to release a tracer continuously over long time periods. However, the combined tracer and modelling approach proposed here open the possibility to continuously monitor the emissions from a site using fixed continuous measurements and routine statistical atmospheric inversion. Sets of tracer release experiments for typical wind conditions would be used to optimize the model and inversion parameters for the continuous inversion of the emissions.

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Table 1 – Weather conditions during the four tests and configuration of the observation vector for the statistical inversion.

Trace gas configuration	Weather conditions (avg.)			Total number of transects	Number of selected transects	Configuration of the observation vector for the statistical inversion
	Temperature (°C)	Wind direction (degree)	Wind speed (m.s^{-1})			
Configuration 1	9.9 ± 0.3	N	3.2 ± 0.6	29	11	Integration of the entire plume
Configuration 2	9.2 ± 0.1	N	3.7 ± 0.8	20	9	Integration of the entire plume
Configuration 3	8.4 ± 0.8	N	2.5 ± 0.7	35	10	Integration of the entire plume
Configuration 4	11.3 ± 0.3	NW	2.0 ± 0.7	40	8	Integration of slices of the plume

Table 2 – Methane emission rates of the different controlled release configurations estimated with the different approaches and methane fluxes actually emitted during these tests. The uncertainties given with the tracer release method are detailed as follows: total uncertainty (standard deviation of the random uncertainty derived from the variability of the results from one transect to the other one ; bias due to the mislocation of the tracer).

	Configuration 1	Configuration 2	Configuration 3	Configuration 4
Controlled methane release (g.h^{-1})	382 ± 7	428 ± 7	360 ± 7	482 ± 7
Tracer release method estimates (g.h^{-1})	454 ± 50 (50 ; 0)	551 ± 299 (44 ; 296)	421 ± 158 (89 ; 131)	760 ± 358 (65 ; 352)
Relative difference to the control release (%)	19	29	17	58
Combined approach estimates (g.h^{-1})	472 ± 2	464 ± 1	360 ± 2	482 ± 25
Relative difference to the control release (%)	24	8	0	0
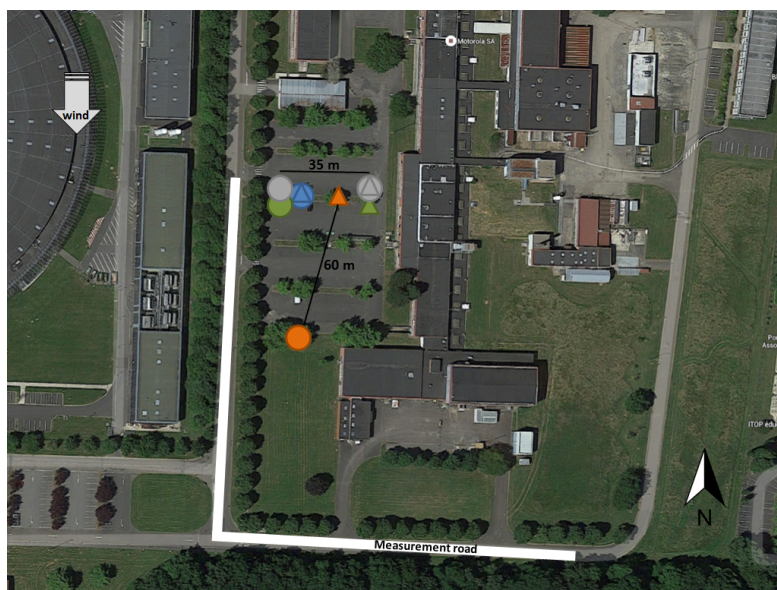
Figure 1 – The four tracer release configurations tested. Triangles represent the tracer source locations, and the circles mark methane sources. Each colour represents a configuration: blue is configuration 1, red is configuration 2, green is configuration 3 and, grey is configuration 4.
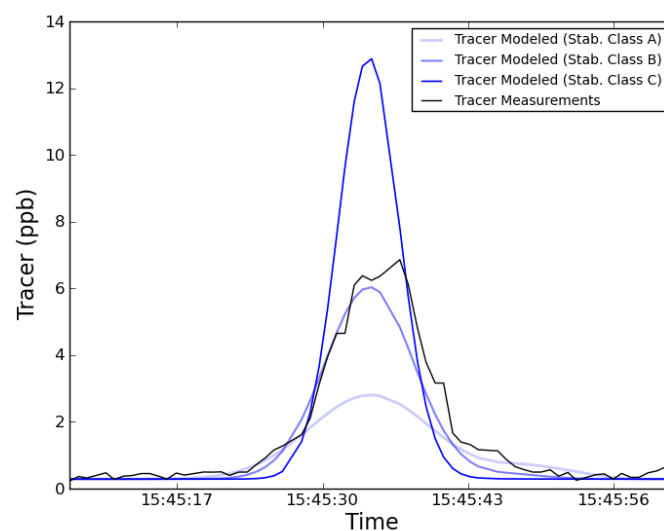
Figure 2 – Example of the Briggs parameterization selection with the acetylene data for peak 5 of configuration 2. The measured concentrations are presented in black, and the modelled concentrations with different stability classes are shown in blue.

(a) *Downwind tracer shift*   (b) *Upwind tracer shift*
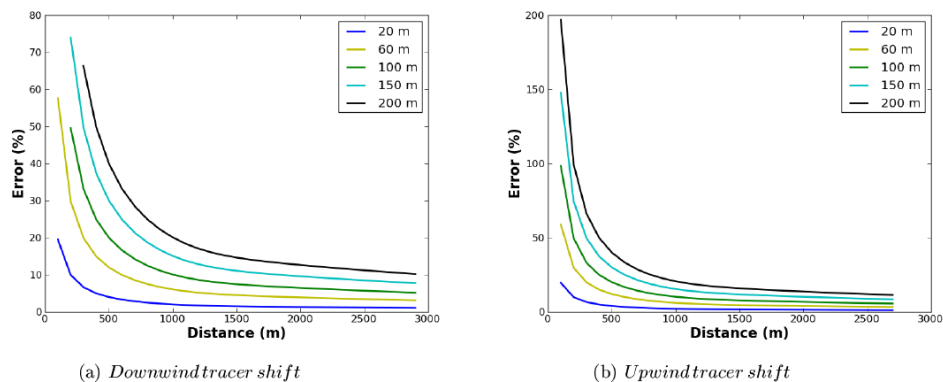
Figure 3 – Error in plume estimation with the tracer method depending on the measurement distance to the methane source and a shift of 20, 60, 100 150 and 200 m of the tracer location relative to the methane source using our Gaussian model.
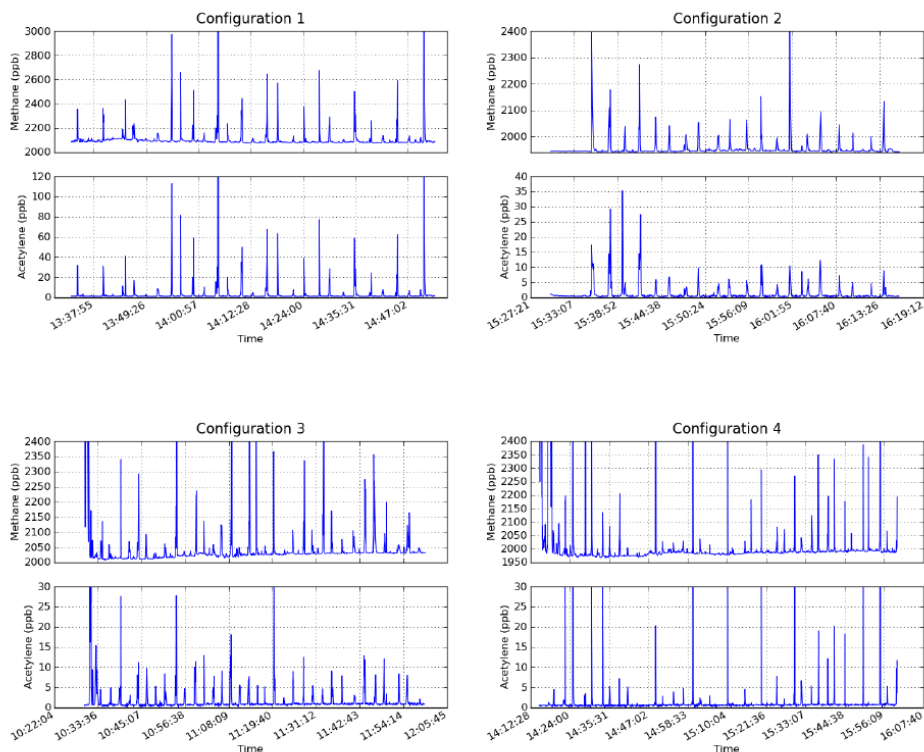
Figure 4 – Concentrations of methane and acetylene during the four tracer release experiments.
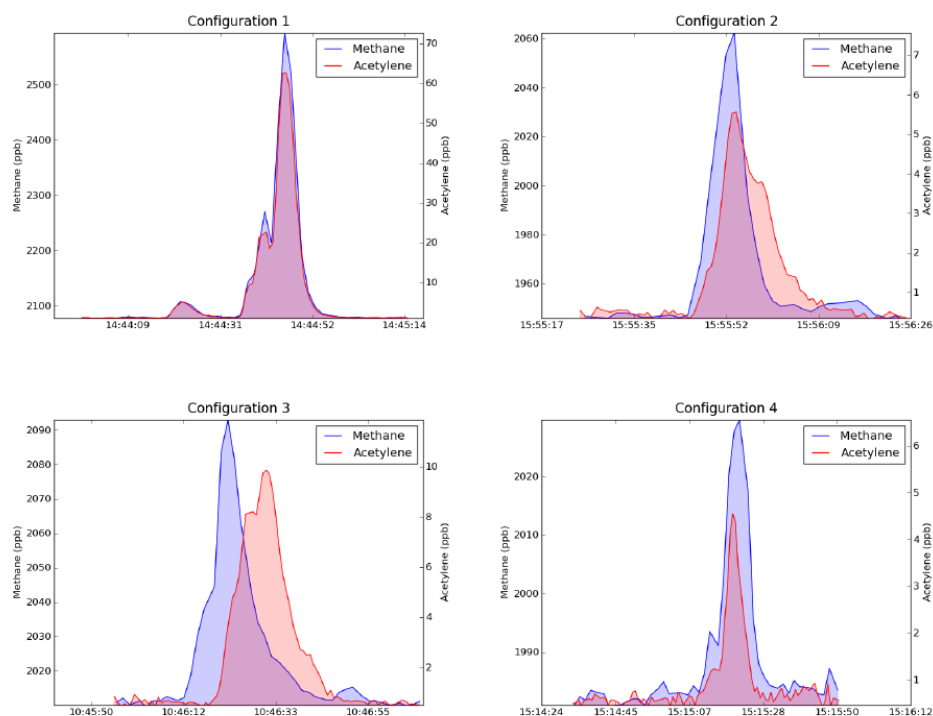
Figure 5 – Examples of cross-sections of the measured emission plumes of acetylene and methane (in red and blue, respectively) for each configuration.
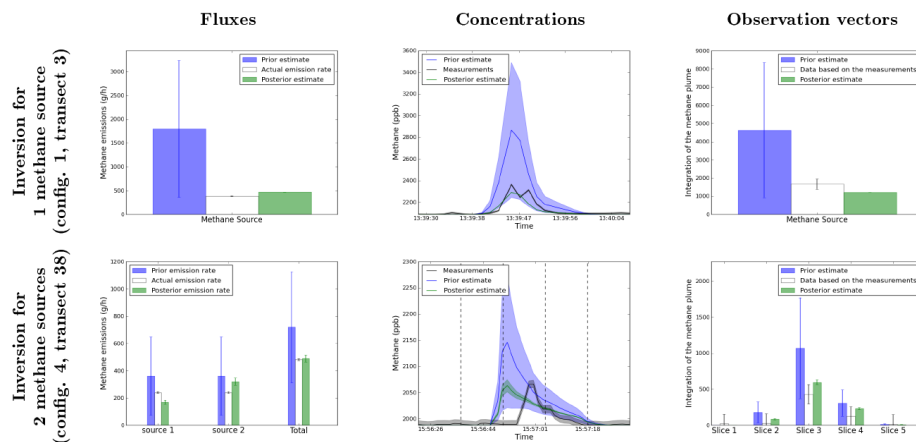
Figure 6 – Examples of prior, posterior and measured values of emission rates, concentrations and values of the observation vector for cases in configuration 1 and 4 (observations from a single transect shown).