Anonymous Referee #1

Received and published: 12 January 2017

The authors propose a new method to estimate emission rates in industrial sites, combining the tracer release method, a Gaussian dispersion model and a statistical atmospheric inversion approach. The method is evaluated through field experiments and conclusions are drawn based on related results.

GENERAL COMMENTS. The idea of the authors is interesting, since it combines and merges different approaches. In their intention, this should bring benefit to the assessment and estimation of emission rates in complex industrial site. The scientific approach is honest, by detailing all possible problems, and rigorous, by trying to address them. Indeed, the method is comprehensive and at the same time complex itself.

We thank the reviewer for this general assessment and for his constructive, detailed and technical comments that will help strongly improve our manuscript, and in particular the presentation of the objectives, concepts, and long-term perspectives of our study.

As the authors discuss thoroughly in several parts of the manuscript, there are many degrees of freedom, many sources of possible uncertainties and errors, many unavoidable approximations, which can affect the reliability of the approach. For these reasons, I have not been fully convinced of the feasibility and applicability of the method. The conclusions are drawn from enough rigorous experimental tests but in a limited number and for a single site. Thus, it is not straightforward to infer whether the method can be effectively generalized, leading to a final novel procedure for estimating even unknown emission rates from industrial sites.

We assume that the success of this method, and, at least, its ability to provide better results than the traditional tracer release technique will depend on the configuration (size and extent of the sources, topography, local transport conditions, positioning of the roads…) of the industrial sites to be investigated. But, at least, this paper demonstrates, both theoretically and for a practical case, that it in principle, it has some potential to behave better than the tracer release technique (which is used quite extensively for the estimate of industrial sources nowadays: Roscioli et al. 2015, Goetz et al. 2015, Taylor et al. 2016). We think that this is definitely worth leading to new studies and tests of such a concept and analyzing how to improve and generalize it. Our OSSEs (see section 3.6) demonstrate severe issues when using the tracer release technique even for simple point source estimation cases, while the use of this method is definitely generalized, highlighting the need for exploring new techniques. The proposed method is new, we naturally acknowledge the need for improving it (see the point regarding the transport model below) in order to limit the impact of the sources of uncertainties that we have discussed.

We will emphasize these points in the introduction and in the conclusion. Of note is that the conclusion will be expanded into a sort of discussion/conclusion section to gather many of the digressions that lengthened the previous sections and to include discussions asked by the reviewers.

I have some specific concerns on the use of a Gaussian model. Certainly, for such short distances it can be reasonably applicable, as the author discuss, and its simplicity allows better dealing with the complexity of the problem. However, choosing a Gaussian model is questionable, since it cannot capture and describe turbulent motions, thus missing the fine-scale structures that can affect the results and their analysis. It has to be considered that turbulence and stochastic motions may produce uncertainties, for which also the tracer release method can fail, since turbulence acts altering the plume spread and pollutant dispersion. I think these aspects need to be better ad-

dressed and discussed in the manuscript, even if already several comments are spread in the text of this version, but too sparsely.

We will expand the discussions on these aspects.

Of note, regarding the use of the Gaussian model in this study:

First, by jointly assimilating the data from the different transects through the methane plumes in the inversion, we attempt at imposing a sort of "average plume" as the main constraint on the emissions. This should limit the weight of fine-scale structures.

Second, the lack of turbulent structure in the Gaussian simulation is implicitly accounted for when assessing the model error based on C_2H_2 model vs. data comparisons. So the uncertainty in the inverted emissions associated with the lack of turbulent structures is included in the diagnostic of uncertainty by the inversion system.

At last, the success of our method with a Gaussian model compared to the tracer release method in our practical test demonstrates the relevance of using such a model for the first assessment of our inversion concept.

Models more sophisticated than Gaussian models could produce turbulent motions but one would hardly manage to control them for capturing (in the sense of having them at right time and location) that seen in the data especially with the available dataset from the type of measurement campaigns we consider. The reviewer criticizes this assumption (see comment on 2.3 and our answer to this comment) and we are ready to be convinced by his considerations on this. Furthermore, using more complex models would allow to account for complex topography, for temporally and spatially varying local meteorological conditions… We thus now prefer to emphasize that testing our theoretical concept first with a model as simple as a Gaussian model before increasing the complexity of the problem made sense since our results demonstrate some advantages of this modeling framework compared to the tracer release technique.

These points will be better emphasized in the manuscript. Furthermore, section 2.3 (which will be split into 2 subsections: a general but short one on local scale transport models and a more specific one on Gaussian models) and the conclusion will acknowledge that some improvement of the method could be achieved by using models more complex than Gaussian models, at least for applications in places with complex topography or situations with complex meteorological conditions, even though we feel that exploiting the capability of such models to generate turbulent patterns would not be straightforward.

Regarding the manuscript and its form, the text is very dense, with a huge amount of information and comments, and avoidable repetitions. In some parts it gets strenuous to keep trace of the work done until first results are presented and finally discussed. The authors made an effort to provide a good organization of the paper structure, yet I think that it needs improvement, optimizing the description, removing repetitions, avoiding verbosity. The content and the form of the manuscript somehow are paired, in that a long and detailed description of the method and of the work done (almost 11 pages) then flow into confined results and conclusions (about 3.5 pages, plus tables and figures).

We will try to use more subsections, use references to such subsections to avoid redundancies and more generally try to be more concise. We want to keep the general structure as it is and gather sections 3 and 4 into a single section 3 since we feel that this paper has definitely two components: the definition of a new theoretical framework in one hand (the present section 2), and its evaluation through an academic experiment (the present sections 3 and 4). It is thus difficult to compare the length of the present section 4 to that of the present sections 2 and 3.

I think that the manuscript needs further revision before considering it for publication: more detailed comments are provided hereafter.

We hope that our answers will provide convincing indications of the improvement we plan for the paper.

DETAILED COMMENTS.

*** Introduction.**

Page 2: why the locations of pollutant sources in industrial sites can be 'not always precisely known'? Because of possible 'fugitive emission' or leakages, or missing information from the industries? It will not be a matter of 'geo-localization', nowadays.

Fugitive emissions (like leakages) that are transitory, or affecting poorly reachable areas or complex buildings, unexpected sources, but also widespread and heterogeneous sources (e.g. livestock in the building of a farm, basins in waste water treatment plants or cells in landfills for which emissions are not homogeneously distributed) makes it difficult to know perfectly the distribution of the emissions within a site. This sentence will be modified and extended to clarify it. In particular we will change "location of pollutant sources" into "spatial distribution of the emissions".

Page 2: 'local atmospheric dispersion models' might be of various type, from simple parametric methods to Gaussian, Eulerian, Lagrangian models. Since the authors cite 'models based on mass conservation' they should better specify that they are here referring to 'simple mathematical inversion' methods. In fact, advanced dispersion models are able to account for 'complex turbulence structures'.

We will improve this introduction on the local dispersion models mentioning families of available models (CFD/LES, Lagrangien…).

Page 3: the authors state that the skill of statistical inversions approach strongly rely on the transport and source modelling. Then, a Gaussian model is used for the study. Gaussian models have strong limitations, especially when accuracy and turbulence structures are important factors, also given that they are designed for homogenous conditions. This is partly discussed later on, but some justification to support the use of a Gaussian model should be given here already.

See our general answer to the similar general comment above regarding the turbulent patterns. We will also try to better discuss the limitations of the method when using a Gaussian model, which could limit its applicability to simple cases in terms of topography and meteorology. The conclusion will remind that, in principle, the general concept of the inversion could be used with other types of models for the sake of improvement and / or generalization, but that the details of such an implementation would still have to be studied.

*** Section 2.2**

Page 5: the authors explain their choice to minimize the impact of the differences between the targeted and released tracer plumes due to a not perfect collocation of the sources. Since turbulent motions can enhance the differences between the plumes further downwind the sources, is this the optimal choice for any distance from the emission points?

We will better indicate that part of the impact due to a not perfect collocation of the sources can be emphasized by incompatible turbulent patterns between the tracer and targeted species. We assume that the author questions our choice of using the area below the plume instead of its maximum to characterize the plume. In principle, whatever the distance from the source, there is more chances that the maximum of the plume is more impacted than its area by the turbulent patterns than the opposite. But it definitely depends on the situation (on the structure of the plume). We have not thought about more sophisticated diagnostics of the plume that would be less sensitive to these turbulent patterns.

*** Section 2.3**

Page 5: I do not understand, and do not agree with the statement: "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." What do the authors mean? Why these models can be hardly set-up and controlled? Due to their complexity? No model can 'perfectly match' the turbulent patterns, but advanced models are in principle the best option, in particular when LES approach is used.

We agree that advanced models are in principle the best options, but we mean that their advantages do not seem easy to exploit for such an inversion problem. Our sentence is not in opposition with this but we will try to clarify and moderate it. We do not mean that these models are difficult to set-up, but that it is not straightforward to control them to ensure that their turbulent patterns are placed approximately well, a requirement to take advantage of it in inversions that make a simple minimization of simulated vs. measured data at different times and locations. Gaussian models seem a good choice for the first test of our method considering their very low calculation costs and the simplicity of their application.

Placed this way, this statement sounds just like a weak justification to use a simple Gaussian model, which on its side has instead severe limitations, since stationary solutions for homogeneous conditions are indeed a strong approximation of real atmospheric processes.

We will now better clarify in the introduction and conclusion that the use of a Gaussian model can be seen as a first step to test our theoretical framework with a simple model before increasing the level of complexity. The successes obtained with this model in our practical case demonstrate that this was relevant. And again, the conclusion will now indicate that future studies should investigate the use of more sophisticated models and the appropriate strategies to take benefits from their advantages for the sake of improvement and generalization of the method to site configurations with complex topography and local meteorological conditions.

Surely, advanced models (maybe available even in the Polyphemus system?) need more established modelling expertise and large computational resources for their application. It would be worth to include some discussion about the expected limitation when applying the Gaussian model in this specific site, where obstacles and buildings affect the flow and dispersion.

We will do it.

Page 6: Please, better explain in the text why "Instead of being deposited, the emission plume rebounds when it reaches the ground" is a 'decent' approximation for the studied gases.

We will replace "Instead of being deposited, the emission plume rebounds when it reaches the ground is a 'decent' approximation for the studied gases" by "As both studied gases are poorly soluble and chemically inert for the dispersion time scale we consider in this study it is relevant to neglect the mass loss due to dry deposition and assume a total reflection from the ground."

*** Section 2.5**

Page 7: the new method intends to overcome the issues associated with the individual usage of the different methods. Could the author foresee possible 'new' uncertainties and issues linked or due to the merging of three approaches? I mean, a sort of propagation of uncertainties, of error propagation? Given the discussion about the limitations of the single method, I wonder whether it is proper to consider 'a priori' that the information on the atmospheric transport from the tracer release method and the Gaussian model simulations can be defined as 'very accurate information'.

We will better demonstrate the moderation of our expectations from this method and its possible limitations. The model uncertainties are explicitly quantified and accounted for in the inversion system. The estimate of prior and model uncertainties which are required by the statistical inversion are definitely not straightforward even though we have data to support the derivation of the latter. Still, weighting such uncertainties in the estimate of emissions is safer than just ignoring them in "deterministic inversion" (based on the tracer or a transport model).

The final part of this subsection ("The statistics of the misfits. . .") is rather verbose, a bit compromising its clarity. The multiple references to topics treated in next sections indicate possible repetitions. I suggest revising this part, optimizing the links with next ones, being more precise and less descriptive.

Following this comment, we will rework the different parts to prevent these references and be more concise.

*** Section 3.1 In which hours of the day were the measurements performed?**

Measurements hours are detailed in figure 4, we will give this information more specifically in the section 3.1.

*** Section 3.3**

Page 8: please check the formalism accepted from the Journal for the units, if to use "l" for litre instead of "L", seconds "s" according to the SI instead of minutes "min" and hours "h".

We have not found the proper formalism of the journal for these units, but it should be corrected during the final steps of the editing process.

*** Section 3.5**

Page 9: regarding the general applicability of the method, 'specific wind conditions of each cross-sections' to estimate the H observation operator are not commonly/routinely available, when not provided from experiments: could the authors comment on this aspect?

We will indicate that the corresponding meteorological variables, if not measured during the experiments, are hardly accessible from local meteorological networks. Analyses by operational centers are available but their spatial representativeness is not always adapted to the characterization of local conditions. We thus strongly support conducting meteorological measurements along with the gas measurements if willing to use a modeling framework as here.

Page 10: some comments and interpretation about the best-fit obtained with stability class B (moderately unstable conditions) would be of interest: what were the atmospheric conditions during the experiment? Was B class effectively representative of them or the best agreement resulted 'by

chance'?

The measured wind speed during this series was 3.7 m/s. For this wind speed, the Pasquill classification indicates that the corresponding stability is whether B whether C depending on the solar radiation. The solar radiation information may be difficult to have, especially at this scale, and it is thus difficult to choose between these two stability classes without the tracer data. Therefore, in this example and for all transect, we have checked that the selected stability class is part of those that are in agreement with what we know of the meteorological conditions.

We will better discuss it in this section.

Here the authors 'admit' the limitation of the Gaussian model in reasonably reproducing the observed motion when turbulence, low wind etc may occur. So the question comes: why using a type of model that may not fit the purpose of its use?

The main purpose is to catch some information about the emissions characterized by the "mean plume" of methane rather than by the turbulent patterns on the top of it. Figure 2 indicates that once its stability class is optimized, the model fits very well with the 1st order pattern from the tracer source which is our aim so we do not agree that our model does not fit the purpose of its use. Again, in a more general way, the success of the method compared to the classical tracer release method for our specific test case demonstrate that the Gaussian model already provides a relevant skill for our objective. But, again, we now indicate in conclusion that more complex models could help improve the accuracy of the estimation and generalize the applicability of the method.

Also: how the threshold of 70% relative error was chosen to remove the data? Saying that the empirical choice has been defined based on the dataset is not enough, what was the reasoning behind?

This threshold fits with our qualitative analysis of the comparisons between measured and simulated tracer concentrations since it removes the transects for which we consider that the shape of the measured and simulated tracer plumes are quite different (simulated transect being wider than measured transects in most of these case) whereas for the other transects the Gaussian model managed to represent quiet well the measured profiles. Still, this conservative choice is not critical since the model error associated with each transect in the atmospheric inversion is based on the fit between the modeled and simulated tracer plumes, so that transects for which this fit is rather low do not bring a significant constraint on the inversion results.

Again, the cross-references to previous section 2.5 and 2.4 suggest that this part of the text should be optimized and harmonized with the previous one to avoid repetitions. Also, the description of the choices for the variances set-up might be improved, making it less descriptive and clearer.

We will make sections 2.4 and 2.5 shorter by detailing the specific concepts for the adjustment of the model data comparisons mostly in section 2.5 and we will improve the presentation of the variances.

*** Section 3.6**

Minor: the title is rather long and descriptive, surely it is possible to shorten it. The authors may consider to combine this section with next section 4.1, since a few items are repeated and 3.6 is in fact functional to the results presented in 4.1.

We will replace this title by: "Estimation of the biases of the tracer release method due to the mislocation of the tracer with theoretical model experiments". We will try to follow the reviewer's suggestion to merge the present 3.6 and 4.1 sections in the new section 3.

*** Section 4.1**

See previous comment on section 3.6.

*** Section 4.2**

Page 13: it is redundant to repeat in the text the numbers already reported in the cited Table 2. Please revise this part to avoid such redundancy and repetition.

We will rework this section to be more concise.

*** Section 4.3**

Same as for Section 4.2 about redundancy with Table 2. It would be worth to discuss more in depth why the tracer release method is better than the combined approach for the configuration 1. Because of fewer sources of uncertainties?

We will also rework this section to be more concise. Yes, with the configuration 1, all the sources of uncertainties for the tracer release method are also sources of uncertainties for the statistical inversion: the measurement error, the difference of time between the C_2H_2 and CH_4 measurements (which implies some uncertainties that were underestimated in the present version of the manuscript), the uncertainty in the background signals (a term which was under-estimated in the present version of the manuscript), and the uncertainty in the debit of methane and C_2H_2 (which is rather low). Since, the statistical inversion is also impacted by the transport modeling error and the uncertainty in the estimate of the prior and model error statistics, it cannot perform better than the tracer release method in such an experimental configuration. We will better emphasize it.

*** Section 5.**

In the results analysis and conclusions there is not a definitive and decisive proof that the combined method provides better and more reliable results than the tracer release method, given the limited number of cases and conditions considered, and the connected uncertainties.

We do not look for a definitive and decisive proof of the success and superiority of our concept (see our answer to the reviewer's first general comment). We just want to demonstrate, in this study, that it has some potential. For the four tests that have been led, the results, considering the uncertainty estimates or not, were in line with what we expected based on our theoretical concept. For each of the four tests, more than 15 transects of measurements are conducted through the plume. This is similar to past studies on industrial sites (Yoshida et al. 2014). We will better discuss the need for far more tests and studies to evaluate more precisely the robustness and potential of the method and improve it, but, according to us, these results are very promising and quite clear regarding the potential of the approach.

In addition, the potential ability of the method for multiple sources could not be fully addressed. The authors honestly recognize this and it becomes clear that these are 'preliminary' results and that more experiments are needed.

Yes, this paper show that in practice, the separation of sources is not straightforward even with the statistical inversion and more studies will be needed to improve it. However, the experiments show that the method can provide more precise estimate than the tracer release technique.

Thus, the paper is mostly the presentation of a method but not a final test of it, supporting its adoption.

Yes, we will better insist on the fact that we just propose a new approach and demonstrate its relevance to encourage further studies on it. We should just insist that it also demonstrates some weaknesses of the tracer release approach (and the underestimation of its uncertainties when deriving it from the STD of its estimate transect by transect).

Figure 2: different colours (or line type) for the curves for different stabilities may better highlight the results.

We will the color lines in order to better differentiate them.