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Ars et al. introduce a method for inverting small-scale emissions using a statistical framework which incorporates a Gaussian plume model and observations from the tracer method (here with acetylene). This is motivated because in real-world environments the exact location of the methane source could be spread out, inaccessible, or not precisely known, limiting the accuracy of the single tracer release technique by itself. Validation of the combined approach is attempted using a controlled methane and acetylene release experiment in four different configurations.

I read this paper with interest since I am also doing work related to quantifying methane emissions at small scale. From the measurement side, the experiment appears to be well conceived and performed. However, I noticed multiple things that could be clarified or improved on the analysis side. I think there are some key revisions needed to helpful clarify the method and results, along with additional suggestions to improve the overall manuscript that are given in this comment.

We thank M. Goldston for his interest in our paper and his constructive and technical comments. They will help us improve the manuscript, and in particular the result section. 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.

Major comments

1. Physical basis: On the basis for the approach, it makes sense a the tracer releases could, for instance, constrain the dispersion in the Gaussian model when the tracer source is collocated with the methane source. However, when the tracer is positioned farther away, for all the reasons outlined in the paper (lack of a homogeneous / stationary atmosphere), the information from the tracer should become decreasingly useful as one has to rely more heavily on the model results. This is both because the diffusion of air along the acetylene path (obstructions, elevation changes, etc.) may not be the same, but also because a Gaussian model is used to bias correct for differences in downwind distance between methane and tracer.

The conclusion will better discuss these points. Overall, our aim is to introduce a new concept (calibrating a statistical inverse modeling framework based on tracer release information for the monitoring of emissions from industrial sites) and to illustrate its potential with a simple implementation using a Gaussian model and a practical test for which the conditions are relatively favorable to such a use of a Gaussian model, with relatively stable and homogeneous wind conditions, and in a field with a relatively flat topography and few obstacles (building) that could impact the emission plumes. Considering the scale of this study (with a maximum distance between methane and tracer sources of 60 m), it seemed relevant to make the assumption that the tracer is a good indicator of the atmospheric stability for both the tracer and methane transport. The results confirmed the good behavior of the Gaussian model calibrating based on the tracer data.

The introduction and conclusion will better support future analysis and improvement of such a concept, in particular by using more complex local scale transport models to generalize its applicability (see also our general answers to the anonymous reviewers on these general topics).

For more complex experimental cases, the Gaussian model would definitely miss the impact of the topography and of variations in space and time of the local meteorology which could make the transport from the tracer source to the measurement location significantly different from the transport from the methane sources to the measurement locations (of note is that we could still run such a model with different wind forcing for different point sources if having relevant measurements, and recombine the full signal for the set of sources thanks to the linearity of the methane atmospheric transport at such a spatial scale). However, in principle, the tracer could be used to control parameters of models that better account for the heterogeneity in space and time of the local meteorology and of the topography during the experiments than the Gaussian model (like CFD/LES ones).

It is difficult (and definitely out of the scope of this first paper on our concept) to assess at which point (e.g. distance between the tracer emission point and the different targeted gas emissions points of the industrial site as a function of the meteorological heterogeneity and of the topography / buildings) it would become difficult to use the tracer to control such models but this issue could be partly handled by emitting tracer at multiple points surrounding the industrial site.

Yet, the test which performed the worst (in terms of relative difference) for the combined approach was actually Configuration 1, with a collocated tracer, while it performed better for all of the three conditions which should have increased the uncertainty of incorporating the tracer information.

We can explain it due to the impact of uncertainties associated with the difference in time between the tracer and methane measurements and with the variations of the acetylene and methane background concentrations which have been estimated by calculating the standard deviations of each series without the plume crossings (for example 0.3 and 9 ppb for acetylene and methane respectively for configuration 1). These two sources of errors that we underestimated in the present version of the manuscript and that highly impacted the computations for the first configuration will now be better accounted for and discussed. In particular, the results will be updated due to using a slightly different method for integrating the methane and acetylene plumes and defining the background concentration above which the plumes are computed. It will decrease the impact of these sources of errors in the first configuration

It makes sense that the uncertainty would be higher for the combined approach than the tracer for Configuration 1, and that the tracer generally was worse for the non-collocated experiments. However, the result for the combined approach is unexpected, both from the perspective of the theoretical basis for the combined approach, and the interpretation of the actual measurement results.

See our answer above. And, more generally, even though the emission conditions where more favorable in configuration 1 than for the other configurations, the transport and measurement conditions (in addition to the background conditions as point out above) could be potentially more challenging. We can only compare the tracer method and statistical inversion for each configuration rather than results from the different configurations in such a context. It will be better analyzed in the result section and discussed in the conclusion section.

Does the combined approach account for uncertainty of the tracer technique when used under ideal (collocated) vs. non-ideal conditions?

Uncertainties for the tracer technique (in our specific case) come from:
1) the problem of colocation between the tracer and the targeted gas
2) measurement errors
3) differences in time between the acetylene and methane measurements
4) variations in the background variations of tracer and targeted gas

Misfits between the estimations and the known emission rate can also be driven by uncertainties in this known emission rate but we did some verifications to ensure that this uncertainty is negligible compared to the misfits obtained in this study.

Uncertainties for the statistical inversion come from: 2), 3), 4) and
5) transport model errors
6) uncertainties in the estimate of the statistics of the errors in the prior estimate of the emissions and of the measurement and model errors.

1) is negligible for configuration 1. In other configurations, strictly speaking, it should impact the tracer technique only (but, actually, it impacts 5) and thus indirectly the statistical inversion; see our answers to the previous comments above). 2), 3) and 4) impact both the tracer technique and the statistical inversion. The estimate of the statistics of measurement and model errors in the statistical inversion system (based on model-data tracer comparisons) should account for 2) and 4) (in addition to 5)). However, in case of 4), it is estimated for the tracer only while the CH_4 background variations may be larger than that of C_2H_2. At last, 3) is ignored in the estimation of the measurement and model errors in the statistical inversions.

We will clarify these points in the new version of the manuscript.

Why where the combined results better (again, in terms of relative and absolute difference) when the tracer is used under less ideal conditions? More physical insight into how the combined approach works would be very helpful.

See our answers above. The new results will show a different picture since better addressing the sources of errors associated with the background concentrations and alternative measurement of C_2H_2 and CH_4.

2. Organization: In general, there is unusually frequent referencing back and forth between Sections 2 and 3 and at one point even a "circular link" between section 2.5 and section 3.2 about the time lag, with neither quite containing the indicated information. Section 2 largely reviews the literature on these three techniques separate from the details of this paper, and could easily be condensed or even combined with section 3 since there are a lot of similarities between the two, and I think it would make it easier for the reader to understand specific aspects of the way the approach and experiment were conducted which is currently tedious going back and forth between the different sections and subsections.

We will improve the concision of the text especially in section 2, avoid redundancies and gather sections 3 and 4 into a single one, within which we should merge some of the subsections that were previously split between section 3 and 4. Section 2.5 will now be more concise and will not enter into its present level of details while the corresponding topic will be better addressed in the new section 3.

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). Many discussions and details will be aggregated into the last section which will now be a discussion / conclusion section rather than a short conclusion section.

3. Relationship to other literature: The effect of non-collocation, including distance of the measurement and magnitude of non-collocation, and the effect of being confined to the road which prevents non-orthogonal slices were discussed. These are all important issues for subsequent people using this method or similar experiments, and is also related to one of the conclusions [L676 - L679],

so several recent papers would also be valuable to cite on these topics:

Goetz et al. 2015, Environ. Sci. Technol. (doi:10.1021/acs.est.5b00452) investigate issue where tracer not collocated and employs a correction based on the Gaussian plume

Roscioli et al. 2015, Atmos. Meas. Tech. (doi:10.5194/amt-8-2017-2015) also look at effect of the tracer and source not being collocated using the dual tracer framework to bracket possible errors, which is an alternative approach to what is given here and is likely applicable to similar types of sites

Albertson et al. 2016, Environ. Sci. Technol. (doi:10.1021/acs.est.5b05059) employs Bayesian framework and also specifically discusses and gives a correction for the issue of the road not being orthogonal to the wind direction also based on a Gaussian plume formulation

We will cite these relevant papers when discussing the imperfect collocation of the tracer and methane sources and its effect on the flux estimations with the tracer release method in the section 2.2 and 2.5.

4. Details missing that are important for understanding the approach/experiment:

- It is vague what information from the tracer is combined with the Gaussian, is it just the (rather coarse) adjustment of the stability class A-F, or more fine scale impact on the parameters (σy, σz, and/or wind)? It would be helpful to see how the parameters were actually affected during these experiments

The wind forcing is imposed based on our local wind measurements. The tracer observations are used for the selection of the model stability class. It appeared that the optimal stability classes in terms of fit between the modeled and measured tracer were systematically in agreement with the range of stability class corresponding to the measured wind speed (according to the Pasquill stability classification, see 2.3). The horizontal and vertical Gaussian plume standard deviations σy and σz are then set-up according to the class of stability. Even if it seems coarse, the model showed a good ability to reproduce the measured tracer concentrations with the selected stability classes while being highly sensitive to the choice of these stability classes.

We will improve the clarity of the text on this topic.

In the future, we expect to use tracer data to control for more complex parameters in potentially more complex models (directly controlling the σy and σz of the Gaussian plume spread might be a good option). However, again, in this first paper, we wanted to demonstrate the potential of our approach with a rather simple implementation case before increasing the level of complexity.

- How the prior uncertainty is determined is not discussed, and the basis for model + observation uncertainty only briefly

The configuration of the prior uncertainty is set high to reflect a lack of prior knowledge on the source as when investigating actual industrial sites. We will better describe and justify it. The presentation of the prior values also needed improvement and clarification. More critically, we will better explain in the conclusion section that the weight of the prior information is relatively low in our statistical inversions (thanks to the choice of high prior uncertainties).

The model and measurement uncertainties are directly derived from the model-data tracer comparison. It is a critical part of our method that was actually discussed in several paragraphs so we will better explain and highlight it.

- Both the method and results for the "multiple sources" inversion is brief other than that the plume is divided into five slices. Can a figure be added to illustrate how this works? Does using five slices mean up to five sources can be quantified? Can the approach resolve multiple sources when the plumes are overlapping, or only when they are basically non-overlapping?

An example of the division of the emission plume and of the integration of the area under each slice was given in the figure 6 (bottom figures).

Since we use a statistical inversion, we do not have to follow a strict relationship between the number of sources than can be solved for and the number of slices (we can solve for more or less sources than the number of observations i.e. slices assimilated by the inversion system). Still, if willing to bring a strong constraint on individual sources, one must have a sufficient number of slices bringing independent piece of information. Having a large number of slices ensures having maximized the potential for catching the different piece of independent information in the signal. But it can give a critical weight for measurement errors, turbulent patterns or background variations, which would dominate in some of the observations. Selecting 5 slices here was considered as a safe trade-off between these two aspects.

If the plumes of all sources strongly overlap, there would hardly be any source of information for individual sources in the signal, and these sources would be difficult to quantify separately. Increasing the number of slices would not help solve for this problem. The level at which the slices can provide independent information for individual sources is difficult to anticipate but it is characterized in the posterior uncertainty that is diagnosed by the inversion system. The more the plumes overlap and the highest the posterior uncertainties on these estimates will be, and the more negative the posterior correlations between these posterior uncertainties in individual sources will be. The analysis of these posterior uncertainties will better highlight it and this general topic will be better discussed in the manuscript.

In a more general way, the analysis of the results will be expanded following the different comments by the four reviewers.

5. Table 2: Two things stand out about Table 2, where results are given from the controlled release experiment.

First, why is there no row given for a Gaussian inversion, separate from combined approach? This is key information in evaluating the difference between the tracer release, Gaussian, and combined approaches.

We could have considered deterministic inversions using the Gaussian model. However, we are not sure that it brings a lot of insights: if using the Gaussian model with a configuration ignoring the tracer results, one would get relatively poor estimates of the emissions. If using a configuration minimizing the misfits between modeled and measured tracer, one would already have used one of the components of our proposed concept (there would already be some combination of approaches). And, when targeting two methane sources, one would have to separate the measured signal into two slices to ensure that the transport is invertible which would be both inspired but different from the choice made in the statistical inversion.

In a general way, we do not think that deterministic inversions based on model handle all of the issues connected to the tracer release technique that are discussed in section 2, so we prefer to compare the tracer release technique to the statistical inversion only. Furthermore we do not really see how analyzing the behavior of such a technique would bring insight on that of the statistical inversion (which would use a different control or observation vector). Therefore we prefer not applying and discussing results from a deterministic inverse modeling framework. We will briefly mention it in the conclusion.

Secondly, the uncertainty given for the combined approach is extremely small, several times smaller even than the controlled release uncertainty for Configurations 1, 2, and 3 which does not make sense.

We will better:

- remind that it is difficult to compare the uncertainty estimates for the tracer release technique and that of the statistical inversion (since they rely on different assumptions even when they are assumed to encompass similar sources of errors)
- discuss the fact that the posterior uncertainties from the statistical inversion are likely under-estimated; our revision of the results (see above) will not change this general fact. Among other explanations, it can be due to an under-estimation of the model and measurement errors due to ignoring spatio-temporal correlations of this error for individual slices, due to ignoring the difference in time between tracer and methane measurements, or due to underestimating the impact of variations in the background concentrations when looking at tracer concentrations (while the variations can be larger for methane). More generally, the translation of model errors when simulating the tracer plume into estimates of the statistics of model error when simulating the methane plume(s) relies on assumptions that may be too optimistic in our experimental case.

This is also noticeable in that for Configuration 2, the statistical chance of the uncertainty ranges 428 ± 7 and 464 ± 1 overlapping is 1-in-a-million, and for Configuration 1 the change of 382 ± 7 and 472 ± 2 overlapping essentially impossible. It is said all of the sources of error are considered, but this is clearly not the case - more thought should be given into how to derive a more representative uncertainty range for the combined approach.

See our answer above. All of this will be better analyzed. A very specific source of errors was actually ignored (the difference in time between tracer and methane measurements). Furthermore, even if a source of error is taken into account theoretically, in practice, it can be very difficult to quantify its impact.

6. Clarification on use of ‘transient’: The time dependence and transience of the problem is mentioned several times, but I do not think a clear explanation is given.

When using this term, we were just referring to the fact emissions for the industrial sites can vary in time over annual / seasonal timescales to daily (and even hourly and smaller) timescales. By conducting relatively short term campaigns (typically on specific days and even over time windows of less than few hours) one may only provide a picture of the emissions that cannot easily be extrapolated into an annual rate for the industrial site, while inventories usually report and focus on such annual rates. This will be clarified.

- In Figure 6 was the release not being run continuously? How was the time dependence of the prior derived? Additionally, the title says “Gaussian plume”, which is formulated for a continuous, averaged value, not a transient release. Clarification is needed on this issue.

Yes, the tracer and methane release are constant in time and if targeted an unknown source, we would use a tracer release that is constant in time. In cases for which the targeted source would vary in time during the measurement time window, this would challenge the tracer release technique. It would challenge our practical implementation of the statistical inversion but we could imagine a more complex statistical inverse modeling set-up (with models that can clearly account for the temporal variation of the sources) to account for potential variations in time of the targeted source. This will be better discussed in the last section.

Minor comments L487-491 and L556-559: Both good points L492-495: Sentence could be clarified

We will clarify it by merging 3.6 and 4.1 in the new section 3.

Table 2: including the bias due to mislocation as part of the " +/- " does not make sense since it is not a random error.

We agree with the reviewer. We will change our way to present these uncertainties as follows: ± 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 ; estimate of the total uncertainty based on the RSS of these two terms)

Also what about the uncertainty of the bias, since presumably this is non-trivial?

All estimates of uncertainties and biases are uncertain (see above). However, our protocol to derive estimates of uncertainty and biases are deterministic and detailed explicitly. The numbers given correspond to these protocols and from that point of view they do not require any uncertainty bars.

Table 1: one of the columns says 'wind direction (degree)', when that column is given in letters rather than degrees

We will remove (degree) from the table.