

Anonymous Referee #2

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In Ars et al., the authors describe a new method for estimating gas emission rates from industrial facilities, by combining 1) tracer flux measurements, 2) Gaussian dispersion modelling and 3) a statistical inversion algorithm. The new method is evaluated using controlled methane/acetylene releases and compared to results from tracer flux. Four tracer placement scenarios are evaluated to demonstrate the improved accuracy of the method in situations where the tracer and the emission source are not perfectly collocated.

We thank the reviewer for his constructive, detailed and technical comments that will help improve our manuscript.

GENERAL COMMENTS

Overall, I was intrigued by the method presented by the authors, since accurately measuring industrial methane emissions remains a challenge, especially as we attempt to find convergence between bottom-up and top-down measurements.

We agree that such an estimation remains a challenge. Our objective is to propose a new approach to support it but we do not claim that it has already reached such a maturity after a first study that it should be considered as fully robust. We rather demonstrate its potential and that it can overcome some of the drawbacks of the tracer release technique, which is widely used nowadays for such an activity. Further studies and tests will be needed to improve it and allow for its generalization, including tests with more complex models than Gaussian ones. The introduction and conclusion will better emphasize it.

However, I am a little skeptical of value of the method in its current form and would like to see more discussion of the method's wider applicability before this paper is published in AMT.

See above. It is difficult at such a stage to provide an objective evaluation about its level of applicability and our main objective is not to present a specific method with a "rigid" configuration but rather to present a general concept and its potential through a rather simple practical configuration. Section 2 already indicated that a wide range of options could be taken to define the observation and control vectors. The conclusion will now better indicate that the number of options for the transport model is large. This will also open many options for the control of the model to fit the tracer data if using models more complex than Gaussian models.

Some areas of the method that I think warrant more discussion include the effect of different methane emission rates (only one was tested, 0.4 kg/h)

Multiplying the number of tests would have provided a more robust evaluation of the very specific configuration of the inversion used in this study. However, again, given our main objective to expose a theoretical concept and demonstrate its potential, and given the difficulty, in practice, to multiply such tests, we find that the number of tests selected was enough. This will be clarified.

Regarding the rate of emissions: see our answers to the reviewer's specific questions about this topic. By using a rather low emission rate, we have challenged the ability to separate the tracer and targeted plumes from the measurement noise and from the acetylene and methane background variations. This impacted both the tracer release technique and the statistical inversions. Due to the linearity of the transport over such distances, results should be better but comparisons between the methods should be similar if using

larger emission rates.

and the role of meteorology – I am particularly concerned that the authors selectively looked at plumes from very specific atmospheric conditions.

Yes, the proposed method could have weaknesses in less favorable conditions. The conclusion will better reflect it. The tracer release technique also requires some favorable meteorological conditions, especially a relatively constant wind direction and sufficient wind speed in order to be able to measure an increase of the concentrations through a clear “emission plume”. Still, the tracer release technique does not require as homogeneous and stationary local meteorological conditions as our practical implementation of our inversion concept in this study that is based on a Gaussian model. Therefore the result section will now discuss the tracer release method results when using all the transects that can be retained for such a method even though they cannot be retained for the statistical inversion, and show that, in our practical case, these “new” transects do not improve the results from the tracer release technique.

The conclusion will better address this general topic. Of note is that this conclusion section will be expanded into a sort of discussion / conclusion section to better address such points, others asked by the reviewers and to gather some of the discussions that lengthened the previous sections.

If this new method is being proposed as an “easy-to-implement” method for operators to employ (as it is described in the Introduction), then I would expect such a method to be robust to different atmospheric stabilities. The quality of the writing is excellent, but the authors would do well to streamline the paper so it is less bogged down in text.

We will improve the introduction and conclusion to ensure that we do not go beyond the objectives and conclusions indicated in our answers above, and that we better present them. We agree that if the method had to stay as “easy-to-implement” as in our practical test with a Gaussian model, it would definitely be challenging to generalize it in terms of the topography, complexity of the facilities and local meteorological conditions of the sites to be studied. More studies and potentially more complexity (using models that are more complex than Gaussian models) will be needed for such a generalization and, if the concept can really lead to operational systems, to find trade-off between accuracy and complexity for being used by operators. However, with this first study we are definitely far from claiming to address such topics.

I tend to agree with Reviewer #1 who described the writing as “verbose”. This complex writing style makes it more challenging to follow the science. Additionally, I think the authors could limit some of the discussion of the methods, particularly the tracer release and Gaussian methods, as these are well-described in the literature, to make room for a more well-rounded discussion of the results, which seemed rushed.

We will improve the concision of the text especially in sections 2 and gather sections 3 and 4 into a single one and better discuss the results in the light of the four reviews we received for this manuscript.

Upon making these major revisions, I expect the publication will be suitable for publication in AMT.

We thank the reviewer for this assessment.

Specific comments follow.

SPECIFIC COMMENTS

Section 1 – L105: Is this method easy to implement for operators?

See our general answer above. This sentence of the introduction was not really aiming at characterizing the specific method presented in this paper but rather at giving a context and long-term objective for the development of its concept and of its practical implementation(s). The method used for our test in this first paper is relatively easy to implement and could definitely be handled by external consultancy groups or by the development department of the operating companies. However, since we acknowledge that first developments may rather go towards complexity than simplification, and since such a consideration is far from the stage of development of our concept, this part of the text will be rewritten.

How have operators historically monitored their emissions?

To our knowledge, most of the landfill, waste water treatment plant or gas operators who derived their own methane emission estimates used standard bottom-up product of emission factors times quantity of waste/wastewater/gas processed and/or emission models. In some cases, some operators ordered measurement campaigns by internal (within the R&D department for large operators) or external laboratories with close chambers, tracer release techniques or some completely different measurement concepts (like the recent flux measurement technique based on LIDAR promoted by NPL, Robinson et al. (2011)) to valid or complement such estimates. However, to our knowledge, the use of real measurements is still rather limited.

If the paper is framed as being in support of industry, then this should be discussed; I am not familiar with many facilities actively conducting tracer flux measurements or those with mobile laboratories to measure downwind emissions.

See our answer to the previous questions. Industries are definitely interested in such a monitoring of their methane emissions and, we should even mention that, since this study, in the context of projects in collaboration with industrial partners, we have led campaigns and tested our method for their industrial sites (it will be documented in future studies). However, the support to the industry is a bit out of the specific focus of this first paper even though it definitely feeds its context and the long-term goal of the development of our monitoring concept. This first paper is exploratory and conceptual and it is not orientated towards the development of an operational approach. Finally, even though the main field of application should be connected to industrial sites, in principle our concept could be applied to natural “point sources”. Therefore, we prefer avoiding spending much time on this topic and, again, the corresponding sentence will be modified.

Section 2.1 – L175: I am not sure how you have demonstrated that your method provides satisfying results over those distances and methane emission rates compared to what you have tested with your controlled releases – please elaborate.

We will remove this sentence and add in the conclusion that the method needs to be tested for all of these parameters (distance, spread of the site...) on real sites.

I am especially interested in how tracer flux and this method differ for large methane emission rates or “superemitters”.

Due to the linearity of the transport, larger sources are easier to invert (i.e. the signal to noise ratio for the concentration measurements is larger) and if the methods succeed in monitoring a given emission rate at a given distance, it should succeed in monitoring a larger emission rate at the same distance (keeping all other emission, topography and meteorological conditions similar). Issues arise for small emission rates, when signals are not high enough to ensure a clear separation between the targeted plume from the noise

of the instrument or from the background variations of the concentrations. This will be discussed in the conclusion.

Section 2.2 – L180: I suggest that the authors conduct a more thorough literature review, particularly of tracer release measurements conducted in various shale gas basins in the United States. Numerous papers have come out on this subject in the past 3 years.

We will refer in the section 2 to the studies of Roscioli et al. (2015), Goetz et al. (2015) and Albertson et al. (2016) who used the tracer release method to estimate methane emissions from different types of sites and sought solutions to overcome the non-colocation of the tracer issue.

Section 2.2 – L210-215: Can the authors speak to how this effect scales with methane emission rate? Does its significance shrink if total methane emissions increase? Or does its importance scale linearly?

In principle, due to the linearity of transport, it should not impact this specific effect. Larger source just increase the signal to measurement noise (+ variations of the background concentrations) ratio and thus the precision of the results due to the lower impact of measurement noise and uncertainties in the background concentration variations. This will be discussed in the conclusion.

Section 2.3 L230-235: Can you expand on this more in the text? I find the model justification to be a little lacking.

We will better insist on the fact that the Gaussian model is a first, simple and low-computational option to test the method feasibility and its ability to improve the estimations when the tracer and the pollutant sources are not well collocated or when there are several sources of the targeted gas. Other options of models are available for more complex topographies but it was not relevant develop a complex inverse modeling scheme with such options for the first tests of our concept in this study.

Section 2.3 L250-253: Is this detail on urban vs. rural configurations really necessary? Especially if you don't mention what configuration was used in this study.

This part of the text gives a general presentation of the Gaussian model so we will keep on giving this information but we will try to give it less weight in the corresponding sentence. And we will indicate that we use the rural configuration in section 3.

Section 2.5 L305-310: I went looking for an explanation of how the spatial offset was treated in Section 3.2, but this section reference Section 2.5. Please make sure this concept is explained. I would strongly caution against routinely referencing other sections, particularly future sections, and instead focus on a linear narrative for the paper.

Section 2.5 will now be more concise and will not enter into this level of details while this topic will be better addressed in the new section 3.

Section 3.1 L342-345: If the authors are going to be highly selective of meteorological conditions, then this should be discussed in more detail. What happens on days with low winds?

When the wind speed is not strong enough, the signal coming from the site is very low and difficult to catch on the roads where the measurements are carried out. Similar wind conditions are requested for the tracer release method. See our answer to the reviewer's general comment on this topic.

Section 3.2 L360-364: Here are some more cyclical references – I don't think the spatial offset is

ever properly described.

We will gather as much as possible the details and improve the clarity of the text on this topic in the new section 3.

Section 3.5 L430-435: Choosing stability class based on best fit to the measurements seems suspect to me. How does this choice compare to the estimated stability class using wind speed and insolation metrics? Furthermore, what were the range of atmospheric stabilities during all your tests? Is this method applicable to all stability classes? This is a main point of concern for me and the authors should better justify their decision regarding the Briggs parameterization.

There could be inconsistencies between the selected stability class and the local meteorological conditions due to the empirical formulation and parameterization of the Gaussian model.

However, for each transect, we have checked that the stability class yielding the best fit between modeled and measured acetylene was systematically in agreement with the measured wind speed according to the Pasquill classification. The solar radiation information may be difficult to have, especially at this scale, and it is thus difficult to choose between the different (in practice for any transect: two) stability classes that were in agreement with the wind speed without the tracer data. We will clarify this and indicate in the text, for each series of measurement, the measured wind speeds and the two stability classes that we could have used according to Pasquill classification and to this measured wind speed, along with the selected class according the model-data tracer comparison. Further tests should be done with stronger wind (> 5 m/s) to make sure the method is applicable to all stability classes as will be discussed in the conclusion.

Finally, even though it did not occur in our experiment, we still think that selecting a stability class that is not consistent with the actual meteorological conditions is not a critical issue as long as the fit (in terms of area and shape) between the modeled and measured tracer plumes is good, since this is definitely a good indicator of the model performance as a function of its parameterization. This will also be discussed in the conclusion section.

Section 3.5 L472-479: I could not follow this; can you explain how these uncertainties translate into those methane emission rates?

Some typos made these sentences difficult to read and we apologize for it. The text will be clarified by better indicating the actual values for the sources, the prior knowledge of the inversion on these sources, and the uncertainty in this prior knowledge both in terms of relative uncertainty (compared to the prior estimate) and in terms of absolute value.

Section 4.2 L584-590: I am not convinced by the argument that the performance of the new model was the worst compared to the actual emission rates due to the low emission source – it seems to me all the other configurations used comparably low emission rates and this problem was not observed. Please provide a better explanation.

This part of the text discussed the results from the tracer release technique. The errors from this method will not be smaller for the other cases, on the opposite. The measurement errors, the difference of time between the tracer and methane measurements (since the instrument continuously shift between the two types of measurements) and the variations of the acetylene and methane background concentrations are the main source of error for this technique when the methane and tracer sources are perfectly collocated as here (the other source of misfit between the results and expected truth being the uncertainty in the debit of the controled emission which is small). We will better explain that the difference between the estimate and the real flux comes from these factors. In particular, the impact of the difference of time between the

tracer and methane measurements and the variations of the acetylene and methane background concentrations were quite forgotten in the present version of the manuscript while they actually have a critical weight in the results. We will update our computations and results to better account for them, in particular for the configuration 1 where they definitely appeared to have a larger impact, and we will better discuss these sources of errors in the new version of the manuscript.

Section 4.2 L584-613: This is repetitive of table 2 and does not to be listed off in the text.

Section 4.3 L628-652: Again, this is repetitive, I would prefer to see more of an analysis vs. repeating of figures in a Table.

We will not provide all these numbers in the text and make it more concise. On the other hand, we will expand our analysis of the results as indicated by our answers to the specific comments of the four reviewers.

Section 4.3 L432: The authors explain the poor performance in configuration 1 does not matter very much due to the fact that the configuration is unrealistic. I am not satisfied with this explanation, if theory dictates that unreasonable or not that configuration 1 should be the ideal case then a good reason should be provided why it was not.

We will remove this part of the paragraph to avoid misinterpretation and we will rewrite this paragraph to better explain that the tracer release method is supposed to give a better estimate of the emissions than the combined approach in the configuration 1 because in this case the tracer is a better proxy of the transport than the model.

Section 5: Nowhere in the conclusions (or in the results) do I see any statements on the performance of this method vs tracer flux for a range of methane emissions. This was introduced in the introduction and I do not think it was adequately followed through on. If this method is currently limited to low industrial emission rates it should be expressly stated. As it stands, I think the usefulness of the method is overstated and the authors should be realistic about what their experiments have demonstrated.

As discussed above, if the method is successful for low emission rates, it should be successful for high ones provided that the size of the source stays the same. We will discuss this in the conclusion. Furthermore, as expressed earlier in our answers to the general question by the reviewer, we do not aim at demonstrating that we have an operational system for monitoring industrial sources yet, and we will better clarify our objective of introducing a new concept and demonstrating its potential through a rather simple practical case (expecting that it could definitely lead to a robust and generalized approach on the long-term).

Table 1: If I understand correctly, on the days where meteorological conditions were explicitly controlled for, the plume capture rate is roughly 30%. This seems very low to me making me question the robustness of the method. Please comment.

In order to compare the results of the two methods, we used in this study only transects suitable for both of techniques even if some of them could have been used only for the tracer release method and others only for the combined approach (strictly speaking, there is no “control of the meteorological conditions” in these cases). That explains the low numbers of transect selected. However, we will now discuss the results from the tracer release technique when selecting all suitable transects for this method even if they cannot be used for the statistical inversion, demonstrating that this does not improve the results when using this technique.

Again, in the future, the concept of the statistical inversion should be improved (in particular by testing it with more complex models) and one of the objectives could definitely be to be able to exploit more measurement transects than in this first practical application.

TECHNICAL CORRECTIONS

Section 2.3, L219: Please define acronym “LES” and possibly “CFD”, as I am unsure if everyone would know what these are.

Section 3.1 L340: Typographical error “serie” Section 3.6: Edit section title to be more succinct.

These corrections will be done.