

Dear authors,

Thank you very much for the revisions. The reviewers had suggested only minor changes and I consider your responses and amendments as adequate.

However, before being able to accept your publication, I have quite a large number of additional suggestions/corrections, most of them addressing issues with grammar and language, but also a few comments/questions on the content. I don't expect you to respond to all these changes point by point, but nevertheless, please try to account for them as much as possible. I would also be happy if you could briefly respond to the most critical comments I had.

Please also note that your paper must finish with a conclusions section. Thus, please split the current discussion section into a discussion and a conclusion.

I think you should be able to incorporate my suggestions (see attached annotated PDF file) rather quickly.

Yours sincerely

Dominik Brunner

We thank the editor for his detailed review of our text and for his comments that helped improve the quality of the text and of the discussions.

L17-19 It would be good if you could expand the sentence a bit explaining why the classical tracer release method is inferior, e.g. with "... than the tracer release method which does not account for xyz".

We prefer to add some sentences earlier (see above) that describe the principle and aims of the new concept, including the aim at overcoming the problems of the classic tracer release technique when the sources are not well collocated.

L24-25 "more complex implementations", not quite clear what you mean.

The sentence has been changed into "Further studies and more complex implementations with more advanced transport models and more advanced optimizations of their configuration will be required to generalize the applicability of the approach and strengthen its robustness."

L36 "Many emitting industrial sites have a typical size of 100-500 m²", this would be 10 m x 10 m or 20 m x 25 m. This is way too small.

We agree with the fact that industrial sites can larger than 500 m², the upper value has been changed into 1km².

L47-50 "mobile measurements can be conducted", tracers (like SF6) have also be released over much longer times (days to weeks) but then in combination with stationary measurements.

We prefer to add "generally" and change "and" into "or" in the sentence and avoid referring to experiments with measurements at fixed sites to avoid complex discussions at this stage and keep the logical flow with the next sentences.

L86-88 “emission spatial distribution”, Depends on how the inversion is setup. Probably “ prior emissions” would be more correct.

We feel that applying the two suggested corrections would lead to a wrong assessment of the typical situations that are discussed in this study. We prefer to rephrase the sentence into “However, the skill of such approaches strongly relies on a good accuracy of the transport modelling and on the ability to characterise the statistics of the modelling uncertainties. It can also strongly rely on the prior knowledge on the emissions, in particular on the spatial distribution of the multiple sources within an industrial site for the type of applications considered in this study, and on the ability to characterise the uncertainties in such a knowledge.”

L104-107 “was no strong incentives to report emissions”, I disagree. The Kyoto Protocol and probably national legislations provided a strong incentive to report emissions (look at the E-PRTR database for point sources in Europe, for example), but there was not strong incentive to actually measure the emissions from this type of sources.

The sentence has been changed: “Until recently, there were no strong incentives to estimate site emissions using dedicated measurements. The reported estimates were usually derived using...”

L156-159 If there is no street within the boundaries of the two criteria mentioned above, then you cannot apply the method, or only by compromising on quality. “Adapt” is a bit misleading here.

The sentence has been changed into: “Finally, the choice of the distance is constrained by the need for conducting measurements on roads located downwind of the site sources (depending on the specific wind directions during the measurement campaigns) when using instruments onboard cars as in this study.”

L160-162 Quite complicated and unclear sentence. How can a “combination” be “linear”? Probably you are trying to say that the relation between the concentrations and the emission rates is linear and expressed by the observation operator H?

The sentence has been changed into: “The simulated relation between the gas emission rates from the single or multiple sources of the site and the atmospheric concentrations relies on the knowledge of the location and spread of each source and on the proxy of the atmospheric transport. It is linear and expressed by the observation operator H.”

L243-247 I would think that the most commonly used models for this are Lagrangian dispersion models driven by flow and turbulence fields either diagnosed or explicitly computed (with a CFD type solver). Spray, AUSTAL2000 or GRAL are a few examples for such models that are used by many engineering companies in Europe and also certified for use in legislative applications.

The sentence has been changed into: “Many types of transport models are used to simulate the dispersion of pollutants at the local scale, i.e. typically over distances from a few metres to 1 or 2 kilometres, from simple Gaussian models to the coupling of Lagrangian and sophisticated CFD

(Computational Fluid Dynamics) models that allow to determine turbulent patterns for complex terrain through an explicit representation of reliefs and obstacles.”

L 322-324 I did not understand this last sentence. I have the feeling that it is merely repeating the statement of the previous sentence and that it can therefore be dropped. I agree with reviewer #1 that your paper is very wordy with unnecessary long sentences stating facts that are already clear from the context. It has much improved, and I hope my suggestions will further help making the paper more concise.

We have deleted this sentence.

L350-353 Too complicated and too long. I can only guess what you are trying to say here.

We have removed this sentence, the underlying idea was too complicated and partly redundant with the previous sentence.

L459-466 Is this last paragraph really needed? I did not fully understand it.

This paragraph has been deleted.

L491-492 30 seconds is not at all "short" but is in fact extremely long for such a mobile system. Probably you used a too large tubing for the small sample flow.

We agree to this comment and replace the sentence by: “The sampled air was sent into the instrument by an external pump system allowing an inlet lag between the sample inlet and the measurements of less than 30 seconds. This more or less constant inlet lag introduced a spatial offset when comparing the measured and modelled tracer or methane concentrations.”

L514-516 Systematically good does not mean anything. If you have really compared, you should be able to give a number for the differences.

The relative difference between flow rate calculated with the mass difference and the flow rate read on the flowmeter was between 1 and 3 % depending depending on the series. These numbers have been added to the manuscript.

L602-604 I found this approach of selecting a specific class a bit weird. Some sort of direct estimation of the dispersion parameters σ_y and σ_z (or of the parameters α , β γ) would have seemed much more logical to me. By selecting a class, you can never fit the measured indices exactly (or only by chance), and will thus always have to deal with a biased simulation. These biases may cancel out with sufficient transects, but I wouldn't take this for granted.

We have added a discussion on this topic in section 4. Such a direct optimization of these parameters could make sense but should be carefully handled to avoid obtaining inconsistent (inconsistent between them and strongly inconsistent with the meteorological conditions) values for these different parameters to fit mathematically the tracer plume indices. By following the Briggs formulation, we ensure a minimum of physical consistency of our model parameterization.

L729-732 If I understand correctly, you are defining a different observation uncertainty for each transect. This makes sense to me, but then I don't understand the following statement in the discussions section (line 880):"unlike the estimate of uncertainties for the tracer release technique, the statistical inversion ignores the variations of the methane observation values and methane model data misfits from one transect to the other one"

Here we speak about defining R based on tracer data, while the sentence from section 4 discusses methane data: it states that the computation of the posterior uncertainties strongly relies on the configuration of R, B and H but not on the values of the methane model – data misfits. We still update the sentence in section 4 since the factor between the measured tracer and methane plume indices enters into the computation of R.

This discussion section is very wordy and not at all to the point. In this way, the main points of the paper are in danger of getting overlooked. Please note that your paper must finish with a conclusions section. Thus, please split this section into a discussion and a conclusion.

We have created a conclusion section.

L806-808 This statement is incompatible with the large biases produced by the tracer release technique in configurations 2 and 4, and actually it is incompatible with the conclusions of the whole paper.

This statement should be placed into a context in which the methane emissions from industrial sites are poorly known and so for which having estimates with 30% uncertainty should already be good. Our correction of the sentence should increase the consistency with the results.

L883-885 I have to admit that I find the low uncertainties very disturbing and, if true, a major deficiency of the method. The method should not only give a good estimate of the flux but also of its uncertainty. It is difficult to understand how you can get from an initial 80% uncertainty of the prior down to an uncertainty of often less than 1% in the posterior. I also don't think that it is a good idea to use the misfit between simulated and measured acetylene tracer index to define the transport uncertainty, because the transport model could (or I would even say should) be optimized in such a way that it more or less perfectly matches the measured index. In this case, the transport error would go to zero.

We agree about the fact that the model error, and thus the overall observation error, can be underestimated since comparing modeled and measured plume indices to derive it, while the model was optimized to fit these indices. By designing such a derivation of R, our assumption is that the main transport error arises from the inability of the selected class of stability to perfectly fit with the tracer plume index. The use of statistics of model - data misfits for tracer plume indices over slices (as in the fourth configuration) rather than over the whole plumes should better reflect the transport errors arising from turbulent, background patterns, and from the inhomogeneity of the wind field. We now discuss it here.