Authors’ answers to RC2’s comments

We thank the reviewer for his/her helpful review of our manuscript. We have carefully considered all the comments and revised the manuscript accordingly. We have noted that the references to specific lines of the manuscript and some remarks correspond to the preliminary version of the manuscript and do not take into account the updated changes to the manuscript after the preliminary review on the current version.

Below our answers to the commentaries raised by the reviewer.

General comments

1. Line106-107 showed that inversion errors from high precision measurements are 23-30% and 8-10m, and inversions from low-cost sensors can reach the same level in the abstract (25%, 9.5m), why is this? And the authors need to point these out in the abstract, which are associated with L336-337 and L410-412: “highlighting the higher impact of the model error on the inversion than the reconstruction error of CH4 mole fractions”. Without high precision instruments (e.g. the background information), can this be achieved? Add the role of high precision instruments in the abstract. Since performances of inversions are associated with wind conditions, the applications also have such limitations, which should be pointed out. Add limitations and conditions of this method and implications of this study in the abstract.

We have updated the abstract considering the comments raised by the reviewer:

“Fugitive methane (CH₄) emissions occur in the whole chain of oil and gas production, including from extraction, transportation, storage, and distribution. Such emissions are usually detected and quantified by conducting surveys as close as possible to the source location. However, these surveys are labour intensive, are costly, and fail to provide continuous emissions monitoring. The deployment of permanent sensor networks in the vicinity of industrial CH₄ emitting facilities would overcome the limitations of surveys by providing accurate emission estimates, thanks to continuous sampling of emission plumes. Yet high precision instruments are too costly to deploy in such networks. Low-cost sensors using a metal oxide semiconductor (MOS) are presented as a cheap alternative for such deployments due to their compact dimensions and to their sensitivity to CH₄. In this study, we demonstrate the ability of two types of MOS sensors (TGS 2611-C00 and TGS 2611-E00) manufactured by Figaro® to reconstruct a CH₄ signal, as measured by a high-precision reference gas analyser, during a 7-day controlled release campaign conducted by TotalEnergies in autumn 2019 near Pau, France. We propose a baseline voltage correction linked to atmospheric CH₄ background variations per instrument based on an iterative comparison of neighbouring observations, i.e. data points. Two CH₄ mole fraction reconstruction models were compared: multilayer perceptron (MLP) and 2nd degree polynomial. Emission estimates were then computed using an inversion approach based on the adjoint of a Gaussian dispersion model. Despite obtaining emission estimates comparable with those obtained using high precision instruments (average emission rate error of 25% and average location error of 9.5 m), the application of these emission estimates is limited to adequate environmental conditions. Emission estimates are also influenced by model errors in the inversion process.”
2. Line 140-141 reported that 2600 are useless, but there are reports that they are useful e.g. in Eugster et al., 2020 (AMT), and it needs more discussions on Rs/R0 ratio, which is sensitive to methane (10-100ppm) from 0.7-1.0 in the datasheet (see below figure), and also RL:

![Sensitivity Characteristics](image)

Indeed, the datasheet shows that TGS 2600 is sensitive to CH4, as other studies (Eugster et al., 2020, 2019; Riddick et al., 2022, 2020) have used it to derive CH4 mole fractions. Our motivation to exclude this sensor from the study was based on the observed response to CH4 enhancements of the controlled releases. Figure A1 shows a comparison of a typical signal measured by the three sensors for one controlled release, as well as the signal measured by the reference instrument. The TGS 2611-C00 and TGS 2611-E00 sensors show voltage drops that correlate with the signal measured by the reference instrument, whereas for TGS 2600, only low-frequency voltage variations are observed. This inability to observe such high-frequency variations of CH4 mole fraction prevents us from applying any reconstruction model and, consequently, from obtaining reliable emission estimates.
Figure A1. Comparison of the voltage measurements from three types of TGS included on chamber A. Upper plot shows the reference CH₄ observations measured from the reference instrument. Lower plot shows the voltage observations from TGS 2611-C00, 2600 and 2611-E00.

3. The writing and expression need substantial improvements. And many parts are very hard to follow. The manuscript needs to be polished by an experienced language editor, to thoroughly improve the fluency and remove grammar errors.

The writing of the manuscript was thoroughly reviewed. As well the methods section was reorganised to present the information in a more logical manner. In the current methods section is structured as follows:

- Section 2.1: We introduce the TADI 2019 campaign.
- Section 2.2: We present the controlled releases and the sampling configuration.
- Section 2.3: We describe the low cost logger system and the meteorological data.
- Section 2.4: We present the preprocessing steps of the TGS data.
- Section 2.5: We describe the reconstruction of CH₄ mole fraction from TGS voltage data and the metric used to evaluate the performance of reconstruction models.
- section 2.6: We explain the rationale in the selection of the training and testing sets.
- Section 2.7: We describe the inverse modelling framework used to estimate the release rates and locations.

4. Discuss why E00 is bad compared with C00, e.g. in Fig.4 and 7.

Our experiment has demonstrated that the TGS 2611-E00 sensor shows lower performance than the TGS 2611-C00 sensor in reconstructing CH₄ mole fractions. This difference in performance was also observed and documented in a previous study (Rivera Martínez et al., 2022), where sensors measured artificial CH₄ peaks under controlled conditions. In both experiments, we attribute the inferior performance of TGS 2611-E00 to the presence of a filter on top of the sensing material, designed to improve its selectivity to CH₄ but with an effect on the sensor's sensitivity, acting as a low-pass filter.

The TGS 2611-E00 signal presents a lower amplitude compared to the TGS 2611-C00 signal, and a noticeable decay after each peak. This decay produces phase effects in the signal, which are also observed in the reconstructed CH₄ mole fraction but absent in the TGS 2611-C00 signal. The lower performance is observed as a phase mismatch and a filtering of high-frequency features
present in the measured peaks. Our suspicions regarding this performance difference were included in the discussion section with the following phrase:

“The fast decay observed for reconstructed CH₄ mole fraction measurements after each voltage spike was attributed to the response time of the TGS sensor. The slow decay observed on Type E sensors was probably due to a filter integrated inside the sensor causing to improve CH₄ selectivity.”

5. **Add designs, and photos on low-cost sensor instrument.**

The logger system used in this study is the same as the one used in our previous studies (Rivera Martinez et al. 2021, 2022). We have added the following sentence in section 2.3:

“The logger system design was previously documented on Rivera Martinez et al. (2021) and Rivera Martinez et al. (2022).”

6. **I suggest the authors provide spatial distributions of simulations and inversions for typical cases, e.g. to show the real emission sources and the inversed sources and their distances.**

The following figure, corresponding to the cost function, was added into the supplementary material to show the spatial distribution of the controlled release and the estimated location using the inversion framework.

![Contour plot of the cost function for release #25 computed using assimilated gradients from TGS reconstructed data. The black and white stars show the location of the actual and estimated location respectively.](Figure A15)

7. **I recommend the authors to make the inversion code publicly available to improve the wide influences and applications of this study.**

We have added the following text to the manuscript:

*The codes developed in the frame of the Chaire Industrielle TRACE ANR-17-CHIN-0004-01. They are accessible upon request to the corresponding author.*
Specific comments

1. Add regression coefficients (slope, intercept and p value) in all related figures (e.g. Fig.5-6; Fig. A4-A12) that are statistically significant.
   The figures were updated adding the $R^2$ and the p-value.

2. Figure 4: Add scatter plots (and coefficients) of the corrected and reference data.
   The figure was updated. Currently it corresponds to figure 3.

Figure 3. Comparison of the voltage signal for one release (#8) from Chamber A before (Uncorrected) and after (Corrected) the baseline correction on (b) TGS 2611-C00 and (d) TGS 2611-E00, on which it is appreciated the correction of the offset preserving the amplitude enhancements linked to CH$_4$ variations. Scatter plot of the corrected (orange) and uncorrected (red) signal vs the reference CH$_4$ observations for (c) TGS 2611-C00 and (e) TGS 2611-E00. (a) Reference CH$_4$ mole fractions, also corrected using the spike correction algorithm.

3. Line99: participate in;
   Section 2.1 was rewritten to improve clarity:
   “In October 2019, TotalEnergies® performed multiple controlled releases at the TotalEnergies Anomaly Detection Initiative (TADI) facility, to investigate the capability of different detection and quantification techniques of CH$_4$ emissions from industrial facilities. The TADI test site is located northwest of Pau, France, with an approximate area of 200 m$^2$. It is equipped with infrastructure typical of oil and gas facilities (pipes, valves, tanks, etc) to simulate ‘realistic’ leaks. The terrain is flat but includes different obstacles that can affect the dispersion of the gases released to the atmosphere. Our experiment consisted of 41 controlled releases of CH$_4$ and CO$_2$, covering a wide range of emission rates of between 0.15 and 150 g CH$_4$ s$^{-1}$, with durations ranging between 25 to 75 minutes. We participated in this experiment
to develop and test inverse modelling frameworks within the TRAcking Carbon Emissions (TRACE, https://trace.lse.ipsl.fr/) program for the estimation of emission location and rates based on CH₄ mole fractions from high precision instruments (Kumar et al., 2022). We presented the inversion results for 26 releases from single point sources based on two inversion approaches, one relying on fixed-point measurements, and the other one on mobile near-surface measurements (the latter had already been documented in Kumar et al. (2021)). In both cases, the emission estimates relied on CH₄ mole fractions from high precision instruments, and on a Gaussian plume model to simulate the local atmospheric dispersion of CH₄. The results from Kumar et al. (2022) for point source emissions yielded an emission rate error of between ~23 to ~30 % and a localisation error (within a 40 m × 50 m area) of between 8 and 10 m. The controlled releases were emitted from heights of between 0.1 m and 6 m above ground level, and inside the 40 m × 50 m ATEX (ATmospheres EXplosibles) zone of the TADI facility (see Fig. 1).”

4. **Line100-101:** ambiguous for “for the estimation of … based on …high precision”, better to separate this for another sentence? “And the TRACE program is …”; See answer for comment 3.

5. **L112:** consist of doing is better to be changed to consist of sth.
   See answer for comment 3.

6. **L115:** You may mean “connected to an upstream chamber which holds the high precision instruments…”
   See answer for comment 3.

7. **L116:** (Picarro CRDS or LGR), or provide specific type; See answer for comment 3.

8. **L122:** better to use the datasheet parameter: “less than 3 ppb per month”
   We have update the phrase: “In a previous study by Yver-Kwok et al. (2015), it was proven that these CRDS gas analysers ensure high precision measurements and a low drift over time, of less than one ppb per month, although the datasheet specifies a drift of 3 ppb per month (Picarro, 2017).”

9. **L124-127:** hard to follow, needs to be rewritten in short sentences; See answer for comment 3.

10. **L129:** redundant, combine sensors: “the CH4 and environmental sensors”
   Sentence corrected:
   “Table A6 shows the TGS and environmental sensors in each chamber, as well as the type of chamber.”

11. **L131:** two sensors other sensors;
   Updated sentence:
   “Each chamber contained at least three TGS units with voltage measurements sensitive to CH₄ and two other sensors measuring relative humidity/temperature and pressure/temperature.”

12. **L134-135:** add “a”…ADC board ;… change “recorded” to records
   Updated the sentence:
   “An AB Electronics PiPlus ADC board mounted on a Raspberry Pi 3B+ recorded the voltage drop across the load resistor, providing observations every 2 s.”

13. **L146:** Why they are used in the training of models?
   To reconstruct CH4 mole fraction from observed voltage from the TGS sensors we employed a data driven approach consisting in minimising the error between the predicted output and the reference measurement on an iterative process, using a multi layer perceptron or a 2nd degree polynomial. This approach requires a sufficient number of examples from which the
model can learn the relationships between voltage variations and CH4 concentration. Contrary to the case of inverse modelling, it is not affected by the wind conditions since reconstruction models do not use wind information to derive CH4 mole fractions.

14. L217: We used
Sentence corrected:
“To assess the performance of the reconstruction models to provide dry CH4 mole fractions enhancements (above the background) from voltage drop measurements corresponding to the TGS sensors, we used a normalised root mean square error (NRMSE) per release, weighted by the inverse of the maximum peak present in the release.”

15. L219 and 222: presented
Correction accepted:
See answer to comment 14 for correction on L219.

“where y_i are the CH4 mole fraction measurements provided by the high precision instrument, \(\hat{y}_i\) are the reconstructed CH4 mole fractions, n is the number of observations present in each release, and \(h_{max}\) is the amplitude of the maximum mole fraction peak enhancement present in the release after removing the background.”

16. L220: the unit of hmax is ppm? And thus the NRMSE is dimensionless?
Yes, the NRMSE is dimensionless. The figures and the text was updated accordingly.

17. L244: change “are” to “were”
The sentence was corrected:
“The reconstruction models were trained and tested only once per chamber, following the distribution of the releases from Table 2.”

18. L245: Table 4
Table 1 and 2 were moved to SM. Table 4 was updated to Table 2 (see previous comment).

19. L275-276: how long is the typical time decay?
The time delay from synchronisation between high-precision gas analysers and TGS chambers varies between 2 to 3 minutes depending on the day of the campaign.

20. L316: discuss a bit on why
The following text was added to the discussion section.
“The combination of both sensors as input produced a reconstruction of CH4 mole fractions similar to using only one of the sensors (TGS 2611-C00). This can be explained by the fact that both of the TGS signals are highly correlated and do not add more information to the model, and the phase mismatch between both input signals produced by the filter on TGS 2611-E00 sensor.”

21. L333-335: redundant and a bit ambiguous
The phrase was corrected to reduce redundancy and ambiguity:
“For most of the cases, the modelled gradients assimilating the TGS data are closer to those assimilating the reference data than to the observed TGS data.”
22. L340, 348: comply with the journal requirements on capitals of figures and keep consistency through the text (Figure A14 and fig 9a).
The manuscript was reviewed to comply with the journal requirements.

23. L366-372: These contents seems to be more suitable for conclusion
The paragraph was moved to the conclusion section.

24. L431-433: The study of how many sensors are needed and the layout of these sensors are also needed.
We have added the following text in the discussion section about the density of the network: “Density of sensor network
In our campaign, we deployed 7 chambers connected to air inlets placed on tripods at distances of between 40 and 50 m from the emission source to capture methane plumes under various conditions. Table 3 details the number of sensors used for emission flux estimations across the controlled releases.
The optimal number of sensors for emission flux localisation and estimation is complex, influenced by varying emission rates, environmental conditions, and setup configurations. Notably, when examining cases with uniform emission rates (1 g CH4/s), such as releases 12, 2, and 21 (with 3, 4 and 5 chambers respectively), a configuration of 4 to 5 sensors consistently produced the lowest errors for both sensor types. Yet, release 21 demonstrated that even five sensors may not guarantee low errors if the plume capture is suboptimal due to environmental factors or sensor placement.
We can contrast our setup with Riddick et al. (2022), who used four sensors approximately 30 m away from the source, but without detailing their individual contributions to emission calculations.
The optimal configuration of such a relatively dense network necessitates a thorough investigation, possibly through simulations of typical emissions and the strategic addition or removal of sensors to assess their impact. However, a comprehensive analysis of optimal network configuration was beyond the scope of our study due to the limited number of data points recorded.”