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Author's response: point-by-point and all relevant changes

Manuscript title: Dynamic quantification of methane emissions at facility scale using laser tomography: demonstration of a farm deployment.

In this document, we provide a point-by-point response to the reviewer comments, including a list of all relevant changes made in the manuscript. The response to each referee comment will be structured as follows: (1) comments from referees, (2) author's response, (3) author's changes in manuscript. The format and text are almost exactly like the author's response uploaded to the Discussion page on EGUsphere, except that point *(3) author's changes in manuscript* will be more detailed with specific section and sentence references to make it easier to connect with the track-changes file. Finally, some minor language and notational inconsistencies have been found and addressed, and will be mentioned at the end of the document.

Reviewer 1

1. - Referee comment:

     Literature overview:

     I feel that your overview of the spectroscopic methods is too limited. It ignores at least the following techniques: NIR long open-path FTIR techniques [1-2], which do not suffer from the limited range as MIR FTIR due to higher brightness.

     Dual Comb Spectroscopy (DCS), which was even already employed for different emission estimations, including from cattle [3-4]

     Other methods utilizing frequency combs or super-continuum sources [5]

     There is also a wider context of tomography using path averaged measurements of gas concentration retrieved from optical measurements going back more than two decades, which would be fair to mention here somehow. Difficult to pick a single one, but for example [6].

     Finally, it seems reasonable to mention other projects with similar methodology, even if the trace gas under analysis or the scale differs. For example [7].

   - Author's response:

     We thank the reviewer for providing these additional references.

- Changes in manuscript:

  We have added the suggested additional references to the text of the introduction, starting from the 5th paragraph (see the marked-change document version for details).

2. - Referee comment:

     General research question:

     Likely as a result of the manuscript being so extensive and addressing so many facets, I feel that at multiple points the concrete research question is a bit unclear. I try to point some of these situations out in my line-by-line comments below. I encourage the authors to try to phrase their research questions precisely and go over the manuscript again, removing details where it is maybe not necessary and adding where the main message needs it.

   - Author's response:

     We agree that in the original manuscript the research questions were not clearly expressed. The research questions of this work are twofold: Primarily, we study the feasibility of LDT to monitoring emissions associated with short-term farm operations – a new application of LDT. The secondary research questions focus on the computational methods used in the reconstruction of the emission sources. Especially, we investigate whether the use of spatial constraints could improve to tolerance of the source characterization to complex wind fields and external sources. The significance of the positive answers we obtained to these secondary, computational questions extends beyond farm operations and agricultural applications overall – complex wind fields and external sources induce biases to source quantification also in other applications of LDT. To clarify the research questions, we have rewritten the last paragraph of the Introduction section.

   - Changes in manuscript:

     The last paragraph of the Introduction section has been rewritten. It now reads:

     "The present paper aims to assess the feasibility of LDT to monitor emissions associated with short-term farm operations. To that end, it reports on the first deployment of LDT at an operational agricultural facility for continuous monitoring. The system is installed at a Finnish dairy research farm to study its performance in characterising CH_4 emissions associated with manure management. To enable reliable emission characterisation, we also further develop computational methods in the BSE framework. Specifically, we introduce spatial constraints to the tomographic reconstruction based on prior knowledge on potential source locations – information often available in facility scale GHG monitoring applications. Such constraints reduce the state dimensionality. We investigate by numerical simulations whether such constraints could improve the tolerance of LDT to approximate modelling of complex wind fields and to unexpected external emission sources. Finally, the numerically validated reconstruction methods are employed to real LDT data from a dairy farm to characterise CH_4 emissions spatio-temporally."

3. - Referee comment:

     L. 47: "high-resolution" is a very flexible term, and seems a bit arbitrary here. I would estimate that the employed resolutions roughly span 5.0 1/cm to 5.0E-3 1/cm, ranging from grating spectrometers via FTIR to most laser solutions and DCS. Maybe you can be a bit more precise here with what you mean?

- Author's response:

  Author's response: In the context of the work here, meaning the spectroscopic measurement of gas concentration at surface pressure, we define high resolution as a spectrometer resolution that insignificantly affects the individual line shapes. So, taking a FHWM broadening of about 2 GHz (0.07 cm-1), this would translate into an instrument spectral response function with a FWHM < 0.007 cm-1. We have added a sentence in the text stating that for high-resolution we meant resolution < 0.01 cm-1.

- Changes in manuscript:

  The following sentence has been added to the 5th paragraph of the introduction:

  "In the context of the present work, we define high-resolution as <0.01 cm^{-1}, so that surface pressure broadened line shapes are minimally affected."

4. - Referee comment:

     L. 48: Tedeschi is an odd choice for the general statement (not limited to methane or emissions from agriculture) concerning the development of high-resolution optical gas concentration.

   - Author's response:

     We agree that Tedeschi may seem odd in the general context of generic monitoring systems, but it is highly topical to the application sector the paper deals with. Many of the general monitoring systems demonstrated would not be suitable for operational deployment in a farm environment, so Tedeschi's review provides a very in-depth review of the actual, deployed systems relevant to agriculture.

   - Changes in manuscript:

     We have kept the reference but moved it to the end of the 5th paragraph in the introduction, and clarified that this was a review relevant to the agricultural sector.

5. - Referee comment:

     L. 53: The limited range is more due to low source brightness than coherence in my understanding.

   - Author's response:

     Coherence covers two benefits as far as range consideration is concerned: 1) temporal coherence, meaning that the source emission spectral bandwidth is very narrow (hence high brightness indeed), and 2) spatial coherence, meaning a high beam quality and therefore a high beam directionality with minimum divergence losses. Therefore, we would prefer to leave the text as is to cover both aspects.

   - Changes in manuscript:

     No changes.

6. - Referee comment:

     L. 54: A citation for the developing hyperspectral remote sensing instruments is needed here. In the context of methane emissions [8] might be an option.

   - Author's response:

     We agree with and thank the reviewer for pointing this out. We have added the suggested reference accordingly.

- Changes in manuscript:

  The reference has been added to the 5th paragraph of the introduction.

7. - Referee comment:

     L. 100: Please state a clear and precise summary of the technical data of your system or at least a single publication where this is provided in a concise manner.

   - Author's response:

     The instrument level description is provided in (Daghestani et al.) and the deployed system description in (Ijzermans et al.). However, we have added a bit more details on the instrument specifications in the section 'Multi-open-path laser dispersion spectroscopy'.

   - Changes in manuscript:

     The final paragraph of Section 2.1 has been expanded with some details on the instrument specifications.

8. - Referee comment:

     L. 116: Typing "ppm x m" as ppm.m seems at least odd to me.

   - Author's response:

     We have updated to use the ppm x m notation.

   - Changes in manuscript:

     We have updated to use the ppm x m notation in the second paragraph of Section 2.1.

9. - Referee comment:

     L. 275f: What do the two noise components describe, in particular the " 1 % of the difference between the maximum and minimum values of the simulated measurements"?

   - Author's response:

     We thank the reviewer for commenting on this statement, and we agree with the need for further explanation. We have included the following statement in the first paragraph of Section 3.1, which explains the thought behind each of the noise components:

     "The first component is proportional to the magnitude of each PAC measurement, and accounts for modelling errors related to discretizing the line integrals in the observation operator (see Eq. 12) and errors related to assuming a stationary concentration distribution during the short duration of each PAC measurement. Owing to turbidity, high gas concentration measurements are associated with more uncertainty compared to those of a well-mixed low background. The second term adds equally distributed noise to every measurement in the inversion, to model a baseline uncertainty level common to all measurements that can be interpreted as instrument noise."

   - Changes in manuscript:

     The above paragraph was added to Section 3.1.

10. - Referee comment:

      L. 302f: Citation for the COMSOL model?

- Author's response:

  We have added a citation for using the k-epsilon model to simulate urban wind flows (https://doi.org/10.1016/j.egypr
  Further, we have uploaded the COMSOL model to the online repository Zenodo and included the DOI as a reference in the corresponding line (https://doi.org/10.5281/zenodo.17857384).

- Changes in manuscript:

  The references have been added to the beginning of the second paragraph of Section 3.2,

11. - Referee comment:

      L. 311ff: I am not certain on this, but your arguments for novelty seem to be things that models like GRAMM/GRAL (e.g. [9]) do for a while now. Is this the case? If so, what makes your approach novel? And why are you not using a model like GRAMM/GRAL to describe the atmospheric transport but write your own? I would suggest you address these questions at a suitable place in your manuscript.

    - Author's response:

      After careful reading of [9], we can highlight the following novelties of our approach compared to models like GRAMM/GRAL:

      First, the timescale and area considered in [9] are very different from what we report on: we aim to specially focus on dynamical (near real time) quantitative emission tomography over the scale of an emitting facility.

      Models such as GRAMM/GRAL rely on Lagrangian particle dispersion models, which track discrete particle trajectories to determine source-receptor relationships. Because of the discrete nature of this method, such models have difficulties in reconstructing continuous concentrations fields, especially in transient cases. Our approach resolves the concentration distribution at the same timescale as the measurement collection (every 7 sec), yielding near-continuous estimates of the concentration distribution. The strength of our approach lies in its ability to generate dynamic 3D concentration maps showing gas plumes moving with the wind (and 2D source maps showing the emission patterns), all with a sub 10 second time resolution (see video supplement, https://av.tib.eu/series/1944). To our knowledge, our approach is the first method currently able to generate such high-temporal resolution 3D concentration maps of greenhouse gases based on time-series of concentration measurements. Most atmospheric inversion models such as [9] have a temporal resolution of 30 mins to 1 hour, meaning that short-lived sources (such as those associated with manure management activities) cannot be resolved accurately.

      Models like GRAMM/GRAL depend heavily on a reliable emissions inventory for simulating steady-state concentration fields. As stated in the conclusion of [9]: "The most critical input is the emission inventory, which is often not available or not of sufficient quality and detail". There is a clear connection between the use of a bottom-up emissions inventory as prior information in [9], and how we are using prior information on emission sources in our laser tomographic approach. To apply our inversion approach, the user needs to provide information on only the location of emission sources, but not any pre-estimated source strengths (contrary to [9]). However, we want to stress that the unconstrained source model can still be applied if no emissions inventory or other prior source information is available.

In this work, we use a novel optical sensor based on laser dispersion spectroscopy (LDS) originally developed by one of the authors (see Wysocki and Weidmann, 2010). The sensor measures CH4 concentrations along several open paths spanning 30 to 110 m using a rotating turret. The tomography is performed out of a single set of radiating beam paths. Our setup operates at a much smaller scale (around 100 x 100 m), than the GRAMM/GRAL model used in [9], which covers more than 10 x 10 km. Further, our approach involves measuring path-averaged concentrations along several beam-paths to give more spatial information on the concentration distribution, compared to measuring pointwise concentrations using a network of low/mid-cost sensors scattered in a larger area (city-scale, such as in [9]). In our work, we have also investigated the effect of external sources contaminating the measurements. This problem affects small facility-scale measurement setups more severely than the larger city-scale setups such as in [9].

Specifically in [9], they rely on the simulation of a catalogue of more than a thousand possible steady state concentration field distributions, determined by different meteorological conditions and source evolution estimates based on bottom-up emission inventories. This is a very heavy computational burden, and the authors report that they need to store about 5 TB of simulated wind and concentration fields. Our data-assimilation based approach does not require the precomputation of concentration field simulations, but only the generation of a suitable computational geometry and mesh. Provided that the mesh is not too dense, our inversion framework and visualization of the 3D concentration and 2D source distributions can (theoretically) run alongside the data collection, with only a 5–10-minute delay from data collection to finished visualization. This makes our approach suitable (with further development) for near-real time source monitoring and leak detection at facility level, which is not achievable by methods such as in [9].

Finally, we note that although the focus of the present paper is on spatial constraining of the sources, the BSE approach generally enables also localization and quantification of emission sources without need for a priori information on the potential source locations (Voss et al., 2024, https://doi.org/10.1016/j.aeaoa.2024.100260 ; Vänskä et al., 2025, https://doi.org/10.1088/1361-6420/adfb29). This feature makes BSE different from the inversion approach described in [9] since the latter relies on inventories to constrain the emission.

- Changes in manuscript:

  No changes.

12.
- Referee comment:

  L. 343: Is N_A your $\bar{N}$ from Table 2?

- Author's response:

  N_A is introduced on L. 174, as the dimension of the source space $a_k \in \mathbb{R}^{N_A}$. The source is only confined to ground level in the mesh, and as such, we do not define the source on all N mesh nodes, but only the N_A nodes that make up the ground level. We have added a clarifying statement after the first use of N_A to make the distinction clearer.

- Changes in manuscript:

  The clarification in the third paragraph of Section 2.2.1 now reads:

"We denote $\boldsymbol{c_k} \in \mathbb{R}^N$ and $\boldsymbol{a_k} \in \mathbb{R}^{N_A}$ the vectors of nodal values of $c(\boldsymbol{x}, t_k)$ and $a(\boldsymbol{x}, t_k)$, where N is the total number of nodes in the FEM discretization and N_A is the number of nodes making up the ground level where the source distribution $a(\boldsymbol{x}, t_k)$ is defined."

13.
- Referee comment:

  L. 361 & Fig. 4: In my opinion the different colour bar in the 5th column is misleading. If you feel that a plot with a shared colour bar might be underselling your work, then maybe consider printing the integrated emissions in the top right corner of each plot in column 4-6.

- Author's response:

  We thank the reviewer for the perspective. We have modified the color bar scale for the 4th and 6th columns in the three affected figures, such that the color bars are consistent between the source columns (4th - 6th columns).

- Changes in manuscript:

  The colorbar in the affected fifth column of Fig. 4 has been updated, and the caption rewritten slightly.

14.
- Referee comment:

  L. 370f: Your argument with the approximate wind field model comes up multiple times in the manuscript and it is not always clear to me. In a synthetic study, I would assume that you still have a correct description of your "physics", assuming the model was used for forward modelling and inversion. Is this not the case? Also, in any case, is this not more a case of your windspeed measurements giving you not the full information on the local wind field?

- Author's response:

  In the synthetic study, the forward modelling and inversion did not use the same wind field model, to account for the fact that the wind field model in the inversion is known to be less representative. For the inversion, we sample the velocity field used in the forward simulation (i.e. the COMSOL generated one) at one location (to mimic a single anemometer measurement) and extrapolate that measurement with a log vertical scaling. This is the numerical equivalent to what is done in the real data study with the extrapolation of the single anemometer measurement. Therefore, we also have a significant misrepresentation of the wind field in the synthetic study. To make the point clearer to the reader, we have added the following paragraph at the end of Section 3.2.

  "In the numerical simulation study, the forward modelling and inversion do not use the same wind field model. For the inversion, we sample the velocity field from the forward simulation (i.e. the $k-\epsilon$ turbulence model) at one location and extrapolate that measurement before applying the logarithmic vertical scaling. This is equivalent to what we do in the experimental study in Sect. 4, where we extrapolate the single anemometer measurement. Therefore, we have a significant misrepresentation of the wind field in both the numerical and experimental studies."

  The reviewer is correct in observing that it is a case of the wind measurements not giving full information on the local wind field. Here, we are dealing with a case where the forward model/inversion discrepancy is caused by the fact that the wind field is sampled a distance away from the buildings, which causes the gas transport near the buildings to be distorted in comparison with the true forward solution.

Finally, we have updated the streamlines in Fig. 3 to give a better visualisation of the difference between the velocity fields used in the forward modelling and inversion.

- Changes in manuscript:

  The paragraph above has been added to the end of Section 3.2. The streamline plot in Fig. 3 has also been updated. We have now used the streamslice function in MATLAB for the visualisation.

15.
- Referee comment:

  L. 418ff: That the spatial extent of the reconstructed source in A4 is only half the width of the true source is just a result of the (spatial) constraints that you put on your "unconstrained" emission model right? Same as with the difficulty for the unconstrained model to allow for two different emission spots – you supress that possibility to make it more stable/solve the degeneracy of the ill-posed inversion problem?

- Author's response:

  The observation that the reviewer makes about the spatial extent of the reconstructed source in A4 is indeed the case. In order to model spatially small slurry tank sources with the "unconstrained" emission model, we have to use a small spatial correlation length of [10m, 10m]. As the reviewer notes, this puts some constraints on how large sources in A4 can get, since the spatial extent of A4 is comparably larger than A1-A3.

  However, we feel the need to address the observation the reviewer makes about the difficulty of the unconstrained model to allow for two sources. The difficulty of the unconstrained emission model to allow for two different emission spots is not due to the prior model suppressing that option. Nothing in the prior model prevents multiple sources from being active at once. However, when two adjacent sources are active at once, the inversion problem suffers from severe non-uniqueness problems, which can cause the model to believe that only one source is active at once. The previous studies on laser dispersion tomography (Voss et al., 2024, https://doi.org/10.1016/j.aeaoa.2024.100260 ; Vänskä et al., 2025, https://doi.org/10.1088/1361-6420/adfb29) have shown the ability of the unconstrained emission model to track multiple sources simultaneously. However, the sources need to be more spatially separated than in the slurry tank case, for the non-uniqueness problems to be resolved.

  Furthermore, as demonstrated in Case 2 of the present paper, constraining the source model makes the source estimates more tolerant to disturbance caused by external sources, i.e., sources outside the computational domain.

  We have added a sentence to clarify that point at the end of the second paragraph of section 3.4.3:

  "Whilst LDT has been demonstrated to resolve multiple sources (Voss et al., 2024; Vänskä et al., 2025), if sources are closer than the expected source correlation length, the inversion will tend to aggregate these into a single source."

- Changes in manuscript:

  The above sentence has been added to the second paragraph of section 3.4.3.

16.
- Referee comment:

  L. 432: See also comment to L. 370f. Some explanation why the misrepresentation gives you the correct magnitude of model error would be beneficial.

- Author’s response:

  We have concluded this observation on the misrepresentation of the wind field based on performing inversions with the true wind field, i.e. using the whole COMSOL generated wind field in the inversion, instead of only sampling it at one location and extrapolating. An example of such a numerical study is illustrated in Fig. 1 below. Here, we have performed the inversion in simulation case 3 using the constrained model and the exact same setup as in the manuscript, the only difference being that we have assumed complete knowledge of the wind field in the inversion. When the complete wind field is known, the emission rate estimate is better than when we only have partial wind information. However, these simulation results are not included in the manuscript, since they do not represent a physically realistic situation, i.e. we are never going to have complete knowledge of the wind field in the entire domain.

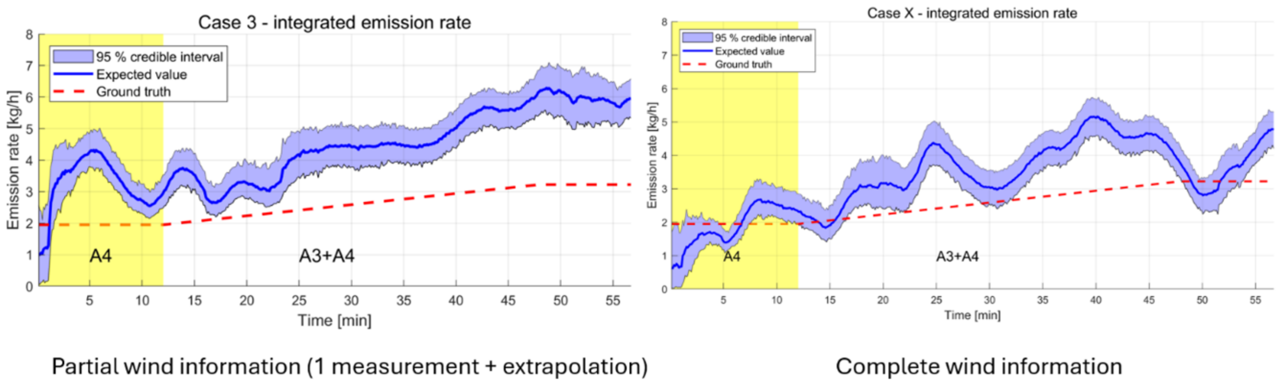

Figure 1: BSE inversion for simulation case 3 with partial (left) and full (right) wind information.

  To illustrate this point, we have performed the inversion of simulation case 3 using the constrained model and the exact same setup as in the manuscript, the only difference being that we have assumed complete knowledge of the wind field in the inversion. When the complete wind field is known, the emission rate estimate is better than when we only have partial wind information, see the figure provided above.

- Changes in manuscript:

  No changes. The above figure is not included in the manuscript since it does not represent a realistic case of what is possible in the field, i.e. complete knowledge of the wind field.

17.
- Referee comment:

  L. 433: Language. Because “it is” trapped. . .

- Author’s response:

  We thank the reviewer for this observation and have modified the text accordingly.

- Changes in manuscript:

  The error was fixed in the final paragraph of Section 3.4.3.

18.
- Referee comment:

  L. 434f: This points again to the point which is unclear to me about the misrepresentation of the wind fields in the synthetic study. Are there different models at work?

- Author's response:

  We thank the reviewer for pointing out that it is not clear which wind field models have been used in the synthetic study. We refer to our response to the reviewer comment made for L. 370f, and the clarification text we added in the manuscript.

- Changes in manuscript:

  See "Changes in manuscript" point for reviewer comment 14.

19.

- Referee comment:

  L. 452: Why did you not include May 21st? This would provide a good test for your method if there is not a large amount of prior knowledge on the emissions (spatial emission pattern and emission time). Including this for the stated reason makes this seem like pretext.

- Author's response:

  May 21st was not included in the analysis, because the current work focuses on reconstructing known emissions events with their near real time dynamic, so that they can be correlated to known farming events/activities.. To do so, in our analysis, we need to establish a clear cause/effect relation in all observed emission events and patterns, such that we would know what emissions are real and what are artefacts. Therefore, we did not deem that including May 21st would contribute to the results in any significant way, since the lack of meta data for the day does not allow us to discriminate between what is real and what is artefactual. However, we recognize that that not displaying this dataset maybe raise concerns. Therefore, we have now added the outcome from the 21th of May inversion in the supplemental material. Some clear emission activities are occurring before noon, but we cannot attribute these to any known management operation as the farmer omitted logging on this day.

- Changes in manuscript:

  The result of the BSE inversion for May 21st has been added to the Supplementary Material, see Fig. S7. Further, the following sentence has been added to the second paragraph of Section 4.

  "In principle, the inversion should be capable of determining whether the elevated concentrations observed on May 21st are attributable to local emissions or transport from external sources. However, the primary objective of this study is to demonstrate the capabilities of LDT in an operational farm setting, rather than to provide a comprehensive characterization of methane emissions from a manure management facility. Accordingly, this event has been excluded from the main analysis. However, we have included the analysis of the measured PAC data from this day without metadata in the Supplementary Material, see Figure S7."

20.

- Referee comment:

  L. 462f: I think your choice of taking the background from the first two days of measurements requires a bit more explanation. Over the course of three weeks the background can of course change. Can you provide an argument/measurement data that the background did not increase during this time period in a way that it significantly interfered in relation with the scale of enhancements you were targeting?

- Author's response:

We chose to estimate the background using the first two days of measurements because the wind speed measurements for the first two days are consistently high throughout both day and night (see measured wind speed in the top row of Fig. 9). This indicates a well-mixed air volume less affected by potential local sources and therefore a representative estimate of the background concentration. We recognize the possibility of the background concentration changing slightly during the measurement period. However, we deem that small changes in background concentration (typically at most around ±50 ppb due to atmospheric stability primarily) will have only a negligible effect, based on the relative differences between the concentration peaks we measure during the manure management activities and the estimated background (peak of several ppm). We have modified the text in the manuscript to include this explanation and improve clarity.

“The model uses a fixed background concentration estimate c_{bg} for the duration experiment, whilst the background may vary slightly during the measurement period, either owing to the atmospheric stability, or medium range sources. However, the emission source estimates are overall not highly sensitive to value of c_{bg} because it is only used as the expected value of the concentration on the inflow boundary of the computational domain. The model for the inflow boundary concentration is stochastic, and it allows for deviations from the expected value – as demonstrated by Case 2 in which the inflow concentration deviates heavily from the background due to an external source.”

Regarding background estimation in the simulation study, we have added a small comment in the second-to-last paragraph of section 3.1, detailing how the background estimation was done in the simulation study. As a result of rechecking the background concentration estimates for all simulation study cases, we found a small mistake in the specification of the background concentration for the unconstrained model in simulation case 2. The affected figures 5 and 6 have been updated, with no changes to the interpretation of the results.

- Changes in manuscript:

  The fourth paragraph of Section 4 has been restructured to include the above statement in quotation marks. The plots in the second and fifth columns of Fig. 6 were updated, as were the plot in the first column and the middle row of Fig. 5. Finally, the text describing the background concentration estimates in Table 2 has been modified.

21.
- Referee comment:

  Fig. 10: I would like to see a more thorough addressing of all the other peaks. A4 shows emissions all the time. Are they real or artefacts? Are the emissions of A4 during the agitation event of A3 real or misallocation?

- Author’s response:

  We thank the reviewer for pointing this out. We have modified the manuscript by expanding the paragraph describing Fig. 10 in Sect. 4.1.1. We added:

  "The reconstructed emissions from A4 during the agitation of A3 are a misallocation that should have been attributed to A3. Throughout the day, the relatively constant but small emission rate observed from A4 characterizes persistent emissions from the manure pile until its removal as part of Event 2. The figure also suggests that the emission rate for the manure pile A4 develops significantly as night falls. The partial misattribution of sources and the emissions from the dry manure pile are discussed further in Section 4.2".

Further, we have expanded the part in the discussion section (4.2) that addresses why A4 is showing emissions all the time, including evening and night.

"As such, the elevated emissions observed from A4 during nighttime are believed to be real, however, the magnitude is overestimated due to the accumulation of concentration caused by a highly stable atmosphere, effect that is not represented by the transport model."

- Changes in manuscript:

  The two above statements were added to the first paragraph of Section 4.1.1 and the third paragraph of Section 4.2, respectively.

22. - Referee comment:

  L. 496 and Fig 11: The scale of (potential?) misrepresentation between A3 and A4 seems to be similar between Fig 10 and Fig 11.

- Author's response:

  We thank the reviewer for highlighting this observation. The experimental results for Events 1-3 generally show a tendency for some emissions to be misrepresented between A3 and A4 during the manure management activities. When first mentioned, we added a note that they are seen in all events.

- Changes in manuscript:

  We have modified the fourth paragraph of Section 4.2 to address the misrepresentation further.

23. - Referee comment:

  L. 512 and Fig. 12: I am not fully convinced that the more flat emissions for A4 is only a result of the removed pile. You always argued with the misrepresentation of the wind fields – could this also be a result of a different wind situation where your model fits better and shows less erratic behaviour? Is this a general observation for all the days after removal of the pile? I feel this needs more argumentation.

- Author's response:

  It is certainly possible that better wind conditions during some time periods of the experimental campaign can provide a better model fit with less uncertainty. However, we argue that the measured wind directions for May 22nd (see bottom row of Fig. 9) are sufficiently varied, for the model not to produce a biased emission rate estimate during the entire day. If the misrepresentation of the wind field would be cause of the more flat emissions for A4, then this bias would only be present in those time periods with superior wind conditions. And since we see consistently more flat emissions from A4 after the manure removal during all the wind conditions, we deemed the manure removal to be a more plausible explanation for the reduced A4 emissions.

  To substantiate the claim further, we computed the BSE inversion for all days of the measurement campaign. We then divided the emission rate estimate for A4 into two sections; one from midnight to 12:59, and one from 13:00 to midnight (since the manure pile removal ended around 13:00 on May 19th). We then computed the mean emission rate of each section for all days and plotted the two timeseries in Fig. 2 (see below). On average, the emissions are lower after the manure pile removal.

On the day of the manure pile removal, we see a spike in the pre-13 mean but not in the post-13 mean. This figure has also been included in the Supplementary Material, and we have referred to it in the main manuscript by including the below sentence at the end of the third paragraph in the discussion section 4.2: "We refer to Fig. S8 in the Supplementary Material for a comparison of the estimated daily mean emission rate from A4 for the entire duration of the experiment."

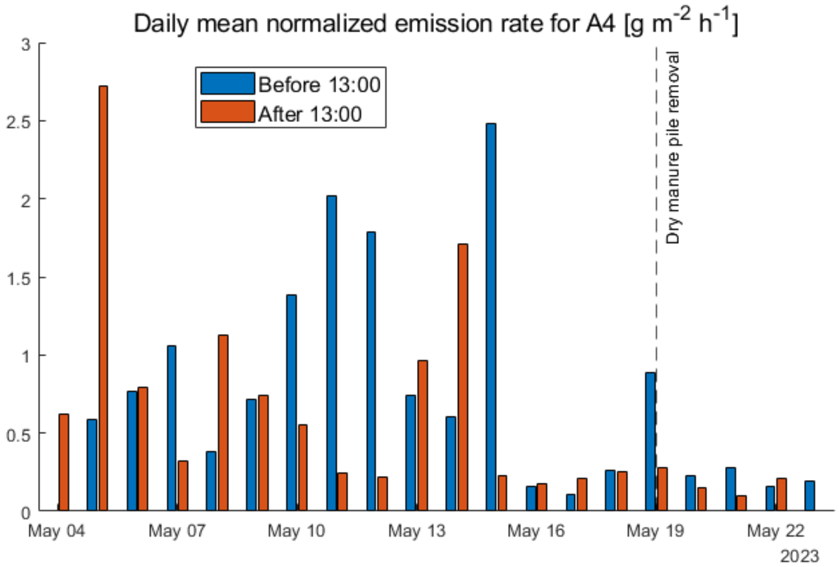

Figure 2: Daily means pre/post 13:00 plotted for every day in the measurement campaign.

Unfortunately, we cannot say with certainty whether this is a general observation for ALL days after removing the pile. Unfortunately, the measurement campaign ended shortly after Event 3, so we cannot give any definitive conclusions about the trends of the emissions from A4 after May 23rd. According to our analysis on the days we did consider, it is a general observation that emissions from A4 have decreased significantly after manure pile removal.

- Changes in manuscript:

  We have added the above figure to the supplementary Material, see Fig. S8. Further, we have added the sentence mentioned above to the third paragraph of Section 4.2.

24. - Referee comment:

  L. 526ff: Neither the mechanism of release (trapped bubbles) nor agitation by heavy rainfall or similar seem to be directly part of your study. To me you mix too much your own measurements, results and conclusions with those of others.

- Author's response:

  We thank the reviewer for pointing this out. We have modified the section to remove these results from other works to be more focused and less ambiguous.

- Changes in manuscript:

  The statements related to agitation by heavy rainfall or similar in the first paragraph have been removed from the first paragraph of Section 4.2.

25. - Referee comment:

L. 533f: “The emission rates inferred correlate very well with the known activity windows.” I do not necessarily agree with this statement, since you only looked at those activity windows (for example ignored the measurements on May 21st.) and in general your timetrace shows (summed) emissions at nearly any time. So please be more precise here what you are actually stating.

- Author’s response:

  This is a good point we agree to. To mention correlation, we would need to compare the like with the like, and we have no emission rate truth-set to correlate the inferred results to. So no correlation can be strictly established. Instead, we wish to state that no actual farming management event we know of has been missed, and all the known activities can be attributed to an emission profile from the inferences. We have no false negatives when considering the activities reported by the farmer. We have therefore updated the statement as:

  "The temporal series of emission rates inferred have all captured signals associated with known farming management activities: there is no false negative in reporting quantitative rates associated with known events. Besides, the reported rates for slurry pumping events are consistent to those reported in previous works (VanderZaag et al., 2014)".

- Changes in manuscript:

  The above statement has been updated in the second paragraph of the discussion Section 4.2.

26.
- Referee comment:

  L. 534ff: I also don’t agree with the statement of the dropping emissions here. Around 14:00 is a time period where a similar low emission is measured. Or it could be a function of other parameters (drop height?). Also I don’t understand why this is important. It seems only relevant if you want to infer in detail the release mechanism on a microscopic level? More general speaking, I think in this paragraph the research question at hand gets a bit muddled.

- Author’s response:

  We thank the reviewer for the comment. The statement was meant to explain why we observe a decreasing emission rate from pumping to pumping. We agree with the reviewer’s opinion that the statement is only relevant if we want to describe the release mechanisms and drivers behind the emissions, which is not the focus of this paper. Based on this lack of relevance for the research questions at hand, the statement has been removed from the manuscript.

- Changes in manuscript:

  We have removed the statement in question from the first paragraph of the discussion Section 4.2.

27.
- Referee comment:

  L. 543: “These signals disappear after the manure pile is removed” – This is a good point which addresses partially earlier questions of mine, but I would encourage you to address this concisely in more detail in the context of the remarks above.

- Author’s response:

  We appreciate the reviewers’ thoughts about the statement. We have moved it to the end of the paragraph and edited it slightly to make the statement more concise.

- Changes in manuscript:

  The statement: "These signals disappear after the manure pile is removed, and no further nocturnal emissions from A4 are seen for the rest of the experimental study. We refer to Fig. S8 in the Supplementary Material for a comparison of the estimated daily mean emission rate from A4 for the entire duration of the experiment." was moved to the end of the third paragraph of Section 4.2. Further, we added a figure to address the point in question further. See response to reviewer comment number 23 above.

28.
- Referee comment:

  L. 555-558: This seems to be the actual main question of your discussion to me, because it is mainly the question “what are real emissions and what are artefacts?”. But I think it requires a more structured argumentation.

- Author’s response:

  We thank the reviewer for observing this point about the main body of the discussion. We have restructured the last two paragraphs of the discussion to better guide the argumentation towards this point.

- Changes in manuscript:

  The last paragraphs of the discussion Section 4.2 has been restructured and rewritten slightly.

29.
- Referee comment:

  L. 584f: The constrained source model should always be more reliable since you add a lot of additional information.

- Author’s response:

  We agree with the reviewer that this is indeed true (given that the constraints are correct), and further, this reasoning was the main driver for us not doing the experimental analysis with the unconstrained source model.

- Changes in manuscript:

  No changes.

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Reviewer 2

1.
   - Referee comment:

     L1:2- Consider focusing on Methane rather than GHGs.

   - Author's response:

     We thank the reviewer for this suggestion. However, whilst we are demonstrating the emission monitoring system based on Laser Dispersion Tomography (LDT) for methane in an agricultural context, the approach is not limited to methane monitoring and is widely relevant to GHG emission monitoring at facility scale. We have already monitored CO2 as part of the Science for Clean Energy project (https://science4cleanenergy.eu/) and are now working on N2O emissions (manuscript in progress). Our overall aim and applicability of the LDT is therefore wider than only methane, and we would like to keep the generality of the GHG monitoring statement.

   - Changes in manuscript:

     No changes.

2.
   - Referee comment:

     L2:3- Consider explicitly stating the gap: facility-scale dynamic quantification under complex wind conditions.

   - Author's response:

     We thank the reviewer for the suggestion and have rephrased the corresponding part of the abstract to include the statement.

- Changes in manuscript:

  The third sentence of the abstract has been rewritten.

3. - Referee comment:

     L3:4- You might clarify that this is an integration of LDS and Bayesian inversion, currently that is implicit.

   - Author's response:

     We agree with the reviewer that this sentence could use some clarification. We have expanded the statement to include the reviewer's suggestion.

   - Changes in manuscript:

     This has also been included in the revision of the third sentence of the abstract.

4. - Referee comment:

     L7:11- This is a very long sentence, and requires to be shortened. Consider specifying that constraints reduce state dimensionality and improve robustness. The phrase "investigate numerically whether such constraints could improve tolerance..." is long and slightly convoluted.

   - Author's response:

     Thank you for bringing this to our attention. The long sentence has been split up to make it shorter and more concise. We have included the phrase about the constraints reducing state dimensionality and improving robustness in the last paragraph of the introduction. The sentence now reads:

     "Particularly, we introduce spatial constraints to the tomographic reconstruction based on prior knowledge on potential source locations – information often available in facility-scale GHG monitoring applications. We investigate numerically whether such constraints could improve the tolerance of LDT to misrepresentations induced by complex wind fields caused by building effects, and/or presence of interfering external emission sources, both highly likely to characterize a real-world farm environment".

   - Changes in manuscript:

     The sentence in the abstract was split up and rewritten.

5. - Referee comment:

     L11:17 - The abstract could benefit from at least one quantitative performance indicator (e.g., reduction in posterior uncertainty by X improvement in bias, etc.).

   - Author's response:

     We thank the reviewer for this excellent suggestion as it leads to a more convincing argument of why the unconstrained source model performs better than the unconstrained model. We have calculated the average reduction in posterior uncertainty for simulation Case 1 and reported this quantitative performance indicator in the abstract and the last paragraph of Section 3.4.1. Simulation Case 1 is the only case where the posterior means for both the unconstrained and the constrained model are close to the true value, and where the posterior credibility interval widths are thus directly comparable.

   - Changes in manuscript:

We included a sentence in the abstract on the reduced posterior uncertainty as the quantitative performance indicator. Finally, we included a sentence on this at the end of the discussion of the simulation case 1 results in Section 3.4.1.

6. • Referee comment:

  L28 - The claim that livestock represents "14.5% of human-induced greenhouse effect" needs careful phrasing — is this global GHG or methane only? Clarify.

  • Author's response: The specific claim in question concerns global GHG, not just methane. We agree with the reviewer that this sentence needs clarification and we have modified it accordingly. The sentence now reads:

  "Agriculture, waste management, and fossil fuel production emit the most methane of all industrial sectors, of which agriculture is the largest contributor (∼150 Tg CH4 per year) to the human-induced methane footprint (Jackson et al., 2024; Smith et al., 2021). Not restricted to CH4, the livestock sector alone represents 14.5 % of the total human-induced greenhouse effect (Tedeschi et al., 2022)."

  • Changes in manuscript:

  The beginning of the second paragraph of the introduction was changed to the above statement.

7. • Referee comment:

  L43 - The sentence: "Methods using static chambers measure emissions directly…" These measure fluxes indirectly via accumulation rate; rephrase this for precision.

  • Author's response:

  We thank the reviewer for this observation and have rephrased the sentence in the manuscript to clarify the statement. It now reads "Methods using static chambers measure emissions indirectly through the accumulation rate of gas concentration within a fixed volume."

  • Changes in manuscript:

  The fourth paragraph of the introduction was modified with the above sentence.

8. • Referee comment:

  L53 - The critique of open-path FTIR ("require very large retro-reflectors") requires a citation.

  • Author's response:

  Following a similar comment from the reviewer 1, we have added two citations where retroflector arrays of about 60cmx60cm are used. Namely, (Deutscher et al., 2021; Schmitt et al., 2023).

  • Changes in manuscript:

  Two references were added to the statement in question in the fifth paragraph of the introduction.

9. • Referee comment:

  L56 - The ">100 kg h^{-1}" detection threshold needs contextual comparison to expected farm emissions.

  • Author's response:

  This is a good point for clarification. The text already mentioned the 1 to 10 kg h-1 range, but we have clarified that this range is the expectation for an average European farm, we also added a supporting publication (Vechi et al., 2022).

- Changes in manuscript:

  The sentence in the fifth paragraph of the introduction was changed, and a reference was added. It now reads:

  "CH_4 observation satellites with high-spatial resolution have reported emission >100 kg h^{-1}, for example, relevant to very large cattle facilities (**?**), but not sensitive enough to resolve the 1–10 kg h^{-1} level of emissions expected from average size European farm (**?**)."

10. - Referee comment:

      L279:285 – How similar are these sources to the actual farm field were measurements were

    - Author's response:

      We chose the emission rates of the simulated sources to yield path-averaged concentration (PAC) measurements that are similar in magnitude to the concentration measurements from the experimental campaign (peak signals between 5-15 ppm). Figures S3 and S5 in the supplementary material show the simulated PACs for the supplementary cases, designed to be around the same level as the experimental data (Figure 8 in preprint). We have added the following sentence to the opening paragraph of section 3.1 to clarify this point.

      "The simulated source strengths were chosen such that the synthetic PAC data were of similar magnitude to the observational data collected in the experimental campaign, see Fig. 8."

    - Changes in manuscript:

      The above sentence was added to Section 3.1.

11. - Referee comment:

      L452 - You exclude May 21 due to lack of metadata. But could inversion detect whether it was real emission vs transport? This requires discussion

    - Author's response:

      From the controlled simulation studies, we are confident the inversion is able to discriminate between real emission and transport from sources external to the facility domain. However, for the elevated concentration data on May 21st we don't have the meta data of the operation log from the farmer. As such, we prefer to omit this analysis in the main article since the focus of the study lies in demonstrating the capabilities of LDT, and how the dynamic nature of the outcomes can resolve farming management events. To be exhaustive, we have nevertheless analyzed the measured data from May 21st without metadata and included the results in the Supplementary Material.

    - Changes in manuscript:

      See "Changes in manuscript" point for reviewer 1 comment 19. A figure was added to the Supplementary Material, Fig. S7.

12. - Referee comment:

      Missing quantitative validation. Major limitation: No independent flux measurement (e.g., chamber, flux balance) for comparison.

    - Author's response:

Indeed, the farm deployment could not be organized with inclusion of any other independent flux measurement systems at the time. To mitigate this aspect, we have provided the references in which the LDT has been validated against controlled released of methane (Voss et al., 2024, and Vanska et al. 2025). Within actual farm deployment we have used published studies to show that the measurements we report are consistent with previous measurements reported in the literature (see response to next point below). The manuscript does report consistent quantification, as well as demonstrating high temporal resolution emission rate evolution, shown to capture the known farm operation events.

- Changes in manuscript:

  No changes.

13.
- Referee comment:

  Emission magnitudes are not benchmarked against expected slurry emission rates.

- Author's response:

  We thank the reviewer for pointing this out. We have added Table 3 to the discussion section that summarizes the comparison between our LDT estimated emission rates with the ones found in referenced literature.

- Changes in manuscript:

  The end of the first paragraph was modified and Table 3 was added.

14.
- Referee comment:

  You should: Compare estimated kg h^{-1} to literature typical values.

- Author's response:

  We refer to L529:532 in the original preprint for this comparison (citations: VanderZaag et al. 2010, 2014), and Park et al., 2006. To reinforce the comparison with the existing data in the literature we added a reference (Arndt et al. 2018). The updated section now reads:

  "The increased emissions we observed are consistent with the measurements from VanderZaag et al. (2010, 2014), who reported an almost 10-fold CH4 emission rate increase from agitation. The reported peak CH4 emission rates during agitation range from 6 g m^{-2} h^{-1} (VanderZaag et al., 2010) to 37 g m^{-2} h^{-1} (VanderZaag et al., 2014). During usual slurry storage and outside any agitation periods, normalized CH4 emission rates of 4.0 g m^{-2} h^{-1} and 1.2 g m^{-2} h^{-1} have been reported from facilities (settling basins and anaerobic lagoons) at 2 different commercial dairy farms in California using an open-path FTIR setup (Arndt et al., 2018). Emission rates of 10.8 g m^{-2} h^{-1} were reported for swine slurry (Park et al., 2006)."

- Changes in manuscript:

  The first paragraph of the discussion Section 4.2 was updated.

15.
- Referee comment:

  Discuss uncertainty credibility.

- Author's response:

The question of how reliable the uncertainty limits are, is indeed important. Ideally, the approximate 95 % credible interval reported for the integrated emissions should contain the true value of the integrated emission 95 % of time. As Figure 5 shows, this is not always the case. The underestimation of the uncertainty is a result of multiple modeling errors. In the simulation studies, we deliberately used approximate, spatially constant, wind field in the gas transport model, because in a realistic setup we do not have information on the complete spatio-temporally varying wind field. This explains majority of the uncertainty underestimation, as demonstrated by the extra figure we provided in response for the first reviewer's comment. The figure shows that in an ideal case of known complex wind field the integrated emission rate in Case 3 would be less biased and the approximate credible interval more representative of the actual estimation error. Another source of error in the estimation is the discretization: Again, in BSE we deliberately used sparser finite element mesh in the transport model than in the model used for creating the synthetic data. The resulting discretization error also contributes to the misspecification of the source's point estimate and its uncertainty. In the real experiment there are also various other potential sources of uncertainty that, if not accounted for in BSE, cause underestimation of the estimate uncertainty. We note, however, that a systematic framework for accounting errors caused by model approximations and uncertainties exists; it is referred to as Bayesian approximation error method (Kaipio, J. P., & Somersalo, E. (2005). Statistical and computational inverse problems. New York, NY: Springer New York.). This framework has already been adopted also to account for, e.g. uncertainties in the velocity fields in BSE, reducing the point estimate bias and correcting the uncertainty estimates (Lipponen, A., Seppänen, A., & Kaipio, J. P. (2010). Nonstationary inversion of convection-diffusion problems-recovery from unknown nonstationary velocity fields. Inverse Problems Imaging, 4, 463-83.) Adaptation of the approximation error method to LDT, however, is out of the scope of the present study.

We have added subsection 3.5 "Discussion on the simulation study" to discuss the uncertainty credibility as per the reviewer's suggestion.

- Changes in manuscript:

  A shortened version of the author's response text was added as subsection 3.5.

Minor changes unrelated to reviewer comments

1. Correction: "Fig." → "Figs.". Second paragraph of Section 3.2.
2. Change italicisation in Eq. 13: "k" → "k". Denote the von Kármán constant by k to avoid confusion with the k-ϵ turbulence model.
3. Fixed typo: "g h^{-1}" → "g m^{-2} h^{-1}". First paragraph of Section 3.4.1.
4. A small note on the different y-axis limits in the middle row of Fig. 5 has been added to the caption.