## Optimizing Methane Emission Source Localization in Oil and Gas Facilities Using Lagrangian Stochastic Models and Gradient-Based Detection Tools

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Response to Reviewer (Anonymous Referee)

Review Comment	Author' Response	Line changed
Editor		
	Thanks for pointing this out. I propose to add the text below to include more description on the Gradient Indicator tool: "Let $C(x,y)$ denote the $CH_4$ concentration matrix where $x,y \in \{1,2,, N\}$ , and N is the number of grid cells in each spatial dimension. Each cell represents a grid cell with a specific resolution. The matrices are reoriented such that the origin corresponds to the sensor location and rows increase in the downwind direction.  To identify spatial gradients consistent with plume structure under a dominant wind direction, a 1D moving window of length (L), varying in a loop starting from one, is applied along each row i. For each window starting at column index j, a gradient sub vector is $g_c$ and the window is flagged as a decreasing gradient if the rank	

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 $g_c = [C(x, y), C(x, y)]$ +1), ..., C(x, y + L)-1) $rank(g_c) = (L, L - 1, ..., 1)$ 

The grid coordinates (x,y) are stored as valid gradient start points if the condition is met. As shown in Error! Reference source not found., the gradient length will increase incrementally with each loop and proceed in both x and y directions until at least one of the top 5% highest concentration points remains on the map. At each step, the number of times each point persists across varying gradient lengths is recorded as a resistance rate. Points with the highest resistance rates—those consistently identified as strong gradients—are then filtered to ensure they fall within the dominant wind direction sector, confirming that they are positioned upwind of the sensor."

Additionally, it's not clear if the main result tables (Tables 1-3) and figure (Figure 7) are for one of the sourcesensor configurations used in the simulation study, or an average over all source-sensor configurations. This makes the results hard to interpret, as different configurations could have very different detection / localization capabilities.

Thank you for the helpful suggestion. To avoid confusion for future readers, we have revised the text in the Results section to indicate that Tables 1–3 and Figure 7 represent aggregate results across all source-sensor configurations, rather than a single case. We now explicitly state that distance-based groupings were used to interpret results across these varied configurations, while other variables (e.g., height difference and stability class) were examined in isolation. Suggested modification: "Thank you for the helpful suggestion. To avoid confusion for future readers, we have revised the text in the Results section to indicate that Tables 1–3 and Figure 7 represent

	aggregate results across all source-	
	sensor configurations, rather than	
	a single case. We now explicitly	
	state that distance-based	
	groupings were used to interpret	
	results across these varied	
	configurations, while other	
	variables (e.g., height difference	
	and stability class) were examined	
	in isolation."	
TP1 1 1 1		
There also appear to be	Table 3 is presented in Figure 7(c).	
inconsistencies between the Table	There is an order issue in Figure 7	
3 and Figure 7(b).	(b) that is suggested to be fixed as	
	follows:	
	1.00 50% CI	
	(Q0.75) (Sp. 10)	
	OD 940 OD 0.50	
	Probability of	
	g 0.25	
	95% CI	
	0.00 25 50 75 100 125 above 125 Relative distance between source and sensor	
I think that the writing could be	Thank you for this valuable	
refined; some ideas are expressed	feedback. To improve clarity and	
imprecisely, making it hard to follow	precision, we are undertaking a	
at times.	thorough manuscript revision.	
	Specifically:	
	Specifically.	
	We will revise imprecise	
	or ambiguous sentences to	
	communicate each idea	
	clearly and concisely.	
	<ul><li>We are having</li></ul>	
	independent colleagues	
	with domain	
	expertise review the	
	revised text to help identify	
	areas where the language	
	may remain unclear.	
	TI CC . 111.1 1	
	These efforts will help ensure the	
	manuscript is more accessible to	

L58-59: The authors should note that gaps in CEMS detections result from wind blowing between sensors. These periods can be identified (and addressed) using an atmospheric dispersion model. For example, see:

https://doi.org/10.1021/acs.estlett.4c00 687

Thanks for pointing this out. Our recent work on sensor placement optimization targets this limitation by showing that, with strategic placement, even a single sensor can effectively capture emissions from multiple directions and sources, thereby reducing the occurrence of non-detect periods. The paragraph was modified to point to this publication "While combining multiple techniques can enhance source localization, stationary sensors still struggle to detect and differentiate emissions from multiple sources. Continuous Emission Monitoring Systems (CEMS), as noted by Jia et al. (2023), tend to mostly measure ambient methane concentrations rather than direct emission rates, thereby raising the issue of source identification when there are more than one emitters. Wang et al. (2022) stated that 1minute average frequency for **CEMS** improves the chance of detecting 24hourlived emissions that occur brie fly. However, additional operator input is often necessary to refine accuracy. It is also important to note that detection gaps in CEMS can occur when wind advects emissions between sensors. resulting in periods where elevated concentrations go unrecorded. As demonstrated by Daniels et al. (2024), such "nondetect times" can be systematically identified and addressed using atmospheric dispersion modeling—specifically through the use of a probabilistic framework that reconstructs likely emission durations by simulating plume transport and wind-driven

sensor coverage."
Daniels, W. S.; Jia, M.;
Hammerling, D. M. Estimating
Methane Emission Durations
Using Continuous Monitoring
Systems. *Environ. Sci. Technol. Lett.* **2024**, *11* (11), 1187–
1192. <a href="https://doi.org/10.1021/acs.estlett.4c00687">https://doi.org/10.1021/acs.estlett.4c00687</a>

L60-64: The authors should note that there is variability in localization and quantification performance across the commercial solutions studied in Bell et al.

2022 (and in the subsequent ADED evaluations, see

https://doi.org/10.1021/acs.est.3c0851 1 and

https://doi.org/10.26434/chemrxiv-2024-f1znb-v2). As the authors note, some commercial solutions struggle to localize emissions, but others perform this task relatively well when evaluated at METEC.

Thank you for pointing this out. We agree that the language should better reflect the variability across commercial CEMS solutions. Not all systems perform poorly—some demonstrate strong localization under ideal testing conditions.

Suggested text for manuscript: "Performance across commercial CEMS solutions varies considerably under controlled testing. While several point-sensor network systems often exhibited low localization accuracy (< 50%) at the equipment unit level (Ilonze et al. 2024, Cheptonui et al. 2025), scanning/imaging solutions reliably achieved higher accuracy and precision (> 50 -90%) (Zimmerle et al. 2025, Cheptonui et al. 2025). Retesting of some solutions showed measurable improvements over time in localization and detection capabilities under METEC protocols (Day et al. 2024). Our analysis focuses specifically on stationary PSN configurations and the inherent limitations in source localization using downwind concentration measurements without supplementary modeling or imaging support."

L82-83: This is not entirely true; there are several open-source methods for CEMS

inversions in the literature. See, for example:

https://doi.org/10.48550/arXiv.2506.0 3395.

https://doi.org/10.1021/acsearthspacechem.2c00093,

https://doi.org/10.1525/elementa.2023. 00110, https://doi.org/10.5194/amt-11-1565-2018. The authors are correct, however, that commercial CEMS solutions

often do not make their algorithms completely open source.

Proposed revised text:

"While several open-source inversion methods for CEMS data have been proposed in the academic literature (Liang et al., 2024; Barkley et al., 2022; Daniels et al., 2023; Fiehn et al., 2018), most commercially available CEMS solutions rely on proprietary algorithms—often involving some form of backtrajectory or dispersion modeling—which limits transparency and independent validation of source attribution."

- Daniels et al.
   2025. <a href="https://doi.org/10.485">https://doi.org/10.485</a>
   50/arXiv.2506.03395
- Weidmann et al.
   2022. <a href="https://doi.org/10.102">https://doi.org/10.102</a>
   1/acsearthspacechem.2c000
   93
- Daniels et al.
   2023. <a href="https://doi.org/10.152">https://doi.org/10.152</a>
   5/elementa.2023.00110
- Aden et al. 2018 <u>https://doi.org/10.519</u> 4/amt-11-1565-2018

The concept of a footprint is discussed in the introduction and methods sections, but is not fully defined. I recommend defining this term in the introduction, given that it is a key idea in the manuscript.

Thank you for emphasizing the importance of fully defining the concept of a footprint. We agree that it is a key term in the manuscript and that the current explanation could be more precise and comprehensive.

We have revised the introduction to include a more detailed definition to address this. The new text explains that a footprint quantifies the spatial sensitivity of a measurement to upwind surface emissions and distinguishes between concentration and flux

footprints, including their respective units and applications. This revision clarifies the role of footprints in our study and reduces the risk of confusion for readers unfamiliar with the concept.

In atmospheric research, a footprint describes the source area from which air is arriving at a measuring point of an eye-friendly gas sensor. This is an illustration of how emissions from the multiple sources spread and be seen at the sensor, considering how atmospheric transport and dispersion work.

There are two common types of footprints:

- A concentration footprint describes the spatial sensitivity of a sensor to unit emissions at different locations and has units of seconds per square meter (s m<sup>-2</sup>). This unit expresses how much a unit emission from a specific area (1 m<sup>2</sup>) contributes to the concentration at the sensor over time. The concentration footprint is especially relevant when interpreting measurements from point-in-space sensors or passive samplers that report mixing ratios (e.g., ppm or ppb).
- On the other hand, a flux footprint indicates the contribution of surface-atmosphere exchanges (e.g., methane fluxes) from different upwind areas to the total flux measured at

the sensor. It is expressed in square meters per square meter (m² m²), effectively a dimensionless ratio that describes what fraction of the measured flux originates from each unit area. This is commonly used in eddy covariance (EC) studies or controlled release quantification.

Footprints are often a product of Lagrangian air parcel models or Gaussian atmospheric dispersion models, which simulate the movement of parcels of air over time and distance. Properties: The dimensions and shape of the footprint are governed by wind speed, turbulence, measurement height (expressed as z), atmospheric stability, and surface roughness. Under stable, low-wind conditions, high localization potential is observed through narrow and elongated footprints (Czs > 0.1) while broad footprints with Czs < 0.01 are associated to enhanced spatial uncertainty in source attribution driven by wind extremes and high turbulence.

The footprint models in this study are important to connect measured concentrations with potential upwind emissions sources, which improves source apportionment from monitoring sensors that have only a single exposure point.

The concept of a "forward simulation" vs. a "backward simulation" should be defined in the introduction

This is a great point. We propose to add the following text manuscript: "In atmospheric transport modeling, a forward simulation predicts the downwind concentration field resulting from a

known or assumed emission source, often using meteorological data to simulate dispersion. In contrast, a backward simulation (also known as a backtrajectory or retro-plume approach) traces the path of air parcels in reverse to identify the potential upwind source regions contributing to observed concentrations at a sensor. These critical concepts support improved localization and quantification practices (Aubinet et al., 2012)."

L145: "source weight function" is not defined.

That is a valid point. This has all been explained in Göckede et al.'s 2004-2006 studies.

"A source weight function describes how sensitive a measurement at the sensor is to potential emissions at different upwind locations over time. Conceptually, it quantifies the influence a unit emission from any given location would have on the observed concentration. That Implies a footprint but not actually a footprint This is the source weight function, which describes the mathematical relationship, while footprint refers to the map or spatial representation of that relationship for visualization and interpretation. In Lagrangian backtrajectory models, the source weight function is typically computed by tracing air parcels backward and assessing their time within each grid cell, adjusted by atmospheric parameters like turbulence and mixing height. For a detailed explanation of these concepts in the context of flux modeling, see Lin et al. (2003) and Kljun et al. (2015)."

Second paragraph of 2.2. More details about the GI method are necessary. I don't understand some key ideas from this paragraph (e.g., the x and y direction and the new vs. original matrices).

This is addressed in a comment above

Each box in Figure 1 needs more detail to be interpretable. I'm not sure what a lot of these boxes mean, even after reading the previous paragraphs in Section 2.2

This is addressed in a comment above that explained the GI method with formulation. Figure 1 can be replaced with this one



The text for section 2.2 can be modified as follows: "The main loop of the GI tool consists of a ranking method to flag the highest concentration points using different gradient lengths (cell numbers) starting from one. This is an assumption due to a lack of information from the source location (meaning that each concentration point is being flagged as high compared to itself). As shown in Figure 1, based on the mathematical concept of the GI method, length L will be selected starting from one, meaning that each (x, y) cell is maximum compared to itself in both x and y directions. The length will increase incrementally until at least one of the remaining points on the matrix surpasses the top 5% CH<sub>4</sub> concentration cells on the original matrix. The recurring gradient loop counts the persistence of points with different lengths and will only keep the high persistence points

	C 1 TEDD A DEX	
	from the TERRAFEX contribution	
	source maps."	
Section 2.2 would be aided by a	Text to be added:	
Figure showing the concentration	"An example of concentration	
maps, and	maps and hot spots located by the	
perhaps an example of a "hot spot"	GI tool is shown in Figure 6."	
Figure 3 is not a "time series," as it is	Correct. We propose this fix:	
referred to on L271. A time series	"Methane concentrations vs wind	
would plot the methane concentrations	direction at a sensor receptor,	
on the vertical axis with time on the	arriving from two points of	
horizontal axis.	emission upwind, driven by real	
These plots show concentration as a	varying winds representative of	
function of wind direction.	four different stability classes"	
Fig 5: Could there be other sources on	Thank you for this thoughtful	
this site besides the four you	comment. You're correct that an	
identified? If so,	overreliance on prior site	
how would this impact the analysis?	knowledge could appear to	
Some discussion of this point should		
be	contradict the potential of	
included.	Lagrangian modeling to identify	
in orange.	unknown emission sources.	
	Our analysis did not rely	
	exclusively on prior knowledge of	
	the four source locations. Instead,	
	the spatial and temporal gradients	
	in the sensor data—interpreted	
	through the Lagrangian back-	
	trajectory model—consistently	
	pointed to four distinct emission	
	regions, which aligned well with	
	the known infrastructure. This	
	agreement serves as mutual	
	corroboration: site records	
	confirmed these were plausible	
	source locations, while the	
	modeling supported their presence	
	based on observed concentration	
	patterns.	
	We believe that if additional,	
	unaccounted-for sources had been	
	present—especially of comparable	
	magnitude—they would have	
	,	
	produced distinct anomalies or	

	unexplained gradients during	
	measurement. For example, a	
	hidden source south of the current	
	sensor array would have likely	
	shown elevated concentrations	
	during southerly winds that could	
	not be attributed to the known	
	four sources. We did not observe	
	any detectable unexplained	
	patterns in the data.	
	Text to be added:	
	"Our analysis revealed four	
	spatially distinct source regions	
	following analysis of modeled	
	upwind sensitivity patterns and	
	concentration gradients observed	
	in the measurements. This	
	coincided with known site	
	infrastructure and assured us that	
	we were correct in our	
	interpretation of the data. While	
	we cannot rule out the existence	
	of minor or intermittent	
	unmonitored sources, we	
	observed no anomalous gradients	
	or unexplained patterns that	
	would suggest significant	
	unaccounted emissions. This	
	suggests that both site inspection	
	and modeling successfully	
	captured the dominant sources."	
Fig 6: It's unclear which source-sensor	It is the second configuration.	
configuration is shown here. Are there	There is only one sensor at x=y=0	
many sensors along the x=0 line? Or		
just one at x=y=0?		
Fig 6: I'm not sure what the	We appreciate the reviewer's	
concentration field on the right is	request for clarification. We have	
showing. I think more	expanded Section 2.1 to describe	
detail needs to be included in Section	better the TERRAFEX model,	
2.1 when describing TERRAFEX,	including how it reconstructs	
because it's unclear what the output from this model shows. Is it the	concentration fields using a back- trajectory framework. We also	
estimated plume shape based solely on	revised the caption of Figure 6 to	
estimated praine snape based solery on	explain that the right panel is not a	
	explain that the fight panel is not a	

the sensor measurements? If so, why is the plume wider on the left and narrower on the right? Also, can the authors explain the other artifacts in this concentration field?

forward-simulated plume but a reconstructed field that represents the spatial influence of upwind sources on sensor measurements. The apparent asymmetries and shape artifacts are due to the underlying grid resolution, smoothing method, and the limited angular sensitivity of sensors under certain wind conditions. The paragraph before the figure will be revised as follows:

"The right-hand panel shows the methane concentration field reconstructed using the TERRAFEX model for the same case. TERRAFEX uses back-trajectory simulations based on wind data to estimate the upwind contribution of potential source areas to each sensor reading. The resulting field is a probabilistic reconstruction of near-surface methane concentrations, derived by combining those back-trajectories with the observed CH<sub>4</sub> enhancements. The color scale represents methane concentration in ppmv, consistent with the lefthand panel. The reconstructed plume exhibits asymmetry and apparent artifacts—such as a wider spread on the upwind side and narrowing near the sensor line—due to the limited angular sensitivity of the sensors, interpolation over a discrete grid, and wind temporal variability during the reconstruction window.

This comparison demonstrates that at the downwind measurement location, the two

Fig 6: It would be very useful to show where the GI method would identify the source given this concentration map.	plumes as separate sources. This illustrates the significance of sensor placement: at measurement locations that are downwind of the two source locations, if measurements had been taken at or near the source (the upwind part of the domain), the spatial separation of the two plumes would be maintained, creating good opportunities for localizing multiple sources."  This figure can be added to show where the Gradient Indicator would identify the source.	
Table 1: Are these metrics averaged across all source-sensor configurations? If so, it would be very hard to interpret these numbers, as diLerent arrangements may have very different detection characteristics.  Figure 7: The 95% confidence interval almost spans the entire range of the POD parameter (0-1). Some discussion of this point should be included. How confident are you in these trends given the very large confidence intervals?	This has already been discussed in a comment above  Despite the wide CIs, the median (50%) trend remains robust, and in most cases, the POD exceeds 50%, even under less favorable conditions. That said, the uncertainty captured in the 95% CI highlights that individual detection events are more variable, and performance under these conditions should be interpreted	

cautiously.

We propose to revise our description in the text: "In panel a, POD increases with atmospheric instability, with the highest detection probabilities observed under stability classes A, B, and C. In these unstable-toneutral conditions, detection rates often exceeded 90% when at least one source directly faced the sensor. Conversely, more stable conditions (D-class and below) showed reduced performance, though POD remained above 50%. Notably, the wide 95% confidence interval—particularly under stable conditions—reflects greater variability and uncertainty in detection performance under such scenarios. This underscores the challenge of consistent detection when dispersion is limited or plume-sensor alignment is suboptimal.

"In panel b, POD increases with relative distance from the source up to around 125 meters, after which it plateaus near 100%. This trend may result from enhanced plume overlap and alignment at mid-range distances, which improves detectability. Shorter distances could reduce POD due to narrow or offset plumes missing the sensor. The confidence interval narrows as distance increases, suggesting more consistent detection performance when sensors are positioned further downwind.

"In panel c Increasing the relative height of the sensor above the source also improves POD, with the highest detection probabilities

	occurring at the maximum tested height of 6 meters. This likely results from improved vertical intersection with the plume, especially under unstable conditions where the plume ascends more aggressively. The narrowing CI at higher sensor heights suggests increased detection reliability when sensors	
Figure 7 Panel a: The stability classes are sorted so that the POD increases from low to high. This needs to be stated directly; otherwise this plot is misleading.	are elevated."  True. We propose to add this text: "In Panel (a), stability classes are intentionally arranged by increasing median POD rather than meteorological order to illustrate the relationship between atmospheric stability and detection performance clearly."	
Figure 7 Panel b: the 100 mark on the horizontal axis is not in the correct order. I think this is also plotted so that the POD is strictly increasing, which is misleading. The horizontal axis should be increasing distances.	We propose to replace that graph with this one, which is closer to the reviewer's description:  1.00  1.00  25  Relative distance between source and sensor	
Figure 8: Is one pixel selected as the source location estimate? Or is the entire red swath the source estimate? The red region covers the sources, but it also covers much of the rest of the site. How do you pick a source location from the concentration maps shown in this figure?	It depends on the specific hot spot map we are looking at, but it can be between 1-4 potential source locations. We are simply showing one example of the Grandient Indicator procedural steps. If it continues, the persistence of points over the site area will retain potential candidate points.  We usually don't search beyond a footprint length exceeding base expectations. For example, using a sensor at 4 meters in height, we	

	would not expect to see anything outside a 40-200 meter distance, so anything outside that will be treated as an artifact. This could be added to the text: "In the Gradient Indicator (GI) maps, red regions indicate areas with high gradient strength across the modeled footprint domain. These do not represent a definitive source location but rather a set of candidate source regions. The final source estimate is typically selected from among the most persistent high-gradient locations, often corresponding to one to four points. We limit the search space based on our expectations of footprint length to limit false detections, so for example, a 4 m height sensor indicates maximum source attribution distance of ~ 40–200 m downwind. Any detection further than this is considered a possible artifact and was dropped out of the selection."	
The POD values in Tab 3 don't seem to line up with the values in Fig 7 panel C. This needs to be checked.	Thanks for pointing this out. There was an issue with the x-axis height difference. The new graph will be added to the script.	
The localization accuracy (LA) acronym is defined twice in the abstract	Thank you for catching this. We propose to fix this.	
Methane is defined as "CH4" twice in the first paragraph of the introduction,	We propose to fix this.	

		1
but the authors continue to use		
"methane" throughout the paper.		
L53: Jia et al. 2023 has been	We propose to fix this.	
published:		
https://doi.org/10.1038/s41598-025-		
99491-x		
L59: There is no Daniels et al. 2022 in	We apologize for the typo. It is a	
the list of references. Do the authors	2024 paper. Corrections will be	
mean to	made to the manuscript.	
say Daniels et al. 2023 or Daniels et		
al. 2024?		
L71: The Gaussian puff and plume	We propose to update our text to	
models studied in Jia et al. 2025 are	read:	
not back-	"Jia et al. (2025) explored the	
trajectory methods. They are forward	application of Gaussian plume and	
models that simulate the transport of	puff models as progressive	
methane	diffusion techniques for CH <sub>4</sub>	
from the source to the sensor.	transport and found them to	
	perform well in assessing complex	
	release events. In comparison,	
	simple back-trajectory approaches	
	as used in commercial CEMS are	
	often incapable of providing the	
	horizontal/spatial scale or time	
	resolution necessary to accurately	
	attribute source events in these	
	types of scenarios (Daniels et al.	
	2023)."	
L91-98: This discussion would be	This added to the script	
clearer if the authors first provided a	"In order to have a more precise	
clear definition	definition, a flux footprint refers to	
of both "concentration footprint" and	the surface area from which	
"flux footprint."	contributions to the measured flux	
	at a given point, typically above the	
	surface, are derived. This area	
	integrates the effects of sources and	
	sinks that influence the net flux of	
	a scalar quantity due to turbulent	
	transport (Kljun et al., 2015).	
	Conversely, a concentration	
	footprint describes the upwind area	
	contributing to the observed	
	concentration at a measurement	
	point, like satellite imagery from	
	above. Conceptually, it represents	
	where the sampled air has come	

	C 11 1 1 1 1	
	from and how much each location	
	contributes to the measured	
	concentration. (Levin et al., 2020)."	
L113: "localization" is defined here	Thank you for pointing that out.	
but used multiple times earlier in the	The definition can be added at its	
introduction. It should be defined at its	first use.	
first use.		
L170: Need to provide some	This has been responded to before	
information about your coordinate	in another comment.	
system before you		
say things like the "x and y		
directions."		
L205: GDM not defined	We propose to add:	
	Gaussian Dispersion Model	
	(GDM)	
Eq 1 is missing a plus sign	We propose to fix this	
Figure 2 needs a legend for the x's and	Figures will be modified	
the red dashed line.	accordingly	
L297: A more precise definition of	We propose to add the following	
POD should be used. It looks like	text:	
there is a	"The most important term used	
Repeated sentence here as well.	here is Probability of Detection	
Repeated sentence here as wen.	(POD), a metric that describes how	
	likely a monitoring system is to	
	detect the presence of an emission	
Your definition of localization	when one occurs successfully."	
	This can be fixed as described.	
accuracy (LA) is more commonly		
referred to as		
"positive predictive value" or		
"precision.		
"It might be better to use these more		
common phrases.		
In the FNF equation, it is not clear	We would propose to add this.	
what the n_c refers to. It would be		
better to write		
out exactly how n_c is calculated (e.g.,		
$n_{tp} + n$		
_fn), as is done with the other		
equations.		