Response to Reviewers

Novel method based on transfer kernel to locate and quantify point-source methane emissions
using ground-based column observations

egusphere-2026-204

Klappenbach et al.

We thank both referees for their careful reading of the manuscript and their constructive comments, which have substantially improved the clarity and rigour of the paper. Below we respond to each comment individually. Reviewer comments are reproduced in *italics*, our responses follow in blue. Line numbers refer to the originally submitted manuscript unless otherwise noted. All changes have been implemented in the revised manuscript.

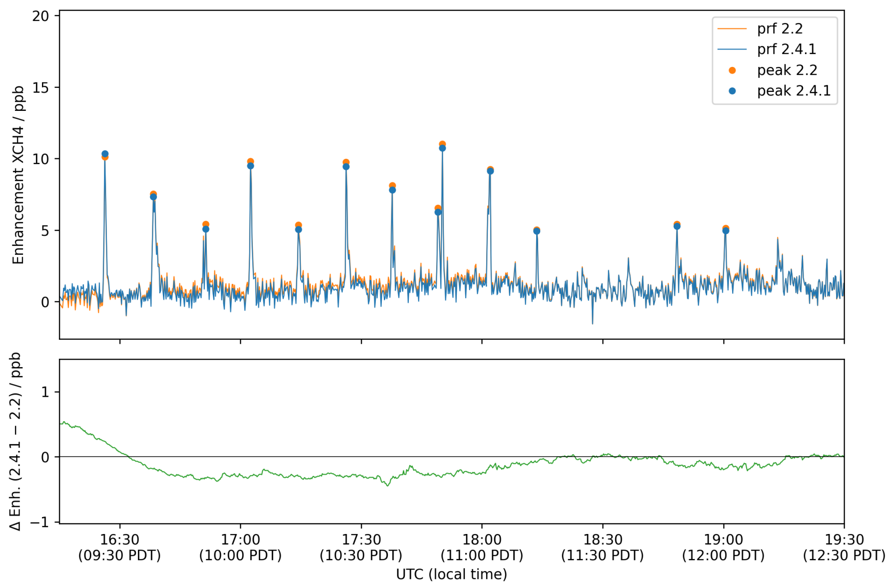
Reviewer 1 (RC1)

General comments

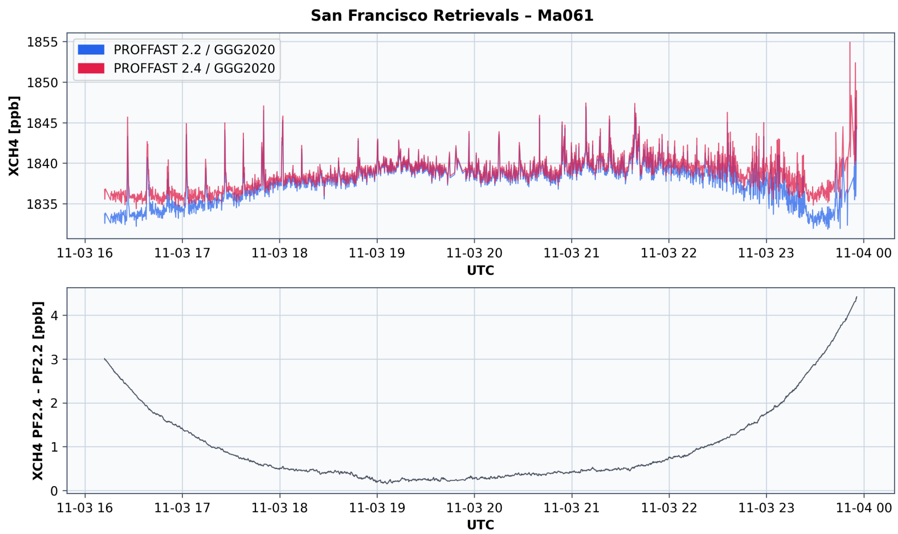
The analysis uses an older PROFFAST subversion (v2.2). The authors must consider updating the analysis to the latest PROFFAST v2.4.1 subversion.

Response: We reprocessed the full dataset with `proffast 2.4.1` as a consistency check. Retrieved peak enhancements change by $-2.9\,\%$ (median across the 13 forenoon peaks analysed, up to $-6.5\,\%$ for individual peaks) relative to the original `proffast 2.2` retrieval, calculated as (v2.4.1 − v2.2)/v2.4.1. The dominant driver is a revised correction for the solar-zenith-angle (SZA) dependence between the two `proffast` versions, which is parabola-shaped with a minimum near local noon. The correction is strongest in the morning and evening, where its slope shapes the baseline most. Because the peaks analysed in this study occur in the forenoon, the resulting difference is close to the maximum of what this effect produces across the day, and it propagates into the retrieved emission strengths in an almost linear fashion.

This shift is an order of magnitude smaller than the 30–70 % uncertainty on the retrieved emission strengths that dominates our error budget (Sect. 2.7), which is driven by wind-field and turbulence-related transport uncertainty rather than by the spectroscopic retrieval. We have added a sentence to the manuscript (Sect. 2.1, directly after the `proffast` citation) reporting this reprocessing check and its magnitude, and retain the original `proffast 2.2` retrieval throughout the manuscript, since the two versions do not lead to different conclusions.



Retrieved XCH_4 enhancement (top) and their difference (bottom, `proffast 2.4.1`−`proffast 2.2`) for the morning of 3 November 2016. All 13 peaks are reproduced almost identically by both retrieval versions, and the residual baseline difference is small relative to the peak amplitudes and does not affect peak identification.



Full-day XCH_4 retrieval comparison between `proffast 2.2` and `proffast 2.4.1` (top) and their difference (bottom), illustrating the parabola-shaped solar-zenith-angle correction with a minimum near local noon that drives the version-to-version difference reported above.

The data availability is currently listed as "can be provided upon request." [...] I would strongly recommend the data used in the analysis to be made publicly available [...]

Response: We agree and have revised the Data Availability statement. The processed data underlying all figures (retrieved column time series, background-subtracted enhancements, STILT footprint output aggregated to the segments used in the inversion, and the retrieved emission and distance estimates) will be made publicly available via GitHub at the time of publication, together with a runnable code example that reproduces the main figures from these processed data. The raw particle trajectories from the STILT ensemble are not included, as their volume (>100 GB for the full campaign) makes long-term hosting impractical. We consider the processed footprints sufficient to reproduce and verify the analysis. The manuscript text (Code and data availability section) has been updated accordingly.

Specific comments

L19: Please capitalize G, W, and P, if capitalizing in the acronym.

Response: Done. "global warming potential (GWP)" has been changed to "Global Warming Potential (GWP)".

L24: Please add citations/references.

Response: We have re-examined this sentence. The CH_4 uncertainty figure (30 %) was already supported by Solazzo et al. [2021]. The adjacent CO_2 uncertainty figure, however, was stated as "about 20 %" without a specific citation and, on closer inspection, did not correspond to a figure we could trace to a specific source. We traced the appropriate value to IPCC AR6 WGIII Chapter 2 [Dhakal et al., 2022], which reports a 90 % confidence interval uncertainty of ±8 % for global CO_2 emissions from fossil fuel combustion and industry (the same report, and the same underlying analysis by Solazzo et al., 2021, gives ±30 % for CH_4, consistent with our existing citation). We have corrected the sentence to state ∼8 % for CO_2 and added the corresponding citation.

L30: Here and in other places, is the order of the citations alphabetical or chronological?

Response: Multi-citations in the manuscript are sorted alphabetically by first author, following the default behaviour of the `copernicus.bst` bibliography style used for this journal. We have kept this convention throughout for consistency.

L30: Please change "by" to "using".

Response: Done.

L33: Just citing the previously published works without listing them as "e.g." should also be fine.

Response: We appreciate the suggestion. We have kept "e.g." at this location, as the citation list is illustrative rather than exhaustive (methane emissions arise from many more source types than the four cited references cover). We use "e.g." consistently elsewhere in the manuscript for the same reason and would prefer to retain this for internal consistency, but we are glad to change it if the reviewer feels strongly about it.

L63: Please expand on this explanation further.

Response: We have expanded the explanation of the black-box/LTI treatment in the Introduction to clarify what is, and is not, resolved explicitly by the transport kernel, and why this is sufficient for the purpose of the method.

L75: Isn't PROFFAST v2.4.1 the latest version? Can the analysis be updated to this PROFFAST version?

Response: Addressed together with the corresponding General Comment above. Please see our response there.

L89: Please change "encapsulates" to "represents".

Response: Done.

Figure 2: Can a different color palette be used? It appears that different altitudes are being represented by the same/very similar colors (e.g., brown).

Response: We would like to clarify the intent of Fig. 2 first, as this also affects RC1's later comment on this figure: colour in Fig. 2 encodes (distance × angle) segments *within a single, fixed altitude layer*, not altitude itself – the figure shows one representative layer, and altitude is not a colour dimension in this panel. We have amended the caption to state this explicitly. We have deliberately left the colour palette itself unchanged: the colours in this panel are purely local, qualitative labels used to make adjacent segments visually distinguishable from one another, and carry no quantitative meaning (segments do not need to be compared by colour across the panel, and two segments sharing a similar colour do not indicate any relationship between them). Since the colours are not intended to convey ordered or quantitative information, we do not think a different (e.g. perceptually uniform, sequential) colour map would improve the figure's usefulness for this purpose. We have updated the caption to state this clearly and concisely.

L118: Please remove "," after "thickness" and put "we use 10 m" within parentheses.

Response: Done: "the segment's vertical thickness (we use 10 m)".

Figure 3: Can a color besides green, such as red or pink be used here? The green and black lines appear to blend into each other.

Response: Thank you for flagging this. We have updated the colour palette in Fig. 3 to improve contrast between the fitted and observed curves. The revised figure is included in this submission.

L168: "This uncertainty can significantly increase..." – Please explain how.

Response: This point is addressed by the restructured uncertainty section (new Sect. 2.7, "Uncertainty analysis"), which replaces the previous brief treatment. Rather than a single qualitative statement, the uncertainty is now split explicitly into a distance-related term (Step 1, governed by the turbulence parametrisation used in the STILT kernel broadening) and an emission-strength-related term (Step 2, governed primarily by wind-speed error and propagated via Eq. 9, $\Delta E \approx E \cdot \sigma_u/u$). We quantify the emission-strength term at 30–70 % and explain explicitly why the distance term is not independently quantifiable with the available data (Sect. 2.7, "Distance uncertainty (Step 1)"), rather than leaving this as an unexplained qualitative statement.

L200: "Nevertheless, uncertainties remain substantial, mainly due to wind-field errors and observational noise." – Please be quantitative.

Response: Addressed by the same restructured uncertainty section. The Conclusion (Sect. 5) now states explicitly: "we estimate a residual relative uncertainty of roughly 30–70 % on the retrieved emission strengths, dominated by wind-field errors, with an additional systematic (and non-quantified) uncertainty on the source distance arising from the turbulence parametrisation." We also clarify that observation noise (quantified via the EM27/SUN Allan deviation, 0.2 ppb at 10-minute integration, Chen et al., 2016) is a *minor* contributor to the emission-strength uncertainty (signal-to-noise ratio of order 50 for the strongest peaks) and only becomes non-negligible for the source-distance estimate at low-amplitude peaks. The previous text conflated these two error sources. They are now treated separately and the wording has been corrected accordingly.

L205: "...a high uncertainty" – Of what value?

Response: Addressed as above: the distance uncertainty arising from the turbulence parametrisation is now explicitly identified as the dominant, directional, systematic error source for Step 1 (Sect. 2.7, "Distance uncertainty"), and we explain why it cannot be assigned a specific numeric value with the data available in this study (no co-located turbulence observation against which the STILT parametrisation could be constrained), rather than asserting a vague "high uncertainty" without further explanation. We consider a dedicated quantification (e.g. via a STILT ensemble with perturbed turbulence parameters) worthwhile future work and now state this explicitly in the Conclusion.

L207: "...method tends to overestimate" – By how much?

Response: We have considered this carefully but decided to leave this statement qualitative rather than attach an invented number. The overestimation in question occurs for sources that deviate from the isolated, point-like, puff-emitter assumption underlying the method (e.g. diffuse or extended sources), and its magnitude would depend strongly on the specific deviation (source geometry, emission duration, degree of overlap with neighbouring sources), which we have not systematically varied in this study. We believe a specific percentage here would suggest a precision the current analysis does not support. We have, however, added this case explicitly to the future-work paragraph in the Conclusion (validation against sources of known location and strength, including less prominent enhancements) as the appropriate route to obtain such a quantitative bound.

Figure 6: Could a different color palette be used [...] Please add a colormap. Please change the color of the purple and white site markers to something bright [...]

Response: We have revised Fig. 6 with a colormap and brighter, higher-contrast site markers as suggested. Specifically, we replaced the previous `jet` colour scale – whose low-residual end (the most important points, corresponding to the best-fitting, most plausible locations) was a dark blue easily lost against the satellite terrain – with a uniformly bright cyan-to-magenta scale, and replaced the dark purple/white site pins with bright yellow star markers. A colour-scale legend is now embedded directly in the figure. This is a plotting change only and does not affect the analysis. The revised figure is included in this submission. Beyond this plotting fix, we have also substantially expanded the candidate-source discussion this figure supports: Table 1 now lists ensemble-averaged emission estimates (mean/min/max, in $\mathrm{g\,s^{-1}}$, kg per peak, and $\mathrm{t\,CH_4\,yr^{-1}}$) for all five investigated upwind candidates rather than only the two most plausible ones, and the corresponding new Fig. 5 shows the retrieved emission strength per candidate for each of the 13 individual peaks with the ensemble range indicated. The main text now gives the physical exclusion criteria (missing blow-down vent for the natural gas valve, stack height $> 25\,\mathrm{m}$ for the two LBNL smoke stacks) alongside the distances upwind (91–938 m across candidates), so that a reader can judge the plausibility ranking directly against the quantitative emission estimates rather than the two primary candidates alone. We note for transparency that, in preparing this revision, we identified and corrected an error in the ensemble emission estimates for one of the two primary candidates (Student Housing). Table 1 has been updated accordingly, and the retrieved values are now in closer agreement with the independent heating-capacity estimate discussed in the main text. Table 1 additionally now reports the geographic location (latitude, longitude) of each candidate alongside its distance upwind of the receptor, so that the candidate positions shown in Fig. 6 can be cross-referenced directly against the quantitative table.

L215: Please ensure that the codes are available at the time of publication.

Response: Addressed together with the Data Availability General Comment above. The Code and Data Availability section has been revised to state a concrete commitment (GitHub, at time of publication) rather than an open-ended "upon request".

Data Availability: Please ensure that the data are publicly available [. . .]

Response: See our response to the corresponding General Comment above.

L228: Please remove "is". It occurs twice.

Response: Done.

L229: Please change to "trajectories discussed in Lin et al. (2003)."

Response: Done: the citation has been changed from parenthetical to in-text form at this location.

L244: Given "an" observed . . .

Response: Done: "a observed" corrected to "an observed".

L245: Please put "r_{max}" within parentheses.

Response: Done.

Please move the Author contributions, Competing interests, and Acknowledgements after Figure G6. [. . .] The Acknowledgements seem to have been split in the footers of pages 20 and 21. Please correct this.

Response: We have added a `\clearpage` before the `\authorcontribution` command in `template.tex`, which should move these sections after the appendix figures and prevent the page-break/footer issue. We will re-check the final PDF layout after all figure replacements (Figs. 2, 3, 6, and the new multi-peak validation figure, see RC2 Major #2 below) are in place, as float placement can shift once additional figures are added.

Reviewer 2 (RC2)

Major comments

[#1] Novelty relative to Patel et al. (2025) is not sufficiently clear.

Response: We have substantially expanded the novelty discussion in the Introduction, and have also given the method an explicit name for clarity and citability: Transfer-Kernel Ranging (TKR). The core distinction is now stated explicitly: TKR encodes source distance in the *shape* of the transport kernel (its progressive broadening with upwind distance) rather than in the amplitude of the observed signal relative to an emission-inventory prior. This makes the localisation step (Step 1) inventory-free and usable with a single time series from a single receptor, in contrast to (a) classical Bayesian inversions, which require a spatially resolved prior emission inventory, and (b) dense-network approaches such as Patel et al. [2025], which localise sources from the spatial structure of the plume sampled across many sensors. We explicitly frame TKR as *complementary* rather than superior: it is restricted to sources that approximate a single, isolated, puff-like emitter, whereas inventory-based inversions remain better suited to diffuse, distributed, or overlapping sources. This trade-off is now stated directly in the Introduction.

[#2] Step 1 (source-range estimation) is theoretically underspecified.

Response: We have added a theoretical justification for why the residual $\rho_s(r)$ is expected to exhibit a minimum at the true source distance: the stochastic turbulent velocity component that STILT superimposes on the mean advective transport causes the transport kernel to broaden monotonically with upwind distance, so that kernels compared against a narrow observed peak yield low residuals only up to the true source distance (Sect. 3.2, “Step 1: Source-range estimation”). We also discuss the worst case explicitly: if wind conditions change during the trajectories' travel time (e.g. a sea-breeze reversal or frontal passage), the ensemble reaching a given distance can split into multiple pathways, and $\rho_s(r)$ may become multi-modal rather than showing a single clean minimum.

To empirically support this argument beyond the single case shown in the main text, we have produced $\rho_s(r)$ curves for all 13 forenoon peaks identified in the campaign and added these as a new appendix figure (Fig. G1, `Figure_multi_peak_validation.png`), together with a new appendix section (Appendix G, "Multi-peak validation of the range-estimation minimum") discussing the result. For 12 of the 13 peaks, both the kernel-only and the full kernel-times-emission residual curves exhibit a clear, well-defined minimum at a comparatively short upwind distance, consistent with the single-pathway argument in Sect. 3.2. The 16:38 UTC peak is the exception, showing a markedly flatter minimum at a substantially larger distance, which we attribute to the worst-case scenario discussed in the same section, where a change in wind conditions during particle transport splits the ensemble into multiple pathways. We retain this peak in the analysis, as its magnitude and periodicity are consistent with the remaining peaks, but note that its inferred source distance is correspondingly less well constrained than for the other 12.

[#3] The manuscript presents a single case study; how representative is this?

Response: We agree this is an important limitation and have made it more explicit rather than downplaying it. The concluding sentences of the Abstract now read: “The study demonstrates the feasibility of this approach for detecting and characterising isolated, point-like methane emitters when transport conditions are stable. Systematic evaluation across more variable conditions remains for future work.” We have also added controlled validation against sources of known location and strength – including cases with less prominent, lower-amplitude enhancements than the exceptionally clean signal analysed here – as an explicit direction for future work in the Conclusion (Sect. 5), and we discuss in Sect. 3.1 why the specific meteorological and signal conditions on 3 November 2016 were necessary prerequisites that were not met to the same degree on other days or at other sites during the six-week campaign.

[#4] Uncertainty quantification is insufficiently developed.

Response: We have restructured the uncertainty section entirely (new Sect. 2.7, "Uncertainty analysis"). Please see our detailed response under RC1 L168/L200/L205/L207 above, which addresses the same underlying concern from a different angle. In summary, the section now (i) separates distance uncertainty (Step 1, dominated by the non-quantifiable turbulence parametrisation) from emission-strength uncertainty (Step 2, quantified at 30–70 % and dominated by wind-speed error), (ii) makes the assumptions underlying the wind-speed propagation (Eq. 9) explicit, (iii) integrates the wind correction (previously in an appendix) into the main text as part of the retrieval itself, (iv) separates the ensemble spread across candidate source segments (spatial representativeness) from the inversion uncertainty at a fixed location, and (v) quantifies observation noise from the instrument's Allan deviation (Chen et al., 2016).

Minor comments

[#1] Typos ("ovserved", "Te ensemble range", "hords").

Response: Corrected throughout the manuscript.

[#2] Eq. B1: symbol Y used before it is defined.

Response: Corrected in Appendix B. Y is now defined before its first use.

[#3] The choice of a 10 % percentile / 60-minute window for the background estimate is stated as "uncritical" without support.

Response: We have replaced this unsupported statement with a quantitative sensitivity test and a physical criterion (Sect. 3.1). Empirically, varying the quantile (5, 10, 15, 20 %) and window length (30, 60, 90, 120 min) changes the mean retrieved peak amplitude across the 13 forenoon peaks by at most −5.6 % to +6.2 % relative to our reference choice (10 %/60 min, 7.38 ppb), an order of magnitude below the 30–70 % emission-strength uncertainty. The full table is given in the new Appendix Table B1. We complement this with a physical criterion, derived from a synthetic test on an isolated Gaussian peak: the background estimate is unaffected once the window exceeds roughly three times the peak's full width at half maximum (FWHM). Below this, the rolling quantile intrudes into the peak itself and both damps the retrieved amplitude and introduces a central dip. In the present dataset, the median FWHM (27 s) is roughly 130 times smaller than the 60-minute window, i.e. about 40 times beyond this requirement, which is why the choice is uncritical here. We also note, as a secondary finding, that for *periodic* peaks such as ours the window length is less critical than for an isolated peak of the same width, since the window then spans several repetitions of the signal rather than a single isolated feature. Two supporting references have been added, Ruckstuhl et al. [2012] (a canonical method reference for robust baseline extraction from atmospheric time series) and Edwards et al. [2025] (a systematic comparison of background-separation methods that independently states the same underlying assumption – that the background varies on a longer time scale than the local source signal).

[#4] Fig. 4: why does the inferred emission increase strongly with distance?

Response: We have expanded the caption of the corresponding figure (dilution: the footprint sensitivity of a segment decreases with its distance from the receptor, so a more distant source must emit proportionally more to produce the same observed enhancement) and note that the emission axis is shown on a logarithmic scale for this reason.

[#5] Only one instrument per day is analysed despite a six-week, six-instrument campaign; please justify.

Response: We have added an explicit justification in Sect. 3.1. The morning of 3 November 2016 was exceptional in two respects that are both necessary prerequisites for the method as presented: (i) wind direction and speed were unusually steady over several hours, a rare condition in this coastal region where sea-breeze circulation combined with complex terrain typically produces variable transport, and (ii) the observed enhancement pattern was strictly periodic and of high amplitude, strongly suggesting a single, consistent source. These conditions were not met to the same degree at the other sites or on other days during the campaign, which is why the present analysis focuses on this single case. We discuss this limitation further under RC2 Major #3 above.

[#6] Appendix A (LTI time-invariance assumption) is discussed too briefly.

Response: We have expanded this discussion. The relevant criterion for time-invariance is a change in the meteorological state between HRRR analysis time steps, not elapsed backward time per se. We clarify that HRRRv1 is updated hourly (not every 3 h, as an earlier draft implied), which sets the effective time scale over which the LTI assumption needs to hold.

[#7] Code/data availability should be more concrete.

Response: See our response to the corresponding RC1 comments above (L215 and the Data Availability general comment): the processed data underlying the figures will be made available via GitHub at publication, together with a runnable code example. Raw trajectories are excluded for practical (volume) reasons.

Summary of manuscript changes

- Uncertainty section (Sect. 2.7) restructured and quantified throughout (RC1 L168/200/205/207, RC2 Major #4).
- Novelty relative to inventory-based inversions and dense-network approaches clarified in the Introduction, and the method given an explicit name (Transfer-Kernel Ranging, TKR) for clarity and citability (RC2 Major #1).
- Theoretical justification and worst-case discussion added for Step 1 residual minimum, together with a new multi-peak validation figure and appendix section covering all 13 forenoon peaks (RC2 Major #2).
- Abstract conclusion softened to state feasibility under stable transport conditions rather than a general claim (RC2 Major #3).
- Background-estimation sensitivity quantified with a new appendix table and physical criterion, two supporting references added (RC2 Minor #3).

- PROFFAST v2.2 vs. v2.4.1 reprocessing check added (RC1, general comment, L75).
- Code/data availability statement made concrete (RC1 L215, Data Availability, RC2 Minor #7).
- Appendix A (LTI time-invariance) expanded (RC2 Minor #6).
- Figure 4 caption expanded to explain the distance–emission relationship (RC2 Minor #4).
- Six-week-campaign / single-day analysis justified explicitly (RC2 Minor #5).
- Figures 2, 3, and 6 revised: Fig. 3 and Fig. 6 recoloured for improved contrast, Fig. 2 caption clarified to explain that its colours are qualitative labels only and intentionally left unchanged (RC1, Figures 2/3/6 comments).
- Numerous typographical and stylistic corrections (RC1 L19/30/33/63/89/118/228/229/244/245, RC2 Minor #1/#2).

Further revisions made during final co-author review

Beyond the reviewer-triggered changes above, the co-authors identified a further round of improvements during final internal review prior to resubmission. We list these here for transparency, since some are visible in the manuscript diff but were not prompted by a specific referee comment.

- The title has been revised to *"Novel method based on transfer kernel to locate and quantify point-source methane emissions using ground-based column observations"*, to foreground the kernel-based mechanism of the method and to streamline the phrasing.
- Jia Chen has been added as a second corresponding author.
- Terminology clarified throughout: the *transfer kernel* (the impulse response of the transport system) and the *transfer function* (its Fourier transform) are distinct quantities and are no longer used interchangeably. The Abstract, Introduction, Conclusion, and Appendix A have been amended accordingly.
- An overstated claim in the Introduction has been removed: we had stated that standard Bayesian inverse frameworks "do not, by themselves, locate an unknown source." On reflection this is not accurate as a general statement – a spatially resolved inversion can in principle detect an unknown source located within its prior grid – so the sentence has been deleted. The surrounding discussion of TKR's inventory-free localisation as a complementary (not exclusive) capability is unaffected.
- The Abstract has been streamlined: the wide illustrative range of continuous-emission-equivalent annual totals has been removed in favour of directly attributing the two per-event mean emission rates to the two candidates (Student Housing, Foothill Dining), and a duplicated phrase ("installed power output of approximately 500–1000 kW installed power") has been corrected.
- Table 1: the two most plausible candidates (Foothill Dining, Student Housing) are now typeset in bold for readability, consistent with how they are already highlighted in the text and in Figs. 5 and 6.
- Uncertainty section (Sect. 2.7): the subsection headings for Distance uncertainty and Emission-strength uncertainty now cross-reference Sects. 2.5 and 2.6 explicitly, an ambiguous internal "(see below)" reference has been replaced by an explicit section reference, and the "Overall" subsection (renamed "Overall uncertainty") now gives an explicit qualitative ranking (major/moderate/minor) of the individual contributions discussed in the preceding subsections, in addition to the existing quantitative 30–70 % estimate.

- A single-item subsection (2.3.1, "Discretisation of the upwind domain") has been converted to an unnumbered heading, since a numbered subdivision requires at least two siblings.
- Minor consistency fixes: the figure-reference order in Sect. 2.7.3 was corrected from "Figs. 5 and 4" to "Figs. 4 and 5"; missing semicolons were added to the Author Contributions statement; the Acknowledgements now name a specific student (Calin Hurley) who assisted with candidate-source verification, in place of a generic mention.

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